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Considerations in the Drying of Spent Nuclear Fuel for Extended Storage

Robert Sindelar,[1,†] Rebecca Smith,[2] Anna d'Entremont,[1] Christoph Rirschl,[3] Keith Waldrop,[4] Darrell Dunn,[5] Robert Einziger,[6]
[1]*Savannah River National Laboratory, Aiken, SC, 29808*
[†]*robert.sindelar@srnl.doe.gov, corresponding author*
[2]*Idaho National Laboratory*
[3]*GNS Gesellschaft für Nuklear-Service mbH*
[4]*Electric Power Research Institute*
[5]*U.S. Nuclear Regulatory Commission*
[6]*Retired*

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INTRODUCTION

Drying of spent nuclear fuel (SNF) retrieved from wet storage to remove water for sealed dry storage in a canister is necessary to avoid problems due to excessive corrosion of the fuel and canister internal components, radiolytic breakdown of the water into gaseous species, and canister pressurization. Drying SNF to avoid those problems to meet general safety objectives for extended storage [1] has been largely addressed by the nuclear community. Figure 1, reproduced from ASTM International C1553-21, an international consensus standard guide for SNF drying [2], provides the logic for considerations in drying that is centered on the determination and mitigation of these effects of residual waters (post drying) within a dry storage canister.

The recently revised drying standard guide [2] lists typical methods to dry SNF; residual water sources and impacts; measurement of dryness; and dryness criteria. The standard guide is comprehensive and detailed on these topics. A brief summary of key topics is provided in this paper. Drying investigations for two examples of SNF-in-canisters, namely, a commercial cask demonstration with zircaloy-clad SNF from a power reactor, and an experiment for the DOE Standard Canister with mockup aluminum-clad SNF from a research reactor, are highlighted.

FORMS OF RESIDUAL WATER

There are three primary forms of water that could remain within the canister following drying: free or unbound bulk water; physisorbed water; and chemisorbed water. Free water retention, due to incomplete drying or to traps, depends on the condition of the fuel and its geometry, the canister design, and the drying process. Physisorbed (adsorbed) water is present on all surfaces including fuel cladding, canister basket, and other canister internals' surfaces. Physisorption is a reversible process; the molecules of water and the surface do not chemically change [3]. The amount of physisorbed water on the surface varies depending on the number of monolayers of water with the outer layers being less strongly bound than the first monolayer [4]. Cracks, open pores, and corrosion products may hinder evaporation during drying and increase the amount of residual physisorbed water by virtue of additional surfaces and capillary action. Chemisorbed water is chemically incorporated as a hydroxide or hydrate in native oxides of components, or in corrosion products that formed on the cladding or other materials internal to the canister [2].

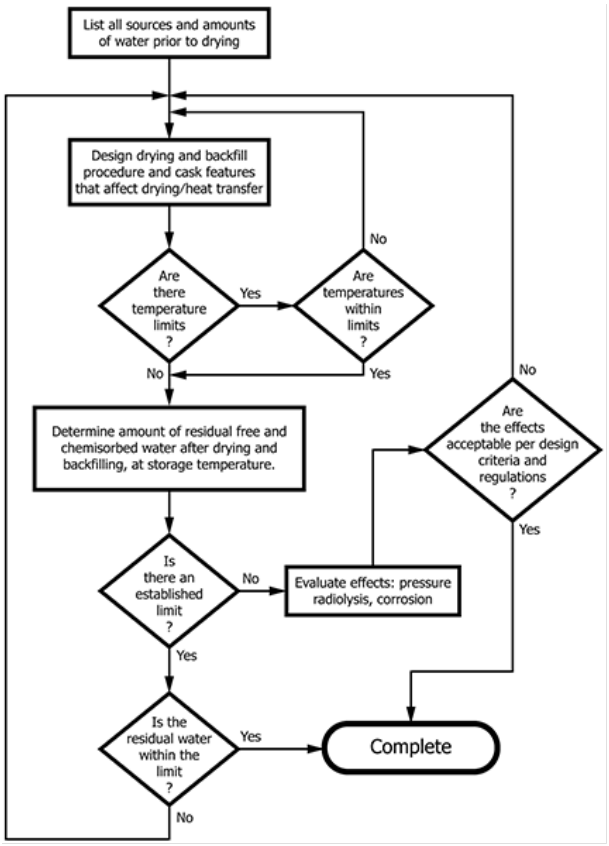

Fig. 1. Flowchart for Evaluation of SNF Drying Acceptability [reproduced from reference [2]].

It is desirable, but not practicable, to entirely remove all these waters in a drying process. Therefore, consideration of the impact of these waters dictates the conditions and rigor of a drying treatment that would be applied.

CONSIDERATIONS IN DRYING SNF

There is no single correct or preferred approach to drying SNF. Each dry storage system vendor applies its own drying procedure specific to an SNF-in-canister configuration. Nevertheless, there are practical and regulatory constraints that must be imposed on drying to meet general safety objectives for SNF storage. To assure cladding integrity, the U.S. NRC imposes a temperature limit for (Zircaloy) cladding of 400°C for commercial reactor SNF; a higher short-term temperature would be allowed if the cladding hoop stress would not exceed 90 MPa [5]. No corresponding temperature limit has been established for DOE SNF; however, a practical limit of 250°C for the aluminum-clad SNF is an example of a suggested limit to avoid phase decomposition and loss of strength of Al6061-T6, a common cladding material [6, 7]. Related considerations include the potential of the drying treatment to impact other components of the canister that, if to be used as a multi-purpose canister, is never to be opened and includes components with functions to be credited for transportation and disposal service.

The important considerations in drying are characteristics of the fuel assemblies and canister internal components such as their surface areas that can hold physisorbed and chemisorbed water and their physical configuration that can impede or even trap free water from being removed. An example of a drying-impacting fuel condition is the presence of crud deposits that could provide additional effective surface area for both physisorbed and chemisorbed waters and pores acting as water traps (see Figure 2 as an example of crud deposits).

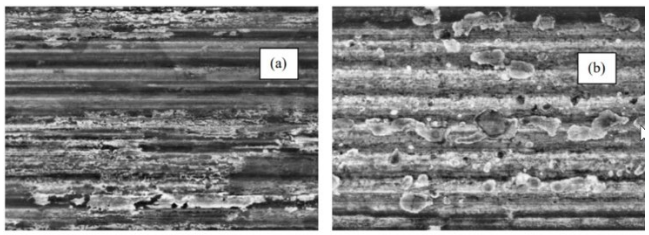

Fig. 2. Examples of observed crud; (a) is noted as a thin peeling crud and (b) is a heavier flaky deposit on top of a thick oxide [8]

Another aspect of drying fuel is to establish two categories of fuel condition, namely "breached" and "intact" [2]. A breached fuel rod (pin) would be a fuel rod that contains a through-cladding defect through which water could enter. The ability to dry such a rod to remove the ingress water is challenging, and no acceptable quantity of ingress water, or the method to dry such a rod, is provided in the standard guide.

As an example of the uncertainty, testing of BWR rods with reactor-induced breaches (>~1 mm holes) and entrained water from service and of BWR and PWR rods with intentional drill hole of 1 mm or 2.4 mm with added water was conducted [9]. The rods with the service-induced holes were judged to have nearly complete removal of the initial estimated 6 to 10 grams of water when the rods were heated to achieve a centerline temperature of ~400°C with vacuum pumping. However, the intentional defect rods with injected water had challenges with the water reacting with the fuel. Recent full-length fuel assembly testing at the University of South Carolina included runs to dry a rod intentionally-defected (1 mm hole) and water-loaded. The rod could be dried if extended heating and drying measures were applied [10]. The information from testing of actual or of simulated breached rods available in the literature would not support assurance that all the ingress water would be removed, and the impact of such water would need to be deliberately addressed if left in the SNF-in-canister sealed configuration. It is recognized that there is an extremely low inventory of modern commercial SNF with failed rods [11].

Another key consideration is the phenomenon of water radiolysis to create products of hydrogen and hydrogen peroxide, or with further decomposition, oxygen. Water radiolysis can occur for bulk water, physisorbed water, and chemisorbed water, with some oxide systems facilitating the radiolytic breakdown of physisorbed water [12]. The observations of radiolytic decomposition of water in a commercial SNF canister, and with testing of DOE aluminum-clad SNF materials, is included in the discussion in the final sections of this paper.

METHODS TO DRY SNF – VACUUM

Vacuum drying is a common method used for drying commercial SNF for cask storage and for dual-purpose canister storage. The following sequence is an example of a process for vacuum drying [2] in use for drying commercial SNF for cask storage:

(1) Load container with commercial SNF in the water pool basin.

(2) Install the lid.

(3) Remove container from the water pool basin. Water may or may not be drained before lifting the container out of the pool.

(4) Remove some amount of water to allow for thermal expansion of the water for sealing the lid. Complete draining of water; most commercial containers do not have bottom drains, so they are drained by pumping or pressurizing with a gas, often called blowdown.

(5) Seal weld the canister lid or bolt the lid in place (for a bare fuel cask). Drying and backfill can't take place until the pressure boundary is established. Welding and lid bolting would preferably take place with water still in the fuel cavity for shielding purposes.

(6) Some systems are designed such that a long tube may be inserted to the bottom of the container after pumping or blowdown, and the residual free water at the container bottom may be aspirated out.

(7) External heating or the flow of heated gas through the container may be used for some systems, especially if the loaded fuel has low decay heat output.
(8) Attach vacuum system to the container port. This may be through a quick disconnect fitting, although the quick disconnects are sometimes removed to improve the conductance of the vacuum system.
(9) Reduce container internal pressure to less than 4×10^{-4} MPa (3 torr).
(a) To minimize freezing, some processes call for pressure reduction in stages and holding pressure for some time during each stage before reducing the pressure to less than 4×10^{-4} MPa (3 torr).
(10) Close vacuum system valves and verify that vacuum remains stable.
(a) One practice is a 30-min hold time "maintaining a constant pressure" $< 4 \times 10^{-4}$ MPa (<3 torr) to verify adequate water removal.
(b) Stricter requirements may be specified for some systems or some fuel conditions.
(11) If an unacceptable pressure rise occurs, open vacuum valves and continue pumping.
(12) Once a stable vacuum is achieved, close the vacuum system valves, backfill the container with an inert gas (for example, helium), reduce the container internal pressure to less than 4×10^{-4} MPa (3 torr) again, then do a final backfill with an inert gas to the positive pressure specified for the system

METHODS TO DRY SNF – FORCED GAS (HELIUM) DEHYDRATION

Forced-gas dehydration (FGD) is another common method used for drying commercial SNF for dual-purpose canister storage. The FGD process removes residual moisture in a fuel storage container after all of the water that can practically be removed by inert gas blowdown has been expelled.

Fuel drying by the FGD process is implemented by circulating dry, heated non-reactive gas (for example helium or nitrogen) through the storage container loaded with SNF. The fuel is dried initially by water evaporation and subsequently more vigorously by boiling due to concomitant heating of the fuel and elevation of the vapor pressure of water above the operating pressure of the canister in the drying process. An example of drying using FGD for a commercial SNF dry storage canister is summarized in reference 13.

The hot circulated gas is controlled and monitored to promote water boiling and evaporation in the system while maintaining fuel and cladding temperatures below some maximum level. Water carried by the gas must be condensed outside the canister prior to the gas being recirculated, and the water content of gas leaving the canister can be used to monitor the rate of water evolution from the fuel and internal structures.

The FGD system is schematically illustrated in Figure 3. The system is defined by series-connected gas conditioning modules as follows: (*1*) the condensing module, (*2*) the demoisturizer module, (*3*) the gas circulator module, and (*4*) the pre-heater module. The condensing module serves to cool the gas/vapor mixture exiting the fuel storage container to a temperature well below its dew point such that water may be extracted from the gas stream. The demoisturizer module, engineered to receive partially cooled gas exiting the condensing module, progressively chills the recirculating gas to a temperature that is well below the temperature corresponding to the partial pressure of water vapor at 4×10^{-4} MPa (3 torr).

In a classical system of the FGD genre, the moisture removal operation occurs in two discrete phases. In Phase 1, the FGD is configured to remove bulk moisture by operating the pre-heater module to heat the circulating gas and the condensing module to remove the evaporated moisture from the fuel storage container. Phase 1 ends when all of the bulk water in the container cavity is expected to have been vaporized. At this point, Phase 2 operation commences by starting the demoisturizer module and monotonically lowering the vapor pressure of gas in the fuel storage container to at or below the 4×10^{-4} MPa (3 torr) criteria. The demoisturizer module is equipped with a facility to chill dry the circulating gas and plays the principal role in Phase 2 dehydration.

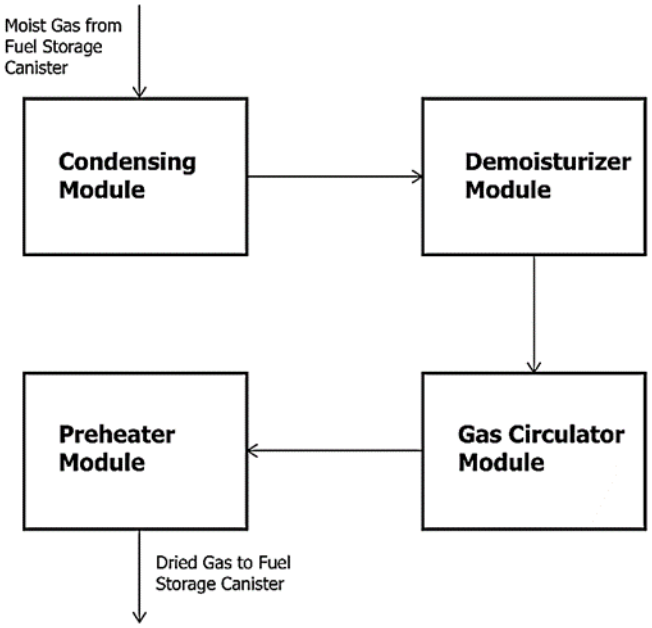

Fig. 3. Process flow for the Forced Gas (e.g. helium) Dehydration drying process [reproduced from reference [2]]. Ultimate dryness in the process is achieved in the Demoisturizer Module by chilling the recirculating gas

MEASUREMENT OF DRYNESS

Residual water detection and measurement inside the canister/cask can be done during the drying or after the drying or both. During vacuum drying operations, measurement is mostly done via pressure gages and pressure rebound tests. During drying operations at atmospheric or overpressure

conditions, chilled mirror hygrometers and capacitive humidity sensors may be used to control and measure the water content in the canister atmosphere. Additionally, condensers used on either the pressure or the vacuum side or both of the connected pumping systems can provide information about the drying status inside the cask cavity.

The residual water measuring devices can be roughly divided in two categories, those measuring the partial pressure of water inside the volume and those measuring the absolute pressure, e.g., different type of pressure gages.

DRYNESS CRITERIA

A typical criterion used to evaluate and measure dryness is the <3 torr, 30 minute pressure [14] that is used for vacuum drying. Using van der Waals ideal gas law and assuming the pressure is entirely from water vapor, 3 torr corresponds to 0.37 moles of H_2O or 6.7 grams of water at 25°C in 2300 L of free volume.

A "falling rate of rise" consideration for dryness was implemented at the Idaho National Laboratory to dry research reactor ASNF with low attendant heat [15]. This method is more involved than performing a simple pressure check at the end of a hold period, but it could show presence of trapped water that would be released at a slow rate.

A comparable quantity of moisture can also be established by specifying the maximum exit gas humidity for the relevant temperature and pressure conditions for the FGD process.

IMPACTS OF RESIDUAL WATER – BULK WATER IN COMMERCIAL SNF CANISTER

The impact of a postulated large volume (up to 55 moles or ~1 liter) of residual free water on commercial SNF and canister internals were evaluated in previous work [16, 17]. A primary thrust of the work was to evaluate the oxidation of the cladding that would occur subsequent to the radiolytic breakdown of water into hydrogen and oxygen. Cladding oxidation models for claddings of Zircaloy-4, ZIRLO and M5 were developed using the rate constants for various cladding alloys in the dry storage temperature range. The results showed the extent of cladding oxidation is no more than 2 μm additional consumption of the cladding metal (Zircaloy-4) even with 10 moles of residual water, indicating that changes in cladding conditions due to water are expected to be negligible.

That work did identify that an initial cladding breach, suggested to be a 1 mm hole, could allow UO_2 pellet oxidation and extension of the initial breach with a flaw instability criterion for flaw extension. Fuel oxidation sufficient to extend the breach could occur even with 5.5 moles of residual water. The results indicate that fuel oxidation vis-à-vis cladding oxidation is more likely under a higher radiation field, i.e., when radiolytic decomposition of the residual water occurs within the first few years compared to several decades, and that the conditions to cause breach extension are increased with fuel and cladding temperatures approaching the peak storage temperature limit of 400 °C.

High Burnup Demonstration Cask – Water Content and Radiolysis Evaluation

The High Burn-Up (HBU) Demonstration Project was initiated by the U.S. Department of Energy to evaluate the effectiveness of fuel drying and the effects of long-term dry storage on high burn-up spent nuclear fuel. As part of the project, samples of the He backfill gas were collected 5 hours, 5 days, and 12 days after completion of drying [18]. At Sandia National Laboratories, the samples were analyzed by gamma-ray spectroscopy to quantify fission product gases and by gas mass spectrometry to quantify bulk and trace gases; water content was measured with several methods. Gamma-ray spectroscopy results indicated no detectible ^{85}Kr, signifying that no failed fuel rods were present after drying. However, water content in the cask was higher than anticipated, with water vapor increasing to ~17,400 ppmv ±10% after 12 days. This is equivalent to about 100 ml of liquid water or approximately ten times the amount of residual water that corresponds to the dryness criterion of 3 torr rebound pressure after a 30-minute hold [14]. As discussed above, this amount of water would not be significant to cladding oxidation.

In addition to molecular water, a high hydrogen content of ~500 ppmv was measured for the 12-day sample as well. A rigorous evaluation was performed [19, 20] of the radiolytic production of hydrogen from estimated free, physisorbed, and chemisorbed sources of water in the HBU Cask. The surfaces hosting physisorbed water included the basket and rails (see Figure 3) in additional to the large surface area of the fuel rods. Physisorbed water on Zr-alloy surfaces was predicted to be by far the most rapid source of radiolytic H_2, due to the assumed energy-transfer mechanism, and thus dominated the short-term estimate [19, 20]. For the case of initial expected physisorbed water content on all surfaces in the canister, and the water vapor, and chemisorbed water, a hydrogen content of 520 vppm was predicted at 12 days. This excellent agreement with the measurement is not considered by the authors of that work to be a verification of the modeling, but it does suggest that the high level of hydrogen in the HBU Demo could have been caused by a radiolysis phenomenon that was not theretofore considered. Additional investigation of radiolytic hydrogen generation from oxidized zirconium has been planned [21].

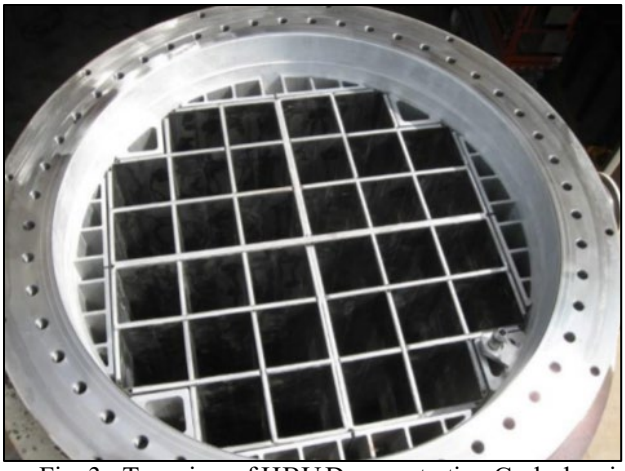

Fig. 3. Top view of HBU Demonstration Cask showing the top of the cask basket and rails. The photo, courtesy of AREVA TN shows cask that EPRI equipped with special instruments to measure the behavior of high burnup fuel

IMPACTS OF RESIDUAL WATER – PHYSISORBED AND CHEMISORBED WATER IN CANISTER WITH ALUMINUM RESEARCH REACTOR SNF

The production of radiolytic hydrogen is expected to be prominent in consideration of extended dry storage of aluminum-clad SNF (ASNF) [22]. Engineering-scale drying tests on ASNF surrogates were performed in a roughly 1/3-height canister mockup to evaluate the effectiveness of vacuum drying and forced-helium drying (FHD) on removal of free and adsorbed water in a realistic canister system [23].

Ten parallel-plate surrogate ASNF assemblies were staged in a single full-size basket in the mock canister, with some assemblies containing single-use aluminum "chemistry surrogate" plates that were corroded in room-temperature water to produce 3–10 μm thick bayerite films [23]. A fixed quantity (105 mL) of liquid water was poured over each assembly to simulate a freshly drip-dried condition, and containers of liquid water were included in some vacuum tests to simulate residual liquid water [23]. Heat sources included heating tapes on the outside canister wall, a heater simulating decay heat inside one assembly, and (for FHD) the heated helium flow [23]. The chemistry surrogate plates were characterized by TGA post-drying to quantify the amount of residual adsorbed water relative to undried control plates [23].

All FHD tests targeted a temperature of 220°C for the fuel surrogates, with target time-at-temperature varying from 6–12 hours (actual achieved temperature and total duration depended on specific test settings of the FHD system and heating tapes) [23]. Vacuum tests compared two vacuum hold durations (cycles with isolation hold times of 5 vs. 15 minutes for successively decreasing steps in pressure), varying wall heating tape setpoints (100–220°C), and preheating using the heating tapes (none, 30–35°C, or 140°C).

For each batch of chemistry surrogate plates produced and for every drying test, at least one plate was preserved and examined as a control. Control plates were handled, packaged, transported, and analyzed in the same manner as the test plates but were subjected to neither FHD nor vacuum drying test conditions. All plates were allowed to air dry after growth of corrosion (to allow weighing) and were handled in air prior to analysis. Surface area normalized TGA mass losses of the respective control plate specimens were nearly the same as many of the vacuum drying test specimens, suggesting that either those tests had little effect on physisorbed water or that re-adsorption of physisorbed water may have occurred between drying test completion and analysis.

Both FHD and vacuum drying processes removed bulk water from the mockup canister [23]. However, there was a clear difference in efficacy for removal of chemisorbed water from the chemistry surrogate plates, with a sharp increase in removal of chemisorbed water for plates that exceeded 220°C during drying, a temperature sufficient to begin thermal decomposition of bayerite [23]. Most FHD tests were able to achieve this threshold. However, vacuum drying tests attained much lower plate temperatures (no more than ~120°C) and consequently removed minimal chemisorbed water.

From mass balance on the storage canister, drying temperatures at and above 220°C stand to reduce the hydrogen gas yield over the duration of storage to the extent that chemisorbed water can be removed before the canister is sealed.

CONCLUSIONS

Drying of SNF for extended storage in a sealed canister is needed to avoid excessive corrosion of the SNF and canister internals, radiolysis, and pressurization that could impact general safety objectives for storage. The ASTM Standard Guide C1553-21 [2] addresses comprehensively the basic issues for drying SNF centered on the considerations of: 1) how dry the SNF needs to be; 2) what drying methods can be used to dry the SNF-in-canister as it is retrieved from wet storage; and 3) metrics for determining adequate dryness.

The many varieties of SNF, their varying post-service conditions, and the many potential dry storage system designs makes a single drying method or dryness criterion impractical. Nevertheless, drying approaches using vacuum drying and using forced gas dehydration have both been proven to be very effective at water removal from various SNF-in-canister configurations to achieve dryness needed for safe extended dry storage with noted expectations of future transportation and repository disposal of the canisters.

For commercial SNF dry storage systems, the "3 torr, 30 minute" dryness criterion [14] can be met; however the actual residual free water may be above that for the corresponding water vapor pressure. For other configurations such as aluminum-clad research reactor SNF, drying to remove physisorbed and chemisorbed waters to a practicable extent to reduce radiolytic H_2 generation is suggested.

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