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AUTHOR: MARVIN RESNIKOFF

AFFILIATION: NY

ADDRESSEE: ANNETTE VIETTI-COOK

SUBJECT: FEB 20TH MEETING ON SPENT FUELPOOL RISKS

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RADIOACTIVE WASTE MANAGEMENT ASSOCIATES

February 16, 2001

U.S. Nuclear Regulatory Commission
Office of the Secretary
Washington, D.C. 20555-0001

re: Feb 20th Commission meeting on spent fuel pool risks

Dear Sir/Madam:

We wish to submit the attached comments regarding the spent fuel pool risk study to the Commissioners for their consideration at the Feb 20th meeting. These comments primarily pertain to a parallel study that the Commission has underway concerning dry cask storage.

If there are any questions, we can be reached at 212/620-0526 and at the address below. An email copy of these comments was also sent to your office today.

Thank you.

Sincerely yours,

Marvin Resnikoff
enc.

526 West 26th St.,Rm. 517 ✦ NY, NY 10001 ✦ 212-620-0526 ✦ Fax 212-620-0518 ✦ email radwaste@rwma.com

RADIOACTIVE WASTE MANAGEMENT ASSOCIATES

Comments on the "Technical Study of Spent Fuel Pool Accident Risk at Decommissioning Nuclear Power Plants, October 2000"

by
Marvin Resnikoff, Ph.D.
Radioactive Waste Management Associates
February 16, 2001

The "Technical Study of Spent Fuel Pool Accident Risk at Decommissioning Nuclear Power Plants, October 2000" provides valuable insights into the length of time that a spent fuel pool must be carefully monitored. The study should guide the Commission to better regulate decommissioning of nuclear power reactors.

But, with more utilities exhausting storage space in reactor pools, the trend is to move towards storage of spent fuel in dry casks at ISFSI's. The Commission therefore needs to produce a parallel study to the fuel pool report that assesses the safety and risk of dry cask storage. It is our understanding that the Commission is doing just that. We recommend that the issues regarding cesium release that we discuss below are taken into account in the Commission's deliberations. Similarly, with the possibility of major shipping campaigns to Skull Valley or Yucca Mountain, the Commission needs to re-examine the safety of transporting spent fuel. For the reasons discussed below, safety and environmental reports that evaluate dry storage and transportation are not conservative. For transportation accidents, the neither the Modal Study[1] nor the more recent NUREG-6672[2] correctly estimates the potential amount of cesium that may be released.

If a shipping or storage cask is opened due to a severe impact or long-duration fire, radioactive materials may be released from the spent fuel rods to the cask cavity and from the cask cavity to the external environment. We focus here on the semi-volatile radionuclide cesium. It is our contention that the amount of cesium that may be released in an accident is greatly underestimated because the amount that exists in the gap between the fuel pellet and the rod cladding, and the amount released from the gap into the cask cavity, have been greatly underestimated in the Modal Study, Sandia studies[3]

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[1] Fischer, LE *et al*, "Shipping Container Response to Severe Highway and Railway Accident Conditions," Lawrence Livermore National Laboratory, NUREG/CR-4829, February 1987.
[2] Sprung, JL *et al*, "Reexamination of Spent Fuel Shipment Risk Estimates," NUREG/CR-6672 March 2000.
[3] Wilmot, EL, "Transportation Accident Scenarios for Commercial Spent Fuel," Sandia National Laboratories, SAND80-2124, February 1981.

526 West 26th St.,Rm. 517 ✦ NY, NY 10001 ✦ 212-620-0526 ✦ Fax 212-620-0518 ✦ email radwaste@rwma.com

and NUREG/CR-6672. A major discrepancy exists in gap inventory estimates prepared for the Yucca Mountain repository project and transportation and storage studies. We urge the Commission to more carefully investigate this issue. In contrast, the percent release of cesium from spent fuel rods in a reactor spent fuel pool fire has been conservatively estimated by staff to be 100%.

All NRC estimates of the percent cesium inventory in the gap and released to the cask cavity in a severe impact or long-duration fire accident are based on flawed experimental measurements by Lorenz and co-workers at Oak Ridge[4]. According to a report by Wilmot[5], used extensively in NRC Staff Guidance and reports, a transportation accident involving impact of a dry shipping cask would lead to an extremely small release of cesium. Under an impact rupture of spent fuel rods, the Wilmot paper claims a fractional release of 2×10^{-6} of the total Cs inventory from the spent fuel rod to the cask cavity[6], the same as for all particulates such as Pu and Ru. If a severe impact is followed by a fire and oxidation of the uranium fuel, the ultimate release can be as high as 1×10^{-3} of the total Cs inventory.

For the fraction of particulates released from spent fuel rods to the cask cavity, Wilmot depends on the Lorenz work, assuming arbitrarily that impact rupture leads to releases that are ½ those due to burst rupture. Burst rupture occurs when fuel rods are overheated and burst due to internal pressure. The burst rupture releases are based on experimental studies by Lorenz and co-workers[7]. In studies that spanned a several year period, Lorenz heated gram quantities of low burn-up fuel with a resistance heater and measured cesium and other radionuclides in air. In addition, higher burnup fuel pellets (33,000 MWD/MTU) from the Robinson reactor were inserted into specially constructed 27" long zircaloy cladding fuel rods and measured in a flowing steam atmosphere. Also, holes were drilled into rod segments that were heated; radionuclide concentrations were measured in flowing air. In all cases, new zircaloy cladding or no cladding was employed in the experiment. Thus, any cesium that previously diffused out of the fuel pellets during reactor operation and plated out on the cladding or remained in the gap during reactor operation was lost. It should be patently obvious that a much greater amount of cesium will diffuse out of the pellets during three years or more of reactor operation than the brief period fuel sits in a resistance heater in the Lorenz experiments. This is a fatal error in applying the Lorenz studies to cesium releases from spent fuel.

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[4] Lorenz, RA et al, "Fission Product Release from Highly Irradiated LWR Fuel," NUREG/CR-0722, February 1980.
[5] Wilmot, 1981.
[6] Wilmot, 1981, p. 33.
[7] Lorenz, RA et al, "Fission Product Source Terms for the Light Water Reactor Loss-of-Coolant Accident," Nuclear Technology, **46**, pp. m404 – 410 (1979).

It is also important to note that the Wilmot paper arose out of a workshop convened by Sandia[8]. The participants were handpicked by Sandia; State and public interest representatives were not allowed to participate. Thus, the consensus developed at the workshop was essentially pre-arranged.

Wilmot assumes 20% of noble gases are in the gap[9], based on a paper by Acey and Voglewede[10]. Curiously, the Acey paper employs the same ANS model used by Rhyne et al, but Acey defines fission gases as noble gases only; cesium, a volatile, is assumed to have the same properties as a particulate, such as Ru and Pu.

A more recent study by Fischer and co-workers at Lawrence Livermore, the Modal Study, estimated the release of cesium vapors from the gap as 2×10^{-4} of the total cesium inventory, assuming forces greater than 100g so that 100% of the fuel rod cladding was damaged. This value 2×10^{-4} for release of cesium from the gap inventory was then assumed for all Type B casks[11]. The same value is also being used for storage casks, such as the HI-STORM 100[12]. The most recent study on the subject by JL Sprung and co-workers at Sandia, NUREG/CR-6672, estimated the release of cesium from the gap as 4.6×10^{-7} of the total cesium inventory for impact only and greater amounts for accidents involving fires, depending on the duration.

The above studies, all based on the flawed Lorenz experimental results, conflict with theoretical models and more recent measurements. A diffusion model developed by ANS, ANS-5.4, estimates 10% of Cs inventory released from fuel pellets to the gap. The noble gas release was predicted as a smaller percentage, 4% of the rod inventory[13]. The Science Applications study, a theoretical study based on the ANS-5.4 model, was ignored by Wilmot in favor of experimental results by Lorenz. Despite the flawed experimental basis, the Wilmot and more recent studies continue to be employed by NRC staff.

More recent experiments conducted at Battelle by Gray and co-workers[14] show much higher Cs levels in the gap. As a function of burnup, between 30,000 and 50,000

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[8] Wilmot, EL et al, "Report on a Workshop on Transportation Accident Scenarios Involving Spent Fuel, May 6-8, 1980," Sandia National Laboratories, SANBD80-2012, February 1981.
[9] Wilmot, p. 33.
[10] Acey, DL and JC Voglewede, "A Comparative Analaysis of LWR Fuel Designs, Appendix I," NUREG-0559, Nuclear Regulatory Commission, July 1980.
[11] Anderson, BL et al, "Containment Analysis for Type B Packages Used to Transport Various Contents," Lawrence Livermore National Laboratory, NUREG/CR-6487, November 1996.
[12] Holtec, HI-STORM 100 TSAR, Report HI-951312, July 27, 1999, Table 7.3.1.
[13] Rhyne, WR et al, "A Scoping Study of Spent Fuel Cask Transportation Accidents," NUREG/CR-0811, Science Applications, June 1979, p. 103.
[14] Gray, WJ and CN Wilson, "Spent Fuel Dissolution Studies, FY 1991 to 1994," Battelle Pacific Northwest Laboratory, PNL-10540, December 1995.

MWD/MTU, the measured cesium in the gap ranged between 0.21% and 9.91% of the cesium inventory[15]. In the Gray experiments, both the spent fuel and the cladding were measured and compared to the total Cs inventory.

The Gray experimental method was the following.

a) to measure Cs in the gap, irradiated fuel was discharged from the cladding and placed in a glass vessel along with the cladding and de-ionized water and heated. After one week, the water was analyzed for uranium and Cs, providing the Cs gap inventory.
b) to measure Cs in the grain boundaries, the fuel fragments were crushed and screened. The finer materials that passed through the screen were dissolved in HCl and analyzed for U and Cs. Since the assumption is that U and Cs inventories are congruent, the grain boundary inventories were derived by subtracting the inventory fractions of dissolved U from the inventory fraction of dissolved Cs.

Gray determined, for fuel with burnup to 50,000 MWD/MTU, that 9.9% of the Cs was in the gap inventory and 1% of the Cs was in the grain boundary. The remaining amount was locked in the fuel matrix.

While the Gray experiments at least measured the Cs in the gap inventory, we note three major problems:

a) the measured quantities are milligram amounts, with no error bars. In projecting from these experiments to casks holding 10 MT of fuel, we project up by a factor of 100 million. A small error could amount to a large amount of cesium released in a shipping cask accident.
b) the Gray experiments were done in support of the Yucca Mountain repository. Dissolving cladding plus fuel in heated water to measure Cs in the gap may simulate Cs release in a repository, but may not capture all the Cs that could be released in a shipping cask accident. Some Cs in the gap may plate out when spent fuel is cool, but may be a gas when cladding temperatures exceed 300 degrees C.
c) to determine the full amount of Cs in the gap, Gray should have also rinsed the spent fuel cladding in HCl.

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[15] Gray, WJ, "Inventories of Iodine-129 and Cesium-137 in the Gaps and Grain Boundaries of LWR Spent Fuels," Scientific Basis for Nuclear Waste Management XXII, eds Wronkiewicz, DJ and Lee, JH (1999), pp. 487 – 494.

Gray determined the range of cesium in the gap as 0.2% to 9.9% of the total cesium inventory, depending on fuel burnup. Since the full amount that plated out on the cladding was not determined by Gray, we regard his numbers as lower bounds. Nevertheless, this is the range of cesium inventory that should be employed to calculate accident consequences for both transportation and storage casks.

Radioactive Waste Management Associates

Memo

To: Marvin

From: Matt

Date: 2/16/01

Re: Notes on STACE report, Appendix IV

Appendix IV to the STACE Report[1] provides the methodology which was used to estimate the release of radionuclides from spent fuel rods in the event of rod failure. Below are some notes on this Appendix.

Section IV.1:
"at temperatures up to approximately 1000°C in an inert or chemically reducing atmosphere, only material associated with the interconnected voids of primarily the fuel-cladding gap and plena will be released from a fuel rod when its cladding integrity has been violated."

Because of this statement, Sanders, et al, conclude that only releases of materials from the void spaces are of concern (since they state that nothing else will get out) because fuel temperatures will not exceed 530°C under regulatory accident conditions. Therefore, the fractional release of cesium which is computed in the Stace report and reproduced in NUREG/CR-6672 comes entirely from that cesium in the fuel-clad gap. Earlier in the Stace Report (Table 4, pg 83) the gap inventory of cesium was listed as 1% for PWR rods and 2.1% for BWR rods.

Section IV.2:
"Although the gap inventories of the cesium isotopes, long-lived noble gases, and (possibly) tellurium isotopes appear to be identical, the way these classes of fission products are distributed differs significantly."

Noble gases are gaseous at all temperatures and are uniformly distributed throughout the interconnected void regions. Elemental cesium has a boiling point of 669°C and therefore most cesium will remain in its condensed phase at temperatures below this, with the amount of gaseous cesium determined by the temperature and equilibrium conditions.

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[1] SAND90-2406, 1992. Sanders, et al. "A Method for Determining the Spent Fuel Contribution to Transport Cask Containment Requirements." Sandia National Laboratories.

"if the fuel rod cladding lost its integrity, virtually the entire gap inventory of the noble gases and tritium would be released, regardless of the location or size of the failure or of the temperature at which cladding occurred."

"the situation is more complicated with cesium...at the instant of cladding failure, the fill and fission product gases purge the void regions of the gas-phase cesium; however...this quantity is sensitive to temperature and the volume of purge gas...after the purge gases are released from the rod so that internal and external pressures become equal, the remaining cesium associated with the interconnected void volume can exit the rod by an evaporation (or chemical decomposition) and diffusion process. Except for very long time periods or exposure of large surface areas of fuel, this diffusional release is dwarfed compared to burst release at temperatures of interest for cask safety assessment. For reasonably small cladding-failure sizes, approximately a few inches, both burst and diffusional release may be regarded as independent of failure location and size."

So, there are two release mechanisms for cesium in the fuel-clad gap. The first occurs instantly, and the second comes as a result of condensed cesium evaporating to regain equilibrium with its surroundings. The second method happens much more slowly than the first, and is negligible unless temperatures are near the cesium boiling point.

The equation used to estimate the cesium mass escaping by the first mechanism comes from the Lorenz paper in Nuclear Technology[2]:

$M_b = 3.49 V_b (M_o/A)^{0.8} \exp[-7420/T]$

V_b = volume (cm^3) of fill and fission gases vented at 0°C and at system cask pressure.

M_o = inventory (g) of cesium nuclides in the fuel-cladding gap around the fuel

A = internal area (cm^3) of cladding associated with M_o

T = temperature (K) at rupture location

M_b = total mass (g) of the particular cesium nuclide released in the burst

The numerical constants were evaluated by Lorenz using data from burst release collected over the temperature range of 700°C to 900°C. Lorenz admits that this equation is applied outside the range of validity (i.e., for lower temperatures than used to fit the equation), and states this error to be no more than a factor of 10 for lower temperatures.

Assuming a PWR rod with a cesium inventory of 2.36 g (for Cs-137), the STACE report assumes the gap inventory to be 1%, or .0236g. Using the equation above and a temperature of 300°C (573 K), the Stace Report concludes that the burst release fraction for cesium (as a fraction of the total inventory) would be 4.6×10^{-7}, or 4.6×10^{-5} of the gap inventory.

[2] Lorenz et al, 1979. "Fission Product Source Terms for the Light Water Reactor Loss-of-Coolant Accident," Nuclear Technology, 46, 404.

According to the Stace report, there would be a greater cesium release if the cladding were stripped away from part of a fuel rod (instead of the small breaches generally assumed for a failure), in which the second cesium release mechanism would be much more important.

A separate equation, also developed in the Lorenz paper, is used to model the diffusional release of cesium (the second mechanism discussed above). I will not discuss that here.

Section IV.3
This section is entitled "escape of nonvolatile species," and discusses cesium release in the form of fuel fines. From this it appears that cesium may be released into the environment as a gas or a fuel fine.

From experimental tests conducted by (who else) Lorenz[3,4] (6 tests on PWR fuel, 2 on BWR fuel) concluded that "ejected fuel fines contained 5 to 7 times more cesium isotopes than the bulk fuel." The STACE report says that this reflects the much higher cesium gap inventory of the BWR fuel employed in the tests (12%) compared with that of the PWR fuel (0.3%). For this reason, the STACE report claims that "if Cs-134 and Cs-137 gap inventories are greater than 1% of their total inventories, concentrations in the fines should be enhanced relative to their respective concentrations in the bulk fuel by a factor equal to the gap inventory, expressed as a percentage. For example, for a fuel with a Cs-137 gap inventory that is 7% of its total inventory,"

$C_{Cs\text{-}137}(\text{fines})/C_{Cs\text{-}137}(\text{bulk}) = 7$

Table IV-2 compares, among other things, the amount of cesium released as a gas and the amount released as a fuel fine. This report assumes that a cladding breach will result in the release of .0003% of the total fuel amount of a rod when the cladding fails. For a PWR rod irradiated to 38.2MWD/Kg U after a 5-year decay period, a cladding breach at 530°C is calculated to result in a release of 6.94×10^{-3} Ci of Cs-137 as a fuel fine, and 4.32×10^{-3}Ci of Cs-137 as a gas. It is also assumed that 10% of the fuel fines will remain airborne for any significant period, so the "effective release" from a fuel rod can be stated as 6.94×10^{-4} Ci of Cs-137 as an airborne fuel fine, and 4.32×10^{-3} Ci of Cs-137 in gaseous form.

Conclusions, Cesium release from Fuel Rod
First, it appears that the low release fraction for cesium from a breached rod (4.6×10^{-7} of the total inventory is released as a gas, less as an airborne fuel fine) is only the release fraction for cesium to the cask cavity. The STACE report concludes that the burst release mechanism of gaseous release is essentially independent of the failure size or location on the rod, meaning that a larger breach would not result in a greater release caused by this mechanism. A larger breach would, however, increase the

[3] NUREG/CR-0722, 1980. Lorenz et al. "Fission Product Release from Highly Irradiated LWR Fuel." Oak Ridge National Laboratory.
[4] NUREG/CR-1773, 1981. Lorenz et al. "Fission Product Release from BWR fuel under LOCA Conditions." Oak Ridge National Laboratory

amount of cesium released by the diffusional release method, owing to the greater surface area available for transfer of cesium to the gaseous phase (the same reason why water evaporates faster if spread out thinly).

NUREG/CR-6672 then assumes that all of the gases released from a rod to the cask interior via the burst rupture mechanism can escape into the environment if there is a pathway.

"deposition processes are assumed to be effective for materials released to the cask interior when rods are failed by impact but is neglected when rods fail by burst rupture. Deposition is neglected following burst rupture because the combustion gases that are assumed to be flowing through the cask during Category 6 accidents are also assumed to carry all materials released to the cask interior out to the environment without significant depletion by deposition to cask interior surfaces." (NUREG/CR-6672, pp. 7-26)

NUREG/CR-6672 also states, again referencing the Lorenz reports, that cesium release during a long duration hydrocarbon fire will be about 3 times higher (due to the increased importance of the diffusion mechanism) than a burst rupture-only release.