Maryland Safe Energy Coalition

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September 19, 1993

Robert M. Bernero, Director
Office of Material Safety and Safeguards
U.S. Nuclear Regulatory Commission
Washington, D.C. 20555

Re: Docket No. 72-8 (50-317/318)

Dear Mr. Bernero:

Pursuant to your denial of our petition of December 21, 1992, dated August 16, 1993, I wonder if you would be so kind as to clarify a few points in the NRC's decision.

Some terminology is used which we do not understand. For instance, on page 7 bottom, you refer to "a hypothetical bounding case accident." What is a "bounding case accident"?

Also, on page 7 and 8, the staff "concluded that the radiation dose to the public is negligible" in a "worst-case DSC leakage accident". Could you please tell us what the staff considers a worst case (e.g., a plane crash into the ISFSI?) and exactly what amount of radiation would be released?

On page 8, it states that our letter of July 27, 1993, is currently under review by the NRC staff. Why did you deny our petition before this letter was reviewed? This inverted procedure does not even have the appearance of objectivity. Does this review have the same docket number as the denied petition of December 21, 1992, a new docket number or no docket number?

When the staff reviews that letter, we would be most interested in knowing exactly what the blast impact, concussion or shock wave would be upon the Calvert Cliffs power plant itself (in precise units of measurement) if 1.5 million barrels of liquified natural gas exploded at Cove Point. We want numbers please.

On page 10, it states "The surface dose rates at the air inlets and outlets locations are less representative than dose rates at the HSM walls and door for assessing the effect on the direct radiation levels to individuals located beyond the controlled area." How would the staff know if the radiation passing through the walls and door is greater than the radiation in a plume emanating from the vents unless the vents were monitored?

On page 11, it states "Because the DSC is made of stainless steel, it is not subject to...embrittlement." Is it not true that stainless steel is more brittle than mild steel and that it can become more brittle by neutron bombardment?

While acknowledging that we submitted 50 questions on February 10, 1993, the NRC never answered most of these questions. Instead, Charles J. Haughney of the Division of Industrial and Medical Safety sent us a list of 14 documents on April 30, 1993, purporting to contain information relating to the 50 questions. He concluded saying "your questions will be considered by the NRC staff in connection with the decision on your petition." We are disappointed the NRC did not answer most of these questions.

Many of these questions were coincidentally studied by the Center for Nuclear Waste Regulatory Analysis for the NRC (Contract NRC-02-88-005) in a paper entitled: "Characteristics of Spent Nuclear Fuel and Cladding Relevant to High-Level Waste Source Term", May 1993, San Antonio, Texas. The conclusions of that study seem to be at odds with those of your staff. I am enclosing a copy of this study for your convenience.

For instance, on page 4-11 of this study, the authors state: "The dry environment has the potential of producing such problems as further fuel cladding oxidation, increased cladding stresses and creep deformation as a result of rod internal pressure, and volume expansion of fuel due to air permeating through any pinholes and incipient cracks in the cladding. These possible spent fuel and cladding alteration modes could be quite accelerated under dry storage conditions, since the temperatures are much higher than in wet storage."

The study estimates that 5,000 fuel rods will have become breached at the time of dry storage and concludes that interactive pellet/cladding/container corrosion is "inadequately addressed in current source-term models."

It is unfortunate that this study was not brought to my attention before the petition of December 21, 1992, was denied. If it was, I would have submitted it as part of the docket record. However, your staff should have been aware of this study. Since it obviously is new information relevant to our original petition, I believe it constitutes grounds to reopen the case for re-examination. Please acknowledge this request.

In any event, we would like to know what the NRC staff thinks of the validity of this study.

Your consideration of all the above concerns and answers to our questions would be most appreciated.

Sincerely yours,

Richard Ochs

Richard Ochs
Director, MSEC

CHARACTERISTICS OF SPENT NUCLEAR FUEL AND CLADDING RELEVANT TO HIGH-LEVEL WASTE SOURCE TERM

ABSTRACT

This report, based on literature study, describes characteristics of light water reactor (LWR) fuel assemblies for boiling water reactors (BWR) and pressurized water reactors (PWR) and the changes that take place in both cladding and uranium dioxide fuel during service in commercial power reactors. This information is provided as a background for the evaluation of important factors related to fuel stability under geologic repository conditions. Data related to discharged fuel storage (both wet and dry) are also provided, along with the condition of the fuel in terms of damaged and leaking fuel assemblies. The degradation of spent fuel and cladding while in service in the reactor and likely degradation in a geologic repository are discussed in terms of cladding oxidation and corrosion, and fuel-pellet cracking, fuel restructuring, microstructure and fission product mobility, inventory and distribution of fission products, fuel pellet rim effect, and fission gas release and pressure increase. The range of attributes of discharged fuel will have an impact on the releases of radionuclides from the engineered barrier system (EBS) and, as a result, on the compliance with the regulations relating to the gradual release from a repository over a period of 10,000 years. A suggestion has been made to characterize a wider variety of spent LWR fuels, including high burnup fuels and fuels with characteristics outside the range of those currently awaiting geologic disposal, with the intent of including the full range of spent fuel characteristics and performance in developing source term models for performance assessment. A significant portion of this report deals with review of studies on oxidation and release of radionuclides from spent fuels. Although the emphasis is on an oxidizing environment, the likely behavior of spent fuel under a reducing environment has also been discussed. The parameters important for assessment of spent fuel behavior in a geologic repository include oxidation state of the fuel at the time of contact with water, geochemistry of the repository (chemical composition of the leachant), water flow conditions, and synergistic effects due to modification of the geochemical environment as a result of releases from vitrified waste form and corrosion products from the waste package. A number of areas requiring additional experimental data have been identified. Issues related to modeling of source term for use in performance assessments have also been discussed.

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developed to give the activity as a function of time after discharge (Locke, 1975). The calculations of the code have been validated against experimental data on spent fuel radionuclide inventories (Heeb et al., 1990).

4.3 CHARACTERISTICS OF SPENT FUEL

Irradiated fuel still contains most of its original ^{238}U, about one-third of its original ^{235}U, almost all the fission products, all the transuranic (TRU) isotopes, and many activation products. Tables 4-7 and 4-8 summarize the most important isotopic features of typical LWR spent fuels. The volume of wastes generated during the service life of LWRs is given in Table 4-9.

4.4 STORAGE OF SPENT LWR FUEL

Prior to permanent disposal in a repository, the spent fuel is expected to be stored for some years (perhaps 10 years or more) to allow the assemblies to cool down as a result of dissipation of the decay heat. Two principle methods are available for storing the spent fuel for extended periods of time, namely, wet storage in water pools and dry storage in air or gases (EPRI, 1986; Bailey et al., 1986; EPRI, 1984; Johnson, 1979). Water pool storage has been used in the U.S. since the first reactors were built in 1943 (Johnson, 1977). However, experience with dry storage, an alternative to extended wet storage, is rather limited. The fuel may be stored unconsolidated or consolidated. By definition, unconsolidated fuel means intact assemblies; consolidated means the fuel rods are removed from the fuel assembly and placed in a grid with closer spacing than that of an intact assembly, or the rods are placed in a close-packed array inside a canister (Zacha, 1988; Johnson, 1986; EPRI, 1989). Volume savings of 2:1 by consolidation have been demonstrated by many utilities (Zachy, 1988). The advantages of fuel rod consolidation are obvious: (i) storage capacity would be almost doubled, and (ii) the number of spent-fuel shipping casks can be halved.

In the wet storage mode, the decay heat from the spent fuel is removed by deionized water (DIW) at a temperature below 40°C. Zircaloy cladding does not experience any significant additional corrosion or hydriding under such conditions compared to that experienced while in core. In the dry storage mode, the decay heat is removed by using air or an inert gas (usually helium or nitrogen) under forced convection. The dry storage facility is simpler and cheaper to maintain compared to wet storage. On the other hand, the dry environment has the potential of producing such problems as further fuel cladding oxidation, increased cladding stresses and creep deformation as a result of rod internal pressure, and volume expansion of the fuel due to air permeating through any pinholes and incipient cracks in the cladding. These possible spent fuel and cladding alteration modes could be quite accelerated under dry storage conditions, since the temperatures are much higher than in wet storage. Temperatures in the range of 300 to 400°C are being considered for the extended dry storage of spent fuel for periods up to 100 years following discharge from the reactor. Because of the stated reasons, any extended dry storage may require evaluation of any additional process of materials degradation or alteration of the cladding properties which may influence its subsequent behavior in a repository. Long-term behavior of extended wet stored spent fuel may need to be modeled differently from assemblies that experience dry storage following discharge from the reactor.

Allowable long-term storage temperatures and times for dry storage of spent LWR fuels will depend upon a number of factors including cladding stress levels, fuel type and assembly design, materials condition of the cladding at the time of discharge, decay heat history of the spent fuel, and heat

6 DISCUSSION OF SPENT FUEL LONG-TERM PERFORMANCE ISSUES

Source-term models are expected to provide estimates of the release of radionuclides from the waste packages under geologic repository conditions. Estimates are required for conditions that are likely to prevail for the 10,000 years of regulatory concern and for additional scenarios which could be anticipated. The reliability of the source-term model will depend on how appropriately the understanding of the quantities and characteristics of the spent fuel, waste package materials, repository environment (such as geochemistry, temperature, radiolysis products), and temporal changes in all these aspects over the 10,000 years after the repository closure are included in the model. Uncertainties in spent-fuel behavior could undoubtedly influence the evaluation (or assessment) of performance in the repository. The output of the source-term modeling exercise can be simply stated as the expression of the 'release rates and quantities' of the radionuclides from the spent fuel as a function of time over 10,000 years. The calculated releases will be compared with the allowable releases per 10 CFR 60 to judge if the repository design will provide the required level of isolation of the spent fuel. There is little uncertainty in the total inventory of the radionuclides in the spent fuel, and the task at hand would be to model release rates and cumulative releases during the 10,000-year period. A similar argument would apply to the case of vitrified (glass) wasteforms.

Based on the discussion of the inventories of the radionuclides in the cladding and spent fuel, the characteristics of the fuel and cladding (i.e., inhomogeneous distribution of the radionuclides), the oxidation behavior of the fuel, and its leaching response when in contact with moist and aqueous environments, it can be concluded that releases of radioactivity in a geologic repository will be greatly influenced by the processes considered. For an unsaturated repository, there could be extended periods of time during which the fuel may not come in contact with water, but the characteristics of the fuel may change due to oxidation in air, leading to increased surface area and release of radioactive gases in the fuel matrix. Subsequent exposure of this 'altered' spent fuel to aqueous leaching would be quite different from the event in which solid UO_2 in the pellet form comes in contact with water. The time at which the waste package fails is also critical, as it will have a bearing on the type and the amounts of radiolysis products present in the groundwaters that will contact spent fuel. Radiolysis products have a direct influence on the pH of the water, which is one of the major parameters that influence spent fuel dissolution characteristics. At the present time, only a few source-term models exist. Although a detailed review of the individual models is outside the scope of this report, a survey of the general capabilities and limitations approaches used for the development of the current source-term models indicate that some significant conclusions drawn from experimental data and analyses of the spent fuel and cladding are absent or inadequately addressed in current source-term models. Some performance factors that could modify or increase the calculated performance of the spent fuel over 10,000 years, are listed below. The literature provides a reasonable amount of data and technical information in these area, except where noted, for their inclusion in current models.

- Processes over 10,000 years of repository performance. Source-term models need to include the degradation behavior of the cladding and spent fuel during the period of exposure to high temperature (above the boiling point of water). Extended exposure at such temperatures could substantially alter the fuel characteristics, which will influence the release of radionuclides upon subsequent exposure to liquid phase groundwater. The degradation of fuel, which may be called 'alteration', will include oxidation of the fuel to higher oxidation states, substantial increase in the surface area, which will lead to increases in the calculated

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release rates per unit volume of fuel, and rapid release of fission gases normally trapped in the spent UO_2 matrix.

- Use of data related to leaching behavior of spent fuel with 'altered' characteristics in modeling releases. The majority of the literature reported data are on release of radionuclides from spent UO_2 fuel rather than fuel with higher oxygen to uranium ratios, such as U_3O_8. The models should also address the scenario where a mix of unaltered spent fuel, semi-altered (quasi-equilibrium state discussed in the report), and fully altered spent fuel are releasing radionuclides in the near-field of the repository.

- Data related to leaching of spent fuel in repository specific geochemistry should be incorporated in the mechanisms which are incorporated in the models. The geochemistry should incorporate the effects of radiolysis products, leading to changes in the water pH, redox conditions, and presence of waste package corrosion products. Since the heat output of the containers with vitrified wastes is lower than containers with spent fuel, it is conceivable that these containers will corrode/breach before spent fuel containers because contact with liquid water will occur at shorter times. Under such a scenario, the geochemistry of the groundwaters will also be modified by the releases from the vitrified high-level wastes. The large number of containers with vitrified wastes (about one-third of the anticipated 40,000 to 50,000 containers) indicates that the groundwaters could contain a substantial amount of radionuclide releases from vitrified wasteforms.

- Although most source-term models acknowledge the radioactivity that might be released in the form of gases, this aspect has generally been addressed by ^{14}C releases from the crud/corrosion products on the cladding and the spent fuel. This approach will result in underestimation of the contribution of the gaseous releases from the cladding and fuel, as less than 1 percent of the wall thickness of the cladding is oxidized. An extended period of exposure of the cladding to high temperature can oxidize the cladding leading to additional release. Similarly, as stated in an earlier item, further oxidation of spent fuel in the repository will break up the fuel pellets and generate particulates leading to release of gaseous and volatile fission products to the near-field environment. Although the volume of the cladding is estimated to be less than 5 percent of the fuel, it contains approximately 50 percent of ^{14}C (the other 50 percent being in the fuel).

- Currently it is estimated that at least 1 out of every 10,000 fuel rods will be in the breached condition at the time of disposal. Although releases of radionuclides from fuel assemblies with such breached rods can be detected while in service in-reactor, individual rods are difficult to identify or remove economically. As such, it is likely that they will be disposed of along with the unbreached rods. Their distribution among the different waste packages can be known with certainty. Based on the number of containers likely to be emplaced in the first repository, one can assume 4,000 to 5,000 breached fuel rods. The radionuclide release characteristics of these rods will be different from the unbreached rods under most repository scenarios. The contribution of the breached rods to the source term will be considerable, and needs to be accounted for. Current models assume no breached rods.

- According to the SCP design, it is estimated that there will be approximately 1,000 fuel rods per waste package. The DOE in assuming 'no-credit' for the cladding assumes that upon failure of a given waste package, fuel contained in all 1,000 rods will be exposed to the

repository environment. This may neither be a realistic situation nor even a conservative one. For instance, if the failure of the waste package is due to flooding of the repository leading to corrosion of the metallic container, we are assuming that spent UO_2 fuel contained in approximately 1,000 rods inside the waste package is exposed simultaneously to groundwater. Under such a scenario, the source-term model will use leaching mechanisms and experimental data related to leaching of 'unaltered' spent fuel to calculate release of radionuclides. This scenario is inconsistent with a distribution of failure of the cladding with time, which would lead to alteration of the spent fuel, e.g., substantially increased surface area per unit volume, different release mechanism, rapid release of fission gases, etc. Therefore, the source-term models need to incorporate a distribution of cladding failure as a function of time.

- Most current source-term models are based on the assumption that maximum release of a particular radionuclide in the leachate (groundwater) will be limited (controlled) by the solubility limit of that radionuclide. This assumption would be valid provided 100 percent of the release of all radionuclides were in solution in the aqueous phase. There is evidence that such is not always the case. A number of elements are likely to occur in the form of colloids and precipitates in a geologic repository. Since the precipitates and colloids are not in solution, they provide an add-on term to the source-term models. Although, the fraction of releases of actinides known to form of colloids and precipitates in not known precisely at present, the assumption used in some elementary treatments in source-term models of a maximum of 30 percent could be a gross underestimation.

- The presence of organic material and microbes in water is known to influence the migration of radionuclides in underground environments. Microbial activities expected in an unsaturated repository may provide a significant add-on term to the source-term modeling and needs to be investigated. Since the studies related to microbial activities and their influence on the alteration of spent-fuel characteristics are practically non-existent, additional experimental investigations in this area are warranted to estimate the contribution of the potential microbial activities to the source-term.

- The EBS components are expected to be constructed from a number of materials. Most studies related to the failure of the waste package concentrated on the corrosion failure of the metallic container as a result of groundwater/container interaction, and do not address electrochemical effects between the fuel and the cladding and the fuel and the container material. Insufficient consideration of potential galvanic effects in selecting waste package materials could adversely affect the fuel cladding failure distribution as a function of time and could also accelerate the spent fuel degradation/dissolution. Therefore, electrochemical interactions between the waste package materials, spent fuel cladding, and the spent fuel need to be evaluated and possibly incorporated in the source-term models.

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