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UNITED STATES NUCLEAR REGULATORY COMMISSION'S
ADVISORY COMMITTEE ON NUCLEAR WASTE

July 18, 2006

The contents of this transcript of the proceeding of the United States Nuclear Regulatory Commission Advisory Committee on Nuclear Waste, taken on July 18, 2006, as reported herein, is a record of the discussions recorded at the meeting held on the above date.

This transcript has not been reviewed, corrected and edited and it may contain inaccuracies.

1 UNITED STATES OF AMERICA
2 NUCLEAR REGULATORY COMMISSION

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4 ADVISORY COMMITTEE ON NUCLEAR WASTE

5 172ND MEETING

6 + + + + +

7 TUESDAY,

8 JULY 18, 2006

9 + + + + +

10 ROCKVILLE, MARYLAND

11
12 The meeting convened at the Nuclear Regulatory
13 Commission, Two White Flint North, Room T-2B3, 11545
14 Rockville Pike, at 8:30 a.m., Michael T. Ryan, Chair,
15 presiding.

16 COMMITTEE MEMBERS PRESENT:

17 MICHAEL T. RYAN	C h a i r m a n
18 ALLEN G. CROFF	Vice-Chair
19 JOHN T. LARKINS	Executive Director
20 JAMES H. CLARKE	Member
21 WILLIAM J. HINZE	Member
22 RUTH F. WEINER	Member

23
24
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1 PANEL MEMBERS PRESENT:

2 RACHEL J. DETWILER Braun Intertec Corp.
3 LESLIE DOLE ORNL
4 EDWARD GARBOCZI NIST
5 FRED GLASSER Aberdeen University, UK
6 DAVID S. KOSSON Vanderbilt University
7 CHRISTINE A. LANGTON SRNL
8 BARRY SCHEETZ Pennsylvania State
9 University

10

11 ACNW STAFF PRESENT:

12 ANTONIO DIAS
13 LATIF S. HAMDAN
14 MICHAEL P. LEE
15 DEREK WIDMAYER

16

17 NRC STAFF PRESENT:

18 DAVID ESH NMSS/DWMEP
19 JOHN FLACK ACRS
20 BRET LESLIE NMSS/DWMEP
21 A.C. RIDGE NMSS/DWMEP

22 VIA TELEPHONE:

23 LANE HOWARD CNWRA
24 LINDA LEHMAN Stanford

25

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P-R-O-C-E-E-D-I-N-G-S

8:31 a.m.

CHAIRMAN RYAN: Okay, if we could come to order, please. The second day of the 172nd Meeting of the Advisory Committee on Nuclear Waste. During today's meeting, the Committee will conduct a working group meeting on predicting the performance of Cementitious Barriers of Near Surface Disposal. The meeting is being conducted in accordance with the provisions of the Federal Advisory Committee Act.

Latif Hamdan is the Designated Federal Official for today's session. We have received no written comments or requests for time to make oral statements from members of the public regarding today's session. Should anyone wish to address the Committee, please make your wishes known to one of the Committee staff.

It is requested that the speakers use one of the microphones, identify themselves and speak with sufficient clarity and volume so they can be readily heard. It's also requested that if you have cell phones or pagers, that you kindly turn them off. I'd like to add a note of welcome to all of panel participants today. I know you've traveled from far and wide to be with us and we appreciate you sharing

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1 your expertise with us today and giving us you
2 insights on this important topic to the Committee.

3 Vice-Chair Allen Croff is chairing our
4 working group meeting, so without further ado, I'll
5 turn that meeting over to Allen.

6 VICE-CHAIRMAN CROFF: Thank you, Mike.
7 Good morning, all. On behalf of the ACNW, I too would
8 like to welcome our speakers and the audience to the
9 working group meeting. By way of background,
10 cementitious materials have been used to stabilize and
11 isolate radioactive waste for many years. However,
12 it's only recently that organizations responsible for
13 the waste have decided to try to take credit for the
14 beneficial characteristics of the cementitious
15 materials. The most visible example of this, I think,
16 is in the Department of Energy in their Tank Waste
17 Management Program, where they're using it to
18 stabilize the tanks and low activity waste both in the
19 near surface.

20 There are also signs that credit may be
21 taken for the beneficial effects of cement in
22 decommissioning applications, although that's a little
23 bit further out on the horizon. All of this raises
24 the issue of how much credit can be taken for the
25 cementitious barriers and for how long. That is, what

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1 do we know about predicting the performance of
2 cementitious barriers into the future. The purpose of
3 this working group meeting is to address this issue by
4 considering three questions.

5 Where are cementitious materials being
6 used in radioactive waste disposal, and what are the
7 potential beneficial effects of the cements? What are
8 the failure modes of the cementitious barriers
9 relevant to the performance over long times, centuries
10 out to the millennia and what's the current state of
11 technology for predicting the performance of these
12 cementitious materials?

13 Information gathered here today will
14 provide the basis for the Committee to provide
15 technical advice to the Commission concerning the
16 reliance on these barriers and identify areas where
17 additional information is needed. I might add that I
18 hope it will produce some information that's relevant
19 to the performance of concretes in applications such
20 as nuclear power plants, which is of interest to the
21 ACRS. This working group meeting will have three
22 sessions corresponding to the questions I outlined
23 earlier. To address these questions, we have a very
24 distinguished group of scientists and engineers. They
25 have extensive experience concerning cementitious

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1 materials. But before launching into the program, per
2 se, I'd like to highlight a few items on the structure
3 of the meeting.

4 Within each of the three sessions, we're
5 going to have two or three speakers followed by a
6 panel discussion where the Committee members,
7 Committee staff, other speakers and NRC staff can ask
8 the panelists questions. It would be useful if the
9 questions for the speakers could be held until the
10 panel discussion, that is try not to interrupt the
11 speakers and their flow.

12 After the three sessions, later in the
13 afternoon, there will be a panel session involving all
14 of the speakers for us to catch up on anything that
15 happens later that needs to be addressed to earlier
16 speakers. And then, as Mike has said, there's an
17 opportunity for the public to weigh in and if you want
18 to do so, you need to sign in and let a member of the
19 staff know.

20 Before going into the agenda, I think we
21 have some folks on the phone. Would you introduce
22 yourselves?

23 MS. LEHMAN: Linda Lehman at Hanford.

24 MR. HOWARD: This is Lane Howard, the
25 Center for Nuclear Waste Regulatory Analysis in San

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1 Antonio.

2 VICE-CHAIRMAN CROFF: Okay, anybody else?
3 Hearing nobody else, I think that's it. Welcome.
4 These introductions out of the way, I'd like to begin
5 the first session concerning the applications of the
6 materials and how they're important to performance
7 assessment. Our first speaker is Dr. Chris Langton
8 from Savannah River National Laboratory. Dr. Langton
9 is a Senior Fellow Scientist at SRNL. Her areas of
10 expertise include waste treatment, cementitious waste
11 form designs, construction grout design, and the geo-
12 chemistry of cementitious systems.

13 She has a master's degree in geo-chemistry
14 and a PhD in material science and engineering from
15 Penn State. Dr. Langton.

16 DR. LANGTON: Is this the format, stand
17 here or sit?

18 VICE-CHAIRMAN CROFF: You can sit there as
19 long as you speak into the microphone or we have a
20 portable mike if you want to use that, someplace.

21 DR. LANGTON: Well, I was asked to speak
22 on designing cement waste forms and the use of
23 cementitious materials for contaminant management in
24 the environment. I'll review some applications for
25 use of cementitious materials in this -- in managing

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1 contaminants. I guess I should look at this. I can't
2 figure out what to look at. The applications include
3 waste treatment, containment, which would include
4 vaults or casks, drums, culverts made out of concrete,
5 environmental remediation, I'm not going to speak too
6 much on that. I'm not really going to touch on that
7 today, just to list it. Facility closure, I'll
8 include some information on functional requirements as
9 I see them from a person designing waste forms, go
10 over material requirements for performance and
11 processing and laboratory qualification of materials,
12 field testing of materials.

13 And then I've included the landfill also
14 because in order to design a waste form, any kind of
15 waste form, including a cementitious waste form, you
16 need to know something about the landfill or disposal
17 environment that that waste form will be placed in.
18 And then I'll talk, just briefly mention uncertainties
19 and touch on risk reduction. The applications for
20 waste include treating liquids, aqueous liquids,
21 sludges, slurries and wet solids, in addition to some
22 particulate material. Another application of cement
23 based containment, the Savannah River site in South
24 Carolina, we use concrete vaults. We also have
25 culverts and casks.

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Concrete at Savannah River site is used as a component for backfill in certain disposal applications and then it can also be used for trench liners and trench covers. For environmental restoration at the Savannah River site, we've used cementitious materials in basin and soil stabilization and in grout curtains for managing the flow of contaminated groundwater, for containing contaminated groundwater. And the grout curtain designs include gates to let the groundwater out after it has been treated or filtered in situ.

We've also done some facility closure at Savannah River site. The one that most people know about or the biggest one has been high level waste tank closures. We've also filled some small 20,000 gallon tanks that were buried underground in our disposal facility. We're looking at decommissioning another facility that was used to make the Plutonium 238 fuel pellets for the space program for heat sources to generate electricity in the space program. That program is just in the infancy stages right now.

As far as -- in this slide, I've just shown a schematic of how the materials are used. The saltstone facility -- why don't I not stand there. I'll stand here. The waste form is a saltstone in a

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1 concrete vault and then we also have a clean cap. The
2 ground material -- the ground material in the slide
3 would be like a containment or a fill material that's
4 not radioactive and that's an example where three
5 applications are used in just one disposal scenario.

6 This is a slide -- these are photos of the
7 saltstone facility. This is the processing room. The
8 mixer is up here and it goes through a series of pumps
9 and finally pumped out to a concrete vault. There are
10 two vaults in this picture, Vault 1, which is a single
11 -- it's got six cells and then the second vault, a set
12 of 12 cells. This is a picture of that first vault
13 under construction. That landfill is very important
14 in the waste form design. If you know what you have
15 and you know where you're going to put it and what --
16 how the two sets of requirements interact, you can
17 design in the middle to achieve your goals.

18 Currently, we're designing another type of
19 vault for the saltstone facility that's a one million
20 gallon prefabricated tank and that will be our --
21 that's the new concept for future designs. This is a
22 schematic drawing of a facility closure. In this case
23 the facility is a high level waste tank. We used
24 several different kinds of cementitious materials.
25 The one that's in contact with the waste which is this

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1 orange material at the bottom and this is residual
2 waste. This is chemically adjusted to control the
3 leaching, minimize the leaching in the -- of the waste
4 on the bottom or the residual heal, a film material,
5 an intruder barrier and then some just engineered
6 features to protect the openings into the tank.

7 This is some photographs of a facility
8 closing. This is one of the 1.3 million gallon waste
9 tanks at Savannah River that's under construction.
10 We had a portable grout plant set up to prepare the
11 film material. These are two 30-cubic yard an hour
12 plants. This photograph at the bottom is the grout
13 going in, the first layer of grout going in. There
14 was a pore over here, another pore and this pore is
15 happening in the center where it's -- and this is the
16 residual, tank residual material that was on the
17 bottom. This is in the same tank closer to the top
18 where we're putting in the bulk fill. That's this
19 line but we had a -- we designed a flowable, self-
20 leveling grout that flowed over the 80-foot diameter -
21 - flowed 45 feet in each direction to make a fairly
22 uniform surface.

23 These are some examples of waste and waste
24 forms. This is a picture inside of a tank. This is
25 not one of the tanks that we closed. This is one that

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1 has cooling coils. The salt waste in this tank is
2 dissolved to a salt solution which is pre-treated,
3 decontaminated and then intimately mixed with a
4 cementitious set of reagents to form a slurry. This
5 is a picture of the slurry after it's gelled. The
6 picture on Slide 2 is a picture of the slurry as it's
7 being -- after it was poured and -- after it's been
8 prepared and being poured into the sample containers.
9 It's very fluid material that, within 60 minutes gels
10 to look like this and it sets within two days.

11 This last slide -- this last photograph is
12 a picture of the residuals. This picture was taken
13 from a test, a field test that we did before putting
14 cementitious material in a tank. A small amount of
15 the cementitious material did go under the waste.
16 These are the solids. There was a little bit of
17 liquid on top that was top dressed with dry grout and
18 you see that material in here and then we -- the
19 addition of more grout resulted in covering it up,
20 encapsulating the waste. So those are some examples
21 of our approaches to radioactive waste treatment that
22 we've used at Savannah River site.

23 Now, the functional requirements of waste
24 forms, of cementitious materials used for contaminant
25 management and mitigation of mobility in the

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1 environment, in disposal environment include
2 stabilization, fixation, we reduce the mobility of the
3 soluble contaminants by either changing the chemical
4 form and/or reducing the surface area that's exposed
5 to the environment. Another function of stabilization
6 fixation is to reduce the mobility of dispersable
7 particles by macro encapsulation and increasing the
8 particle size, again, to decrease the exposure to the
9 environment. And then there's another function that
10 we like to accomplish and that's to create a
11 recognizable material that's distinct from the
12 environmental media with the idea of reducing exposure
13 and dispersion by human intrusion.

14 Waste form solidification, sometimes we
15 treat liquid waste, tritiated water for example, to
16 eliminate the liquid nature of that waste so it
17 qualifies for disposal in a landfill. And then the
18 functional requirements of landfills in which -- of
19 concrete materials or cementitious materials that are
20 used in landfills are to provide engineered barriers
21 and those engineered barriers are intended to reduce
22 infiltration of water and human intrusion and support
23 the over-burdens to maintain the integrity of the
24 capping material.

25 The waste form plus the landfill plus the

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1 environment make up the disposal system. And those
2 disposal systems also have requirements. One of them
3 or two of them are that they can be operated and that
4 they can be constructed. I guess in the opposite
5 direction, they can be constructed, and then they can
6 be operated. And finally, that they perform for the
7 required duration.

8 Now, if we talk about the design process
9 for designing waste forms or other disposal
10 applications using cementitious materials. First, we
11 need to characterize the waste and get a waste
12 classification. The waste classification is very
13 important to determining the path that's chosen. We
14 also need to select a disposal unit. Different
15 disposal units come with different concentration
16 limits, packaging requirements, handling requirements,
17 and once we get through this process, we would down
18 select a type of waste form based on an alternative
19 study or some testing, past experience, and down
20 selecting the waste form involves identifying
21 potential materials for the waste form and processing
22 for the waste form.

23 The processing depends on the amount of
24 material that needs to be processed, the rate at which
25 it needs to be processed, the location of the disposal

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1 site relative to where the waste is generated. And
2 the material requirements typically depend on the
3 contaminants. The next thing we do is test the waste
4 forms to optimize waste loading and to measure
5 properties. Properties are important with respect to
6 demonstrating that we meet the regulatory requirements
7 and properties that are important to the overall
8 performance of the landfill.

9 The disposal -- the treatment disposal
10 system is designed, that means, designing the
11 processing building or the process equipment for
12 making the waste form and making changes or modifying
13 the disposal site if necessary. Backfill performance,
14 we run performance assessment, modeling, calculation
15 that evaluate how the waste form containment in the
16 landfill and the environment interact. We address the
17 risks and approaches to risk reduction and determine
18 if there's a need for additional specifications or
19 modifications. Sometimes we're in this process for a
20 long time and if there's regulatory or -- regulatory
21 guidelines that we meet easily, we're in this loop for
22 a couple of years, two years. If we're in new
23 territory, it could be a few decades.

24 Waste form testing; the important thing to
25 get out of this slide is that the testing protocols

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1 that are in blue are in the literature. They're
2 standard. We don't have any problems running those
3 tests and we run the tests. You need to characterize
4 the waste, again, select the ingredients and the
5 proportions. Cure the samples, curing the samples, we
6 cure according to ASTM methods. Sometimes we have to
7 modify those methods but I'm talking about laboratory
8 curing, not field testing here.

9 We test the samples. Processing
10 properties, that's straightforward. Rheology
11 measurements, leachability on a crush sample, we do
12 EPA TCLP. Sometimes we change the leachate but run a
13 similar type of test. There's an ASTM test where the
14 leachate can be selected but again, it's a crushed
15 sample. Monolithic sample, on the monolithic sample,
16 we get an effective diffusion coefficient. Sorption
17 is going the other way, from extraction. We take a
18 material like a concrete and put the contaminants in
19 simulated groundwater and absorb them or sorb them
20 onto the material to get numbers, values for our
21 predictions. Mechanical properties are
22 straightforward.

23 The next one, the hydraulic properties, we
24 have issues in measuring permeability, hydraulic
25 conductivity of our materials or our waste forms,

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1 especially the waste forms that contain salts that
2 interact with the leachate and in getting moisture
3 characteristic curves for materials like concrete and
4 our waste forms in the cases where they have very low
5 hydraulic conductivities. Thermal properties, we
6 know how to do that. I don't know if they're ASTM
7 methods. We don't use ASTM methods, but we feel we
8 have the general properties down.

9 Degradation to susceptibility, there are
10 ASTM methods for concrete and there are standard
11 concrete practices for designing materials that have
12 low susceptibility to degradation for a variety of
13 mechanisms. The one thing to point out here is that
14 waste form are not concrete. They may have cement in
15 them but they're not concrete. There's a whole body
16 of literature, extensive literature on concrete but
17 that's not the same as a -- as waste forms.

18 Long term aging; accelerated test methods
19 and the evaluation and effects of the long term aging.
20 A lot of these concrete methods, there might be a
21 length change or usually it's an expansion that
22 indicates cracking has occurred. Now, that's
23 susceptibility -- that measures susceptibility to
24 degradation, not exactly how that's going to perform
25 in the long term or the rate at which that effect is

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1 going to take place. And property geometry changes
2 we call cracking geometry changes, whether it's
3 independent or coupled with -- independent, just one
4 mechanism being evaluated or coupled with several
5 mechanisms.

6 Now, in the landfill evaluation, again,
7 there are standard practices. For the material
8 property characterization and testing, for the
9 cementitious materials and for the environment, there
10 are standard tests. Constructability and
11 offerability, there are engineering practices, there
12 are codes, available materials, ALARA considerations
13 and costs. Performance of the landfill, we evaluate
14 landfills with lysimeters. At Savannah River we've
15 had a series of lysimeters over the years. Some were
16 -- we had three 30-ton blocks of our saltstone
17 material and then some smaller 20-pound blocks, 30 of
18 those 20-pound blocks disposed of in the environment
19 where different parameters were measured over time.

20 The performance of the landfill can be
21 evaluated in terms of sampling and monitoring of the
22 waste form and barriers and the environment as the
23 landfill is being used before it's closed. And then
24 to evaluate the landfill again, we use this predicted
25 modeling. We use flow and contaminant transport. We

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do this evaluation by commercial computer codes. We use deterministic to do the base case and sensitivity cases and we're using -- we've started to use probabilistic or uncertainty analysis. This predictive modeling besides data that's produced in the waste form and environmental characterization studies, the predictive modeling relies on scenarios and a number of assumptions, many assumptions.

There are uncertainties in the waste form design and data collection. At the top of the list is the analysis of the waste. Often we have limited samples or we design the waste form based on pilot scale or laboratory scale processing. There is sample variability. Detection limits, we have problems with low detection limits, low concentrations, and interferences. We have in the leaching absorption category, contaminant distribution and interaction between contaminants, competition between contaminants, the chemistry of the leachate. We can have leachates from groundwater to rainwater, the TCLP leachates, the ionized water.

In some aspects the chemistry leachate has a significant effect and in other -- on other parameters, it has very little effect. So there's no -- there's no worst case because -- or there's no

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1 conservative case for all parameters that are of
2 interest. And then we have laboratory versus field
3 conditions. There are issues with equilibrium,
4 attainment of equilibrium, evaporation, flux extremes,
5 exposure to leachate, and coupled effects.

6 For hydraulic properties, the low
7 permeability makes the measurements difficult. The
8 poor size distribution in the cementitious materials
9 also makes the measurements and interpretation of the
10 measurements over the long term difficult. There are
11 scaling factors, hydraulic conductivity of a
12 laboratory sample is not the same as a hydraulic
13 conductivity over a 100 by 100 by 25-foot block in the
14 ground enclosed in concrete. There's again,
15 laboratory versus field aging. The curing conditions
16 used in the laboratory are standardized to eliminate
17 a variable so that other parameters can be studied.

18 Field aging is field aging. It's variable
19 through the course of the time that the sample is
20 exposed. And again, there are the coupled effects;
21 hydraulic properties effect leachate chemistry,
22 leachate chemistry effects hydraulic properties and
23 all this is taking place over time. Thermal
24 properties are not a particularly big problem or
25 insurmountable problem. The impact of thermal

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transients during curing on the other properties -- the curing time is short. For concrete we would think of a few months. For the waste forms we might think of a few months to a few years. But the bottom line here is the effects of those thermal transients can be measured in a relatively -- in a reasonable amount of time and incorporated into the modeling. However, this isn't always done, again, because field conditions and laboratory conditions are different.

The durability, we have chemical and physical durability. There are events that occur, catastrophic events, continuance events. The timing of the catastrophic events is significant and the rate at which continuous events, if they are continuous in fact, are difficult to determine. It's relatively easy to identify susceptibility to degradation, but it's difficult to turn that susceptibility into a rate and extrapolate for years, 1,000 to 10,000 years.

There are coupled effects and that's the manifestation and the impact on the rates of other changes that are taking place. And I just -- I listed a few, again, mineralogical changes impact the mechanical properties, the hydraulic properties, the leaching properties. There's alkaline environment in some of our waste forms. There's a chemically

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1 reducing environment, all of which -- some of these
2 specs could lead to cracks, the crack location,
3 pattern, dimension, inter-connectivity and in-filling.
4 Healing of the cracks is a good possibility over long
5 time periods.

6 Now, I included the landfill because it is
7 an important part of the overall objective here which
8 is disposing of certain wastes. The landfills,
9 performance time is up to 10,000 years or the time of
10 maximum contaminant -- the maximum of the peak
11 concentration in the compliance well, wherever the
12 compliance well happens to be. The factors that
13 effect that, extrapolate field conditions, there are
14 seismic events, climate change, landfill use, intruder
15 scenarios, toxicology, and we joke about it at
16 Savannah River, but maybe in some 1,000 years, people
17 will have evolved to be radiation resistant. We may
18 be doing something, over-kill here with our
19 requirements.

20 Also there are programmatic changes and
21 regulatory and public perception changes that we have
22 to deal with and these last two are not technical but
23 they do impact the design of waste forms. Now,
24 approaches to risk reduction; this is what we're doing
25 at Savannah River site. We're statusing our current

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1 state of knowledge and our programs with respect to
2 the current state of knowledge. Technical information
3 on rates of change, I don't want to say degradation,
4 just rates of change of materials over long time
5 periods. We're reviewing regulations and DOE orders
6 and interpreting how those changes impact our
7 programs. We're reviewing to improve -- we're
8 reviewing our strategy with the idea of improving our
9 current and future disposal systems, performing
10 sensitivity analysis for our current and proposed
11 units, disposal units.

12 An example of a way of reducing the risk
13 is to better capture the initial conditions and the
14 initial material properties. So right now we have an
15 emphasis on better descriptions and incorporating
16 those descriptions into our long-term predictions, as
17 a starting place for long-term predictions. Where
18 there's a need is to develop innovative approaches for
19 aging and waste form testing and then to develop a
20 framework for interpreting the test results. This is
21 probably -- this is a bigger issue than one would
22 think.

23 Obtain peer review approvals for the
24 current and future work and establish priorities for
25 future work. And I've summarized it in this slide.

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1 We know the waste treatment requirements. We know the
2 disposal site requirements. We pre-treat the waste to
3 get from here to here. We may need to modify a
4 disposal unit. We design a waste form, a waste form
5 production process and a packaging process, packaging
6 system, perform the performance assessment and through
7 testing and inputting that would be testing models.
8 Then we evaluate the risk and if we find there's
9 acceptable risk, we can treat and dispose and if we
10 don't we come back up here, start with the pre-
11 treatment.

12 Pre-treatment is really inventory
13 reduction. We can remove some contaminants, remove
14 bulk waste, remove waste, and start the process over.
15 Now, this looks simple but issues to be pointed out
16 here are that it's unclear at this time what it takes
17 to get through this risk, acceptable risk decision
18 point to a yes. We don't have a clear set of
19 criteria. This would be like going into court and
20 you'd need to know what evidence needs to be presented
21 to get a certain verdict or what level of evidence,
22 how much of that evidence. And I'm not sure that we
23 have this right now. For example, what case needs to
24 be made for this yes decision.

25 Right now, we're looking at using

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1 predictions, using scenarios for intruder analysis,
2 using extrapolations of groundwater contaminants, but
3 we don't look at the ability to remediate and is our
4 situation that we're coming up with if there was a
5 problem, if we didn't fully address the risk or if we
6 didn't analyze the testing, if we didn't do a complete
7 set of testing and I don't know how you'd do a
8 complete set of testing for 10,000 year predictions,
9 but if we could do that, what if we come up to the
10 acceptable risk and find out that remediation, if
11 there is a problem is known technology and people have
12 been remediating groundwater for 50 years using the
13 technology.

14 Remediation is not included in this risk
15 analysis to get us over here. And risk reduction --
16 I'm sorry, remediation is a viable strategy for
17 shallow land disposal, maybe a lot more so I would
18 think than for geologic disposal. But we know sort of
19 who makes this analysis, it's DOE, NRC, National
20 Academy of Sciences, and other state and federal
21 regulators, the public, our peer group. We know who
22 the judges are, and that's expert witnesses, our peer
23 reviews, but they can come down on either side of an
24 issue. I mean, how you present enough case to get
25 into the -- to go -- to treat and dispose is unclear

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1 to me. And that's something I pose to this Committee
2 is to help us figure out. That's it. Are there any
3 questions?

4 VICE-CHAIRMAN CROFF: Thank you very much.
5 If we could, I'd like to defer questions until after
6 out next speaker and then we'll have at it. I suspect
7 we'll have one or two.

8 Before moving on, to the speakers, you
9 have your choice up front. You can either stand
10 fairly fixed as Dr. Langton has done and speak into
11 the microphones that are there, which means you can't
12 move around. Or there is a portable microphone there,
13 if you like to wander around a bit when you speak,
14 that's an option also. So it's your choice.

15 With that, our next speaker is Dr. David
16 Kosson. Dr. Kosson is a Professor and Chairman of
17 Civil and Environmental Engineering at Vanderbilt
18 University. He's carried out research on leaching
19 processes and performance assessment including
20 cementitious and other waste forms for approximately
21 20 years. Dr. Kosson will talk on conceptual models
22 and approaches of understanding long-term performance
23 of cementitious waste forms.

24 DR. KOSSON: I have trouble standing still
25 so I might as well use this one. Okay, what I wanted

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1 to do was to lay the framework for how -- oh, that's
2 no good at all, can't get too close -- about how we
3 think about the various waste forms and their long-
4 term performance with respect to leaching. I want to
5 acknowledge that the work I'm presenting today and the
6 concepts come out of collaborations within my own
7 group at Vanderbilt and the department, other faculty
8 members there, collaborations that we've had with the
9 Netherlands Energy Research Center for about 20 years
10 as well, working very closely, DHI in Denmark and then
11 also working with some folks at the Savannah River
12 National Lab. You just heard from Chris, and also
13 from Pacific Northwest National Lab, dealing with some
14 of the Hanford programs over the various years.

15 When looking at it, Chris gave an overview
16 of the various disposal scenarios involved but what I
17 wanted to make a point with this slide is that you're
18 looking at the performance of an overall disposal
19 system and that system includes both the waste form,
20 it can be concrete structures that the waste form is
21 deposited into and other barriers and caps and
22 drainage layers that can be important. So you're
23 looking at overall performance, not only the waste
24 form and the influence of the external conditions on
25 that waste form, but also migration of constituents

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1 from that waste form such as sulfate or chloride which
2 may, in fact, influence the containment structure such
3 as degradation of reinforced steel and concrete
4 systems.

5 The motivation why we look at these things
6 is for a number of reasons. I'm sure you're familiar
7 with many, if not all of these, but the perform
8 assessments and the waste incidental to reprocessing
9 determinations or 3116 determinations and Chris went
10 through a number of these different applications. But
11 what I also think is important is to recognize that
12 cementitious waste forms are one of many waste forms
13 that are being considered for near surface land
14 disposal systems and as a result, you need an equal
15 comparison base to understand what the performance of
16 these are. And frequently what's used as over-
17 simplified methods, do not give you a good
18 understanding of the various processes the occur over
19 time and their interactions to give you a comparative
20 base say between a cementitious waste for and say a
21 steam reforming waste form that may be under
22 consideration.

23 So ultimately, then you have the
24 performance assessments, but then you have your waste
25 acceptance criteria which can be backed out of that.

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1 Your operational controls, how do you deal with day-
2 to-day quality control and performance and then also
3 when you're trying to make decisions about future
4 waste forms. We're at a time right now where the
5 country is considering reprocessing again. We're
6 considering different future nuclear fuel cycles.
7 Looking forward, these fuel cycles are going to
8 generate waste. Some of those wastes are going to be
9 targeted for near surface land disposal and how do we
10 evaluate them?

11 Often for the waste that we see right now,
12 the constituents of concern are some are long-lived
13 and mobile. Tc-99 is certainly one of the key ones
14 that's of concern. Iodine 129 often. And then in the
15 mobile and not as long-lived cesium-137, strontium-90
16 and then often at some of these wastes we see nitrate
17 in very high concentrations and tritium which are not
18 radio nuclides but also have the potential to impact
19 both the performance of the system and some of the
20 constituents of interest down gradient.

21 So some of the broader questions that
22 you're typically trying to address is what is the
23 appropriate type of waste form disposal system and
24 location for specific waste, because, as I mentioned
25 earlier, it's that overall system and the local

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1 conditions become important; to estimate the long-term
2 waste form performance and disposal system
3 performance, to establish the treatment criteria and
4 to define monitoring requirements. If you understand
5 the mechanisms, I would suggest what you want to do is
6 to monitor pre-cursors to impact so that you can be
7 pre-emptive in your system rather than going into
8 remediation mode after the fact.

9 So what I want to do then is overview what
10 we consider to be the primary factors and mechanisms
11 that you have interactions as you view these systems
12 and one is your system integrity and you've got to
13 look at the coupled effects of the engineered and
14 institutional systems. And often you see in the
15 regulatory environments and some of the other
16 regulatory programs, you engineered barriers are
17 separated in terms of performance monitoring,
18 integration from your institutional performance and
19 bring those two together, I think is very essential as
20 you look at the integrated system.

21 From the waste form performance itself,
22 I'm going to talk about the physical integrity, water
23 contact, the moisture status, your oxidation rates and
24 extent and the chemistry and mass transport and then
25 sum that up with how we view it in an integrated

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1 approach.

2 So with the physical integrity, you can
3 start off with an attack monolith but over time, you
4 go to a stressed matrix and you have some micro-
5 cracking, macro-cracking occurring and then ultimately
6 a spalled matrix. So what may become initially a
7 diffusion controlled release, your mechanisms of
8 release vary in your varying degrees of infiltration
9 over time as you look at the system degradation. So
10 over time you may see convective flow through it and
11 your ultimate release at this end state may be based
12 on what the equilibrium conditions are, local
13 equilibrium in that long-term, even though your
14 initial release and for some considerable period of
15 time, may be dictated by the boundary conditions and
16 those diffused responses.

17 With moisture transport, it is an
18 important controlling variable which I think is often
19 overlooked in these systems. The reason why it's
20 important is not only because of the percolation
21 through the system, but your moisture status in the
22 system, your pore structure and whether you're
23 saturated, say a capillary saturation or whether
24 you're unsaturated has an important impact on the
25 transport processes that occur.

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1 You can go through several different
2 scenarios. One is where you're saturated and you have
3 a continuous liquid phase and discontinuous gas phase.
4 At that point, you have liquid diffusion processes
5 which become very important and gaseous diffusion
6 processes become relatively unimportant because it's
7 in isolated pockets of vapor space. However, you have
8 a transition phase where you have continuous liquid
9 and a continuous gas process phases and at that point,
10 your liquid diffusion and your vapor diffusion become
11 important because you can have gasses for example,
12 oxygen or carbon dioxide infiltrating into the
13 material both through the liquid phase and through the
14 gaseous phase.

15 And then you have completely dry or --
16 which you'd probably never get to in field conditions,
17 but you have what's also referred to as insular
18 saturation where you have a discontinuous liquid phase
19 and a continuous gaseous phase. When you have a
20 discontinuous liquid phase, you don't have leaching
21 occurring obviously, because you have no complete
22 pathway but you still have vapor transport occurring
23 so you have oxidation, carbonation, other reactions
24 still occurring.

25 And if you look at this from an overall

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1 perspective, if you look at the physical resistance or
2 D/D0 for your diffusivity, what you see is you go
3 through these boundary conditions. As you increase
4 saturation, your liquid diffusivity increases and your
5 gaseous diffusivity decreases.

6 Now, how is that impacting? If we look at
7 oxidation rates, and that's been a very important
8 aspect with respect to Tc-99 looking at the overall
9 performance of that because of the difference in
10 mobility under reducing conditions where it's
11 relatively immobile, we're under oxidized conditions,
12 it's much more mobile. And what you find here is your
13 oxygen is really -- has to consider two-phase system
14 that depends on the moisture content, as I mentioned
15 earlier, and your oxygen transport via gaseous
16 diffusion can be important depending on the
17 saturation.

18 If you're in a less than saturated state
19 and you have a continuous vapor pathway, then your
20 diffusion flux of oxygen in the gaseous phase can be
21 up to five orders of magnitude greater than diffusion
22 through a liquid phase. So you've got a great
23 disparity between the two diffusive rates. If you're
24 in a continuous liquid phase and discontinuous gaseous
25 phase, then only liquid phase diffusion should be

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1 considered. But understanding the moisture content
2 and the moisture status becomes very important, as I
3 mentioned where you are in that curve.

4 In most cases from what I've seen that
5 moisture status is not well-defined or well understood
6 under the field conditions. Carbonation again, your
7 moisture impacts it and the way that that does is your
8 carbonate formation, you're basically reacting carbon
9 dioxide with the alkalinity of the matrix and forming
10 a carbonate or under extreme conditions a bicarbonate
11 precipitate. If you are under saturated conditions,
12 that tends to be on the exterior boundaries that that
13 occurs. If you're in partially saturated conditions,
14 then you get migration further in.

15 The actual rate and extent that this
16 occurs is a consequence of the alkalinity of the
17 material and your external carbonation, carbon dioxide
18 source, be it through soil gas, which can be elevated
19 over basic atmospheric gas because of biological
20 processes in the adjacent soils or also you can be
21 talking about coming from carbonate saturation in
22 groundwater such as some of the systems that you see
23 out at Hanford, for example. You have a carbonate
24 saturated groundwater and that will react and
25 precipitate with that.

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1 Now, what is the impact of the
2 carbonation? One is that it can change the
3 speciation or the absorptive sites that you have in
4 the matrix. I illustrated this here looking at a
5 cement matrix, one that is uncarbonated and the
6 arsenic liquid solid equilibrium that occurs, that
7 typical curve. And then under the carbonated
8 circumstance, you see that this was the typical curve
9 for that same material. It can be very dramatic.
10 It's ph dependent behavior. Also, you have the
11 potential for pore blocking because of precipitation
12 that occurs at the interface and it changes the ph
13 grading within the system.

14 So that also if your release is coupled to
15 ph in your system, then that's changing in a gradient
16 within the material itself. So your carbonation can
17 either have positive or detrimental impacts on the
18 long-term performance of a material. The leaching of
19 major constituents, basically, you've got to look at
20 it in terms of gradients that you have within it and
21 your local chemistry is controlled by the major
22 constituents; your alkalinity, your calcium, other
23 major constituents that are being released and that,
24 in turn, is setting up what you have in terms of your
25 pour water chemistry, oxidation fronts and ph

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1 gradients that you have within the system.

2 So when you look at this, if you don't
3 consider that, then you can lose the effectiveness of
4 some of these highly non-linear processes within the
5 material and some of the boundary layer effects. The
6 trace constituents, then follow some of the bulk
7 constituents and that local pour water equilibrium
8 chemistry. So your release would then be dependent on
9 the moisture conditions because that effects your
10 diffusivity, your continuity of your liquid base, your
11 ph gradients and your oxidation or your redox
12 chemistry within the material itself, and boundary
13 layer formation. I'm going to talk more about that in
14 a few minutes.

15 But I just wanted to illustrate the
16 complexity of the process a little bit and how over-
17 simplification can result in misleading results. If
18 you took -- this is a cement stabilized waste form, a
19 sariate (phonetic) waste form that was analogous to
20 one that's being considered out at Hanford. And in
21 turn we did monolith or tank leaching for more than a
22 year on that study. And what you see here, this green
23 line, is the projection that you would get off of
24 using ANSI 16.1, a short-term rapid study and a
25 simplified diffusivity that would come out of that or

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1 leaching index and if you projected that over for a
2 year, and then the actual data as to what the flux
3 conditions are. And what you find is after just one
4 year, you're off by two orders of magnitude. So and
5 if we're looking at projecting much longer, if you
6 don't take into account the coupled effects within the
7 matrix, and that pour water chemistry, those two
8 projections diverge considerably.

9 So ultimately, what you're look at is
10 integration of the chemical degradation and the
11 physical stress and they have integrated effects.
12 They are synergistic. Some are antagonistic that you
13 have to look at and you see the physical stresses in
14 terms of loading, drying, shrinkage, seismic effect,
15 settlement, but then also you have expansive reactions
16 which can couple with that, including carbonation,
17 sulfate attack or corrosion reactions that are
18 happening at rebar. And those expansive reactions
19 both change the local chemistry and can open up
20 cracks, micro or macro cracks, within the system.

21 So what I want to do is just illustrate a
22 few of these and wrap it up with how we look at these
23 as an integrated system. And this is just an example
24 where we were using the Tc-99, Iodine 192 and
25 secondary waste from vitrification with a prototype

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1 matrix to understand the different factors that would
2 be involved and using a reducing grout and we used
3 rhenium as the surrogate for Tc-99 and stable iodine
4 as the surrogate for iodine-129 to simplify the
5 laboratory handling. Also I want to point out, we're
6 using distilled water as one leaching extractant as is
7 typically used in ANSI 16.1 but also we're using a
8 synthetic groundwater which is a stimulant that's
9 often used at Hanford to reflect the carbonate,
10 bicarbonate effects that you see out there.

11 Now, when you look at equilibrium, what we
12 -- the way we view this is understanding that leaching
13 behavior as a function of ph, and then what we do with
14 that is back infer from looking at the whole range of
15 constituents that are present, a geochemical
16 speciation model for that system so that you can look
17 at the coupled chemistry within it that considered
18 both solid phase dissolution and saturation, solid
19 phase dissolution kinetics, absorption processes on
20 the iron, aluminum, silica type surfaces, iron
21 exchange processes and coupled with redox and the
22 various ionic strength effect. So for example,
23 looking at high ion strength that you have initially
24 in the pour water using a modified pitzer (phonetic)
25 type of approach.

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1 So what you see here, just to illustrate,
2 for strontium for example, your initial waste form is
3 about ph 11.5 to 12.5 and you'd see initial solubility
4 constraint of strontium or leaching at about 100
5 micrograms per liter but as you go towards the fully
6 carbonated waste over time, you would shift along that
7 curve and that can increase by two, three orders of
8 magnitude. Here you see it at about 10,000 micrograms
9 per liter. And you seem some, the rhenium does not
10 have that characteristic behavior. Uranium, for
11 example, has also a very typical characteristic
12 behavior, not impacted in speciation by the
13 carbonation as I'd shown earlier for the arsenic case,
14 but is impacted dramatically by the ph changes that
15 are occurring.

16 When you look at the mass transfer results
17 and these are again, in the distilled water type
18 testing, here I'm just comparing again, for various
19 species, the rhenium, the selenium, calcium and
20 strontium, the difference between the ANSI 16.1
21 projection and what you actually see over a year of
22 testing in that matrix. And again, you can see up to
23 two orders of magnitude variance over just one year of
24 testing.

25 Then when you go to synthetic groundwater,

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1 it gets interesting. What we're looking at over here,
2 and I just want to focus on selenium because it's the
3 best example to illustrate it quickly, is over time
4 you see the carbonate precipitation on the material.
5 Here you see a little bit of the white on the edges of
6 the surfaces that are occurring and we saw that
7 carbonation that precipitation beginning right around
8 the six leachate and what you see here, this is the
9 distilled water line, for ANSI 16.1. This is your
10 simulated groundwater and you see the dramatic effect
11 of the precipitation in terms of pore blockage and the
12 change in some of the chemistry at that interface.

13 The typical ANSI 16.1 type models are
14 effected and cannot capture these boundary layer
15 effects and what you see here in comparison where
16 before with distilled water we were about two orders
17 of magnitude, now we add on about another two orders
18 of magnitude of variance from that type of projection
19 that can occur. So how do you pull all this together
20 and begin integrating these different processes. One
21 thing that I want to make sure to separate in people's
22 mind is the difference between the observational
23 experiments that are used to develop the conceptual
24 model, your parametric experiments that are used to
25 parameterize models and projections and integrative

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1 experiments which are independent to do validation of
2 models, because often we see model verification but
3 not validation, even short-term validation over one to
4 10 years, given what the field conditions are allowed
5 much less none of us are going to wait around to see
6 validation over 10,000 years.

7 So then once you have at least short-term
8 validation that you've coupled the processes
9 correctly, then I think it's appropriate to look at
10 your field scenarios and your long-term performance
11 estimates, sensitivity analysis and uncertainty
12 analysis. We published an overarching framework for
13 leaching assessment which is currently under adoption
14 by EPA right now as well as the European EU DG
15 environment is concerning a framework under a number
16 of applications over regulatory development and what
17 this framework does is it looks at integrated approach
18 to leaching assessment. It says that you measure the
19 intrinsic leaching characteristics of a material and
20 then you use that in conjunction with mass transfer
21 and geochemistry models to project different field
22 scenarios.

23 So you're looking at the leaching is a
24 function of ph and liquid to solid ratio or tank
25 leaching monoliths as I showed earlier and coupling

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1 out of that what your model would be and your model
2 parameters for the various conceptual models. Then
3 what you're doing is you're evaluating the release in
4 the context of the field scenario. What are your
5 important external factors such as carbonation,
6 oxidation, mineralogical changes and hydrology and
7 then using coupled geochemical speciation, mass
8 transfer models to estimate release for alternative
9 scenarios.

10 Why this integrated approach? TCLP, for
11 example, may have regulatory standing for some of
12 these applications, has no valid technical basis for
13 application to cementitious waste forms in these
14 applications. Similarly, oversimplification and some
15 other approaches, as I pointed out, can lead to some
16 misleading results. But importantly from this
17 approach, you can take multiple scenarios from a
18 common data set and project them out. So what we do
19 is use coupled laboratory testing to get model
20 parameterization and then develop that into a
21 geochemical equilibrium model that considers the
22 various factors I mentioned earlier and then take that
23 into field scenarios and mass transfer modeling.
24 You've got this so you can look at it.

25 The way that we do it, we use a program

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1 that's been developed and in development right now
2 called Leach XS which is based on ORCHESTRA as the
3 geochemical speciation reactive transport modeling,
4 couple it with a materials data base, a scenarios data
5 base, and then regulatory criteria and ultimately that
6 models the various materials and scenarios. EPA is
7 evaluating this right now as a decision support pool,
8 looking at it going forward in some of its
9 applications.

10 Why? If you take a look, what it allows
11 us to do is to follow the speciation, the geochemical
12 speciation, as a function of ph and location and
13 conditions at each location in a matrix over time.
14 This is just an example of how you can follow the
15 chromium speciation, the aqueous phase, and see the
16 difference between the contributions of the calcium
17 chromate, for example, that's dissolved, the chromium
18 3 that's dissolved at various conditions, comparing it
19 to the experimental data, the bright red dots, but
20 also, it's a partitioning between a liquid and a solid
21 phase, where it takes into account the free dissolved
22 which is this is that whole thing in the green over
23 here, but then, for example, that which is absorbed
24 onto iron oxides.

25 This yellow fraction over here, for

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1 example, is barium sulfate chromate speciation over
2 there and over in here you see the ettringite
3 formation with the chromium. Now, if you couple that
4 with your reactive transport, then you start capturing
5 the boundary conditions. If you start looking, for
6 example, that interaction between the Hanford
7 groundwater and the matrix interface, and here I've
8 got the leachate cement material, simulating that but
9 what you see is that that interface you get brucite
10 precipitation and calcite precipitation, again
11 shutting off the pore structure and capturing those
12 changes over time that I showed experimentally.

13 And then when we couple this fully, we
14 look at all of the different species that are present
15 and can do it in a layered effect and then for
16 example, for this case, we were looking at cement MSW
17 bottom ash and soil, but you see areas where you get
18 precipitation, where you get the boundary layer
19 effects and you're tracking the full speciation at
20 each point in the matrix over time. So that that
21 allows you to capture these various long-term effects
22 that are occurring.

23 So just to wrap it up, then the suggested
24 path forward from my perspective is to process a
25 continuous improvement, trying to capture the state of

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1 the art in these assessments and both for current
2 assessments and to future nuclear waste management,
3 and to define both the short term and the long term
4 needs and to look at experimental studies, coupled
5 with model development and validation. I can't say
6 that validation being important enough. And really
7 trying to capture the formation, the effect of the
8 boundary layers. The moisture transport and status,
9 I think is critical and often overlooked.

10 The oxidation rates are closely coupled
11 with that. And to bring that together in terms of a
12 full geochemical model that couples your local
13 equilibrium mass transfer for the key systems and then
14 as you go down, trying to couple that with the
15 physical effects as well as you change from an intact
16 matrix to a spalled matrix progressively.

17 So in conclusion, there are a number of
18 these processes currently aren't included in the
19 current DOE performance assessments that can impact
20 constituent release. In some cases, I think it's
21 over-simplified being very conservative. I showed
22 several orders of magnitudes difference. In some
23 cases it may not be conservative such as if oxygen
24 diffusion is important in this moisture status or
25 these materials and the effect of technetium

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1 potentially. So it's important over time to improve
2 the understanding for both near term and long term DOE
3 waste management decisions and we've been working with
4 Savannah River National Lab as well as others, as I
5 mentioned earlier, to develop the various system
6 components and integrate this for model systems. So
7 thank you very much for your attention. I know I went
8 through this quickly but it was a lot of ground to
9 cover.

10 VICE-CHAIRMAN CROFF: Thank you. I'll
11 give you a moment to be seated and then we'll go with
12 the questions.

13 Dr. Hinze?

14 MEMBER HINZE: Well, I was very much
15 struck with Dr. Kosson's comment that over-
16 simplification can lead to misleading results. I
17 think that's the bottom line to what you've said here
18 today. The -- I was also struck with your concern
19 about short-term versus long-term validation in the
20 performance assessment and that's really where the
21 rubber is attached to the road. Can you give us your
22 expert opinion on the major sources of uncertainty in
23 the long-term validation and what are their impacts
24 and how do you know this?

25 DR. KOSSON: Right now, the major

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1 uncertainties that I've seen -- I'm sorry, does that
2 help?

3 MEMBER HINZE: That helps.

4 DR. KOSSON: All right, the major sources
5 of uncertainty that I've seen looking at these various
6 systems and how they're implemented, one is dealing
7 with the boundary layer effects, which could be very
8 important. The second is the neglect of the
9 geochemistry within the pore matrix and the boundary
10 and the gradients that actually exist which are highly
11 non-linear in these systems. A third is understanding
12 the moisture status of these systems over time, both
13 initially and over time and how that impacts vapor
14 phase processes, both in terms of carbonation,
15 oxidation. And then in terms of validation, there has
16 been very limited comparison of modeling results both
17 with data that may be in hand or obtainable in the
18 short term meaning one year to a decade type data,
19 which to me is reasonable time frames when we're
20 looking at the lifetimes that we're looking at on
21 these and the rate of movement on some of these
22 processes.

23 The long-term is extraordinarily
24 difficult. You can look at historic systems. We've
25 looked at concrete, for example, from Roman ruins and

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1 things of that sort to validate different pieces of
2 it, but I think long-term, you don't know the initial
3 conditions well, you don't know your boundary
4 conditions well. So getting back to your question,
5 those are what I think are your major uncertainties.

6 MEMBER HINZE: Hardly any at all.

7 DR. KOSSON: Hardly any at all. But I
8 tried to point out the magnitudes that some of these
9 have.

10 MEMBER HINZE: That's very important. Let
11 me ask about the vapor aspect of it and the mobility
12 through the cement. We see -- whenever we see the
13 picture of the cement we see this model and you've
14 shown how the cracking can lead to spalling and so
15 forth. How well do we know those
16 mechanical/hydrologic properties and their change and
17 how well can we model those in cement?

18 DR. KOSSON: I think some of the other
19 speakers are going to address that in more detail but
20 the micro cracking and the larger cracking has been a
21 separate area of model development in cement based
22 system reliability. The integration of those models
23 with the chemistry, the coupled effect, is very
24 limited in its implementation right now, I believe.

25 MEMBER HINZE: I was very struck with both

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1 you and Dr. Langton commenting about the coupled
2 processes which is a major factor in the whole
3 uncertainty in the performance assessment.

4 DR. KOSSON: If I can just add one more
5 comment; it's what you'll see typically in models,
6 either strength comes from the physical durability
7 side or strength comes from the leaching side.
8 Getting the coupled strength from both of those, is
9 much less prevalent.

10 MEMBER HINZE: Yeah, thank you.

11 CHAIRMAN RYAN: Dr. Langton, on your Slide
12 16, you had the diagram there for acceptable risk and
13 you talked a little bit about that. Have you ever
14 used a probabilistic risk analysis technique to try
15 and get at what the range of risks might be and you
16 know, based on what you know and don't know and how
17 that might help?

18 DR. LANGTON: We have -- yes, at Savannah
19 River we have but I was getting at a larger issue that
20 -- at Savannah River we have started using
21 probabilistic analysis. I was trying to get at an
22 even larger issue and that would be -- an example
23 would be, so exactly what is the risk of exceeding
24 four millirem at the monitoring well, hypothetical
25 monitoring well, for technetium, for groundwater that

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1 contains --

2 CHAIRMAN RYAN: Sure.

3 DR. LANGTON: What is that risk at 5,000
4 years? Is it one -- well, what is it and how do we go
5 about mitigating it. All of the modeling contains a
6 series of assumptions and scenarios that are
7 hypothetical scenarios.

8 CHAIRMAN RYAN: Well, I think that's one
9 of the values of PRA is that you can actually analyze
10 that and you can evaluate each parameter for its
11 contribution to the total system risk.

12 DR. LANGTON: That's right, but if you --
13 when we do that, some of the scenarios result in
14 failure and I don't know if those scenarios are valid
15 or -- I mean, we can always come up with scenarios
16 that result in failure.

17 CHAIRMAN RYAN: Sure.

18 DR. LANGTON: It's who's going to evaluate
19 that evaluation? I guess that's my question.

20 CHAIRMAN RYAN: Yeah, no, and I understand
21 the point that sometimes the -- a regulatory
22 requirement tends to be deterministic and it's hard to
23 match a probabilistic analysis against a deterministic
24 requirement. That's fair enough. But I think it's
25 hard to gain insight into what risks are and what

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1 uncertainties are without doing some kind of a more
2 rigorous analysis, say a one off, a boundary case.

3 DR. LANGTON: No, we agree with that --

4 CHAIRMAN RYAN: Okay.

5 DR. LANGTON: -- and we're doing that type
6 of analysis but the question is even -- is bigger than
7 that. Again, we don't take any sort of remediation
8 strategy into account to mitigate any risk. We just -
9 - just looking at risks, not just, we are identifying
10 risks and we're making improvements but that doesn't
11 mean we can't even improve our scenario that resulted
12 in negative risks.

13 CHAIRMAN RYAN: There's lots of good work
14 to do. Sure. No, I appreciate that point. Turning
15 to the second presentation, Dr. Kosson, your Slide 16,
16 I took away from -- 16 and 17 actually, it's your
17 curve showing mass transport tests and synthetic
18 groundwater tests, I took away from that, that your
19 prediction, your green line, which is a prediction of
20 the model, maybe we could throw that up on the screen,
21 is that possible? 16 or 17 either one. Thank you,
22 Michelle.

23 All the data diverged from the model in
24 every case. Wait a second, let's get it up there.
25 No, that's not it. There it is. Slide 16, please.

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1 There we go. And as you pointed out, you know, it
2 rapidly becomes orders of magnitude of deviation, in
3 this case in probably the positive direction. It's
4 less getting out than the model predicts but I would
5 say a model that has that pattern of divergence isn't
6 conservative. It's just flat out wrong. It doesn't
7 match the data. So help me understand these results
8 in the context of uncertainty analysis.

9 DR. KOSSON: Sure. What I was trying to
10 do was the green line is the model that is typically
11 applied in these cases, the ANSI 16.1 projection of
12 constant diffusivity.

13 CHAIRMAN RYAN: Right.

14 DR. KOSSON: So I wanted to compare what -
15 - even over a short term, over one year, what the
16 typically assumed model that's implemented would
17 project in comparison to what actual data is. If you
18 looked at the full coupled geochemistry mass transfer
19 model which I didn't have time to really get into in
20 detail here, just gave you an overview, that would
21 track with the -- that's a very different type of
22 modeling approach.

23 CHAIRMAN RYAN: Gotcha.

24 DR. KOSSON: My goal in putting up that
25 slide and the next is to show some of the deviations

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1 between -- that can be caused by over-simplification
2 some of which is occurring now. That's your green
3 line which you typically see and what reality is or at
4 least on one year, you know, short term experiment --

5 CHAIRMAN RYAN: And I --

6 DR. KOSSON: -- which is longer than
7 usually carried out.

8 CHAIRMAN RYAN: I take that as a real
9 important caution that, you know, unless you have some
10 fairly detailed understanding of your uncertainties
11 and their behaviors, for lots of reasons,
12 phenomonologic reasons, chemical, physical, scale, you
13 know, all those reasons, you really don't know -- you
14 really have the risk of not knowing whether you're
15 diverting from what reality is when you model and
16 that's a great caution to carry away.

17 And I guess maybe we'll get into it later
18 on through the day is to think a little bit more and
19 talk a little bit more, and I'd be curious to hear all
20 of your views on how do you get at system risk or
21 system uncertainty. That's a big question, I know.

22 DR. KOSSON: Do you want me to comment?

23 CHAIRMAN RYAN: If you want to take a
24 crack at it now, have at it.

25 DR. KOSSON: Okay, just a little bit, I

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1 think you start off to get to system risk and you're
2 starting point is the best conceptual models that you
3 can have for the system and then look at the
4 individual uncertainties and lump them together trying
5 to get an overall system uncertainty.

6 CHAIRMAN RYAN: Step-by-step.

7 DR. KOSSON: Step-by-step.

8 CHAIRMAN RYAN: Gotcha. Okay, thanks.
9 It's great, appreciate it.

10 MEMBER WEINER: I'd like to thank both of
11 you for something that -- for very interesting and
12 revealing presentations. I'd like to ask Dr. Kosson,
13 since you get transport through the concrete,
14 transport of solutions basically, can you use that to
15 bring about chemical reactions which will stabilize
16 your waste? In other words, can you -- can you make
17 a precipitation reaction happen in the grout itself?

18 DR. KOSSON: I believe that by
19 understanding the chemistry in some of these reactions
20 that occur, you can design and control your systems to
21 take those into account and to -- without going too
22 far in the way I want to comment, but I think what
23 you're suggesting, Dr. Weiner, is indeed possible but
24 if you're basing your evaluation on models or
25 evaluations that don't even capture these effects,

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1 then there is no incentive to design in that way
2 because you can't take credit for it.

3 MEMBER WEINER: Well, shouldn't we be
4 looking at models that do -- I'll tell you what
5 prompted the question is that many years ago in a
6 study of how do dispose of nerve gas at an arsenal in
7 Denver -- outside Denver, Rocky Mountain Arsenal, the
8 suggestion was made and it was quite a good one, drop
9 it in the ocean because the salt water infiltrates and
10 hydrolyzes the stuff and make a relatively benign
11 product. Certainly, you get rid of the toxicity. So
12 I was just thinking, couldn't you use the opposite
13 effect and shouldn't that be incorporated into some
14 models?

15 DR. KOSSON: I think so, yes.

16 MEMBER WEINER: Okay.

17 DR. LANGTON: At Savannah, we're looking
18 at landfill amendments. The schematics that we're
19 shown are simple but there are other features that
20 could be added to make improvements and then the
21 follow-up on that would be that there are amendments
22 that could be made post-closure, post-completion of
23 the landfill which would fall into the remediation
24 category and yes, that's out there as something that's
25 available and uses the same chemistry and known

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1 engineering practice but is not to the point of being
2 included in any modeling or risk assessment at this
3 time.

4 MEMBER WEINER: Which brings me to the
5 question that I have for you, Dr. Langton. And I
6 suppose you've partly answered it. What is preventing
7 you from including remediative factors? Is it
8 regulation, is it approved modeling, why can't you go
9 there?

10 DR. LANGTON: It's at least a perception
11 that we will -- we need to design today and construct
12 today so that when the landfill is closed or 100 years
13 after it's closed at some period into the future, a
14 short time into the future, we can walk away from it.
15 It doesn't require any maintenance and it's a big
16 problem in durability assessment because one way of
17 defining durability is performance of design function
18 for design lifetime with a certain amount of
19 maintenance, however much maintenance is decided upon
20 up front and we, for some reason, historically, don't
21 know for some reason, have the idea that that's zero
22 maintenance.

23 And 10,000 years is a long time to design
24 for and to address risk over that time period and
25 eliminate maintenance at the same time.

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1 MEMBER WEINER: I think you've raised a
2 really interesting point to look at in designing
3 regulation, both of you have. I just had one more
4 question for Dr. Kosson. And that is that I'm a
5 little surprised that with uranium the ph effects
6 overshadow the carbonate effects. Is that what you've
7 observed? Have you done this with other actinides?

8 DR. KOSSON: The results that I showed for
9 this matrix was the first matrix that we did uranium
10 evaluation on. When you look at the geochemical
11 modeling of this system, that's not surprising for the
12 system, but we have not scored it more broadly for
13 other waste systems.

14 MEMBER WEINER: I see, thank you.

15 MEMBER CLARKE: I have a few questions and
16 I think they may be a little premature, because I know
17 we have several more talks. Allen has planned a full
18 day of really interesting stuff, but let me just kind
19 of give you a heads up of some of the things I'm
20 interested in. One of them was that until Ruth asked
21 her question, I had yet to hear the term maintenance
22 from either of you. And I would suggest that at least
23 in an ideal world, free from other constraints, that
24 it would go containment, intervention, and remediation
25 would be the last resort.

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1 So in other words, if we want the systems
2 to perform for a long time, I can't see anybody doing
3 that without intervening at times and maintaining them
4 and that suggests to me that we should design them to
5 facilitate that. Now, I know that goes against the
6 school of thought that says we don't want to burden
7 future generations with things that we've done and we
8 want to design these facilities so that we can
9 implement them and they'll take care of themselves.
10 So I'd just throw that out as kind of food for
11 thought.

12 And the other thing I didn't hear much
13 about was monitoring. And Dr. Kosson raised the
14 importance of moisture content. That is being looked
15 at as something that can be monitored in landfill
16 covers and there is a prototype systems, there's a
17 prototype at Hanford and I think there's a full-scale
18 system at Fernald, where moisture content is being
19 monitored in the landfill cover. Is that something
20 that could be done in these vault systems, in these
21 concrete containment structures? Like I said, I just
22 throw that out for both of you.

23 DR. LANGTON: I don't see why not.

24 VICE-CHAIRMAN CROFF: Microphone.

25 DR. LANGTON: Yes, it certainly could be

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1 done. We are not doing -- we are not monitoring our
2 concrete vault moisture characteristics currently or
3 the saltstone, but I don't think it's impossible. I
4 think it's doable. For the longer time periods that
5 we're interested in, which a long time period for
6 field monitoring might just even be a year, longer
7 would be better but certainly programs that need to be
8 developed and implemented.

9 MEMBER CLARKE: That is certainly the
10 case, I'm sorry, David. Certainly the case from what
11 we've seen at landfills that if they're going to fail
12 the high probability of failure is assumed because
13 they weren't constructed properly, they weren't
14 designed properly.

15 DR. KOSSON: I just wanted to comment,
16 Jim, that it's also important to keep in mind that
17 it's easy to project these as steady state conditions
18 or constant conditions, but we've got to recognize
19 that moisture as well as infiltration and other -- a
20 host of other conditions area periodic or intermittent
21 type of effects in response to climate cycles, you
22 know, winter/summer type effects but also
23 precipitation events, for example, impact the moisture
24 content.

25 MEMBER CLARKE: Episodic.

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1 DR. KOSSON: These episodic events are
2 very important, I think to be captured because your
3 outcome from an episodic event modeling can be very
4 different than what you get from continuance average
5 event.

6 CHAIRMAN RYAN: Can any other panel
7 members address Jim's question?

8 VICE-CHAIRMAN CROFF: Barry?

9 DR. SCHEETZ: Your concern about
10 monitoring; through FHWA there are programs going on
11 through our organization at Penn State right now,
12 where we are instrumenting bridge decks with a 25-year
13 monitoring plan where the embedding instrumentation
14 has a -- is connected to a cell phone which calls home
15 every day or every week or every month and then
16 automatically downloads. So these autogenous sensors
17 and the ones that you referenced out at Hanford are
18 available for pressure, for tilt, but they're just now
19 coming on line for moisture. I don't know of any
20 decent moisture sensor, particularly one that you're
21 going to be able to embed long-term and leave it there
22 for 20 years or more. Rachel may have -- it looks
23 like she has some ideas there.

24 The specific conductants that the folks at
25 Hanford are using are very good. They're very

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1 durable, but they're -- you know, they're not going to
2 function in the long term that we're looking at there.
3 These so-called intelligent aggregates is something
4 that needs to -- you know, that's a very specific need
5 that is out there to be developed that you can put
6 into concrete and just applications here are one
7 application but they're in -- in the civil
8 infrastructure they'd be invaluable.

9 MEMBER CLARKE: Thank you. How are we
10 doing on time, Allen? Can I ask one more question?

11 DR. DETWILER: I would like to add
12 something on monitoring, just as a general conceptual
13 look at it. I think that over the long term, Dr.
14 Scheetz is right, you're not going to be able to use
15 instruments over an extended period. For example, if
16 you embed something in the concrete, you can expect
17 that after a certain time, whether the battery runs
18 out or the leads corrode or whatever it is that makes
19 it impossible for it to continue to work and it may be
20 something as simple as electronic computational
21 methods that we use just become obsolete and so they
22 can't talk to each other any more.

23 And so I think if you were really -- if
24 you really wanted to look at long-term modeling as in
25 decades and centuries, you're not talking about active

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1 systems but more like passive systems where, for
2 example, you have access ports that you can put
3 different kinds of probes in when you want to or you
4 put benchmarks that you can sight on and then you use
5 whatever instruments you have that -- you know, in
6 that year or that century but you have something
7 permanent that you can sight on so that that's always
8 the same. And your records, then, may be something
9 very different from electronic records or something
10 that you use in addition to electronic records, like
11 pencil and paper because you can keep those longer or
12 you have -- you can still read them 10 years, 20
13 years, 50 years later even though it may have faded or
14 started to deteriorate. At least it can be read much
15 longer than say a floppy disk.

16 So you really would have to consider some
17 of those long-term things as well and then have some
18 overlap when you're switching from one measurement to
19 another so that you at least are satisfied that you're
20 still getting the same thing.

21 MEMBER CLARKE: Those are good comments.
22 I appreciate them. I think the approach that is being
23 taken in the way of recovery is just to put in access
24 ports and incorporate that into the design. As I
25 recall, I think in another site they actually

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1 retrofitted the moisture content measurements. You
2 know and it strikes me that even if we couldn't do
3 this for, you know, short-term versus long-term, what
4 does that mean? You know, if we did it for as long
5 as we could, it would be valuable information on some
6 of these systems as we begin to implement them and
7 look at their performance. So my concern is that
8 everything that's being done up front is good stuff
9 and it's good science, but if we don't get it right up
10 front, then where do we go from there and I'd hate to
11 go right to remediation. So I'm raising questions
12 about monitoring, designing to monitor, which I don't
13 think we do, and designing to maintain, which I don't
14 think we do either.

15 So those are my comments. Thank you.

16 VICE-CHAIRMAN CROFF: I've got a couple of
17 questions here. I think the first one I may struggle
18 to articulate but I'd like to address this to Dr.
19 Langton. In one of your early slides you had a list
20 of applications of cements, where they have been used.
21 It focused on Savannah River but I know you're
22 familiar with what's going on in a lot of other
23 places. What wasn't so clear to me is in how many of
24 those applications did the Department of Energy or
25 whomever try to take credit for more than just filling

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1 up a void space, try to take credit for the hydrologic
2 barrier or the chemical barrier or something like
3 this.

4 DR. LANGTON: I don't think we have tried
5 to take credit for hydrologic barrier for waste forms.
6 We've taken credit for chemical stabilization of
7 specific radio-nuclides and by taking credit it's
8 reduced the leaching in our low level waste disposal
9 facility. Physical integrity, we've taken credit for
10 waste forms maintaining their integrity and that would
11 be drummed waste forms that went into this low-level
12 waste disposal facility.

13 Taking credit for hydraulic conductivity
14 is typically the function of the concrete vault, the
15 barrier in our disposal facility, not the saltstone
16 facility but in our low level waste disposal facility.
17 That -- the concrete structure is intended to reduce
18 infiltration.

19 VICE-CHAIRMAN CROFF: And the performance
20 assessments reflect some credit for this?

21 DR. LANGTON: Yes, the -- it's the E area
22 performance assessment takes credit for that, yes.

23 VICE-CHAIRMAN CROFF: Okay. So --

24 DR. LANGTON: There are two places, E area
25 and Z area performance assessments for the concrete

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1 vault -- we have several different types of concrete
2 vaults. We also have a disposal system referred to as
3 containment ingrouts where we're poured grout around
4 a large object that's disposed of, so that is an
5 infiltration barrier also. But there are waste forms
6 that we've made that are drummed where that's not
7 taken into account. We solidify the liquid, stabilize
8 contaminant, reduce the surface area but not so much -
9 - it's not related to hydraulic conductivity, I don't
10 believe.

11 VICE-CHAIRMAN CROFF: Okay, and in those
12 cases it doesn't really factor into a performance
13 assessment.

14 DR. LANGTON: Right, right. Small cement
15 waste forms, small volumes that go into our disposal
16 facility aren't included. It's only the large
17 concrete vaults or the large volume waste form.

18 VICE-CHAIRMAN CROFF: Okay.

19 DR. LANGTON: One's a monofill of --
20 saltstone is a monofill essentially where one -- a
21 large volume of a waste stream is solidified in a
22 cementitious matrix but we do have small volumes, a
23 few drums here and there over time.

24 VICE-CHAIRMAN CROFF: Okay, so is it fair
25 to say that in much of this and in trying to take

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1 credit for the effects or the beneficial effects of
2 cements beyond just, you know, sort of filling a void
3 space or something, that we've started down that path,
4 we've done some of it but it's a relatively new
5 experience for us in how to do it and how much?

6 DR. LANGTON: Well, cementitious waste
7 forms are best developed available technology for
8 hazardous constituents. So EPA has a much broader
9 application of cementitious waste forms for non-
10 radioactive waste.

11 VICE-CHAIRMAN CROFF: I know they use them
12 a lot on that side of the house, if I can call it
13 that, but I didn't think they did much performance
14 assessment. They sort of say use it and hope it works
15 or --

16 DR. LANGTON: No, it's use it and it needs
17 to go, depending on its characteristics, whether it's
18 characteristic or listed waste, whatever the details
19 are of its waste classification, it goes to a
20 landfill, whatever kind of landfill, it goes to that
21 landfill. It's monitored. It's got leachate
22 collection. There's a plan. The plan doesn't extend
23 that far into the future. I think it's 50 years, is
24 that right, Les, or Subtitle C landfill but at the end
25 of 50 years, it doesn't say it's closed. We haven't

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1 gotten to 50 years for any of these landfills yet.

2 So there is a leachate collection and a
3 groundwater monitoring plan and it is regulated.

4 VICE-CHAIRMAN CROFF: Okay.

5 DR. LANGTON: Thirty years, yeah.

6 VICE-CHAIRMAN CROFF: Okay, Dr. Kosson, a
7 very different question; if -- I sort of sweep across
8 all of what you said and you know, let me suggest a
9 broad generalization, it's sort of a true/false thing.
10 It sounded like if the cement waste form was saturated
11 with water, and it was carbonated, the carbon dioxide
12 had reacted to sort of seal up the surface and this
13 kind of thing as you described, it sounds like that
14 would be a good thing, a desirable thing to have.

15 Are there any down-sides to this? In
16 other words, at some point does carbonation begin
17 degrading the waste form or the water or something
18 like this?

19 DR. KOSSON: I think there are a number of
20 different processes that you have to balance across
21 that and let me give you a couple of examples that you
22 just mentioned and why. Ideally, you would keep the
23 matrix saturated to avoid vapor phased diffusion and
24 to limit it, but at the same time, when you're
25 saturated, you have to be concerned about not

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1 maximizing some of your liquid phased diffusion and if
2 you're past saturation, suddenly you've got capillary
3 -- I mean convective movement as well.

4 So there are careful balancing acts for
5 the design conditions that you want there. Similarly,
6 carbonation as a capping, a pore blocking phenomena
7 can be very beneficial. Also the ph effects of it in
8 some circumstances can be very beneficial the way it
9 effects the leaching behavior of some constituents.
10 However, for some constituents, as I pointed out
11 earlier, it also can have detrimental effects so
12 because of the ph geochemistry effects. So what
13 you're really looking for is looking for your best
14 operational window that you have of these combined
15 processes and then designing to maintain that
16 operational window.

17 VICE-CHAIRMAN CROFF: Okay, I was --

18 DR. DETWILER: May I add something here?
19 Carbonation also causes shrinkage so that could result
20 in cracking.

21 VICE-CHAIRMAN CROFF: Okay, I was struck,
22 maybe generalizing off this specific point by
23 something Dr. Langton said and that is that there is
24 no conservative case for all parameters. It's, you
25 know, your phrase is windows. It's something is good

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1 on one hand, bad on the other and that makes
2 performance assessment very interesting kind of an
3 exercise.

4 DR. LANGTON: Especially for 10,000 years
5 work.

6 VICE-CHAIRMAN CROFF: Yes, at this point,
7 I'd like to offer the other panel members. Do you
8 have any questions of our two first speakers? All
9 right.

10 DR. DETWILER: For Dr. Kosson, I saw you
11 had a couple of slides in here at page 7 and also page
12 13, where you are showing cracking and the development
13 of cracks and I was wondering how you put that into
14 your model, because for the most part, that would be
15 something that's not going to be continuously growing
16 but either -- it's caused by something and that
17 something is probably fairly short term. How are you
18 incorporating this into your model, if you are?

19 DR. KOSSON: Well, I've got to be candid
20 that that's a very high challenge in these kinds of
21 modeling. The way that we're dealing with it right
22 now is that micro-cracks are dealt with as they form
23 from the structural perspective as localized effects
24 on diffusivity because they're not through cracks.
25 Larger cracks, the rate and formation of them, we have

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1 not been able to capture well, but what we've done is
2 assume that they've formed as a matrix within the
3 matrix and then basically what you do is you have a
4 dual model where you have diffusion to the boundary of
5 the macro-crack and then transport through the macro-
6 crack.

7 MEMBER HINZE: May I ask a follow-up
8 question to that? In terms of this monolithic cement,
9 is the separate pours, are there at the interface
10 between pours that may be separated in time by some --
11 by days or even longer, do we see a lot of micro-crack
12 there? Is there infiltration along those pours?

13 DR. KOSSON: I would suggest -- we've done
14 some observations and I would suggest some of the
15 other folks here have done much more extensive work on
16 the crack evolution and they'll probably talk about
17 that later.

18 DR. LANGTON: I didn't quite understand
19 your question.

20 MEMBER HINZE: Well, if you pour concrete
21 in and you don't immediately pour in further concrete
22 but then at a later time have another pour, what is
23 the interface like at the position between the pours?

24 DR. LANGTON: There is an interfacial
25 region, interfacial zone layer. It's more than a

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1 layer, it's got dimensions. What it looks like
2 depends on the details of the situation. If drying
3 takes place between pours, there's drying, shrinkage,
4 cracking. If drying doesn't take place, in some cases
5 there might be condensation of water, in our case we
6 have vaults that have roofs and moisture will condense
7 on the roof and fall back into the -- onto the
8 surface. So if we have a day between placements, we
9 have a wet surface.

10 So there's always an interfacial region.
11 It could be a high water region. It could be a very
12 dry region, with cracks. It depends.

13 MEMBER HINZE: Yeah, how do you handle
14 that in the performance assessment?

15 DR. LANGTON: Right now, that's a de --
16 that's in the noise. That's a detail that's not
17 handled. The performance assessments are built on so
18 many assumptions that when we ask to have the
19 assumptions listed, people just get weak kneed, there
20 are so many assumptions.

21 VICE-CHAIRMAN CROFF: Peace. Okay,
22 anybody from NRC staff have a question?

23 MR. FLACK: Yeah, John Flack from ACRS.
24 Getting back to acceptable risk in the long-term, and
25 I'm actually coming from reactors where they have

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1 safety goals and they talk about .1 percent of other
2 risks that the individual is exposed to around nuclear
3 facilities. Have you thought of, in the long-term
4 that type of risk as exposure to individuals, certain
5 frac percent of all the environmental risks that one
6 would be exposed to at that time in the future and
7 using that as some goal, then working backwards to see
8 how one achieves that goal through this short-term
9 monitoring that has extended into the long-term
10 effect, that sort of thing, that sort of thinking?

11 DR. LANGTON: I don't specialize in the
12 details of the performance assessments but I haven't
13 seen anything that addresses what you're talking
14 about.

15 MR. ESH: This is Dave Esh with the NRC
16 staff. I have a question that might be for the two of
17 you or it might be for the whole panel; but one of the
18 items that was addressed was the use of standard
19 method for particular materials or assessing the
20 materials, I guess. In the working group's opinion,
21 are the standard methods suitable for the very longer
22 term performance that we're getting at or are they
23 more focused on shorter term performance, common in
24 low level waste applications where you're looking at
25 a cesium, strontium dominated source where hazard is

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1 on the order of a few hundred years instead of longer
2 term? So that's kind of my question is, are the
3 methods suitable or is there a need for maybe some
4 longer term methods or are there longer term methods
5 to get at it?

6 I'm kind of thinking that some of the
7 processes that occur, the standard methods you have
8 now, they might be focused on the processes that the
9 best response on the time scale that we're looking at
10 in the laboratory but are there long term mechanisms,
11 processes, that aren't really amenable to the short-
12 term test that you might need this set of longer term
13 procedures or tests to look at those long-term
14 mechanisms?

15 DR. KOSSON: I think what we're seeing is
16 standard methods in a number of cases from the
17 leaching assessment, from things I talked about at
18 least, were developed for other applications or for
19 very short-term type of phenomena or over-simplified
20 assumptions and are very inadequate for projecting a
21 long-term behavior. Don't capture a lot of phenomena,
22 don't capture a lot of processes.

23 For example, as I mentioned earlier, TCLP
24 has regulatory standing, has no technical basis for
25 its application here even though I know that, for

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1 example, Dr. Langton listed is as one of their
2 evaluations because it's a regulatory standard.
3 There's no basis for projecting off of that.

4 MR. ESH: Then a second question I had is
5 related to scale. In the working group's opinion how
6 much validity is there to testing laboratory samples
7 on this very small scale when your application might
8 be a much larger system that brings in all sorts of
9 discrete engineered features and different conditions
10 for those large scale samples compared to the
11 laboratory samples? Is -- are you aware of any work
12 that has been done to try to take results from the
13 small sample -- small samples and show that they've
14 accurately or at least over-estimated the
15 deterioration compared to the larger scale, because I
16 think that maybe the larger scale has some features
17 that can be detrimental to performance that of course
18 aren't represented in the laboratory analysis.

19 DR. LANGTON: We recognize the same issue.
20 To take it out of the waste form business and into
21 ordinary concrete construction, there are applications
22 where concrete hydraulic conductivity is important.
23 It's measured underwater concreting like the Chunnel
24 or underwater tunnels. The material property is
25 measured and it's something that the people that make

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1 the concrete, the suppliers of the concrete have
2 specifications on or if they don't have
3 specifications, they have a correlation between some
4 other property and the permeability. So it gets to --
5 specifying a job and constructing the job. Material
6 properties are important.

7 Performance of the final product is a
8 separate issue and the two are related but one doesn't
9 necessarily predict the other except in extreme cases.

10 So we do need some sort of evaluation, I think, of
11 the structure.

12 MEMBER CLARKE: If I could interject here,
13 David has articulated very well, you know, of why 25
14 years of my in-system monitoring would be valuable,
15 because now we have a large full-scale system. We've
16 done our best in the front end to project long-term
17 performance. If we can link the front end to what
18 we're finding, what we're actually measuring, we can
19 make that loop, design to get monitoring data that can
20 help us build model confidence, which is the title of
21 a working group meeting we're going to have in
22 September, just throwing out a plug.

23 Then, you know, we can improve the models
24 as we go. I mean, we don't have to do this everywhere
25 but maybe we'll learn a lot if we do this, and that's

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1 the reason I keep raising monitoring. And the other
2 reason is that I guess I'm not a believer. You know,
3 I guess I'm just concerned that anything is going to
4 last 10,000 years without intervention and if we have
5 to intervene, then we ought to plan on intervening
6 now.

7 DR. SCHEETZ: I think to address Dave's
8 question, it's a matter of the crinkly green
9 lubricant. If we look at the performance of large
10 scale engineering structures in the civil engineering
11 realm, you know, we've placed -- we have an investment
12 of what, 1.7 or \$1.8 trillion in placed concrete
13 within the United States in the civil infrastructure
14 realm. We looked at big structures. We built the
15 Grand Cooley Dam. We've build enormous structures.
16 So the engineering community has had to look at, they
17 have been forced to look at, what you do in the short-
18 term ASTM or ACI test protocol to place the concrete
19 and then what the long-term performance is.

20 We have this so-called analogue, if you
21 want, out there. What we don't have is the same thing
22 in the nuclear community. No one has ever -- I mean,
23 Chris is the only one that's ever placed a tank full,
24 a 1.3 million gallon tankful to close a hazard -- or
25 one of these storage tanks. We don't have that

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1 experience and the driver isn't there, hasn't been
2 there from and economic standpoint to develop those
3 understandings on these rather unique and unusual
4 circumstances. But I think, you know, to answer
5 Dave's question, some of the knowledge is there but
6 we're going to have to pull it from the civil
7 concrete.

8 MEMBER WEINER: Could I ask a final
9 question? It's on another topic, so I don't want to
10 interrupt the discussion but uranium is mined as we
11 just learned, by leaching with an oxygenated carbonate
12 solution. It seems to me that this is an intruder
13 scenario that you might consider with grout because
14 you get cracks and you have carbonate and you have
15 water and here is a possible intruder.

16 VICE-CHAIRMAN CROFF: Okay, I think -- I'm
17 sorry, is there a comment?

18 DR. DOYLE: I think I'll address that in
19 a later -- I have a slide that addresses that because
20 that's exactly the scenario that lay in grouts from
21 Yucca Mountain. They failed to look at the aluminum
22 silicate chemistry associated with that system and
23 just isolated the carbonate and the ph. And you get
24 a different answer when you do a little more complex
25 geochemical model.

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1 MS. RIDGE: This is Christian Ridge from
2 the NRC and if I could follow up on something that
3 Chris talked about a little earlier was the interface
4 between various grout pours and the various ways that
5 that interface can occur, a dry surface, a wet
6 surface. I was wondering if work was being done with
7 respect to the interface between the grout and the
8 waste itself in a sort of DOE specific case of closing
9 a tank if you have not just various grout pour
10 interfaces to look at but also what to my mind seems
11 like would be a more radical interface between
12 cementitious material and the sludge itself, you know,
13 of metal oxide precipitate and whether or not you
14 think that that interface would be, perhaps more
15 important to look at.

16 To me it seems like there would be for
17 different physical properties of the two, which would
18 facilitate cracking and separation of the materials,
19 and I was wondering if there was work ongoing or if,
20 in your opinion and the rest of the panel's opinion,
21 if maybe I'm not understanding and that interface is
22 not as much of a problem as it seems like it might be.

23 DR. LANGTON: Actually, I proposed doing
24 work on actual tank residual material and cement and
25 grout in a layered effect to get to look at the

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1 interface and a column effect. But that was -- that's
2 primarily concerned with the chemistry of the system.
3 As far as cracking is concerned, there are reasons for
4 cracking.

5 Cracking is dimensional change, either
6 from expansion or contraction -- well, not contraction
7 but from expansion. So dimensional changes and I
8 don't really see dimensional changes in the sludge as
9 it exists now, which is soft material or not
10 completely rigid material. It's solid but it's not
11 bound together. It's a rigid mass, a particulate
12 material. There's plenty of room for expansion in
13 that material.

14 Now, whether it changes with time as a
15 result of leachate chemistry or interaction diffusion
16 of materials in a cement pour solution to form a rigid
17 material that then cracks, that then could crack as a
18 result of expansion, I don't know, but we have
19 proposed work from a chemistry standpoint. There's
20 talk of cleaning the tanks, removing more of the
21 residual material from the tanks which would negate
22 that need to do the work. So that's where we're at
23 right now, but if we do leave material in the tanks,
24 I think we would certainly be looking at the chemistry
25 and the chemistry, in turn, impacts the dimension or

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1 creates dimensional changes or could create
2 dimensional changes.

3 MS. RIDGE: Right.

4 DR. LANGTON: You would get that also.

5 MS. RIDGE: And then, I suppose, the
6 temperature you'd expect after the curing would be
7 fairly constant. I was thinking perhaps that with
8 respect to any thermal gradient, the sludge would
9 react very differently than the cement and you could
10 get them -- a separation in that interface.

11 DR. LANGTON: We did thermal analysis for
12 the next two tanks to be closed and there's such a
13 small temperature rise from radioactivity in the
14 waste. Now the temperature rise from the grout is
15 something that we can -- if we find it's a problem or
16 if we identify it as a problem, we can control but
17 again, it's moving across the surface. It's
18 expanding, contracting, whatever the grout is doing,
19 it's happening on a surface that is not rigid.

20 MS. RIDGE: Right, thank you.

21 DR. LANGTON: We're not anticipating a big
22 effect there in grout temperature.

23 MS. RIDGE: Right.

24 VICE-CHAIRMAN CROFF: I think with that,
25 we're slightly over, not a problem. Let's take a

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1 break till about 10:35. I'd like to thank our two
2 speakers this morning, look forward to your
3 participation in the rest of the show. 10:35, please.

4 (A brief recess was taken at 10:23 a.m.)

5 CHAIRMAN RYAN: On the record. If we
6 could come back to order please.

7 (Discussion off microphone.)

8 VICE-CHAIRMAN CROFF: Let's go ahead and
9 proceed here with our second session and our first
10 speaker here is Dr. Rachel Detwiler. Dr. Detwiler is
11 a Senior Engineer at Braun Intertec Corporation in
12 Minneapolis. Her areas of expertise involve the
13 durability, transport properties, microstructure and
14 test methods of concrete and cement based materials.
15 She also previously served in an advisory role for the
16 initial development of the grout formulation for the
17 stabilization of radioactive and hazardous waste in
18 storage tanks in Savannah River. Take it away.

19 DR. DETWILER: Thank you, Allen. I'll
20 stand up too, so I don't have to be tied to one place.
21 I have several areas that I'm going to try to cover in
22 this talk. First of all, I just wanted to go quickly
23 over the difference between grout and concrete because
24 sometimes people who are not in this field, they get
25 confused about which is which and it's nice to get it

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1 defined a little bit first. Then I'd like to talk
2 more about getting concrete to do what we want and
3 I'll use the term "concrete" throughout. To an
4 extent, you can put grout in the same category but
5 just for simplicity, I'll refer to concrete primarily.
6 Then I have a few ideas at the end about monitoring.
7 So the bulk of the talk actually is this central
8 portion right here.

9 To distinguish between grout and concrete,
10 first of all, grout is a mixture of cementitious
11 materials with water. It may also contain fine
12 aggregate which the rest of you know as sand and fresh
13 grout would have a pourable consistency. So when, for
14 example, you're doing that you're going to be able to
15 pump it very easily and place it in a form and it's
16 probably going to be more or less self-consolidating.

17 Concrete is similar, but it also contains
18 coarse aggregates. That would be larger pieces of
19 either gravel or crushed rock and you would probably
20 use this in larger applications. So grout would tend
21 to be for smaller things just because usually in
22 normal construction we use the coarse aggregates
23 because they are the cheaper ingredient. But it's
24 also something to consider when you're doing
25 applications like this where the cost of the materials

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1 is probably not the biggest issue. But the coarse
2 aggregates also provide you with a lot of dimensional
3 stability. So if you're concerned about the overall
4 shrinkage of the material for example, you're going to
5 get less overall shrinkage if you have concrete
6 because you have this big component in here that is
7 not going to shrink but instead will restrain the
8 shrinkage.

9 As I mentioned, concrete is generally more
10 economical and more stable dimensionally than grout
11 and so probably on large pours that's going to be what
12 you would rather use, although not necessarily.
13 Generally speaking because the grout does not have the
14 coarse aggregate in it, it has much more cementitious
15 paste, it's going to have a greater tendency to crack
16 and that can have a very significant effect on such
17 things as the transport properties. It may not be
18 that big a deal in terms of some of the structural
19 characteristics because of the kinds of configurations
20 that we'll be using it for. Either grout or concrete
21 could be made as a waste form if that were desired.

22 Now how do we get concrete to do what we
23 want? First of all, I would like to go over some hard
24 facts about concrete. One is it almost always cracks
25 and so when you're modeling transport characteristics,

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1 for example, you need to take that into account or you
2 need to figure out what you're going to do to make it
3 not crack or to limit the size of cracking which is
4 probably more to the point.

5 It is subject to deterioration and that's
6 what I'm going to go over. I call this talk, it has
7 to do with durability. That's because it's durable
8 against something. It's resisting deterioration
9 hopefully.

10 Most of what we know about concrete comes
11 from the construction industry. There are researchers
12 who have done a lot of work in the area of specific
13 applications for nuclear materials but they are far
14 fewer than those who have done work in the
15 construction industry and we in the construction
16 industry have a great deal of advantage in that we
17 actually build stuff.

18 VICE-CHAIRMAN CROFF: Rachel, we just had
19 a problem here. Your slide vanished.

20 DR. DETWILER: You're on my screen. I
21 don't know what your problem is.

22 (Laughter.)

23 DR. DETWILER: Okay. There it goes.

24 VICE-CHAIRMAN CROFF: There it is.

25 DR. DETWILER: Most of what we know about

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1 concrete comes from the construction industry. So
2 that's sometimes good that you have people like me who
3 mostly work in construction and occasionally dabble in
4 nuclear applications that can sort of bridge the gap,
5 but mostly people like Dr. Langton can review the
6 literature and say, "This is what they're doing in
7 construction and this may apply to us in the following
8 ways."

9 But there are certain disadvantages about
10 that. First of all, we are dealing with a very
11 different context in the construction industry. For
12 us, a long time is 50 years. Now we're starting to
13 look at hundred year lives of structures, design
14 lives. And nobody know whether we can actually make
15 that. Yes, we have Roman concrete and some people
16 even claim that the Egyptian pyramids were made of
17 concrete which is kind of silly but there you go.

18 But the problem with that is that those
19 things were made of a very different kind of material
20 than we are using today. So in fact, we don't really
21 have that long a track record with modern materials
22 and modern concrete technology. In any case, when we
23 talk about these 50 year lives or 100 year lives and
24 actually we don't know if we can have a hundred year
25 life, we're just talking about it at this point, we're

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1 specifying it, but we haven't actually seen that it
2 lasts that long, we are considering that we are going
3 to be doing regular maintenance. So that is very
4 different.

5 This question has already been brought up,
6 but we in construction assume we are going to be doing
7 regular maintenance and what people are assuming in
8 the nuclear area is that we're not going to touch it
9 again or we may upgrade in 50 years or 100 years and
10 then not touch it again. So that's a very different
11 set of criteria than we would be using in construction
12 and so there's a lot less experience in that.

13 Now because we are looking at much shorter
14 life times in construction, when we mean durability or
15 when we say durability, we're actually talking about
16 generally postponing or slowing deterioration, not
17 prevention and that's something else that's a very
18 different mind set. It's good enough if you have to
19 make something last for 50 years to simply postpone
20 deterioration. You can slow down things like
21 corrosion by just limiting the permeability or the
22 diffusivity of the concrete to those harmful
23 constituents.

24 For example, with corrosion, you want to
25 keep chloride ions from reaching the steel. But

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1 that's relatively easy to do if you all have to do is
2 make a thicker cover of concrete over it and that will
3 work. But if you're talking about even 1,000 years,
4 let along 10,000 years, postponement and slowing of
5 deterioration are really not going to be that helpful
6 unless you can say there are certain, say, very highly
7 radioactive materials that will lose enough of their
8 radioactivity over that time that in 50 years we don't
9 care as much and then we can allow certain kinds of
10 deterioration to have happened. So we may have to
11 look a little bit more carefully at what the criteria
12 are or decide that we can actually prevent
13 deterioration but we really don't know a lot about
14 that.

15 The other concern that is a little bit
16 difficult when you're trying to use knowledge from
17 construction to help in applications that it has not
18 been designed for effectively is that there are
19 certain criteria that matter tremendously for the
20 Department of Energy, for example, that have no
21 relevance to us. So we don't know what they are. So
22 what that means is that in that case, you cannot draw
23 from this larger group of people who are doing
24 research in the construction industry. You really
25 have to rely on those people who are looking

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1 specifically at applications that have to do with
2 radioactive materials. So suddenly you have a much
3 smaller pool of people and much less in terms of
4 resources to look at them.

5 Now the next thing is what criteria are we
6 imposing on the concrete. Now one thing that we do
7 have in here is structural support. Now really this
8 turns out in most cases not to be that difficult to do
9 because if you are actually just using it, for
10 example, as tank fill material, you could have used a
11 completely incoherent material like gravel and that
12 would have done if all you're trying to do is keep
13 your cap from collapsing or keep that tank from
14 collapsing after the fact. So you don't actually need
15 much from your concrete to do this. In that case, you
16 wouldn't care if it was riven with cracks because it
17 would still hold it up.

18 You may need it as a barrier to intruders
19 whatever those intruders are, whether they are plants
20 or animals or humans. So in that case, you're talking
21 about something that's hard to get your roots into or
22 hard for an animal to dig into or just something that
23 looks unnatural because humans can dig through almost
24 anything if they're really determined.

25 Sometimes people want it as a physical

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1 barrier to infiltration, in other words, hydraulic
2 isolation from the environment. There may also need
3 to be a chemical barrier to transport. This is where
4 we get into the reducing capabilities and the pH and
5 that sort of thing. Basically for most purposes, we
6 want a high pH and a low E_h .

7 And I think often times you haven't
8 brought up the politics of it, but really a lot of it
9 is public perception. I think in a lot of cases
10 especially for low level waste it probably isn't
11 necessary but it makes people feel better because they
12 thing concrete is really solid, even though every time
13 you see it, it has cracks in it. But, hey.

14 Going back again to specific criteria, for
15 structural support probably what we're going to
16 measure is the compressive strength. That's usually
17 something that's easy to measure and everybody
18 understands that. We may care about the stiffness.
19 That is the resistance to deformation when it's under
20 load. And as I mentioned, these criteria are fairly
21 easy to satisfy. We just need to make sure that
22 cracking is controlled and we have the durability we
23 need so that, for example, if we still need to have a
24 certain measure of compressive strength in 100 years
25 or 300 years or whatever that may be, that it has not

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1 deteriorated so much that we have lost that. And
2 actually, it has to deteriorate pretty thoroughly
3 before it doesn't have any compressive strength left.

4 Now as a barrier to intruders, if we're
5 talking about plants, animals or humans, here we
6 probably need to be more careful about limiting
7 cracking because of course a plant can find its way in
8 if there's something to be had and then the problem
9 there is that once the root gets in it could start
10 wedging it apart. So we need to get sufficient
11 resistance which could be strength in or thickness, in
12 other words, to make it difficult for an animal to
13 burrow in and then you wouldn't care if there were
14 some cracks in it. You would just have to have
15 something that was solid enough and even really a
16 thick enough layer or rubble would probably do for a
17 lot of burrowing animals.

18 For humans, you might be looking more at
19 things like adding a pigment or to do something to
20 make it look really unnatural. And remember that
21 after awhile, concrete that was pretty well broken up
22 might not look as unnatural as all that. So you might
23 really want to look at a pigment of some kind of stuff
24 that was not seen locally so that people would know it
25 was really unnatural.

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1 Now concrete as a physical barrier to
2 infiltration is probably not all that wonderful. Even
3 without the cracks, the concrete is porous and
4 permeable and all the models if they have any relation
5 at all to reality will show this. I mean they're all
6 talking about some sort of permeability or
7 diffusivity. So you're always going to have even in
8 the small scale if you have no effective cracking
9 you're still going to have this. With cracking, of
10 course, it is in orders of magnitude more permeable.

11 So if your barrier has to be completely
12 impermeable, you really need to have some other system
13 and you may be able to do a good deal with things like
14 site grating so that the water runs off rather than
15 leeching through. You may want to do a lot of things
16 with clay barriers and other things like that that are
17 more appropriate for that and also using vegetation to
18 take up what water is there. So you're really looking
19 at a total system, not that concrete is doing
20 everything and I think it makes more sense to let it
21 do what it's good at and not expect it to do what it
22 really isn't capable of doing.

23 Now as a chemical barrier to transport in
24 general as I mentioned, we want it to have a high pH
25 and a low E_h to make the constituents as immobile as

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1 possible. Now Portland cement and concrete naturally
2 has a high pH because of the calcium hydroxide which
3 is one of the products of the hydration reactions.
4 The reducing characteristics, this low E_h , would be
5 imparted by slag cement. This is the ground
6 granulated blast furnace slag.

7 Over time, leeching may alter these
8 characteristics and that's where I'm not sure that we
9 have as good an understanding of this as we really
10 need and remember we know a fair amount about the pH
11 because we care in construction about pH because high
12 pH protects steel from corrosion. And so we do have
13 a pretty decent understanding about that in the
14 relatively short term, you know, the first few
15 decades. We probably can say something sensible about
16 that, maybe not everything you want to know, but at
17 least something.

18 Reducing capabilities, if you ask most
19 people in construction, they would have no clue what
20 that even was. There just isn't any need for it in
21 our normal lives. So we don't deal with this. I know
22 about it because I've worked in nuclear things before.
23 But that's the only reason.

24 Now going back to public perception and
25 I'm not a politician, so I don't want to get too far

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1 into this but just to bring it up because it is part
2 of the context and it is something that will often
3 dictate what gets done. In a lot of cases, the use of
4 concrete isn't really necessary. You could probably
5 get away with especially in the case of low activity
6 waste you could probably design your landfill very
7 nicely without it. You don't really need it. But it
8 does give people a good feeling about it.

9 In particular though, concrete is ill-
10 suited as a long term barrier against water and this
11 may seem counterintuitive if you're thinking of dams
12 that hold back water. But in fact, they're not
13 necessarily holding back all of it. It's probably
14 getting through somehow and you don't particularly
15 care. It's capable of fulfilling its function as a
16 dam while still allowing some water through. But if
17 we don't want any water through in the applications
18 that we're talking about, then it's not concrete
19 that's going to do this for you. It has to be
20 something else.

21 It's probably appropriate as an intruder
22 barrier. So it is a way of making it difficult for
23 plants to get through and particularly burrowing
24 animals. It can also provide structural support.
25 That's something it fairly easily does, although there

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1 certainly are other ways to do this. So it isn't
2 necessarily the first thing I would think of if I were
3 trying to provide that.

4 So the criteria then we've sort of
5 discussed. Now look at the kinds of characteristics
6 of the concrete we're after. We probably want a
7 certain measure of strength, although this is unlikely
8 to be the controlling factor. In the construction
9 industry, we have been able to make strong concrete
10 for a long time and that's not really the issue
11 anymore even for us. We tend to be much more
12 concerned now about durability.

13 We want to do things to minimize the
14 cracking and the reason I would like to de-emphasize
15 strength is that normally the things that we do to get
16 strong concrete ironically enough will tend to make
17 more cracks. So we need to be careful that we don't
18 decide if some is good, then more is better in terms
19 of strength because the more things we do to get high
20 strength the more likely we are to have some serious
21 problems with cracking and that may be detrimental to
22 the application that we're considering.

23 We probably also want minimal permeability
24 and it happens a lot of times people use strength as
25 a surrogate for permeability and that's a mistake.

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1 They really need to look at the transport properties
2 as directly as they can to obtain those and if it's
3 done at the expense of strength, so what because we'll
4 probably have plenty of strength anyway.

5 We are also concerned about favorable
6 chemistry and this is obviously chemistry over the
7 long term and also favorable microstructure which is
8 to say as low permeability as we can, as low
9 diffusivity as we can. We need to stabilize
10 radionuclides and something that really hasn't been
11 mentioned here but it is often an issue is the toxic
12 heavy metals because a lot of times these wastes do
13 have other components that are not radioactive, but
14 they can be just as nasty to the environment and these
15 don't go away. So it's not like you're going to --
16 These things don't have a half-life. They're just
17 there unless they are transported somewhere and you
18 don't want that either.

19 So these things have to be kept in mind
20 and some toxic heavy metals actually do have certain
21 effects on the ability of the cement components to
22 set, for example. So you may have some sort of a
23 waste form where you make it and it never sets because
24 you had say too much lead in it. So there are things
25 like that that will affect how these waste forms work.

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1 Then the next question is how long do we
2 actually want to the concrete to be able to do these
3 things. It may be that we don't need structural
4 integrity for the entire 10,000 years. It may be that
5 it's good enough if we have it for the first 50 years
6 and then it's so riven with cracks that maybe then
7 we're concerned more with the chemistry or whatever.
8 So I think we really need to look at this in more
9 detail and it seems like some of the models that I've
10 seen they are starting to do this in sort of a
11 rudimentary way but I don't think they've really
12 considered this as well as they probably need to.

13 Now if we look at how to minimize cracking
14 which obviously is a big issue here, in a model cracks
15 are, just as in real life, going to form for a
16 specific reason and so it isn't just that you have
17 deterioration over time and it's sort going on at a
18 steady rate and you get so many millimeters of cracks
19 per year or anything like that. A lot of it happens
20 in a very short time and then those cracks may
21 propagate, but they start for a particular reason and
22 they propagate for a reason.

23 Now we have thermal cracking which is
24 basically what happens when you get differences in
25 temperature which will result in some of the concrete

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1 expanding and some wanting to contract. So they are
2 basically fighting each other until the crack forms
3 and that relieves the stress. This is relatively
4 early on and basically what happens mostly is that it
5 has to do with heat that is generated during the
6 hydration of the cementitious components. So that's
7 basically what you're dealing with there is how much
8 heat are you generating and when it is being
9 generated. But most of this is occurring within the
10 first hours, days and weeks, not later. So that's
11 something that you're going to deal with soon and then
12 those cracks may propagate for other reasons, but this
13 is early relatively speaking.

14 Plastic and drying shrinkage both occur
15 due to the drying of your cementitious materials and
16 that again is very early on. Plastic shrinkage
17 happens before it sets and drying shrinkage will
18 happen following setting usually within -- I mean you
19 can still measure it after a year, but it's just so
20 minimal by then you don't really care. It's again
21 something that happens earlier and not so much later.

22 If you allow the concrete to shrink as
23 much as it likes, then you will not generate stresses
24 and you will not generate cracking. But normally in
25 any real system, you have some degree of restraint and

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1 so basically if it wants to shrink and you don't let
2 it, then you'll generate stresses that could result in
3 cracking.

4 Structural overloading, again basically
5 just something that would happen episodically. It
6 could happen because of an earthquake. It could
7 happen because during construction your construction
8 vehicles drove over it. It could be that you're
9 casting some sort of pre-cast concrete and the
10 greatest stresses it ever sees are when you're
11 assembling this thing and then just picking it up may
12 be the most stressful thing you ever do to it.

13 So how do we control thermal cracking?
14 This chart is an example of what we did. This
15 happened to be from construction of a hospital MRI
16 facility and they're using relatively thick concrete,
17 approximately seven feet thick, as radiation shielding
18 around the structure. So it forms the floors, the
19 walls and the ceiling of this facility.

20 This shows you what are thermal couple
21 measurements measured at different locations in the
22 section over time and you can see this is something
23 that -- Our peak in this case happened approximately
24 at 200 hours. So you can see that this is a
25 relatively short term phenomenon. But the design of

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1 this concrete was specifically developed for mass
2 concrete. So it was supposed to generate relatively
3 little heat. That was a good design.

4 Basically, what you're trying to do is
5 keep the thermal stress less than the tensile strength
6 at all times and so that's how to prevent any cracking
7 actually is to keep the stress less than the strength.
8 Here ACI has a short-cut method in which you just
9 maintain the temperature difference between the
10 surface and interior to less than 35 Fahrenheit
11 degrees or 20 Celsius degrees and that's what we were
12 doing here is just monitoring to make sure that
13 happened and the reason that monitoring is useful in
14 this case is that we know what the interior
15 temperature is and then we can say, for example, if
16 the contractor wants to remove the forms and we know
17 what the air temperature is and we say that difference
18 here is more than 35 degrees, no, you leave the forms.
19 Or if you want to take them off, you can tent and heat
20 the structure and you can keep the heat on so that you
21 don't have more of a temperature difference than we
22 can tolerate.

23 This is something that might conceivably
24 be used in some of these applications because you
25 certainly do have mass concrete. And basically the

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1 way you do this is you select materials that don't
2 generate a lot of heat. That's one reason why you
3 like slag and fly ash as opposed to lots of cement
4 because it's cement that generates the most heat and
5 the earliest heat.

6 You will also do things to minimize the
7 amount of cementitious material overall. The
8 aggregate does not generate heat at all. So the more
9 aggregate you use the better off you are in this case.
10 So you would look at things like can we use larger
11 aggregate that takes up more space, can we get a more
12 efficient aggregate grating that fills up more space.
13 So there are ways to formulate these things that I
14 think would be very relevant to a lot of the mass
15 concrete that you're using in these applications.

16 How do we minimize the cement content?
17 We have aggregate size and grating as I mentioned, fly
18 ash and/or slag cement and preferably in large
19 percentages. You don't really care about things like
20 how fast does it gain strength. If it takes weeks or
21 months, so what. But you can get a much better
22 product if you use these materials that don't generate
23 a lot of heat and react much more slowly. So they are
24 generating their heat over a much longer time.

25 Another thing that you can do is have a

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1 low placement temperature. There are ways of cooling
2 concrete before you place it. The extreme case would
3 be to use liquid nitrogen and people do that where
4 they'll actually jet liquid nitrogen through there so
5 that it cools it off considerably and you can place it
6 at any temperature above freezing and you will be
7 better off because of course just like any chemical
8 reaction or most chemical reactions, the hydration
9 reactions are a whole lot slower when it's cold and so
10 if you can keep them slow, then the heat will be
11 generated much more slowly over a much longer time.
12 It gives it a chance to dissipate. So that's
13 something that's relatively easy to do and again can
14 be done commercially on a regular basis.

15 Insulation is also helpful. This is
16 probably more of an issue for construction above
17 ground. If it's underground and you're just casting
18 against soil, the soil will be good enough insulation.
19 You don't need to worry about it. But you probably
20 have a free surface that's not against the soil and
21 that would be good to insulate. Then you want to have
22 insulation that can be removed gradually so that you
23 can gradually lower the temperature on the cool
24 surface rather than just take it off all at once.

25 Another thing that's really important is

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1 you need to protect it from rain because of course if
2 you splash cold water on it, it will cool it very fast
3 and you will probably see cracks because of that.

4 Plastic and drying shrinkage both result
5 from drying and sometimes what will often happen in
6 construction is that the crack will initiate by
7 plastic shrinkage and propagate through drying
8 shrinkage. You may not even see it during the plastic
9 stage, but it has in fact already started to crack.
10 So a lot of people will just call it shrinkage cracks
11 because it is in fact hard to distinguish between
12 them.

13 Plastic shrinkage will occur before it
14 sets and as I say, you may not even see this but it's
15 there and then the drying shrinkage is after the
16 concrete sets. A colleague of mine sometimes says,
17 "Plastic shrinkage occurs when the concrete is of the
18 texture of a Three Musketeers bar and drying shrinkage
19 occurs when the concrete is more the consistency of a
20 Butterfinger bar." So it's actually brittle at that
21 point.

22 Basically with this, you minimize both of
23 these by preventing the drying and so that's mostly
24 just good curing practices. If you're really
25 concerned about this and you really want to do the

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1 best job you possibly can, you will also fog the air
2 above the concrete even while you're placing it and
3 finishing and other things just to keep it from losing
4 water while it's being handled.

5 Now this is a little example of how
6 restraint of shrinkage works. Concrete generally
7 speaking does want to shrink and it will do so for a
8 variety of reasons. If you just let it happen, it's
9 not that big a deal. It gets smaller but it's not
10 going to develop a lot of stress. But normally, we
11 have some measure of restraint and that may be because
12 we've cast the concrete against the ground or it has
13 to be joining two things that aren't moving, say, it's
14 a floor and it's going between two columns. So we
15 actually do have measure of restraint.

16 And then of course, the aggregates
17 themselves are a form of internal restraint too.
18 Basically what happens is if it wants to shrink and it
19 can't, then somehow the shrinkage has to be
20 compensated and that's what it's going to do. So a
21 lot of what we do in normal construction is just bow
22 to the inevitable and figure out how we're going to
23 handle the cracking rather than try to prevent this.

24 As a general rule, concrete is roughly ten
25 times as strong in compression as it is in tension and

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1 because of some of the things I've talked about, it
2 tends to develop cracks even before any load is
3 imposed. Certainly the plastic and drying shrinkage
4 could cause cracking before loads are imposed. So
5 could the thermal stresses. So those are basically
6 the things that would tend to cause cracking before
7 loads are imposed.

8 Typically what we do is we minimize the
9 crack width by closely spacing reinforcing bars and
10 what that does is it gives us a lot of narrow cracks
11 rather than a few wide cracks. We may also design in
12 the structure and include prestressing or post-
13 tensioning to keep the concrete in compression. So
14 that's a way a structural engineer might handle it and
15 that's something that's worth looking at. Note though
16 that if you do have prestressing or post-tensioning
17 that over time that will be reduced by creep. So it's
18 more effective at the beginning than at any other
19 time. Also this prestressing steel will be in tension
20 and it will be more susceptible to corrosion because
21 of that. So that's kind of a tradeoff.

22 Structural overload, you could have
23 structural overloading occurring in construction or in
24 service due to any one of a number of causes and you
25 really need to consider all these possible sources of

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1 loading. A lot of them do take place only during
2 construction and at no other time, but even things
3 like driving construction vehicles over something and
4 remember that the concrete is weakest when it's young.
5 It's continuing to gain strength over time. So here
6 you have big heavy vehicles driving over it early in
7 its life. That's bound to be detrimental. Even
8 before you put any other load on it, a structure has
9 its own weight to carry and often that's more than any
10 live load it will ever see.

11 You have bearing pressures from the soil
12 and any groundwater that's there. Soil settlement can
13 be a big problem because the concrete is rigid, the
14 soil is not and so if you have differential
15 settlement, that can be very problematic.

16 Earthquakes which would be of course a
17 single event, but certainly in 10,000 years it's not
18 difficult to imagine how you could have multiple
19 earthquakes. And there probably are plenty of other
20 things that I haven't thought of that you would need
21 to.

22 Now going to permeability and durability
23 which is probably more of an issue for most of the
24 things that we're concerned about. This is something
25 I found when reading through an old concrete

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1 engineer's handbook. You know, of course, remember
2 that when something gets to a handbook it's already
3 old news. But back in 1918, it was apparently well
4 established that water penetration is directly or
5 indirectly the cause of the majority of
6 disintegrations in concrete and the degree to which
7 water penetration is permitted by the texture of any
8 concrete is a direct measure of its strength and
9 endurance. And this is ironic because this happened
10 to be the year when Duff Abrams developed the
11 relationship between water-cement ratio and strength
12 and so we've been concerned about strength since about
13 1918 and it's only been relatively recently that
14 people have started to concern themselves again with
15 permeability and durability. So now we're back to
16 1981 again.

17 This slide gives you some idea about the
18 relationship among strength and permeability and
19 porosity. Normally in concrete we're looking at
20 porosities in this range, but you could conceivably
21 have higher porosities. And you can see that the
22 compressive strength tends to go fairly smoothly
23 along. It gets lower and lower and lower as the
24 porosity increases which stands to reason.

25 But the permeability is more interesting

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1 because you don't see a steady increase. What you see
2 is a steady increase and then a sudden rapid increase.
3 So basically what this is telling you, roughly
4 speaking this break point is around 30 percent
5 porosity. So in general what we would like to do is
6 get the porosity below 30 percent if we're concerned
7 about permeability and diffusivity and all that sort
8 of stuff.

9 Now we reduce porosity, again here is our
10 porosity and here is 30 percent. We have just for
11 theoretical reasons this 100 percent hydration curve.
12 This never happens. Ever. I have seen 100 percent
13 hydration in the lab and it was not done in concrete.
14 Basically they put cement and water in a bottle with
15 some ceramic balls and keep turning it and turning it
16 so those balls keep breaking up the hydration products
17 and what you have at the end is something like a 100
18 percent hydration and nothing like concrete.

19 In real life, 75 percent hydration is
20 probably as good as you're going to get in a real
21 system where you give it a reasonable curing and for
22 that then to get to 30 percent porosity you're looking
23 at a water/cement ratio of about 0.45. So that gives
24 you an idea. Once you get above 0.45, you probably
25 will not see low porosities and therefore you will

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1 always have a continuous pore system no matter what
2 else you do. So water/cement ratio is really key to
3 getting this to work.

4 If we have a water/cement ratio below
5 0.45, we also need to get an extended moist curing
6 time even to get to the 75 percent. If we had 0.45
7 water/cement ratio at 50 percent hydration, we're
8 looking at almost 40 percent porosity. So that's not
9 going to be acceptable.

10 Supplementary cementitious materials,
11 again fly ash, slag and also silica fume can really
12 help here, but again, they need this extended moisture
13 curing time to be able to work.

14 Now looking at some of the deterioration
15 mechanisms, we have carbonation which has already been
16 mentioned and also leeching of soluble materials. We
17 have cycles of freezing and thawing which probably is
18 not an issue once the concrete is buried. We have
19 sulfate attack. We have alkali-silica reaction. We
20 have corrosion of reinforcement and we have
21 irradiation. So these are all things that I'm going
22 to go over fairly quickly to give you an idea of what
23 we might do about some of them.

24 Carbonation has already been mentioned.
25 Carbon dioxide and moisture in the air react with

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1 calcium hydroxide to form calcium carbonate. It is
2 true that you might have also, you might get this from
3 dissolved in water and some other things. So it is
4 possible, but normally we'd see it in air. The result
5 is shrinkage and also a reduction in pH and I would
6 consider both of these things to be undesirable for
7 this application because remember shrinkage probably
8 means cracking.

9 Once the concrete is buried, this is
10 probably not an issue unless you do happen to have
11 water that's carrying carbon dioxide in it. If the
12 concrete is above ground, a coating may be helpful and
13 remember that for most of these systems if you
14 starting with the concrete above ground, it's not
15 staying there. It's going to be buried eventually.
16 So your coating doesn't have to be a permanent thing.
17 It could be something that lasts for just the time it
18 takes you to build the thing and get it buried. So
19 you don't have to have something that lasts 10,000
20 years. Five years is maybe enough. Maybe you need it
21 for ten years.

22 The way we test for carbonation, you can
23 actually see this, you squirt some phenolphthaleine on
24 a freshly broken surface and you can see where the
25 color change has not happened. This is all carbonated

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1 and you can see that normally it's done, it comes from
2 the surface and it will go irregularly into the
3 concrete depending on whether there were cracks or
4 other voids or other things for it to go in.

5 Leeching is an issue because some
6 components of hydrated cement paste are solubles,
7 primarily the calcium hydroxide which is soluble in
8 both water and acid. So if you have any acidic water,
9 your groundwater may be acidic or you may have acid
10 rain, this is more of an issue even than just in
11 water.

12 The loss of the calcium hydroxide will
13 leave open pores and also locally reduce the pH. Over
14 the long term, it really is best to keep the water out
15 and you need to do that by some other means than just
16 the concrete itself.

17 Cycles in freezing and thawing because
18 water expands approximately nine percent on freezing,
19 if you have saturated concrete when it freezes you're
20 in trouble and you can see what happens here. It
21 completely destroys the structure. Again, this is
22 probably not going to be an issue once the concrete is
23 buried because it's probably not going to freeze and
24 hopefully it's not going to be that wet.

25 What we do to prevent freeze/thaw damage

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1 is we entrain air, you can see all these little air
2 bubbles here, and that basically provides some place
3 for the water to go. We also need to make sure though
4 to get this that we have done our job in terms of mix
5 design, mixing, transport and placement so that we get
6 the air in and we don't take it out again. It's
7 extremely difficult to do all of this in hot weather
8 and do it properly.

9 Sulfate attack, this shows you a photo
10 microgram of the sulfate front coming in in this
11 direction and you see the kind of deterioration that
12 occurs and these are, of course, cracks that would
13 come in and that's obviously something that you don't
14 want to have. The tricalcium aluminate in the cement
15 is actually the component whose hydration product is
16 vulnerable to this. You need to be careful that
17 aluminates in some supplementary cementing materials
18 is specifically Class C fly ash can also supply this
19 and so you may end up with just as bad a problem wit
20 them. There are certain kinds of Class C fly ash that
21 can make the problem worse than it was.

22 If you have sulfates, for example sulfate
23 groundwater, and water present, there's no preventing
24 sulfate attack. It's just a question of postponing
25 the deterioration or slowing it down. So this is

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1 something where you really want to make sure that you
2 don't have this in your environment.

3 Alkali-silica reaction, you see an example
4 here of the characteristic cracking that occurs. For
5 your application, you would want to use nonreactive
6 aggregates. What we normally do in construction is
7 we'll use reactive aggregates and we'll mitigate the
8 reaction by using fly ash or slag. Again, though that
9 is a slowing down or postponement of the inevitable.
10 It is not avoiding it and you need to avoid it for the
11 applications you're looking at. So you really need to
12 be aware that if you have a reactive aggregate you're
13 going to have to import something else.

14 Corrosion of reinforcement, this is
15 actually a familiar site for anybody who lives in
16 northern states where it freezes and we use deicing
17 salts on the roads. There are two problems with
18 corrosion. One is the actual loss of the steel area.
19 If you needed the steel for structural integrity,
20 you've obviously lost that. What's probably more of
21 a concern though is that corrosion of steel is an
22 expansive reaction. The products of corrosion are
23 anywhere from seven to ten times the volume of the
24 original steel and of course, that causes a lot of
25 spalling and that's much more of an issue for most

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1 people.

2 The high pH of the Portland cement when
3 it's hydrated does help protect the steel, but
4 chlorides are catalysts for the corrosion reactions
5 and so that's something that you really have to be
6 concerned about. If you have chlorides present, they
7 will continue with this and they're not consumed.
8 They are free to continue to do this.

9 Corrosion is slowest in the absence of
10 oxygen or if there's a shortage of oxygen and that may
11 pertain to a lot of the applications that you look at
12 because most of these things would be underground. So
13 there would be less access for oxygen. But there's
14 always some corrosion going on. It's slower or it's
15 faster. You may reduce the rate by a couple of orders
16 of magnitude if you do the right things, but it's
17 still happening. So eventually you will see this
18 deteriorate.

19 Irradiation, if concrete receives
20 sufficient gamma radiation, it can deteriorate. But
21 what information I was able to obtain came from the
22 use of concrete for radiation shielding where it's
23 actually getting a lot more radiation than it would in
24 our applications. So I'm not sure this is even an
25 issue, but it's possible that it would be.

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1 It would take the form of cracking and
2 loss of strength and stiffness. Again, maybe that
3 that doesn't matter too much over the long term
4 depending on what the concrete was used for.

5 Carbon steel which is what you would be
6 using for the reinforcement and prestressing can also
7 lose ductility with irradiation. It may or may not be
8 an issue. Here this application where they determined
9 this information is very different from what we have.
10 It may not be all that relevant and it may be worth
11 investigating what are the effects at much lower
12 levels of radiation over much longer times.

13 The best model that I know of for modeling
14 deterioration mechanisms is at NIST and I think it's
15 because it's been going on so long and they've made a
16 very conscientious effort to keep it tied to the
17 physics and chemistry and the material science and
18 also to validate it against actual tests. I think
19 they've done a really good job.

20 Otherwise in general, there seems to be
21 rather little connection between the models and the
22 material science. I see an awful lot of models that
23 they just kind of assume things and you wonder how
24 they get there and if you ask, they just kind of look
25 embarrassed and wonder why you want to know.

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1 Some of the assumptions that I've seen in
2 the models that are being used by DOE are very
3 conservative, but it's hard to estimate the safety
4 factors even to the order of magnitude and I think
5 that's just because our knowledge isn't that good in
6 a lot of areas and some of the assumptions are kind of
7 wild. There is naturally very limited knowledge of
8 the long-term behavior because we're using a lot of
9 short-term tests or no tests at all.

10 We have some recommendations coming up in
11 our National Academy of Science's report that I'm
12 participating in. I think it should be out any day.
13 I'm not sure what's happened to it.

14 Just to quickly reiterate a few ideas that
15 I had about monitoring. First of all, the instruments
16 are not going to last decades let alone centuries. So
17 you need to be aware of that and either decide that
18 you're only going to monitor certain things for a
19 short time or make provision for monitoring with
20 something else and changing several times during the
21 course of things.

22 Electronic data would need to be
23 transcribed to some long-term formats just because you
24 can't read old formats of electronic data for very
25 long. The things themselves deteriorate or they're

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1 just so incompatible with what you have now that you
2 can no longer read it. So I think that may end up
3 pointing people to stuff that may be very old-
4 fashioned and simple, but at least you can still use
5 it.

6 It may be best to install access ports,
7 benchmarks and so on, things that don't change very
8 much over time and then let whoever is monitoring it
9 use the instruments of their own day. So that way
10 they can just get in there and cite on the benchmark
11 or whatever they need to do citing on it, however they
12 like.

13 Something else that could be done is you
14 could put materials coupons in certain exposures just
15 to see what happens over time and then you can observe
16 them, pull them out and take a look and see what's
17 happened to them.

18 I guess we weren't going to do questions
19 at this point. So I'll just end.

20 VICE-CHAIRMAN CROFF: Okay. Thank you
21 very much. Our next speaker is Dr. Barry Sheetz who
22 is a Professor of Civil and Environmental Engineering
23 at Penn State. His research activities focus on
24 cementitious materials for civil infrastructure,
25 reclamation and nuclear waste management.

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1 PROFESSOR SHEETZ: Thank you, Allen. In
2 preparing this presentation, I've talked to Allen and
3 we did try and get an idea of where I fit into the
4 grand scheme of things today and we were talking and
5 he suggested talking about failure in cementitious
6 materials and he suggested the potential of discussing
7 cracking and you'll find out as Chris has pointed
8 earlier in her conversation that there are synergisms
9 throughout this entire process and considering that
10 Rachel just gave one-third or more of my presentation
11 you'll see the synergisms develop.

12 I'm going to talk about cracking but I'm
13 also going to give you a disclaimer. As we discussed
14 with Dr. Esh's question earlier, most of our
15 knowledge, most of our intuition, most of our
16 expertise, resides from civil infrastructure
17 applications. We just haven't done enough of these
18 types of tank closures in sequestration of nuclear
19 materials in order to gain the broad scope of
20 knowledge that we're wanting and that you're asking
21 for right now.

22 So we have to go back to what we know.
23 You always want to start with what you know and then
24 extend it out into the unknown and we're starting with
25 civil infrastructure. We have a very significant

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1 background in concrete. We have a very significant
2 background in Portland cement concrete. We have a
3 very significant background in construction and we
4 have this to call upon.

5 A fundamental tenant of anything that we
6 do with Portland cement concrete is that "all concrete
7 cracks" and that's exactly what we heard. You expect
8 it to crack. The civil engineers when they're
9 designing, now I'm a civil engineer but I've been
10 raised as a geochemist and my entire career up until
11 about Thursday was in Materials. So I look at things
12 differently than my colleagues in civil engineering.
13 I look at it from chemistry and phase composition
14 rather than how close to the functioning specs can you
15 get your material. If it cracks they can engineer
16 around that. But just because it cracks, that doesn't
17 say that all cracks are bad. We can mitigate those.
18 We can control them and this is what we need to
19 discuss.

20 Why are cracks bad? Well, I'm not going
21 to go through all of this, but generally if you're
22 looking at hydraulic conductivity, the hydraulic
23 conductivity is composed of two parts. It's composed
24 of the matrix conductivity that David talked about it
25 and of course, it's controlled by the crack and the

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1 flow of water through the crack. And what you'll find
2 out is that this water flowing through this crack is
3 the predominant force or the predominant factor
4 controlling the movement of water through the concrete
5 object.

6 The crack width becomes very important.
7 You can see width down there versus hydraulic
8 conductivity in centimeters per second and you get the
9 linear relationship just from this and not taking in
10 the permeability. ACI 224 on cracking, it's a
11 committee on cracking, gives a guideline. It used to
12 be more than a guideline until the lawyers go involved
13 with it. But they give you crack widths, two sets of
14 crack widths, one for water retaining structures and
15 another one for protective membranes and these span
16 the amount of allowable crack width within a structure
17 and you can see that we're talking about hydraulic
18 conductivities that are much, much higher than what we
19 would consider acceptable for the applications that
20 we're discussing today.

21 If it's a partially-saturated system, the
22 Wabash equation comes in, we understand how this
23 behaves and then we can determine the width of the
24 crack that will drain water. Not all cracks will
25 drain water. So not all cracks are necessarily bad.

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1 And again, this is probably a third
2 derivative of the 1918 Handbook that Rachel just
3 talked about. We know that the premature
4 deterioration of concrete structures, it can be traced
5 to cracking. It can generally be traced to cracking
6 and it allows ingress of deleterious agents into the
7 interior of big cementitious bodies. That's how it
8 fails. The hydraulic conductivity through the
9 tortuous microstructure of a concrete is generally
10 slow enough that if you go from a two inch cover over
11 a rebar to a four inch cover over a rebar most of the
12 corrosion of rebar disappears. So it's a relatively
13 slow process in engineering time frames, but when it's
14 crack, you get it through quickly.

15 We've heard from Rachel a little bit of
16 the cracking and time frame. I've tried to give that
17 in some sense here and I wish these were years, but
18 they're hours and you can see that there's a thing
19 called placement settling that will contribute to
20 cracking and this is while it's still plastic
21 aggregate flowing around objects, around rebars. You
22 see this a lot in parapets where you're forcing
23 concrete through a parapet and it will settle and flow
24 around the rebar. But you get these various phenomena
25 that contribute to cracking occur as a period of time

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1 in hours, tens of hours and in obviously my log scale
2 and we get to the freeze/thaw and alkali-silica
3 reaction and the sulfate attacks that we talk about
4 and in these cases, you're talking about years to
5 reach the full degradation effects of cracking
6 associated with those.

7 The underlying reason is very
8 straightforward. Rachel talked that as a general rule
9 of thumb concrete is ten times stronger in compression
10 than it is in tension. We know that. That's not a
11 big deal. So that any time the tensile stresses
12 exceed the mechanism strength, a crack develops. So
13 the onset of cracking looks like this with time after
14 drying and stress level.

15 If we did not take into consideration the
16 behavior of creep in a concrete what you would follow
17 is this kind of a behavior here where the stresses
18 exceed the crack resistance and cracking would occur
19 here. But because cement creeps, creep will locally
20 diffuse the microcracking. It will locally
21 disseminate this, the stresses, and we extend the age
22 of cracking from basically here to here. So the
23 residual stress level when we take into consideration
24 creep has to be evaluated when you begin to design
25 your structures.

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1 Now how does creep respond to strength?
2 It's important. That's what I just said. And creep
3 is inversely related to strength. If you want to
4 build a (Inaudible.) strength concrete, it's going to
5 have very low creep. So in this particular case, I
6 think we want creep. So what kind of a strength
7 should be recommended to design into this? Basically,
8 what we would recommend is you design the strength in
9 there to be just enough for what you need. You don't
10 need 15,000 psi compressive strengths. Basically you
11 need something that's probably a compressive strength
12 of a little over 40 psi for if you're going to fill an
13 underground tank and I'm looking here like and I'm
14 approaching the whole presentation focused on large
15 cementitious bodies, a.k.a. underground tanks.

16 So creep's important. We know how to
17 control it. We know what that creep will do for us.
18 We know the benefits that it will give us. We don't
19 need to make high strength cement.

20 When we sit down, we start looking at the
21 factors that impact cracking in cementitious bodies.
22 We see that there's chemical and physical properties
23 of the cement itself. We have to look at the cement
24 itself. This is becoming very, very important and
25 then there are the external conditions. There's

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1 mechanical and there's environmental. I'm going to
2 step down through these and when I started to prepare
3 these, this I have 20 different mechanisms or 20
4 different things that can contribute to creep and that
5 sort of sounds daunting like we could never make a big
6 piece of concrete that would survive.

7 Properties of cement that impact the
8 cracking. These are the cements itself. High
9 tricalcium silicate content suggests that there's low
10 dicalcium silicate content. With time, what has
11 happened is the market who's using, this is the
12 construction market who's doing the civil
13 infrastructure, has demanded from the concrete
14 manufacturers, demanded from the specifying agencies,
15 that what we do is we want a concrete that we can pull
16 the forms off of faster when we're constructing
17 because the faster we pull the forms off, the more
18 money I can put in my pocket.

19 So what they're doing is there's a subtle
20 draw, there's a subtle market draw, on the industry
21 saying faster concrete, faster setting, faster setting
22 and what this does is it impacts the chemistry. These
23 are not unrelated. You will find the fact that we're
24 putting more tricalcium silicate in our cements means
25 that the heat of hydration of tricalcium silicate is

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1 higher by also a factor of four than the dicalcium
2 silicate. So we're producing more heats of hydration.
3 If you go back, this ratio is changing and we'll talk
4 a little bit about that.

5 We're also tending to get more tricalcium
6 aluminate into the Portland cements and as a
7 consequence, we're having to put in more sulfate to
8 ameliorate the early age reactions of the hydration of
9 tricalcium silicate. We're also finding that we're
10 grinding, we're tending to grind, these cements finer
11 and that means a higher surface area. That means a
12 higher chemical reactivity. That means faster
13 strength gain. That means I can get my forms off
14 quicker.

15 The other thing that we're finding is that
16 deleterious is a high alkali content and this is from
17 the other end of things. This is from the
18 environmental end of things. Most cement
19 manufacturing in the United States has now shifted to
20 the dry kilns as opposed to wet kilns and in dry
21 kilns, what you end up doing or what you were getting
22 -- I'm sorry. In the wet kilns and in the dry is
23 cement kiln dust. You're getting dust that comes off
24 and it's collected in the bag house during these
25 processes.

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1 In the wet kilns, you couldn't put that
2 back into the kiln. You could only put a very small
3 percentage in. But in the dry kilns, you can take
4 this dust that you've collected and you can insulflate
5 it back into the kiln. This insulflation gets rid of
6 your waste product that you don't have to dispose of.
7 But that cement kiln dust is very high in potassium
8 and potassium is our killer in the alkali content of
9 cements, potassium and sodium.

10 So a manufacturer today is saying I don't
11 want handle. I don't have to deal with EPA. I don't
12 want to have to deal with the local environments,
13 environmental regulations on handling this separate
14 waste stream. I'm just going to blow it back into my
15 kiln and as a consequence what we're doing is we're
16 raising the alkali content. This is good and it's
17 bad. The problem with this game is that there's no
18 black and white.

19 We did a study, an FHWA study, that was
20 Della Roy was the lead author on it. It was Paul
21 Takowski (PH) and I were also on this project and when
22 we compared this data just to back to 1969 to Blaine's
23 study, we found a 40 percent increase between 1969 and
24 2003 in the sulfate content. We found a 45 percent
25 increase in the potassium and that directly reflects

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1 the conversion of going from wet kilns to dry kilns.
2 We see the calcium, this ratio of dicalcium/tricalcium
3 silicate, has gone much higher. We have a eight
4 percent rise in the sodium equivalent and we found the
5 cements are much finer.

6 In a lot of authors and Burrows is the one
7 that I read in preparation, this is ACI Monograph No.
8 11, and Burrows is a hoot. If you haven't read it,
9 you need to get that book and read it. Burrows, he
10 has a quirky sense of humor and he's opinionated and
11 it comes through very, very quickly and I respect a
12 lot of what he says, but Burrows and a lot of other
13 people think that what we're doing by catering to the
14 construction industry to change and get the cement to
15 hydrate faster and set up faster that we're making
16 poorer quality cements. We're causing the
17 microstructure to change because it's hydrating so
18 fast the microstructure of this. The hydration
19 products in concrete is poorer than it was back when
20 in the '20s and '30s when we had a very high dicalcium
21 silicate content and were hydrating much more slowly.

22 This is also stolen out of Burrows and you
23 get an idea of how things have changed over this 70
24 year period. Sid Mendez in one of his publications
25 have a very, very good graphic which I stole. I used

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1 in my classes and since I've moved offices recently,
2 I can't find anything and I couldn't find it. But he
3 found two inch cube strength data that span from like
4 about 1870 through current times and it's a log linear
5 increase. It's an increasing logarithmically of time
6 and the strength and you can see that here. You can
7 see the fairly significant changes that are taking
8 place.

9 We made some pretty damn good concrete
10 back here and why? Because it hydrated slower.
11 Burrows would have you think it also was better
12 because we didn't cure it as long. He would also have
13 you believe that it was better because of the higher
14 water to cement ratio. Now when you read Burrows, you
15 have to take him with -- You have to understand him.
16 There are some things in there that nah --

17 But the bottom line on it is that the
18 cement that we're manufacturing today is changing. We
19 may not in the applications that we're going to talk
20 about if we do and use Portland cement, we may not
21 have control over any of this. But you have to know
22 it's changing and you have to know what these changes
23 are doing to the performance. Cement that hydrates
24 rapidly generally gives you a more poorly developed
25 microstructure. Slower hydration is better, the moral

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1 to that story.

2 Let's look at practices. Water to cement
3 ratios, we've been pushing. You used to if you
4 specify FHWA it would specify a 0.5 water to cement
5 ratio for bridge deck. Then it was down to 0.7. Then
6 to 0.4 or 0.5, I'm sorry. 0.47, 0.45. We're doing an
7 FHWA study at Penndot right not where some of our
8 decks are down at 0.43. We're pushing it to lower,
9 lower water to cement ratios and this may not be good.
10 Once you get it down below 0.4 you tend to get into
11 another situation where the hydrating cements will
12 suck up all of the water that's there. It's called
13 autogenous shrinkage and you'll remove all of the
14 water. You'll severely reduce the pore structure, the
15 pore fluids and then deleterious other reactions open
16 up.

17 We are also adding accelerators in many
18 cases because as soon as I can get these forms off I
19 can make more money. So we're trying to accelerate
20 these reactions. We'll go farther out into the winter
21 season beyond the end of the concreting placement
22 season and we can accelerate this by adding calcium
23 chloride which would be a death knoll if you had any
24 kind of reinforcements in there.

25 And then we're pushing for very long

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1 periods of curing and I put this in there. This is a
2 Burrows statement and I find it interesting because
3 he's concerned in his statements about the use of
4 hydrating all of the cement. Rachel has shown that
5 with a 0.47 water to cement ratio in her graph you
6 still have 25 percent unhydrated clinker phases in
7 your concrete.

8 If you have hydrated everything, it's
9 considered bad and Brian Mather is one who expressed
10 a great deal of concern about the consumption of all
11 of the anhydrous phases in your concrete because
12 microcracking can be an autogenously healed. If you
13 microcrack and you get water in and you have on
14 hydrated cement present and one of these microcracks
15 goes by an unhydrated part, a piece of Portland
16 cement, one of the cement minerals and it gets wet,
17 there's no reason why it won't rehydrate. So we have
18 to be cautious and we can indeed recover a lot of our
19 failure and a lot of our cracking by autogenous
20 healing of concrete. Don't necessarily build that in,
21 but it is a reality.

22 Let's talk about these external or the
23 mechanical loading. I haven't stress a lot of this
24 because I'm looking at a buried underground tank at
25 Savannah River or a buried underground tank at Hanford

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1 and short of an earthquake, we're not going to get
2 cyclic loading on it. We're not going to get a great
3 deal of static loading on it. It's in a stainless
4 steel tank or not a stainless -- It might be. It's in
5 an iron tank. It's contained. The tank may have a
6 hole in the floor. It may leak. But basically you
7 have a rigid containment.

8 Loading is a problem because in a
9 stress/strain space concrete is nonlinear and as soon
10 as you load it in any manner it's going to start
11 microcracking and when it fails, of course, is when
12 all these microcracks coalesce into failure. So I'm
13 done playing this for the scenario that we're talking
14 about today. The one episodic event that we have to
15 keep in mind which I didn't think of was earthquakes.

16 Shrinkage. There are two types we've
17 heard about. We've heard about plastic and drying
18 shrinkage. Plastic shrinkage is a short-term
19 phenomena. It results in part from aggregate
20 settling. It's in part from evaporation from a higher
21 temperature and low humidity. You're doing this in
22 Hanford. You could get high temperature and low
23 humidity. It's something to be concerned about.

24 But are you going to get that in a tank
25 where you're pumping this? Probably not. The

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1 temperature is going to be reasonably stable. It's
2 going to be less than the -- I've been out there at
3 103 or 104 with about 20 percent relative humidity.
4 It's actually fairly comfortable. It's not like
5 outside today.

6 The other thing that we have to worry
7 about is wind because wind is going to contribute to
8 the evaporation rate and we need to control that. So
9 any time you're doing these kind of placements whether
10 it be in an open vault at Savannah River where the top
11 is off while you're pouring you have to take this into
12 consideration. And with the finest of cement, all of
13 this tends to increase.

14 Drying shrinkage is a slightly longer term
15 and it's basically related to evaporation. If you
16 control the scenario and you control the environment
17 of placement you have the chance of controlling this.
18 If you're pouring it in a tank through a hole, most of
19 this is a non-consideration. Just to give you an idea
20 of the effects of shrinkage on evaporation, this is
21 kilograms per meter squared and this is the measured
22 shrinkage and we have fairly decent understandings and
23 relationships to what's going on there.

24 Autogenous shrinkage is a consideration
25 that in the '20s they never even thought of. We now

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1 need to take that into consideration because we're
2 pushing for high performance cements and I think what
3 you have to do is define what high performance is.
4 High performance is not necessarily high strength, but
5 most people correlate them. When you go to high
6 strength concretes, you saw from one of Rachel's
7 figures that you go to low water to cement ratios and
8 you can get enormous strengths.

9 I have a concrete that I had patents on in
10 the early '80s that had 70,000 psi compressive
11 strength in a concrete. It didn't have any water in
12 it. We cooked all the water out of it. So if you can
13 push the water down, your strength goes up. But once
14 you get down below 0.4 what will happen is that the
15 influence of the hydration phase is going to pull that
16 water and going to pull that water out of the pore
17 fluids and you'll get cracking associated with this
18 removal of the water and that's also with water to
19 cementitious materials.

20 Silica fume is particularly noted for
21 this. When Silica fume first came onto the market and
22 was being used, they were pushing it at 30 percent, 25
23 percent. If you go back to that patent that I had in
24 '83 or something like that, we said five to seven
25 percent. And if you look today, it's in the three to

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1 five percent that people are recommending.

2 Thermal behaviors. Rachel did a marvelous
3 job on this, so I'm going to go through it quickly.
4 The equivalent alkali content has a strong impact on
5 thermal cracking, fineness. The tricalcium aluminate
6 which you heard, the tricalcium silicate content also
7 has a fairly significant impact on it and we've
8 discussed those.

9 Internal and external restraints we've
10 talked about. This is I think going to be a big
11 problem because the tanks that we're going to fill,
12 you know you picture a tank and when the layman thinks
13 of a tank, they think of a tank that's a right
14 circular cylinder with nothing in it. In reality, we
15 know that these tanks are penetrated by dozens of
16 cooling vents and pipes and all of kinds of probe
17 ports and whatnot and as the concrete or grout of
18 whatever we put in here starts to dry, starts to cure,
19 against these, these are going to act as restraints.
20 These are going to act as points of restraints and
21 these have the potential therefore to enhance
22 cracking.

23 For external behavior, of course, it's
24 contained in a tank. It's contained in this iron
25 shell and it's going to thermally expand against that.

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1 Preferably we won't have an -- Or maybe we want an
2 expanse of concrete in there to put a positive force
3 against that and we can engineer around this in many
4 cases by having a positive internal stress left in the
5 concrete when it finally does cool down.

6 Freeze/thaw. We've gone through this.
7 Nine percent expansion. When that water expands in
8 the pores, it puts a hydraulic pressure on the
9 interconnected pore water and it has to go someplace.
10 If it doesn't have a place to expand, you're pumping
11 a hydraulic pressure on the inside pore structure and
12 you're going to get microcracking. When these
13 microcrack coalesce, you get failures.

14 What we do is we introduce bubbles. We
15 introduce about five to nine percent bubbles that are
16 about 100 microns with a less than 0.2 millimeter
17 spacing and we give it a place for that water to
18 expand into as the ice freezes. A well developed
19 aerated concrete, freeze/thaw is a non-problem
20 anymore. Are we going to see a freeze/thaw problem at
21 Hanford? Probably not even if we're casting in the
22 wintertime because we're going to be casting
23 underground. So again, I don't think that this is a
24 major concern.

25 Corrosion. I'm not going to go through.

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1 I think Rachel did a wonderful job there. The
2 corrosion is a problem. The corrosion products are
3 expansive. You have to keep chloride out. There is
4 a threshold in civil infrastructure concrete above
5 which that we know chloride begins to initiate the
6 corrosion of the rebar. But commensurate with that,
7 we have to get the pH of the pore fluid down so that
8 we go from a passive to an active surface. And there
9 is a lot of, as Rachel pointed out, concrete engineers
10 out there who don't know what slag does for them or
11 don't really realize that it's controlling the E_h of
12 that solution as well and extending the durability of
13 it.

14 The alkali aggregate reaction, they come
15 into two forms. They come as alkali-carbonate
16 reactions and alkali-silica reactions. Basically this
17 is the reaction that you've all seen where carbon
18 dioxide is entering the pore fluids and reacting with
19 calcium hydroxide. The reality of the matter is that
20 there isn't one phase in the mineralogy of clinker
21 that is stable with respect to carbon dioxide.

22 So let me put that another way. The
23 thermodynamic end products given enough exposure to
24 moisture and to carbon dioxide, the thermodynamic end
25 products are calcium carbonate, quartz, silicon

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1 dioxide, aluminum oxide, water and calcium carbonate
2 and Chris in her graduate student years had an
3 opportunity to go over Crete and look at some of these
4 with -- Who did you go with? Was that Geographic?

5 DR. LANGTON: Smithsonian.

6 PROFESSOR SHEETZ: Smithsonian study to
7 look at these old concretes as a function for long
8 term use in the applications that we're talking about
9 and the Colosseum is glued together with calcium
10 carbonate and silica. So carbonate chemistry is a
11 very interesting concern, however, it's all relative
12 humidity driven. So if we can keep the relative
13 humidity up, it's a non-problem. If we keep it up in
14 the 50 to 100 percent range, we minimize the effects
15 of the deleterious carbonation effect.

16 (Microphone noise.) labs they've actually
17 had a program out there where they are specifically
18 carbonating Portland cement in order to change its
19 properties and enhance its properties and they were
20 basically making a moral when they finished their
21 reactions. But we do have some experience with rate
22 kinetics and things like that from those studies.

23 The alkali-silica reaction you've seen as
24 these polygonal cracks. Basically what happens here
25 is that the rock contains forms of silica that are

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1 more water soluble. The pore fluids, we've talked
2 about the pore fluids being controlled by calcium
3 hydroxide at 12.45. The reality of the matter is that
4 it's not controlled by that, at least not initially.
5 It's controlled by the alkali, mostly the potassium
6 hydroxide content and the pore fluids are actually up
7 at about 13.3 or 13.4. That's why if you play in
8 concrete with your bare hands at the end of the day
9 your hand feels like a piece of sandpaper because
10 you're dissolving. It's dissolved all the fluids out
11 of your hand, the oils out of your hand and you become
12 rough.

13 These high pHs will dissolve certain forms
14 of silica, crystabolite for instance, and they tend to
15 have a higher solubility and they'll take those into
16 solution. What they'll do is the whole process is
17 fairly complex. I'm not sure that the model is fully
18 developed, but what you'll find is it gets an osmotic
19 pressure in this silica gel. Water is absorbed. As
20 that water comes in, the gel expands and microcracks.
21 Sid Diamond found that there's this pessimum and I
22 sort of like that idea. There's an optimal value of
23 silica where it favors the deleterious expansion and
24 again that's something you can determine.

25 You can minimize it. You can generally

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1 eliminate it by the use of blended cements and these
2 are the supplemental cementitious materials that are
3 put in and that will stop it. Size of the aggregate
4 also affects the alkali-silica reaction and the use of
5 lithium. Lithium does a lot of things, but it also
6 cures ASR.

7 One of the other carbonate reactions is
8 the so-called dedolomitization where the alkali
9 hydroxide if you have any amounts of dolomite in your
10 aggregate you can get it to decompose and form
11 brucite, an alkali carbonate and calcium carbonate.
12 Brucite is not necessarily very good because if it
13 goes from magnesium oxide to brucite there's a very
14 significant swelling in it. That's how we seal
15 geothermal wells with the paraclay brucite reaction.

16 There are other environmental effects from
17 sulfate attack. There is alkali ingress into the
18 concrete. Magnesium ingress. The so-called delayed
19 ettringite formation and that may be a can of worms.
20 When I speak, I'll open it and then duck and
21 ettringite formation. I'll go through these really
22 quickly.

23 The ingress of sulfates will react with
24 the tricalcium aluminates and the tricalcium
25 aluminates are in the cement but as Rachel pointed

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1 out, you have to look at very carefully your
2 applications with other supplemental cementitious
3 materials because a Class C fly ash has a mineralogy
4 that has a lot of the cement phases in it and that
5 could put that into it. So you could get some
6 tricalcium aluminate from your supplemental
7 cementitious material.

8 But generally they form gypsum, ettringite
9 and monosulphoaluminate and if these are formed while
10 the cement is a plastic, while it's fluid, ain't no
11 problem. But let it get hard, let a structure set up,
12 and let them begin to form and now you have a problem.
13 They'll start to expand. Their molar volume is larger
14 than their constituent starting materials and you'll
15 initiate microcracking. I don't need to go through
16 the chemistry of all that. I'll go through it very
17 quickly, but you can have it there as a reference.
18 And you can assume that the rate of sulfate attack is
19 almost directly proportional to the calcium aluminate
20 content.

21 Magnesium attack is particularly onerous
22 because what magnesium does is magnesium attacks the
23 C-S-H. It attacks the glue that's holding your
24 structure together. So what will happen is the
25 magnesium sulfate solutions will come in. They'll

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1 react (Microphone noise.) gypsum or you form --
2 Something's not right there. Yes, you form gypsum and
3 magnesium hydroxide and you'll get a double whammy
4 from the expansion of the magnesium hydroxide and the
5 loss of the glue that's holding your structure
6 together.

7 Delayed ettringite was a big phenomena
8 here a few years ago and I'll be very gentle here I
9 think. Basically we need to have ettringite in our
10 Portland cements. It controls that reaction rate with
11 tricalcium aluminate. So we have to have it formed.
12 But it's not stable above 65 degrees. So above 65
13 degrees, it decomposes and it decomposes, ettringite
14 will decompose to maybe hydrogarnet, depending again
15 on how hot it gets, some form of a hydrated calcium
16 silicate, either one of those four polymorphs, and
17 water.

18 So it sits in these forms and they're in
19 spacial or they're in a close spacial relationship
20 because they just fell apart. So if your concrete is
21 sitting here, it's gotten very warm. First of all,
22 you've deteriorated the microstructure and you're
23 going to have long-term problems with the loss of that
24 microstructure. But what will happen is if upon
25 rehydration water gets back into these, this is going

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1 to be generally taken out into the microstructure, if
2 water gets back into these what you do now is you have
3 a hardened structure and you're reconstituting the
4 ettringite and it expands and it cracks and it fails.

5 Tank closures aren't unique. Chris has
6 done two. She's going to do two more. How many
7 20,000 gallon tanks did you do?

8 DR. LANGTON: Oh a lot. Twenty-five.

9 PROFESSOR SHEETZ: Lots. You know they've
10 done the vault. We have experience within the
11 community. We have a broader experience in mass
12 concrete in the civil engineering community. We have
13 bridge piers. We have all kinds of structures. So
14 this isn't unique. But what is unique is that we have
15 all these damn things in there. We have to take that
16 into consideration. We have to be aware of it.

17 So size and geometry. We've talked about
18 this. We started to bump up against this, what we do
19 in the lab versus what we do in the field. We have to
20 be aware of that. I can make the best concrete in the
21 lab, but if I don't have my proper QA/QC in effect,
22 the engineer in the field will say, "Gee, what did he
23 know" and he's going to do his own thing and you get
24 a different product and then you now don't know the
25 performance.

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1 Do we know the scale factors for all of
2 the concerns that we have? No. Can we guess at them?
3 Yeah. We have, the engineering community has, ideas
4 on what to do. Geometry is a very, very interesting
5 question that we haven't talked about. Most of what
6 we've talked about and most of our knowledge on
7 cracking comes from lenticular slabs, lenticular
8 structures, not from mass concrete pores. We've done
9 a lot of these but we've done an awful lot more.
10 We've laid a lot more highway and bridge decks than we
11 have big massive structures.

12 So we've done bridge decks, parapets, foot
13 walls, slab grades. These are where we're getting a
14 lot of our data from, but we do know how to do the big
15 stuff. So if you were going to go and you were going
16 to do a 1.3 million gallon tank closure, who would you
17 go to? Take out a piece of paper and a pencil. This
18 is a quiz. Who would you go to? What component of
19 the engineering community out there is interested in
20 producing mass concrete that doesn't leak? Dam.
21 You'd go to the dam builders. Right? These people
22 are out there. We know we do this. These guys are
23 concerned about leakage. They know how to handle the
24 infiltrating water. They know how to handle the heat.
25 So we should engage them.

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1 Are there a lot of those guys out there?
2 We're not building dams anymore.

3 PARTICIPANT: Then you go to Brazil.

4 PROFESSOR SHEETZ: We go to Brazil. Go to
5 China. Maybe not. But the point is that we have a
6 body of knowledge to look at. So are there unknowns
7 and I'm here to tell you that there are and the
8 unknowns are this. We're going to use Portland
9 cement. It's going to hydrate and I'm using
10 tobermorite as the surrogate because it's easy because
11 it has stoichiometry. But this is the formula for
12 tobermorite and we can substitute crystal chemically
13 into that.

14 So David is going to be putting all kinds
15 of sluck from radioactive material into his waste
16 forms into this grout to sequester and it's going to
17 take up different mobile, labile species into the
18 structure and it's going to do it according to this.
19 Do we know what the properties are? No. Will there
20 be significant effects on the mechanical performance
21 of the concrete? Maybe. Maybe not. Has anybody
22 looked at building structures with this kind of
23 substitution into it? Yeah, but not nearly as
24 complete as what we're talking about and what we need.

25 Ettringite, we have to have ettringite

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1 into our cements. David talked about the uptake of
2 the hexavalent chrome, oh no, trivalent chrome into
3 the ettringite. These will all go into ettringite.
4 There are some of the nitrates that will go into
5 ettringite. Iodides will go into ettringite. In
6 fact, you can't put iodide into ettringite. It will
7 oxidize. It will take it to iodate going into
8 ettringite.

9 Do we know its performance and the
10 behavior of these? Not really. But let me give you
11 an example. Ettringite is a crystal with symmetry 6m
12 and 2m 2m. It's a dihexagonal dipyramid. It looks
13 like this and this is where the problem comes because
14 it elongated along the zed direction and if you have
15 a hardened concrete structure and this starts to grow,
16 this can put enough forces that it will exceed the
17 tensile strength of concrete and crack.

18 However, what we found in that Della Roy
19 project of '03 is if we substitute trivalent iron for
20 trivalent aluminum we change the morphology. The
21 morphology doesn't go elongate. It goes something
22 like this. We suppress the growth in the zed
23 direction. So I can get rid of ettringite degradation
24 just by raising the iron content in my cement and it
25 works. So this was an unintended consequence of that

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1 study, but it's the surprise and I'm presenting this
2 just to give you an idea of what we don't know. I
3 don't know what would happen to this morphology if we
4 substitute trivalent chrome into it. Maybe instead of
5 going from a clam to a needle, it goes somewhere in
6 between. There is some unknowns.

7 Let me wrap up. So there's a list of the
8 factors. I didn't even include wet/dry cycling in
9 here and efflorescent salts. So let me just quickly
10 go through this. Tricalcium silicate, we may not be
11 able to have any control over. Because if you're
12 buying cement at Savannah River, they're not going to
13 reburn the cement for you. So you have to live with
14 it.

15 The sulfate that sort of came and went,
16 same deal. May not be a problem.

17 Fineness, we can probably get them to
18 grind it for us the way we want it if we really
19 insist. If you tell them you're going to buy their
20 total year's production if they grind it the way you
21 want it, they'll knuckle under.

22 Alkali content, again you're not going to
23 have much control over. You're going to have to live
24 with it.

25 Low water to cement ratio, you can control

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1 it. You can engineer that.

2 Accelerating add mixtures you can
3 certainly engineer that. You don't care if this thing
4 hydrates in ten hours or if it hydrates in ten months
5 as long as it gets hard and as long as it gets 40 psi,
6 you don't care.

7 Long-term thermal cracking, that may be a
8 concern. How do we control that? Can we control
9 that? That's going to be one of the bigger concerns.

10 ASR, you can control that. You can
11 control that by the choice of aggregates that you use.
12 You can control it by the choice of supplemental
13 cementitious materials you use.

14 Sulfate attacks, this is going to be a
15 tank and it's a lot likely that you're going to get
16 external groundwater into this tank until that tank
17 dissolves.

18 Sulfate attack from magnesium, sulfate
19 attack from ettringite.

20 DEF may be a problem. If you can't
21 control the thermal behavior you may have future
22 susceptibility for the delayed ettringite attack. How
23 long those sleeper cells are going to remain in place
24 in there and be reactive is anybody's guess at this
25 point.

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1 Drying shrinkage is probably not going to
2 be a problem because you're going to place this in a
3 tank underground in a relatively constant temperature
4 and constant humidity. Plastic shrinkage, likewise.

5 Thermal contraction is a problem.

6 Freeze/thaw, if you do it and you do it
7 underground, it's probably not going to be a problem.

8 Corrosion, as long as you keep the pH up
9 it's not going to be a problem. I don't see really
10 that you're going to reinforce these tanks with
11 anything. So that's probably not a problem.

12 Static loading, cyclic loading, they may
13 be a problem. The only problem you get with
14 mechanical loading may come from earthquakes and
15 frankly I didn't look at any of that.

16 And internal and external restraint is I
17 think going to be your biggest problem

18 So if we look at these, what can we do?
19 That's going to go away. I just said that you really
20 don't have any control over that. The alkali content
21 you really don't have any control over. The long-term
22 contraction you may have, you may be able to handle.
23 So DEF, my guess is that DEF's not going to be a
24 concern.

25 So what we're left with is we're left with

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1 the thermal contraction that you have to be concerned
2 of once this thing heats up and then it's going to
3 swell because of heating and it's going to shrink when
4 it cools down and of course, the external/internal
5 restraints associated with the tank. These I believe
6 are going to be the two things that we really have to
7 be concerned about for closure of these large tanks in
8 order to minimize the cracking.

9 I believe we can control everything else.
10 I believe we can control these. I think it's going to
11 take a bit of effort to control this because of all of
12 the pipes and protrusions and things inside, but I
13 believe it's possible. They do it with dams. They
14 pump liquid nitrogen through cooling pipes in dams and
15 they pump water through it to cool the individual
16 components. If we really need to, we could probably
17 do that.

18 Everything you wanted to hear about
19 cracking.

20 VICE-CHAIRMAN CROFF: Thank you very much.
21 It was just a tad long here and I do want to adjourn
22 for lunch at 12:30 p.m. as planned because the
23 lunchtime is relatively short even at that. But we'll
24 try a few questions here. Jim.

25 MEMBER CLARKE: I had a question I wanted

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1 to ask before but we were running short on time on
2 that and we're hearing a lot of interesting stuff and
3 a lot of the pieces and I'm sitting here trying to put
4 the pieces together that both of your talks, I think,
5 were very helpful in identifying what we could be
6 concerned about and what we need to be concerned about
7 if those are any different.

8 The question I wanted to ask was about
9 accelerated testing that you mentioned, Christine, and
10 the question is if Rachel can give me 25 years of
11 monitoring data and I can keep that in an information
12 management system that we can all understand and I
13 think that's possible, what do we get out of the
14 accelerated testing? What tests are available and how
15 far do they accelerate? David showed us a model and
16 real data and after one year, there were two orders of
17 magnitude apart. So how can the accelerated testing
18 help us with this?

19 DR. LANGTON: I just identified the need
20 for accelerated testing.

21 MEMBER CLARKE: Okay.

22 DR. LANGTON: I didn't say what they were.

23 PROFESSOR SHEETZ: Accelerated testing is
24 obviously something that is of interest elsewhere than
25 just here. There are some procedures that are

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1 floating around the literature right now where you
2 raise -- I mean how do you accelerate any reaction.
3 You can do it three ways. You can add a catalyst, you
4 can increase the surface area or you can increase the
5 temperature.

6 DR. LANGTON: Or you can increase the
7 concentration.

8 PROFESSOR SHEETZ: Or we can increase the
9 concentration. So we can't increase the concentration
10 here. We're not adding catalysts. We can't grind
11 this thing up. We're left with increasing the
12 temperature. There is a procedure that is being
13 circulated in the literature where you would take your
14 ASTM protocols, let's say if you want to compressive
15 strength, and you cure it for seven days at room
16 temperature and then you take and you cure it for
17 another seven days at 38 degrees Centigrade. That
18 will give you about 3X acceleration in time and then
19 you go crack it and there's very good correlation that
20 that accelerated test works and you can just spend the
21 time and take the room temperatures for the period of
22 time and the values are pretty reasonable, I mean,
23 within the limit of error of the particular test
24 method. They will accelerate.

25 But you're only accelerating a factor of

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1 3X. If you go to 68, you accelerate by about, what,
2 7X or 8X. So what we need is a test that's going to
3 accelerate by orders of magnitude not by --

4 MEMBER CLARKE: Maybe, but I still think
5 the further out we go the better off we are and we're
6 not going to go to 10,000 years. So I'm not looking
7 for that, but I'm looking for ways to get monitoring
8 data and correlate it with model predictions and
9 improve the model and go through that cycle.

10 PROFESSOR SHEETZ: I believe we can
11 probably -- Some of that data is out there.

12 MEMBER CLARKE: And then I just keep
13 saying that because I don't see it being done.

14 DR. DETWILER: One concern you have though
15 when you accelerate phenomena is that you may
16 eventually get to different failure mechanisms than
17 you had or that would happen in real life or it may
18 just be that if you had multiple mechanisms already
19 occurring, that by your acceleration method you may
20 favor one over another and so that it becomes a poor
21 representation of what's going to happen.

22 So it's not simple and especially when
23 you're -- You know it's one thing if you're dealing
24 with just chemical reaction. But if you're also
25 dealing with things like diffusion and a number of

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1 physical phenomena at the same time that you may end
2 up with something that's very, very different. I
3 think that's where maybe using your accelerated test
4 to choose from among several formulations to decide
5 this is the best one and then making an effort to do
6 some of these long-term monitoring things so that you
7 can kind of verify your models and go back and see
8 does this make sense or do we need to adjust or
9 correct and then if we have the possibility --

10 I mean I've seen even in something like
11 when I was in my first job. I went out to a
12 precasting plant where the people who had started this
13 plant were very interested in investigating things.
14 They instrumented every member that they had. Their
15 plant was built of prestressed concrete and they had
16 instrumented absolutely everything so that they would
17 have long-term data and then later they could go back
18 and look and that kind of thing then if you did it in
19 a systematic way and you shared it you could provide
20 a lot of useful information for yourselves and other
21 people who were doing that same kind of thing.

22 MEMBER CLARKE: That's where I'm coming
23 from and help sorting out which because the complexity
24 is obvious and if you were focusing on one property,
25 the accelerated test would probably be extremely

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1 helpful, but sorting out which ones are the ones you
2 can focus on.

3 PROFESSOR SHEETZ: To supplement Rachel's
4 statement, what you're doing from the materials
5 standpoint, when you accelerate you have to stay
6 within the same compositional range, the same
7 mineralogical stability range when you're accelerating
8 and if you exceed that range and you go into the
9 stability range for another mineral phase in your
10 concrete, then all of a sudden all bets are off
11 because you're not really, you're accelerating it into
12 something that doesn't exist.

13 MEMBER CLARKE: I understand. In the
14 interest of time, thank you. Ruth.

15 MEMBER WEINER: Would you prefer that I
16 just wait, hold my questions till we have the
17 roundtable?

18 MEMBER CLARKE: Your choice. I'll give
19 you one.

20 MEMBER WEINER: Okay. They're quick
21 questions. But the first is, Dr. Detwiler, you
22 mentioned that you go out to 100 years. How about 500
23 to 600 which is 20 half lives of strontium-90 and
24 cesium-137? Do you think you could have structures
25 that could be predicted to retain something for that

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1 long?

2 DR. DETWILER: I'm just saying what our
3 experience is and that's well beyond our experience.
4 You could predict anything. I won't be around to see
5 it.

6 MEMBER WEINER: Nor will I.

7 DR. DETWILER: One job that I was on where
8 one of the geologists that was consulting on it said
9 and he was probably in his 60s already, he said, "I
10 don't care about 500 years. I only care about five
11 years." So we could say anything, but as far as do we
12 have a solid basis, no we do not.

13 MEMBER WEINER: The other question I have
14 is what about, you mentioned gamma degradation of
15 concrete, what about alpha degradation?

16 DR. DETWILER: I don't know of any
17 information about that.

18 MEMBER WEINER: Dr. Sheetz, do you have
19 any?

20 PROFESSOR SHEETZ: Alphas don't penetrate
21 very much. They'll just -- I mean they won't even
22 penetrate your skin.

23 MEMBER WEINER: Thank you.

24 CHAIR RYAN: I'll defer.

25 VICE-CHAIRMAN CROFF: Okay.

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1 MEMBER HINZE: Briefly. In the face of
2 this blizzard of information, I have taken away
3 several things and one is that there's concrete and
4 then there's concrete.

5 (Laughter.)

6 MEMBER HINZE: And the concrete that one
7 might use in Savannah River versus Hanford versus
8 Minnesota, you might wish to have considerably
9 different attributes. And one of the things I'm
10 taking away from this is that there is the possibility
11 of custom blending of concretes.

12 To focus it on the Hanford problems, for
13 example, you might want to change the rigidity of the
14 concrete because of the seismic hazard there, but you
15 don't have the problem associated with all of the
16 moisture that you have. So it seems to me that what
17 I'm taking away and tell me if I'm right or wrong is
18 that there is a lot of potential here for customizing
19 concretes to the particular environmental attributes
20 that you anticipate over a 10,000-year time period.

21 PROFESSOR SHEETZ: You are correct.
22 Concrete is an engineering substance.

23 DR. DETWILER: Basically even for things
24 that are much less complicated, we still design it.
25 It's basically custom designed for anything. So

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1 certainly with this, you would do the same. I mean if
2 you do that to make a box store, you have a different
3 mix design for the footings and for the columns and
4 for the slab on-grade. You could certainly and you
5 should do this in this kind of application, but you're
6 probably going to be looking at more sophisticated
7 criteria and criteria that not everybody deals with.
8 So there may be things where we really need to do some
9 very different kinds of testing, very different kinds
10 of investigations to come up with how best to meet
11 those criteria. But that's well within the purview of
12 what normal concrete engineering is about.

13 CHAIR RYAN: Bill, if I may just add on.
14 There's an important, I think, addition to your
15 thought which I think is right on target and that is
16 that if you can somehow do that tailoring of the
17 material and then take credit for it in a modeling and
18 performance sense and then add Dr. Clarke's comment
19 about if I can now figure out a way to monitor it over
20 some reasonable period of time and see that if it's
21 behaving in that environment as tailored, as
22 anticipated, now we have something where confidence is
23 a possibility.

24 MEMBER HINZE: (Inaudible.)

25 CHAIR RYAN: Absolutely. So I just wanted

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1 to throw that in.

2 MEMBER HINZE: I don't want to get at this
3 now because we don't have time, but one of the things
4 that perhaps we can take up in the final is the
5 direction and status of research in concretes and
6 Barry tangentially hit that.

7 VICE-CHAIRMAN CROFF: You'll hear a lot
8 this afternoon.

9 MEMBER HINZE: We will hear that. Okay.
10 Thank you.

11 VICE-CHAIRMAN CROFF: I think a couple of
12 points. First, I've taken away from two or three of
13 these talks that air is the enemy of cement. Cement
14 is not stable in front of the CO₂ in air in particular
15 and over time it will degrade, air and water, humid
16 air, which tells me air ingress is a problem and that
17 leads to a couple of thoughts and that is whether any
18 consideration has been given to let's say sealing a
19 tank or a saltstone vault or something like that,
20 sealing it to air. We think a lot about water
21 ingress, not so much about air, but I'm hearing that's
22 fairly important over the long term.

23 Secondly, we're talking about monitoring
24 and we're talking about penetrations. The
25 penetrations are avenues for air to get into this

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1 thing potentially and so I'm sort of seeing a
2 possibility of a tradeoff there. Any thoughts on
3 that?

4 PROFESSOR SHEETZ: Carbonation attack in
5 concretes in northeast is pretty rare. Would you
6 agree?

7 DR. DETWILER: Well, I think that of
8 course that's part of that and I think where people
9 really see lots and lots of carbonation is more in
10 Europe where there's a lot less cement in their
11 cement. After World War II, there was a real shortage
12 of cement because they were trying to rebuild. So
13 there was a tremendous demand and basically what they
14 did was they intergrounded a lot of unburned limestone
15 into the cement and so that was basically a dilution.
16 That's where you really saw a lot of carbonation.

17 But it is nothing like that bad when
18 you're talking about the amount of cement that we have
19 in ours. It's almost all clinker and we should have
20 enough residual in there to deal with that. So I
21 don't see it as a huge problem and remember that if
22 it's an underground tank you don't have that much
23 access to air. I mean the soil around it or the steel
24 while the steel is there and the soil around it, it's
25 not something where you have lots and lots of air and

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1 chances are some of these other things that you hope
2 are not in the soil like sulfates would definitely be
3 worse.

4 PROFESSOR SHEETZ: As long as the humidity
5 is up in the pore fluid, it's a non-problem and
6 nothing is independent of one another. But if the
7 pore fluid in that 50 to 100 percent range and it's
8 sealed off, there won't be any problems. If it's up
9 and there's carbon dioxide present, it still probably
10 won't be a problem.

11 We've looked at some structures from a
12 parking garage at Duquesne University. It's been in
13 25 years and did the phenolphthaleine test and you can
14 see some effect on the surface. It's not quite clear
15 if it's carbonation or if it's just rain and other
16 events washing out of the surface, but the measurement
17 on that is just a millimeter or two after 25 years.

18 DR. KOSSON: I think it's very important
19 that you differentiate between structural concretes
20 and waste forms because in waste forms you do see
21 significant carbonation and the outcome carbonation
22 often is in the 60 to 80 percent relative humidity
23 range and when you talk about these very long time
24 frames where you do have cyclic wetting and drying
25 conditions, then you may be talking slow processes

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1 relative to your 50 year duration of structural
2 concrete. But when you're starting talking hundreds
3 of years into a waste form which is not designed like
4 a structural concrete the carbonation ingress can be
5 rather significant.

6 PROFESSOR SHEETZ: Yes, the cyclic wetting
7 and drying we see efflorescence coming out of concrete
8 and that's limestone. That's calcium carbonate. So
9 you do have the potential of sealing your pores with
10 the wet/dry cycling as well.

11 DR. LANGTON: I have to emphasize that
12 when you're looking at the system -- Well first, waste
13 forms are not concrete and our waste form at Savannah
14 River, our saltstone waste form, already contains
15 carbonate as sodium carbonate salt. I can't say that
16 air is the nemesis of concrete. Air is not
17 necessarily bad. That's the wrong conclusion to come
18 to, air with carbonate.

19 The other thing to say is that monitoring
20 units of a landfill is one thing, but so far all I've
21 seen done is monitoring of the landfill. We're
22 monitoring well either in near field or someone more
23 distant, not too far away, is what's monitored and you
24 can wait hundreds of years before you see anything in
25 those wells unless something really catastrophic goes

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1 wrong. But what we need is monitoring of the
2 components that go into the landfill, the soil. Well,
3 even the soil is too far away to be affected when
4 we've have large --

5 MEMBER CLARKE: Christine, if I can
6 interrupt. The monitoring I was referring to is
7 placing access probes inside the cover over the
8 landfill and then monitoring -

9 DR. LANGTON: But we won't put the cover
10 on for years, for 20 or 100 years.

11 MEMBER CLARKE: As you close this
12 landfill.

13 VICE-CHAIRMAN CROFF: We need to terminate
14 this.

15 MEMBER CLARKE: I just wanted to make that
16 distinction. I agree with you on the --

17 VICE-CHAIRMAN CROFF: We can continue it
18 on later on this afternoon. I think we'll hear some
19 things that bear on it, but it's a very relevant
20 discussion. Let's come back at 1:20 p.m. if we can.
21 It's a quick lunch but thank you. Off the record.

22 (Whereupon, at 12:35 p.m., the above-
23 entitled matter recessed to reconvene at 1:24 p.m. the
24 same day.)

25 VICE CHAIRMAN CROFF: I'd like to come to

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1 order, if we could. We've got a number of interesting
2 things to do this afternoon, and time is fleeting. I
3 think we've got everybody here in the room.

4 All right. We're into the third session
5 that's to address the state of the art in predicting
6 long-term performance of cementitious materials.
7 We've got three speakers here, although I suspect many
8 of our former speakers are going to have more than a
9 couple of opinions as we get into the question and
10 answer later in the afternoon.

11 The first speaker in the session is
12 Professor Fred Glasser from Aberdeen University in the
13 United Kingdom. Professor Glasser received a Ph.D. in
14 Geochemistry from Penn State. In his many years of
15 service at the University of Aberdeen, he has directed
16 a group working on ceramics, glass, and cement, with
17 emphasis on fundamental and applied studies. He has
18 been working on radioactive waste since the late
19 1970s, mainly on application of cementing, but also on
20 longer term planning and repositories and their post-
21 closure performance.

22 Professor Glasser, take it away.

23 DR. GLASSER: Thank you, Chairman. It's
24 a pleasure to be here. I do have to say that almost
25 everything that I wanted to say has in fact been said

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1 by previous speakers.

2 (Laughter.)

3 But I'll try to go through that part
4 quickly. I think I have a few things that haven't
5 been said, and I'll try to make my oral comments
6 relate to some of the things that I think haven't been
7 said.

8 I'll briefly talk about the role of cement
9 in nuclear waste disposal, but I think I'm teaching my
10 grandmother to suck eggs here.

11 (Laughter.)

12 Resistance to degradation, mechanisms and
13 processes, some synergies with civil engineering and
14 civil engineering practice. I want to talk about what
15 we mean by "performance," what we mean by words like
16 "durability," to give you some data on solubility
17 studies, showing how cements perform in response to
18 different aggressive substances in the natural
19 environment, about the need to develop a new paradigm
20 and to implement it about testing and test methods,
21 some special hazards to cement studies in nuclear
22 applications, to cements and the source term about
23 which not much has been said, remediation activities,
24 and, finally, conclude with a brief summary.

25 By "cement" -- and I'm going to talk about

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1 cement. When I do venture into mortars, grouts, and
2 concretes, I'll try to use the appropriate correct
3 term, but mainly the focus of durability studies is on
4 the cement. It's the matrix that binds other
5 materials together. And when that matrix is
6 destroyed, the functionality of the resulting
7 material, be it mortar or concrete, is destroyed.

8 Now, cement is a standard product. It's
9 almost the same the world over. The detailed
10 specifications in the United States, ASTM
11 specifications, are mirrored very closely by worldwide
12 practice, not surprising perhaps when you think that
13 the bigger cement producers operate on a worldwide
14 scale. The U.S. is one of the few places in the world
15 where there's still lots of independent companies, but
16 increasingly the market is being dominated by the big
17 international players.

18 There are codes of practice that govern
19 the composition of both cements and also blended
20 cement specifications, and they are important.
21 They're treated second only in importance to the Bible
22 by civil engineers who work with cements and
23 concretes. And you find that if you want to make
24 changes you do so at your own risk.

25 In nuclear waste disposal, cements and

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1 concretes have a long history of use. Obviously, it's
2 primary matrix material for solidification and
3 stabilization of liquids, sludges, or particulates.
4 Because of their relatively high physical density and
5 the possibility of including aggregates, they give
6 good shielding for personnel and good protection
7 against the exigencies of storage and transport.

8 They can be used in repository
9 construction, both as liners and seals. Sometimes
10 they're necessary to permit retrievability, which may
11 be a requirement. But although cements are relatively
12 similar in specification, they can be modified by
13 adding supplementary materials as well as, of course,
14 coarser aggregates. And we'll have course to deal
15 with some of those in the not-too-distant future.

16 Now, cements are perhaps unique in respect
17 of other barrier materials. Whereas other barrier
18 materials, such as steel or glass or clay have
19 primarily a physical function to play, cements also
20 exhibit chemical activity, so it's not directly
21 comparable with other barrier materials -- for
22 example, metals that have a mainly physical role to
23 play in the immobilization process, although as we
24 will see there are certain factors in common between
25 the deterioration of cements and those of metals.

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1 This chemical conditioning role is always
2 present and has both positive and negative
3 implications for performance. For example, portland
4 cements will react strongly with glass. They
5 condition a high pH, and this is detrimental to the
6 longevity and durability of glass wasteforms. And so
7 it's important that the two not be mixed, or if they
8 are mixed that there are special circumstances which
9 will permit effective isolation of one from the other.

10 Now, we've talked about the change in
11 cement, and many people will question whether this is
12 inevitable. And I think the answer is yes. I think
13 almost all environments that I can think of in which
14 cement and concrete will be used, both in nuclear
15 engineering and in civil engineering, they are in fact
16 thermodynamically stable -- unstable, sorry.
17 Therefore, they will change by reaction with their
18 environment. So this is inevitable.

19 What we cannot always predict is the
20 consequences of a reaction with the environment, which
21 range widely, and we also cannot readily predict the
22 rate at which these changes will occur. So a purpose
23 of this meeting is to define and, if possible,
24 quantify these changes, and I will try to stick,
25 whenever possible, to generic considerations in

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1 presenting material to you.

2 I'm not going to present a vast amount of
3 factual material. I don't want people to go on to
4 overload. So the examples that I'll offer I hope are
5 selective and easy to understand, although not always
6 comprehensive.

7 Well, if we compare resistance to
8 degradation of different materials, I have the feeling
9 that many scientists are more comfortable with the
10 degradation of metals than they are with the
11 degradation of cement. Perhaps if I call it corrosion
12 of metals, that will strike home exactly what I mean.

13 Now, there are differences between the
14 behavior of corrosion of cement and corrosion of
15 metals. Metallic corrosion clearly involves oxidation
16 and with it electron transport, whereas cement
17 corrosion does not generally involve oxidation
18 reduction, although I would make an exception for slag
19 cements and, of course, the materials that are
20 embedded within cements.

21 But certain features are common to both.
22 For example, we have the formation in some regimes of
23 soluble corrosion products, and we -- and insoluble
24 corrosion products. Now, in metallic corrosion, this
25 gives rise to regimes of active corrosion where the

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1 products are soluble, and passivation where the
2 products are insoluble and tend to accumulate at the
3 surface of the metal undergoing change.

4 So we can see this analogy in the next
5 slide. I've shown here on the left a metal which is
6 partly active, soluble products are forming and going
7 off into the aqueous phase, but there are also an
8 accumulating layer of insoluble products of corrosion,
9 the substrate metal being -- ah, yes, the substrate
10 metal being, well, I think -- yes, the substrate metal
11 being to the left and insoluble products of corrosion
12 accumulating as well as some soluble products.

13 Now, this is mirrored by cement, which can
14 also have these different regimes of corrosion. Here
15 the aqueous solution is on the left. The unaffected
16 cement is to the right, and we have insoluble products
17 of corrosion accumulating at the surface as well as
18 solid products going off into solution.

19 But a difference between metals and
20 cements is that in aqueous solution water and salts
21 that might be present in the aqueous solution can
22 migrate into the rather more permeable and porous
23 cement matrix. So alteration indicated by the hatched
24 lines may extend to greater depth within the cement,
25 whereas with the metal we generally see a clean

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1 contact, an interface, although not necessarily as
2 geometrically perfect as I have shown it.

3 So the strength of this interaction
4 between cement solids and water, or species that may
5 be dissolved in water, or with atmospheric
6 constituents varies greatly. As with metallic
7 corrosion, there is a need to preserve local
8 electrostatic charge balances.

9 So when we measured diffusion coefficients
10 of species in and -- in and out of cement, we are
11 measuring apparent diffusion coefficients, because the
12 driving force may be partly dictated by the need to
13 maintain local electrostatic balances to maintain the
14 ionic potential of the pervading aqueous phase, and so
15 on.

16 So it also is a difference between metals
17 and cements that because these matrix diffusions occur
18 in cements that there is a quality factor associated
19 with the diffusion. It's a complex function of the
20 matrix formulation -- for example, how much porosity
21 there is and how interconnected this porosity is to
22 give rise to permeation. It's a function of age of
23 the material, and it's also a function of the thermal
24 history of the cement, mortar, or concrete.

25 Now, this quality factor has been much

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1 studied by engineers. You heard a lot about the
2 influence of water cement ratio, for example, in
3 previous talks. And, clearly, engineers have been
4 looking for decades now for specific measurable
5 features to -- with which to quantify the cement that
6 they can introduce into equations relating to
7 degradation in order to define the rate at which
8 degradation occurs and its consequences.

9 Now, these relationships are almost
10 entirely empirical. Again, we've had descriptions of
11 some of them today. And the problem with these
12 empirical relationships -- and this is one of the nubs
13 of the problem -- is that although they are cherished
14 by engineers, protected, beloved by engineers, they
15 don't actually yield a predictive capability.

16 So that the questions that have been asked
17 today over and over again, how do we -- but how do we
18 -- all right. You've told us what happens in five
19 years, but what happens in 500? We can't answer that
20 question, by and large, from the existing body of
21 knowledge. And that's a problem that we'll have to
22 live with, and so I'll be talking later about new
23 paradigms.

24 So given the long history of civil
25 engineering, and the scientific study of cement and

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1 concrete, was begun well over a hundred years ago,
2 famous names like LeChatelier, and so on, were very
3 active, and Henry Sorbe, the discoverer of the
4 petrographic microscope, these people very active in
5 elucidating the constitution and deterioration of
6 cement.

7 And one can understand, given the times,
8 the qualitative nature of their approach. So although
9 it might be expected that quantitative models now
10 exist, this is not so, and there are a variety of
11 reasons for this, some of which are valid, but some of
12 which are -- really fall in the class of excuses.

13 So just to recap our durability standards
14 in civil engineering, now, these stand like a great
15 monolithic body. And if you're going to do anything
16 new or different, you run into this obstruction on the
17 highway of progress. The standards are legally
18 binding, and they are prescriptive. And no self-
19 respecting engineer will go against them, because his
20 or her professional reputation is riding on the way in
21 which they implement these.

22 So this experience of test standards,
23 which themselves are often the products of experience,
24 do have a lot to teach us. I'm not dismissing this
25 body of wisdom. It has been acquired, much of it, in

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1 a hard way, and much of it is very useful. But we
2 have another problem to overcome, and that is that
3 cements and concretes are often made on the job at
4 site, and they're not necessarily subject to the same
5 rigorous quality assurance that you get if you bought,
6 say, a steel column or eyebeam.

7 Another problem that we'll have to face is
8 the extreme complexity of natural environments. Even
9 steady-state environments are very variable from one
10 spot to another on the face of the earth, the
11 atmosphere, the oceans, the groundwaters, the earth.

12 Temperatures vary, and there are physical
13 processes as well, which affect the durability and
14 performance of cement, the phenomena like Freestar has
15 been menticned earlier, and they create a virtually
16 infinite spectrum of service conditions that require
17 to be tested. And, of course, the tests have not been
18 done for this variety -- or infinite variety of
19 conditions, simply because there's not infinite
20 manpower and there's not infinite time.

21 So having said that, engineers fall back
22 on a few simple measurable parameters -- for example,
23 compressive strength. Again, we've heard that
24 mentioned, and it is, after all, for material which is
25 intended for use by engineers in structural. It's

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1 hardly surprising that compressive strength is a
2 strong arbiter of your success in creating a proper
3 structure, proper material. But as we'll see -- as
4 we've seen in the nuclear field, compressive strength
5 is not such an important parameter in many
6 circumstances, not all but many.

7 All right. Now, I've been using words
8 like "performance" and "durability," and I haven't
9 defined them. And, indeed, I haven't heard them
10 defined earlier today, and the reason for that is that
11 some of these words like "performance" and
12 "durability" are themselves rather abstract words,
13 like patriotism or love or whatever. They're abstract
14 words.

15 And the only way which you can define them
16 is through separate attributes like, did I remember my
17 wife's birthday?

18 (Laughter.)

19 No, for us the attributes may be a bit
20 more complicated, and you may be able to express them
21 numerically. And I think engineers have been good
22 about realizing this. This overhead is borrowed from
23 Professor Geyorv in Norway. And he would say that we
24 -- we define performance or durability in terms of
25 separate attributes.

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1 And we plot a numerical value of the
2 selected property. The selected property has to be
3 one that is capable of quantification, and we plot
4 this as a function of time. I'm sorry, that has gone
5 missing off the lower axis here. This is time in this
6 direction, possibly log time as you will. And we note
7 an interesting feature about cements and concretes --
8 that after they are made they oftentimes improve in
9 quality for quite a while.

10 I don't want you to be totally negative
11 about cements and concretes. They oftentimes, with
12 maturation, actually improve in properties for some
13 time. But eventually, the value of the selected
14 parameter declines, and we agree some lower limit of
15 that parameter -- and that is the time to failure. A
16 very simple concept, but a very useful one that
17 enables you to relate words like "performance" and
18 "durability" to actual measurable quantities.

19 All right. Let's change subjects slightly
20 now and look at some conditions in which cement
21 performs well and some conditions in which cement
22 doesn't perform so well.

23 Now, my first example is to look at the
24 durability of cement as modeled by taking a very
25 simple substance -- calcium hydroxide -- and we know

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1 that one of the reasons why cement fails to perform
2 over the long term is it dissolves. It's somewhat
3 soluble in water, and so dissolution is an important
4 reason for its failure.

5 Now, here on the left-hand side I've
6 plotted the solubility of calcium hydroxide in
7 millimoles at equilibrium. And you'll see that the
8 solubility of calcium hydroxide actually decreases
9 with rising temperature. So if you're looking at the
10 impact of temperature alone, in the range up to 85
11 degrees, the solubility decreases by not quite half,
12 from 20 millimoles down to 12.8.

13 This decrease in solubility actually
14 carries on up to about 180 where the solubility curve
15 then turns positive again. So this is unusual
16 behavior, because relatively few substances have
17 decreasing solubility with rising temperature.

18 Now, if we want to look at the impact of
19 sodium chloride, a common constituent of many
20 groundwaters and, of course, of the oceans -- and I've
21 marked for reference the approximate seawater
22 concentration sodium chloride, just a little bit less
23 than a half-molar, you can see that -- and I've
24 extended data points up to 1.5 molar sodium chloride,
25 a little more than three times seawater concentration,

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1 you can see that there's a considerable increase in
2 solubility.

3 But taking into account the overall scheme
4 of things, not a huge increase, going to .5 molar
5 sodium chloride. So the solubility at 25 degrees
6 increases from 20.1 to 27.7 millimolar, and at 55 from
7 14.4 to 21.9, and so on. There is still the same
8 tendency with rising temperatures for falling
9 solubility, but the effect is about the same
10 percentage-wise as it is in distilled -- initially
11 distilled water.

12 But you see that if I go on to higher
13 sodium chloride concentrations, the solubility is
14 really very little affected. So the question about
15 whether cements will perform well in brine is answered
16 for you -- that certainly with respect to the
17 dissolution they will not be significantly more
18 affected by sodium chloride concentrations up to
19 1.5 molar. And, indeed, they will dissolve less at
20 high temperatures than they will at low temperatures.

21 Now, I haven't put the data for pH onto
22 this diagram, but because the solubility of calcium
23 hydroxide is not greatly affected, and because calcium
24 hydroxide continues to ionize in sodium chloride
25 solution, the pH is -- at 25 degrees is virtually

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1 unchanged.

2 Now, it's true that the pH decreases at
3 high temperature, but that's because the whole nature
4 of the pH scale itself is temperature-dependent. And
5 I don't want to get on to that in this discussion.
6 We'll be bogged -- we'll be thoroughly bogged down.
7 The ion product of water changes with temperature, so
8 the basis of the pH scale changes.

9 But on a relative basis, at any conditions
10 on this grid, the pH under comparable conditions, i.e.
11 same temperature, the pH remains about 12.4, something
12 like that, plus or minus .2 unit.

13 So these are conditions under which cement
14 will perform well. Well, I've got the same data for
15 CSH, which is -- makes -- the gel phase, which makes
16 up 80 percent of cement. But the same lesson is true
17 here as from the previous diagram. The trends are
18 exactly the same, so I'll pass over this.

19 Now, the role of carbon dioxide has been
20 touched on by almost all of the speakers today.
21 Rainwater is saturated with respect to atmospheric
22 partial pressure of CO₂, about 10^{-3.5} atmosphere at sea
23 level. And when this water contacts cement,
24 especially where the water can be sucked into pores in
25 the cement, most of the CO₂ dissolved in rainwater

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1 will react with carbonation of cement and the reaction
2 is as was depicted earlier.

3 So moist atmospheres carbonate cement, and
4 all of the cement phases will react with formation of
5 calcite, silica gel, alumina, and ferric oxide
6 hydrates. And the rate of carbonation is quite
7 dependent on relative humidity. The CO₂ has to
8 dissolve in a film of water in order for it to react
9 with cement. So humidities lower than 50 percent by
10 and large don't generate this film of water.

11 At humidities above 80 or 85 percent
12 approximately, water condenses, the pores are filled
13 with water, and so the surface area available for
14 transport of CO₂ from the atmosphere to water
15 decreases. The geometric surface area remains the
16 same, but the pore surfaces get filled, and so the
17 rate of carbonation decreases.

18 For reasonable quality concrete, the rate
19 of carbonation in normal air -- and I realize this
20 varies with exposure. For example, north sides of
21 buildings tend to carbonate faster than the south
22 side, because they don't experience so much solar
23 insolation. Depending on exposure, carbonation rates
24 are sort of in the range .2 to 2 millimeters per year.

25 Now, the interesting thing about this

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1 carbonation process is that the strength is not
2 impaired. Indeed, in the past, possibly in the
3 future, there has been quite a productive industry of
4 taking fresh cement products, like brick and block,
5 and so on, and letting them carbonate in flu gas. Not
6 only does this help pay back some of the CO₂ penalty
7 that you've incurred by making cement, but for
8 relatively porous and permeable objects they are
9 actually strengthened by allowing carbonation to
10 proceed.

11 So it's not necessarily a bad process to
12 happen in cements, although I'll show in some
13 circumstances it is harmful. But, of course, the
14 problem arises in that most cements are used as
15 composite materials, not only in the form of concrete
16 but also in the form of steel reinforced concrete.
17 And there, of course, if the pH decreases, the
18 passivation that these objects enjoy in the high pH
19 regime of cements is lessened or eliminated and
20 corrosion tends to be much more rapid.

21 Now, because calcium carbonate is several
22 orders of magnitude less soluble than calcium
23 hydroxide, or CSH, it tends to form a protective
24 carbonate skin in or on the near surface layers of
25 concrete. And this is the passivating film that I was

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1 referring to earlier, and it really does have a
2 remarkable effect on improving and promoting the
3 durability of constructional materials.

4 The medieval cathedral builders in Europe
5 quickly learned by the 12th and 13th century not to
6 make their lime for setting brick and stone from pure
7 limestone, but to use impure limestones containing
8 alumina and silica. So it was easier to build up a
9 protective layer containing amorphous alumina,
10 amorphous silica, as well as calcium carbonate. The
11 properties of that layer were much more durable.

12 So, in general, with models of cement and
13 concrete performance in the atmosphere, which rely on
14 solubility in initially pure water, will tend to
15 overpredict the importance of dissolution. So we
16 don't find concrete bridge decks, for example, slowly
17 dissolving away. They may wear away through abrasion,
18 and solubility will contribute but only to a minor
19 extent.

20 So rapid carbonation of these near surface
21 layers will result in about a two order of magnitude
22 decrease in solubility with the result that lime is --
23 particularly impure lime is a successful construction
24 material, whereas gypsum, which has about the same
25 solubility as lime, is not a successful construction

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1 material when exposed to rain or drip or whatever.

2 The only ones who have built successfully
3 in gypsum were the ancient Egyptians who used it for
4 facing pyramids that were built in the Nubian Desert
5 where rainfall is typically a few millimeters a year.
6 There it's successful, but otherwise not, because, of
7 course, gypsum doesn't develop this self-protective
8 skin on it.

9 Now, seawater contains a certain amount of
10 dissolved carbon dioxide. Obviously, it picks up CO₂
11 from the free surface, the contact with the air, the
12 waves, and so on. You get a lot of aeration at the
13 surface. But on the other hand, there are organisms
14 living in the ocean that very effectively extract
15 carbon dioxide as carbonate from seawater and use it
16 to construct their own homes, i.e. mollusks and snails
17 and things of that sort.

18 And so the result is that the Henry's Law
19 considerations that you might think would apply to the
20 concentration of CO₂ in seawater don't really apply,
21 because, first, the only place that seawater can pick
22 up fresh CO₂ is at the surface, by and large. And,
23 secondly, that CO₂ at greater depth is scavenged by
24 shell-building organisms. So the actual effective CO₂
25 pressure in seas is somewhat less than atmospheric

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1 saturation, not much but somewhat.

2 Now, natural waters are a different case
3 entirely, because in natural waters the rainwater
4 filters through a biolayer in soil where CO₂ is an
5 important metabolic product from -- coming from
6 microorganisms. So the result is that groundwaters
7 may start out life where water permeates the soil and
8 hits the subsoil -- effective CO₂ pressure in those
9 waters may already be 10 to 100 times higher than in
10 rainwater.

11 Now, some of the CO₂ dissolved in
12 groundwaters can be remarkably aggressive to cement
13 and concrete. These aggressive conditions arise in
14 several sets of circumstances, mainly chemically when
15 there is more CO₂ than can be effectively combined
16 with calcium as calcium bicarbonate, but also where
17 the water is rendered acidic by passage through peat
18 or other types of bioaccumulation.

19 There are special circumstances, of
20 course, like mine drainage, but I won't go into those.
21 So there are both empirical approaches to this
22 question of CO₂ aggressivity, and there's also a
23 mathematical model which we published in Advances in
24 Cement Research some years ago. And we also
25 demonstrated that computer-based interactions handle

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1 these calculations well, although, having said that,
2 1992 was a long time ago, and we probably ought to
3 revisit this.

4 So summing up on CO₂, CO₂ can be neutral
5 in passivating in some circumstances, but it can also
6 be aggressive and cause rapid dissolution in cement
7 components in others. So we mustn't stick labels on
8 these substances. We mustn't say CO₂ is not
9 aggressive or CO₂ is aggressive. It all depends on
10 the physical chemistry of the situation that you
11 faced.

12 Right. Having criticized existing test
13 methods, and having said that these are to varying
14 degrees inaccurate or inadequate, and let me give an
15 example of this and talk about sulfate resistance. I
16 don't want to condemn ASTM, so I've included our
17 Canadian cousins in this.

18 (Laughter.)

19 But there's also a draft European standard
20 on sulfate resistance, which will probably be
21 published towards the end of this year, which is very
22 similar in spirit to the ASTM. Naturally, being
23 European, it can't be exactly the same as the U.S.
24 standard, but there you are.

25 Now, what one does to test the sulfate

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1 resistance of cement under the ASTM specifications is
2 to make up prisms or cylinders of a known composition,
3 and you immerse them in dilute sodium sulfate. Now,
4 the first question is: well, why sodium sulfate? If
5 you look at groundwater now, see, sodium is a common
6 constituent in groundwaters. And so, too, is sulfate,
7 but almost always balanced by a whole lot of other
8 cations and anions.

9 So I don't know of any groundwaters
10 anywhere in the world which are predominantly sodium
11 sulfate. So why sodium sulfate? Why have they
12 selected this? Well, the answer is -- and I've talked
13 to some old timers about this -- and they say, "We
14 didn't want to use sulfuric acid. We knew that was a
15 non-starter." It's like testing the resistance of
16 sugar cubes to putting -- immersion in pure water. We
17 know what the outcome is. You don't have to do it.
18 They'll all fail.

19 What we wanted is to combine the sulfate
20 into a salt which was neutral or near neutral in terms
21 of pH, but also one in which the cation did not
22 interact strongly with cement, so we chose sodium
23 sulfate. Well, you have to start out, then, in the
24 knowledge that these tests are unrealistic with
25 respect to the composition of almost any natural

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1 water.

2 All right. You say, "Okay. We'll do
3 these tests." Well, what you then do is to measure
4 the -- because there's usually an expansion in sodium
5 sulfate, you measure length of your prisms or
6 cylinders, and you measure the compressive strength as
7 a function of time at a fixed temperature, typically
8 up to one year.

9 Now, because it is difficult to make
10 cement cylinders or cubes reproducibly, you need a
11 large number of specimens to get statistical
12 reproducibility. And because you're going to sample
13 at perhaps different temperatures and perhaps
14 different times, the number of samples rapidly grows
15 extremely large.

16 So if you visit a typical testing center
17 using these engineering protocols, you will see great
18 big tanks known as swimming pools, which are filled up
19 with cubes and cylinders and what not, and every so
20 often they're taken out and wiped with a paper towel
21 and weighed and measured for length and then put back
22 to continue the test.

23 So obviously we're not going to get a huge
24 variety of formulations, and we're not going to get a
25 huge variety of temperatures, and we're not going to

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1 get a huge variety of sodium sulfate concentrations.
2 And we're certainly not going to be able to pick up
3 mixtures with other salts.

4 Now, besides what I've said, what's the
5 matter with these tests? Well, first off, because the
6 cement samples absorb sulfate out of the solution, you
7 may have started out with a precise concentration of
8 sodium sulfate, and it usually is prescribed in the
9 test method you're following. The sulfate
10 concentration will decrease with time.

11 So if your test has lasted three months,
12 six months, or a year, what is the effective sulfate
13 concentration in the life of the test? Is it what it
14 was at the start? Is it what it was at the finish?
15 Or is it what it was in between? I don't know the
16 answer.

17 Sodium, on the other hand, is not
18 depleted. Because it's a weakly interacting cation,
19 it stays behind. Now, you can't have sodium in
20 solution on its own. It's a positive charged ion. It
21 requires some negative charge. So what happens, well,
22 it takes hydroxide from the cement. That's the one
23 soluble anion which the cement can contribute. So,
24 bingo, you've got sodium and hydroxide ions in
25 solution, and the pH goes up.

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1 And in many of these simulations, if the
2 sample is put into a sealed container, the pH will
3 rise perhaps as high as 14. So have you seen the
4 mineralogical changes that would occur in real cement?
5 Because the pH is now 1.5 orders of magnitude higher
6 than what it would be in the real cement. Well, you
7 don't know.

8 Moreover, the situation is compounded by
9 the fact that most tests do not require you to do the
10 test in a sealed container. And when the container is
11 left open to the atmosphere, as the pH goes up, the
12 solution absorbs atmospheric CO₂ very rapidly. The
13 kinetics of CO₂ absorption from the atmosphere
14 increase with increasing pH, and the equilibrium
15 concentration will increase with increasing pH.

16 So lo and behold, that carbonate that now
17 appears in the solution reacts with the cement to
18 precipitate calcium carbonate. And what's worse, the
19 point at which precipitation occurs is not necessarily
20 at the surface of the cement. You may get cement
21 substance dissolving, going to near the surface where
22 the CO₂ concentration is being increased and refreshed
23 by absorption, and then it precipitates like a gentle
24 rain of calcium carbonate.

25 So you've coupled a new leaching mechanism

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1 for removing calcium from the cement that you did not
2 anticipate in the course of the test.

3 Well, I think I've told you enough to
4 convince you that the sodium sulfate tests are not
5 only extremely variable depending on the size of the
6 container, the surface area available for contact with
7 the atmosphere, and a host of other factors that are
8 not specified in the test to indicate why you get such
9 large interlaboratory variations. And even more
10 important with respect to present cases, why these
11 tests don't have any predictive ability.

12 Well, I've run ahead of myself. Well, we
13 have looked at the physical chemistry of these tests,
14 and we will be reporting to RILEM, the International
15 Pre-Standards Committee on Cement Testing, in Quebec
16 in September. So there will be a paper from us giving
17 chapter and verse and numerical calculations related
18 to these.

19 Well, just coming back to this depletion
20 of sulfate, with time you generally come down to the
21 -- at the end of the test, the depletion of sulfate
22 stops, because you get down to the solubility that's
23 controlled by gypsum, not the sodium sulfate, the
24 solubility of gypsum at the relevant pH.

25 Right. Now, magnesium sulfate is an

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1 interesting case, because here you have a cation and
2 an anion that both react strongly with cement. And
3 testing with magnesium sulfate is -- which is a
4 standard test method is fraught with problems, because
5 if you don't watch the solution concentration and
6 monitor it continuously, and that's not generally a
7 requirement of the test, you will get virtually
8 quantitative removal of magnesium from the solution.

9 The equilibrium solubility of magnesium
10 hydroxide at pH 12.5 is on the order of 10^{-7} molar,
11 which means you will need a very sensitive analytical
12 technique to detect any remaining magnesium in the
13 solution.

14 So I hope I've given you enough evidence
15 to convince you that standard tests and test methods
16 are not the way to go if we're to set standards for
17 cement durability and performance in the nuclear waste
18 field.

19 I'm not suggesting that you ignore this
20 body of knowledge. It represents experience in its
21 identification of destructive agents and the mechanism
22 of destruction, and it is going to be legally binding
23 that the findings of those investigations are paid
24 good and careful attention by any of the civil
25 engineers you employ on design.

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1 But it does not measure, I assert,
2 reliability or quantitatively what it purports to do.
3 So these tests are really in need of refreshment. We
4 need supplementary calculations about how they work.
5 We should analyze them from the physiochemical point
6 of view, and focus them better if we're to extract any
7 useful information from them.

8 Now, I want to say a little bit about
9 thermal hazards to cements and concretes. People have
10 mentioned delayed echingite formation in other words,
11 and much of the problem arises -- and it's going to be
12 particularly a hazard in nuclear waste immobilization
13 -- where large masses of cement-rich formulations are
14 used -- is the thermal excursion that results in the
15 first few days or weeks of cement hydration.

16 It really is strongly exothermic. If you
17 take a standard barrel, U.S. barrel, and fill it with
18 a fresh mixture of cement and water in appropriate
19 proportions, and keep a thermocouple in the center,
20 you'll quick find the centerline temperatures will
21 reach the boiling point, and cement won't set properly
22 because steam will be evolved before the cement has
23 had a chance to harden, not at the outside but at the
24 inside.

25 And this -- the resulting cooling in the

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1 post-hardening stage of course then results in a
2 thermal contraction to -- and then the result is
3 cracking, and usually not microcracking but usually
4 macrocracking. And, you know, we heard a lot this
5 morning about microcracking, but I don't consider
6 microcracking too serious a hazard to cement
7 performance, because, first, the microcracks may not
8 go right through the cement. And, secondly, the
9 transport properties of torturous microcracks are not
10 very good, but macrocracking is certainly at all costs
11 to be avoided. You undo all the good that you've
12 done.

13 So this has to be managed by attention to
14 formulation design, sequential emplacement, and that
15 entails problems. It's not free from problems and
16 junction boundaries. But certainly the importance of
17 thermal cracking on transport properties is, in my
18 view, a neglected area of study and one which everyone
19 needs to take into heart.

20 Now, there's also a situation that could
21 arise, and that is that in some situations we may have
22 heating as a result of discharge of heat from waste.
23 I'm not aware that cements themselves will be used as
24 a primary immobilization matrix for highly active,
25 heat-generating waste, but the repository itself may

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1 be lined with concrete, the integrity of which is
2 important.

3 And in the post-closure phase, we may get
4 hazards to the concrete which include crystallization,
5 if it's a concrete reaction between the cement and the
6 aggregate. Now, we heard this morning about inert
7 aggregates and reactive aggregates, but you must
8 remember that with the exception of calcium carbonate
9 aggregate no mineral aggregate is inert in the
10 equilibrium sense in contact with cement.

11 No mineral aggregate is inert -- granites,
12 schists, gneisses, sandstones, shales. All are
13 potentially reactive with cement. You only get away
14 with this in ordinary construction, because you don't
15 raise the temperature. But when you have a prolonged
16 thermal excursion you must expect reaction to occur in
17 the course of time. If you're worried about it, you
18 need to specify limestone aggregate concretes.

19 So we can consider briefly the
20 consequences to mineralogy pH conditioning ability,
21 resistance to dissolution, strength permeability, and
22 dimensional stability.

23 Now, just to remind you briefly that 90
24 percent of cements are two solids. It's calcium
25 hydroxide and there's this gel-like amorphous phase

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1 cryptically known as CSH, shorthand for calcium oxide,
2 silica oxide, water.

3 Now, the CSH is nearly amorphous to X-
4 rays, and although it doesn't necessarily have a fixed
5 composition, in cement where it coexists with calcium
6 hydroxide it is effectively calcium saturated, which
7 means it has a molar calcium to silica ratio of about
8 1.8 to 2.0.

9 Now, we do in industry do a lot of work
10 with autoclave cements. And to cut a long story
11 short, we know that if you autoclave unmodified
12 cements -- that is, they're not chemically modified by
13 adding any other material -- they will emerge very
14 porous and very weak, because at high temperatures the
15 CSH phase crystallizes and the products of
16 crystallization are denser than the CSH itself.

17 So the physical dimensions, exterior
18 dimensions of a shape, remain the same, but the volume
19 change is accommodated by increasing the porosity and
20 with it the permeation of -- the permeability.

21 So that's not a good way to go, but we
22 know that in industry what's done is to add around 50
23 mole percent of finely ground quartz silica. And
24 then, when you autoclave you shift the bulk
25 composition from the molar calcium to silica ratios of

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1 portent cement, which is typically about 2.7, 2.8 for
2 most industrial products, you shift it down to about
3 somewhere between 1 and 1.5, which means adding quite
4 a lot of silica to it, and then you get
5 crystallization phases like tobermorite, for example,
6 with a calcium silica ratio of .87.

7 And the density of tobermorite is very
8 similar to that of the mixture of CSH calcium
9 hydroxide in quartz that you started with, and the
10 result is that tobermorite autoclave products have good
11 strength and low permeation. So we know some of the
12 answers from industrial practice about what happens
13 when you heat cements.

14 Now, the question is: what happens if a
15 cement is used in a repository, experiences a
16 prolonged thermal excursion, and then cools down
17 again. What will its pH conditioning ability be?
18 Will be it permanently affected, or will it not? And
19 I've simplified a phase -- complex phased diagram for
20 a lime silica water system only to show you the range
21 of compositions bounded by yellow that will still
22 contain, at the high temperature and upon cooling back
23 down to room temperature, calcium hydroxide.

24 Now, commercial cements have a ratio about
25 here. So you can see that after heating, while they

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1 might crystalize to have a brandite or jaffyite, I'm
2 assuming no silicious aggregate is present -- that
3 when they're cooled back down portandite calcium
4 hydroxide will remain continuously stable throughout
5 this region bounded by a ratio of 1.5 and stepwise
6 function of temperatures close to 160.

7 So I conclude from this that all cements,
8 provided they're not reactive with silica or aluminum
9 materials, will, after prolong thermal excursion, come
10 back down to room temperature and still have a good
11 reserve of calcium hydroxide left with which to
12 condition the pH.

13 Right. I think I want now to come to some
14 conclusions. I won't say anything about the source
15 term, except to remark that I'm very surprised that
16 after decades of study of the interaction between
17 cements and radioactive waste species that we don't
18 have a library or an agreed dictionary of
19 solubilities.

20 We don't know the nature of the
21 solubility-limiting substance that represents the form
22 of binding between cements and radionuclides. We
23 don't know how that particular mineral or minerals
24 respond to changes in temperature, to humidity, or CO₂
25 or other dissolved constituents in groundwater.

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1 We don't have this information scattered
2 throughout the literature. Never been pulled together
3 into any sort of collective form, a real mission in
4 our ability quantitatively to model cements, but one
5 that could be partly repaired by a really good survey
6 of the literature.

7 All right. QA -- enough has been said
8 about QA in the course of this meeting to convince you
9 that we're -- that laboratory concretes, so-called lab
10 cretes -- are different animals than field concretes.
11 What you make in the laboratory may not be duplicated
12 in the field unless you take extraordinary precautions
13 to see that that is done.

14 It's not enough -- and we've visited many
15 sites that -- where concrete is being used. And
16 what's happening? Well, Joe down on the job with his
17 mixer is getting on with making the concrete. The
18 engineer in charge is sitting in his construction
19 shack drinking instant coffee and filling up
20 timesheets and health and safety forms and God only
21 knows what. But he's not checking the quality of the
22 concrete.

23 And I have seen examples of this in
24 documentation that has been sent to me for assessment
25 by the U.S. Nuclear Regulatory Commission and its

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1 agents where this QA is not being done, to my mind at
2 any rate, to proper satisfaction.

3 Okay. Summary. Well, it appears
4 frustrating that we can't at present predict the
5 lifetime performance of cement barriers or matrices
6 for that matter. In fact, I think that people are
7 oftentimes putting unreasonable demands on cement and
8 concrete barriers. They're wanting to say, "Well,
9 tell me how it will perform. I don't know what the
10 conditions will be and to which -- particularly in
11 shallow burial the conditions may fluctuate, there may
12 be an Ice Age in a thousand years, it may turn
13 tropical and people have got a banana plantation on
14 top in another thousand years."

15 You know, we don't know what the
16 conditions are. We don't know what the groundwater
17 level is. We can tell you what the groundwater
18 chemistry is now, but not in the future. Never mind,
19 we don't have this information; just tell us how the
20 cements will perform.

21 Well, you're not going to get good
22 predictions unless the whole mechanism for evaluating
23 repository performance is firing on all cylinders.
24 People have got to work together on this, and they've
25 got to come up with a holistic scenario.

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1 So to conclude on an optimistic note,
2 though, the purely chemical models of cement
3 performance are reasonably well advanced. That's why
4 so many people have been talking about the chemical
5 alteration or the chemically-induced alteration of
6 cements, including myself in that category, because
7 these models are reasonably well advanced.

8 Okay. They're like a child taking their
9 first steps. They can't yet run, but they can toddle,
10 they can walk. Now, where we're not good and where we
11 also have to improve the links, weak areas include
12 linking the change in chemical and mineralogical
13 changes with mechanical properties. We're not good at
14 that.

15 The engineers say they don't understand
16 the chemistry, and the chemists say, "Well, we're not
17 terribly interested in what the compressive and
18 tensile strengths are." You go away and sort that
19 out, and the answer is nothing gets done.

20 We don't have a basis for accelerated
21 testing that is free from suspicion that it doesn't
22 alter the fundamental mechanisms involved. That's the
23 rub of the problem. I can tell you from experience
24 that the mineralogy of cement is remarkably sensitive
25 to temperature. Even differences like between

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1 5 degrees and 35 degrees are crucial. Big changes
2 occur over short ranges of temperature.

3 So you can't just accelerate things by
4 heating up the system. Sure things go faster, but are
5 they really the things you want to know about? We
6 have people that rely on computer-based model
7 predictions, but we also need those same laboratories
8 to develop methods for experimental verification of
9 their computer-based results. You can't wait years
10 for somebody else to pick it up and do it. You've got
11 to have a dual capability in your laboratories.

12 And, finally, we need a better integration
13 of data into deterministic performance models. I
14 don't think very much of the probabilistic models --
15 I think situations are much better off as far as
16 developing predictive capability if we can have a
17 quantitative model. I feel much more comfortable with
18 these than I do about probabilistic models applying to
19 something where my gut feelings say we ought to be
20 able to calculate this.

21 So that's -- thank you for listening. My
22 final thoughts are that material performance in non-
23 steady states will always be difficult to quantify,
24 that performance has to be quantified by a subset of
25 parameters, each of which can be determined and then

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1 modeled, and we need to all work together on this
2 problem of delivering reliable predictions about
3 cement performance in the longer term. We need more
4 holistic whole of repository performance models.

5 Thank you very much for your attention.

6 VICE CHAIRMAN CROFF: Thank you.

7 With that, we'll take just a moment or two
8 here to change speakers. And our next speaker is Dr.
9 Les Dole from Oak Ridge National Laboratory.

10 Dr. Dole studied corrosion and nuclide
11 propagation in Westinghouse nuclear powerplants,
12 directed research on engineering barriers for some
13 predecessors of the current Office of Civilian
14 Radioactive Waste Management, served as the Technical
15 Director of Call Tech, a Superfund remediation
16 contractor, and led a group at Oak Ridge National
17 Laboratory for more than 10 years that develops and
18 tests wasteforms for various hazardous and radioactive
19 waste from across the DOE complex.

20 Dr. Dole, take it away.

21 DR. DOLE: Allen is giving me the unique
22 possibility here of having -- following people in the
23 program from whose work I've stolen from for at least
24 three decades.

25 (Laughter.)

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1 So not surprisingly, you're going to hear
2 some of these themes repeated. I thought maybe I'd
3 give them cards and they'd hold up and put a number on
4 it, and I could see what my score is for how much
5 material I've stolen.

6 But nevertheless, Fred will be an empiric
7 hard act to follow.

8 Basically, you know, it's like what a
9 woman learns in marriage, you know. The secret of
10 happiness is to lower your expectations.

11 (Laughter.)

12 So the question is: what do you really
13 expect the cement to accomplish in the system? And
14 basically, we're looking at the wasteform package, the
15 bulk, the liner, whatever configuration we're looking
16 at, as a system where different parts of the system
17 contribute to the overall thing.

18 And some of those are -- some of those are
19 mechanical, and some of those are just basically a
20 geochemical buffer between essentially you influencing
21 the local geochemistry to minimize the transport from
22 that site. Now, whether that's 1 meter or 50 meters
23 or 1,000 meters, you can at least include those
24 concepts into your selection of materials. And that's
25 what I'm really going to talk about.

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1 Rather than go through a litany of
2 pathologies that I've seen over my career, I thought
3 I'd try to talk about the elements which we use to
4 choose to try to avoid problems. And most of the
5 problems are really the wrong choice of materials or
6 poor mixing. And that's really not the issue we want
7 to address here.

8 I think we want to look at the idea that
9 cement is a complex material that forms sequentially,
10 and that we have choices in what level of C3A or C3S
11 we choose. We have a choice of the calcium to silica
12 ratio. And based on the experience of my friends, we
13 have a -- somewhat of a guideline, if not
14 phenomenological, at least experiential guidelines to
15 make the right decisions.

16 And so we really have a choice of how we
17 adjust the calcium to silica ratio, the alumina silica
18 ratio, and, you know, how that affects the
19 permeability, how that affects the choice of high
20 density or low density CSH, increase the internal ion
21 exchange capacity or effect reducing conditions or to
22 buffer the pH.

23 So all of these things, from a waste
24 formulator standpoint, are controllable, perhaps
25 unlike construction where you essentially have to work

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1 with a commodity material. In most cases in a waste
2 management context the major cost is not a materials
3 cost. It's an operational cost. So you can afford to
4 go wide and far to find the material that meets your
5 needs. And that's really going to be a small
6 component of the overall cost of your disposal
7 operation.

8 For instance, for years we have used a
9 Type 1-2 low alkalized cement for the northwest,
10 Washington State near Hanford. We're currently now
11 happy with a Type 5 cement from Southern California
12 which is now our favorite. So in waste management you
13 have more luxury. The volumes are smaller. And like
14 I say, relatively, the cost of materials is a smaller
15 component.

16 Now, this is right out of the Barry
17 Scheetz student's handbook. This is the Roy school of
18 grout in Penn State. But the idea is that we have
19 this suite of materials -- pozzolans, slags, and
20 various types of cement. And we can blend those in
21 such a way that we can achieve different properties in
22 the wasteform.

23 Now, the question is: given that ability,
24 what do we use? What do we want? We're looking for
25 a tall ship and a star to guide her by. Well, this is

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1 the star I use more often than not is this phase
2 diagram, so we may have to like refer back to it. But
3 the idea is we want to steer the system into a phase
4 -- part of the phase diagram that prevents the
5 formation of calcium hydroxide, which is the most
6 labile component.

7 And we usually formulate right about this
8 upper line, because a lot of the times the materials
9 we use we formulate based on total silica alumina and
10 calcium. And because silica comes in a variety of
11 minerals, not all of it participates in the pozzolanic
12 reaction.

13 So by working up at this end there may be
14 a fraction of unreactive material that keeps us
15 actually effectively down in this region. But this is
16 kind of the sweet spot which we use, and we'll talk
17 about what we hope to achieve by picking this sweet
18 spot, in addition to reducing the free calcium
19 hydroxide.

20 Now, this is a formula -- now, I've always
21 said that I've been a dirty water cement chemist in
22 which I mix the waste directly with cement, and that
23 there's a whole body of civil engineering experience
24 out there that I call clean water cement. And I've
25 never had much to do with them.

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1 Suddenly, as a cruel twist of fate in my
2 career, I'm suddenly trying to formulate an
3 engineering material. And we're looking at coming up
4 with a blend of live cement, blasfernous slag, Class F
5 fly ash, and silica slew that controls over the
6 sequence of curing the level of calcium hydroxide.

7 So, in principle, we want some calcium
8 hydroxide, because that's what drives many of the
9 reactions. It helps etch the aggregates, and so we
10 want -- we don't want to eliminate calcium hydroxide
11 immediately, but at the end of the curing cycle we
12 want it all gone. So what we've done is we've found
13 a blend of these components, these specific
14 components, that give us the effect that we want.

15 So the idea, then, is that by the
16 judicious selection of materials you can formulate a
17 cement that avoids calcium hydroxide yet still has the
18 properties you need to develop strength and bonding
19 with aggregates and other properties that you would
20 like.

21 Another issue is that by reducing the
22 calcium to silica ratio, I think we're -- anyway, this
23 is the dehydration. Obviously, when you heat up
24 cement to about 200 to 300 degrees, which is about the
25 estimated heat pulse that you would get at Yucca

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1 Mountain, you dewater the CSH. And you have to go all
2 the way out here to 5-, 600 degrees before you
3 actually decompose the calcium hydroxide.

4 Well, we're eliminating the calcium
5 hydroxide, but the other aspect we're looking at is
6 controlling the low density CSH and the high density
7 CSH. Fred refers to this I think as microcrystalline
8 CSH, but it doesn't matter, CSH forms with two packing
9 densities. And I think a lot of the work out of NIST
10 points to this as well. In fact, this is data from a
11 NIST-based work that shows the distributions.

12 And so by adjusting the formulation, what
13 we're hoping to do is reducing the fraction of low
14 density CSH, which is responsible for shrinkage, both
15 from dehydration and from curing, because what happens
16 then with aging is that the low density CSH
17 redistributes and repacks itself into dense CSH, which
18 -- so if you can slow down the formation of low CSH,
19 extend the curing so that when it forms in its early
20 stage you get the high density CSH, then it is less
21 prone to shrinkage on curing, it's less -- it won't
22 shrink on aging, and it's more resistant to
23 dehydration.

24 And, again, this talks about the two
25 packing densities of high density and low density CSH.

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1 And they really don't change much below 250 degrees,
2 so that gives us some indication that the wasteforms
3 will be resistant or the structural materials that we
4 choose for the repository couldn't be expected to be
5 reasonably resistant -- in other words, maintain its
6 physical integrity over the thermal pulse.

7 Now, that's not related to time, but Fred
8 has talked about the time, in that if you do form this
9 high density CSH, the reactions that redistribute the
10 materials are slowed down because they're now solid
11 diffusion limited. So if you create the micro-
12 nanostructured CSH or the high density CSH, you've
13 essentially increased the diffusion barrier or the
14 rate at which they recrystallize with aging or aging
15 under thermal conditions.

16 Now, there's really no -- there's no
17 modeling at this point. Maybe if I have time at the
18 end I'll talk about some of our methods of modeling.
19 Certainly, Ed is following on with the NIST, but the
20 other aspect of lowering, as Fred alluded to, is if
21 you reduce the calcium to silica ratio to -- you know,
22 from 1-1/2 down to 1, you -- the soluble silica that's
23 available in leaching, either for healing microcracks
24 and later wetting episodes, or forcing the
25 geochemistry of the system in insoluble components is

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1 improved by lowering the calcium to silica ratio.
2 And, again, it's a balance between early strength and
3 this calcium to silica ratio.

4 So what we're really doing, then, is that
5 if we raise the silica counts -- and that's -- some of
6 the earlier discussions talked about the carbonate and
7 the pH. They left out the part about the silicates,
8 because when you add aluminum silicates then you force
9 these insoluble compounds, and even then those initial
10 insoluble compounds which precipitate on the surface
11 of the fuel or on the surface of the cement, which
12 shuts down diffusion, shuts down oxygen transport, may
13 perhaps at this point not -- we don't know how to
14 quantify those or model those kinetically, but we
15 certainly have a good intuition that these things form
16 and that they are very, very effective at reducing the
17 mobility and solubility.

18 And even then, more complex than that is
19 that it's really a sequence. We talked about
20 kinetically we get soddyite and urethane forming, but
21 then over time they alter themselves into even more
22 stable compounds of haywetite and ursalite. So,
23 unfortunately, nothing in this system is simple.
24 Nothing comes out of the solution directly in forms.
25 It comes out as an intermediate and slowly undergoes

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1 a transition.

2 So even phosphates or carbonates are
3 silicates all come out in a very complex sequence.
4 And we're just starting to unravel that. We're just
5 starting to get the tools to get -- to start to get
6 a handle on that. But it -- as Fred points out, we're
7 in the infancy of identifying precisely which
8 compounds and the sequence of which they form.

9 The other issue it talked about is we
10 talked about stress cracking and load. Certainly in
11 the -- we have a great predilection for using steel
12 fibers for two reasons. One, they increase the area
13 under this curve, so they increase the fracture
14 toughness by sometimes 10-fold. And if you really go
15 out, maybe 20- or 30-fold. This does several things.

16 First, the metal adds a reducing quality
17 to the mix. And, second, it increases the
18 flexibility, the flex strength, and its ability to
19 absorb energy. And it's quite critical from the
20 standpoint of using silos for hardening against
21 missiles and things like that.

22 So there's a tremendous body of knowledge
23 on how to harden facilities and how to harden
24 materials that at some point or another will be
25 germane to waste disposal.

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1 Okay. And this is the one we've all seen
2 before. I've used this slide a dozen times. But the
3 point is is that when you try to accelerate the tests
4 for temperature, as we pointed out nothing is that --
5 nothing is in a nice linear simple step. And that as
6 I said -- others have said before is that when you
7 change the temperature you change the reaction path of
8 the system. And so you have to be very skeptical
9 about what you see in an accelerated test.

10 So is there a way around that? Hopefully,
11 I'll talk about that. So what's missing, then, is a
12 mass transfer, coupled thermodynamic model, a
13 deterministic model. And NIST has part of that, and
14 unlike Fred I -- as a physical chemist, I'm not so
15 uncomfortable with stochastic methods. But I think
16 we're just now starting to really get a serious
17 handle, and NIST has led the way.

18 The team at Lawrence Berkeley -- John Epps
19 and Carl Stieffle -- have been working with Tough
20 React, which is another variation of a coupled
21 thermodynamic and kinetic model. And we're trying to
22 come to grips first with just getting the literature
23 data in it.

24 And, again, I -- my experience with
25 culling the literature is the literature was taken for

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1 another purpose. And we'd go back and look at that
2 data, and there's such a range of experimental
3 conditions and these measurements are so sensitive to
4 your choices of those conditions that it's really hard
5 many times to just gather this up and put it in a
6 coherent database. But it's something that needs to
7 be addressed, and, skeptical or not, it needs to be
8 tried.

9 So then it gets to the issue of
10 anthropogenic and natural analogs. I found it
11 curious, I was looking at some background material,
12 and there are guidelines from the IAEA, what
13 constitutes a good analog and not. And Pettit has
14 worked in this area also.

15 What I found curious was that in the
16 German waste disposal program they chose that the most
17 important thing, number one on the list to be solved,
18 is looking at anthropogenic and natural analogs, which
19 my reaction was that somebody was smart enough to pick
20 out the fuzziest, most unreliable, and most difficult
21 task and put that as number one to disposal.

22 But it's very difficult, and it can't be
23 done independent. There has to be an organized
24 approach to your laboratory work and you analyze these
25 materials and you are constantly looking at the

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1 information. You're not going to see validation from
2 this.

3 You're going to get ideas about what to
4 look for and what not to look for or find things that
5 you found in the laboratory, you didn't see in natural
6 materials, and try to resolve the differences. But
7 you're never going to converge on the answer with this
8 system, but you need to do it to at least find bounds,
9 to get some sense of where the systems go and -- with
10 these very long times, even though it's very
11 difficult.

12 Now, we've located -- most people are
13 familiar with the Gallo-Roman. Naboseans are kind of
14 unique, but we found in the U.S. we have a formation
15 in Marble Canyon, Texas, there's the Hatrium formation
16 in Israel, and the Scawt Hill in Northern Ireland that
17 are fairly well documented areas where you could find
18 materials that -- where through heat events, whether
19 it was magma or an underground fire, that you baked
20 the formation into a cement clinker, it hydrated
21 naturally, and you can go back and then try to unravel
22 what happened to these systems over the last million
23 or so years.

24 So it at least gives you some sort of
25 benchmark, but that benchmark alone only works if you

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1 have a concomitant or a parallel study in the
2 laboratory to take a look. And I'll talk a little
3 about the link -- mass transfer.

4 What may save us is that besides the
5 better models that are evolving is that we also have
6 much better microtechniques where we can start to look
7 at modifications to crystalline layers, almost a
8 molecular layer at a time.

9 And so if we have these extraordinary
10 methodologies that gives us -- so we don't have to
11 wait 10 years to see an effect, we can wait maybe one,
12 two, three years and we can start to look at the
13 direction in which the surface of the crystals are
14 starting to evolve. That gives us an early indication
15 of where the system is going to go in longer time.

16 So somewhere between the natural analogs
17 and applying these extraordinary analytical techniques
18 we might be able to start to link up the thermodynamic
19 and kinetic deterministic models and then link it.
20 And also, with the help of the NIST in their
21 stochastic modeling, at least come up with a better
22 picture of what's going to happen.

23 So we can't do it now. Just the elements
24 necessary to do it better are just now starting to
25 come out, and no one is putting them all together in

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1 a coherent way at the moment.

2 I didn't talk much about leaching. But
3 think about leaching two ways. If you disrupt cement-
4 making -- we usually look at leaching from the
5 standpoint of trying to keep some nuclide or some bad
6 guy in the mass. But the other turned that around and
7 said if you have an aggressive agent in the
8 environment, and it wants to -- in other words, for
9 cement to degrade, some environmental element has to
10 diffuse into it.

11 So the same mechanisms perhaps that
12 control the release of things also control the ingress
13 of things, like diffusion, effective diffusion
14 coefficients, porosity, permeability. And so some of
15 the leaching approaches may or may not be as
16 successful.

17 The problem of -- okay, let's take the
18 issue of permeability. What do you really expect?
19 All you really need from permeability from a wasteform
20 is it has to be 100 times less than the permeability
21 of the adjacent formation. And if there's any
22 advective scenario, then a particle of water goes
23 around rather than through. So most times we're
24 looking at soil permeabilities of 10^{-5} , and we can
25 generally -- we can make wasteforms at 10^{-7} , 10^{-10}

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1 microdarcies.

2 So we know how to make impermeable
3 wasteforms. We use the pozzolans so they cure very
4 slowly. The plastic state is extended over long
5 periods of time. Ed will talk about, you know, the
6 creation of internal tension or stresses that cause
7 microcracking. It relates to the placidity of the mix
8 and the time at which it sets.

9 So if you extend the placidity of the mix
10 way into the setting process, you relax all those
11 interior stresses. You allow those interior stresses
12 that cause microcracking to relax, and it reduces it,
13 and that's another virtue of choosing cements with low
14 C3A or C3S and the use of pozzolans.

15 Okay. Diffusion -- again, as David
16 pointed out, using a single diffusion coefficient to
17 describe the complex sequence of chemical events is
18 pretty imaginary. I mean, the term "effective
19 diffusion coefficients" is one I use a lot, because
20 it's something we measure in the laboratory, and it
21 reflects many processes, not simply -- it's not a
22 simple diffusion coefficient.

23 It's a coefficient that may at one point
24 in time in the life of a wasteform reflect the
25 sequence of events that controls the release at that

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1 point in time. Now, that changes with time. And as
2 David pointed out, if you pick a diffusion coefficient
3 early in the age of a wasteform, it's generally very,
4 very conservative. And you're right. If it -- if it
5 overestimates the release by two orders of magnitude
6 it's the wrong answer.

7 And so we've looked at that. It gets back
8 to the use of static KDs and static leach tests. It
9 goes back to exactly what Fred was talking about is
10 that the results reflect the experimental design. So
11 that the time is really driven by the solid surface to
12 liquid ratio.

13 And so, again, Fred wants to go back and
14 look at the literature, but very seldom if you compare
15 these either -- either from leach data or partition
16 data can compare those unless they are taken under
17 similar experimental conditions, similar experimental
18 configurations. So it's very difficult to interpret
19 those results.

20 It's good news and bad news. The bad news
21 is that when you use static KDs and static leach tests
22 and effective diffusion coefficients, you get the
23 wrong answer. It's there any good news in there,
24 generally it overestimates the release.

25 And if you can accept that, then it's

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1 fine. But if you can't, then you have to go back into
2 the more deterministic models like David was talking
3 about and that we're trying to develop with Tough
4 React and some of the other geochemical models where
5 it's a sequence of geochemical processes that
6 ultimately determine the release. And it's not a --
7 it's never going to be very simple.

8 Okay. So we do have a lot of experience,
9 and we've taken these data. And we have a good sense
10 that they are fairly conservative, but we also know
11 they're the wrong answer.

12 I think I'll leave it go at that. Again,
13 we're beginning to use Tough React and build more
14 geochemistry into our model. Our particular modeling
15 is looking at the near field of the repository, the
16 impact of the cement and the cement chemistry on the
17 immediate contact with the formation. NIST is working
18 at looking at the inside of the cement fabric and its
19 effect. And eventually hopefully we'll meet. It's
20 our intention to do that.

21 Questions?

22 VICE CHAIRMAN CROFF: At this point, as
23 before, I think we'll defer the questions to the panel
24 discussion. And we're doing reasonably well here, but
25 I think it's time we took a short break. That's what

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1 we've got scheduled. So let's come back about five
2 minutes after 3:00 if we could.

3 (Whereupon, the proceedings in the
4 foregoing matter went off the record at
5 2:52 p.m. and went back on the record at
6 3:07 p.m.)

7 VICE-CHAIRMAN CROFF: Last leg of the
8 race. At this point, I'd like to introduce our final
9 speaker who is Dr. Ed Garboczi from NIST. Dr.
10 Garboczi is a physicist and leader of the Inorganic
11 Materials Group in the Building and Fire Research
12 Laboratory. This group carries out a combination of
13 experimental and computational materials science on
14 cement and concrete to supply measurement and
15 prediction technology to industry and helps support
16 the development of science and performance-based
17 standards. It's this group that developed the Virtual
18 Cement and Concrete Testing Laboratory software which
19 is a tool for predicting the performance of concrete
20 from fundamental materials science.

21 With that, please.

22 DR. GARBOCZI: Is this coming through all
23 right?

24 Okay, so I was here last year, I remember,
25 at a similar meeting. This year is much more focused

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1 on concrete science and I'm in a very distinguished
2 crowd of speakers here, so I feel honored to pick up
3 the end here.

4 I'm glad we had a break because it woke
5 you up a little bit, I hope. I hope you had some
6 coffee and it's been a long day.

7 I just use the title that Allen said, the
8 last two talks before Dr. Glasser and Dr. Dole had the
9 same title, but they focused on other aspects of it.
10 I'm going to focus on the computation aspect of it.
11 And just to give you a little bit of background, I
12 think some of my introduction will seem a little
13 familiar to you by now, but I was talking to Dr.
14 Glasser the other day. I said I think a lot of these
15 talks overlap each other. He said oh, that's good to
16 see the same thing with different points of view. So
17 he said it must be okay, so --

18 (Laughter.)

19 For a long time we've been providing
20 scientific and technical foundations with performance-
21 based selection use of concrete. The whole -- try and
22 drive the whole world to performance-based
23 specifications. We do that through material science,
24 trying to give a better material science basis for
25 tests, performance-based standards, etcetera.

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1 Part of our -- fairly unique to us, at
2 least a while ago it was the use of computational
3 materials science. We have always felt that the
4 complexity of concrete demands computational materials
5 science along with experimental materials science to
6 back it up. Jeff Frohnsdorff, my former boss who
7 passed away this year, was really the driving force
8 behind that for many years. In fact, in the '60s, he
9 helped write the first computer-based model. So it's
10 a little testimony to him. He passed away in March of
11 this year.

12 We look at the length of scales of
13 concrete from meters down to nanometers. Our main
14 work has been in the micrometer and millimeter scale.
15 Some early stage work on the nanometer scale and some
16 of the prediction tools we'll talk about today
17 generally fall in this area.

18 We have a structural group which
19 concentrates more in this area as well.

20 I'm going to set up, trying to find the
21 problems we're looking at in terms of time scales and
22 structural complexity. It's just my point of view.
23 I remind you that's my point of view and if I say
24 something, it's kind of runs counter to the current
25 norms in this whole area, please excuse me, because of

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1 my ignorance.

2 So for me, when we talk about prediction,
3 we're looking first at shorter time scales. Shorter,
4 I mean that 100 to 200 years type scale. And that
5 region there's two kinds of problem. There's the high
6 structural complexity problems like new nuclear power
7 powers, for example. There's a lower structural
8 complexity, things like low-level reactive waste --
9 vaults kind of thing. They're much less structurally
10 complex than say new nuclear power plants.

11 I don't really mean crack free. That was
12 sort of stupid to put it there. I mean sort of
13 control crack or not too many cracks or doing the best
14 you can with cracks. But for this kind of concrete,
15 like the usual construction industry concrete which we
16 really serve the construction industry, so that's my
17 point of view, just like Rachel was saying earlier
18 today.

19 The use of degradation processes, you
20 should remain pretty immune to them for 100 to 300
21 years and that will make your barrier work right. It
22 will make your concrete in your nuclear power plant
23 work right.

24 There's also an additional problem though.
25 It's not just the usual processes like Rachel talked

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1 about, but also the other chemicals that could be
2 there like the waste, stuff in spent fuel pools or
3 some WR problems. There's other chemicals too.
4 There's other chemistry involved as well.

5 And for all of this, as you've gathered
6 today, I think, prediction is not so easy. Then
7 there's the other point of view which I'm not so
8 familiar with, even though we worked at NRC for the
9 last 15 years or so, it's not -- it's a little bit far
10 for me is the longer time spans, the thousand years,
11 the 2 thousand, 10 thousand year time spans. We talk
12 about thermodynamics.

13 That time span, the concrete must continue
14 to function in its physical and functional
15 containment. The degradation processes are a little
16 bit different. You have to avoid the usual shorter-
17 term degradation processes. If you want something to
18 last 10 thousand years, you can't have it fall apart
19 by alkaline silica reaction in 50 years.

20 I think there's some ways to make progress
21 on this and we've heard some today as well. I'll talk
22 more about it later. And if anything, prediction is
23 probably even harder for this time scale than the
24 shorter time scale which is no surprise to anybody.

25 Was it Yogi Berra who said about the hard

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1 thing about prediction is it's about the future?

2 (Laughter.)

3 Why is prediction difficult? I mean the
4 time scale gives difficulty as well, but I'd argue for
5 this material, it's even more difficult. I would say
6 the reason is anything you're going to predict about
7 the concrete, how it's formed, what its properties
8 are, how it degrades, interaction of chemistry,
9 microstructure, micromechanics. It's not just things
10 reacting, look at the reaction product. See if they
11 look at the mechanics and they interact with each
12 other. I'll show you some examples of this.

13 Of course, you can't ignore cracks.
14 That's part of the micro mechanics as well and the
15 micro structure, and sometimes the chemistry.

16 Let me give you some examples of what it
17 means, how closely these things interact. Just plain
18 hydrating cement. Cement, paste mixed up or you have
19 a concrete mixed up, if you have a lower water to
20 cement ratio, probably around .4 or lower, or any
21 hydration involves chemical shrinkage. The products
22 take up less space than reactants, volume-wise.

23 If not enough water comes in from outside,
24 you tend to start using up the water inside the mix
25 and you end up with air/water menisci. Menisci have

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1 forces in them which induce tensile stresses. Tensile
2 stresses can induce cracking. That's the autogenous
3 shrinkage cracking that was mentioned a couple of
4 times. So that's chemistry and mechanics missed up
5 together. Chemistry induces mechanics which can
6 affect the material. Every cement has that. It's
7 just a lower ratio, tends to be a bigger problem.

8 Degradation is -- one example of
9 degradation. Alkaline silica attack and alkaline pore
10 solution reacts to the morphosilica and the
11 aggregates. This is a gel. The gel can swell and
12 cause induced tensile stresses, cause cracking.

13 Carla Ostertag at Berkeley has introduced
14 fibers and tried to control the fibers going in near
15 the aggregate paste interfaces. It turned out the ASR
16 was controlled a lot, but it's merely mechanical
17 means. There is the chemo-mechanical problem.
18 Usually people do a chemistry to alleviate ASR. She
19 did fibers, so she controlled the swelling. The
20 chemistry changed. So into the mix of chemistry
21 mechanics, this is one example of that.

22 I'll say a word about cracking, another
23 difficulty in this material. If you have a block of
24 concrete and you wonder what the transfer properties
25 are, if you can spatially and topologically

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1 characterize the cracks, there are ways to predict
2 transfer properties. You do 3-D calculations, find
3 out whatever. And you can predict transfer
4 properties. If you can do this, this is probably the
5 harder job, characterizing spatially and topologically
6 in the cracks.

7 It's a lot better to, of course,
8 predicting the occurrence of cracking is a lot hard to
9 do. It's much better to -- much easier to prevent it
10 in the first place. The ways you do that structural
11 design, you know, obviously good constructional design
12 means you don't have settlement cracking, that kind of
13 thing. That's beyond my field of expertise.

14 You mix designs, ways to look at mix
15 design and proper curing. And this was mentioned.
16 I'm going to try to mention a few things that weren't
17 talked about earlier. One way to alleviate cracking
18 which can alleviate cracking sometimes, is curing, but
19 internal curing. Instead of applying water from the
20 outside, especially high density concrete, high
21 performance concretes, high strength concretes which
22 might be used a lot in say nuclear power plants or
23 some places it may be in waste disposal. It's so
24 dense that you can't really pour in water from the
25 outside. The transfer rate is so slow the water can't

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1 get in, so you can run that water inside through this
2 cell desiccation I talked about before.

3 So one way is to try supply water
4 reservoirs inside the concrete. This thing can be
5 drawn on as the emergency supply of water when the
6 regular water, mixing water runs out. Well, there's
7 many ways to do that. One way is if you replace some
8 of the fine aggregates with porous fine aggregates,
9 quite porous, they have water tied up in them. You
10 mix it in and then water can be pulled out of the fine
11 aggregates when the water is needed in hydration
12 process to alleviate some of the cell desiccation.
13 And that's been shown to work well.

14 Dale Bentz in my group did an x-ray
15 tomographic study where these are the porous fine
16 aggregates. The blue is showing where water has left
17 the aggregate. So as curing went on water is pulled
18 out of the aggregates into the space where the water
19 is needed in curing. So he can definitely show that
20 this water was being used as a water reservoir for
21 internal curing. In fact, it could be very helpful.
22 There's other ways to do it, first kind of polymers,
23 you can use that too. But that can alleviate cracking
24 and it's shown to alleviate cell desiccation shrinkage
25 quite a bit.

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1 That's a fairly new technique that's out
2 there, but something that could be very useful for any
3 time you're going to use high density concretes.

4 I'll say one word about mineral admixtures
5 before I get on to more prediction stuff. I'll say
6 it's a monkey wrench. You know, it's very common to
7 use mineral admixtures today. Pore cement chemistry
8 is hard enough, chemistry mixed in with dirty water
9 type stuff, is even harder. You mix in some of these
10 other things, you get problems even harder. I would
11 say that chemical interactions are not well known.

12 For instance, you can pretty well measure
13 the amount of cement that's hydrated versus time. The
14 reaction of cement versus time. When you have cement
15 mixed up with flyash or silica fume or blast furnace
16 slag you can make a measurement of the reaction of the
17 flyash versus time. That's not that much data on
18 that. There's some, but you have to work very hard at
19 it.

20 So I would say when you're mixing this
21 stuff, your durability predictions are probably even
22 less steady than before. We saw before that just the
23 durability predictions for Portland cement and
24 concrete are not that great.

25 These mineral admixtures are often used to

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1 -- if you can avoid cracking -- to get low
2 permeability, low hydration. You also tend to
3 ameliorate the usual degradation stuff. This was
4 mentioned -- I think you mentioned that. You also
5 tend to lower the pH of pore solutions, so you have a
6 trade off of lower permeability with low pH.

7 Also, chemical shrinkage involved with
8 these kind of admixtures are more than Portland cement
9 so even the higher tendency toward cell desiccation
10 shrinkage cracking with mineral admixtures than you do
11 with just plain Portland cement.

12 And so internal curing is probably even
13 more important when you use those materials than just
14 plain cement. And the whole world is using these
15 mineral admixtures right today. Anything new built
16 with these materials, built with this cement, it's
17 going to have mineral admixtures in them.

18 Okay, let me get to prediction now.
19 Predicting durability failure for first principles,
20 that's sort of a grandiose title to the slide. We'll
21 just pass on by that title and I'll give some details
22 for it. But you need to correctly predict
23 transporting reaction mechanisms, ions moving through
24 cement paste pore solution, how they move, how they
25 react.

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1 As they move and react, how they change
2 the material micro structure, cause cracking or close
3 cracks or fill up the pore space or open up the pore
4 space, how that changed material microstructure
5 changes the properties, mechanical properties and the
6 transfer properties which then change how the ions
7 transport and react and change into a micro structure
8 and so forth and so on, how everything changes with
9 time.

10 You expect the surface environment is not
11 so easy. That's beyond my research, my level of
12 expertise, but that's a key thing to characterize how
13 something is going to react over even 100 years. And
14 of course, the relevant thermodynamics which is not
15 totally well known for all these materials.

16 And any predictions you make must be
17 accompanied by the results of valid -- I say auxiliary
18 tests for valid experiments, accelerated or not. And
19 the point Fred made is well taken that the models and
20 the verification need to go together.

21 Actually, I'm not going to talk about this
22 very much today, but just to mention in passing that
23 to kind of highlight for my group for our current
24 abilities and prediction, the Virtual Cement and
25 Concrete Testing Laboratory, we're trying to build up

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1 a software laboratory, just like a physical testing
2 laboratory, but only on the computer so you build
3 concrete on the computer, test it on the computer,
4 etcetera.

5 Try to make it as fundamental as possible.
6 It's not empirical at all. And we have an industrial
7 consortium we're working with and they're focused on
8 producing concrete and selling concrete. So it's
9 mainly focused on predicting current properties of
10 concrete, short-term properties. Eventually, we'd
11 like to be able to use it for durability in the
12 industrial construction 50 to 100 year time frame. In
13 fact, Rachel and I didn't talk together, but 50 to 100
14 seems what she said. Fifty now, 100 hopefully next
15 decade or something.

16 So that's what the focus is on now.
17 There's pieces of it though which can be used for the
18 kind of prediction more interesting to nuclear
19 industry. I'll get to that in a second.

20 That's the people we're working with and
21 I need to put them up because they do supply funding
22 and research over the last six years. So it's
23 important to mention them.

24 Software base tool. It just looks like a
25 web page and you just pick various things. Just like

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1 you go into a laboratory and pick various instruments
2 to measure. It works the same way.

3 Okay, the rest of the talk now, I want to
4 go through and just highlight some topics of things we
5 can currently predict or hope to predict, things we're
6 working on which could be relevant to the topic of
7 today.

8 We'll look at hydration, micro structure
9 formation and chemical interactions, rheology of
10 cements and concrete, micro mechanics, transfer
11 properties of ions and pore solution, thermodynamics,
12 a little bit and a little bit about soil durability
13 tests.

14 Fred pretty much trashed them, but I'll
15 try to trash them a little further.

16 (Laughter.)

17 Now cement hydration. There's been models
18 of cement hydration for a long time. Like I said,
19 Jeff Frohnsdorff passed away recently. His 1960s
20 model was a computer model of hydration and solved for
21 differential equations and that's been carried on a
22 lot around the world. In the '80s, there was work at
23 NIST led by Jeff Frohnsdorff to develop a
24 microstructure model, how a 3-D microstructure form
25 for hydration. Unfortunately, it didn't have any of

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1 the fancy chemical kinetics. It was more kind of
2 empirical, but more of a structural model.

3 We've improved on that with our current
4 model. But still, it doesn't have the kinetics in.
5 It forms a nice microstructure, it has sort of pseudo
6 kinetics in, but doesn't have what you really want in
7 a model.

8 What we're working on now is we're trying
9 to marry the two and have a real model that gets you
10 a real three-dimensional micro structure that does
11 real chemical kinetics, real chemistry and physics of
12 the cement reacting. That's the way forward to clean
13 water cement and for dirty water cement.

14 The way we do it is to break up particles
15 into rock seals and do a three-dimensional digital
16 model where you actually represent the cement
17 particles and the formation of products and get a
18 three-dimensional microstructure.

19 You have to start with a -- with the real
20 cement. If you just start with idealized cement you
21 get the wrong properties, so we characterize the
22 cement very well with SEM and backscattered electron
23 imaging. You get the atomic elements, mix them all
24 together and come up with a three-dimensional version
25 of the real shapes, the real sizes and the realistic

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1 distribution of clinker phases among the cement
2 particles. And that, we found is crucial to start out
3 with -- if you don't start with that, the model is not
4 going to be very good. It might not be very good
5 anyway, but it sure won't be any good with -- if you
6 start with that.

7 Our current cement hydration model is rule
8 based. The rules are based on chemistry, real
9 chemistry and there's various chemical phases in
10 there, but there's no true kinetics. We're now
11 working on, Jeff Bord in my group is working on
12 HydratiCA which is a chemi-physical model which hopes
13 to marry all this stuff together. I'll tell you a
14 little bit about that soon.

15 I should note that working with mineral
16 admixture companies in our consortium, they have lots
17 of experience with mixing organic and inorganic
18 chemicals, effecting cement hydration. It's possible
19 that we could learn from their experience to help us
20 with their -- work with dirty water cement and the
21 various stuff that occurs in the nuclear waste
22 containment.

23 HydratiCA, what we're working on now is
24 object-oriented code. And you don't really know what
25 that is, just a new way of computer program which is

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1 very adaptable.

2 Algorithms designed the model that's
3 reaction and transfer equations of the ions
4 dissolving, reacting, foreign product, it's all in
5 there. It's real kinetics. If you run the model for
6 an hour, you can tell exactly how many hours, minutes,
7 days it was in real life. There's an exact
8 relationship between the kinetics.

9 All that stuff is just to impress you. I
10 don't really want to talk about it, but just lots of
11 stuff there and it goes in the model. Jeff has done
12 a very nice job with it.

13 Let me just mention before I leave that
14 because there's real kinetics in there, because the
15 model is set up to easily add new materials, for us
16 it's been adding new cements or adding flyash or
17 something, but it works for any materials. So if you
18 want to start reacting, some uranium complex, if you
19 know what the ionic species is, you know how it reacts
20 with stuff, you can put it in this model and let it
21 react. It will react to full microstructure. If you
22 know the laws, if you know the various coefficients
23 needed, that will work. So it's very easy to add new
24 materials in, any materials at all, as long as you
25 know some basic information about them.

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1 We'll switch to rheology. Rheology is
2 important for construction issues, obviously, for
3 placing concrete. We talked about the tank problem,
4 filling in the grout and i guess Barry mentioned all
5 the rods and stuff in there. Rheology is going to be
6 a big -- it's going to play an important role for that
7 as well, to get the concrete, the grout to flow into
8 all the interstices and fill up without voiding, large
9 voids, filling up the tank. So rheology is important
10 for both problems.

11 We can quantify roughly the rheology of
12 any suspension by viscosity and seal stress. What
13 we're working on is measuring experimentally,
14 predicting computationally and I'll tell you something
15 about both. The modern drive for rheological research
16 has really taken off in the last 10 years or so and
17 it's been self-consolidating concrete. That's the
18 thing that came out of Japan, I guess Japan about 15
19 years ago or so, where you make concretes with very
20 low yield stress.

21 So typical concrete, you are trying to
22 pour it into a heavily reinforced region. It has to
23 flow between the small holes and reinforcements. It
24 gets stuck. If it has a non-zero yield stress, it
25 gets stuck. You have to kind of stick in vibrators or

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1 push it along or guys with shovels and boots slam it
2 along. But self-consolidating concrete has been
3 lowering the yield stress, keeping the viscosity high
4 so the rocks don't just drop right out in the bottom
5 of the form and self-consolidating means that it can
6 flow anywhere, sort of go in and fill up and you're
7 done. You get rid of voids. You get rid of the need
8 for guys in boots with shovels and vibrators pushing
9 concrete around.

10 And so the usual way of looking at how
11 concrete flows is some tests are not good enough for
12 self-consolidating concrete, hence, the drive toward
13 a more sophisticated rheological measurements for
14 concrete.

15 All right, so rheology applies right from
16 the mixing stage through the mixing of truck stage
17 through the flowing and placing. We measured in the
18 lab at the mortar scale. Also measure in the lab the
19 concrete scale and then we simulate it using real
20 shapes of rocks, shapes of rocks that are scanned from
21 x-ray tomography and put into the models and we have
22 code to let the matrix flow and the rocks spin and
23 tumble and bump into each other.

24 And then the results from these
25 experiments, preliminary results compare to the

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1 experimental data and then we can tell, basically we
2 can tell where the data is wrong or we need to do
3 better experiments. Because the theory is always
4 right, but the experiments are wrong sometimes. It
5 works both ways.

6 Although I must say in my experience, it's
7 been a little easier to get the modeling right than to
8 get the measurements right because spinning a
9 suspension around and you get rocks in the fly out to
10 the outside, it's hard to keep suspension uniform and
11 measure at the same time. So there's difficulties in
12 both, but I think we're overcoming them.

13 So we have some -- I think we can do a
14 decent job of predicting rheological parameters.
15 We're starting to do any way.

16 Let's look at micro displacements in
17 concrete, I mean micro mechanics. We're real good at
18 compression strength measurements. We put a block
19 something and smash it. That's sort of macro. But if
20 we really want to look at degradation you have
21 chemistry happening at the pore level. You have
22 growth of cracks and growth of phases. It shrinks in
23 phrases. You have micro mechanics. You have stress
24 at the local level and that's really what I'm talking
25 about. That's a hard problem to get at. I think we

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1 had some of the pieces for it, but it's a particular
2 problem.

3 So several different mechanisms of
4 degradation involve some kind of localized
5 displacement. Aggregates expand or shrinks, various
6 things. They all set up stresses in the micro
7 structure. Tensile stresses drive cracks to form.

8 So this is a piece of real concrete that
9 actually was cracked and just some mechanism. We cut
10 out a piece of it computationally. This is in 2D. We
11 can do this in 3D. It's just a lot more visual in 2D,
12 obviously. You take this and put it in the computer,
13 specific elastic properties for each phase, realistic
14 properties and then you can test various mechanisms.
15 You can say well, was this cracking caused by all the
16 aggregate expanded? Let's try and see what happens.
17 So we did that hypotheses and said let all the
18 aggregate expand a little bit. That's going to drive
19 stresses in the cement base matrix which didn't
20 expand, just gets squeezed. So where would the cracks
21 go?

22 So you saw the fine element problem. Saw
23 stresses everywhere. Find principal stresses
24 everywhere. Map of tensile stresses. And then this
25 shows where the cracks would be. This is the probable

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1 crack directions that is based on the tensile stress
2 is up by that mechanism of having aggregate expand.
3 And so you can go back and then compare this to the
4 real one and say well, was this how the cracks
5 actually went? If it didn't, then that mechanism was
6 wrong. You go back and try another one.

7 This is a 2D simple way of looking at
8 micro mechanics. We have three dimensional models.
9 We can look at stresses in 3D and hopefully coupling
10 that into the models of HydratiCA with degradation
11 reactions going on and start coupling this together.

12 It's really what our kind of window of
13 work is, trying to couple the microstructure and the
14 chemistry and the properties together via the micro
15 structure. We're not real great at any one of them,
16 but all together, we kind of do I think fairly unique,
17 in trying to couple together those things into one
18 kind of model.

19 Let's look at transport and reaction
20 degradation. This is a piece of cement paste matrix.
21 We have unhydrated cement particles. You have all
22 kinds of hydration products. You have cracks and you
23 want to know as you flow in ions, ions react and
24 what's going to happen to them as they go through the
25 pore solution.

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1 You want to calculate transporting the
2 concrete pore solution. It's a pretty nasty problem.
3 The reason is several times today we mentioned
4 effective diffusion coefficient. The pore solution in
5 cement paste is very high ionic strength, much higher
6 than -- if you look in the handbooks for transport
7 properties versus pH or versus ionic strength, usually
8 they run out before they get to cement paste. So Ken
9 Snyder has worked on this a lot and has had to go
10 beyond that and look up in the literature to get how
11 the transport rates depend on ionic strength.

12 So that's hard. High pH is hard and the
13 many chemical species involved, even in clean water
14 cement is pretty hard too. And so if you want to look
15 at the ionic species going through the pore solution,
16 you have a couple of problems because you have
17 reactions going on as you have transport. You have
18 degradation happening and then the degradation changes
19 transfer properties, so the code that he's worked on
20 for a long time is called 4SIGHT and that's been
21 worked on at the NRC for many years.

22 We're trying to handle it, those things,
23 those complications that set about at a continuum
24 level. I don't actually have a picture of the
25 microstructure in the code, but it's more of a

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1 continuum level where you know how much of each of the
2 things you have there.

3 The model tries hard to avoid empiricism.
4 I don't think there's anything empirical in it. It's
5 basic physics and chemistry. The fundamental
6 treatment of a concentrated ionic solution is probably
7 the hardest part of it. We encountered many ionic
8 species. We can predict the onset of severe
9 degradations. It's not a model of mechanical failure
10 though. It doesn't have mechanics in it. Right now
11 it can be extended. It doesn't have radionuclide
12 chemistry in it right now. It can be extended to that
13 so the prediction capability as a model could be
14 extended to radionuclides.

15 Other species like boric acid things like
16 that could be put in as well with surface complexation
17 which you probably have to have if that could be put
18 in as well.

19 And those you have printouts that you can
20 look up those references.

21 I'm going to briefly say something about
22 thermodynamics. Looking back at my graduate
23 education, I think the weakest part of it was
24 thermodynamics, so I'm not going to say very much at
25 all.

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1 But looking at longer term problems, these
2 thousands of years problems, one thing you worry about
3 is what C-S-H forms do we have for a long time? Dr.
4 Glasser said that C-S-H is only meta stable. It's
5 going to change. What's it going to change into?
6 What are the properties that those things are going to
7 change into over many years?

8 One thing you might try is if you can get
9 an idea of what it will change into, you can
10 synthesize those forms in the laboratory and it
11 measures properties like do radionuclides bind to them
12 for instance. Right now, we're synthesizing some of
13 these minerals for different projects. We're not
14 measuring binding on radionuclides on them, but you
15 can't synthesize as minerals and that might be a way
16 to get at that long-term problem.

17 And I think Dr. Glasser mentioned, this
18 was -- if you go to high temperatures and pressures,
19 like an autoclave or the oil well cementing people,
20 they pump cement down two miles so you have high
21 temperature and pressure down there, you get
22 crystalline phases of a set of meta stable amorphous
23 C-S-H. Maybe we can make use of some of their
24 experience in our sorts of problems. The same C-S-H,
25 some of the same crystalline forms it's transforming

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1 into.

2 Okay, here's my opportunity to trash these
3 more. Current durability tests we have and quote
4 unquote accelerated and I don't have to re-explain
5 that to you, but it's pretty much empirical. I'm
6 probably less kind than Dr. Glasser is to them, but
7 you make a bar. You drop it in a bucket of bad stuff.
8 You measure length change ever so often and hope for
9 the best and repeat it again as many times as you need
10 to because the first six months don't tell you
11 anything. I was probably being a little severe on
12 that, but I'm a physicist. I'm allowed to be severe
13 on that stuff.

14 We're trying to get ahead of that. We're
15 trying to get a different kind of test and this is
16 just the beginning. This is not the problem solved,
17 but looking at a sulphate attack, we've done some
18 collaboration with the Portland Cement Association
19 where we don't - -we still don't know how to really
20 accelerate it. It's still empirically accelerated,
21 but we can at least improve it somewhat.

22 We don't really need a one foot long
23 mortar bar and dump it in a bucket. You can do better
24 than that. You understand microstructure. You
25 understand what the aggregates do. You don't really

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1 need to do that. So going on to smaller cement paste
2 samples, it turns out it's faster than the old method,
3 but also coupled with the SCN investigation of the
4 mechanisms of attack, what forms in the space, how
5 does attack happen and that can be used to help us
6 understand the results of the test.

7 So I worked with Paul Stutzman in that
8 group, worked on this and by looking at that, the
9 damage on the surface of these big mortar bars found
10 that most of the damage, even after many weeks was
11 confined to the first quarter millimeter or so. So we
12 thought well, why have an inch-wide, 11 inch long bar
13 and have the quarter millimeter, you might as well
14 have a small sample. That's all the degradation
15 you're going to have in the typical time of the test.

16 So we're able to change a foot long mortar
17 bar to about a four centimeter long cement paste
18 sample. It's much more controlled, better temperature
19 control, better statistics because it's a small sample
20 and you get better results much quicker. Still
21 empirical, but it's a lot better than empirical and I
22 think it's the way to go for these tests.

23 Again, repeating -- we're going to do
24 other things besides sulphate attack the same way.
25 Enumerate possible reactions. Use SEM to quantify

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1 microstructural effects. We understand the chemistry
2 of a lot of these, but we don't understand the
3 microstructural effects with the binding chemistry,
4 learn how to correctly accelerate degradation
5 mechanism and that's the hard one right there. I
6 don't know how to do that right now. We need to learn
7 how to do that.

8 And then we have small samples and do high
9 through put and if you need good statistics, you need
10 to do 200 samples. We'll do a small sample, high
11 through put measurement. You can do it a lot faster,
12 a lot quicker and get the results out. Now do it just
13 as good as before. If you can do it faster, it's
14 going to help industry a lot more. I hope we can do
15 it better as well.

16 Let me finish up with some thoughts of
17 research needs and then a summary. I think you've
18 seen a lot of research needs today. This is my
19 personal opinion for something else that's needed
20 worked on. This is my area.

21 Need to develop HydratiCA better and if
22 we're going to apply it to nuclear waste type
23 simulations, we're going to need aggregate information
24 on ionic species of interest to reactive waste
25 containment problems. We need to develop 4SIGHT more,

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1 looking at a continuous scale, but hopefully link up
2 4SIGHT and HydratiCA together so we have a
3 microstructural at a continuum level married together.
4 I think it would be a very powerful combination tool.

5 I should note here as well that in the
6 Virtual Cement and Concrete Testing Laboratory, you're
7 given a microstrucure and it predicts properties. It
8 doesn't care where the microstructure came from. So
9 if we modify HydratiCA and be able to get new
10 microstructures that are based on reactor waste
11 problems, we can use the tools already existing to
12 calculate things like lasting modulus and tensile
13 strength and tensile properties, etcetera. So I think
14 we can start attacking the problem of linking
15 microstructure mechanics, chemistry and transport
16 together into one model. I think we have the pieces
17 to do that. It's going to be hard, but I think the
18 way is there.

19 We certainly need fundamental research on
20 property accelerating degradation mechanisms.
21 Properly accelerating correct degradation mechanisms.
22 I'd like to see a lot of research done on that.
23 That's something that we can do. We can do some of
24 that, but other people have much more expertise than
25 we do on that. And it would be nice to have more

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1 research in the chemistry of this kind of applications
2 to build into these models. And I'd like to see
3 experimental measurements on the crystalline
4 endpoints, thermodynamic endpoints to C-S-H because
5 you need that kind of information to do computational
6 modeling as well at the Thomas scale or the continuum
7 scale.

8 Then the idea of the talk is then the
9 prediction tools must be based on carefully
10 characterized materials and fundamental physics and
11 chemistry, thermodynamics. It is nice to have good,
12 materials science-based standard experiments,
13 crystalline C-H-S, controlled environment, etcetera,
14 you need to have that to get prediction to be anything
15 worthwhile at all.

16 If we combined those kind of computations
17 and experiments, we should be able to make better
18 predictions, I won't say accurate, better predictions
19 of the durability of cementitious materials whether
20 used in a construction industry or used in a nuclear
21 waste, reactive waste type of application.

22 We're primarily focused on the concrete
23 industry. That's our goal, 1600-year durability. But
24 overall we're still interested in NRC, because we're
25 a government agency and want to help the country. So

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1 I think the same kind of tools can apply to this long
2 term globular prediction that NRC cares about.

3 Okay, so that's all. Thanks.

4 VICE-CHAIRMAN CROFF: Thank you very much.
5 Thank you for some very interesting talks. I think at
6 this point we'll move into the Q&A. I'd like to take
7 the first little while to just focus on the last three
8 speakers and then we'll have sort of another session
9 where we broaden out maybe the considerations and the
10 victims, I guess.

11 So with that, Bill?

12 MEMBER HINZE: Sorry. Very interesting
13 talks and particularly interested in hearing about the
14 various research lines depending on the areas of
15 expertise. There were a couple of questions that came
16 out of your presentation, Fred, that I'd like to ask.

17 You criticized a couple of things and one
18 was the QA problem in the production of cement. I
19 wondered if you could expand on that a bit in terms of
20 what are the causes. Are we lacking in protocols?
21 Should we have protocols for waste types of -- for
22 concretes used in nuclear waste problems? Are we just
23 not adequately enforcing QA standards? Could you
24 expand on that a bit?

25 DR. GLASSER: I don't consider myself an

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1 expert on QA so I would have difficulty in giving you
2 a prescriptive answer. But I what I pick up from
3 reports that I assess are descriptions of how grouts
4 and concretes are in place. I don't find any evidence
5 that the people who did the work knew what the
6 objectives were, what quality was expected, and how it
7 could be measured, and it doesn't seem at intermediate
8 stages where process was stopped and started to have
9 been any checks in quality.

10 So really what I'm talking about I think
11 is sort of a common sense approach to it. I think if
12 you want anything fancier than that, you'll have to go
13 to a genuine QA expert.

14 MEMBER HINZE: Okay, fine. Let me ask
15 another question to you. You criticized, and rightly
16 so from what you said the standard test. I had the
17 question immediately to my mind of what do you have as
18 an alternative? And you actually had a slide which
19 you kind of approached the alternatives and one of the
20 things was the tests should be better focused.
21 Standard tests have a great place in all of this, I
22 believe, and we can't just completely eliminate them.
23 But can you give us better insight into how we could
24 improve those? We heard a little bit of this from Ed
25 in his presentation.

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1 DR. GLASSER: Yes, I don't want to totally
2 preempt what we're going to say in Quebec in
3 September, but I think I can tell -- and some of
4 things we're going to recommend are quite complicated.
5 But one is quite simple and let me deal with just that
6 one alone.

7 Now you, Ed has said you take a piece of
8 concrete and you put it in a bucket. Well, it turns
9 out that depending on the concentration of the bad
10 stuff in the bucket, that how big the bucket is has
11 the important role to play in what you get out of the
12 test.

13 So we're going to recommend that if you
14 use the ASTM standard of sodium sulfate, which I think
15 from memory is 32 grams per liter, but I might be
16 wrong on that. Don't put it down in the minutes
17 without checking. You need to --

18 MEMBER HINZE: It's already in there.

19 (Laughter.)

20 DR. GLASSER: You need to use a mass ratio
21 of at least 10. That is the mass of the aqueous
22 solution has to be at least 10 times greater than the
23 mass of cement. I don't mean the mass of concrete.
24 If it's concrete you're using you're allowed credit
25 for the aggregate is being inert, 10 times greater

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1 than the mass of cement. Otherwise, you run the risk
2 of serious depletion of sulfate in the solution.

3 And we will also be making recommendations
4 about how you control the change in pH.

5 MEMBER HINZE: One of your alternatives
6 related to the need for supplemental calculations, and
7 if you start putting caveats on these results of
8 standards tests, I wonder how really standard they
9 become because people will use different calculations
10 and some will consider them, some will not. Is this
11 really an alternative to this?

12 DR. GLASSER: I think what we're trying to
13 do is make the test more reproducible, which is in the
14 spirit of things. Not to complicate it or put non-
15 standard features into the test. But I mean, I can't
16 change what ASTM have in their test specification.

17 MEMBER HINZE: Let me ask one more
18 question if I might, and this is to Ed. When you
19 talked about small sample sized testing as a geo-
20 scientist, that immediately raised red signal flags
21 because there is always a problem of representative
22 sample. In fact, Fred and some of us at lunch were
23 discussing the size factor in some of these
24 determinations.

25 Can you give us a better insight into how

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1 you can justify using the small sample and what kind
2 of restrictions should be placed upon the results?

3 DR. GARBOCZI: The reason you can do that
4 is in the -- I'll just say sulfate attack only.
5 You're not really testing a structure at all, you're
6 testing the cement. That's really all you're doing is
7 testing the cement. Is the cement conducive to
8 sulfate attack? How susceptible is it to sulfate
9 attack? So the things that you worry about is the
10 length scale of cement piece micro-structure. So you
11 can probably get away with three millimeter samples
12 because that's big enough to be representative of a
13 cement paste micro structure. The four centimeters
14 may even too big, because you're trying to test the
15 concrete then you have to go to a bunch of bigger
16 sample, concrete size sample. But the current tests
17 the mortar. You don't build things out of mortar
18 anyway. You're really only testing the cement.

19 So it's very similar. There's an ASTM
20 strength test for cement strength. You make a two
21 inch mortar cube and break it and that gives you a
22 feel for the cement strength. So it's really the
23 same kind of thing. If you're only testing the
24 cement, then why not use a cement paste size sample.
25 That was our point of view. So you know, structure of

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1 concrete is different. But cement only you can stay
2 with a small sample.

3 DR. DETWILER: I'd like to add a comment
4 here related to that and that is there may actually be
5 some things that would be somewhat different depending
6 on exactly how you mixed it for example, because the
7 mixing can effect the rate of hydration at the
8 beginning. You know, so there are certain things that
9 might affect.

10 And also I supposed in the size sample
11 you're talking about, bleeding is not really an issue.
12 But it's conceivable that it might be in that also and
13 could open up some things. So there are some
14 possibilities there and same with if you had the
15 presence of sand in there, you would have those
16 transition zones and that would have more to say about
17 the rate. Although I don't know that is such an issue
18 if you're comparing apples with apples. But certainly
19 the mixing would have an effect.

20 DR. GARBOCZI: Right, you wouldn't get --
21 your small space samples wouldn't necessarily give the
22 exact same results as the big mortar samples, but you
23 don't care so much because you're just testing the
24 cement. Yes, I agree with you.

25 DR. SCHEETZ: The other thing is that I

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1 guess I was talking to him, I guess, offline is that
2 he's doing more samples so he's getting a bigger body
3 of statistics to work with. So part of that is
4 compensated. But certainly the mixing effect and the
5 energy that a mixer puts into the mortar or into the
6 concrete is a very, very significant concern.

7 I mean, when we do our developmental
8 samples at the laboratory, we have a one-third yard
9 mixer, but when we get the data, when the rubber hits
10 the road, when we get that data for PennDOT, we have
11 contracted with the ready-mix companies to bring it in
12 and mix it in their truck, drive it into our
13 laboratory and dump the truck to do the measurements
14 on. So those scale problems are real.

15 VICE-CHAIRMAN CROFF: Mike.

16 CHAIR. RYAN: The last two presentations
17 were interesting from several points of view.
18 Professor Glasser, I was taken by your comment that
19 there really isn't a good compendium of all this
20 information. That's striking to me and that's
21 probably part of the problem that we struggle with.
22 The second part is kind of a synthesis from all three
23 talks and that is that I think it is troublesome to me
24 that we rely on tests where it's clear as a bell to
25 everybody at the table that the models are wrong.

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1 That really creates a ripple into
2 performance assessment for which there's probably a
3 high penalty being paid, that if we're overpredicting
4 failure or underpredicting success of behavior, that's
5 a bad thing in performance assessment. And the third
6 gets to the statistics issue, the uncertainty analysis
7 issue that we're kind of it sounds like we don't have
8 a very good rudder on where we are relative to the
9 center line of what we think is the best answer.

10 And without a real serious and
11 comprehensive treatment of uncertainty and probability
12 of one answer over another being right, we really need
13 to think about how to work on that. Of course I would
14 jump on Dr. Garrick's risk triplet and try to
15 catalogue them by some ranking of how important they
16 are to overall importance or risk assessment or
17 whatever it might be. But I think we need a
18 thoughtful review of what would be a really good
19 approach to do a systematic approach of these
20 uncertainties.

21 It's interesting that when we've asked you
22 to do this, though it's not really a bad thing, but
23 you've compartmentalized into your own areas of
24 specialty the risks and the uncertainties. And while
25 I appreciate the fact that this is certainly valid

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1 based on how all of you work on different aspects of
2 this area, it doesn't facilitate the bringing together
3 all the information or the systematic analysis of
4 uncertainty which then could flow into a performance
5 assessment.

6 I guess the one question I ask is do you
7 think I'm making any sense or is this crazy? I mean,
8 am I on the right track of maybe how to move ahead
9 here a bit or not?

10 DR. GLASSER: No, I agree with what you
11 said. I think that's a very good synthesis from the
12 standpoint of regulator and overseer. You have a
13 different role to play than many of the other
14 participants in this process. I think anything that
15 we can do to assist in the discharge of those
16 obligations, it is our duty to do it and I think
17 that's a very clear statement of what you need to do.

18 CHAIR RYAN: I came at it from perhaps
19 that perspective but also as a former applicant and
20 licensee, I think it's helpful from that side of the
21 fence too because then you know what the expectations
22 are and you don't have to worry if the two order of
23 magnitude difference between a test and your answer is
24 going to be good, recognized as reasonable, or
25 recognized as wrong.

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1 You know, again I want to give credit to
2 Tony Malinoskos who worked at ORNL because he's the
3 fellow that I quoted when I said if it is off by two
4 orders of magnitude, it's not conservative it's just
5 wrong. So but it really helps both sides of the
6 fence, whether it is the applicant who is trying to
7 describe reality or the regulator who is trying to
8 evaluate that assessment of a reality.

9 DR. GLASSER: These two order of magnitude
10 differences that we were looking at and I think you're
11 probably referring to the, what was it Professor
12 Kosson showed earlier this morning. I think there the
13 prediction was based on a rate model for the
14 prolongation of a particular process that was
15 effectively using fixed laws of diffusion and applying
16 them to a situation. So you've got a profile which
17 was a constant times the square root of time, that
18 governed the slope. But then Professor Kosson's own
19 data showed that in many cases you didn't get a
20 Fickian profile of diffusion, you got steps.

21 So even without the mathematical analysis,
22 you can tell there's a serious divergence. There are
23 processes occurring within the cement that have not
24 yet been built in to the model. So it's not that the
25 model is wrong so much as the model is inappropriate,

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1 which is the way I would put it.

2 CHAIR RYAN: That would be close enough to
3 wrong. I would say it is wrong.

4 (Laughter.)

5 CHAIR RYAN: But I appreciate your point.

6 DR. KOSSON: If I can comment on that a
7 little bit further, I just wanted to point out the
8 divergence between what is currently being assumed in
9 performance assessments versus what we recognize to be
10 the phenomena. And to go a little bit further on
11 something you said a little bit earlier, Mike, is that
12 I think what is really needed is a concerted effort
13 over a committed period of time. Not six months, not
14 tomorrow, hurry up and get it done today, but to
15 develop an integrated research development program
16 that feeds into incrementally into performance
17 assessments and other applications so that you take
18 advantage of data that evolves as Jim mentioned from
19 field monitoring and the like and you really put it on
20 a continuous improvement basis.

21 CHAIR RYAN: Absolutely. I couldn't agree
22 with you more.

23 DR. KOSSON: That's not what is happening
24 now.

25 CHAIR RYAN: I couldn't agree with you

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1 more. I think that is well said. There's one other
2 aspect to all this that I just think to add as a
3 comment is that I'm always nervous when people are
4 satisfied with deterministic overestimates of failure.
5 Well, it is conservative so we feel pretty good about
6 the number, we're okay. Conservative and ultra-
7 conservative estimates mask the true uncertainty. They
8 ask for phenomenology and all sorts of other things,
9 so I think there's a real tendency to rely on that
10 perhaps and I challenge that as being a little risky.

11 DR. GLASSER: They also end up with a
12 situation that you can never contain radioactive
13 waste.

14 CHAIR RYAN: Right.

15 DR. GLASSER: It's like trying to put gas
16 into a sieve. No barriers really work.

17 DR. KOSSON: I contend even further as if
18 you mask the phenomena and your assessment protocol is
19 misleading in terms of phenomena and the results, then
20 you are missing the incentive to improve and the
21 opportunities to improve what you're doing and the
22 insights to lead to improvements for a much better
23 performing system itself.

24 CHAIR RYAN: Well said. I think that kind
25 of captures the essence of it.

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1 DR. DOLE: There's a push-me, pull-me.
2 Certainly the EPA regulations sort of prescribe a
3 methodology for assessing the transport and we know
4 from our experience that the essence of that, the
5 heart of that is based on phenomena that we observe.

6 Okay, so the question is we have so many
7 different conflicting observations. Are we confident
8 enough to tip over the current methodology. I don't
9 think we're ready for that when we know, in fact, that
10 is the Agency ready to accept that they based all
11 these assessment on the wrong basis.

12 CHAIR RYAN: Then again, maybe somewhere
13 and I'm not trying to exactly be the champion for oh,
14 let's just turn on the PRA switch and go all wild with
15 that. But there is a way to get at your question I
16 think, Les, and I think that is to systematically
17 assess these uncertainties. Which ones are important
18 to performance, which ones are less important to
19 performance, and somehow line them up in a way where
20 I think we can attack the tough ones that need to be
21 answered first and maybe order them in some way after
22 that.

23 If a phenomenon is interesting but not
24 important to outcome of issues related to performance
25 assessment, it's kind of a secondary thing.

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1 DR. KOSSON: I think it's important to
2 recognize that you have to make the best decision
3 based on the information that you have and
4 understanding that you have available at the time.
5 However, a lot of what we were seeing and what things
6 propagate right have an appearance of being stuck in
7 time. That not taking the last decade of
8 understanding of computational advances, of analytical
9 advances, and incorporating them into the next
10 generation.

11 Inherently, there's a lag time in these
12 things, but also there's got to be a commitment to
13 incorporate them. Not just maintain the status quo.

14 CHAIR RYAN: I have one final question and
15 sort of off this topic, but we've talked about
16 concrete in terms of small, medium, large, and really,
17 really large constructions. And with the issue of
18 seismic, it's a very specific point but why do we
19 build such big structures if we're interested for
20 waste disposal, if we're interested in seismic
21 control? I know very little about seismic analysis,
22 but I know one big block is not as good as five little
23 ones.

24 DR. DETWILER: I'm not sure how important
25 seismic activity is in some of these things. I

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1 realize that the Hanford site, for example, does have
2 some, you know, is subject to seismic activity and I
3 would assume that the Idaho one is too just from
4 having grown up in that area. But I think actually
5 for underground structures, I don't think it really
6 matters all that much.

7 By the time it is actually underground, if
8 you're talking about underground tanks and that sort
9 of thing, I'm not sure that the stresses transmitted
10 are really all that big a deal. So something we make
11 an issue of, but I'm not sure that it really matters.

12 I would assume that the really big
13 problems with what might happen underground would have
14 more to do with soil settlement, differential
15 settlement and that kind of thing.

16 CHAIR RYAN: Okay, thanks for the
17 clarification.

18 DR. GLASSER: I'm not like Rachel. I'm
19 not an expert on it, but from the work that we've done
20 on cements intended to be used in seismic areas, what
21 the geophysicists tell me is that what's much more
22 important than size is coupling. Not to leave void
23 spaces, not to leave gaps, but to ensure that concrete
24 is in contact with -- well, I've only worked over hard
25 rock mines in this context, but the contact is good.

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1 And that's more important than the actual
2 size of the structure, because the structures that I
3 have experienced with gold mines have miles of large
4 diameter tunnel and very large underground crushing
5 plants. So scale is large.

6 CHAIR RYAN: Okay, thanks. That helps.

7 Ruth?

8 MEMBER WEINER: We all come at these
9 questions from our own backgrounds, and I've just
10 learned more about cement than I ever thought I would
11 know and I sure can't remember it all.

12 (Laughter.

13 MEMBER WEINER: But this is fascinating.
14 It seems to me that this is for the physical chemists.
15 It seems to me that the concrete has a lot of surface
16 area. Does it act ever as an absorbent? Can you
17 absorb and desorb water contaminants from concrete?
18 Could it ever act like an absorption column or an ion
19 exchange column or something like that?

20 DR. GLASSER: I think the answer to that
21 is yes.

22 MEMBER WEINER: Well, could we then use
23 that property to isolate, sequester if you will,
24 radionuclides in these tanks?

25 DR. GLASSER: I think cement will have a

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1 response for almost all nuclides where at low
2 concentrations there will be absorption is what you
3 refer to. But then depending on the particular
4 species, you shift into a regime of precipitation
5 where they form some solubility limiting phase with
6 the cement.

7 Now there is a huge spread of
8 concentrations over which that occurs. For example,
9 for cesium, you never really attain the point where
10 you get a precipitate phase. There's not solubility
11 control. But for most di- and trivalent radionuclides
12 and possibly for some anionic species, you will have
13 a boundary somewhere and it's not been found possible
14 to predict where that boundary is. But especially if
15 you don't have to do the experiment with the
16 radioactive species, if you do it with an active
17 stimulant, nickel or chromium or something like that.

18 We have seen examples today where the
19 precipitation phase was noted, but the concentration
20 is lower than that. Yes, you will get absorption.

21 MEMBER WEINER: Well, since radionuclides
22 that have stable analogs behave chemically and
23 physically, according to what they are chemically and
24 physically and not what they are radiologically,.
25 would this be a fruitful area to examine for

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1 sequestering radionuclides?

2 DR. GLASSER: Well, there's two comments
3 that I would make. First of all, the worry always is
4 in the minds of people that what you absorb can easily
5 be desorbed. That tends to be an element of
6 reversibility about many of these processes. So the
7 worry is that while there might be binding by
8 absorption, if conditions were changed you would get
9 desorption. Nonetheless, there's a hold up in the
10 transport process.

11 Secondly, at the -- I'm surprised nobody
12 mentioned the natural analog site at Makaren in
13 Jordan, because there has been a lot of work done on
14 the absorptive potential of the calcium silicate
15 hydrate for various species. Now obviously you can't
16 do it on everything. You have to do it on what nature
17 has provided you with. But there are quite a bit of
18 data in that area as a result of the joint Swiss-
19 Swedish-British initiative to study the area. And I
20 have been onsite and it is fascinating.

21 It's on a huge scale and you can actually
22 see if you go during the rainy season, you can see
23 springs coming out at the base of information, stick
24 a piece of pH paper in and bingo, you get pH 13. And
25 it's wonderful, all these predictions that you made

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1 you can see them happening.

2 DR. DOLE: You know, the idea that --
3 certainly the work at NIST can talk about the fabric
4 of the C-S-H is simulated by discrete granular
5 entities. And then there is the surface of the cement
6 and I don't know whether it was a miscommunication is
7 that cement as a fabric of a mass may be granular and
8 have internally have very high ionic exchange capacity
9 or capacity to promote insolubles, but as an exchange
10 media to outside flowing water, there's not ready
11 transport within the mass of the cement, because the
12 apparent diffusion coefficient is the best thing we
13 can use to describe are exceeding low. Ten to the
14 minus twelve, 10^{-16} , almost imaginary numbers for
15 actinides and many of the materials.

16 So the effective transport within the
17 small masses, now you could postulate that you could
18 have little balls, you know, through which you could
19 percolate like backfill or something like that. And
20 then they would be very effective.

21 MEMBER WEINER: Thank you, that's very
22 clarifying.

23 Another question I had and I think this is
24 for more than the last three speakers. Is clearly, in
25 looking at the tanks using as cement to stabilize the

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1 underground tanks at Savannah River and Hanford,
2 you're not looking at structural strength. You're
3 looking at the ability of cement to hold back the
4 radionuclides. Do you design the composition?

5 What processes go into that design? Is
6 there some overriding formula that you use or is it
7 empirical, you try it out?

8 DR. LANGTON: Well, at Savannah River, the
9 tank fill material, there's multiple layers. And the
10 layer in contact with the waste was actually designed
11 on a concept that the concept was cement hydrates to
12 C-S-H, the more C-S-H, the better the sorption.

13 That lower layer also contained slag
14 cement to achieve reducing properties. And silica
15 fume to control microstructure to act as a pozzolan.
16 So yes, there was a thought process that went into
17 that. Actually, Rachel is the one that came up with
18 it.

19 MEMBER WEINER: So it's basically a
20 designer cement, if you will, for the particular
21 purpose.

22 DR. LANGTON: A blended cement and there
23 were some leaching tests performed.

24 Now unfortunately for the contaminants of
25 most concern which are the long-lived anionic species

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1 for technotate, the neptunium, iodine sorption in
2 cementitious materials, whether they're -- I won't say
3 they're reducing, nonreducing, just ordinary
4 cementitious concrete mortar materials is 50 percent
5 at the most or less. Fifty percent or less.

6 So that would be a KD absorption
7 coefficient of between 1 and zero. Now if you add a
8 reducing agent, I can't say that you're having
9 sorption by a different mechanism by precipitation, by
10 chemical reduction and subsequent precipitation.
11 You're lowering the source term in solution by
12 precipitating it rather than absorbing it.

13 So the effective KD when there's a
14 reducing agent present would be lower. And I measure
15 values like 6,000, 5,000, 6,000, that range.

16 But absorption is the wrong mechanism.

17 MEMBER WEINER: I wasn't suggesting either
18 a single mechanism or that sorption would be -- I
19 thought there might be. But you're quite right that
20 what you're looking at is the effective case D. And
21 that's -- so you design your -- the question is you
22 design your cements -- you design the system to
23 provide you with the effective case of D. Is that
24 correct?

25 DR. LANGTON: Yes, the two knobs that are

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1 tweaked right now are ph and KD. Oxidizing, oxygen
2 potential and --

3 MEMBER WEINER: And KD.

4 DR. LANGTON: Well, I'm sorry, pH and EH
5 are the two knobs that are tweaked. Now other things
6 can be done. Additives, getters could be added for
7 cesium, add a zeolite for cesium or for strontium. So
8 there are other knobs that could be tweaked, but we're
9 not doing that at the moment.

10 DR. DETWILER: We also have some other
11 considerations that when we were formulating that
12 particular route and some of them just had to do with
13 can we pump it into place, will it flow? Because we
14 knew we were going to be placing it at very limited
15 number of entry points. And so we had to make sure
16 that it could be placed there and that it would flow
17 to the edges of the tank and still retain its
18 integrity as grout.

19 So that was one reason why the silica fume
20 was in there. Had it done nothing else, it was doing
21 something very important in maintaining the integrity
22 of the liquid grout to get all the way out to the
23 edges of the tank and not have segregated into its
24 separate components.

25 So there were some engineering aspects of

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1 it as well and we did mock ups. there were a number
2 of mock ups that were done that tested that ability
3 for the grout to be mixed and trimmed into place and
4 that it would flow out. It would flow over obstacles.
5 All that sort of stuff was part of the testing. And
6 that's really more of an engineering concern and just
7 constructability.

8 MEMBER WEINER: That's a very good point,
9 by the way.

10 DR. KOSSON: If I could just comment also,
11 when -- getting back to Mike's comment about wrong
12 models and your comment about KDs, when the mechanism
13 of retention in the cement matrix or cement paste is
14 one of precipitation/dissolution, modeling it as an
15 absorption process, as a KD approach which I've seen
16 frequently done is just plain wrong.

17 Or as an effective diffusion coefficient.
18 You need to couple dissolution --

19 DR. SCHEETZ: What you have to look at
20 when you're doing your designer concrete is you have
21 to look at the different mechanisms by which the
22 radionuclides are sequestered.

23 You have sorption. You have
24 precipitation. You have a raisin bread model where
25 it's -- where the waste is a raisin in a raisin bread

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1 model and it's just physically encapsulated.

2 So what you need to do is you need to look
3 at all of the potential ways in which the waste is
4 going to be sequestered. Not all components will be
5 sequestered in the same manner.

6 Chris was talking about the anions. Anion
7 hydroxy anions like selenite, arsenate, chromate,
8 protectnate, they tend not to be tied up very readily
9 and they -- and we see this in other environmental
10 fields.

11 This is why we go to the use of slag, so
12 it reduces those down from an anion to a chadian and
13 chadians are retained. So you need to look at the big
14 picture. You need to look at what mechanisms are
15 available. You will need to look at what elements are
16 -- you're trying to tie up and which ones are going to
17 best suited to which mechanism and then you try and
18 integrate all of those mechanisms into your grout.

19 And you have the other thing to do and
20 what we've been hitting upon here, we've been bouncing
21 back and forth, but nobody has enumerated it. We have
22 grout people. We have structural engineers. What
23 sets in between is material scientists. These are
24 materials problems. They're not -- and that materials
25 chemist has to have his foot in engineering as well.

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1 So we have to balance the mechanical and
2 the engineering constraints with the fundamental
3 materials properties of what you're trying to do. And
4 that's what Chris does.

5 MEMBER WEINER: Thank you.

6 DR. LANGTON: Designing waste forms to
7 past tests at 28 days to demonstrate that contaminants
8 have been stabilized is straight forward. The
9 contaminate chemistry of the various species that
10 we're interested in is well known.

11 Designing waste forms for placement. It's
12 another set of tools, another set of techniques and
13 strategies. That's well known. And can be worked
14 around. Can be engineered around.

15 What's missing is long-term performance
16 predictions. For cement waste forms, the long-term
17 issues depend on migration or movement of water and
18 air. And the air contains two constituents of
19 concern: oxygen and CO2. That's what it gets down
20 to.

21 How do you predict how the transport of
22 water and air in the environment in the land fill, in
23 the waste form, through the containment, what
24 conditions that containment or waste form are going to
25 be in, what conditions the cap, the cover are going to

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1 be in; how it's shedding water, how is it transmitting
2 water. And how do you do that for 10,000 years? Or
3 for 1,000 years?

4 DR. DOLE: Yes, just to pick up on a
5 small, delicate thread that she touched on is that,
6 was that designing a waste form to pass the test, and
7 take you down the wrong path, specifically the TCLP,
8 comes with a chelating agent, acetic acid, so if I
9 design a waste form that blinds the acetic acid, and
10 I apparently pass the test, I had generally made a
11 waste form that's very geochemically unstable. That's
12 a caveat you have to look at. If you specify a test,
13 it can sometimes push, have unintended consequences of
14 driving you to waste forms that --

15 MEMBER WEINER: Yes.

16 DR. DOLE: -- less desirable.

17 DR. LANGTON: The chemistry is known for
18 people that want to look into it.

19 DR. DOLE: Yes.

20 MEMBER WEINER: Could you talk into the
21 microphone? We'd hate to miss any of this.

22 DR. LANGTON: Oh sure. But the chemistry
23 in the qualification, we call it the qualification
24 testing, the chemistry that needs to be adjusted or
25 that shouldn't be adjusted, is well known for the

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1 contaminants that we're interested in.

2 MEMBER WEINER: I have a final question
3 for Dr. Garboczi. And that is, in your model, how do
4 you model your chemical reactions? Do you model an
5 equilibrium? Is it based on minimizing the Gibbs free
6 energy? What's the overriding way that you do that?

7 DR. GARBOCZI: That might be a detail
8 beyond me. This is for Jeff Bord. All the chemistry
9 takes place in a node. I will ask him to email you
10 the answer to that.

11 MEMBER WEINER: Thank you.

12 DR. GARBOCZI: It's better than me trying
13 to wing one.

14 MEMBER WEINER: Thank you.

15 VICE-CHAIRMAN CROFF: Thanks. I think
16 that before going to Jim, we seem to have merged out
17 of the, to the questioning the entire group here. So
18 I think we should assume with it at that point.

19 MEMBER CLARKE: I ask permission to
20 officially begin the round table.

21 VICE-CHAIRMAN CROFF: Yes, I think we're
22 in the round table.

23 MEMBER CLARKE: And the rule that he or
24 she speaks last is also applicable to he or she
25 questions last. So, a lot of the things that I wanted

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1 to ask have already been asked.

2 But, picking up on the recipe, one of the
3 things I heard from all of you was that these
4 processes may be important, this may be important,
5 that may be important. What I took away from that was
6 this is very waste-specific and this is very site-
7 specific, and so the value of having a recipe that
8 addresses as many things as you can with that
9 knowledge is very important and that, I think, gives
10 us what we need for the QA, because we need to tailor
11 the QA obviously to the recipe and, you know, what
12 performance we're looking to.

13 And I think, Christine, you do this, I'm
14 sorry, do you not? You don't, you know, you mix your
15 cake and apply it and you can control the things you
16 need to control. Is that a fair statement?

17 DR. LANGTON: Yes. A problem with the Q.
18 We do. And the QA, we have as good a QA for concrete
19 vaults or for waste forms as any construction job has.
20 The problem with QA is that, for concrete, for
21 concrete, in general, compared to a product coming out
22 of a factory. The factory process is over in a short
23 time. The raw material, temperature, particle size
24 features can be adjusted to meet any specifications
25 that are required by the process.

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1 To a large extent, the reactions that take
2 place for the cement materials and for the waste
3 forms, the cementitious materials like concrete in
4 waste forms, take place in ambient conditions. And
5 the placements are done under outdoor ambient
6 conditions, which are variable.

7 So, the QA that's done is to get a certain
8 placement property, a certain flow or a slump or some
9 placement at whatever conditions exist outdoors. And
10 they'll be a range on the amount of water that can be
11 added, the amount of admixture, the need for
12 admixture, and that takes care of the placement.

13 But the curing, and so ambient conditions
14 over the range of ambient, of normal ambient
15 conditions, makes a big difference. And there are
16 adjustments made, just like routinely they're make on
17 the fly for concrete. The curing process also takes
18 place over a range of ambient conditions, and the
19 curing period is not like a product in a factory where
20 it's a few hours. It's weeks, months, or longer.

21 So, the QA problem is a lot bigger. And
22 it's a lot different than what a construction job
23 would experience. Construction jobs can test the 28-
24 day strength, if that's what it's designed for, and if
25 it passes, they're finished.

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1 For our QA, for waste form QA, do we test
2 at five years, and then take the waste form out if it
3 didn't meet the specification? When people are
4 talking about QA, they're trying to mix concrete and
5 disposal and they don't mix.

6 MEMBER CLARKE: So you test as you go.
7 That's the best you can do, I guess.

8 DR. LANGTON: You test as you go and I
9 would say that the requirements are met, the
10 specifications for placement are met. But there's a
11 lot that happens after placement and things need to
12 happen in order to achieve placement. As I said, more
13 water, admixtures, more vibration necessary, and
14 that's how concrete jobs work also.

15 DR. SCHEETZ: Let me add a cautionary note
16 here. A lot of what we talked about when we
17 criticized QA was done on, you know, on engineering
18 structures and we're having just fits with this right
19 now building Interstate 99 out through central
20 Pennsylvania, where we're placing a ternary mix of
21 flyash and slag concrete on a bridge deck and it comes
22 out and the people who are placing it judge it to be
23 sticky. So, not knowing, and this is back to what
24 Fred's point is, not knowing that this was an
25 experimental design, not knowing the properties of it,

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1 not knowing the objective of it, not knowing its
2 performance, they arbitrarily decide to mist it. So
3 they've changed the water to cement ratio. And the
4 concrete is not what was intended to go down. And I'm
5 sure that Rachel, with her vast experience, probably
6 can enumerate dozens of these types of cases.

7 You have the situation where, on the fly,
8 this is out in industry, where you have on the fly
9 people making decisions without the full background
10 and scope of what the consequences of those decisions
11 are. And you know, if you're the PennDOT inspector
12 standing there, what do you say? They may or may not
13 see it. They may or may not, you know, have anything
14 that they can do about it at the time, because he's
15 already added the moisture.

16 So it's those types of QA concerns are
17 less manageable in the real world than in, under a
18 controlled condition where, you know, everybody, if
19 you're going to pour a tank, you're going to stand
20 around and look it, look at it. And I would think
21 that, under those circumstances, your ability to
22 control the QA is better than it is a three o'clock in
23 the morning in central Pennsylvania.

24 DR. DETWILER: I would like to add to
25 that, though. I now work for a company that does

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1 construction testing, you know, for more routine
2 things than what we're talking about here. But, we
3 have tried to position ourselves in the market as a
4 company that really cares, that always provides a
5 qualified technician and all that. And I can tell you
6 that a lot of that has to do with providing good
7 training for all of your people.

8 And we have one of the issues that we have
9 is that every summer we hire a lot of student interns
10 to come in, and so we'll increase our number of
11 personnel from 400 and some to 500 and some over the
12 summer.

13 So, we have a big training program that
14 goes on where we teach everybody and we make sure that
15 they all get certified by, in our case ACI, but, you
16 know, you would have your own program whatever that
17 was. And then, every one of those junior people is
18 assigned to a senior person. And that senior person
19 is also on-site with them. So that every junior
20 technician has a senior technician that he can go to
21 if he doesn't understand something and who is watching
22 over him. And that senior technician reports to a
23 project manager and I serve as a resource to these
24 people.

25 A lot of the senior technicians have my

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1 number programmed into their cell phones so they can
2 call me from the site, and I can provide them with any
3 kind of information or anything else if there's any
4 question.

5 So that, these things have to do with, you
6 know they're not really technical issues any more.
7 They're management issues. They have to do with how
8 you organize it, how you support people, how you train
9 them, how you communicate, make sure that they know
10 where to go if they have a question, that you have
11 somebody there who's willing to answer the question
12 without biting their heads off. You know, all of that
13 sort of thing goes into that and I think that it may
14 not be done perfectly every time.

15 It certainly is not as easy as in a
16 factory because, for example, we can't control the
17 weather. But, certainly, you know, we can tell our
18 technicians if we are trying to design and often it is
19 more of a prescriptive specification. In our case you
20 have to meet a certain mix proportion. That batch
21 ticket will tell you what went into that particular
22 batch. If they ask you whether you can add water and
23 you see that there's already all the water they're
24 allowed, you say no, it's not allowed.

25 MEMBER CLARKE: Reminding me that there is

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1 a distinction between QA and QC and we started out
2 talking about QC and you're talking about QA so I
3 think we're covering it. It, also this need for a
4 site-specific, waste-specific recipe impresses me that
5 there's a need for a site-specific, waste-specific
6 leeching test as well. And so we've talked about the
7 TCP and how the TCP doesn't do any of this. The other
8 standard tests are probably appropriate. I mean I
9 don't know of impressibility and things like that.
10 But maybe they're just fine. But if we're looking at
11 the potential for leeching under certain conditions,
12 I would think that would be --

13 DR. KOSSON: Jim, I couldn't disagree with
14 you more.

15 (Laughter.)

16 MEMBER CLARKE: I set you up as best as I
17 could.

18 (Laughter.)

19 DR. KOSSON: Rather than a site-specific
20 leeching test which I think would be an unmitigated
21 disaster, because of all the different permutations
22 and the like, you want to have leeching tests that
23 have net measure intrinsic parameters, properties of
24 the material and you want a closely couple of both the
25 laboratory measurement as Fred inferred, the

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1 interpretation of the data and the data reduction as
2 well.

3 It seems a bit crazy that we can use
4 computational models and simulation in a host of
5 different ways, but if you want to take a test result
6 from a laboratory that you have to interpret it by
7 hand, but I think there are appropriate algorithms for
8 interpreting tests and that the rigor that goes into
9 a design basis for a building or for anything else
10 also has to go into the design basis for a test.

11 Both how you're going to use with the
12 output is, all the parameters being modeled and then
13 verified for the test itself so that you can
14 appropriately get the parameter estimates out of the
15 test that you want and then use it in a feed-forward
16 way and to either as quality control or in your
17 performance assessments models.

18 MEMBER CLARKE: And the other thing I
19 would just throw out to all of you -- by the way, one
20 other thing let me throw out. I enjoyed the exchange
21 on deterministic versus probabilistic. I think that's
22 an interesting area to look at and what I took out of
23 that maybe I didn't take out what I should have, but
24 for certain valuations, where we're looking at a
25 process and maybe we can control it fairly well, we

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1 want to really understand the underlying science
2 deterministically.

3 It would make a whole lot of sense, when
4 we're scaling out to a facility where we have
5 variability and uncertainty of large proportion and
6 then maybe a probabilistic route is the way to go. So
7 I throw that out to see what you think of it, if we
8 need to talk about it.

9 And the last thing I would throw out would
10 be how do we put all this together? I mean we have
11 heard so many different things, so many processes that
12 may be important under some conditions, maybe less
13 important under other conditions. To use a word
14 that's a little overworked, you know, how do we have
15 a road map that takes us to what we need to do? I'll
16 say it again, for a specific waste type and a specific
17 environment?

18 And do this in a risk-informed way so that
19 we know what's important, what is less important.
20 What's the best way. We've all agreed that predicting
21 -- we're driving way beyond our headlights and our
22 ability to predict much beyond our experience, it's
23 questionable, way beyond our experiences, very
24 questionable. So how do we deal with all of this?

25 I just throw that out to all of you.

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1 DR. SCHEETZ: With an integrated program,
2 with an integrated program that's got some foresight
3 to longevity, you can't work on a six-month contract
4 on a six-month basis and do it hodge podge and willy-
5 nilly.

6 We've all worked at this and all of us at
7 this table at various times and in various conditions
8 for various periods of time. And I don't see the
9 integrated program that needs to be -- somebody has
10 got to do it and unfortunately that takes the crinkly
11 green lubricant and the commitment to stand behind it.

12 But what we need to do here is look at
13 Yucca Mountain as an example. I mean look at all of
14 the vast diversity of backgrounds and fields and
15 models and everything that went into coming up with
16 the performance assessment for Yucca Mountain,
17 effectively we've got to do that here, but not
18 necessarily on such a grand scale.

19 But we have to make a commitment to bring
20 the material scientists together, bring the civil
21 engineers together, bring the people who are doing the
22 thermodynamic modeling, bring the people together who
23 are doing the computational modeling and integrate
24 them into a program that the output of which will --
25 we need to get them to talk together and the outcome

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1 of that program then can be used for the ultimate
2 performance assessment.

3 CHAIR RYAN: And again, I'd offer my
4 friendly amendment to your summary that that has to be
5 structured so that we're touching on the things that
6 are important to risk in the context of performance
7 assessment.

8 DR. SCHEETZ: Absolutely.

9 CHAIR RYAN: As a priority from the top
10 down.

11 DR. GLASSER: I think you might have to
12 face some hard choices, for example, I am not
13 optimistic that we can cope with conditions which are
14 fluctuating, wet one moment and dry the next.

15 It might be necessary to come back and say
16 well, in the sort of time scale that you envisage,
17 even allowing for a substantial component of
18 additional research, we're not going to be able to
19 cope with fluctuating conditions. You must choose a
20 repository siting which is going to be permanently dry
21 or permanently wet or whatever, but not fluctuate back
22 and forth between states. This is one of the things
23 I was referring to earlier when I said there needs to
24 be a more holistic dialogue.

25 Cements are not like a bandage to cover up

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1 other deficiencies and inadequacies . It's not true
2 just for cements. It's true for any barrier material
3 that you're going to use. You have to have a fairly
4 particular and relatively constant set of conditions
5 if you really want to know what the future performance
6 is going to be. You can't deal with too much
7 fluctuation, too much erratic, unpredictable behavior.

8 MEMBER WEINER: I could make a comment.
9 We have an example in this country where we actually
10 did it, did a performance assessment and acted on it
11 and are putting wastes into a repository which in
12 theory is going to sequester it for 10,000 years and
13 that's the Waste Isolation Pilot Plant in New Mexico.
14 And it does exactly what Dr. Scheetz said. It took 28
15 years and a great deal of money and an integrated
16 performance assessment with a huge variety of
17 disciplines and the license application fills several
18 miles of library shelves, and that's it.

19 CHAIR RYAN: I think that's an interesting
20 example. You know, I really grudge that folks think
21 about what is in the waste before you decide that's
22 the model to follow. There's a lot of actinides and
23 lots of long-lived materials. Those time horizons are
24 meaningful, but if you look at say pretty much the
25 commercial low-level waste, with the exception of

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1 source material uranium, the show is over in 300 years
2 pretty much.

3 MEMBER CLARKE: Also, I think we're
4 talking primarily today for the waste determinations
5 applications about near surface disposals.

6 CHAIR RYAN: Right. So it's a whole
7 different kind of setting and I think the point is
8 that, and I take the point clearly that the setting,
9 you know, very much can drive the bus. Whether it's
10 the fluctuations of, you know, is the water table
11 going up and down through your waste zone? I mean,
12 that's always fun to figure out. Or is it static
13 either in the saturated or unsaturated zone and so
14 forth.

15 I mean those things all help shape the
16 framework in what you're going to, I think, be well
17 served by trying to assess uncertainty. And then the
18 waste forum and all the rest of the things that have
19 been talked about today. Again, I see a framework
20 shaping up here as kind of the way to think about.

21 And again Jim, along the lines that you
22 said that I think you can -- I very strongly believe
23 that a waste site is qualified in the first phase of
24 its life through licensing or permitting, whatever it
25 might be. But I think there's a tremendous

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1 opportunity that is often not taken advantage of when
2 people do monitoring.

3 Monitoring is done for two reasons:
4 compliance demonstration with something, a
5 concentration at a location or a concentration you
6 plug into a dose calculation or something of that
7 sort. But if you take the second step and monitor for
8 modeling confidence building, you know you can do a
9 lot to say well, we were on the right track or we can
10 make a course adjustment and may get on a better
11 track, or whatever it might be with regard to
12 performance.

13 I use the example in my own class where
14 are the most stream samples taken in a large facility?
15 Well, on the bridge where it crosses the road because
16 it is the easiest place to get to it. So often the
17 sampling that's done for compliance, that might be
18 perfectly fine. But is that the best place in the
19 surface seismologic system to get a system so that you
20 can do other things to understand the model? Perhaps
21 yes, perhaps not. So I think that second step can
22 help break the conundrum that we're stuck with and we
23 really don't know enough.

24 I mean, there are ways to get at it. It
25 might be incremental. I think that word was used

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1 before but it's a way to proceed. Again, if anyone
2 thinks I'm crazy just pour a little cold water on me.
3 It seems like a good idea to me.

4 MEMBER CLARKE: Well, yes, and you know
5 how I feel about that as well. But I wanted to see if
6 this is going on. Is there an integration effect?
7 How do we sort all of this out? Do the best thing for
8 the best situation and the best location.

9 VICE-CHAIRMAN CROFF: Are you done?

10 (Laughter.)

11 DR. GLASSER: A further example is
12 integrated approach. It's for a repository which is
13 in clay and the clay is pyritic and then the
14 operational phase, well the testing in pilot plants
15 has been going on since 1974 or 1975 and the
16 repository will shortly become operational in three,
17 four years, something like that. So it's going to
18 have -- and then it will be another 30 years before
19 it's closed. A long life.

20 And it turns out that in the performance
21 assessment that the serious worry is that pyrite in
22 the clay will oxidize by leakage of oxygen through
23 tunnel linings.

24 We know that this result in production of
25 thiosulfate and the impact of thiosulfate on canister

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1 corrosion and concrete durabilities, we're not very
2 certain about it. So at quite an early stage it was
3 decided that what should be done was to simply limit
4 oxygen leakage into the repository surroundings. Now
5 all sorts of things were considered including way out
6 things like people going around in space suits because
7 the whole tunnel was filled with argon or something.

8 But in the end it came down just to common
9 sense because the measured leakage of oxygen through
10 the tunnel lining was extremely low. So it was
11 decided to leave the tunnel in air or fresh air, but
12 simply to make sure that there were no unsealed access
13 ports where the tunnel atmosphere could come into
14 direct contact with clay. And that is working very
15 well.

16 So sometimes if you deal with these
17 problems on a one-off basis, but if you deal with them
18 in good time and integrated into the overall
19 operational plan, testing plan and the operational
20 plan for the repository itself, you avoid problems.

21 DR. KOSSON: Let me jump in also. If
22 somebody could put back up slide 18 that I used,
23 because I think that that in turn serves as a
24 framework for looking at an integrated approach for
25 this that you might want to be thinking about as you

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1 go forward in terms of coupling mechanics with
2 chemistry and the various aspects.

3 (Pause.)

4 DR. KOSSON: No, the Power Point, please.
5 Lower right. Slide 18, I believe. But I think you
6 want to be in the Power Point one because Adobe --
7 there you go. If you go to 18 and go to the slide.

8 Basically, integrating the various
9 conceptual models of testing in the simulation, I
10 would suggest that this an approach to think about how
11 you might want to tie it all together.

12 CHAIR RYAN: Yes, and I would add a couple
13 of steps. I think lots of folks, and I don't mean
14 this as a criticism specifically, but tag and
15 sensitivity in certainty analysis boxes at the end is
16 part of it. The other part of it is figuring out what
17 is sensitive and what isn't, what is certain and what
18 isn't, and one of those things is important to
19 whatever your measure of risk is through the system.

20 DR. KOSSON: I agree completely.

21 VICE-CHAIRMAN CROFF: I think at this
22 point, if it would be good, there are a few other
23 people we need to get questions from. I'm going to
24 try one and I'd like to try to follow on something
25 that Dr. Langton said a little bit earlier in response

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1 to some question. And that was concerning the, you
2 know, basically there's two materials coming at this
3 cement that can affect it. There's the water and the
4 air with the two components of the air.

5 And I was thinking with an analogy, about
6 analogies, with the Yucca Mountain Project, where of
7 course corrosion of metals and the waste package is a
8 great concern and an equally problem and maybe a
9 greater problem is predicting the chemistry or the
10 atmosphere, if you will, inside that repository over
11 long times. And I'm wondering is it possible that the
12 greater issue for cements is predicting the
13 environment that it is in as opposed to its behavior
14 given that you know the environment?

15 DR. LANGTON: The environment to a large
16 effect determines the condition that the cement waste
17 form of the concrete will be in. If there is no water
18 coming into the system, none of these degradation
19 processes are going to take place. They all require
20 the presence of water. Chemical durability of a
21 contaminate that's chemically reduced, like
22 protectnetate going to technetium sulfite, technetium
23 hydroxide, is dependent on whether or not oxygen gets
24 to it, is transported to it.

25 And then the contaminants need to be

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1 transported out through the fluid phase, an
2 interconnected fluid phase in the pore structure of
3 the waste form of concrete and soil.

4 VICE-CHAIRMAN CROFF: At this point, what
5 would you say is our predictive capability for the
6 I'll call it the environment that's coming at the
7 cement waste form? Is it mostly assumptions or do we
8 have some, are we actually able to get at it from some
9 predictive capability?

10 DR. LANGTON: There are sensitivity
11 studies, uncertainty studies, but the sensitivity
12 studies I think are covering a larger range. And no,
13 that's really the big problem. We have a lot of
14 information to design for situations that we know
15 about. The problem is that movement of air and water
16 into and through the system control the performance
17 and we don't, that's unknown.

18 One of my questions was if you have a
19 waste form that you were happy with, was performing
20 right on target, right just the way you wanted it to,
21 improving for 100 years, do we have a way of saying it
22 is going to be okay in a thousand or five thousand or
23 ten thousand years? So I personally would look at,
24 I'd reevaluate the risks. How bad is it if technetium
25 leaches out faster than what is acceptable by current

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1 calculations? Exactly what is that, not exactly but
2 how much do we want to pay for mitigating that risk?

3 And it's really the long-lived isotopes
4 that we're concerned about. Long-lived isotopes and
5 stable isotopes, I supposed.

6 VICE-CHAIRMAN CROFF: Okay, thank you. I
7 think at this point, do any of the speakers have
8 anything that they want to follow up on or a question
9 they would like to pose to another speaker?

10 Okay, seeing none there, NRC staff
11 questions?

12 Pete?

13 MR. HAMDAN: Just one question or comment
14 and they are to many of us we've got a good education
15 on cement here, me included. Thank you very much.
16 Because of the time frames we are talking about, it
17 seems to me that the solution may lie in us having to
18 change our approach to performance assessment. And by
19 that I mean instead of doing it as a one time shot,
20 uncertainties and all, and come up with results that
21 probably we cannot defend, and assumptions we don't
22 know enough about, it seems to me if we follow an
23 approach like what David Kosson hinted on earlier, a
24 PA, a performance assessment, that's carried over many
25 years, let me call it -- I'll give it a name "a High

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1 Maintenance PA". And that would allow you to give you
2 a framework to work on and evaluate, to systematically
3 evaluate your uncertainties over time as you go along.

4 It will allow you to make more use of your
5 monitoring data. It allows you perhaps to even take
6 advantage of new PA technologies and you don't have to
7 defend it so much at the very beginning. And you
8 know, we can choose to go and do that approach
9 ourselves or I think ultimately we'll be forced to do
10 that. And I would love to hear your comments.

11 VICE-CHAIRMAN CROFF: Anybody going to
12 leap into that one?

13 DR. KOSSON: Well, I'll leap into it. I
14 think you do performance assessment, you ought to
15 revisit to see how well your predicting even the near
16 term and updating it on a regular basis. I wouldn't
17 say continuously, because that would be a nightmare,
18 but at regular intervals and take the short term
19 monitoring data that Jim was referring to and update
20 your models based on the science, will certainly help
21 you understand that site and the next one you have to
22 do. Because we're going to be doing these for a long
23 time when you look across the complex.

24 DR. SCHEETZ: I think what we have to do
25 is we have to take it and look at a paradigm fit, and

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1 I think Chris was the one who brought this up. I
2 mean, what we've been looking at now is the situation
3 where we're going to close it and we're going to walk
4 away from it and we have to be able to leave it
5 without looking at it.

6 And that's not going to happen. If you're
7 going to look at, if you're going to take and use
8 advantages of evolving skills and evolving knowledge,
9 as Mike said, you can monitor for two reasons. You
10 can monitor for compliance or you can monitor for
11 knowledge. And you can use the long-term monitoring
12 that you're doing and apply it to evolving knowledge.
13 I mean, let's not be conceited that what we know today
14 is going to be valid 50 years from now.

15 So that within our discipline, we have to
16 look at the potential that we need to change the way
17 we're going to look at this. We have this problem of
18 stewardship and legacy wastes out there. These
19 things, we would like to get it out of DOE's hands
20 into someone else's hands. But the reality of the
21 matter is it's going to be there, somebody is going to
22 have to maintain it, somebody is going to have to look
23 at it.

24 And what we have to do is decide we need
25 to look at this. We need to be able to look at it and

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1 see how it is changing with time. If it breaks, we
2 need to fix it. That's maintenance. But there's
3 going to be the evolution of understanding. There's
4 going to be evolution of models. Our science is going
5 to get a hell of a lot better than it is now. And we
6 have to have a mindset that we're going to go back and
7 we're going to adapt to the new changing, evolving
8 technologies. So I think it is a fundamental mindset
9 that's going to have to change.

10 CHAIR RYAN: I guess that I would offer
11 that one very important part to this is the John
12 Garrick so what question. There's lots and lots of
13 work that gets done and that's specifically in the
14 waste area, but it really doesn't get at this idea
15 we're managing risk, we're managing some endpoint of
16 impact hypothesized down the line.

17 There's lots of "ology" work that gets
18 down to get to that endpoint, but it's got to be
19 focused on what it is contributing to our
20 understanding of whatever the risk endpoints are that
21 we want to measure. We can't lose sight of that
22 strike zone, because without that we're just --

23 DR. SCHEETZ: But that's all part of it.
24 If you understand what's going on, you can calculate
25 the risk from it.

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1 CHAIR RYAN: But I just want to keep
2 driving that point because I think often we lose track
3 of the fact that we're doing this for that endpoint,
4 not for the, not alone for the intrinsic value of
5 whatever the research project might be.

6 MEMBER CLARKE: I would just like to add
7 to that. I've used the term monitoring a lot. I've
8 used it and I've heard what comes back from that I've
9 been reminded that it's one of those words that you
10 can throw out and everybody might have a different
11 understanding of what you mean by that.

12 I'm talking about monitoring not
13 necessarily groundwater, although clearly we want to
14 do that and that at least tells us that something has
15 gone wrong. But it tells us something has gone wrong
16 too late.

17 So we want to look at monitoring that can
18 help us with our monitoring so we can cycle through
19 that from time to time whatever is appropriate. But
20 the monitoring and the re-upping the performance
21 assessment need to be based on consequences I think
22 and I think risk. Is this something that's going to
23 be a serious problem if it fails and we have exposure,
24 is this something where failure may not be as serious.
25 In other words, I would suggest that the monitoring

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1 need to be risk-informed.

2 DR. SCHEETZ: But the monitoring also it's
3 not that it has failed. The monitoring can alert to
4 an impending failure, if you know the system and the
5 --

6 VICE-CHAIRMAN CROFF: Right, the
7 precursor. That's the precursor. Because what should
8 we be tracking that tells us that things aren't going
9 exactly as planned and we don't really, I don't know
10 what those are yet. We know water is a big component.
11 I mean in this kind of a waste world there are
12 probably other things that we should be looking at as
13 well. But in a land fill, we know infiltrating water
14 is something we don't necessarily want. So what are
15 precursors?

16 The other thing that hasn't been
17 mentioned, I'll just throw it out, is that the
18 institutional controls that are needed need to be
19 monitored as well and they need to be evaluated for
20 their performance. But all of this is as Mike has
21 articulated very well, needs to be risked-informed.
22 That's so hard.

23 CHAIR RYAN: Dr. Langton, go ahead.

24 DR. LANGTON: We do have a, DOE does
25 support a PA monitoring plan, just to use monitoring

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1 one more time. But for example, Saltstone had a 1991
2 performance assessment and we're getting around to a
3 2006 update. In the interim, we've had special
4 analyses performed, but every year there is a plan for
5 work that will be done to support the performance
6 assessments at the Savannah River site.

7 Now right now it is a year-to-year plan.
8 Our funding comes year to year. Some items have
9 carryover into subsequent years, but probably a longer
10 term performance assessment updating plan would be
11 very useful. Performance assessment roadmap to
12 updating performance assessments over a longer time
13 period to allow us to quality work, not on the yearly
14 annual budget schedule. But we're all faced with
15 those problems so I don't know how much hope there is
16 for that.

17 DR. SCHEETZ: It's a crinkly green
18 lubricant.

19 DR. LANGTON: Over more than a year.

20 VICE-CHAIRMAN CROFF: Anymore from NRC
21 staff?

22 MR. LESLIE: Yes, this is Bret Leslie from
23 the NRC staff. I appreciate Mike's focus to going
24 back to risk informed. But I also want to remind
25 folks of the regulatory constraints in which the

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1 process is working.

2 You know, we're in terms of Savannah River
3 and Idaho, we're governed by the Nuclear Defense
4 Authorization Act which specifies certain things. We
5 have been conducting and using independent performance
6 assessments in conducting our reviews of the waste
7 determinations.

8 In fact, if you look at the standard
9 review plan, there's a portion on monitoring. And
10 what goes into those factors that we think are
11 important is based upon our risk-informed review using
12 our analyses. And so part of this generic, there is
13 site specificity associated with it. The cementitious
14 barriers are not treated monolithically across the DOE
15 sites.

16 Savannah River takes a very different
17 approach than Idaho, okay? Consistent with the NRC
18 approach where the applicant decides what its safety
19 case is, we, the staff, are forced to review how much
20 credit they will take for concrete. We can't give
21 them more credit than they're willing to take.

22 So part of this is that for a particular
23 site, Idaho, where they might not be taking any credit
24 at all, a hundred years performance for concrete.

25 We might not have any monitoring because

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1 they have decided that they're not going to take any
2 credit. Whereas if you go to Savannah River, they
3 might be taking a lot of credit. So to understand
4 what needs to be focused on, what the focus of the
5 monitoring should be or the support for long-term
6 performance, you have to look at what is the outcome
7 of those factors from our review.

8 So for instance, we've looked at based
9 upon our reviews to date, we think the long-term
10 chemical and physical stability of concrete is one of
11 the things that has very little support for. The
12 effect pH diffusivity and hydraulic conductivity of
13 these monoliths over long periods of time is something
14 that needs to be evaluated.

15 So I wanted to bring in that perspective
16 that it is risk informed but it also is a factor of
17 two regulatory aspects -- the law and the NRC's
18 perception and policy. The applicant decides what
19 kind of credit they want to take. We can research and
20 can inform and suggest and identify that they might
21 not be taking as much credit as they could, but
22 ultimately it's DOE and its particular sites that
23 determine how they want to make their safety case.

24 CHAIR RYAN: And I think Bret, just so I'm
25 clear, you identified two things that have fallen out

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1 of that as being important -- diffusivity and the
2 block behavior and some of those things. But I think
3 that observation where you are right now is dependent
4 on those choices that the applicant made. So if the
5 applicant went back and made different choices, for
6 example, or another applicant made different choices,
7 you may end up in a different place.

8 The point is is that it's a case. I think
9 it's a case where there's in a way a partnership in my
10 view for what's in the application and what you have
11 to do in the regulatory constraint that you mentioned.
12 That's a good point, because it is a world we got to
13 live in.

14 But the science case you develop in your
15 assessment is strongly dependent on what the applicant
16 gives you. So if they go through the process of
17 saying we're going to take credit for concrete in a
18 different way, in an extended period of time, for
19 these reasons with this information to justify that,
20 and you satisfied, you can end up with a whole bunch
21 of different things that are important versus where
22 you are at the moment with the current cases.

23 I think in a way that's not exactly
24 completely risk informed, but it's risk informed based
25 on what choices you're presented with. I think that's

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1 maybe a view that what we're getting at and that's
2 part of my comment of I think it is risky to just
3 assume the bounding cases. But we're only going take
4 credit for concrete for the 100 years, for the 50
5 years, which is what construction people do.

6 Well, you've shut off a whole world of
7 things you might better understand that could give you
8 margin or confidence. I just wonder, you know, how to
9 get at that. So it is a very flexible exercise of
10 trying to nail pieces of Jello to the wall, but I
11 think it's been a real informative discussion and
12 thanks for that Bret. That was a good comment.

13 VICE-CHAIRMAN CROFF: I think at this
14 point, does the Center have any questions? I think
15 they've been on all day.

16 MR. HOWARD: Yes, we have and that's very
17 much appreciated. But no questions other than what
18 has been asked.

19 VICE-CHAIRMAN CROFF: Okay, thanks. I
20 think with that we're at the end of it. I'd like to
21 thank the speakers for much. You've given us a wealth
22 of information to consider and we will consider it
23 going forward.

24 As you know, our products are letters and
25 I feel fairly certain we're going to see a letter out

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1 of this and we'll talk just a little bit about that
2 right now. But, you know, it's been like drinking
3 from a fire hose is all I can tell you. There's a lot
4 of information to digest. I'm sure Latif is going to
5 have great entertainment while going through the
6 transcript. I think most of you know, this is
7 recorded and there will be a full transcript like a
8 legal transcript and it will have all the view graphs
9 in it some place at the back of it. So that will be
10 out in a month or two, I think, in complete form, on
11 ADDAMS, the NRC Information Management System.

12 And my thanks to Latif for helping to
13 organize this and I think with that again, my sincere
14 thanks and the working group meeting I think will
15 close and I guess talk just momentarily about a
16 letter?

17 CHAIR RYAN: Sure and I want to add my
18 thanks on behalf of the entire Committee for your
19 generous time and talent that you presented to us
20 today and also to Latif and the other staff and to
21 Allen for putting together this fabulous working group
22 that's covered an awful lot of ground in a short
23 period of time. And again, I sincerely thank you on
24 behalf of the Committee. So we'll leave this in a
25 letter discussion?

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1 VICE-CHAIRMAN CROFF: Yes, and it's going
2 to be fairly short. It's late in the day. Is there
3 any disagreement that we need a letter?

4 CHAIR RYAN: None.

5 (Laughter.)

6 VICE-CHAIRMAN CROFF: At this point --
7 what I'd like is something like middle of next week.
8 If you'd email observations and any recommendations
9 that you think are important or any other thoughts.

10 CHAIR RYAN: I think for the benefit of
11 our panel members, I think in the last half hour or so
12 or maybe in the last couple of hours of summary
13 comments and points by you all and by us will be
14 organized into the body of our letter. I don't think
15 there is any real need to rehash all those over again,
16 but we will be mining the transcript with a little bit
17 more detail to get the good words down.

18 VICE-CHAIRMAN CROFF: Definitely. We'll
19 be mining the transcript, and just the big points or
20 distillation or integration of things or this kind of
21 thing is what I want from you.

22 And I'll try to prepare a letter and we'll
23 try to bring it into the August meeting which is where
24 we're going.

25 CHAIR RYAN: Our subcommittee meeting.

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1 All right, with that no other business before the
2 Committee. We'll adjourn the record and adjourn the
3 meeting. Thank you all very much.

4 (Whereupon, at 5:10 p.m., the meeting was
5 concluded.)
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This is to certify that the attached proceedings
before the United States Nuclear Regulatory Commission
in the matter of:

Name of Proceeding: Advisory Committee on

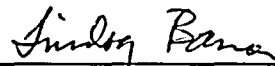
Nuclear Waste

172ND Meeting

Docket Number: n/a

Location: Rockville, MD

were held as herein appears, and that this is the
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States Nuclear Regulatory Commission taken by me and,
thereafter reduced to typewriting by me or under the
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Lindsey Barnes
Official Reporter
Neal R. Gross & Co., Inc.

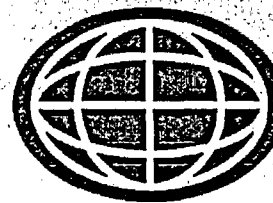
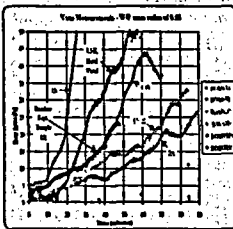
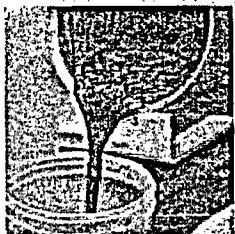
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Designing Cement Waste Form



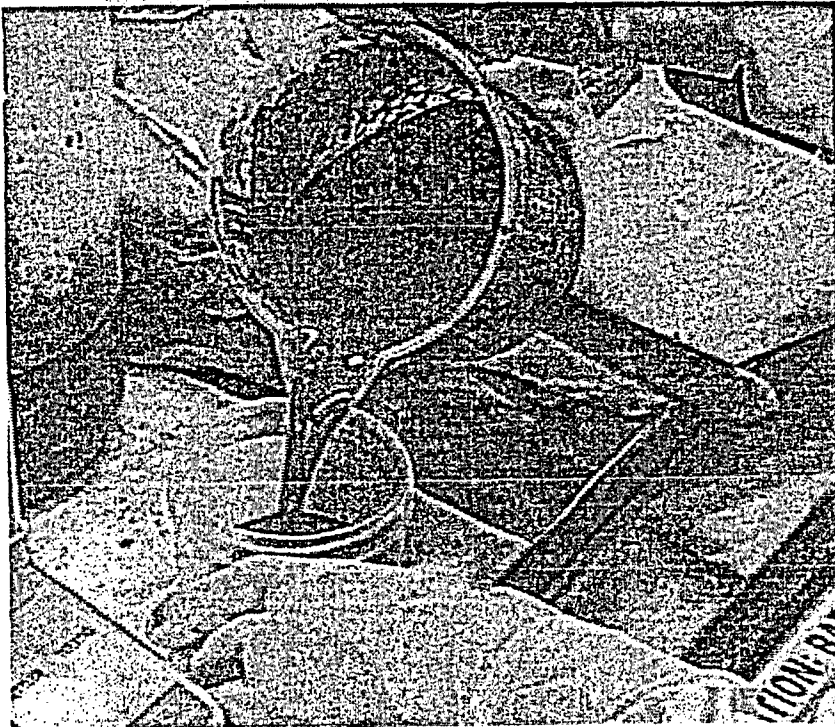
SRNLTM
SAVANNAH RIVER NATIONAL LABORATORY

We Put Science To Work

Christine A. Langton, Ph.D.
Savannah River National Laboratory
July 18, 2006

Presented to:
Advisory Committee on Nuclear Waste
Nuclear Regulatory Commission

OUTLINE



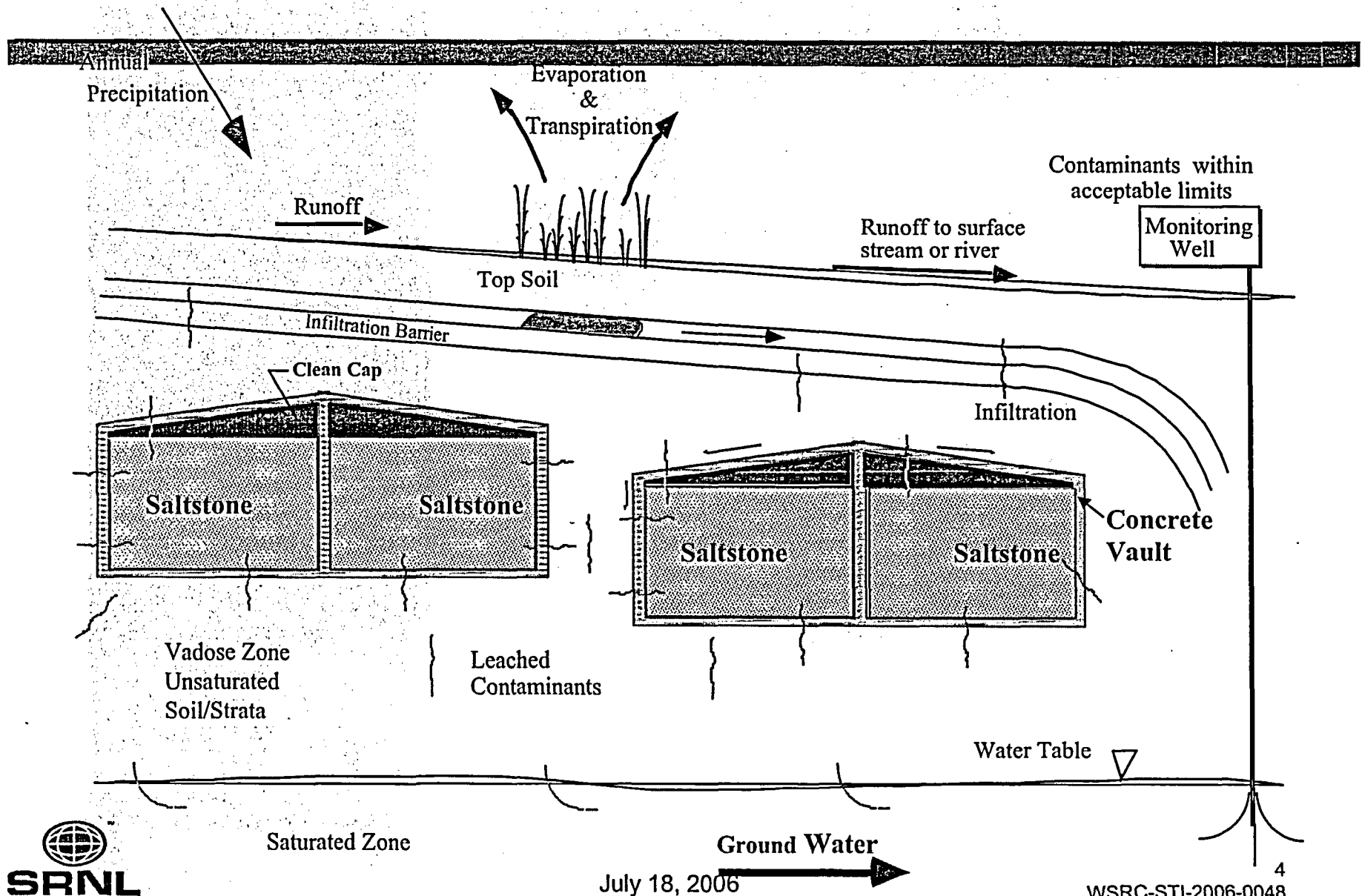
**SRS Tank 50 Batch 0 Saltstone
Formulation Confirmation**

- **Applications**
 - Waste Treatment
 - Containment
 - Environmental Remediation
 - Facility Closure
- **Functional Requirements**
- **Waste Form Design**
 - Materials Requirements
 - Performance
 - Processing
 - Laboratory Qualification
 - Field Testing
- **Landfill Evaluation**
 - Modeling (Performance Predictions)
- **Uncertainties/Unknowns**
- **Risk Reduction**

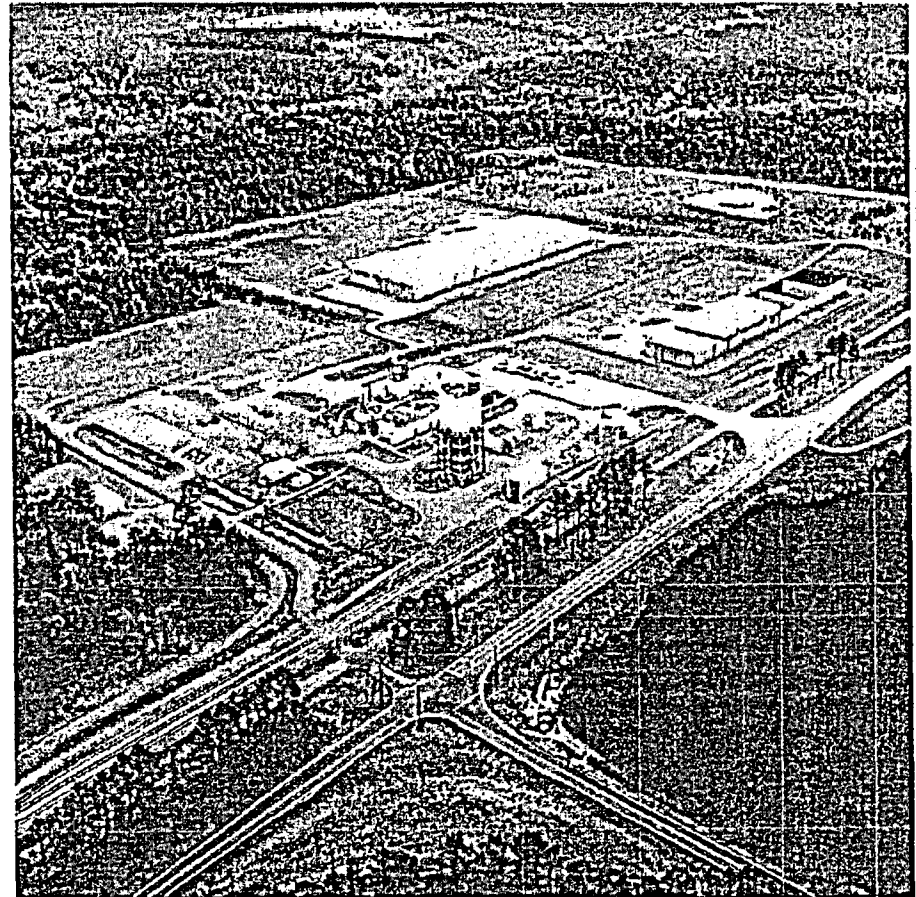
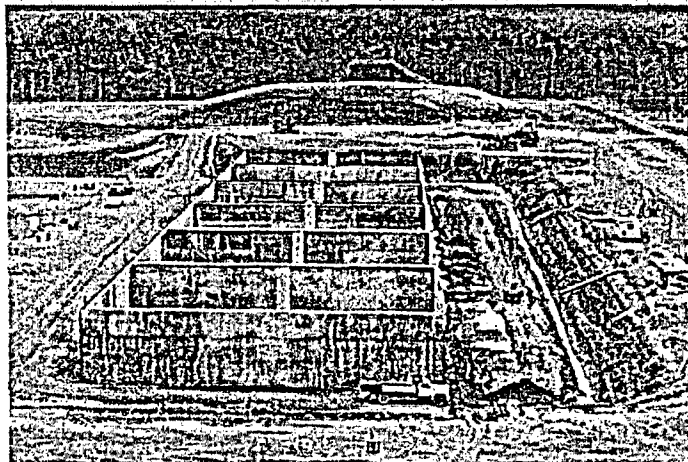
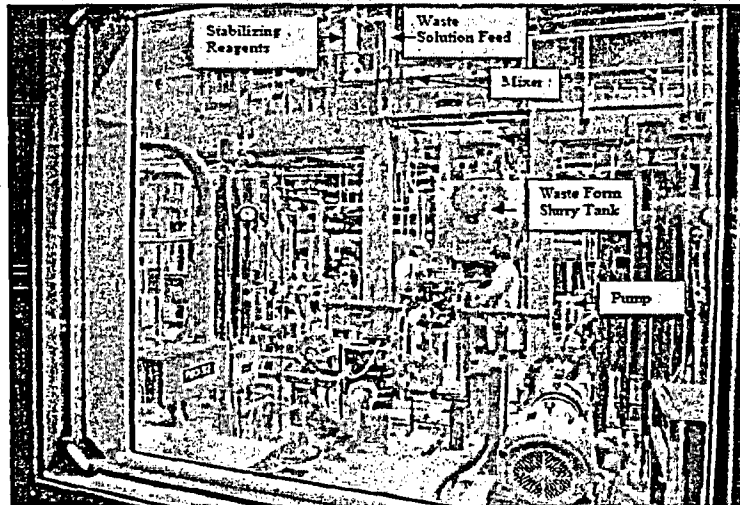
APPLICATIONS

- **Waste Treatment**
- **Cement-Based Containment**
 - Concrete Vaults (SRS LAW Vault, ILT, ILNT)
 - Concrete Culverts, Casks, Drums
 - Concrete/Grout Backfills (SRS E-Area Components in Grout)
 - Trench Liners and Covers
- **Environmental Remediation**
 - Basin Sludge/Soil Stabilization (SRS Reactor Basin Closures)
 - Grout Curtains (SRS Separation Area Curtain wall)
- **Facility Closure**
 - Tank Closure (SRS Spent Solvent Tanks and HLW Tanks F-17 and F-20)
 - Facility Decommissioning/Entombment (SRS PuFF)

Saltstone Conceptual Model

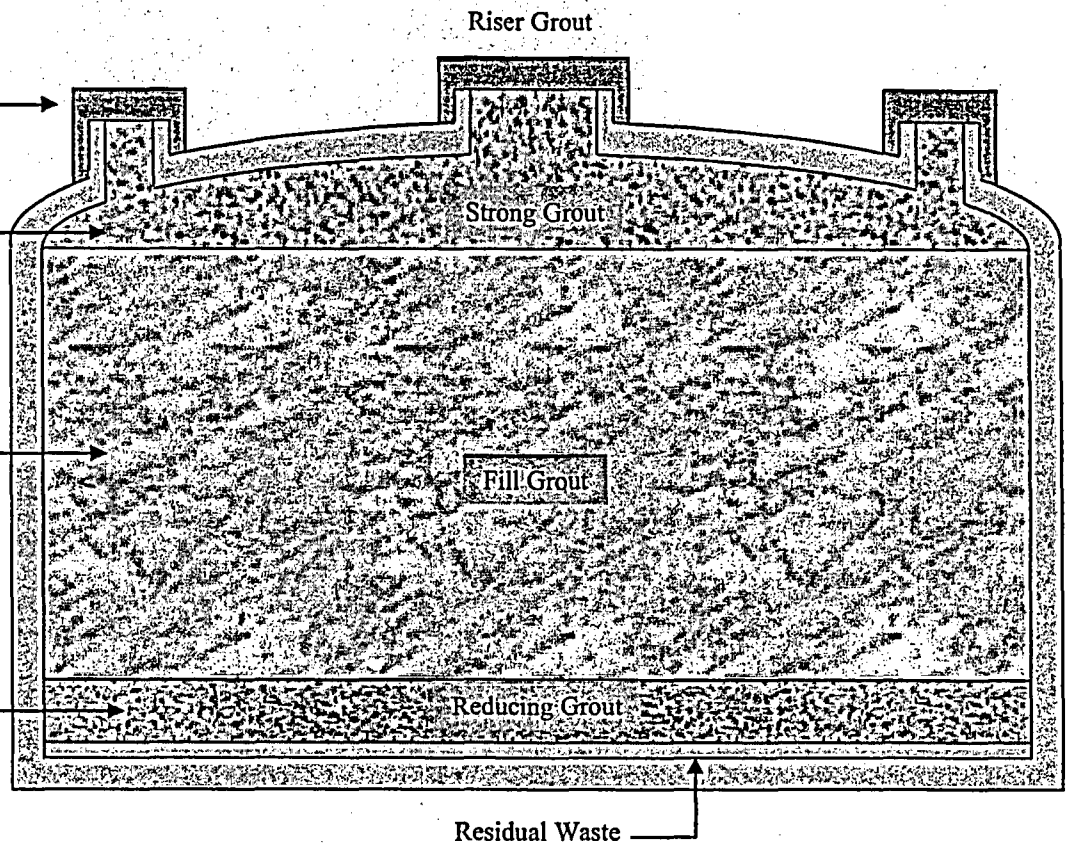


SRS SALTSTONE FACILITY

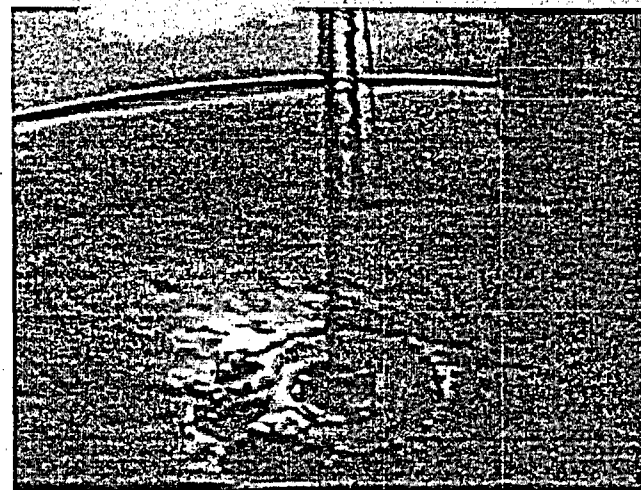
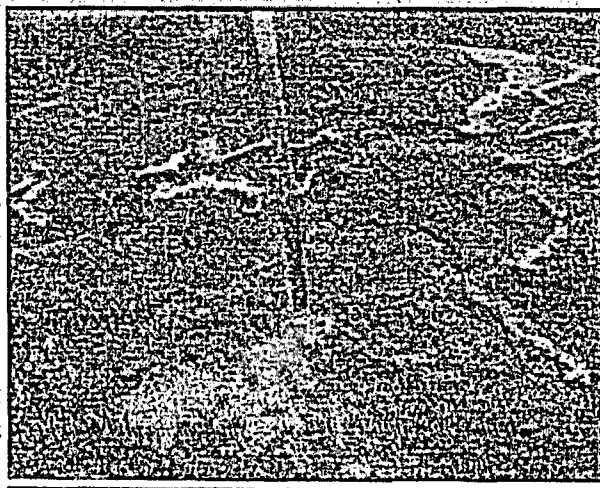
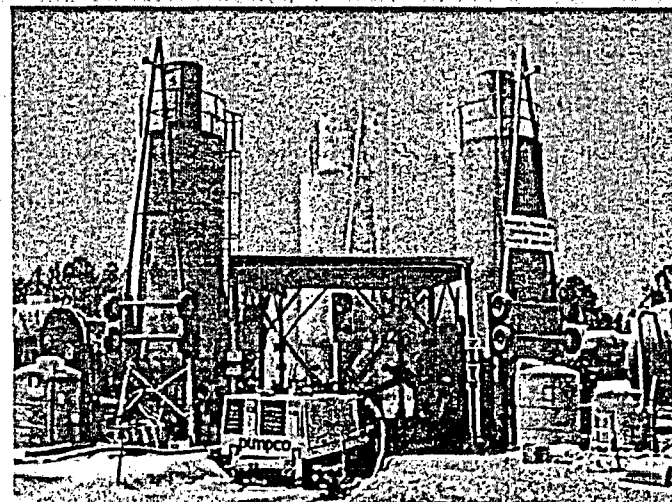
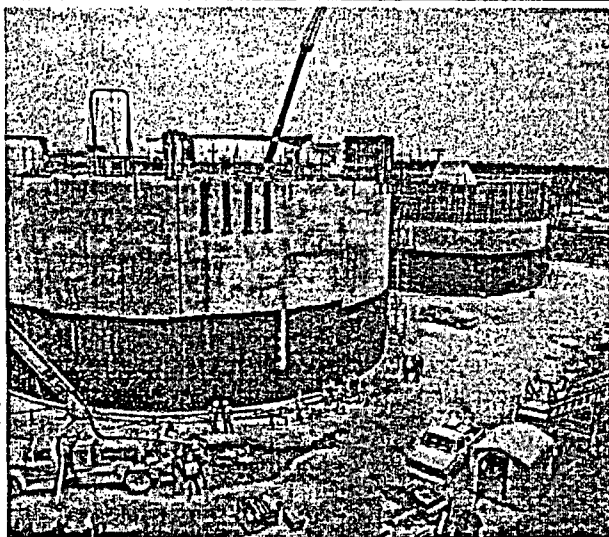


CLOSED HLW TANK

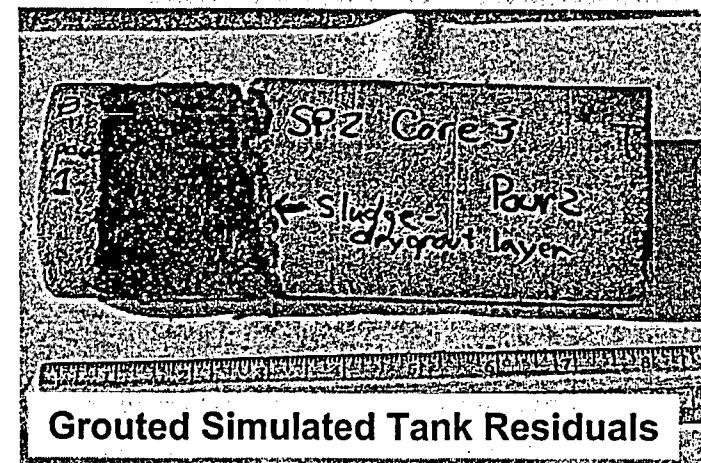
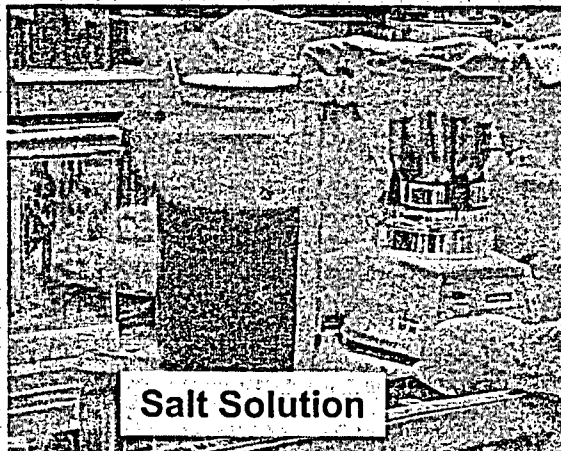
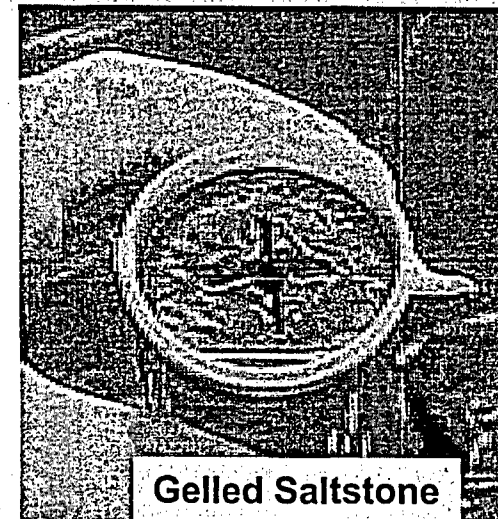
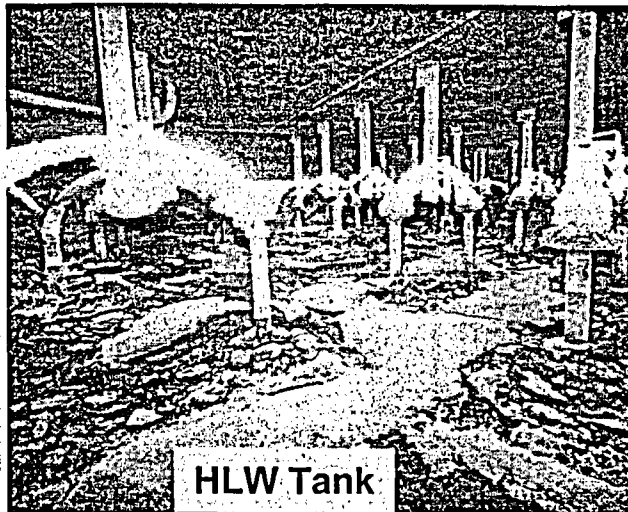
Requirement	Tanks 17 & 20	Tank 19 w/All Reducing Grout option
Riser grout >5,000 psi @ 28 days	Riser grout >5,000 psi @ 28 days	Riser grout >5,000 psi @ 28 days
Strong grout >2,000 psi @ 28 days	Strong grout >2,000 psi @ 28 days	Reducing Strong grout >2,000 psi @ 28 days
CSLM >50 psi @ 28 days	CSLM >1,800 psi @ 90 days	Reducing CLSM 1,800 psi @ 90 days
Reducing grout >500 psi @ 28 days	Reducing grout 10,000 psi @ 90 days	Reducing grout 1,800 psi @ 90 days



SRS TANK CLOSURE



Examples of Waste and Waste Forms



FUNCTIONAL REQUIREMENTS

- **Waste Form Stabilization/Fixation**
 - Reduce mobility of soluble contaminants
 - Change chemical form; precipitate low solubility phases
 - Reduce exposure/contact to dispersing media (leachate)
 - Reduce mobility of dispersible particulate contaminants
 - Micro/macro encapsulation
 - Increase particle size
 - Create recognizable material distinct from environmental media
 - Reduce exposure/dispersion by human intrusion
- **Waste Form Solidification**
 - Eliminate free liquids (<0.5 vol. %)
- **Landfill Stabilization**
 - Provide engineered barriers
 - Support overburden (minimize landfill cover collapse)
 - Reduce water infiltration and human intrusion
- **Disposal System (waste form, containment and landfill)**
 - Operation and Construction
 - Perform for required duration

WASTE TREATMENT DESIGN PROCESS

- **Characterize Waste**
- **Classify Waste**
 - Identify regulatory treatment and disposal requirements
- **Select Disposal Unit**
 - WAC (concentration limits, packaging and handling)
- **Down Select Waste Form (tests, alternatives study)**
 - Material and Process
- **Test Waste Form Samples (optimize loading and properties)**
 - Leachability
 - Long-term Aging (durability)
- **Design Treatment/Disposal System**
 - Evaluate Constructability (cost, location, codes and standards)
 - Evaluate Operations (regulatory, personnel, attainment, logistics)
- **Model Landfill Performance (PA)**
 - Evaluate landfill features + waste form interactions
- **Address Risks and Risk Reduction Approaches**
 - Determine additional waste form and/or landfill specifications
 - Modify design if needed and re-evaluate

WASTE FORM DESIGN AND TESTING

- Characterize Waste: EPA and nuclear methods
- Select Mix Ingredients and Proportions
- Prepare and Cure Samples: Modified ASTM methods
- Test Samples (material properties)
 - Laboratory Measurements
 - Processing Properties (rheology)
 - Leachability/Extraction
 - Crushed: EPA TCLP
 - Monolithic: ANS/ANSI 16.1 (D_{eff})
 - Sorption: ASTM column and batch: K_d
 - Mechanical Properties: ASTM UCS, density, porosity
 - Hydraulic Properties: permeability, moisture characteristics
 - Thermal Properties: calorimetry
 - Degradation Susceptibility: ASTM concrete methods, ACI Standard Practices
 - Long-term Aging
 - Accelerated aging methods and evaluation of effects
 - Property/geometry change rates: Independent and coupled

LANDFILL EVALUATION

- **Material Property Characterization/Testing: ASTM Methods**
 - Containment
 - Environmental
- **Constructability and Operability: Engineering Practices**
 - Suitability
 - Material Availability and Compatibility
 - Codes and Standards
 - ALARA
 - Cost
- **Performance**
 - Lysimeters
 - Disposal Unit Sampling and Monitoring
 - Waste Form
 - Containment (engineered barriers)
 - Environmental
 - Predictive Modeling: Flow and Contaminant Transport: Commercial Computer Programs
 - Deterministic – Base Case and Sensitivity Cases
 - Probabilistic – Uncertainty Analysis

WASTE FORM UNCERTAINTIES

- **Waste Analyses**
 - Limited Samples, Sample Variability
 - Detection Difficulties, (Low Detections Limits, Low Concentrations, Interference)
- **Leaching and Sorption**
 - Contaminant Distribution
 - Leachate Chemistry
 - Laboratory vs. Field Conditions (equilibrium, evaporation, flux extremes, coupled effects)
- **Hydraulic Properties**
 - Low Permeability – difficult measurements
 - Scaling Effects: sample vs. structure
 - Laboratory vs. Field Aging
 - Coupled Effects: Hydraulic Properties, Leachate Chemistry, Aging
- **Thermal Properties**
 - Impact of thermal transients during curing on other properties (mineralogy, cracking)
- **Durability (Chemical and Physical)**
 - Rates and Timing of Events
 - Coupled Effects (manifestation and impact on rates of other changes)
 - Chemical and mineralogical changes impact mechanical, hydraulic, leaching properties
 - Alkaline, Chemically Reducing
 - Crack location/patterns, dimensions, interconnectivity, infilling

LANDFILL UNCERTAINTIES

▪ Long Performance Time Requirements

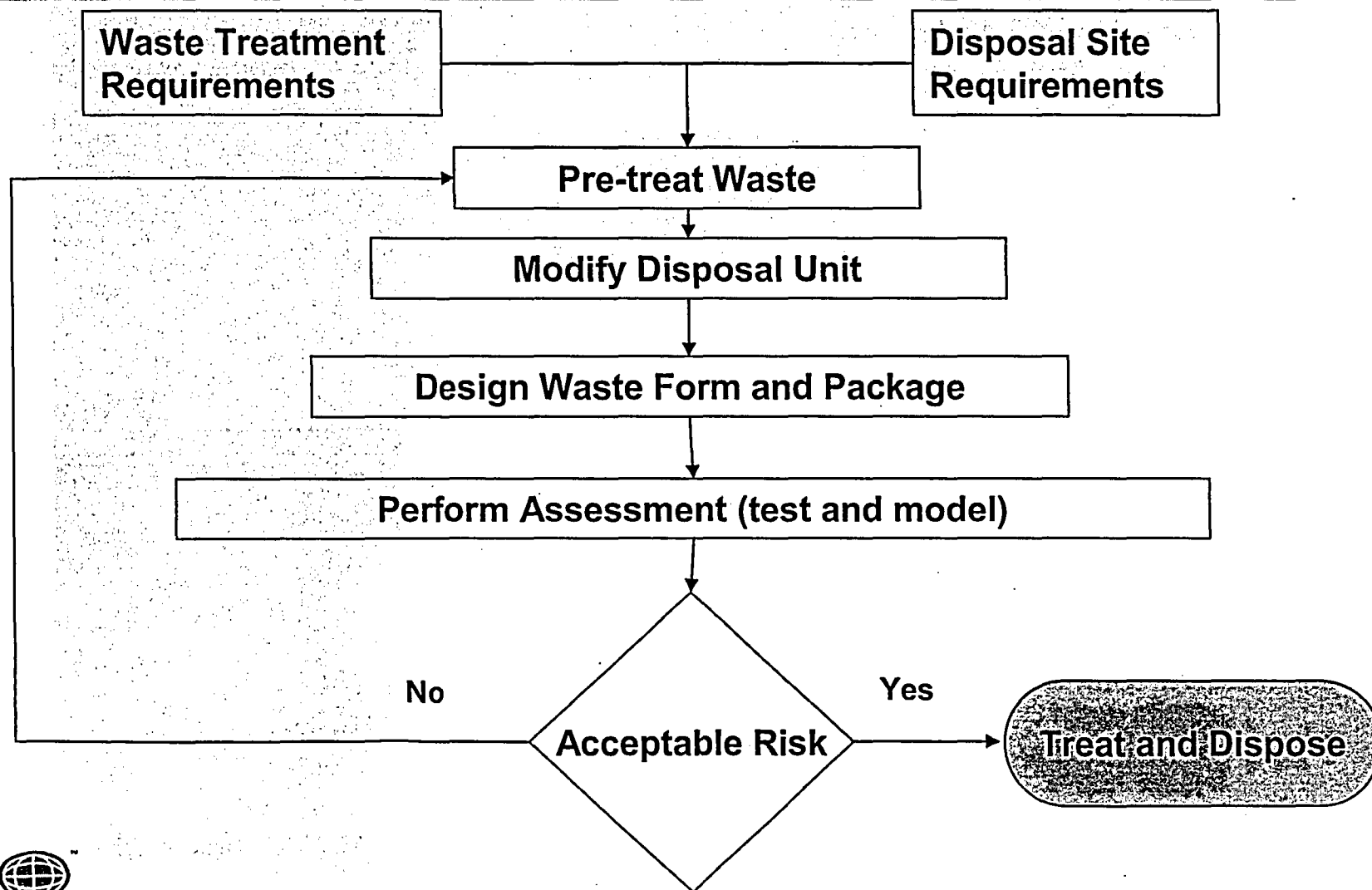
Up to 10,000 years or time of maximum contaminant concentrations in the compliance well

- Extrapolation of Field Conditions
- Seismic Events
- Climate Changes
- Land Use
- Intruder Assumptions
- Toxicology
- Programmatic Changes
- Regulatory and Public Perception Changes

RISK REDUCTION APPROACH

- Status current state of knowledge and programs
- Review regulations and orders; interpret changes and effects on programs
- Review and improve strategy for future disposals
- Perform sensitivity analyses for current and proposed disposal unit configurations to help identify data needs
 - Review and improve conceptual model (e.g., descriptions of initial conditions and material properties)
- Develop
 - Innovative approaches for aging and testing waste forms
 - Framework for interpreting test results
- Obtain peer review approvals for future work
- Establish priorities for future work

SUMMARY



SUMMARY



Conceptual Models and Approaches to Understanding Long Term Performance of Cementitious Waste Forms

Presented by

**David S. Kosson, Ph.D., Professor
Vanderbilt University**

**Consortium for Risk Evaluation with Stakeholder Participation
(CRESP)**

To

**Advisory Committee on Nuclear Waste
Nuclear Regulatory Commission**

July 18, 2006

Collaborations

Vanderbilt University

D. Kosson, A. Garrabrants,
S. Mahadevan, F. Sanchez,
J. Clarke, S. Lopez

Netherlands Energy Research Centre (ECN)

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J.C.L. Meeussen, A. van
Zomeren, P. Seignette

DHI (Denmark)

O. Hjelmar

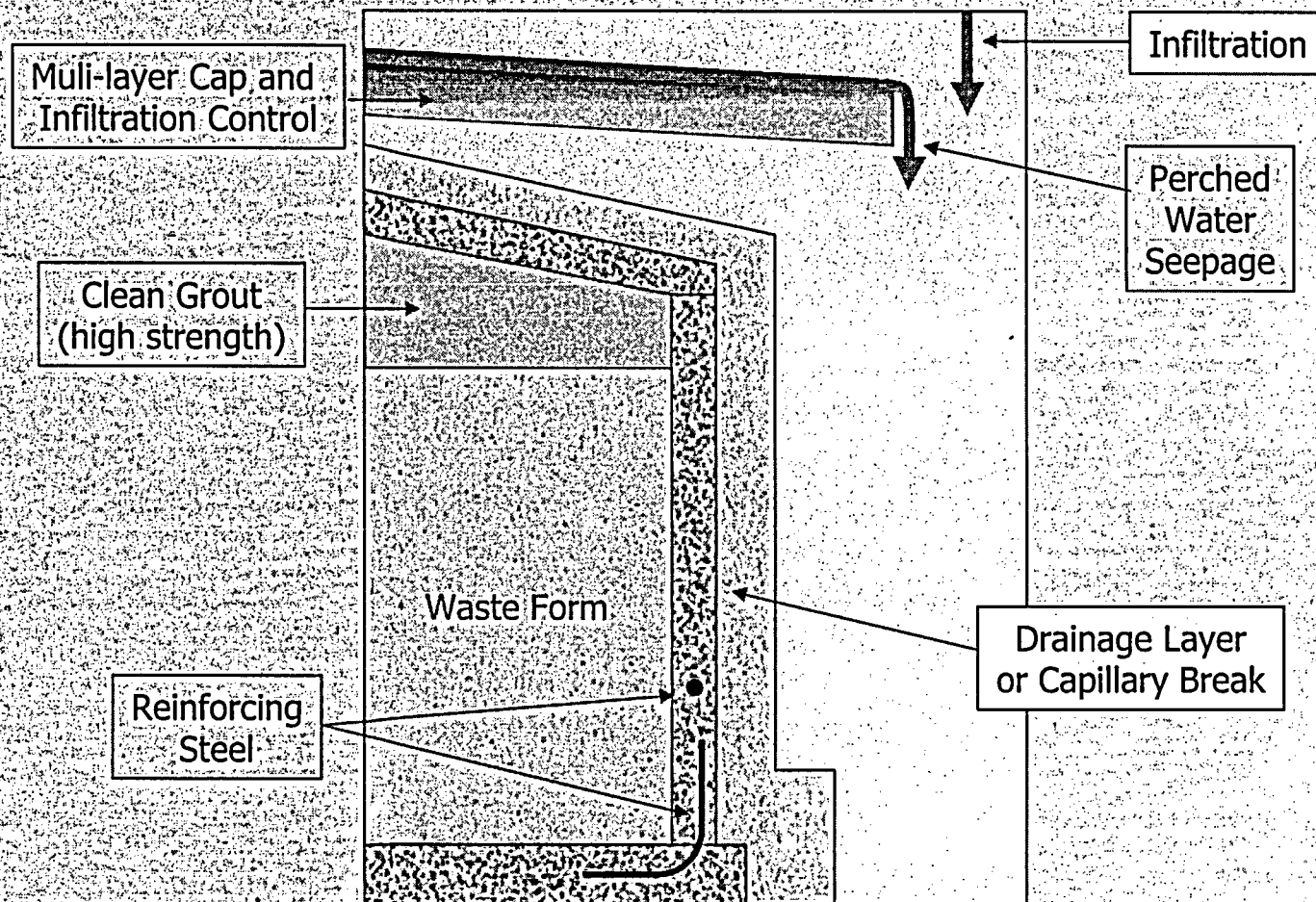
Savannah River National Lab

C. Langton, G. Flach

Pacific Northwest National Lab

J. Serne, T. Brouns

Generic Vault Disposal System



Motivation

Need for better estimates of long-term constituent release for near-surface disposal of cementitious and other non-vitrified waste forms.

Applicability

- Performance Assessments and 3116 Determinations
 - HLW tank closure using grout
 - Disposal of saltstone & similar wastes at SRNL, INL, Hanford (ORP)
 - Primary and secondary waste streams from steam reforming
 - Secondary waste streams from vitrification
- Waste Treatment Acceptance Criteria
- Operational Controls
- Management of future wastes from reprocessing/recycling (GNEP)

Primary Constituents of Concern

- Long lived & Mobile: Tc-99, Np-237, Se-79, I-129, C-14, U
- Mobile: Cs-137, Sr-90, Nitrate, tritium



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Broader Questions

What basis should be used to

Define the appropriate type of waste form, disposal system and location for specific wastes?

Estimate long-term waste form and disposal system performance?

Establish treatment (operational) criteria?

Define monitoring requirements (pre-cursors) that enable pre-emptive action to avoid system failure?



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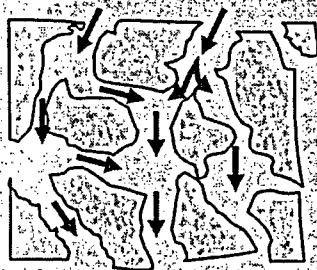
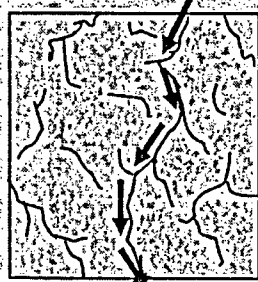
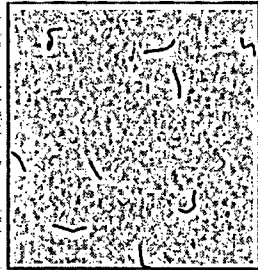
Constituent Release by Leaching

Primary Factors

- System Integrity
 - Engineered and Institutional System
- Waste Form Performance
 - Physical Integrity
 - Water Contact
 - Moisture Status
 - Oxidation Rates and Extent
 - Constituent Chemistry and Mass Transport

Processes and Impacts

Physical Integrity & Water Contact



Monolithic Matrix

- Flow-around
- Low interfacial area
- Diffusive release

Stressed Matrix

- Flow-around/through
- Higher interfacial area
- Diffusion-convection

Spalled Matrix

- High permeability
- Very high interfacial area
- Equilibrium-based release

Conceptual Model

- Micro-cracks develop, increasing solid-liquid surface area
- Bridging of micro-cracks create macro-cracks
- Through-cracks develop over time, leading to convective flow
- Ultimate end state may be permeable matrix – release based on local equilibrium

Impact

- Need to account for the sequence of physical states and rate of changes
- Influences chemical reactions and constituent release
- Both "intact" & "degraded" cases are simplistic and may not be realistic



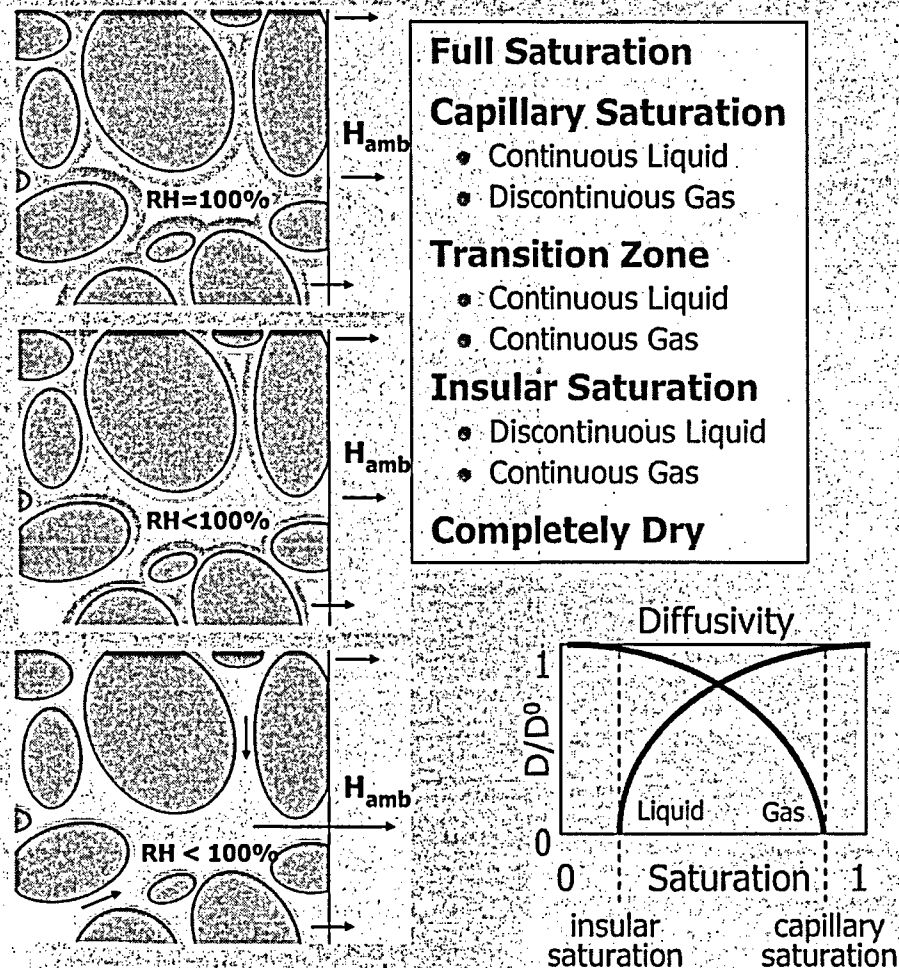
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Processes and Impacts

Moisture Transport



Conceptual Model

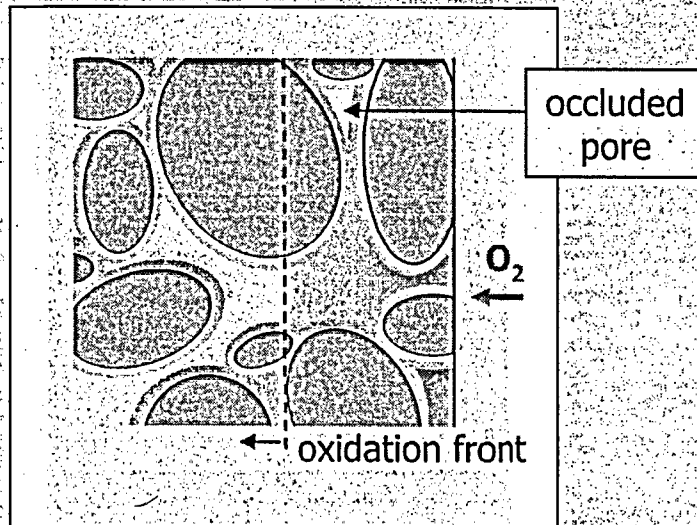
- Waste form consumes water via hydration reactions
- Moisture exchange w/environment
 - Evaporation/condensation
 - Capillary suction
 - Intermittent wetting (precipitation)
- Water content determines
 - Gaseous degradation processes (oxidation, carbonation)
 - Constituent diffusion pathways

Impact

- Diffusivities are not constant over moisture regime
- Fractional saturation
 - Increases the importance of gas phase transport & reactions
 - Decreases rate of liquid phase transport

Processes and Impacts

Oxidation Rates and Extent



Conceptual Model

- Waste form pores – two phase system of gas and liquid; depends on moisture content (saturation)
- O_2 transport via gaseous diffusion may be important depending on saturation.
- Oxidation may lead to change in leaching behavior
 - Increased Tc-99 release; other constituents

Impact

- Gas phase transport not considered
 - Flux of O_2 (gas) $\sim 10^5 >$ liquid phase flux
- Impact to Tc-99 oxidation minimized

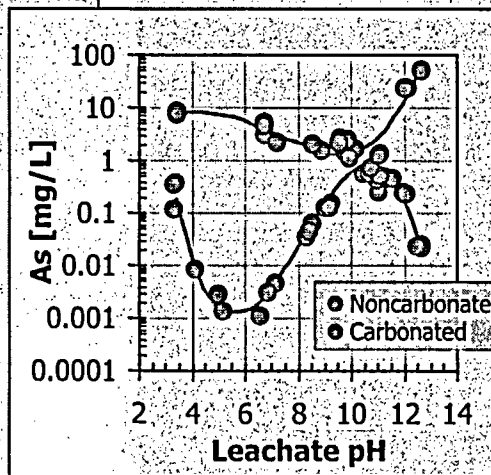
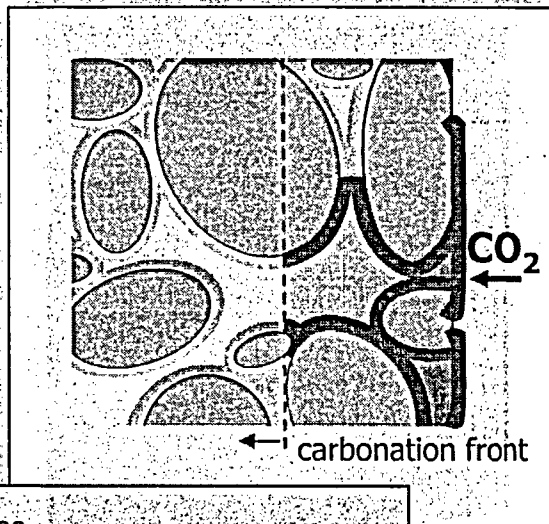
	Air	Water	Ratio (A/W)
D_{O_2} [cm^2/s]	0.21	0.000019	1.1E+04
Conc of O_2 [mole/L]	8.9E-03	2.6E-04	1.4E+01

(1) Wilke and Chang, 1955

(2) www.swbic.org/education/env-engr/gastransfer/gastransf.html

Processes and Impacts

Carbonation



Conceptual Model

- $\text{CO}_3^{-2} + \text{Ca}^{+2} \rightarrow \text{CaCO}_3 (\text{s})$
 - Gas phase diffusion of CO_2
 - Liquid phase diffusion of HCO_3^-
- Pore water pH decreased
 - Alters solubility of constituents (increase or decrease depending on species).
- Carbonation
 - Expansive precipitate – internal stress (cracking)
 - Pore blocking – increases diffusional resistance (decreases oxidation, release rates).
 - Extent and pore effects depend on waste form alkalinity and saturation

Impact

- Potential for speciation changes (e.g., As)
- Pore structure changes
- May have either positive (e.g., pore capping) or detrimental (i.e., increased solubility) impacts



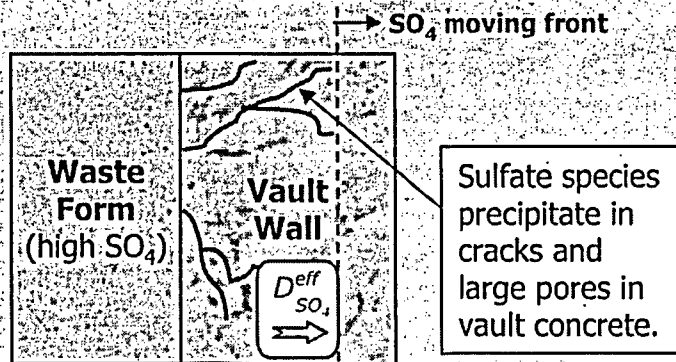
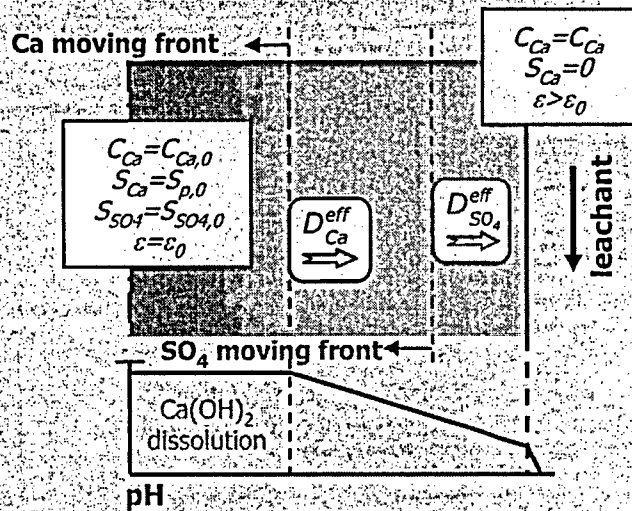
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Processes and Impacts

Leaching of Major Constituents



Conceptual Model

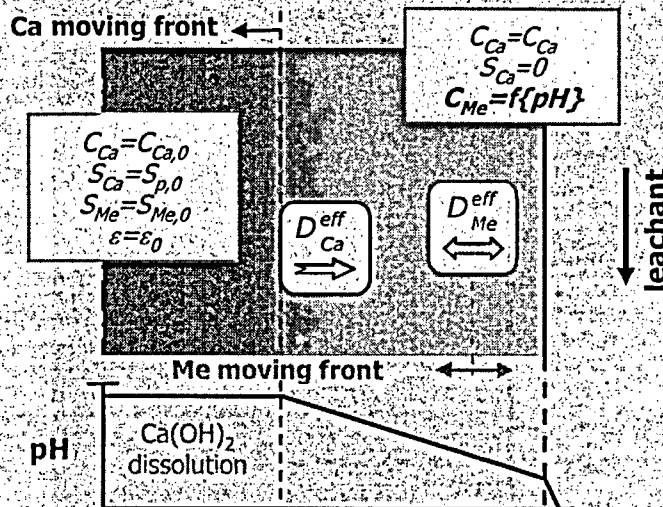
- Transport described by moving dissolution fronts
- Precipitation/reaction processes near external boundaries may significantly impact release (+ or -)
- Dissolution/diffusion of Ca(OH)₂ and CSH control pore water pH
 - pH gradients alter trace species release
- SO₄ leaching from waste into vault attacking concrete physical structure.
- Source of SO₄ may be waste or external environment

Impact

- Mass transport estimates may not reflect the dynamic chemistry and mineralogy of the waste form.
- Release rates and extents mechanistically different from simplified assumptions, effecting predictability.

Processes and Impacts

Leaching of Trace Constituents

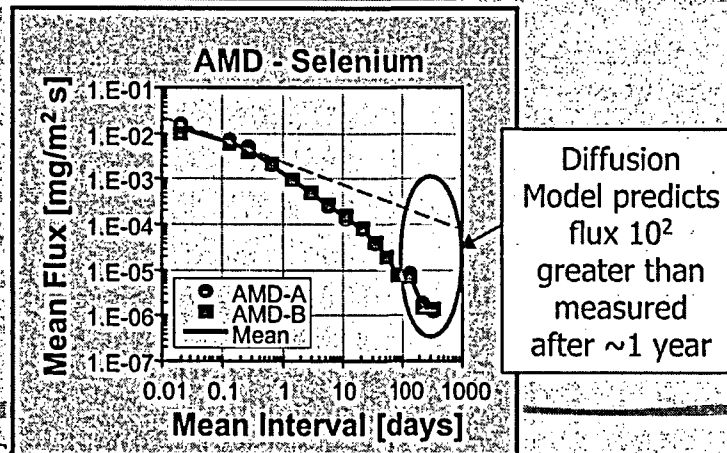


Conceptual Model

- Release based on coupled chemistry and mass transport.
- Release dependent on:
 - Moisture conditions
 - pH gradients
 - Redox chemistry
 - Boundary layer formation

Impact

- Performance assessments may over- or under-predict release



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Integrated Long-Term Degradation

Chemical degradation and physical stress effects are coupled and integrated.

Physical stress

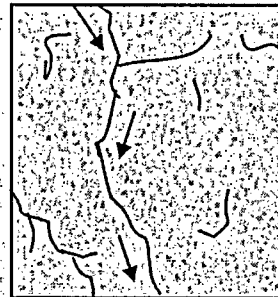
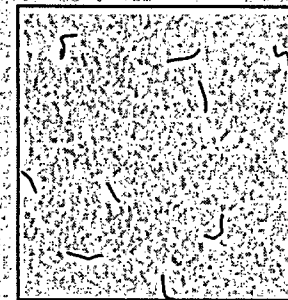
- Cyclic loading
- Flexural bending
- Drying shrinkage
- Seismic events
- Settlement

Chemical degradation

- Oxidation
- Leaching
- Expansive reactions
 - Carbonation
 - Sulfate attack
 - Rebar corrosion

Microcracks

- Increase porosity
- Increase interaction pore water/surface



Through-cracks

- Preferential flow path
- Diffusive and convective release
- Loss of strength

Spalling

- Loss of cohesiveness
 - Two body problem
- Eventual release from "granular" material



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Current Studies on Secondary Waste from ORP

Motivation

- Tc-99, I-129 in secondary wastes from vitrification

Objective

- Leaching assessment of reducing grout for secondary waste treatment.
- Comparison with "ANS16.1-type" testing in synthetic ground water.

Reducing Grout

- Ground Steel Slag 43 wt%
- Class F Fly Ash 42
- OPC 7
- DI Water 7

Synthetic Hanford Groundwater

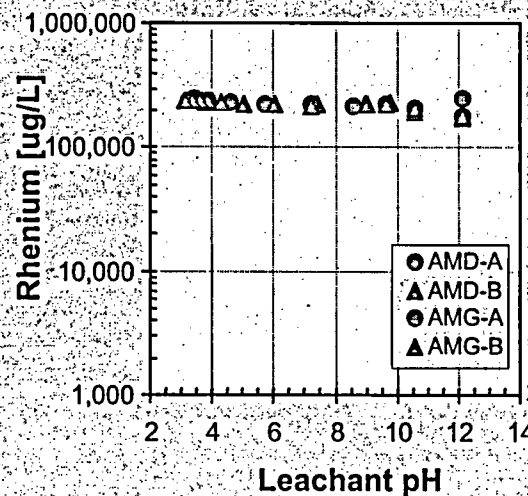
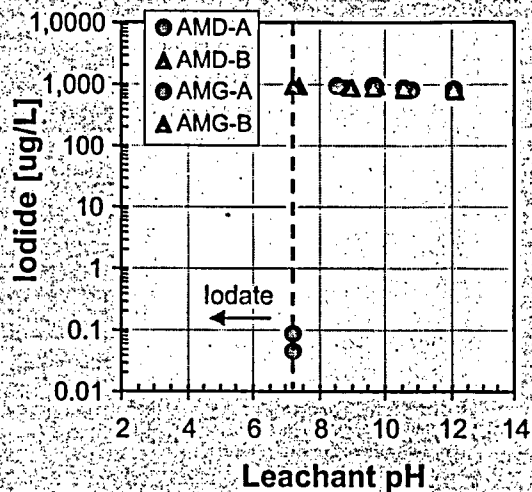
- CaSO_4 1.20 mmol/L
- NaHCO_3 1.04
- $\text{Mg}(\text{HCO}_3)_2$ 0.62
- CaCl_2 0.34
- KHCO_3 0.19
- $\text{Ca}(\text{HCO}_3)_2$ 0.18

Contaminants in Reducing Grout

	mg/kg	Added As
Ag	243	AgNO_3
As(V)	1000	$\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$
Ba	500	$\text{Ba}(\text{NO}_3)_2$
Cd	1000	$\text{Cd}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$
Cu	1000	$\text{Cu}(\text{NO}_3)_2 \cdot 2.5\text{H}_2\text{O}$
Cs	1000	CsCl
I	1214	NaI
Pb	1000	$\text{Pb}(\text{NO}_3)_2$
Re	971	KReO_4
Sb	952	Sb_2O_3
Se	751	KSeO_4
Zn	1000	$\text{Zn}(\text{NO}_3)_2$

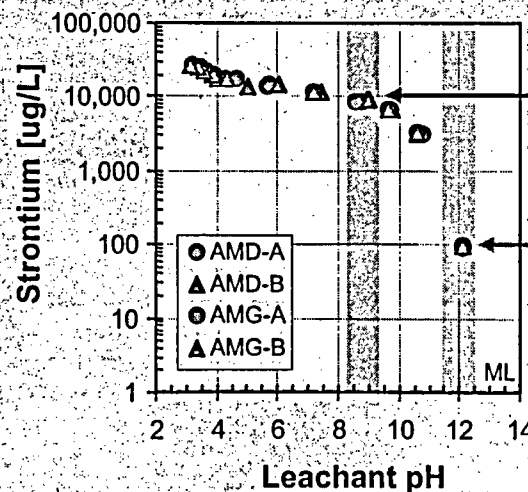
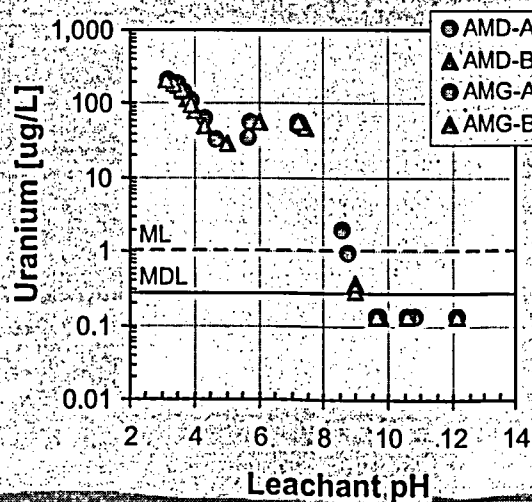


Equilibrium – Trace Species



Solid-Liquid Partitioning Comparison

- AMD – DI Water
- AMG – Synthetic Hanford Ground Water

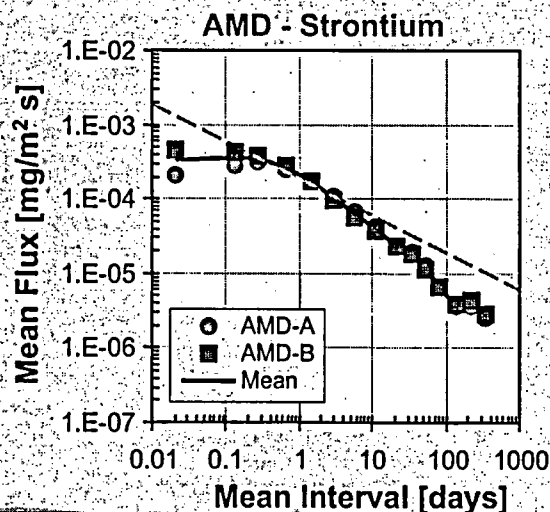
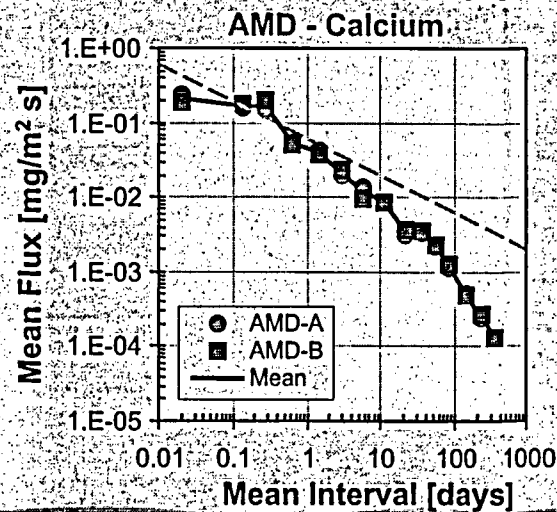
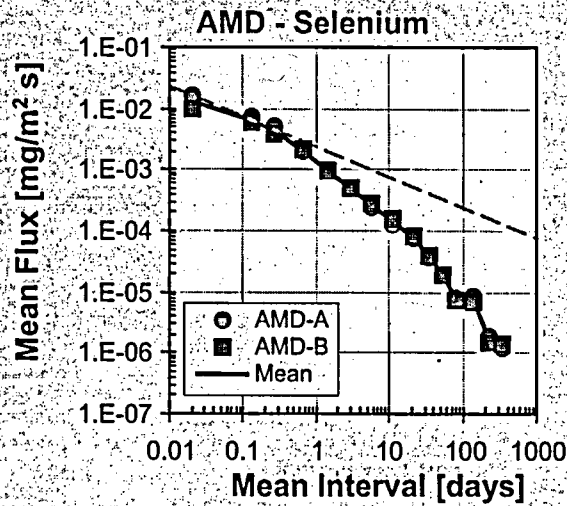
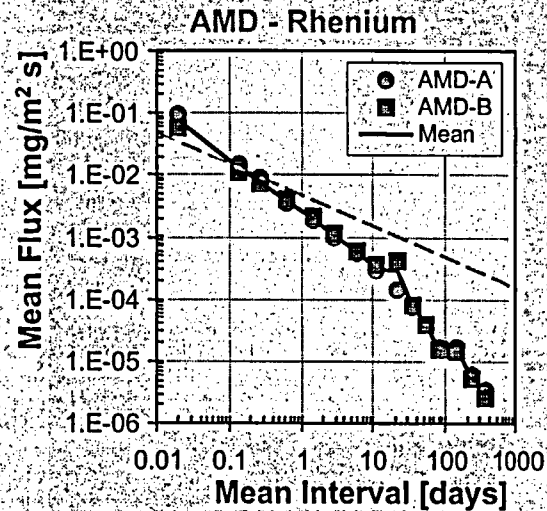


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Mass Transport Tests



MT001 Test

- Tank Leaching in DI Water
- Constituent Flux
- Constituent Release

Comparison

- AMD - DI Water
- Flux @ constant diffusivity (green dash)

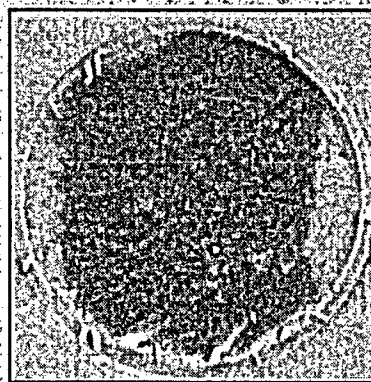
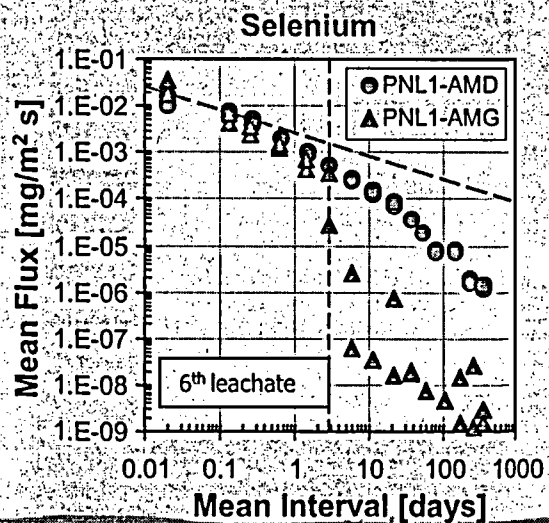
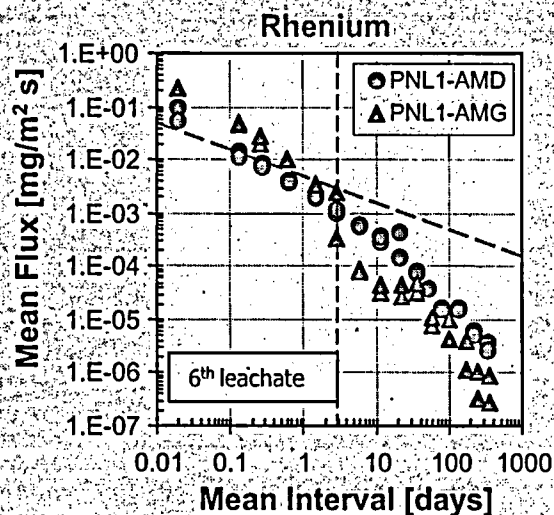
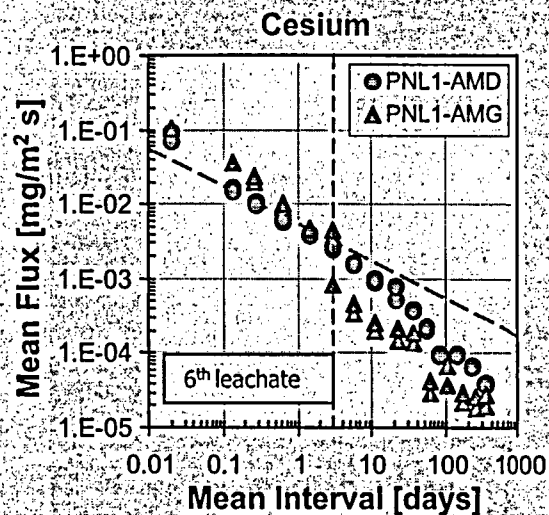


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Synthetic Groundwater



Precipitate on sample AMG-B
after 6th leaching interval

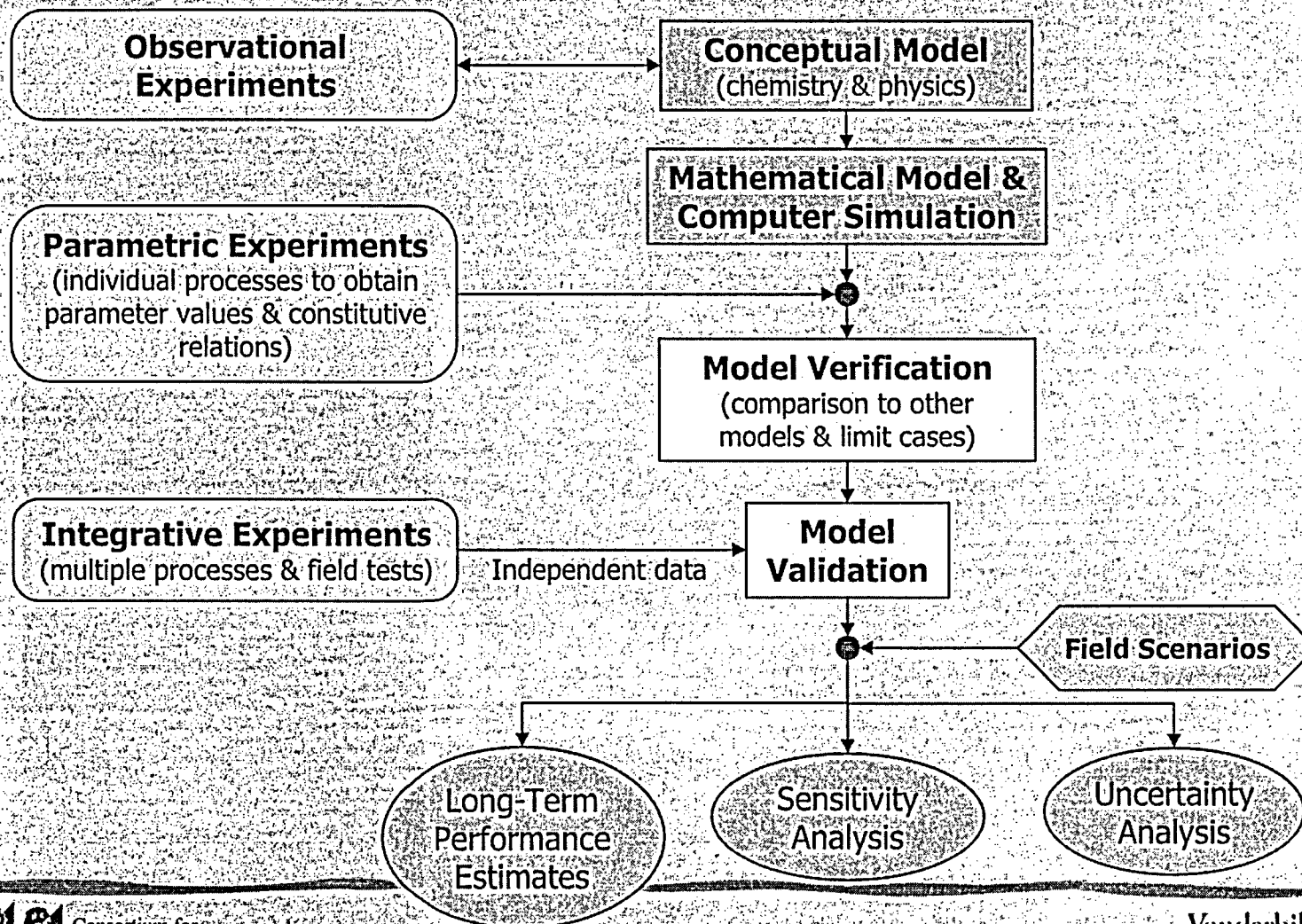
MT001 Test Comparison

- AMD – DI Water
- AMG – Synthetic
Hanford Groundwater

Hanford Groundwater

- CaSO_4 1.20 mmol/L
- NaHCO_3 1.04
- $\text{Mg}(\text{HCO}_3)_2$ 0.62
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Process- and Mechanism-Based Experimentation & Modeling



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Overarching Framework

(Kosson, van der Sloot, Sanchez & Garrabrants, 2002, Environ. Engr. Sci., 19, 159-203)

Measure intrinsic leaching characteristics of material (aqueous-solid partitioning (pH and LS); release kinetics)

- Batch extractions & tank leaching (monoliths)
- Constituent fraction readily leached
- Controlling mechanism for release (mineral dissolution and solubility, solid phase adsorption, aqueous phase complexation)
- Release kinetics for mass transfer parameters

Evaluate release in the context of field scenario

- External influencing factors such as carbonation, oxidation
- Hydrology
- Mineralogical changes

Use geochemical speciation and mass transfer models to estimate release for alternative scenarios

- Model complexity to match information needs
- Many scenarios can be evaluated from single data set

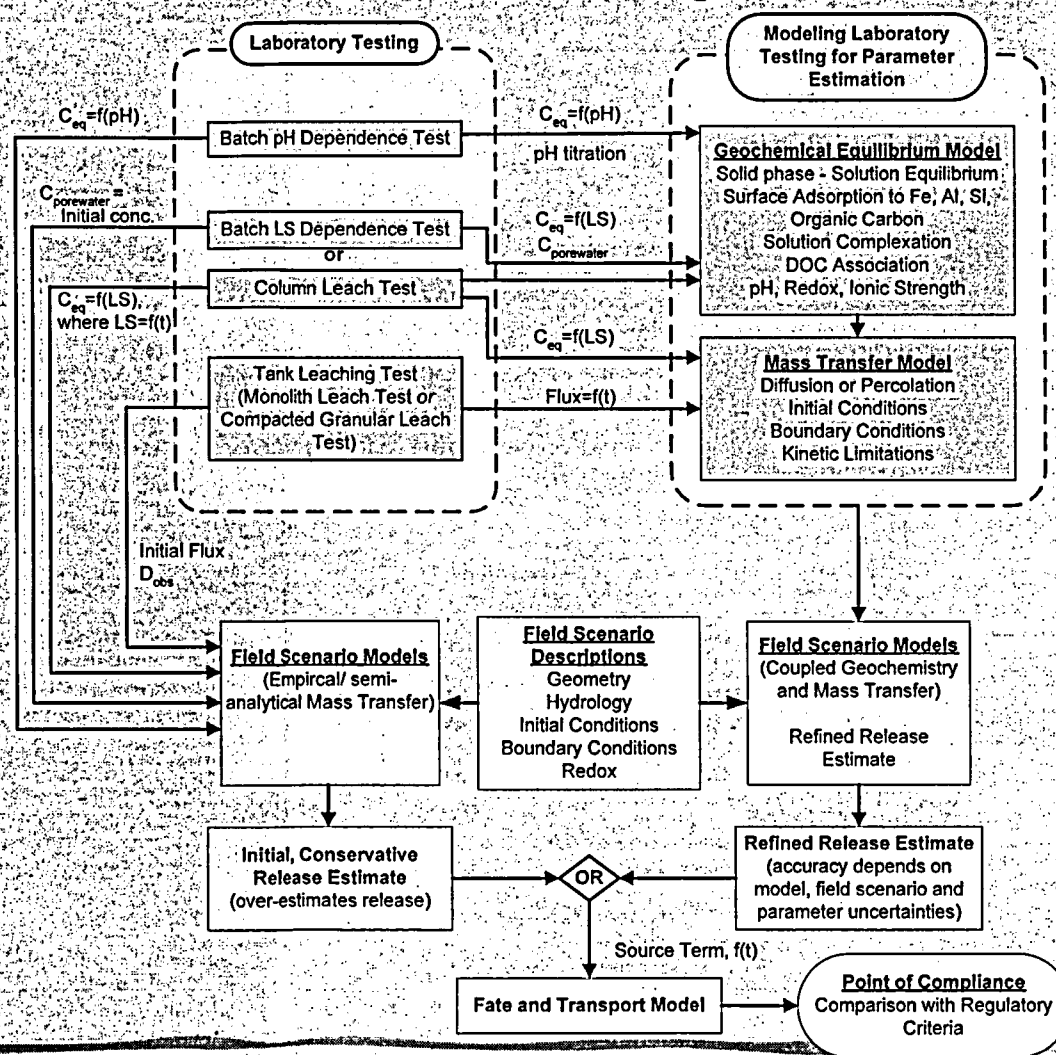


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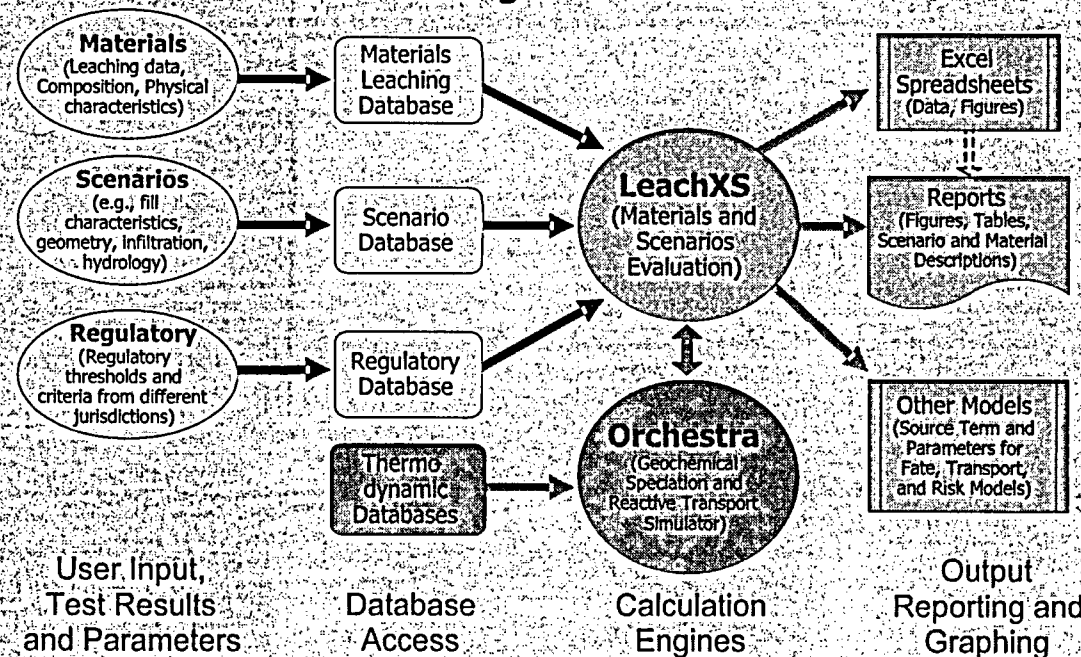
Integrated Use of Testing and Simulation



LeachXS – A tool for integrating experimental data, model development, parameter estimation and prediction

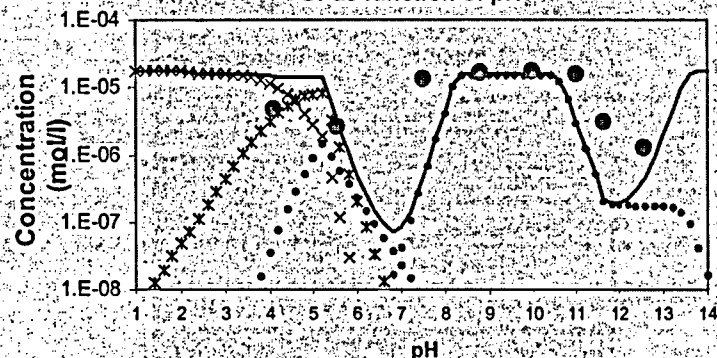
Software-based system for evaluating leaching

- Incorporates multiple processes and system configurations
- Data management/interpretation
- Geochemical analysis via ORCHESTRA (Meeussen, 2003)
- Database of material leaching information



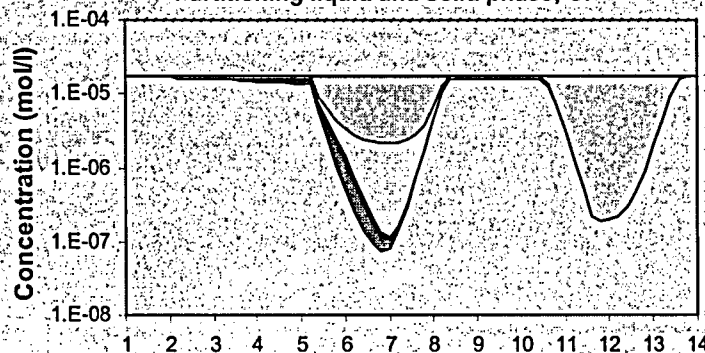
CHROMIUM SPECIATION IN MORTAR AND WATER

Cr as function of pH



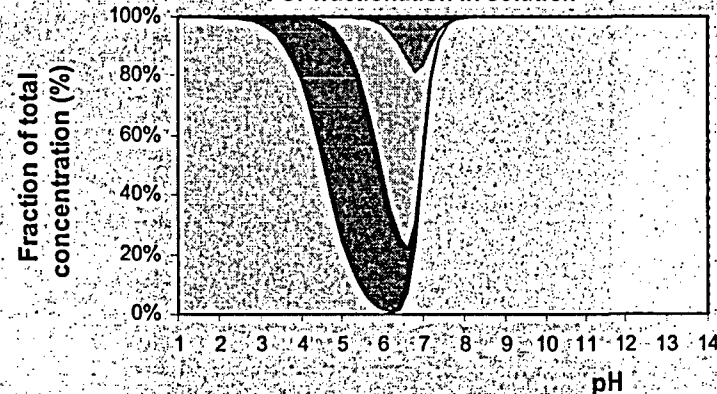
- Cement mortar
- $\text{CaCrO}_4[\text{aq}].\text{diss}$
- × $\text{Cr}[\text{OH}]_2+.\text{diss}$
- Model prediction
- × $\text{Cr}+3.\text{diss}$
- $\text{Cr}[\text{OH}]_2+.\text{diss}$

Partitioning liquid and solid phase, Cr



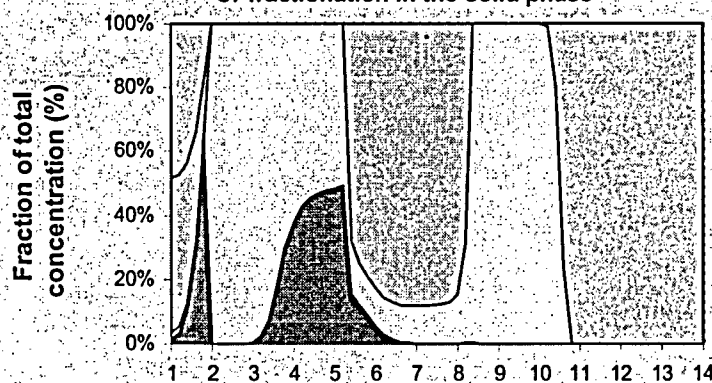
- Free
- POM-bound
- Clay
- $\text{Cr}[\text{OH}]_3[\text{A}]$
- DOC-bound
- FeOxide
- $\text{Ba}[\text{SCr}]\text{O}_4[96\%\text{SO}_4]$
- Cr-Ettringite

Cr fractionation in solution



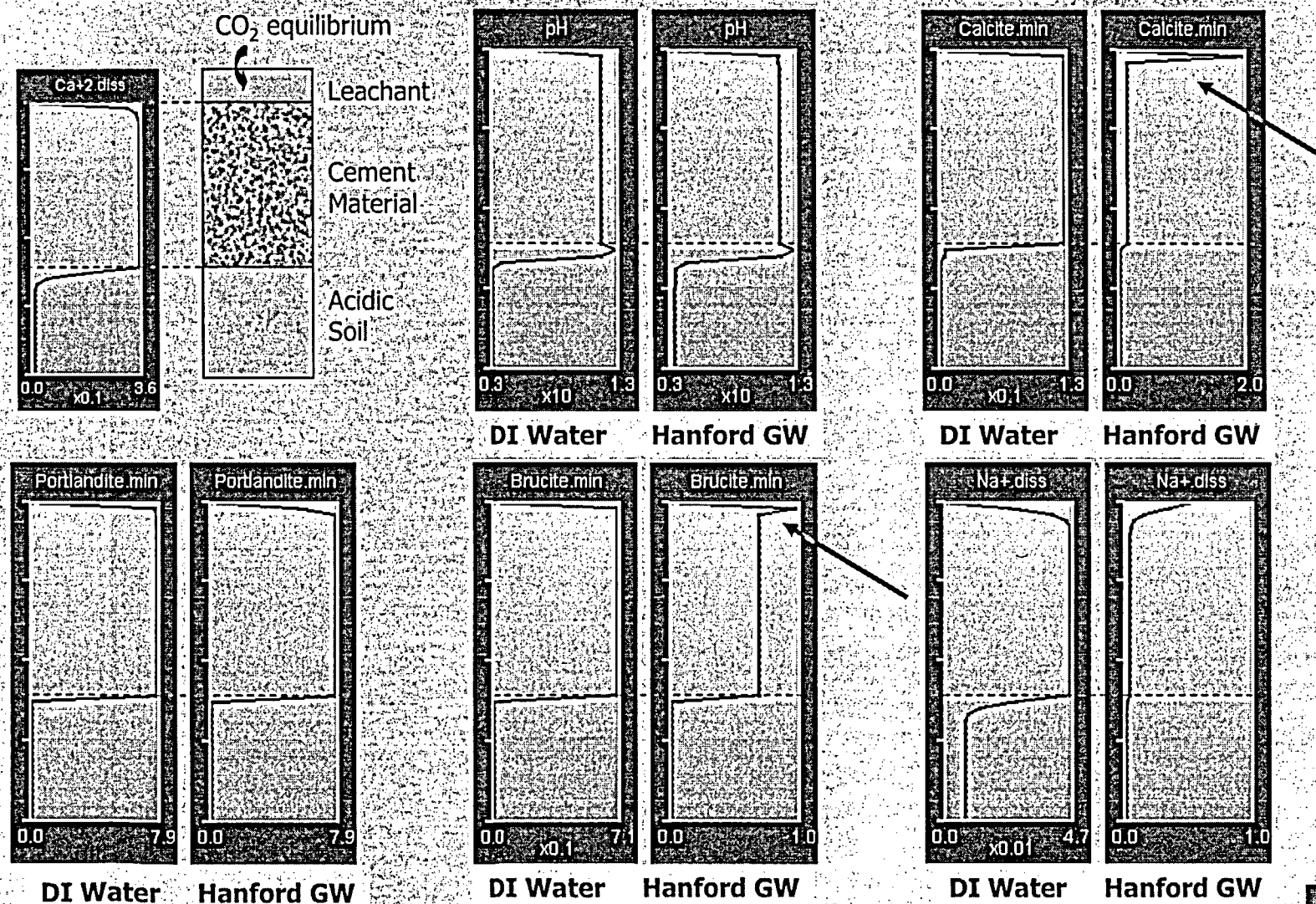
- DOC-bound
- $\text{Cr}+3.\text{diss}$
- $\text{Cr}[\text{OH}]_2+.\text{diss}$
- $\text{CaCrO}_4[\text{aq}].\text{diss}$
- $\text{Cr}[\text{OH}]_2+.\text{diss}$
- $\text{Cr}[\text{OH}]_3.\text{diss}$

Cr fractionation in the solid phase

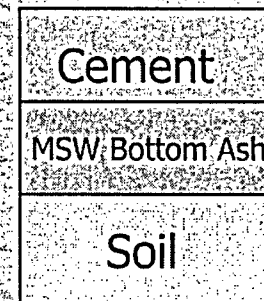
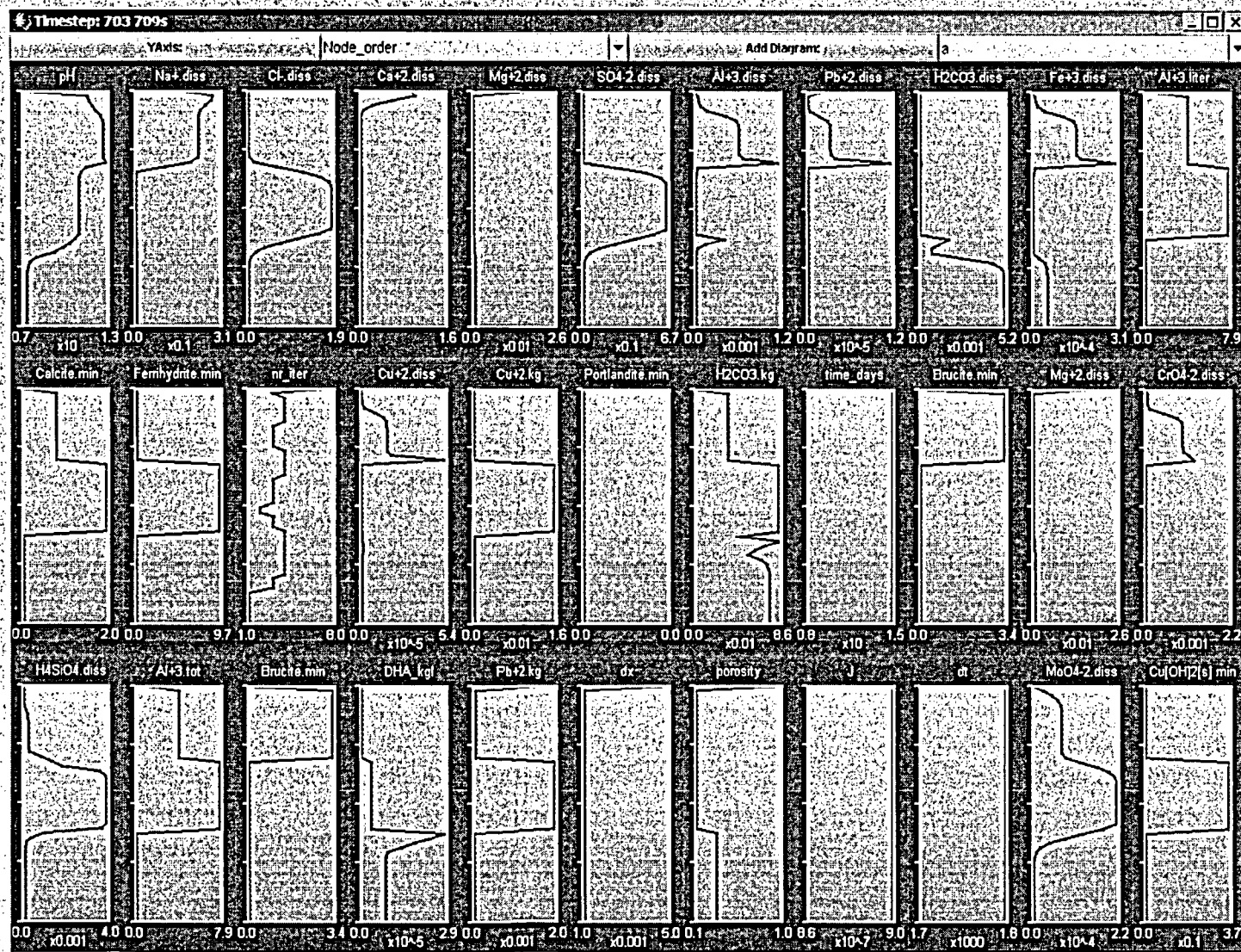


- POM-bound
- Clay
- $\text{Cr}[\text{OH}]_3[\text{A}]$
- FeOxide
- $\text{Ba}[\text{SCr}]\text{O}_4[96\%\text{SO}_4]$
- Cr-Ettringite

Leachant Simulation – Boundary Effects



MODELLING OF 3 LAYER SYSTEM WITH FULL CHEMICAL SPECIATION AND TRANSPORT



Suggested Path Forward

Process of continuous improvement, such that assessments incorporate "state of the art" understanding to extent practical

- Important for current assessments and future nuclear waste management (legacy and future wastes)
- Need to define short-term and long-term needs

Experimental studies coupled with model development and validation

- Formation/effect of boundary layers (e.g., CaCO_3 , oxidized layer)
- Moisture transport and status
- Oxidation rates
- Full geochemical model (equilibrium & mass transfer) for key systems
- Physical changes considering key geochemistry and mass transfer

Conclusions

Processes are not included in current DOE performance evaluations that can impact constituent release.

It is important to have an improved understanding and model(s) for near-term and longer-term DOE waste management decisions.

CRESP and SRNL, along with others, are currently working together to provide the needed evaluation system components.