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Engineered product storage under the advanced fuel cycle initiative. Part II: Conceptual storage scenarios ☆,☆☆

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Abstract

The Advanced Fuel Cycle Initiative (AFCI) of the US Department of Energy stresses waste minimization within an integrated nuclear fuel cycle. In one possible future fuel cycle scenario, the radionuclides ^{244}Cm and ^{241}Am are placed in decay storage until a capability for fast-spectrum transmutation is available. We discuss the scale of hypothetical wet and dry storage facilities for cesium/strontium and americium/curium products. Because of the extended storage period for cesium/strontium products, underground storage appears to be the option most acceptable to regulatory policy. Several underground designs are discussed. To reduce the required underground storage space, we recommend a pre-staging period where the cesium/strontium products are cooled in a small pool for 30–50 years. Americium and curium products would require storage for not more than approximately 50 years. For this reason, and because of the high thermal power density, pool storage appears to be the best option for americium and curium.

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1. Introduction

The Advanced Fuel Cycle Initiative (AFCI) program of the US Department of Energy has begun research and development of a fully integrated nuclear fuel cycle for the future. This could include a spent fuel processing and separation scheme that will produce, among other streams, two streams of high-heat radioactive products containing cesium and strontium, and americium and curium. In order to maximize the effective use of a single geologic repository for high-level waste, the short-lived fission products of cesium and strontium will be placed in decay storage prior to burial as low-level waste. Transmutation radionuclides ^{244}Cm, whose daughter product ^{240}Pu can be burned, and ^{241}Am will be placed in decay storage until the transmutation reactor fleet is constructed.

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In Part I [1], we described a model that computed the maximum size of cylindrical radioactive products consisting of cesium, strontium, americium and curium. Herein we summarize the properties of the storage material and conceptually adapt industry experience to describe the scale of centralized storage facilities for the engineered products. We do not describe facility design in detail.

At full deployment, operations under the proposed AFCI fuel cycle are assumed to recycle approximately 2000 MTHM per year. The mass yield (Fig. 1) and thermal decay power (Fig. 2) of Cs (^{137,134}Cs primarily), Sr (^{90}Sr), Am (primarily ^{241,243}Am), and Cm (primarily ^{243,244,245}Cm) are dependent on the burnup of the fuel and its age. Since rubidium and barium remain in the cesium and strontium product stream after fuel processing and barium is the daughter of ^{137}Cs, the total mass of the Cs/Sr/Rb/Ba stream does not change appreciably with fuel age. However, the mass yield of each element is significantly different between high-burnup fuel (50 GWd/MT) and more common low-burnup fuel (33 GWd/MT). The americium stream increases with fuel age due to the in-growth of ^{241}Am from ^{241}Pu, while curium will decay according to the 18.1-year half-life of ^{244}Cm. Thermal decay (Fig. 2) follows closely the half-life of the primary heat-generating radioisotopes (i.e., ^{137}Cs, ^{90}Sr, ^{241}Am, ^{244}Cm).

When opened, a facility will begin to accept engineered products for decay storage. We assume a single national storage facility for each product stream. To reach Class C LLW classification, storage for cesium and strontium is necessary for 300 years, at which time steady-state physical capacity is reached as the oldest products are moved to a LLW site. However, older storage products will be much cooler than new products so a steady state in total thermal output is reached well before 300 years (Fig. 3). For the Cs/Sr product, steady-state capacity is reached in 180 years. The facility, therefore, must accommodate 20–80 MW thermal capacity, depending on fuel age prior to initial processing. For Am/Cm, we assume that no product will remain in the storage facility longer than 50 years before being processed for transmutation. Then, the capacity is 7.6 MW for americium and 17 MW for curium products from 10-year cooled fuel.

We provide an overview of approximate facility size by which to judge options for going forward in the development of engineered product storage for the AFCI. We consider dry and wet storage facilities and underground tunnel storage. We argue that interim pool storage of fresh Cs/Sr/Rb/Ba products for 30–50 years significantly reduces the footprint of long-term, dry, underground storage facilities that will be needed for the remaining storage period of approximately 300 years. For Am/Cm or pure Cm, an above-ground pool is preferred to dry storage because of the intense decay heat, small pool footprint, and shorter storage period.

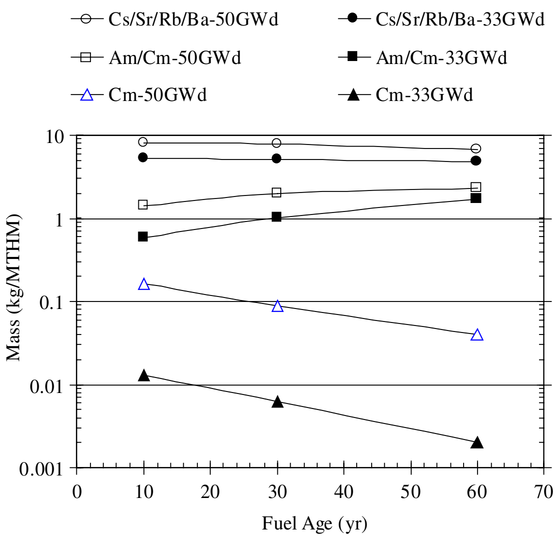


Fig. 1. Mass quantities of high-heat radionuclide products requiring storage (from ORIGEN2).

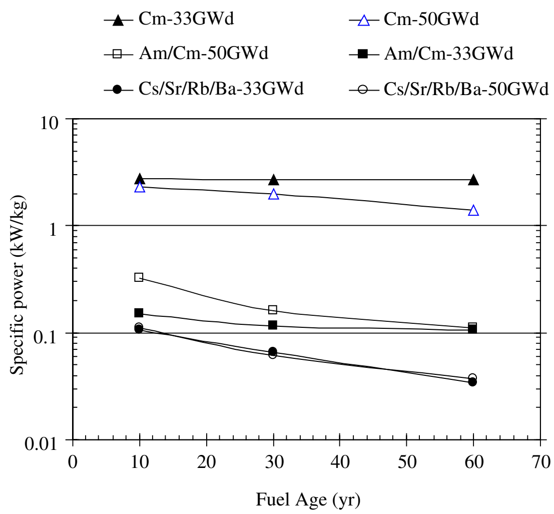
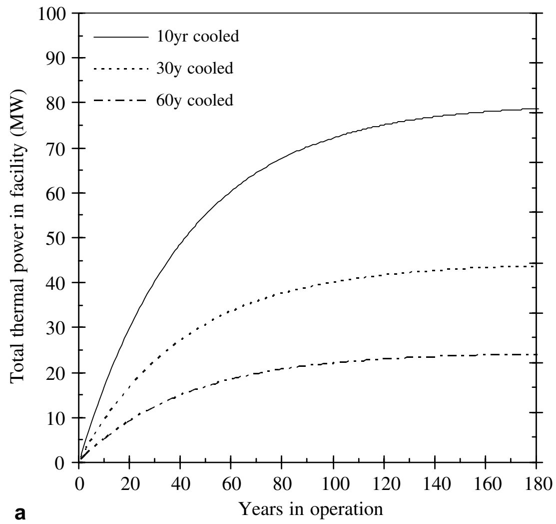


Fig. 2. Specific thermal power of radionuclides for 50-GWd/MT and 33-GWd/MT fuel.

2. Methodology

Unless stated otherwise, the reference fuel is from a pressurized water reactor (PWR, 4.25% enrichment and 50 GWd/MT burnup) cooled 10 years prior to processing. ORIGEN2 calculations were performed in-house. The maximum diameter of right, cylindrical storage canisters is given in Ref. [1] and repeated here in our discussion.



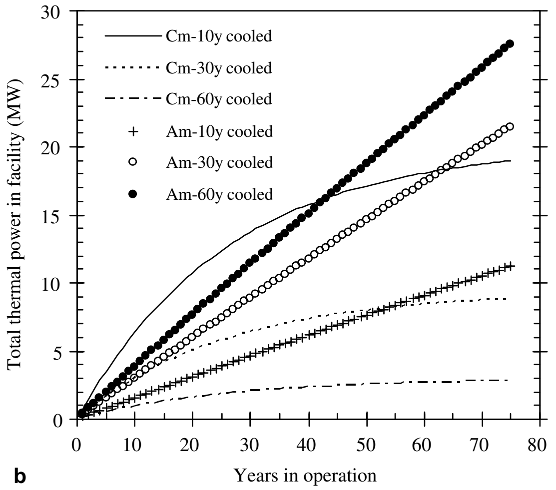


Fig. 3. Thermal power from the decay of engineered storage products for (a) cesium/strontium and (b) americium/curium during the operation of the storage facility. Assumes 2000 MTHM processed per year with a constant accumulation of material from 50-GWd/MT PWR fuel.

2.1. Facility scale

Because the design of a decay storage facility will be similar in many aspects to radioactive waste facilities, we can compare decay storage concepts with current radioactive waste facility design and philosophy. The design limit of a decay facility is expected to be the thermal power density, so we estimate the footprint using Eq. (1)

$$\text{footprint } [\mathrm{m}^2] = \frac{2000\dot{P}f_{\mathrm{mass}}}{R_{\mathrm{thermal}}}, \tag{1}$$

where R_{thermal} is the thermal rating for the facility in terms of $\mathrm{kW/m^2}$, $\dot{P}$ is the specific power of the radionuclide stream in kW/kg (Fig. 2), f_{mass} is the mass output of the radionuclide stream in kg/MTHM (Fig. 1), and the factor of 2000 accounts for 2000 MTHM processed per year.

2.2. Storage pools

Spent nuclear fuel assemblies are routinely discharged from the reactor and transferred under water to a spent fuel storage pool. Pools are kept at approximately 40 °C by active cooling. At shutdown, the thermal power is approximately 1600 kW (from ORIGEN2) per assembly, with 60–80 assemblies removed per cycle. Pools are 100–3000 m^3 and vary in capacity from 3 to 6 $\mathrm{MTHM/m^2}$, where storage limits are dictated by criticality, space, and decay heat [2]. We assume storage capacities of 5 $\mathrm{MTHM/m^2}$. The thermal rating, 5.6 kW/MT, is from ORIGEN2 calculations for 5-year cooled fuel (50 GWd/MT). Thus, $R_{\mathrm{thermal}} = 28\ \mathrm{kW/m^2}$.

2.3. Dry cask and vault storage

An alternative to pool storage is to store the material in dry storage casks or vaults. Each system has been in safe, reliable service for spent fuel and/or low-level storage in many countries. Dry-storage casks and vaults provide a passive means of cooling, which can decrease cost and increase reliability over the long term.

Spent fuel casks are rated according to thermal output primarily to avoid excessive cladding temperatures. We assume casks (2.08 m diameter) are stored with a 3.5-m pitch and NRC-approved 40-kW capacity. Thus, at 12.25 m^2 per cask plus an additional 25% to account for service aisles, we arrive at $R_{\mathrm{thermal}} = 40\ \mathrm{kW}/(12.25(1.25)\ \mathrm{m}^2) = 2.61\ \mathrm{kW/m^2}$.

Vault storage units are massive bunkers for multiple waste products or spent fuel casks. Many low-level waste (LLW) facilities are of this type, as they are generally reserved for large-volume needs. One system, the NUHOMS modular vault design, is claimed to reduce the dry cask footprint by 30% [3].

2.4. Borehole (small tunnel) storage

One concept [4] describes a series of long, small-diameter boreholes drilled horizontally into a drift to create a large radiator-type facility. In Canada, they have evaluated a similar design for low-level and intermediate-level waste, where small tunnels are drilled into the soil from below-grade access ramps [5].

In our analysis, we assume high-heat radionuclide canisters are packed linearly within a system of horizontal boreholes drilled into the subsurface. The radial dimension of the canister is as discussed in Ref. [1] for air-cooled canisters ($r_{\mathrm{max}} = 0.18$ m for 10% Cs/Sr/Rb/Ba loaded in alumino-silicate zeolite). Inlet and outlet vents in the borehole permit natural convective cooling across the air gap between the canister and the borehole wall. Each canister would contain a mass of radioactive material m_{rad} given by

$$m_{rad} = \pi r_{max} H f_{loading} \rho_{rad}(1-p), \tag{2}$$

where r_{max} is the radius of the storage material (excluding canister), f is the volume fractional loading of radioactive material in the storage material, ρ_{rad} is the density of the storage material, and p is the porosity of the storage material (assume 20%). The reference height H of the canister is arbitrarily set to 10 times the diameter. Minimum borehole lengths L are

$$L = 10 \times (2r_{max}) \times N, \tag{3}$$

where N is the number of canisters per shaft. The number of canisters packaged per year depends on the product stream and the storage form composition (Fig. 4).

The thermal rating can be estimated from the minimum heat flux from Yucca Mountain, 137 kW/ha, which is based on conductive cooling through the drift. The heat flux at a boundary is $k\frac{dT}{dR}$ for conduction and $h\Delta T$ for convection. If we assume conduction through the earth, we may observe a temperature drop of $\frac{dT}{dR} =$ 20–200 K/m and $k = 3$ W/(m K) or $k\frac{dT}{dR} = 60$–600 W/m^2. For natural convective cooling, we assume $h = 6$ W/(m^2 K). For a calculated temperature drop of $\Delta T = 140$ K (440 K surface temperature for Cs/Sr/Rb/Ba loaded to 10% in zeolite and cooled by air at 300 K) the heat flux $h\Delta T = 840$ W/m^2. We will assume convective cooling is 10 times more efficient than conduction or $R_{thermal} = 137$ kW/ha × 10 = 1370 kW/ha or 0.137 kW/m^2.

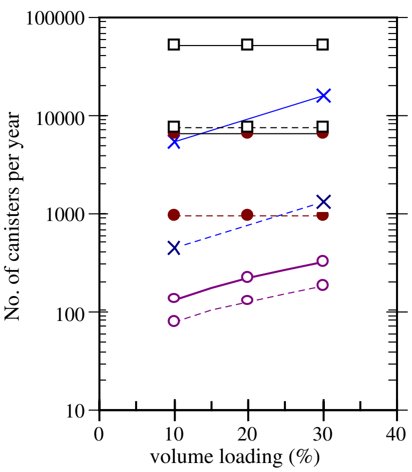


Fig. 4. Yearly canister production rates for decay storage material from 50-GWd/MT (solid lines) and 33-GWd/MT (dashed lines) fuels. Cm oxide (□), Cm oxide in UO_2 (×), Am/Cm oxide (●), and Cs/Sr/Rb/Ba in zeolite (○) are shown.

2.5. Gamma-ray dose

The gamma intensity from ^{137}Cs is calculated from

$$I(x) = I_0 e^{-(\mu/\rho)_{earth}\rho_{earth}x} \tag{4}$$

measured in photons/cm^2/s, where $(\mu/\rho)_{earth}$ is the mass absorption coefficient for 662-keV gamma rays in earth $= 0.030 \pm 0.005$ cm^2/g, $\rho_{earth} = 5.15$ g/cm^3, and x is the thickness of earth above an infinite planar source of photons, I_0

$$I_0 = \frac{m_{Cs} f_{137}}{2 \times \text{area}} \frac{N_A}{\text{MW}} \lambda, \tag{5}$$

where m_{Cs} is the mass of cesium in a year of processed fuel = 8160 g; f_{137} is the fraction of cesium that is ^{137}Cs ($f_{137} = 0.45$); area is the footprint of the borehole space per year of storage = 6500 m^2, MW is 137 g/mol; N_A is Avagadro's number; λ is the decay constant for ^{137}Cs ($\ln 2/30.1y$), and the factor of 2 divides the source strength in half for a skyward fluence rate. At a depth of 6–8 m of earth, the dose is reduced to approximately 10 μR/h.

3. Results

3.1. Engineered product composition

For Cs/Sr/Rb/Ba storage, we must ensure safe interim storage for up to 300 years. Afterward, the storage form should be suitable for direct disposal as Class C LLW[1] without additional packaging or conditioning. Cesium and strontium oxides or salts tend to be hygroscopic, a deleterious property when a dry form is required to reduce radiolysis products and canister pressurization. For this and other reasons such as melting point and unit operations, the most favorable option is a product diluted in a stable adsorbent. Although a decision remains, storage within an alumino-silicate zeolite is attractive. The alumino-silicate product can then be fabricated to a convenient dimension, where the maximum radius is determined by the volume fraction of radioactive material (Fig. 5).

For a combined Am/Cm product, a pure form is most desirable. This material, after decay storage, will need to be conditioned for transmutation so additional bulk material and impurities would complicate the conditioning process and increase cost. Thus, a pure oxide is most favorable. From Ref. [1], a combined pure oxide product has a much smaller maximum diameter compared with the Cs/Sr/Rb/Ba product, 0.0094 m at 50 GWd/MT and 0.013 m for 33-GWd/MT from 10-year cooled fuel. From 60-year old fuel, product dimensions are 0.016 m

[1] Calculations show that a time period of approximately 300 years is necessary for ^{137}Cs and ^{90}Sr levels to reach 4600 Ci/m^3 and 7000 Ci/m^3 limits, respectively, for Class C waste.

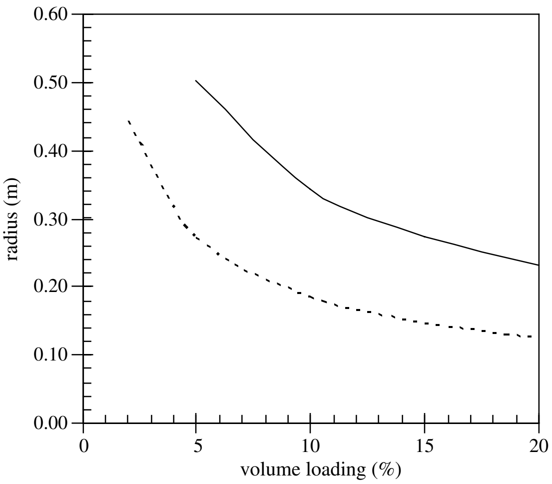


Fig. 5. Maximum radius for Cs/Sr/Rb/Ba products from 10-year (dashed line) and 60-year fuel (solid line) for various waste loadings (50 GWd/MT fuel). The difference between 50 and 33 GWd/MT (not shown) is small. *Note*: 10% by volume is equivalent to 21% by mass.

for 50- and 33-GWd/MT fuel. But, similar to nuclear fuel pin fabrication, the small Am/Cm elements can be stacked into long pins for storage.

Within the AFCI program, it has not been decided whether to separate curium from americium for storage. If curium is isolated for decay storage, the maximum radius of the pure oxide would be prohibitively small to avoid surface temperatures >700 K (0.004 m diameter [1]). Unless thin, plate-type storage forms are used, this material would have to be diluted in a suitable matrix for decay storage. Depleted uranium or zirconia may be appropriate. In France, they have considered diluting curium in an inert matrix fuel [6], storing it until complete decay of ^{244}Cm, and then introducing it into a transmutation reactor with little pre-conditioning necessary.

3.2. Production-scale output

The AFCI will produce as much as 16300 kg of combined Cs, Sr, Rb, and Ba and 2860 kg of Am and Cm (320 kg of which is Cm) at full deployment. It is important to point out the small volume of radioactive material requiring storage compared with current spent fuel volume and LLW production rates. Less than 300 m^3 of cesium and strontium-loaded zeolite will require storage per year compared with 40183 m^3 in LLW waste volume in the US, of which 360 m^3 was Class C waste in 1998 [7]. The Am/Cm oxide volume is 0.3 m^3 per year.

3.3. Storage facility options

In our discussion of storage facility options, we eliminate facilities from consideration where retrieval and natural convective cooling systems are not possible. These include trench designs often used for LLW and silos like the Swedish and Finnish designs for HLW [8].

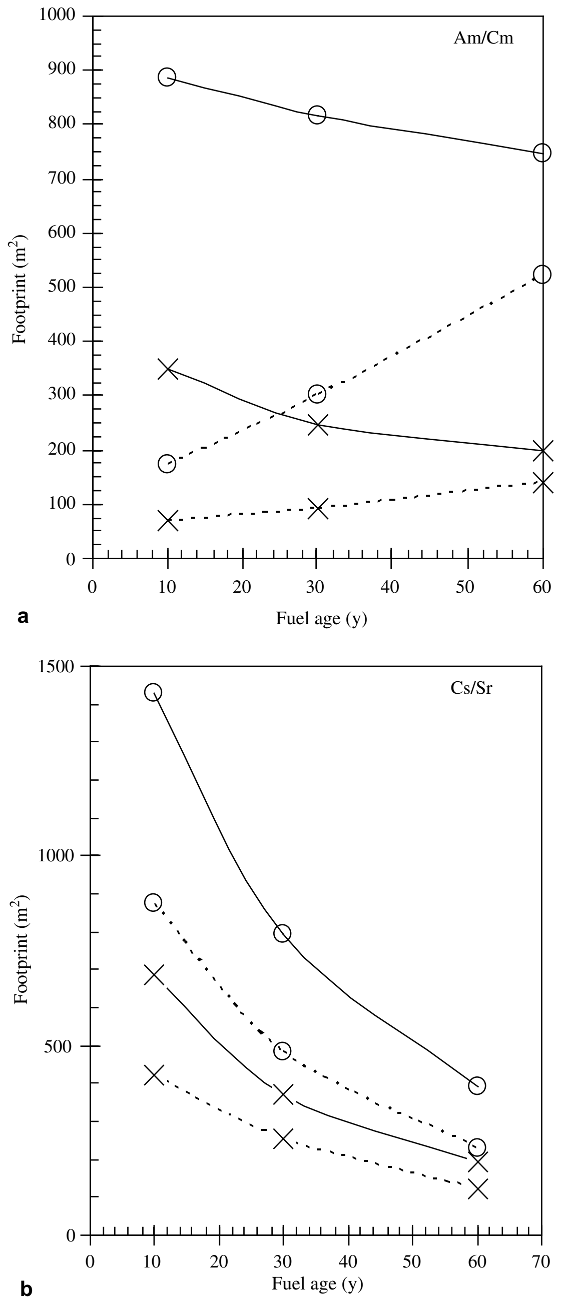


Fig. 6. Extrapolated lifetime pool capacity (○) and yearly dry storage area footprint (×) for (a) Am/Cm and (b) Cs/Sr storage products (50 GWd/MT solid lines, 33 GWd dashed lines). Lifetime pool storage period for Cs/Sr is 30 years (see text for explanation).

3.3.1. Americium/curium forms

For short-term storage, water storage appears to be best. The technology has a long history of safe operation and stable economics. The water coolant provides excellent radiation shielding for neutrons and very efficient cooling, resulting in a small storage pool. Because the anticipated storage period is short (<50 years), concerns over stainless steel corrosion are low, and maintenance costs of the pool may be offset by the small facility footprint. Importantly, though, special provisions are necessary to remove the tremendous buildup of helium from alpha decay of the product. For the combined stream, the required pool size is 890 m^2 for 50-year capacity (Fig. 6(a)) or 600 m^2 for Cm (not shown) from 10-year cooled fuel.[2] In comparison, a dry storage system consisting of Am/Cm in dry casks would require a 350 m^2 footprint per year or 17 500 m^2 over 50 years due to the high heat density (Fig. 6(a)). Clearly, this is a dramatic difference in favor of pool storage. An above-ground facility is recommended.

3.3.2. Cesium/strontium products

3.3.2.1. Stage 1 storage. For cesium and strontium products, a two-stage storage concept is proposed. Pools can be used to store the fresh and most heat-intense Cs/Sr/Rb/Ba products for approximately 30 years, thereby reducing the size of a longer-term dry storage facility by roughly one-half. A maximum pool capacity of 1400 m^2 is needed for this purpose (Fig. 6(b)). Because of the short-term decay period, an above-ground pool is recommended to reduce cost.

3.3.2.2. Stage 2 storage. For long-term storage, maintenance operations are the dominant factor in decision-making [2]. For this reason, passive, dry storage systems are preferred. We must consider also the consensus paradigm in radioactive waste management as it should apply to a storage facility – we should limit the burden of this generation's decay storage products on the next generation. This ethical viewpoint teaches us not to rely on the stability of government for monitored long-term storage (approximately 300 years for cesium/strontium before emplacement in a LLW site), which leads us to recommend that a dry facility be located underground. An underground facility offers better security and the possibility of safe closure during political uncertainty. Moreover, if the facility is sited in a locale that can be permitted as a LLW site in the future (the NRC and public may require it), then environmental safety can be further guaranteed in case of an early forced closure. The facility certainly need not be located in deep caverns such as the practice for HLW, spent fuel, and greater-than Class C waste, but can be near the surface, analogous to Class-C LLW waste sites. Calculations show that a soil depth of 10 m is sufficient to make the surface dose negligible. The question then becomes 'what kind of underground facility should be built for the bulk of cesium and strontium products?' One can envision several design concepts that offer natural convective cooling to dissipate the high heat production, reduce costs over forced cooling, and guarantee retrieval. Designs include underground caverns housing dry storage casks or underground vaults, or an open tunnel storage system.

3.3.2.2.1. Dry cask and vault storage option. A large, open cavern can house dry storage casks. Based on spent fuel cask thermal ratings, the size of the concrete pad needed to house casks containing high-heat radionuclides would be manageable (Fig. 6(b)). Compared with a football field (5000 m^2), the pad size for a year's capacity of cesium/strontium is small, 200–690 m^2. The 300-year storage capacity required for Cs/Sr is 58 000–200 000 m^2. A system of underground vaults would be slightly smaller in size than an above-ground vault system or dry cask system. For example, the footprint of NUHOMS vaults (Transnuclear, Inc.) are approximately 70% that of a comparable dry-storage cask system [3]. There is no reason to believe that the NUHOMS system could not be implemented below-ground.

The major drawback to the cask or vault system is the large capital cost for excavation of the large open caverns needed to house the products. Certainly, caverns with many years of storage space would be expensive, but economy of scale should improve unit storage costs. To reduce the upfront capital costs, we consider a modular system that allows for cessation of storage activities prior to reaching storage capacity with limited capital investment.

3.3.2.2.2. Borehole (small tunnel) storage option. A series of horizontal tunnels can contain rows of storage products much like the drift system described for Yucca Mountain. But, unlike Yucca Mountain, there is no reason why the tunnel diameter cannot be much smaller since the Cs/Sr storage products are quite small (diameter <1 m). Moreover, the depth of the holes need not be similar to Yucca Mountain because of the drastically different isolation periods. Drilling small-diameter horizontal or vertical holes into a suitable near-surface geologic formation can be an attractive option (see Fig. 1 in Ref. [5]). Studies suggest that upfront costs for this type of facility would compete with LLW trench designs, a significantly lower cost over cavern excavation [5]. To establish natural convective cooling, a number of vent holes would be connected and the earth would provide radioactive shielding and security.

[2] This analysis does not include criticality. With the thermal density being much higher than spent fuel, high-heat radionuclide products will have a much higher pitch than spent fuel, so k_{eff} is expected to be much smaller than for spent fuel. Criticality calculations will be needed to verify that preventative measures such as borated pool water or steels are not necessary.

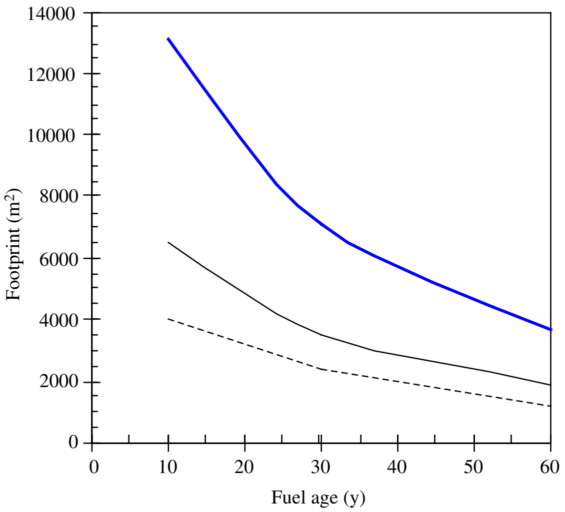


Fig. 7. Horizontal borehole facility footprint based on thermal density of Cs/Sr storage product. Products from 50-GWd/MT (thin, solid line) and 33-GWd/MT (dashed line) fuels are compared with footprints for a facility without stage-one pool storage (thick, solid line).

Storage of cesium and strontium-loaded zeolite would require 0.5 and 0.6 km of borehole space annually for a product loaded to 5% and 30%, respectively (50 GWd/MT fuel). Spacing between boreholes can be estimated from the minimum heat density values for Yucca Mountain (137 kW/ha) as explained in the methodology. Then the maximum storage facility footprint can be estimated (Fig. 7). Borehole space for a single year of cesium/strontium product could occupy as much as 0.65 ha or 6500 m^2. For example, a series of 10 boreholes 50 m long and spaced 15 m apart would accommodate a single year of product (132 canisters).

An additionally attractive feature of this design is the possibility of re-licensing areas of the storage facility as a LLW site as old products reach Class C limits. The stable product form, canister integrity, depth of the facility, modular design, and monitoring capabilities are consistent with Class C waste site designs. Indeed, the NRC and/or public may require that the storage site be located in an area suitable for a LLW waste site as further protection against early or forced closure.

4. Summary and conclusions

We compared the scale of wet and dry storage facilities for cesium/strontium and americium/curium engineered products. Storage in above-ground pools much like those used for discharged spent fuel is well-suited for americium/curium products. A small pool roughly the size of average spent fuel pools operating at US nuclear power stations is needed for a 50-year capacity.

For cesium/strontium, a two-phased storage approach has attractive features. The hottest products would be stored in a single 1400-m^2 pool for 30–50 years and then transferred to an underground dry storage facility for long-term storage. A shallow-depth facility constructed of passively cooled boreholes has attractive features as well as a potential for significant cost savings over an underground cavern facility. Designs consist of a series of horizontal holes totaling 0.5 km in length. An important consideration for further study is regulatory ruling. It is important that proposed designs be conferred to the NRC with sufficient time to establish the regulatory guidelines needed for this unique storage concept.

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