IDENTIFICATION OF POTENTIAL DEGRADATION PHENOMENA FOR SPENT FUEL DRY CASK STORAGE SYSTEMS

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ABSTRACT

The U.S. Nuclear Regulatory Commission (NRC) licenses the dry storage of spent nuclear fuel at operating and decommissioned nuclear power plants and certain other stand-alone facilities. NRC is currently updating the regulatory guidance for spent fuel dry storage license renewal and evaluating the technical information needs to ensure that spent fuel can be safely stored and transported up to and beyond 60 years. As part of this initiative, NRC is identifying aging-related degradation mechanisms that could affect components important to safety in the dry cask storage systems. The degradation phenomena are identified from reviews of the existing technical literature, previous knowledge gap analysis reports, operational experience from analogous systems, and generic guidance for reactor license renewal. NRC may use the findings to help determine the scope of aging management programs or analyses that would provide an adequate basis to ensure that the components continue to perform their intended safety function, as well as to highlight issues for which further research is needed to address limitations in current knowledge.

Keywords: Spent nuclear fuel, waste, storage, stainless steel, concrete, aging, stress corrosion cracking

1.0 Introduction

Beginning in the 1980s, a number of operating and decommissioned reactor sites in the U.S., as well as some other facilities, began to place spent nuclear fuel (SNF) in dry cask storage systems (DCSS). The U.S. Nuclear Regulatory Commission (NRC) licenses dry storage of SNF under Title 10 of the Code of Federal Regulations (10 CFR), Part 72 “Licensing Requirements for the Independent Storage of Spent Nuclear Fuel, High-Level Radioactive Waste, and Reactor-Related Greater than Class C Waste.” Under the provisions of 10 CFR, Part 72, licenses for specific facilities or Certificates of Compliance (CoC) for cask systems are typically issued for an initial term of 20 years, after which they may be renewed for additional terms of up to 40 years. When DCSS were initially placed into service in the 1980s, it was anticipated that a permanent geological repository would be available within 20 to 40 years. To date, however, a permanent disposal facility has not been licensed and spent fuel is likely to remain in dry storage longer than was expected. As such, the industry and NRC are addressing issues related to renewal of specific licenses and CoCs, and at the same time, identifying technical information needed to ensure that SNF can be safely stored and transported for extended timeframes.

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The regulatory approach to ensure that structures, systems, and components (SSCs) in the cask system continue to perform their intended safety function during license renewal terms can be broadly described as aging management, as described in 10 CFR, Parts 72.42 and 72.240. Aging management can include what are referred to as time-limited aging analyses (TLAAs), which are calculations or analyses used to demonstrate that a structure, system, or component will continue to perform its intended function during an extended operating life. An example is a fatigue calculation showing that the number of anticipated load cycles within the licensed life is less than the number that would cause failure of that component. The other main approach to aging management is the implementation of aging management programs (AMPs), which are intended to monitor for and detect aging-related degradation of SSCs before a loss of intended safety function, and then to provide an approach for mitigation or other corrective action if such degradation occurs. NRC guidance for form and content of TLAAs and AMPs is found in greater detail in NUREG-1927, "Standard Review Plan for Renewal of Specific Licenses and Certificates of Compliance for Dry Storage of Spent Nuclear Fuel," the first revision of which was posted for public comment in 2015 [1].

The development of TLAAs and AMPs for a license renewal application requires a thorough and systematic evaluation of the SSCs for the particular cask system design, to identify their intended safety functions, their materials of construction, and the operating environment (e.g., temperature, atmosphere, humidity, radiation field). The guidance in NUREG-1927 states that the degradation phenomena should be considered which, "…could be reasonably expected to occur, as well as those that have actually occurred, based on industry and site-specific operating experience and component testing." Suggested sources to review for identification of degradation phenomena include maintenance and inspection records, corrective action reports, consensus codes and standards, operating experience from outside the nuclear industry, among others.

While it is expected that applicants will provide sufficient information to demonstrate the adequacy of their licensing basis, NRC needs to perform an independent safety review to confirm that the regulatory requirements are satisfied. NRC has already issued renewals for several specific licenses and CoCs, but in the course of performing the application reviews, NRC recognized that enhancements to generic guidance were needed to improve the consistency and efficiency of technical reviews and to clarify expectations for the industry. As mentioned above, the first revision of NUREG-1927 was released for public comment, and includes example AMPs for localized corrosion and stress corrosion cracking (SCC) of welded austenitic stainless steel canisters and for reinforced concrete structures. Over the next few years, NRC envisions issuing further generic guidance, potentially taking form similar to NUREG-1801, "Generic Aging Lessons Learned (GALL) Report," for reactor license renewal [2]. NRC, with the support of contractors at the Center for Nuclear Waste Regulatory Analyses, is reviewing various DCSS designs to independently develop a listing of aging-related degradation phenomena that could affect components fabricated from certain materials and in certain environments. This paper describes the process for this evaluation and summarizes the initial findings of the activity, which is still ongoing.

2.0 DESCRIPTION OF MATERIALS EVALUATED

NRC has issued CoCs for over 15 different DCSS designs and SNF is stored at more than 70 sites throughout the U.S. The design concept for a DCSS involves SSCs that are intended to perform various safety functions, including confinement, structural stability, radiation shielding, thermal management, and criticality control. Typically, spent fuel assemblies are placed into basket assemblies inside metallic

canisters sealed by welding or bolting. In a number of designs, the sealed canister is placed inside a concrete overpack or storage module with external air vents to facilitate passive cooling, as illustrated in Figure 1 [3]. The entire system sits, usually unanchored, on top of a level, reinforced concrete pad. In determining the priority of systems for the evaluation described in this paper, NRC focused on the most commonly used systems and those which are closest to the expiration of the initial licensing term. These include the Areva TN Standardized NUHOMS System (Docket Number 72-1004) and the Holtec International HI-STORM 100 Cask System (Docket Number 72-1014). A detailed evaluation of these systems is not presented here. Rather, this is a description of material-environmental interactions that may be broadly applicable to these and other DCSS designs. Further, it should be noted that this paper primarily focuses on the storage canister and overpack or storage module. The fuel assemblies themselves may require aging management, as well as the transfer cask in which the fuel canister is transported from the spent fuel pool to the storage pad. NRC is also reviewing these items but these are not addressed here.

2.1 Metallic Materials

The Standardized NUHOMS, HI-STORM 100, and other storage systems employ a welded Type 304 or 316 (or the corresponding L grades) austenitic stainless steel cylindrical canister with an internal basket structure to hold the fuel assemblies. The number of fuel assemblies in each canister can range from 20 to 70 depending on whether the fuel is from boiling water reactors or pressurized water reactors, whether the fuel is damaged, and the fuel heat load, which is determined by enrichment, burnup, and cooling time. The internal basket structure includes components such as sheaths, plates, and spacers to maintain the positioning of the fuel assemblies, provide for heat conduction, and to maintain criticality control. Most of the internal components are fabricated from austenitic stainless steel, though some, such as heat conduction elements, are fabricated from aluminum or carbon steel. Finally, neutron absorbing material, including aluminum boron carbide cermets and borated aluminum metal matrix composites, may be inserted into the canister for criticality control.

Metallic materials are also used for the overpacks or storage modules into which the welded canisters are placed. Some storage modules use concrete reinforced with carbon steel rebar, while others use unreinforced concrete fill between carbon steel liner plates. Aluminum, stainless steel, or carbon steel may also be used for concrete heat shields inside the storage module. Lastly, various structural elements, such as rails, and support beams, as well as attachment hardware, are also fabricated from carbon steel. Table 1 lists some of the common uses of metallic materials.

Table 1: Common Metallic Materials Used in DCSS

Material	Specifications	Use
Austenitic stainless steel	304, 304L, 316, 316L	Canister shell, lid, canister internal basket structures, concrete heat shields
Aluminum	1100, 6061	Canister internal basket structures, concrete heat shields
Carbon steel	SA-533, SA-537, SA-36 , SA-516, A615, A706	Canister internal basket structures, concrete reinforcing bar, concrete structural liner, support elements, hardware
Metal matrix composites and cermets	Boralyn, Boral, Metamic	Neutron absorbers

2.2 Concrete

Concrete in DCSS is primarily used for the pad on which the systems sit, and to provide structural integrity and radiation shielding for the overpack or storage module in which the sealed metallic canister is placed. Regulations in 10 CFR, Part 72 require that the DCSS be designed to withstand external natural phenomena loads, as well as other design basis external man-made events. In NUREG/CR-6407, the storage pad is categorized as a Category C component, which means that its failure would have only a minor effect on safety [4]. Nevertheless, its failure could affect the functionality of Category A and B components, so it is considered within the scope of this analysis. The concrete structures are typically fabricated according to ACI specifications, including ACI-318, "Building Code Requirements for Structural Concrete" [5] and ACI-349, "Code Requirements for Nuclear Safety-Related Concrete Structures" [6]. The aggregates are intended to conform to ASTM C33, "Standard Specification for Concrete Aggregates" [7]. Aggregates are typically locally sourced and are thus geographically varied in their mineralogy. Aggregates may include limestone, dolomite, marble, basalt, granite, gabbro, and rhyolite. The concrete is designed to meet a specified compressive strength with water-to-cement ratio generally less than 0.5 and low water-soluble chloride ion content.

3.0 DESCRIPTION OF ENVIRONMENT

For the purposes of this evaluation, there are four general environments to which the materials described in Section 2.0 will be exposed (apart from those which are embedded, as in reinforcing bar in concrete): the canister internal atmosphere, the air outside the canister but inside the storage module or overpack, the outdoor air external to the storage module or overpack, and fill or soil for below grade support structures. These are briefly described as follows.

3.1 Canister Internal

After the fuel assemblies are placed into the canister in the spent fuel pool, it is dried by vacuum or forced gas and backfilled with helium to provide an inert environment for the storage period. Assuming that there is a minimal amount of water in the canister, the primary environmental parameters of concern for the canister internals are the temperature and the radiation field. The fuel generates decay heat which must be managed to avoid thermally-related degradation of the fuel cladding and canister internal components. NRC previously analyzed temporal temperature profiles for fuel in dry storage, which depends on fuel burnup and other parameters [8]. Calculations indicated that cladding temperature may decay from in excess of 300 ^{o}C in the first years after loading, to around 200 ^{o}C within 20 to 40 years, to below 100 ^{o}C beyond 60 years. The metallic internal components will be at even lower temperatures than the fuel cladding. With respect to the radiation field, previously published reports describe gamma fields of about 10^5 R/hour and neutron flux of $10^4 – 10^6$ n/cm^2-s [9].

3.2 Inside Air

The term inside air refers to the atmosphere outside the sealed canister, but within the confined internal space of the overpack or storage module. Components exposed to inside air include the external surface of the canister, the inside surface of the overpack or storage module, heat shield assembles, and other internal support structures, such as rails and beams. The overpack or storage module is ventilated to the outside air to allow airflow for passive cooling, otherwise there is no climate control. The environmental parameters of concern for the inside air include the temperature, humidity, radiation field, and the composition of atmospheric particulates which may enter through the vents. Modeling suggests that the

surface temperature of the canister could be in the range of 120 to 200 ^{o}C for the few first years after loading, would drop below 100 ^{o}C within 20 years, and below 60 ^{o}C within 60 years [10]. To date, only limited attempts have been made to inspect the interiors of the overpack or storage modules, but there is evidence of dust or other particulate materials on the canister surfaces, as well as of rainwater intrusion through the vents [11].

3.3 Outside Air

The term outside air refers to the uncontrolled environment around the overpack or storage module. Components exposed to outside air include the external surfaces of the overpack or storage module and the pad on which the systems are placed. The radiation field is not a concern in this environment, so the important parameters include the temperature, humidity, moisture (e.g., rain, snow), and composition of atmospheric particulates.

3.4 Below Grade

The term below grade refers to natural soil or engineered fill placed on top of natural soil which supports the storage pad. The primary parameters of concern for below grade structures are the temperature, moisture content, and soil and groundwater chemistry, particularly the concentrations of chemical species such as chloride, sulfate, and magnesium.

4.0 DEGRADATION PHENOMENA

To identify relevant degradation phenomena for the materials described in Section 2.0 exposed to the environments described in Section 3.0, a range of information sources were reviewed. These include primary sources in the technical literature, gap assessments previously prepared by NRC [12], Department of Energy [13], Nuclear Waste Technical Review Board [14], and the Electric Power Research Institute [15], and operational experience summarized in the GALL Report for reactor license renewal. This section provides a brief discussion of those phenomena thought to be most likely to affect the structural functions of the DCSS as well as the rationale for determining that other phenomena are of lower significance.

4.1 Phenomena Affecting Metallic Materials

4.1.1 SCC and Localized Corrosion of Stainless Steel in Indoor Air

The potential for SCC of austenitic stainless steel exposed to inside air is primarily attributed to the potential presence of atmospheric chloride species which could deposit on the cask outside shell or other structural support surfaces inside the overpack or shielding structure. There is operational experience of chloride-induced SCC on outdoor piping and tank systems at a number of nuclear power plants, particularly those in near-marine environments where the concentration of atmospheric chloride species is relatively high [15]. The process for initiating SCC involves the deliquescence of deposited species on the material surfaces in humid conditions to form a chloride-rich brine. Stresses to propagate cracking may arise from weld residual stresses or from other work imparted to the components. The operative temperature range for this process appears to be between about 30 and 80 ^{o}C, above which the humidity required to sustain deliquescence is unlikely to be reached [16]. Pitting and crevice corrosion in localized regions, including tight spaces where components are in contact with one another could occur under environmental conditions similar to that of SCC and, in fact, may serve as a precursor for crack initiation.

Further details about these phenomena can be found in a number of comprehensive analyses and studies by NRC [16], the Electric Power Research Institute [17], and Japanese researchers [18], among others.

4.1.2 Creep and Thermal Aging of Aluminum Components in the Canister Internal

Time-dependent deformation under applied stress, or creep, is generally understood to occur in metals at temperatures above 0.4 times the absolute melting temperature. For pure aluminum, this is in the range of 100 ^{o}C, though it is likely to be somewhat higher in precipitation-hardened aluminum 6061 [19]. For aluminum canister internal components, this phenomenon is of main concern for the first relicensing period, as it is expected that the temperature will drop below 100 ^{o}C within 60 years. Aluminum components internal to the canister could also be affected by thermal aging leading to loss of strength. Precipitation hardening of aluminum 6061 is performed between 163 and 204 ^{o}C, suggesting that overaging may occur when the canister temperature is high during the years after loading. Similar to creep, thermal aging is not likely to persist beyond 60 years.

4.1.3 Corrosion of Carbon Steel in Inside Air

Corrosion processes that could affect carbon steel components, such as structural supports and hardware elements, inside the overpack or shielding module include general corrosion, localized corrosion, and galvanic corrosion. Both general and localized corrosion will be affected by the temperature of the components, the presence of moisture from humid air or the ingress of rainwater, and the composition of atmospheric species that deposit on the surfaces. Inspections of cask systems have identified regions where there is evidence of rusting or surface staining on carbon steel components [11]. Component configurations where there are lap joints, bolted connections, or other restricted spaces also offer the potential for crevice corrosion. Finally, there may be certain locations where a stainless steel and carbon steel structural elements are in contact. A galvanic couple between the materials in which the carbon steel element is anodic could cause corrosion.

4.1.4 Wet Corrosion and Blistering of Boral in the Canister Internal

Boral is material with cermet core consisting of a mixture of boron carbide and aluminum 1100 particles between cladding of aluminum 1100 sheets. The core is not fully sintered and is porous, allowing water to ingress when it is saturated. Blisters are known to form between the cladding and the core of Boral. One cause of the blister formation is due to reactions between water and aluminum in the pores which generate aluminum oxide and hydrogen gas [20,21]. The extent of blistering is expected to lessen over time, and likely arrest within 60 years, as the temperature decays to the point at which the aluminum oxide film is largely protective, and as available water is consumed by the reactions.

4.1.5 Phenomena Not Thought to Have a Significant Effect

In addition to those described above, a number of other degradation phenomena were evaluated, but are thought to have potentially lesser effects on the metallic SSCs. A lack of moisture in the evacuated canister, the inert environment, and the decreasing temperatures over time will preclude many corrosion processes, such as microbiologically-influenced corrosion. Additionally, neutron flux analyses indicate that the fluence experienced by metallic components is much lower than what would be needed to degrade the mechanical properties [9]. Further, there do not appear to be significant thermally-induced cyclic stresses that would cause fatigue. Finally, the thermal embrittlement of welded stainless steel internal to the canister was evaluated. It was recognized that spinodal decomposition of ferrite present in welds to form Fe-rich alpha and Cr-rich alpha prime phases and further aging to form an intermetallic G-

phase could reduce the toughness of the welds [22]. The relatively small amount of ferrite in the welds however, suggests that substantial degradation of fracture toughness is unlikely [23].

4.2 Phenomena Affecting Concrete Structures

4.2.1 Freeze-Thaw Degradation in Outside Air and Below Grade

Freeze-thaw degradation refers to a process wherein moisture in the pores of saturated concrete freezes and expands thereby imparting a tensile stress that causes cracking of the concrete. Successive freezing and thawing cycles can cause an accumulation of damage that manifests as surface cracking or scaling. Freeze-thaw degradation is commonly mitigated by the entrainment of air in the concrete to form small voids into which water can migrate to relieve the pressure in the pores [24]. Operational experience indicates that cracking of SNF storage structures at Idaho National Laboratory was caused by freeze-thaw degradation [25]. This phenomenon is of primary concern on the outside surface of the overpack or storage module, assuming that heat from the canister will prevent it on the inner surface, as well as in below grade structures above the frost line.

4.2.2 Aggregate Reactions in Inside Air, Outside Air, and Below Grade

The primary aggregate reaction of concern for concrete structures is alkali-silica reaction (ASR). Reactions between hydroxyl ions in the cement and reactive forms of silica in the aggregates produce a gel that increases in volume by taking on water [26,27]. The expansive pressure of the gel can cause an irregular pattern of cracking on the concrete surface. The requisites for ASR to occur are a sufficiently high alkali content of the cement, a reactive aggregate, such as chert, and the presence of water. Aggregates are typically tested for reactivity prior to construction to mitigate the potential for ASR. There is operational experience for ASR of concrete structures in operating reactors [28].

4.2.3 Aggressive Ion and Acid Attack of Concrete and Reinforcing Bar in Inside Air, Outside Air, and Below Grade

Aggressive ions that can attack and cause degradation of concrete structures include sulfate and chloride. For sulfate attack, species such as K_2SO_4, Na_2SO_4, $CaSO_4$, and $MgSO_4$ which are present in groundwater and rainwater, penetrate the concrete and react with alkali and calcium ions. Sulfate attack can promote cracking by expansive pressure from reaction products, as well as increase the porosity [29,30]. Chloride ions have a particularly deleterious effect on reinforcing bar by degrading the protective oxide layer which is typically maintained in the presence of basic pore water [31]. This compromises the integrity of the bond between the concrete and the reinforcing bar, and makes the latter more susceptible to corrosion. Acid attack could be caused by rainwater containing sulfur, nitrogen, or chloride-bearing species. Low pH acids can dissolve cement compounds and aggregate to form water-soluble compounds which are washed away by aqueous solutions [32].

4.2.4 Carbonation and Leaching of Concrete in Inside Air, Outside Air, and Below Grade

Gas-phase carbonation involves the reaction of atmospheric CO_2 with $Ca(OH)_2$ in the cement to form calcium carbonate [33]. By consuming $Ca(OH)_2$, carbonation lowers the local pH and compromises the passivity of the reinforcing bar. Moreover, carbonation can cause shrinkage of the cement paste which leads to cracking. Carbonation typically occurs at moderate humidity levels, as the saturation of the pores at high humidity slows the diffusion of CO_2. Leaching also involves the loss of $Ca(OH)_2$, but in the aqueous phase of concrete exposed to water. Evidence of leaching includes the presence of white mineral

scale on the concrete surface or powdery deposits if efflorescence occurs [6]. Leaching can erode the concrete as it passes through porous pastes or cracks and can lower the pH at the reinforcing bar, making it more susceptible to corrosion.

4.2.4 Differential Settlement of Concrete in Inside Air and Outside Air

Differential or structural settling of concrete is caused by uneven deformation of the supporting foundation material [34]. Factors affecting the settlement include the type of foundation soil (clayey, sandy, etc.), thickness of the foundation, water table level, and load, among others. It can also be caused by sudden liquefaction of supporting soil, triggered by an earthquake. Most settlement problems are caused by poor construction practices, and would be identified early in the life of the affected structure. Some evidence of uneven settlement was identified at the Three Mile Island fuel storage facility at Idaho National Laboratory [25].

4.2.5 Phenomena Not Thought to Have a Significant Effect

In addition to those described above, a number of other degradation phenomena were evaluated, but are thought to have potentially lesser effects on the concrete SSCs. Certain phenomena are thought to be relatively transient such that any effects would be evident within the first few years of operation. This includes shrinkage cracking and creep deformation. In the case of the latter, it is also not expected that there will be any substantive, sustained loads on the passive structures. The phenomenon of thermal dehydration was also evaluated and it was concluded that the design basis temperatures for the system are low enough to preclude loss of moisture to the extent that would compromise the concrete properties. Finally, analyses of the expected radiation doses for the concrete structures indicates cumulative fluence much lower than the limit specified in the ACI code, and well below those of reactor concrete structures, such as the biological shield wall surrounding the reactor pressure vessel [9].

5.0 IMPLICATIONS OF THE FINDINGS FOR AGING MANAGEMENT

As discussed in Section 1.0, NRC regulations require that licensees describe an approach for managing aging-related degradation of the SSCs, which may involve monitoring, periodic inspections, or engineering analyses to demonstrate that the functionality of the SSCs still meets the design basis. The listing of degradation phenomena in Section 4.0 is not intended to be comprehensive, nor inclusive of the full range of DCSS. It is the responsibility of the licensee or CoC holder to make an assessment for a specific system and environment. Nevertheless, NRC will follow such an analytical approach in the development of generic guidance for cask system license renewal. Of note, NRC has already described example AMPs for welded austenitic stainless steel canisters based on the potential for localized corrosion and SCC, as well as for concrete structures. It is expected that these will continue to be refined and other AMPs developed as additional testing data and operational experience are accrued.

6.0 SUMMARY

This paper describes the NRC approach to identify aging-related degradation phenomena that could affect austenitic stainless steel canisters and concrete overpacks or shielding structures for DCSS. This analysis is part of an ongoing NRC process to update the generic guidance for license renewal of cask systems, and to identify issues for which further research is needed in the context of long-term storage and transportability. For metallic components, which include the stainless steel canister shell, canister internal elements, and support elements for concrete structures, the phenomena that were identified include SCC of the canister shell and other stainless steel components exposed to indoor air, thermal aging and creep of

aluminum elements internal to the cask, corrosion of carbon steel components exposed to indoor air, and blistering of Boral neutron absorbers. For concrete structural and shielding components, the degradation phenomena include freeze-thaw, ASR, aggressive ion and acid attack, carbonation and leaching, and differential settlement.

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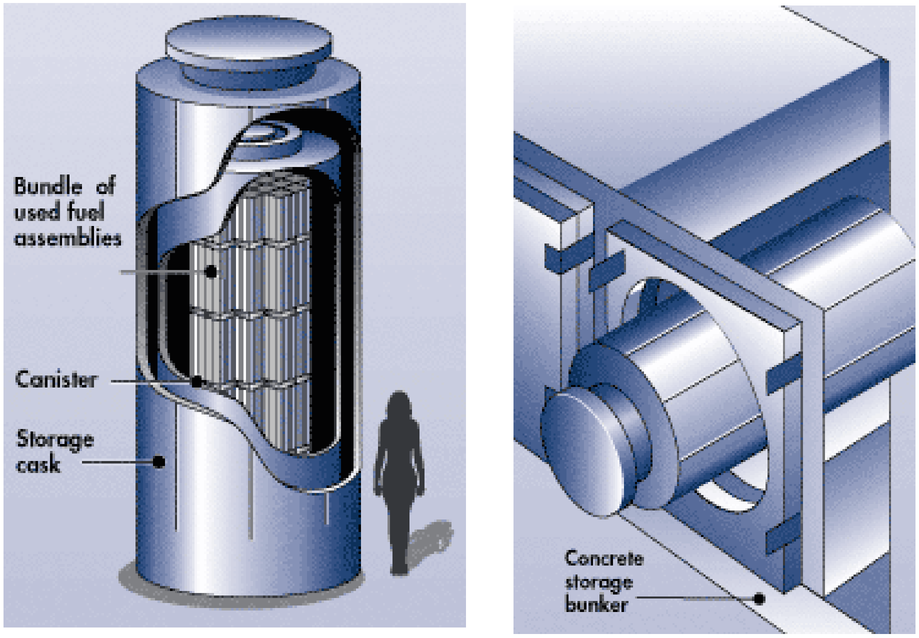

Figure 1: Illustrations of vertically oriented (left) and horizontally oriented (right) spent nuclear fuel dry cask storage systems [3].