October 18, 1996

MEMORANDUM TO: Richard A. Weller, Section Leader
Engineering and Material Section
NMSS/DWM/ENGB

FROM: Tae M. Ahn, Materials Engineer /S/
Engineering and Material Section
NMSS/DWM/ENGB

SUBJECT: PROCEEDING FOR SYMPOSIUM ON THE SAFETY OF RADIOACTIVE WASTE MANAGEMENT (SSRWM '96), KOREA

Attached is the proceeding to be presented at SSRWM '96. The title of the presentation is "SPENT FUEL BEHAVIOR IN THE YUCCA MOUNTAIN REPOSITORY." The justification for this trip (memo to J. Blaha from C. Paperiello) is also attached. As shown in the justification, one mission would include NRC's cooperative support of and assistance to Korea's nuclear safety program; therefore, I would like to present this proceeding in Korean.

Attachments: As stated

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Attachment 1

Attachment 2

UNITED STATES
NUCLEAR REGULATORY COMMISSION
WASHINGTON, D.C. 20555-0001

MEMORANDUM TO: James L. Blaha
Assistant for Operations
Office of the Executive Director
for Operations

FROM: Carl J. Paperiello, Director
Office of Nuclear Material Safety
and Safeguards

SUBJECT: FOREIGN TRAVEL FOR TAE M. AHN TO TAEJON, REPUBLIC OF KOREA

Attached is a request for approval of foreign travel for Tae M. Ahn, Materials Engineer, in the Division of Waste Management. Mr. Ahn has been invited to present a keynote address on spent fuel behavior associated with the disposal of high-level waste in the U.S. at the Symposium on the Safety of Radioactive Waste Management (SSRWM '96), which will take place in Taejon, Republic of Korea (ROK), November 4-5, 1996.

The symposium is being organized by the Korea Institute of Nuclear Safety, the technical expert agency which carries out all of the day-to-day regulatory responsibilities of the ROK Ministry of Science and Technology (MOST), the national regulator. The symposium will focus on the primary safety issues of radioactive waste management in relation to the Korean program of nuclear waste management. Mr. Ahn's participation would allow us to share our information and update our knowledge of what is being done internationally, as well as in the ROK to address the problem of nuclear waste management. It would also further the U.S. Nuclear Regulatory Commission's cooperative support of and assistance to the ROK's nuclear safety program, which the Office of International Programs advises is carried out under the auspices of both the NRC-MOST information exchange and cooperation arrangement and the US-ROK Joint Standing Committee on Nuclear and Other Energy Technologies.

The symposium organizers have offered to cover Mr. Ahn's transportation and per diem expenses. Because NRC will benefit as well as the symposium organizers, I support the request. However, the invitation was not received until September 9, therefore, the travel was not anticipated in the Office of Nuclear Material Safety and Safeguards' foreign travel projection for the first quarter of FY 97. I recommend NRC approval.

Attachments: Forms 445/279

SPENT FUEL BEHAVIOR IN THE YUCCA MOUNTAIN REPOSITORY

Tae M. Ahn
U. S. Nuclear Regulatory Commission
Washington, DC 20555-0001, USA

ABSTRACT

The U.S. high-level waste (HLW) program is introduced, emphasizing permanent geologic disposal. Detailed site characterization, total system performance assessments, and designs are the bases for implementing the disposal program of HLW. Various technical issues associated with site characterization are addressed. Detailed failure modes associated with spent fuel and its packaging materials are highlighted. The importance of international cooperation is emphasized to utilize limited resources effectively.

BACKGROUND

The Nuclear Waste Policy Act [NWPA, 1983] and the Nuclear Waste Policy Amendments Act [NWPAA, 1987] enact the safe transportation, interim storage, and permanent disposal of high-level nuclear waste (HLW) in the U.S. The U.S. Nuclear Regulatory Commission licenses and regulates the safe transportation, interim storage, and permanent disposal of HLW. The HLW in the U.S. includes: (1) spent nuclear fuel from commercial nuclear power reactors; (2) vitrified HLW from Liquid HLW stored at defense and commercial reprocessing sites; and (3) others such as enriched spent nuclear fuel from research reactor, surplus plutonium, and depleted uranium. The NRC regulatory requirements for the HLW management have been developed for packaging and transportation with 10 CFR Part 71 [10 CFR Part 71], for storage at a Monitored Retrievable Storage with 10 CFR Part 72 [10 CFR Part 72], and for permanent geologic disposal with 10 CFR Part 60 [10 CFR Part 60].

The U.S. Department of Energy (DOE) is implementing the safe disposal of HLW. DOE's disposal implementation is under the licensing authority of NRC and the applicable environmental standards of the U.S. Environmental Protection Agency. All affected parties such as States are involved in the implementing and licensing processes. Previously, DOE had considered three candidate sites for the geologic disposal of HLW. They were Salt, Basalt, and Tuff repositories. Currently, the tufaceous Yucca Mountain (YM) is the proposed repository site in the U.S. It is planned that the repository will be located in a horizon above the water table. Among the advantages of locating a repository in this so-called unsaturated zone, is less severe corrosion of waste packages because of the small amounts of water.

The site characterization of the proposed YM site determines the suitability of the YM as the permanent disposal site of HLW. 10 CFR Part 60 mandates the total system requirement and subsystem requirements for radionuclide behavior in geological times. Recently, the technical bases for the current HLW standards are being reviewed for a dose-based amendment. To comply with this rule, geology, hydrology and engineering have been studied. Site Characterization Plan [DOE] describes the detailed geology, hydrology, engineering, and other related activities. The total system performance assessment (TSPA) [examples: NRC, 1995; TRW Environmental Safety Systems, 1995] evaluates numerically the performance of the YM repository. The subsystem performance assessment [example: Mohanty et al., 1996] is used for the compliance assessment of subsystems and for modules for TSPA. The TSPA is based on engineering designs [TRW Environmental Safety Systems, 1994] of waste packages and geotechnical underground facilities. The TSPA and design activities are iterative each other or iterative in themselves as characterizations progress.

Whereas designs are taking the defense-in-depth approach, the TSPA is a probabilistic assessment based on the principle that risk is probability times consequence. Currently, NRC considers following issues in the risk assessment: (1) igneous activity; (2) structural deformation and seismicity; (3) unsaturated and saturated flow under isothermal conditions; (4) thermal effects on flow; (5) evolution of the near-field environment; (6) container life and source term; (7) radionuclide transport; and (8) repository design and thermal-mechanical effects. NRC is implementing license application review plan for HLW [NRC, 1994]. This presentation focuses on the spent fuel behavior in waste packages under anticipated repository conditions for the assessment of container life and source term.

CONTAINER FAILURE

The container confines radionuclides within the container until the container is breached. The current DOE repository concept proposes two concentric containers (overpacks) surrounding the multi-purpose canister or the uncanistered spent fuel [TRW Environmental Safety Systems, 1994]. The primary candidate material for the inner container is alloy 825, which is a Ni-based and highly corrosion-resistant alloy. The outer container will be made from a corrosion-allowance material. Corrosion-allowance materials are thick-walled, and are expected to corrode slowly and predictably over time in aqueous environments. DOE selected three candidate materials for the outer container: (1) a wrought C-Mn steel, A516 Grade 55; (2) a centrifugally cast C steel, similar in chemical composition to A516 Grade 55 steel; and (3) a low-alloy steel, A387 Grade 22.

The container performance for the initial several thousand years is extremely important in the current DOE strategy for waste containment and isolation for the proposed YM repository. There are several hypotheses that could demonstrate that radioactive waste can be isolated for long periods of time. One of them is that environments near the container could be dry, to prevent aqueous corrosion of the container. The heat from the radioactivity decay will cause low relative humidity and thus prevent an aqueous film from forming on container surfaces. The surface temperature of container ranges from 300 to 150 C over a period of 1,000 years. Nevertheless, aqueous environments are expected to be encountered on waste packages by fracture flow of groundwater in host rocks or by large relative humidity. Currently, an RH of about 70 percent is considered necessary for the onset of aqueous corrosion.

Under aqueous conditions, thick corrosion-allowance materials will corrode over an extended period. Once the first pit penetrates through the wall of the outer container, the inner corrosion-resistant container will be cathodically protected over another extended period. After the outer container is completely consumed, the inner corrosion-resistant container and the canister (multi-purpose or glass pour canister, mainly austenitic stainless steels) will contain radionuclides for another extended period.

NRC considers many failure modes during the container/canister performance. Although containers have been considered immune to dry environments, material stability and dry oxidation are potential failure modes in dry environments. The outer container may be susceptible to phosphorous segregation along grain boundaries during the long-term performance under repository conditions. This segregation will decrease the fracture toughness, leading to an earlier mechanical failure of container. The inner container/canister may be sensitized by forming secondary phases along grain boundaries. This sensitization will also lead to easy corrosion failure of the container. Localized dry oxidation along grain boundaries is another potential failure mode under dry conditions.

Under aqueous conditions, the groundwater chemistry may vary from mild tufaceous solution to concentrated solutions by wet-dry cycles of groundwater on waste packages. Corrosion is the major failure mode in aqueous environments. Corrosion is, in turn, divided into uniform corrosion, localized corrosion (pitting and crevice corrosion), stress corrosion cracking, hydrogen embrittlement, galvanic corrosion, and microbial corrosion. The major problem associated with corrosion is that the kinetics is not linear with time. Often, there are long initiation times and short propagation times. This makes the prediction of corrosion kinetics very difficult.

RADIONUCLIDE RELEASE

The inventory of U.S. HLW is composed mainly of approximately 63,000 MTHM spent fuel of pressurized-water reactor and boiling-water reactor and 7,000 MTHM of HLW glass [TRW Environmental Safety Systems, 1995]. The radionuclide activity is approximately 97 percent spent fuel and approximately 3 percent HLW glass. Therefore, this presentation emphasizes the spent fuel behavior.

During the initial dry and high temperature period of repository, most spent fuel is likely to be contained in the container. However, some containers may fail during this period, caused by initial fabrication defects or by external mechanical forces. Therefore, the spent fuel behavior needs to be considered in both dry and aqueous environments.

The radionuclides of concern are long-lived ones: fission products such as Tc-99, I-129, and Cs-135; activated products such as C-14 and Cl-36; and actinides such as Pu-(239+240), Am-(241+243) and Np-237. Abundant fission products, Cs-137 and Sr-90, are short-lived and expected to decay during the containment period.

There will be two types of release of these radionuclides. The first one is prompt release of radionuclides from gaps and grain boundaries. This prompt release will take place instantaneously once the container is breached. Approximately one to two percent of radionuclides are expected to be released in this way. Most of radionuclides will be released slowly over time mainly as the spent fuel matrix is degraded.

In dry environments, C-14 is expected to be released by solid state diffusion in the unoxidized spent fuel. As the spent fuel matrix oxidizes, C-14 is expected to be released rapidly. There are two steps of spent fuel oxidation. UO_2 will oxidize to U_4O_9 and eventually become U_3O_8. Once U_3O_8 forms, Cl-36, Tc-99, I-129 and Cs-135 are also potentially diffused out in the matrix. Also, as U_3O_8 forms, the spent fuel matrix will become powered by a large volume expansion. The large surface area associated with powering will increase the radionuclide release significantly. These radionuclides will not be expected to be retarded in hostrocks by sorption.

In aqueous environments, there are two groups of radionuclides in release. The first group consists of fission products and activated products. These radionuclides are released as the spent fuel matrix dissolves. Under near-static conditions, they do not reach solubility limits and do not reprecipitate on the spent fuel surfaces. Therefore, the radionuclide releases are governed by the dissolution rate of the spent fuel matrix. These radionuclides are not expected to be retarded in hostrocks by sorption.

The second group consists of actinides. These actinides are expected to reach solubility limits and reprecipitate on the spent fuel surfaces. Therefore, radionuclide releases are governed by the solubility limits and groundwater flow. Whereas Np-237 is not expected to be retarded in hostrocks, Pu-(239+240) and Am-(241+243) have been considered to be retarded significantly. However, recently it is believed that Pu-(239+240) and Am-(241+243) form colloids. The formed colloids are unlikely to be retarded in hostrocks. If fractures of hostrocks are responsible for radionuclide transport, both ionic or colloidal Pu-(239+240) and Am-(241+243) are potentially released without retardation.

Regarding cladding, the majority is Zircaloy-2 or Zircaloy-4. C-14 is present in cladding and expected to be released during oxidation. Cladding is credited to contain radionuclides in spent fuel matrices. Failure modes of cladding include: uniform oxidation; creep and rupture; hydride-reorientation leading to brittle fracture; and localized corrosion such as pitting.

Whereas container is primarily responsible for radionuclide containment, the source term for radionuclide release is mainly governed by spent fuel degradation. The overall source term, therefore, evaluates container and spent fuel. Additional components, responsible for the overall source term, are also emplaced. There may be backfill of crushed tufaceous rocks surrounding containers. Also, HLW glass will be emplaced. When HLW glass dissolves faster, its contribution to radionuclide release will become important. The radionuclide behavior during HLW glass degradation may be grouped as in spent fuel dissolution.

Lastly, criticality should be prevented during the performance of spent fuel in the repository. A few critical times are being considered for the criticality assessment. Examples are, water intrusion in the container that is breached by pitting corrosion, or geometry changes of spent fuel during the spent fuel degradation.

CONCLUDING REMARKS

This presentation has introduced the U.S. HLW programs, emphasizing permanent geologic disposal. Detailed characterization, performance assessments, and designs are the bases for implementing the disposal program of HLW. Among various technical issues, failure modes associated with spent fuel and its packaging materials are highlighted. The HLW problem is an unprecedented difficult task, mainly because of long geologic compliance time. Therefore, it is important to cooperate internationally to utilize limited resources effectively.

REFERENCES

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Code of Federal Regulations,"Packaging and Transportation of Radioactive Material," Part 71, Chapter I, Title 10, "Energy."

Code of Federal Regulations,"Licensing Requirements for the Independent Storage of Spent Nuclear Fuel and High-Level Radioactive Waste," Part 72, Chapter I, Title 10, "Energy."

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