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LIFE CYCLE MANAGEMENT OF RADIOACTIVE MATERIALS PACKAGING

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ABSTRACT

The objective of life cycle management of radioactive materials packaging is to ensure the safety functions (i.e., containment of radioactivity, protection against radiation, and criticality safety for fissile contents) during the entire life cycle of the packaging in storage, transportation, and disposal. A framework has been developed for life cycle management regarding Type B radioactive and fissile materials packaging, drawing upon current U.S. Department of Energy (DOE) storage standards and examples from interim storage of Pu-bearing materials in Model 9975 transportation packagings. Key issues highlighted during long-term storage of Pu-bearing materials included gas generation and stability of PuO_{2+x}; other operation safety issues highlighted for interim storage of Model 9975 transportation packagings included the need to consider a facility design-basis fire event and the long-term behavior of packaging components such as Celotex and elastomeric O-ring seals. The principles of aging management are described, and the key attributes and examples of effective aging management programs are provided based on the guidance documents for license renewal of nuclear power plants. The Packaging Certification Program of DOE Environmental Management, Office of Safety Management and Operations, plans to expand its mission into packaging certification for storage and aging management, as well as application of advanced technology, such as radiofrequency identification, for life cycle management of radioactive materials packagings.

INTRODUCTION

Currently, thousands of Model 9975 transportation packagings have been certified by the U.S. Department of Energy (DOE) for shipment of Type B radioactive and fissile materials in accordance with Part 71, Title 10 Code of Federal Regulations (CFR), or 10 CFR 71, *Packaging and Transportation of Radioactive Material*. When these transportation packages are not in transit, they are in de-facto interim storage. Tens of thousands of U.S. Department of Transportation (DOT) Specification 6M transportation packagings are also being used for interim storage of radioactive materials at DOE facilities. The use of the 6M transportation packagings will be phased out by October 1, 2008. Three possible replacement packagings for 6M are the ES-3100 packaging designed by the Y-12, and the Model 9977 and Model 9978 packagings designed by Savannah River National Laboratory. The ES-3100 packaging received certification from the U.S. Nuclear Regulatory Commission (NRC) in April 2006; the Model 9977 packaging received certification from DOE in October 2007. Both ES-3100 and Model 9977 are certified as a transportation packaging in accordance with 10 CFR 71. (Model 9978 is currently under DOE certification review.) Prolonged interim storage of Type-B radioactive and fissile materials in certified transportation packagings, if not properly managed, can raise safety and security concerns that directly impact operations and economics.

This paper will describe a framework of life cycle management (LCM) strategies for the Type-B radioactive and fissile material transportation packagings that are also used for interim storage. The objective of the LCM is to ensure the safety functions of the radioactive materials packaging (i.e., containment of radioactivity, protection against radiation, and criticality safety for fissile material contents) during the entire life cycle of storage, transportation, and disposal. The paper will cover existing storage standards and examine the need for considering the aging of contents, interactions between contents and packaging and between packaging and the environment, under normal conditions of storage and facility design-basis accidents. The paper will also describe the key attributes in the programs for managing aging degradation of materials and packaging components, including periodic inspection and monitoring, and the application of advanced technology such as radiofrequency identification (RFID). The RFID technology appears particularly suitable for monitoring and tracking high-value assets and in nuclear materials management with greatly enhanced safeguard and security.

STORAGE STANDARDS

There are two DOE standards for long-term (i.e., up to 50 years) storage of nuclear materials: STD-3028-2000[1] for uranium-233-bearing materials and STD-3013-2004[2] for plutonium-bearing materials. DOE-STD-3013-2004 was developed over a period of 10 years, with significant revisions in 1996, 1999, 2000, and 2004. It covers stabilization, packaging, and storage of Pu-bearing materials, including metals and oxides. Plutonium metals and alloys do not need to be stabilized, provided the pieces have a mass greater than 50 g and do not include turnings or wire, but they must be free of non-adherent corrosion products, liquids, and organic materials. Oxides must be calcined at 950^{o}C for at least 2 h and shown to have residue moisture content $\leq$ 0.5 wt.%, or weight loss $\leq$ 0.5%, at the time of packaging following calcination. DOE-STD-3013-2004 does not limit chloride content, but recognizes that materials could have significant chloride content following calcination.

Packaging of the Pu-bearing materials after stabilization relies on the use of a minimum of two individually welded, nested stainless-steel containers known as 3013 cans. These ductile, corrosion-resistant, 300-series stainless-steel cans isolate the contents from the environment; the outer can provides a pressure boundary with a minimum design pressure of 4,816 kPa (699 psig), which is much higher than the estimated buildup of internal pressure from all known sources except water desorption and vaporization. (The sources of internal pressure considered include temperature rise, helium from alpha decay, and hydrogen from radiolysis of residue moisture in the stabilized oxide.) The design of the outer 3013 can also considered mechanical damage from accidental drop and abnormal temperature excursions during a loss of cooling in a facility's storage vault.

Development of DOE-STD-3028-2000 for ^{233}U-bearing materials was modeled after DOE-STD-3013-2004 for Pu-bearing materials, but took into account the significant physical/chemical differences between the two materials. For example, the stabilization of ^{233}U-bearing material by heating in an oxidizing atmosphere is at a lower temperature (>750^{o}C) and shorter duration (>1 h) than those for the Pu-bearing material, and the design pressure for the stainless-steel outer can for the ^{233}U-bearing material after stabilization is 2,067 kPa (300 psia), compared with 4,816 kPa (699 psig) for the Pu-bearing material after stabilization. It is of interest to note, however, the following statement in the purpose and scope section of DOE-STD-3028-2000:

"*Major particulars for the safe storage of separated ^{233}U are preventing nuclear criticality, containing radioactive materials, protecting personnel from undue exposure to penetrating radiation, and safeguarding the SNM [special nuclear materials]. The storage facility plays a* *primary role in addressing all of these safety elements except containment. The facility plays a principal backup role (i.e., defense in depth) in confining radioactive contaminants during upset conditions. Material stabilization, consolidation, access limitation, low maintenance storage, and reliability in verification of the inventory are the Department's present goals for the ^{233}U-bearing materials*."

This paragraph sums up the three major safety concerns for the storage of ^{233}U-bearing material (i.e., containment of radioactivity, protection against radiation, and criticality safety), which are the same as those mentioned earlier for packagings during storage, transportation, and disposal of radioactive and fissile materials. The paragraph also mentions the safeguard and security concerns and the important role of the storage facility. The principles involved in the two DOE storage standards are nearly identical and universally applicable to other storage situations, i.e.,

- Stabilize contents, e.g., by high-temperature calcination, oxidation, or other means.
- Remove liquids, corrosion products, organic materials, and gas-generating species.
- Isolate contents in corrosion-resistant, stainless-steel cans from the environment.

Both DOE storage standards also include requirements for a surveillance program that addresses site-specific operating conditions, surveillance frequency, package selection, sample size, non-destructive examination, acceptance criteria, and corrective actions when the criteria are not met. In addition, the surveillance program for Pu-bearing materials in DOE-STD-3013-2004 is required to include provisions for evaluation of any observed off-normal behavior or unanticipated condition.

Gas Generation

There was limited information on gas generation from actual Pu-bearing material during the development of DOE-STD-3013-2004. It was assumed that the only mechanisms for gas generation are the alpha decay of plutonium generating helium and the radiolysis of water (i.e., residue moisture in the Pu-bearing material) generating hydrogen, whereas oxygen was assumed to be taken up by the plutonium oxide. On this basis, it was relatively straightforward to calculate the total pressure from the fill gas, the alpha-decayed helium, and the hydrogen from the mass of the stabilized plutonium oxide and its moisture content (< 0.5 wt%), as shown in Appendix B of DOE-STD-3013-2004. If the storage temperature is kept low (e.g., $\leq 250^{o}$C), the gas pressure in the 3013 can, under static pressure loading conditions, will remain sufficiently low compared to the design pressure of 4,816 kPa (699 psig).

The possibility that a flammable mixture of hydrogen and oxygen may form is a safety concern, if the oxygen from the radiolysis of water is not taken up completely by the plutonium oxide, as is assumed in DOE-STD-3013-2004, to form PuO_{2+x} , or if PuO_{2+x} is, by itself, not completely stable over the prolonged period of storage. In "The path to PuO_{2+x},"[3] Luis Morales states

"*The stage is set for the next act in this unfolding adventure in actinide-material science. Much work remains to be done to fully understand the relationships between the thermodynamic, crystallographic, and electronic properties of the actinide oxides. ... A systematic examination of all actinide oxides, with emphasis on uranium, neptunium, and plutonium (UO_2, NpO_2, and PuO_2) and their ability to accommodate extra lattice of oxygen, is needed.*"

Researchers at Los Alamos National Laboratory have also studied gas generation by pure and impure plutonium oxide materials and found substantial differences in hydrogen and oxygen.[4] Typical Los Alamos impure oxide materials in the packaged 3013 cans contain plutonium oxide with salt mixtures of sodium chloride (NaCl), potassium chloride (KCl), and magnesium chloride ($MgCl_2$). Comparison of the pressure versus time plots of high-purity and impure PuO_2 with 0.5 to 2.0 wt.% added water showed much higher gas generating rates, 12 and 1.7 kPa/day, for impure PuO_2 with 0.5 and 2.0 wt.% added water, respectively, than the rate of 0.25 kPa/day for high-purity PuO_2 with 0.5 wt.% added water. For the impure PuO_2, the ratio of hydrogen to oxygen decreased from ≈ 8 to 4 at longer times; at these ratios the gases are flammable. Recent data from a 3013 test unit showed a maximum gas pressure of ≈ 220 kPa (32 psia) after one year, and a flammable mixture of 46% H_2 and 14% O_2.

Researchers at Argonne National Laboratory and Michigan Technological University have been studying the long-term stability of PuO_{2+x} by performing *ab initio* all-electron density functional theory calculations.[5] Such electronic structure calculations, coupled with advanced synchrotron techniques, may help determine the valence structure of the various Pu-containing phases and the role of impurity salts on gas generation and the long-term stability of PuO_{2+x}.

Worth noting here is that hydrogen gas generation, deflagration, and detonation in a confined space have long been a safety concern for transportation packagings containing transuranic (TRU) waste, such as TRUPACT-II.[6] In fact, any radioactive waste type (especially alpha-bearing waste, waste containing hydrocarbons, and biological waste that has not been adequately treated) will generate gases that may pose the risk of fire or explosion, if the gases are flammable. Venting, filtering, or other devices may be employed to prevent overpressure in the waste packages for long-term storage;[7] however, these devices are not allowed for Type B radioactive material transportation packagings per 10 CFR 71.43(h).

Interim Storage of Pu-bearing Materials in Model 9975 Transportation Packagings

As noted earlier, thousands of Model 9975 transportation packagings, shown in Figure 1, are currently used for interim storage of Pu-bearing materials in 3013 cans. Such usage created several operational safety issues: (1) the need to perform annual leakage tests for Model 9975

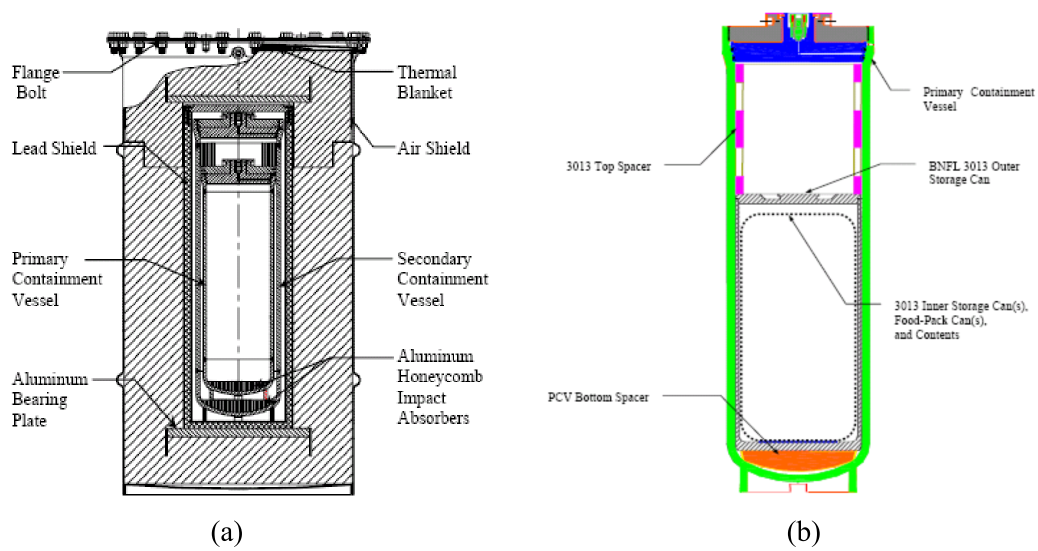

Figure 1. Schematic of Model 9975 transportation packaging: (a) cross-sectional view and (b) primary containment vessel containing a 3013 can assembly. The hatched area in (a) between the lead shield and the drum surface is the Celotex thermal insulation.

transportation packagings as part of the maintenance requirements, (2) the thermal response of Model 9975 packaging to a facility design-basis fire event, and (3) the long-term behavior of the Model 9975 packaging components due to exposure to the storage environment. With respect to the annual leakage test, that requirement may be waived with justification such as a monitored storage program.[8] With respect to the thermal performance of the Model 9975 packaging under a facility design-basis fire event, one study[9] has shown that even in an 800°C fire with a duration of up to 4 hours, the Model 9975 packaging would provide adequate thermal protection, and the 3013 can temperature would stay significantly below the Pu/Fe eutectic temperature of ≈ 400-410°C. The 3013 cans are protected from the fire mainly by the Celotex insulation in the Model 9975 packaging.

With regard to the long-term behavior of the Model 9975 packaging components that are exposed to the storage environments, the monitored storage program has reported corrosion of lead shielding due to acetic acid, which is an off-gas vapor product from degradation of the Celotex and the water-based polyvinyl acetate glue. The rate of the corrosion attack is very low (≤ 0.05 mm/y) and will not challenge the safety function of the lead shielding. As the source and mechanism of the corrosion were identified,[10] a design change was made to insert a stainless-steel sheathing between the Celotex and the lead shielding. This design change should effectively eliminate corrosion of lead shielding for the future Model 9975 transportation packagings.

As part of the monitored storage program, laboratory studies are being conducted by researchers at the Savannah River National Laboratory to determine the properties of Celotex following thermal aging treatment[11] and the long-term performance of elastomeric O-ring seals for the Model 9975 packagings.[12] The goal of these studies, which use the actual Model 9975 packaging components but under an accelerated aging environment, is to develop models for predicting the service life of the components under long-term storage conditions. Such predictive capability, coupled with the continuing surveillance, monitoring, and verification of the long-term behavior of the key components (e.g., Celotex and the elastomeric O-ring seals), will ensure the safety functions of the Model 9975 transportation packagings during storage. These laboratory studies under simulated and accelerated aging environments are conducted in the same spirit as those carried out by the researchers in Los Alamos's Materials Identification and Surveillance program on gas generation in impure plutonium oxides.

AGING MANAGEMENT

At least three aging-related studies were reported in PATRAM 2004: the thermal behavior of neutron shielding material, NS-4-FR, under long-term storage conditions,[13] aging management assessment of Type B transportation packages,[14] and corrosion protection of containers for the radioactive waste-storage requirement,[15] by authors from Japan, Canada, and Germany, respectively. In addition, Sassoulas et al.[16] described an experimental study of aging of metallic gaskets for spent fuel casks, and made a century-long life forecast based on 25,000-h-long experiments. In the United States, the license for the first Independent Spent Fuel Storage Installation (ISFSI) at Surry was renewed in 2005 by NRC for another 40 years; other dry cask storage ISFSIs are certain to follow in future license renewals.

During the late 1980s and throughout the 1990s researchers at Argonne National Laboratory assisted NRC in the development of guidance documents for license renewal of U.S. commercial nuclear power plants. The lead author of this paper is the project manager for NUREG-1800, *Standard Review Plan for Review of License Renewal Applications for Nuclear Power Plants*,[17] and NUREG-1801, *Generic Aging Lessons Learned (GALL) Report*.[18] Both reports were first published in 2001 and updated in 2005. The basic principle of license renewal of a nuclear power plant is that aging shall be effectively managed such that the components can continue to provide their intended function in a manner consistent with the current licensing basis for the extended period of operation.

One needs to understand the mechanisms of aging degradation of reactor materials and components in order to manage (i.e., slow down) aging or the rate of aging degradation. Time, environment, and use are the three primary factors affecting aging, and the rate of aging degradation always increases with prolonged exposure to an aggressive environment and/or frequency of use. For the systems, structures, and components (SSC) in nuclear power plants, corrosion, irradiation, and fatigue are the three primary mechanisms of aging degradation, and each area has been and will continue to be studied extensively because of the safety concerns and potential consequences from the unexpected failure of SSCs. For spent fuel casks and radioactive materials packagings, the conditions of the environments (e.g., temperature and irradiation) are generally not as severe as those in the nuclear reactor; however, the principal attributes of an effective aging management program (AMP) developed for SSCs in a nuclear power plant are applicable and worth repeating:[17]

1. Scope of the program: The scope of the program should include the specific structures and components subject to an aging management review.

2. Preventive actions: Preventive actions should mitigate or prevent the applicable aging effects.

3. Parameters monitored or inspected: Parameters monitored or inspected should be linked to the effects of aging on the intended functions of the particular structure and component.

4. Detection of aging effects: Detection of aging effects should occur before there is a loss of any structure and component intended function. This includes aspects such as method or technique (i.e., visual, volumetric, surface inspection), frequency, sample size, data collection and timing of new/one-time inspections to ensure timely detection of aging effects.

5. Monitoring and trending: Monitoring and trending should provide for prediction of the extent of the effects of aging and timely corrective or mitigative actions.

6. Acceptance criteria: Acceptance criteria, against which the need for corrective action will be evaluated, should ensure that the particular structure and component intended functions are maintained under all current licensing basis (CLB) design conditions during the period of extended operation.

7. Corrective actions: Corrective actions, including root cause determination and prevention of recurrence, should be timely.

8. Confirmation process: The confirmation process should ensure that preventive actions are adequate, and appropriate corrective actions have been completed and are effective.

9. Administrative controls: Administrative controls should provide a formal review and approval process.

10. Operating experience: Operating experience involving the aging management program, including past corrective actions resulting in program enhancements or additional programs, should provide objective evidence to support a determination that the effects of aging will be adequately managed so that the structure or component's intended functions will be maintained during the period of extended operation.

Chapter IX in NUREG-1801 contains selected definitions and use of terms for describing and standardizing structures, components, materials, environments, aging effects, and aging mechanisms. Chapter XI in NUREG-1801 lists fifty-four (54) AMPs for the SSCs in nuclear power plants, e.g., water chemistry (XI.M2), bolting integrity (XI.M18), BORAFLEX monitoring (XI.M22), fire protection (XI.M26), reactor vessel surveillance (XI.M31), one-time inspection (XI.M32), structure monitoring (XI.S6), and electrical cables and connections not subject to 10 CFR 50.49 environmental qualification requirements (XI.E1). Each of these AMPs contains the formalized 10 attributes that can serve as examples in the development of aging management programs for the components of Type B radioactive and fissile materials packagings.

Chapter X in NUREG-1801 contains several time-limited aging analyses (TLAA): evaluation of aging management programs under 10 CFR 54.21 for metal fatigue of reactor-coolant pressure boundary (X.M1), concrete containment tendon prestress (X.S1), and environmental qualification of electrical components (X.E1). Each of these TLAAs also contains the 10 attributes of an AMP, and these TLAAs are specifically related to the frequency of use (e.g., cycles of fatigue) and exposure time (e.g., relaxation of pre-stress) that one may find parallels in packaging during transportation (e.g., vibration), bolted closure (e.g., preload torque) and elastomeric/metallic seals (e.g., compression set and relaxation).

The Appendix of NUREG-1801 describes the quality assurance (QA) of the aging management programs that follow the QA requirements of Appendix B to 10 CFR Part 50, which consists of eighteen (18) elements that are also in Subpart H of 10 CFR 71 for packaging and transportation of radioactive material, as well as Subpart G of 10 CFR 72 for storage of spent fuel and high level waste. This Appendix emphasizes corrective actions, the confirmation process, and administrative controls because of the stretched time horizon needed to evaluate the effectiveness of the AMPs for managing the aging effects, and the important role of confirmation by independent audits and verification. In fact, the NRC staff conducts audits of AMPs at utility plants as part of the license renewal process. DOE also conducts packaging QA audits per DOE Order 460.1B, *Packaging and Transportation*. Highlights of recent DOE packaging QA audits and source verification at selected DOE sites are provided in a separate PATRAM 2007 paper by Fabian et al.[19]

ADVANCED TECHNOLOGY

In 2006, the Packaging Certification Program (PCP) of EM-60 initiated a project to evaluate the potential application of RFID technology as part of a long-term strategy in the life-cycle management of radioactive materials packaging.[20] A small number of high-performance, commercial RFID tags (ST-676) were obtained from Savi Technology. These tags were irradiated with a gamma source at Argonne National Laboratory and continuously monitored for performance over three months. The surprisingly robust radiation resistance of the tags led to discussions between Argonne (on behalf of PCP) and Savi that culminated in a collaboration on the development of a prototype RFID system, an effort that involved both hardware modification (form factor, seal sensor, and batteries) and software development. A limited number of prototype tags have been developed for the Model 9975 packaging, shown schematically in Figure 2, and preliminary system testing has been completed with encouraging results.[21] The RFID system appears to be able to achieve all potential benefits envisioned originally for the management of nuclear materials packaging: enhanced safety and security, reduced need for manned surveillance, real-time access of status and history data, and overall cost-effectiveness. In the near future, research engineers at Savannah River National Laboratory and Argonne will field test the RFID system on Model 9975 drums, which are used for storage and transportation of radioactive and fissile materials.

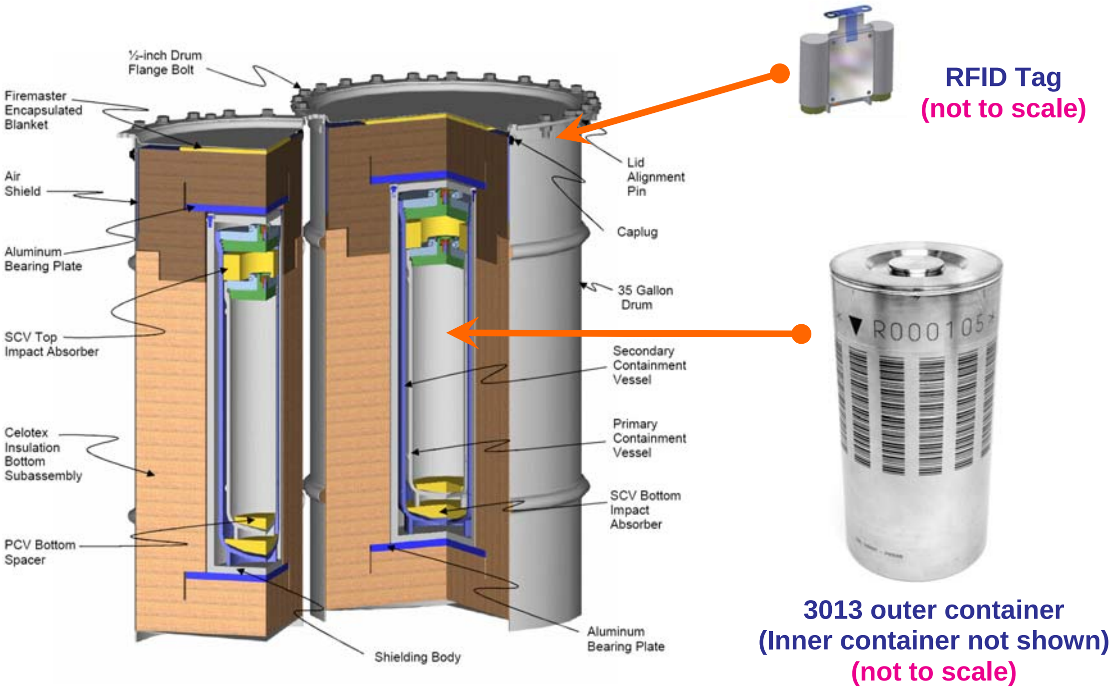

Figure 2. Schematic cutaway view of Model 9975 transportation packaging showing a 3013 outer container and an RFID tag.

Any non-destructive method that allows interrogation of the conditions of components inside a packaging will be useful, for example, x-ray digital radiography or magnetic pressure detection. Powerful x-ray sources, solid-state semi-conductor detectors, and 3-D reconstruction computed tomography are being explored as means for packaging development and testing[22] and for the detection of aging effects.[23]

DISCUSSION

In this description of the framework of LCM strategies, storage standards, and aging management of radioactive materials packagings, occasional references were made to waste packages and dry cask storage systems for commercial spent nuclear fuel. Storage, transportation, and disposal of high-level waste packages are generally considerably more complex because their contents are more radioactive, and less well characterized, than Pu-bearing and/or ^{233}U-bearing materials that have been stabilized and packaged in accordance with DOE-STD-3013-2004 and DOE-STD-3028-2000.

Storage, transportation, and disposal of commercial spent nuclear fuel are perhaps the most complex of all waste types, because not only the reactor irradiation history matters, but also the history of the spent fuel since its discharge from the reactor, e.g., the time the spent fuel underwent cooling, drying, dry storage, transport, and disposal. Particularly for high burnup ($\geq$ 45 GWd/MTU) fuel, the NRC Interim Staff Guidance (ISG)-11, Rev. 3, describes the cladding considerations for transportation and storage of spent fuel.[24] The concern for potential radial hydride formation in the cladding led to the following statement: *For all fuel burnups (low and high), the maximum calculated fuel cladding temperature should not exceed 400^{o}C (752^{o}F) for* *normal conditions of storage and short-term loading operations (e.g., drying, backfilling with inert gas, and transfer of the cask to the storage pad)*. The ability to calculate the peak cladding temperature in a spent fuel storage and transportation cask is, therefore, important to conform with the guidance of ISG-11, as shown in the PATRAM 2007 paper by Li et al.[25] Other concerns for storage and transportation of commercial spent nuclear fuel are the classification of fuel conditions, burnup credit, moderator exclusion, and potential rod splitting discussed in NRC ISG-1, -8, -19, and -22, respectively.[26-29]

In June 2007, the DOE Office of Civilian Radioactive Waste Management issued a document titled *Transport, Aging and Disposal Canister System Performance Specification*, Rev. 0.[30] The basic unit of the transport, aging, and disposal (TAD) canister system is the TAD canister itself, which is sealed by welding after being loaded with commercial spent nuclear fuel at reactor sites or the repository. The loaded TAD canister may be stored in a storage system certified by NRC per 10 CFR 72; transported in a transportation overpack certified by NRC per 10 CFR 71; and aged in an aging overpack or disposed of in a waste package at the disposal site. All three of these functions will be covered by the repository license granted under 10 CFR 63. Similar to the multi-purpose canister concept in the early 1990s and the 3013 cans used for the Pu-bearing materials, the TAD canisters will isolate the spent fuel from the environment after being loaded into the canister by welding. Standardized leak-tight canisters are also being adopted for the DOE spent nuclear fuel. These welded, leak-tight canisters not only improve worker safety by eliminating future repackaging needs, but also provide a barrier for isolation of the spent fuel from the environment during storage, transport, and disposal.

SUMMARY

A framework has been developed for life cycle management strategies regarding Type B radioactive and fissile materials packaging, drawing upon current DOE storage standards and examples from interim storage of Pu-bearing materials in Model 9975 transportation packagings. Key issues highlighted during long-term storage of Pu-bearing materials included gas generation and stability of PuO_{2+x}; other operation safety issues highlighted for interim storage of Model 9975 transportation packagings included the need to consider the facility design-basis fire event and long-term behavior of packaging components such as Celotex and elastomeric O-ring seals. The principles of aging management are described, and the key attributes and examples of effective aging management programs are provided based on the guidance documents developed for the NRC license renewal of nuclear power plants. The DOE Packaging Certification Program plans to expand its mission into packaging certification for storage and aging management, as well as application of advanced technology, such as RFID, for life cycle management of radioactive materials packagings. Preliminary testing has shown RFID technology to hold strong promise for life cycle management of radioactive materials packaging.

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