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Mr. Samuel J. Collins
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Dear Mr. Collins:

The industry has performed an evaluation of the behavior of spent fuel stored in spent fuel pools following a catastrophic event, i.e., a beyond design basis seismic event that drains and so damages the spent fuel pool that replacement of cooling water is impossible. The industry has expressed its views on several occasions regarding the questionable value of focusing resources on the consequences of such an unlikely event. Nevertheless, if the NRC intends to base its regulations on this event, it is imperative that it be characterized using realistic phenomenology.

I am enclosing a technical report prepared by Dr. Robert Henry of Fauske and Associates. Dr. Henry's report evaluates the phenomenology and consequent releases of fission products with results that differ substantially from those postulated by the Advisory Committee on Reactor Safeguards (ACRS). The substance of this report was presented to the ACRS on November 2, 2000.

I urge you to consider the views expressed in the attached technical report and incorporate them in finalizing the NRC report on the risk of spent fuel pools. If I can be of any assistance to you or your staff please call me or Lynnette Hendricks (202 739-8109, or, LXH@NEI.org).

Sincerely,

Ralph E. Beedle

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***THE RESPONSE OF THE SPENT
FUEL POOL TO POSTULATED
ACCIDENT CONDITIONS***

Submitted To:
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1.0 INTRODUCTION

When considering the risk of nuclear power installations, it is also appropriate to evaluate the defense-in-depth for assuring cooling of the fuel assemblies in the spent fuel pool. This fuel has been used in a reactor and generally has exhausted its useful life for the current Light Water Reactor (LWR) core designs. Typically, fuel assemblies in the spent fuel pool have been out of the reactor for a number of years and the decay power has decreased over time. Nonetheless, this fuel is cooled by submerging the fuel assemblies in water which also provides the necessary shielding for plant personnel working near the pool.

The spectrum of accident conditions considered are those where this cooling function could be lost, which means either the loss of cooling of the spent fuel pool and the subsequent boildown of the water inventory or a postulated leak in the pool which enables the water inventory to drain down and uncover the fuel pins. Of these, the latter results in the fastest uncovering of the fuel, but with the design of the typical pool, this leakage must result from a postulated catastrophic event. For example, it has been assumed to result from a very strong seismic event that destroys the pool integrity, including its stainless steel liner. In contrast, for those cases in which the accident sequence is postulated to be a loss of spent fuel pool cooling, this loss would have to be sustained for several days before the fuel pins would be uncovered. Consequently, comparably simple corrective actions would be sufficient to sustain the pool cooling by providing makeup water to the pool.

Such a large seismic event is highly improbable. As a result, those conditions leading to releases from the spent fuel pool are very low probability events. Nonetheless, evaluations of such conditions should focus on realistic failure conditions and the subsequent heatup and degradation of the fuel assemblies. Without such focus, it is difficult to provide quantitative assessments of the accident behavior and the risk to the public or draw any meaningful risk insights from the study.

2.0 SPENT FUEL POOL EVALUATIONS

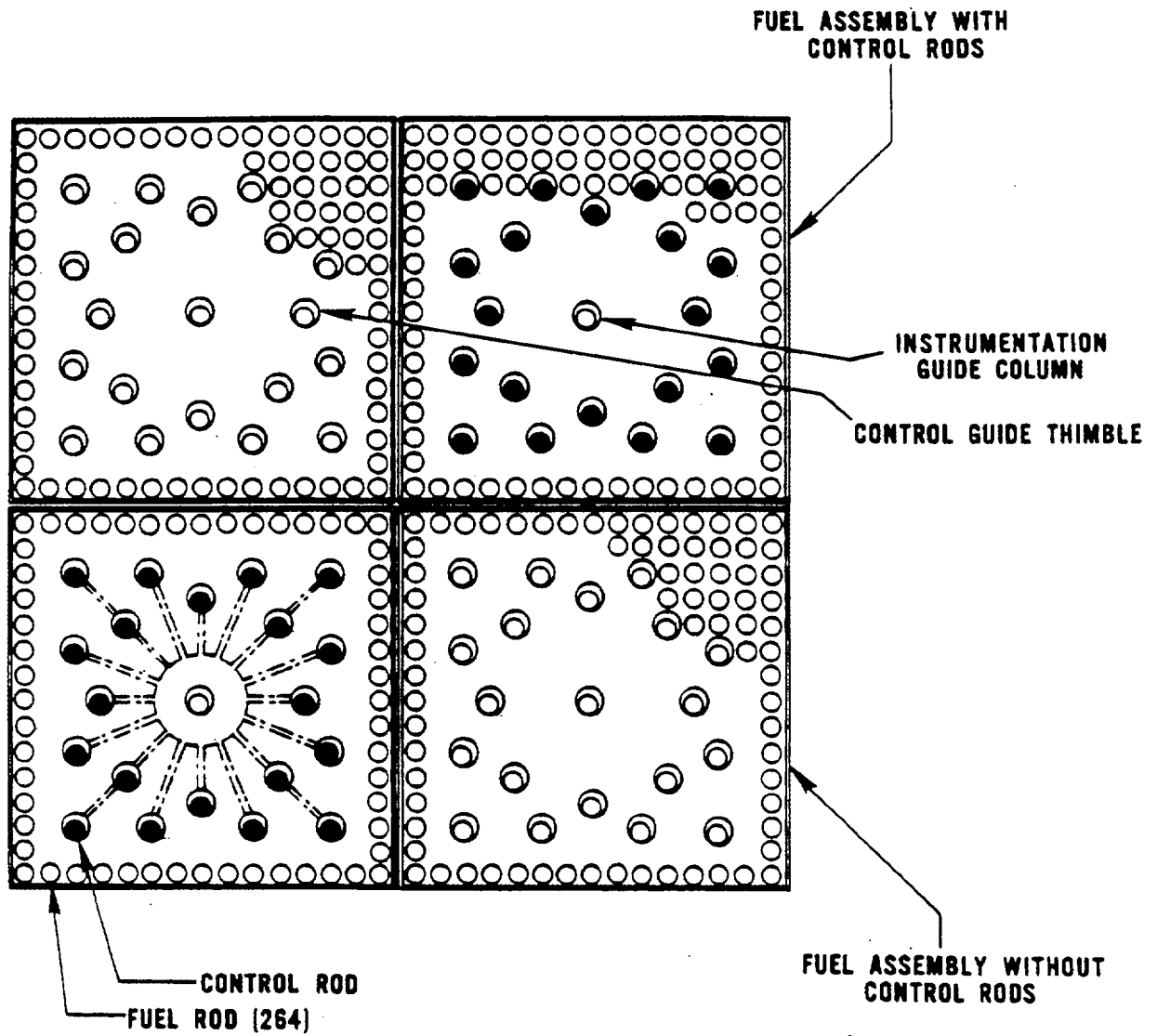
Table 1 gives guidance for such evaluations to assure that the results are useful. In particular, the results should address those issues relevant to both operating plants and those being decommissioned to enable the operating organization and the NRC to use the results in the most productive manner.

Table 1: Approach to Evaluations	
<ul style="list-style-type: none">• All evaluations should use a mechanically identified failure condition.• Evaluations should assess the results of potential recovery actions consistent with the postulated accident initiator.• Evaluations should consider all mechanisms for cooling and for energy generation, including the influence of water vaporization in the lower regions of the pool as well as natural circulation of air.	

When assessing the response of fuel assemblies to the loss of water inventory, the elements listed in Table 2 need to be considered. The first and second elements are the most important. The third element identifies those possible open channels within individual fuel assemblies where control rods may have been inserted for PWR designs. Figure 1 illustrates such a configuration and the possibility that openings exist or perhaps are only partially filled with spent control rod assemblies. The configuration will depend on the plant specific utilization of the spent fuel pool. However, if these openings are available, they should be part of the evaluation since they are regions of comparatively low frictional pressure drop and therefore permit more local flow.

Table 2: Focus for Analytical Models	
<ul style="list-style-type: none">• Spent fuel pool is at atmospheric pressure.• Flow within the fuel assemblies is laminar, i.e., resistances are well characterized by standard representations.• Opening in individual fuel assemblies are influential flow paths and should be considered.• The fuel assembly distribution within the pool does not matter for those accident conditions where the water inventory decreases below the top of fuel until the water is at about 70% of the fuel assembly height. The fuel assembly distribution would matter in the multi-dimensional flow pattern that would develop at lower water levels, i.e. if a thermal plume would develop.	

Figure 1 Schematic of Fuel Assemblies in a High
Density Racking for the Spent Fuel Pool



The fourth element addresses the extent of cooling for partially uncovered fuel assemblies. The analysis used here is from the evaluations performed in the Technical Basis Report (TBR) for Severe Accident Management Guidance (SAMG) (Henry, 1992). Specifically, the analyses in Section Z of Volume 2 of the TBR show that when considering a condition like that illustrated in Figure 2, the peak cladding temperature can be approximated by the equation listed in Table 3. The assumptions for this evaluation are included in Table 3. Figure 3 is taken from the TBR and shows the results of this calculation, which is (for practical purposes) independent of the decay power in the fuel assemblies. For this steady-state representation, the peak cladding temperature remains below levels where significant oxidation can occur as long as approximately 70% of the fuel is covered. Note that this evaluation does not characterize the time required for the fuel temperature to increase to the levels shown in Figure 3. With the low decay power in the fuel and the effective heat removal, this would occur over many hours at the very minimum. However, the important element is that sufficient heat removal can be achieved even with part of the fuel uncovered.

Table 3: Estimation of Peak Cladding Temperature for Assumed Accident Conditions Where the Top of the Fuel is Uncovered
-- Assumptions --

1. The process is assumed to progress quasi-steady manner.
2. Steam and water are the only fluids in the core.
3. The inlet water is at the saturation temperature T_{sat} .
4. The decay power (QD) is constant along the fuel pin length.
5. The collapsed water level (y) can be used to represent the covered portion of the fuel assemblies.
6. The cladding temperatures remain low enough that the energy released by Zircaloy oxidation is an insignificant fraction of the decay power.
7. This results in
$$T_o - T_{sat} = \left[\frac{1-y}{y} \right] \frac{h_{fg}}{c_{pg}}$$
where h_{fg} is the latent heat of vaporization for water and c_{pg} is specific heat at constant pressure for steam.

Figure 2: Quasi-Steady Heat Removal

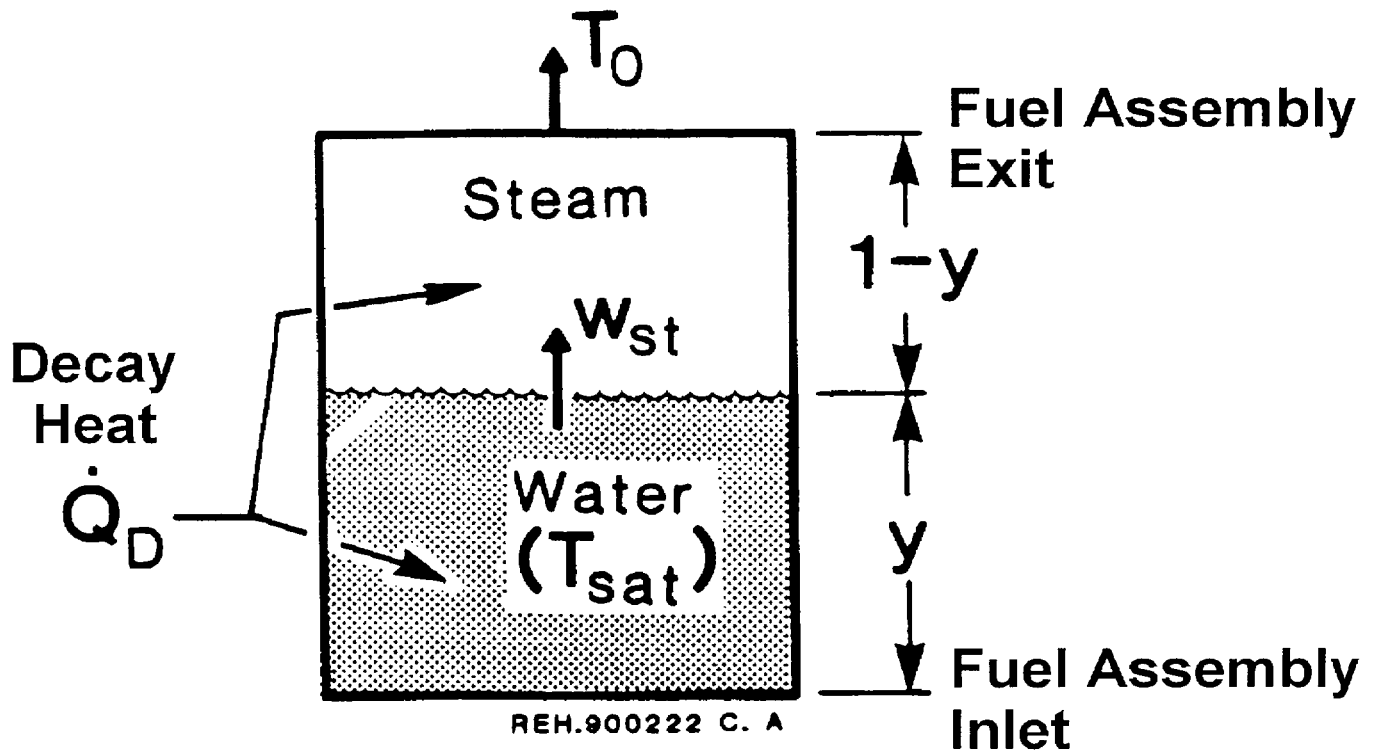


Figure 3: Quasi-Steady Cladding Temperature for a Partially Uncovered Group of Fuel Assemblies

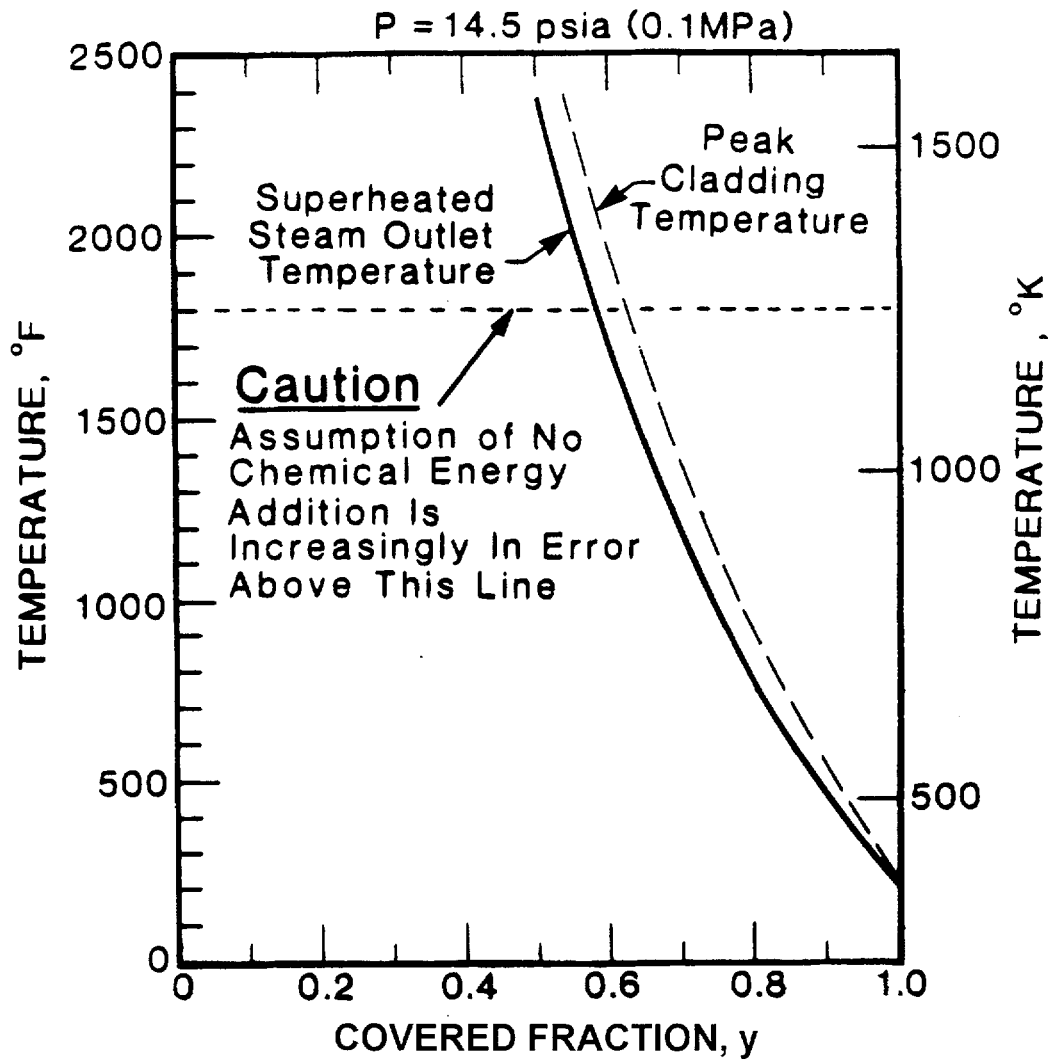


Table 4 provides a perspective on the rate of water inventory depletion. This shows that with an average of 5 kw/assy and 1000 fuel assemblies, the total power in the spent fuel pool would be 5 MW. Given the assumed pool dimensions, the boildown rate for that part of the pool above the top of the fuel assemblies would be approximately 5.4 in/hr assuming that the steaming rate from the spent fuel pool did not experience any condensation process and reflux (return) of the condensate back into the pool. As illustrated in Table 4, this water inventory loss can be stopped with the water addition rate of approximately 35 gpm.

Table 4: Example of Postulated Accident Condition and the Response Boildown Rate	
•	Assume an average power of 5 kw/assy and 1000 fuel assemblies = 5 MW.
•	Assume the pool is 27 ft. (8.2 m) x 23 ft. (7.0 m).
•	Boildown rate when the water level is above the fuel is about 5.4 in/hr. (14 cm/hr).
•	If the water level progresses into the fuel assembly, this rate is then about 9 in/hr. (23 cm/hr.).
•	This boildown can be stopped with a water addition rate of about 35 gpm.

With a decay power of 5 MW and 1000 fuel assemblies ($\sim 5 \times 10^5$ kg) the adiabatic heatup rate is about 0.017°C/sec. Using this as a conservative assessment (understates the interval), the interval for the fuel assembly temperature to increase from 400 K to 1000 K is 10 hrs. With the cooling provided by heat of evaporation of the decreasing water inventory and air natural circulation, the time period would be even longer.

For those assessments in which the fuel is uncovered, the natural circulation flow of air through the fuel bundles can be approximated by

$$\Delta P = f \left(\frac{L_1}{D_h} \right) \frac{\bar{\rho} U^2}{2} = \Delta \rho g L_h \quad (1)$$

where D_h is the hydraulic diameter of the fuel assemblies, L_1 is the path length for the gases traveling through the core, $\bar{\rho}$ is the average gas density for the fluid, U is the fluid velocity, g is

the acceleration of gravity, L_h is the length over which the effective density difference occurs and $\Delta\rho$ is the density difference in the gas flow through the core. These density differences are due to temperature differences as well as changes in the gas mixture as a result of oxidation in the fuel pin cladding, i.e. changing air to nitrogen. The laminar flow friction factor can be expressed by

$$f = \frac{64}{N_{Re}} \quad (2)$$

where N_{Re} is the Reynolds number for the flow defined by

$$N_{Re} = \frac{U D_h}{\nu} \quad (3)$$

with ν being the gas kinematic viscosity.

Considering that the gas density continuously changes along the fluid path, the density difference that is responsible for the flow can be approximated as half of the maximum density difference which would be derived from the maximum temperature difference. Hence, this density difference can be expressed by

$$\Delta\rho = \frac{\Delta\rho_{max}}{2} \quad (4)$$

with the heat removal rate for the gaseous flow described by

$$\dot{Q}_R = \bar{\rho} A_F U c_p \Delta T_{max} \quad (5)$$

Substituting and solving for the maximum temperature difference in the flow results in

$$\Delta T_{\max} = \bar{T} \left[\frac{\dot{Q}_R}{A_F} \left(\frac{L_1}{L_h} \right) \right]^{1/2} \left[\frac{64 \nu}{g D_h^2} \frac{R}{P M_w c_{pg}} \right]^{1/2} \quad (6)$$

where \bar{T} is the average temperature of the gas in the spent fuel, A_F is the flow area, R is the universal gas constant, P is the local pressure (1 bar), M_w is the molecular weight of the gas and c_{pg} is the gas specific heat. With this expression, one can assess the extent of heat removal as well as predict when the lack of adequate heat removal would enable the fuel temperature to escalate to conditions where cladding oxidation could be anticipated. To assess the possible flow paths, the height of the fuel bundle can be taken as the parameter L_h whereas the length of the flow path through the fuel is either assumed to be a vertical height for those loading patterns with an open slot next to the assembly or twice the vertical height for a densely packed geometry. In the latter case, the flow is considered to be downward through the bundle in one location and vertically upward in an adjacent location. For this case, the vertical flow area would be taken as one-half of the total flow area and the length of the flow path is twice the length of the fuel assemblies.

One of the pool loading strategies discussed by Sailor et al. (1987) is to provide more open regions (up to one foot) on all sides of recently discharged fuel (less than two years). This configuration would mean that the flow path length for the higher power (more recently discharged) fuel assemblies is the height of the assemblies with the older fuel experiencing a length twice this long. Hence, this would be a hybrid configuration that could be examined with these simplified calculations.

As the water inventory in the spent fuel pool decreases, the fuel pins begin to heatup. For the Zircaloy fuel pin cladding exposed to air, as the temperatures exceed approximately 1000°K, the oxidation will begin to contribute a nontrivial energy increase to the process. As a result, the fuel pin cladding will heat up faster and the oxidation will continue to increase with increasing temperature. With this unstable configuration, the chemical energy release from the cladding oxidation becomes the major contributor to the core heatup and degradation. This is also the case for those accident sequences which begin with an "at power" condition as was the case for the TMI-2 accident. Therefore, any extensive core damage is attributed to the chemical energy

release even though the inadequate removal of decay power was the initiating condition that caused the cladding to reach temperatures where such rapid oxidation could occur.

Substituting the parameters given in Table 4, the temperature for the gas flow through the bundle would eventually reach values in excess of 1200 K and significant oxidation would occur between the Zircaloy cladding and the air flowing through the bundle. Given these conditions, the chemical energy release would dominate the power generation within the spent fuel bundles and as a result, the heatup, melting and relocation of material would be expected to be similar to the condition for the "at power" evaluations that have been performed in the past. In this regard, the heatup rate of the fuel pin and cladding is limited by the flow rate of air circulated into the bundle to supply oxygen for the oxidation process. For the "at power" evaluations, the limitation is one of steam starvation since this is the oxygen supply to sustain the oxidation process. For the assumed condition of a completely drained spent fuel pool the heatup would be limited by air starvation since this is the major oxygen source for cladding oxidation.

Conceptually, the general characterization of the fuel heatup, oxidation of the fuel pin cladding, clad melting and relocation as well as liquefaction of the uranium dioxide by the unreacted molten Zircaloy would be expected to be similar to the fuel damage configuration observed for the TMI-2 accident. For the spent fuel pool, the fuel pin heatup would be limited by the oxygen supply to the cladding and the fuel pin configuration. Those configurations presenting the most extensive surface area would more fully support the oxidation process. With the natural circulation being driven by the temperature differences between the incoming gas and that in the reaction zone, the oxygen supply rate would initially be increased by the higher cladding temperatures caused by oxidation. Subsequently, the cladding surface area would rapidly decrease as the materials reach melting and liquefaction temperatures and relocate downward into a more compacted configuration. This relocation diminishes the oxidation effects in two ways: (a) the available surface area for oxidation decreases, and (b) the open areas to support natural circulation gas flow decrease as the fuel geometry becomes more pin compacted. At some point the oxidation is self limiting and results in a highly compacted configuration that supports essentially no continued oxidation.

The reconfiguration of fuel assemblies has two important influences on the fission product releases. First, the release of ruthenium is strongly affected by its chemical state with the metallic ruthenium being far less volatile than the oxides (Williamson and Beethan, 1990 and Iglesias et al., 1990). For example, the Handbook of Physics and Chemistry (Hodgman et al., 1958) gives the boiling point of ruthenium metal as greater than 2700°C whereas it lists the oxides of ruthenium dioxide (RuO_2) and ruthenium tetra oxide (RuO_4) as decomposing at very low temperatures. Similar large differentials are quoted in Lange's Handbook of Chemistry (Dean, 1985); namely a boiling point of ruthenium metal as greater than 4000°C with the dioxide decomposing at a very low temperature. Of particular importance is the unreacted Zircaloy that would remain within the relocated core materials. From the experiments performed with Zircaloy cladding (Barrand et al., 1999) it has been observed that the zirconium metal would getter all of the oxygen such that the ruthenium would remain in its metallic form. Consequently, minimal ruthenium releases would be expected during the interval before the cladding is completely oxidized since the ruthenium volatility would be the low volatility of the metal state. Since the core material relocation strongly decreases the air circulation paths, it decreases the cladding oxidation rate and thereby greatly increases the interval before the ruthenium can be oxidized and also slows the release rate of this fission product.

There is substantial evidence of the gettering capabilities for Zircaloy from the CODEX out-of-reactor experiments (Mathus et al., 1999) as well as in the fission product release experiments of Barrand et al. (1999). In the CODEX tests, a nine rod PWR type fuel bundle with UO_2 pellets was oxidized in an air environment. The information developed in the CODEX experiments is particularly relevant and meaningful since it was of a much larger scale than previous air oxidation tests, was performed with (a) full length fuel pins, (b) Zircaloy cladding and (c) the appropriate initial flow area to permit air flow through the fuel pin configuration. The thermal excursion resulting from the unstable Zircaloy oxidation was observed and was continued until the central region experienced melting. It was observed in these tests that the Zircaloy gettered all the oxygen available and there was no indication of any higher order oxides of uranium other than the uranium dioxide for the fuel pellets. For the above reasons, the experiments which have been performed without Zircaloy cladding (e.g. the unclad UO_2 experiments performed by Williamson and Beethan, 1990) represent only the potential for

further oxidation of either or both of the uranium dioxide and fission products retained in the fuel matrix in the absence of cladding or after complete oxidation of the cladding. However, these do not represent the influence of an oxidation behavior in the presence of zirconium, a very reactive metal. Furthermore, such experiments were performed at very small scale and are not capable of demonstrating the geometry changes and the consequential changes of the gaseous flow paths through the fuel pin region that were observed in the CODEX experiments and the TMI-2 accident. The scale of the experiment is essential in demonstrating this composite result of core degradation.

For the fission product release experiments reported by Barrand et al., the results are particularly relevant to evaluations of ruthenium release in the spent fuel pool. Specifically these irradiated fuel studies included the Zircaloy sheath (cladding). The authors state that:

“Rapid Cs release was delayed after the addition or ingress (e.g., test H04) of oxidizing atmosphere until oxidation of the sheath was complete.”

Two paragraphs later they add:

“The solid fraction of fission-product Ru would have been present almost entirely in the metallic state at the temperatures and $P(O_2)$ values in the HCE3 experiment, and release probably occurred by oxidation to RuO_x gaseous species. As observed previously for fragment samples, release of Ru in test H02 (and probably also in tests H01 and H03) began a significant length of time (> 2000 s) after oxidative release of Cs began. This delay was due to the competition among Zircaloy, UO_2 and Ru for oxygen. Test H02 showed the largest percentage release of Ru from any complete Zircaloy-sheathed sample tested at CRL, due to the high test temperature and the comparatively long exposure to air.”

These observations are consistent with those of the CODEX experiments which demonstrate the controlling role of unreacted Zircaloy in determining the chemical state of both the ruthenium and the uranium dioxide.

The likelihood of having degraded but unmelted fuel on top of the debris and the total mass of these fuel pellets was a subject of discussion at a November 2 ACRS meeting. As the fuel degradation occurs, the compaction of core materials could also be accompanied by debris on top of the collapsed remnants of the fuel assemblies. This debris would remain as unmelted fuel pellets without cladding as was seen in the TMI-2 accident. This phenomenon has also been observed in the PHEBUS FPT1 test (Ortega-Bernardo and Fichot, 2000). For example, the author's evaluation of their calculations for this PHEBUS test are characterized as follows:

- Gas fluid flow is strongly reduced inside the debris bed, therefore there is no convective cooling.
- Due to the disordered geometry, particles in the center of the debris bed have no radiation heat transfer with the periphery. This kind of heat transfer becomes less important. In rod like geometry, there is a significant view factor between the central fuel rods and the shroud.
- The accumulation of materials, especially fuel fragments, which causes a lower porosity yields a local power increase.

For the spent fuel pool condition that could result from the postulated initiating event, solidified debris on top of the collapsed fuel assemblies would lose energy by convection and thermal radiation to the environment. The evaluation of the potential material mass that could accumulate on top of the debris which has been stripped of its Zircaloy cladding is best estimated from the TMI-2 post accident investigations. According to Akers and McCardell (1989) the upper core debris bed was 20% of the original core material inventory. To estimate the release histories for the fission product inventory, the surface temperatures associated with the thermal radiation of this fraction of debris inventory are most relevant. Let us assume that 20% of the fuel material is in the form of degraded but unmelted fuel pellets that are collapsed into a debris bed within the area of the fuel pool. This debris bed would principally lose energy by convection through the bed and radiation to the air above. If this debris bed only radiated to the environment, this would result in a surface temperature in the range of 800-850°K. Using the

release rate information developed by Iglesias et al. for the ruthenium release rate in air at this temperature, the release rate would be less than 0.01%; a small rate of release. Consequently, the impact of this limited amount of material on the top of the debris would need to be evaluated in terms of the amount of material and the temperatures that could be achieved with the upper regions of the debris pile considering energy loss to the environment by convection and thermal radiation. While this can result in some exposed fuel, the mass of fuel involved is substantially less than the total mass of material. In fact, based on the experience in the TMI-2 accident, it is expected that this would be no more than 20% of the core material with a limited release rate, less than 0.01% per minute. This translates to a value of 0.002% per minute based on the total core inventory.

3.0 CONCLUSIONS

The conclusions for this evaluation are straightforward from the assessments performed. These can be stated as:

1. Each evaluation should have a well defined failure condition and recovery actions.
2. For the spent fuel pool there are long intervals available for recovery actions to be implemented.
3. For postulated accident conditions that preclude any recovery actions, the fuel assemblies would eventually increase in temperature sufficient for significant Zircaloy clad reaction. Under these conditions the chemical energy release would dominate the fuel bundle response which would lead to cladding melting and relocation (compaction) of the core materials. This type would be similar to those accident conditions considered for "at power" states but would occur over a somewhat longer interval.
4. The behavior under these severe accident conditions is clearly influenced by the scale of the fuel assemblies. Hence, the representation for this response must be consistent with the technical basis but must also consider the differences between very small scale tests and those which represent the compaction behavior of fuel assemblies under these postulated conditions such as the TMI-2 experience, the CODEX tests and the PHEBUS experiments such as FPT1.
5. There is a potential for some parts of the fuel material to be accumulated on top of the debris in a declad state, i.e. without Zircaloy cladding. This can be estimated from the behavior observed in the TMI-2 response. Evaluations of the fission product release from these unmelted fuel pellets must also consider the thermal radiation to the environment from the surface of this debris bed. This surface

temperature would limit the fission product releases. Preliminary estimates show that this temperature would be in the range of 800-850°K which results in a very slow release rate of ruthenium in air based upon the published experimental information.

6. Considering the composite behavior of the fuel assembly response and the potential slow release of debris from clad fuel pins on top of the debris, the release rates of ruthenium would be limited by the unreacted zirconium in the molten region and by the low surface temperatures for any overlying solidified debris. Hence, the technical basis for evaluations of the public risk resulting from these very low probability events should be consistent with these observations. Of particular note here are the observations of:

- Iglesias et al. (1990) that “Ruthenium release did not start until the sample stoichiometry was close to its equilibrium value and, hence, oxygen was available for the less thermodynamically favorable reaction of oxidizing ruthenium.”
- The CODEX experiments (Mathus et al., 1999) that “There was no evidence at all any transformation of the UO_2 to higher oxides” for the oxidation of Zircaloy clad UO_2 overheated in air. This shows that essentially all the zirconium must be oxidized for the ruthenium to begin oxidizing.
- Barrand et al. (1999) summarized the results of the HCE3 fission product release experiment from Zircaloy sheathed CANDU fuel as:

“The release rates of Kr, Xe, I and Cs were very low before complete sheath oxidation; their peak release rates in steam after complete sheath oxidation were not very temperature-dependent. Ru release was delayed

by more than 2000 s after oxidative releases of the volatile fission products.”

- The TMI-2 and PHEBUS observations that the fuel pin configuration will compact under this postulated severe accident state and there are limited flow paths for cooling and/or air ingress to support continued oxidation.

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