

## **CENTER FOR NUCLEAR WASTE REGULATORY ANALYSES**

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### **TRIP REPORT**

**SUBJECT:** International Workshop on the Prediction of Long-Term Corrosion Behavior  
in Nuclear Waste Systems  
Project Number 20.01402.571, AI Number 01402.571.021

**DATE/PLACE:** November 26–29, 2001  
Cadarache, France

**AUTHOR:** D.S. Dunn

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## **TRIP REPORT**

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**DATE/PLACE:** November 26–29, 2001  
Cadarache, France

**AUTHOR:** D.S. Dunn

**PERSONS PRESENT:** D.S. Dunn [Center for Nuclear Waste Regulatory Analyses (CNWRA)] T.M. Ahn [U.S. Nuclear Regulatory Commission (NRC)] and about 50 representatives from various countries and organizations. See attachments 1 and 2 for a meeting agenda and a list of attendees.

### **BACKGROUND AND PURPOSE OF TRIP:**

The International Workshop on the Prediction of Long-Term Corrosion Behavior in Nuclear Waste Systems was organized to provide a forum for researchers working in the area of radioactive waste disposal to present methodologies, models and supporting data for predicting the lifetime of radioactive waste containment systems. The workshop attendees represented many countries including Japan, France, Belgium, Germany, India, Sweden, Finland, United Kingdom and the United States. The main goals of attending the workshop were to:

- Present the approach used by the NRC/CNWRA for the prediction of long-term performance of waste packages in the proposed repository at Yucca Mountain.
- Examine approaches used in other countries to predict long-term performance of radioactive waste containers.

The workshop was divided into sessions focused on: research and development program, models, research and development approaches and results, and analogues. A round table discussion on the prediction of long-term corrosion behavior was also held near the end of the workshop.

### **SUMMARY OF PERTINENT POINTS:**

The opening remarks were provided by Professor D.D. MacDonald From the Pennsylvania State University and Th. Lieven from Commissariat a l'Energie Atomique. The Technical symposium was divided into sessions for: R&D, Models, R&D Approaches and Results, and Analogues. A round table discussion was also held during the workshop.

Presentations that focused on the U.S. high-level waste program included a key note lecture from T.M. Ahn from the NRC, which provided a background to the risk-informed performance-based approach used by the NRC to evaluate a potential license application from the U.S. Department of Energy (DOE), as well as examples of issue resolution. J.S. Lee from Sandia National Laboratories described the DOE approach to modeling long-term performance of waste packages. Much of the material presented was identical to the material in the recently released Supplemental Science and Performance Analyses report. Some recent data was presented where the localized corrosion susceptibility of Alloy 22 was determined using cyclic polarization in calcium chloride and calcium chloride with calcium nitrate solutions. Localized corrosion was observed in concentrated calcium chloride solutions. The addition of calcium nitrate was observed to prevent the initiation of localized corrosion. D.S. Dunn from the CNWRA presented the approach used to model waste engineered barrier system performance in the Total-system Performance Assessment code.

Presentations by G. Engelhardt from OLI systems and D.D. Macdonald were focused on the determination of accumulated damage to Alloy 22 high-level waste container materials. G. Engelhardt presented a model for the deterministic prediction of localized corrosion of Alloy 22. D.D. Macdonald presented an approach for predicting damage to Alloy 22 high-level waste containers based on the prediction of localized corrosion susceptibility and passive corrosion rates using the point defect model. The accumulated damage function is conceptually similar to the present approach used by in the NRC/CNWRA performance assessment code. Uniform corrosion rates are determined to be the operative degradation mode in the absence of localized corrosion. The initiation of localized corrosion is based on the pit initiation potential that can be predicted using the point defect model. At present, the necessary parameters used in the model for the prediction of localized corrosion initiation are being obtained in tests conducted at the Frumkin Institute in Russia. Preliminary results suggest that localized corrosion is not initiated based on a pure sodium chloride environment and the damage to the container in 10,000 years by uniform corrosion results in a penetration of 1.6 to 1.8 mm [0.063 to 0.071 in.]. When asked about the possibility of selective dissolution, D.D. Macdonald responded that he does not believe that selective dissolution will occur over a long period.

Presentations from researchers in the French high-level waste program were focused on the prediction of oxidation and corrosion of carbon steel. Oxidation is anticipated in interim storage. Several presentations from the French researchers were also included in the analogues session. These presentations included characterization of corrosion products of buried iron-based archaeological artifacts and archaeological artifacts which were continuously indoors such as structural members in the Pope's Palace. The results of long-term testing of steels in compacted clay was also presented and compared to models. A presentation by P. Combrade from Framatome ANP cited many results of repassivation potential measurements conducted at the CNWRA as part of the NRC HLW program. His analyses of the data from the CNWRA with Alloy 825 and Type 316L stainless steel concluded that the critical chloride concentration observed in the repassivation potential versus chloride concentration plots may define regions where the repassivation potential measurement is dependent on crevice geometry (low chloride concentrations) or independent of crevice geometry (high chloride concentrations). Tests to confirm this analyses have not been conducted.

Presentations from Sweden and Finland were focused on the corrosion of copper. Data was presented from tests conducted in the Aspo hard rock laboratory where in situ corrosion tests have been conducted. Some of the instrumented test coupons are quite elaborate and include

many individual specimens and heaters to simulate the heat produced by radioactive decay. Several monitoring methods have been used to measure the corrosion rates.

A general observation from the workshop was that in almost all of the high-level waste programs outside of the U.S., the choice of material is based in part on the expected composition of the environment in terms of aggressive species and redox potential. This approach is particularly useful for copper where it can be shown that under suitable environmental conditions, long container lifetimes are predicted

A presentation by Professor Balasubramanian showed the history of ancient iron artifacts in India as well as the results of attempts to characterize both the base metal and the oxides and corrosion products. The long life of the iron artifacts in India was attributed to the inclusion of slag that contained impurities which contributed to the formation of a protective oxide film.

A round table discussion was held to address the limitations and needs for making long-term predictions of nuclear waste systems. The approach of the repassivation potential used in the NRC/CNWRA TPA code was discussed and compared to alternate methods to predict the initiation of localized corrosion. Professor D.D. MacDonald, with funding from the DOE is working on a localized corrosion initiation model based on pit initiation potential and the point defect model of passive films. Professor MacDonald suggested that the use of the repassivation potential for the long-term prediction was a strictly empirical approach which may be too conservative. In the subsequent discussion, it was stated that there is much evidence in many systems that suggests the repassivation potential is not an overly conservative parameter. Nevertheless, mechanistic models that can be used to predict either the initiation or repassivation of localized corrosion on passive materials do not exist. The present effort to develop such a model by professor MacDonald may yield a useful tool for DOE to evaluate container performance.

#### **SUMMARY OF ACTIVITIES:**

Following the workshop, the workshop attendees visited the CEA Cadarache facilities. The two areas visited included the development of fusion energy, and solar cell research and development. During the visit, it was stated that the development of fusion energy systems is anticipated to take several decades. Meaningful power from solar energy is still expensive compared to other energy sources, however the cost is continuously decreasing. Most of the applications for solar power systems are in areas where no centralized power grid exists.

Literature collected during the visit to the CEA Cadarache center is available from the author.

#### **CONCLUSIONS:**

The workshop provided an excellent avenue for obtaining most current information on different approaches to predict corrosion behavior. In addition, the workshop provided an opportunity to discuss various aspects of the modeling approaches and measurement methods. Discussion of the methodology to predict the long-term initiation of localized corrosion of highly corrosion resistant material such as Alloy 22 provided an excellent forum for the peer-review of repassivation potential criteria that is presently used by the NRC/CNWRA. It should be noted that the alternative approach using the accumulated damage function where the initiation of localized corrosion is determined by the localized corrosion initiation potential relies on

experimental data for the statistical distribution of localized corrosion initiation and death as a function of the environment. Professor McDonald did not provide a time frame for abstracting parameters from the experiments for the development of the model. It was stated that the initiation of localized corrosion on Alloy 22 was difficult.

As noted earlier, the approach used in other programs outside of the U.S. have performed an assessment of the expected repository environment. In many cases the susceptibility of the material to a particular degradation mode such as localized corrosion is directly compared to the range of expected conditions in the repository environment. In the case of the Swedish radioactive waste disposal program, the selection of materials for the waste containers are tested in the expected repository environment using multiple test specimens that allow long-term measurement of corrosion rates. The corrosion rates measured during testing are then verified at the conclusion of the tests. A similar approach may be useful to determine the range repository conditions after closure and assess the expected degradation modes for waste packages in the proposed repository at Yucca Mountain.

**PROBLEMS ENCOUNTERED:**

None.

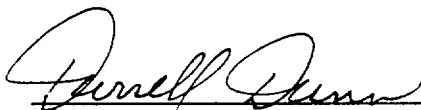
**PENDING ACTIONS:**

None.

**RECOMMENDATIONS:**

Attendance at such meetings provides an excellent opportunity to discuss the methodologies, concerns and problems encountered in testing and modeling the long term performance of materials for the containment of nuclear waste. It also provides an opportunity to present the approach used in the NRC/CNWRA high-level waste program and obtain valuable criticisms and suggestions as well as gain insights into the corrosion behavior of waste containment materials proposed in foreign programs.

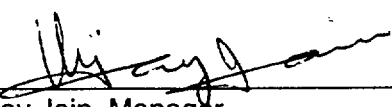
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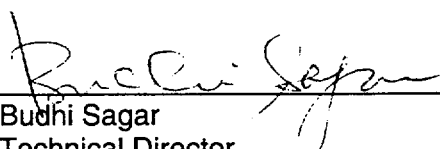
Darrell Dunn  
Senior Research Engineer

12/13/01  
Date

**CONCURRENCE:**

  
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Vijay Jain, Manager  
Corrosion Science & Process Engineering, Element

12/13/01  
Date

  
\_\_\_\_\_  
Budhi Sagar  
Technical Director

12/13/2001  
Date

Attachment

DD:jg

EFC event 256

# **International Workshop on "Long term corrosion behaviour in nuclear waste systems"**

26-29 November 2001, Cadarache, France

<b>MONDAY</b> 26 <sup>th</sup> November 2001	<b>TUESDAY</b> 27 <sup>th</sup> November 2001	<b>WEDNESDAY</b> 28 <sup>th</sup> November 2001	<b>THURSDAY</b> 29 <sup>th</sup> November 2001
	8h00-8h30 Registration		
	8h30-9h00 <b>Opening addresses</b>	8h30-10h00	8h30-10h00
	9h00-10h00 <b>R&amp;D program session</b>	<b>Model session (cont'n)</b>	<b>R&amp;D approaches &amp; results</b>
	10h00-10h30 coffee break	10h00-10h30 coffee break	10h00-10h30 coffee break
	10h30-12h30 <b>R&amp;D program session</b>	10h30-12h30 <b>R&amp;D approaches &amp; results</b>	10h30-12h00 <b>R&amp;D approaches &amp; results</b>
			12h00-12h30 <b>Closing session</b>
	12h30-14h00 Lunch	12h30-14h00 Lunch	12h30-14h00 Lunch
	14h00-16h00	14h00-16h30	14h00-17h00 <b>Visit of the CEA Cadarache Centre</b>
	<b>Model session</b>	<b>Analogue session</b>	
	16h00-16h30 Coffee break		
	16h30-18h30	16h30-17h00 Coffee break	
	<b>Model session (cont'n)</b>	17h00-18h30 <b>"Table Ronde"</b>	
18h00-20h00 Registration & welcoming	19h30 Dinner	19h30 Official Dinner	

ATTACHMENT 1

# **International Workshop on "Long term corrosion behaviour in nuclear waste systems"**

26-29 November 2001, Cadarache, France

## **FINAL PROGRAM**

<b>Monday 26<sup>th</sup> November</b>			
<b>Registration &amp; welcoming 18h00-20h00</b>			
<b>Tuesday 27<sup>th</sup> November</b>			
	<b>Registration 8h00-8h30</b>		
<b>Opening session 8h30- 9h00</b>			
<b>Welcome addresses:</b>			
<p><b>LJEVEN Th.</b>, Chef du Service de la Corrosion et du Comportement des Matériaux dans leur Environnement (Head of the CEA corrosion department)</p> <p><b>MACDONALD D.D.</b>, Director of the Center for Electrochemical Science Technology (Pennsylvania State University, USA)</p>			
<b>Opening address:</b>			
<p><b>BERANGER G.</b>, Université de Technologie de Compiègne (France), Membre de l'Académie des Technologies (<i>Member of the French Academy of Technologies</i>)</p>			
Few words about the organisation : <b>FERON D.</b> , Head of the CEA Aqueous Corrosion Laboratory			



Tuesday 27th November			
<b>R&amp;D Program session</b> 9h00 – 12h30			
<b>Chairman : PINARD LEGRY Gérard</b>			
<b>AHN Tae M. (US Nuclear Laboratory Commission, USA)</b> U.S. Nuclear Regulatory Commission (NRC) Corrosion Program for High-Level Nuclear Waste (HLW) <i>Key note lecture</i>			
<b>PLAS Frédéric (ANDRA, France)</b> Containers and overpacks for high-level wastes in geological disposal conditions : French corrosion program <i>Key note lecture</i>			
<i>Coffee Break 10h00-10h30</i>			
<b>KURSTEN Bruno (SCK-CEN, Belgium)</b> In situ testing of waste forms and container materials: contribution to the identification of their long-term behaviour <i>Key note lecture</i>			
<b>SANTARINI Gérard (CEA-Saclay, France)</b> COCON : Corrosion research program for long term interim storage conditions			
<b>SAARIO Timo (VTT Manufacturing Tech., Finland)</b> Approaches chosen to predict the effect of different forms of corrosion on copper in disposal conditions of spent fuel			
<i>Lunch 12h30 -14h00</i>			

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<b>Tuesday 27th November</b>	
<b>Model Session 14h00-18h30</b>	
<b>Chairman : AHN Tae</b>	
<b>MACDONALD Digby D. (Penn State University, USA)</b>	The holy grail : Deterministic prediction of corrosion damage thousands of years into to future <i>Key note lecture</i>
<b>FOCT François (EDF, France)</b>	Semi-empirical model for carbon steel corrosion in long term geological nuclear waste disposal <i>Key note lecture</i>
<b>ENGELHARDT George (OLI Systems, USA)</b>	The deterministic prediction of localized corrosion damage to alloy C-22 HLNW canisters
<b>BADIALI Jean Pierre (CNRS, France)</b>	Numerical simulations of simple processes associated with corrosion, diffusion and formation of a passive layer
<b>Coffee Break 16h00-16h30</b>	
<b>LEE Joon H. (Sandia National Laboratories, USA)</b>	A stochastic model for long term performance of waste package for high nuclear waste disposals
<b>BATAILLON Christian (Laboratoire d'Etude de la Corrosion Aqueuse, CEA-Saclay, France)</b>	Application of the point defect model to modelling the corrosion of iron based canisters in geological repository
<b>URQUIDÍ-MACDONALD Mirna (Penn State University, USA)</b>	Transients in the growth of passive films on high level nuclear waste canisters
<b>HOERLE Stéphane (Laboratoire d'Etude de la Corrosion Aqueuse, CEA-Saclay, France)</b>	A contribution to the modelling of atmospheric corrosion of iron
<b>Dinner 19h30-21h00</b>	

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<b>Wednesday 28th November</b>	
<b><u>Model Session (continuation)</u> 8h30-10h00</b>	
<b>Chairman : AHN Tae</b>	
DESGRANGES Clara (Laboratoire d'Etude de la Corrosion Non Aqueuse, CEA-Saclay, France)	Model for low temperature oxidation during long term interim storage-
DUNN Darrel (Southwest Research Institute, USA)	Modelling corrosion of alloy 22 as a high level radioactive waste container material
POURBAIX Antoine (CEBELCOR, Belgium)	Long-term prediction of corrosion of passive materials for high-level waste containers based on protection potential
<i>Coffee Break 10h00-10h30</i>	
<b><u>R&amp;D approaches &amp; results</u> 10h30-12h30</b>	
<b>Chairman : LEE Joon H.</b>	
WALTERS Steve (AEA Technology, UK)	A methodology for assessing the integrity of spent nuclear fuel in long term dry storage
ALVES Helena (KRUPP VDM, Germany)	Long-term thermal stability and corrosion behaviour of welded and unwelded alloy 59 for rad waste containers
NAKAYAMA Guen (Ishikawajima-Harima Heavy Industries, Japan)	Assessment of Crevice Corrosion and hydrogen-induced stress corrosion cracks in Titanium-carbon steel composite overpack for geological disposal of high-level radioactive waste
WOOD Peter (United Kingdom Nirex Ltd, UK)	The choice of stainless steel for ILW waste containers
<i>Lunch 12h30-14h00</i>	

<b>Wednesday 28th November</b>	
<b><u>Analogue session</u> 14h00-16h30</b>	
<b>Chairman : POURBAIX Antoine</b>	
<b>DAVID Daniel (UTC, France)</b>	Archaeological analogue studies for the prediction of long term corrosion on buried metals <i>Key note lecture</i>
<b>BALASUBRAMANIAM R. (Indian Institute of Technology, India)</b>	Corrosion resistance of the Delhi iron pillar - Scale characterization and passive film growth models <i>Key note lecture</i>
<b>TICE David (Serco Assurance, UK)</b>	Expansion due to anaerobic corrosion of steel and cast iron : Experimental and natural analogue studies
<b>NEFF Delphine (ANDRA, France)</b>	An analytical study of corrosion products formed on buried ferrous archaeological artifacts
<b>DILLMANN Philippe (CEA-Saclay/LPS, France)</b>	Rust characterization of ancient iron artifacts exposed to indoor atmospheric corrosion
<b>PONS Emmanuelle (UTC, France)</b>	Long-term behaviour of iron in clay soils : A study of archaeological analogs
<i>Coffee break 16h30-17h00</i>	
<b><u>Table ronde</u> 17h00 – 18h30</b>	
<b>« How to predict long term corrosion behaviour ? »</b>	
<i>Official Dinner 19h30-21h00</i>	

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<b>Thursday 29th November</b>	
<b><u>R&amp;D approaches &amp; results</u> (continuation) 8h30-12h00</b>	
<b>Chairman : LEE Joon H.</b>	
<b>SELLIN Patrick (SKB, Sweden)</b>	The performance of a copper canister for geologic disposal of spent nuclear fuel in granitic rock <i>Key note lecture</i>
<b>ROSBORG Bo (Rosborg Consulting, Sweden)</b>	Measurements of copper corrosion in the LOT project at the Äspö hard rock laboratory
<b>JULLIEN Michel (CEA-Cadarache, France) — <u>CANCELLED</u></b>	Low-alloy steel behaviour in compact clay: two long term tests for corrosion prediction
<i>Coffee break 10h00-10h30</i>	
<b>COMBRADE Pierre (Framatome, France)</b>	Crevice corrosion of passive materials in long term geological nuclear waste disposal
<b>WOOD Peter (United Kingdom Nirex Ltd, UK)</b>	Control of environmental conditions during storage of ILW waste packages
<b>CORBEL Catherine (CEA-Saclay)</b>	Effect of irradiation on long term alteration of oxides and metals in aqueous solutions
<b>PRITCHARD Andrew (Corrosion &amp; Fouling Consultancy, UK)</b>	An approach towards assessing the effects of microbially-influenced corrosion in nuclear waste systems
<b>Closing session 12h00- 12h30</b>	
<b>MACDONALD D.D.,</b> Director of the Center for Electrochemical Science Technology (Pennsylvania State University, USA)	
<b>FERON D.,</b> Chef du Laboratoire d'Etude de la Corrosion Aqueuse (Head of the CEA aqueous Corrosion Laboratory, France)	
<i>12h30-14h00 Lunch</i>	
<b>14h00-17h00 Visit of CEA Cadarache Centre</b>	

International Workshop "Long term corrosion behaviour in nuclear waste systems"  
26-29/11/2001, Cadarache (France)

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