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ABSTRACT

This paper presents bounding pressure and flammability evaluations for DOE Standard Canister loaded with DOE-managed aluminum-clad spent nuclear fuel (ASNF). The objective of these evaluations is to gain confidence in the safety and feasibility of possible loading configurations for extended (>50 years) periods of dry storage, with particular focus on the dry storage canister pressures and potential for formation of a flammable atmosphere. The primary concern about the extended dry storage of ASNF is radiolytic gas generation. The aluminum cladding of these materials tends to corrode, and these corrosion products—typically aluminum oxides, such as boehmite, bayerite, or gibbsite—could carry water. This makes ASNF dry storage canisters difficult to dry. The gamma radiation field in dry storage environments could cause a radiolytic breakdown of residual water, forming chemical species such as molecular hydrogen (H_2). The release of these species could increase the canister pressure and lead to the generation of a flammable canister atmosphere. The bounding evaluations presented within this study surmise conservative, but credible, conditions and processes. This includes the assumption of a full breakdown of a large quantity of free, physisorbed, and chemisorbed water (bound in a trihydrate, i.e., $Al_2O_3 \bullet 3H_2O$, layer). The considered dry storage configurations include a ~3 m (10 ft) long, ~46 cm (18 in) diameter (10 × 18) DOE Standard Canister loaded with 32 Advanced Test Reactor (ATR) ASNF elements, and a ~3 m (10 ft) long, ~61 cm (24 in) (10 × 24) diameter DOE Standard Canister loaded with 40 ATR ASNF elements. While the results of this study indicate the possibility of atmospheric hydrogen concentrations above the lower flammability limit, insufficient concentrations of oxygen will prevent the formation of flammable atmospheres. The maximum credible pressures remain well within the structural limits of the DOE Standard Canister.

INTRODUCTION

DOE manages more than 13 metric tons of heavy metal of aluminum-clad spent nuclear fuel (ASNF). Much of this material is stored, either wet or dry, in aging facilities, such as Idaho National Laboratory's (INL's) CPP-603 facility or Savannah River Site's L Basin. In recent years, DOE-sponsored technology development initiatives are underway to enable safe, extended (>50 years) ASNF dry storage in a range of novel configurations [1, 2]. One configuration under consideration is ASNF dry storage in DOE Standard Canisters [3].

The DOE Standard Canister

The DOE Standard Canister (Figure 1) is a family of sealed, standardized, stainless-steel dry storage canisters designed to accommodate a range of DOE-managed spent nuclear fuel types, including ASNF generated by the Advanced Test Reactor (ATR). Current design variations include cylindrical canisters in two different lengths (3 or 4.6 m, i.e., 10 or 15 ft), two different outer diameters (46 or 61 cm, i.e., 18 or 24 in.), and two different cylinder wall thicknesses (9.5 or 12.7 mm, i.e., 3/8 or 1/2 in.). The canister body is manufactured with a welded bottom lid. After canister loading, the top lid is welded to the cylindric canister body at the location of the backing ring. A vent port in the top lid enables canister conditioning after closing. Lid skirts function as impact limiters and lifting rings are attached to the skirts for canister hoisting. Impact plates protect the lids from damage in a canister drop scenario. The maximum gross weight of the design ranges from 2,270 kg (5,005 lb) to 4,540 kg (10,000 lb). This extremely robust standardized system was rigorously tested, simulated, and evaluated, demonstrating the ability to withstand a range of extreme accident scenarios without confinement failure, such as an impact from a free fall height of 9 m (30 ft) [4, 5, 6].

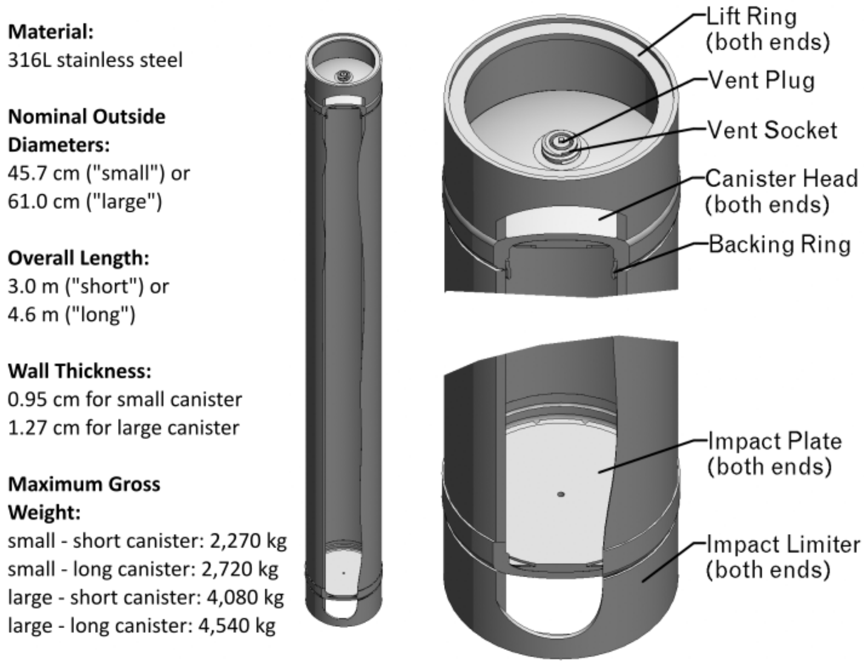

Figure 1. Sketch of the DOE Standard Canister design.

ASNF Dry Storage—Challenges

Because the cladding of aluminum-based fuel is susceptible to in-reactor and post-operational corrosion, aluminum oxides layers can commonly be found on these materials. These layers—which are typically a mix of boehmite, bayerite, or gibbsite—hold significant amounts of physisorbed or chemisorbed water, making ASNF dry storage canisters difficult to dry. The key challenge to extended dry storage of ASNF is radiolytic gas generation associated with the breakdown of residual water within the canister due to exposure to a strong gamma radiation field. This breakdown typically yields gaseous species, such as hydrogen (H_2). Thus, the formation of a flammable canister atmosphere or canister overpressurization (in sealed systems) is of major concern.

DOE Office of Environmental SNF Technology Development Efforts

To address these concerns, and to develop a technical basis for extended dry storage of ASNF, the DOE Office of Environmental Management (EM) initiated technology development (TD) activities which include expansive radiolytic gas generation bench scale experiments and predictive modeling and simulation. These efforts, completed by INL in close collaboration with the Savannah River National Laboratory, point to unproblematic canister conditions [7, 8, 9, 10]. Conclusions drawn from previously completed, bounding hand calculations align with these findings [11].

Novel ASNF Dry Storage Configurations and Findings—Need for Reevaluation

A recent redesign of the DOE Standard Canister internals enables denser ASNF dry storage configuration [12]. Specifically, these new ASNF baskets allow loading 32 ATR ASNF elements into a 10 × 18 DOE Standard Canister and loading 40 ATR ASNF elements into a 10 × 24 DOE Standard Canister. These changes require a reevaluation of the safety of ASNF dry storage in DOE Standard Canisters. This includes the computation of the bounding canister pressures and the potential for the formation of a flammable atmosphere. Further, the extensive efforts completed within the scope of the DOE-EM Spent Nuclear Fuel TD efforts provided deep insights into the corrosion processes and expected conditions within ASNF dry storage conditions, including data on expected oxide layer thickness, oxygen concentration, and gas temperature [13, 14, 15, 16]. The following reevaluations—completed under consideration of these latest data points and newly gained insights—will improve the accuracy of the existing bounding estimates and can be used to support making the case for ASNF dry storage safety in DOE Standard Canisters.

METHODOLOGY

Assumptions

The following assumptions are made to compute the bounding pressure, hydrogen concentration, and oxygen concentration in the considered ASNF dry storage configurations. Additional details on the bases for these assumptions can be found in the associated technical report (INL/RPT-22-68212, Rev. 2) [17].

1.) All chemisorbed, physisorbed, and free water is broken down in a radiolytic gas generation process.
2.) The breakdown of chemisorbed and physisorbed water leads to a release of H_2. Oxygen remains trapped in the oxide layers [13]. However, the breakdown of any residual, free water could lead to the formation of oxyhydrogen.
3.) A DOE Standard Canister loaded with ATR ASNF is the bounding case for the range of DOE-managed ASNF, providing a total of 1,152,000 and 1,440,000 cm^2 of potentially corroded ASNF surface in the 10 × 18 and 10 × 24 DOE Standard Canister, respectively (adapted from [11]).
4.) The bounding, average oxide layer thickness is limited to 25 μm [14].
5.) The trihydrate oxide ($Al_2O_3 \cdot 3H_2O$) layer density is 2.43 g/cm^3 and the water content is 34.6% [18].
6.) The DOE Standard Canister drying process removes all free and physisorbed water other than residual steam due to an imperfect vacuum evacuation, before backfilling it with helium (He).
7.) The average, credible gas temperature and DOE Standard Canister shell temperature is 220°C (428°F) [15, 16] during storage.
8.) The considered DOE Standard Canister void volumes, after loading and reduction of a safety margin of 10% of the empty canister void volume, are 0.21 and 0.42 m^3 for the 10 × 18 and 10 × 24 DOE Standard Canister, respectively. These values were estimated based on an ATR ASNF element volume of 2,928 cm^3 [19].
9.) DOE Standard Canister corrosion is equivalent to 10% of the nominal canister shell wall thickness.
10.) After evacuation, the DOE Standard Canisters are backfilled with helium at 27.6 kPag at 21°C [3].
11.) The atmospheric pressure of the DOE Standard Canister storage environment is 50.7 kPa.
12.) There is no credible scenario leading to a DOE Standard Canister confinement barrier breach [20].

13.) Both a minimum hydrogen concentration of 5 vol.% and a minimum oxygen concentration of 4 vol.% are required to reach flammability (adapted from [21, 22, 23, 24]).[1]

The parameters of the associated bounding cases are summarized in Table 1.

Table 1. Considered parameters in the bounding cases.

Canister	**10 × 18**	**10 × 24**
Number of ATR ASNF elements	32	40
Free void volume	211,018 cm^3	418,043 cm^3
Total ASNF surface area	1,152,000 cm^2	1,440,000 cm^2
Canister corrosion	10%	10%
Oxide layer type	Trihydrate	Trihydrate
Oxide layer thickness	25 µm	25 µm
Oxide layer density	2.43 g/cm^3	2.43 g/cm^3
Oxide layer water content	34.6%	34.6%
Amount of physisorbed water	None	None
Amount of free water	None	None
Vacuum quality	3 torr at 25°C	3 torr at 25°C
Helium backfill pressure	27.6 kPa at 21°C	27.6 kPa at 21°C
Atmospheric pressure environment	0.5 atm at 21°C	0.5 atm at 21°C
Gas and canister material temperature	220°C	220°C

Amount of Gas and DOE Standard Canister Pressure

Eq. 1 is used to quantify the amount of molecular hydrogen ($n_{hydrogen}$, in moles) formed due to the radiolytic breakdown of chemisorbed water bound in the corrosion layers (i.e., the aluminum oxide) found on the cladding of the ASNF:

$$n_{hydrogen} = \frac{\rho_{oxide} \times t_{oxide} \times A \times WC}{MR_{water}} \quad \text{(Eq. 1)}$$

In this equation, ρ_{oxide} describes the density of the oxide layer, t_{oxide} is the oxide layer thickness, A is the surface area of the ASNF, WC is the water content, and MR_{water} is the molar mass of water.

The Ideal Gas Law (Eq. 2) can be used to compute the quantity of oxyhydrogen (2:1) ($n_{oxyhydrogen}$, in moles) potentially released due to the full radiolytic breakdown of residual water steam within the canister:

$$n_{oxyhydrogen} = \frac{p_{vacuum} \times V}{R \times T} \times 1.5 \quad \text{(Eq. 2)}$$

where p_{vacuum} is the vacuum quality achieved during dry storage conditioning, V is the DOE Standard Canister void volume (in loaded condition), R is the ideal gas constant, and T is the temperature

[1]Note that a recently completed data review point to H_2-air-He mixture flammability at oxygen concentrations below 4 vol.%. The impact of these findings on the conclusions of the present study has yet to be evaluated.

associated with the achieved vacuum quality. The factor of 1.5 accounts for the quantity of gaseous oxygen (O_2) in addition to the H_2 in the 2:1 (H_2:O_2) mix in the oxyhydrogen.

Further, the Ideal Gas Law (Eq. 5) can be used to compute the amount of helium (n_{helium}, in moles) backfilled into the canister after evacuation:

$$n_{helium} = \frac{(p_{backfill} - p_{vacuum}) \times V}{R \times T} \quad \text{(Eq. 3)}$$

where $p_{backfill}$ is the absolute backfill pressure at the specified temperature T computed under a specified atmospheric pressure at the same temperature, $p_{vacuum}$is canister pressure after evacuation, adjusted for the backfill gas temperature, V is the DOE Standard Canister void volume (in loaded condition), and R is the ideal gas constant.

Eventually, the Ideal Gas Law (Eq. 4) can be used to compute the canister pressurization:

$$p = \frac{\sum n \times R \times T}{V} \quad \text{(Eq. 4)}$$

where $\sum n$ is the total amount of the individual gaseous species within the canister computed with Eqs. (1)–(3), p is the absolute canister pressure at the specified temperature T under the DOE Standard Canister void volume V (in loaded condition), and R is the ideal gas constant.

Permissible Canister Pressure

The permissible canister pressure of the DOE Standard Canister is computed according to ASME BPVC.III.3-2021 at the assumed gas temperature (i.e., 220°C) and considering the parameters listed in Table 2.

Table 2. DOE Standard Canister parameters used to compute the permissible pressure.

Canister	**10 × 18**	**10 × 24**
Material	Stainless steel 316L	Stainless steel 316L
Internal corrosion layer thickness	0.95 mm	1.27 mm
Canister shell wall thickness, corroded, 12.5% tolerance	7.5 mm	10 mm
Canister shell inner radius, corroded	221.1 mm	294.8 mm
Canister head wall thickness, corroded	14.9 mm	19.4 mm
Canister head sphere radius, corroded	454.98 mm	607.7 mm
Canister head diameter at shell interface location, corroded	433.7 mm	578.8 mm
Canister head knuckle radius, corroded	28.7 mm	38.6 mm

RESULTS

Table 3 summarizes the results of the bounding pressure and flammability evaluations. The calculated bounding pressure in the 10 × 18 DOE Standard Canister reached 2,696 kPag, which is higher than the calculated bounding pressure in the 10 × 24 DOE Standard Canister, it remains well below the permissible pressure of 1,729 kPag.

Note that the permissible pressures (3,553 kPag) of the 10 × 18 and 10 × 24 DOE Standard Canister versions are identical, despite their different geometries. This is due to the similar proportions of the geometrical parameters (such as wall thickness to radius) used in the design of both versions.

The total amount of gaseous species is limited to 141 and 181 mol in the 10 × 18 and 10 × 24 DOE Standard Canisters, respectively.

The amount of helium due to canister backfilling is 7 and 13 mol in the 10 × 18 and 10 × 24 DOE Standard Canisters, respectively.

The total amount of generated gaseous hydrogen could reach up to 134 and 168 mol in the 10 × 18 and 10 × 24 DOE Standard Canisters, respectively. The corresponding maximum credibly achievable gaseous hydrogen concentrations could reach 95 and 93%.

Only 0.02 and 0.03 mol of gaseous oxygen is generated, in the 10 × 18 and 10 × 24 DOE Standard Canisters, respectively. This keeps the corresponding oxygen concentration in the atmosphere below 0.01 and 0.02%.

Note that these results and findings have been confirmed through recently completed sophisticated modeling utilizing similarly conservative assumptions [25].

Table 3. Results of bounding pressure and flammability evaluations.

Canister	**10 × 18**	**10 × 24**
Bounding pressure	2,696 kPag (391 psig)	1,729 kPag (251 psig)
Permissible pressure	3,553 kPag (515 psig)	3,553 kPag (515 psig)
Structural integrity concern?	No	No
Total quantity of gaseous species	141.18 mol	181.42 mol
He quantity	6.72 mol	13.31 mol
H_2 quantity and concentration	0.05 * 2 / 3 + 134.41 = 134.44 mol (95.23%)	0.10 * 2 / 3 + 168.01 = 168.08 mol (92.64%)
O_2 quantity and concentration	0.05 * 1 / 3 = 0.02 mol (0.01%)	0.10 * 1 / 3 = 0.03 mol (0.02%)
Flammability concern?	No	No

DISCUSSION

The calculated bounding pressure in the 10 × 18 DOE Standard Canister reached 2,696 kPag, which is higher than the calculated bounding pressure in the 10 × 24 DOE Standard Canister of 1,729 kPag; however, both pressures remain well below the permissible pressure of 3,553 kPag. Thus, no canister overpressurization issues are identified.

The total amount of gaseous species is limited to 141 and 181 mol in the 10 × 18 and 10 × 24 DOE Standard Canisters, respectively. The total amount of gaseous hydrogen could reach 134 and 168 mol in the 10 × 18 and 10 × 24 DOE Standard Canisters, respectively. The corresponding credibly achievable gaseous hydrogen concentrations could reach 95 and 93%. In both cases, these values exceed the hydrogen lower flammability limit of 5%. However, only 0.02 to 0.03 mol of gaseous oxygen is generated. This is equivalent to atmospheric oxygen concentrations of 0.01 and 0.02%, which are well below the required minimum oxygen concentration of 4%. Thus, no canister flammability issues are identified.

The presented computations were made under a range of conservative assumptions. Most importantly, a 25 μm-thick trihydrate layer, and a full radiolytic breakdown of water and release of all available hydrogen as H_2 into the canister atmosphere were assumed. Due to these assumptions made, the computations in the present study yielded hydrogen concentrations well above a lower flammability limit of 5%. Thus, the demonstration of ASNF DOE Standard Canister safety under the presented conservative assumptions relies on the limited generation and availability of oxygen, i.e., the nonflammability of a H_2-air-He mixture in presence of oxygen concentrations below 4% [24]. This safety basis appears to be well supported. For instance, recently completed research efforts support the assumption of oxygen absence the considered ASNF dry storage configurations. I.e., this research indicates that the available oxygen remains trapped within the aluminum oxides [13]. Furthermore, less conservative, sophisticated computer models predict significantly lower hydrogen concentrations but confirm the assumption of a negligible oxygen concentration in the considered DOE Standard Canister ASNF dry storage configurations [26]. Additional supply of oxygen from the environment can be reasonably excluded due to the demonstrated robustness of the DOE Standard Canister confinement barrier, making canister breach a hypothetical, highly unlikely event [4, 5, 6, 20].

Note that the technical report associated with the present work describes a comprehensive set of sensitivity studies. These studies identify the free void volume of the canister after loading, the oxide layer thickness and associated properties (i.e., density and water content), and gaseous canister atmosphere and canister material temperatures as parameters with significant relevance to safety [17].

CONCLUSIONS

Under credible, bounding conditions, no canister overpressurization or flammability concerns are expected during the extended long-term storage of DOE-managed ASNF in DOE Standard Canister.

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