

Annotated Bibliography for Drying Nuclear Fuel

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September 2011



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**Prepared for the
U.S. Department of Energy
Assistant Secretary for Environmental Management
Under DOE Idaho Operations Office
Contract DE-AC07-05ID14517**

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ACKNOWLEDGEMENTS

The author would like to thank Matthew Marston, Charlie Pennington, Joy Russell, Robert Einziger, and John Abrefah for sharing their insights, experience and constructive comments on this work. Special appreciation also goes to Sandy Birk for her strategic advice on this task and introductions to helpful contacts.

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INTRODUCTION

Internationally, the nuclear industry is represented by both commercial utilities and research institutions. Over the past two decades many of these entities have had to relocate inventories of spent nuclear fuel from underwater storage to dry storage. These efforts were primarily prompted by two factors: insufficient storage capacity (potentially precipitated by an open-ended nuclear fuel cycle) or deteriorating quality of existing underwater facilities. The intent of developing this bibliography is to assess what issues associated with fuel drying have been identified, to consider where concerns have been satisfactorily addressed, and to recommend where additional research would offer the most value to the commercial industry and the U. S. Department of Energy.

THE SEARCH STRATEGY

The supporting literature search was pursued in two distinct phases. The initial phase was mostly limited to the operation of drying nuclear fuel. A broad preliminary request was made of the INL Library, hard copy and electronic personal files were reviewed, a modest internet search was undertaken, and select secondary references were sought. In the first phase, nearly 200 items were deemed relevant for this review. Each was perused and an associated comment was recorded. The search was not exhaustive. Internet sources included NRC, the International Atomic Energy Agency (IAEA) and nuclear community (conference-specific) web sites where responses were in some cases untenably slow. Material of interest may have been obscured by tens of thousands of hits or not recognized at all with the attempted variations of pertinent search terms.

The strategy for the second phase of the literature search was to identify and evaluate common sources referenced in those documents found in the first phase. These common sources provided a more detailed understanding of the issues and in the context of their significance to drying fuel.

In general, several observations were clear. First, drying as an industrial process is a well established field with engineering publications and forums devoted exclusively to the topic: a couple of general sources (cited in the sources reviewed) were identified among the many thousands available. Second, search efforts including “dry”, as well as, or instead of “drying”, tend to include the much broader topic of dry fuel storage without efficient means to distinguish or prioritize between them. Also, the initial search response was overwhelmed with reports of the incremental development of technical data to support the Hanford N-Reactor fuel relocation project.[95] To manage the search in a manner consistent with the requested scope, emphasis was put on items defining, limiting, or defending fuel drying processes. Third, some fuel drying designs and patents came up in the search and are included for completeness, without confirmation of whether such concepts have been successfully implemented. And finally, the topics of corrosion mechanisms and corrosion products arise repeatedly in conjunction with the discussion of drying. The drying process and operational temperature limitations, must consider the constituents and configuration of the materials to be dried. The more general corrosion citations have been included in the context of dryness achieved, safe handling practices, and the effects on cladding or containment integrity, but specific corrosion topics can (and have been) the subject of their own separate reviews.[106][107][165][251]

SYNOPSIS OF FUEL DRYING ISSUES

The issues fall into several categories: regulations and guidance, demonstrations and experience with dry storage; corrosion (rates, mechanisms, microbially facilitated; fuel, cladding, and containment materials; modes of failure), residual water (estimates, waterlogged fuel, tolerance for adsorbed/absorbed/free water), pressurization concerns (radiolysis, decay product, corrosion, thermal excursion), process limitations (burnup-, temperature-, and hoop stress-related), and handling potentially pyrophoric materials. While there is a lot of overlap among these categories, they provide an outline for a discussion of where concerns have been identified and whether they have been satisfactorily addressed.

Regulations and Guidance

In the United States, federal requirements under the jurisdiction of the Nuclear Regulatory Commission (NRC) prompt drying to preserve fuel and cladding integrity (10 CFR Part 72, Section 120 Paragraph (h)(1) and Section 122 Paragraph (d)). Also, guidance from the NRC directs drying as the basis for a long-standing licensing device relied upon by the commercial nuclear industry (NUREG-1536). By contrast, the Department of Energy (DOE) has custody of much fuel for which the cladding is no longer intact. In either case, the most desirable benefit of drying is the mitigation of the liquid phase corrosion environment that enables or accelerates so many of the fuel, cladding, and containment degradation mechanisms.

A consensus standard has been developed to incorporate drying practices that mitigate chemical reactivity and over-pressurization potential, and (to the extent possible) preserve cladding integrity.[25] However, from a regulatory perspective, commercial interests are currently focused on accumulating data to support re-licensing existing dry storage casks. While there has been some discussion of unforeseen quantities of liquid water in the dry storage environment, there is relatively little concern with the efficacy of current fuel drying practices.

Demonstrations and Experience

Dual purpose casks have been developed to satisfy NRC requirements for storage and transportation of commercial fuel. Patents and cask vendor publications illustrate broad system design considerations, and experimental and operational data benchmark the practical experience for various materials and configurations. Several successful demonstration programs have been undertaken.[135] Hanford has loaded, drained, dried and conditioned thousands of multi-canister overpack (MCO) containers. INL has dried, packaged and relocated canisters of TMI-2 fuel debris from underwater to (sealed) horizontal dry storage and has dried and consolidated the inventories of the MTR Canal and CPP-603 Basin to (vented) dry storage in the Irradiated Fuel Storage Facility (IFSF). Several nations have taken similar steps and others have dried and encapsulated fuel for continued wet storage (in a new location).[41][47][441]

The drying methods are varied, but generally favor vacuum drying technology to facilitate the mass transfer, often in combination with a purge or cyclic backfill to improve heat transfer between the heat source and any remaining water. Forced helium gas drying has also gained acceptance.[453]

The determination of vacuum drying endpoint was usually influenced by, if not established outright by the NUREG-1536 [299][300] illustration of drying in accordance with PNNL-6365 [217], calling for evacuation to ≤ 3.0 mm Hg and having the pressure retained through a 30 minute isolation period. The expectation is that any liquid water remaining in the isolated volume would produce a distinct increase in pressure within that time. The forced helium gas drying system establishes dryness based on the moisture content (water vapor pressure) in the outlet gas; this endpoint can be estimated with the number of turnover exchanges of cavity free volume.[396] The perception is that the basis for these criteria for dryness was somewhat arbitrary.

PNNL-6365 documents an analysis of the cover gas constituents measured for representative storage casks in service for < 1 year and shows that reactions over a 40-year operating period would not cause significant cladding degradation for PWR and BWR fuels.[217] Note that the data reflect gas analysis of the in service casks and are intended to show preservation of cladding integrity. These data do not

account for equilibrium water vapor pressure over hydrated corrosion products or physically adhering water within a high surface area particle bed (that might be present in badly degraded fuel or fuel debris but the resultant vapor pressure might take longer than 30 minutes to develop).[105] Neither do these data account for a situation where the vacuum impedes the heat transfer from the heat source to any residual water, although NUREG-1536 Rev. 1 does caution against masking effects of icing adding the suggestion of a staged draw down.[299] Likewise, there could be unanticipated moisture holdup after forced helium drying if occluded internal volume is not at equilibrium with the outlet gas.

Corrosion

Corrosion is a fundamental consideration to wet and dry storage and much attention has been given to estimating rates and establishing mechanisms to enable dry storage and repository performance modeling. The corrosion environment in sealed dry fuel container is a complex equilibrium, so predictions incorporate assumptions that may be overly conservative.

Fuel Corrosion

Oxidation of the fuel must be considered in the context of the fuel material. Consistent with their contribution to the overall used fuel inventory, corrosion of uranium metal, UO_2 , and uranium aluminide fuels were well represented among the items reviewed. Failure mechanisms included the potential for gross cladding rupture with the expansion that occurs with oxidation to the less dense U_3O_8 . Pyrophoric byproducts of uranium metal corrosion (discussed separately) were another consideration. In general, corrosion mechanisms and rates are reasonably well described for temperatures and materials of interest.

Cladding Degradation

Multiple mechanisms have been considered supporting predictions that supported the initial dry storage licenses: primarily creep, stress corrosion cracking, delayed hydride cracking, and oxidation. And significant work has been invested in their study. Within the licensed storage (and transportation) environment, cladding is a redundant means of containment. However, given the history of robust zircaloy cladding performance for commercial light water reactor fuel, there is ongoing interest in the understanding and accurate prediction of cladding failure to enhance confidence in the safety performance of commercial nuclear fuel. Temperature restrictions for drying and dry storage have been instituted to preserve cladding and fuel integrity.

Containment Material Aging

Drying discussions typically focus on fuel and cladding performance, but some consideration has been given to containment throughout storage and transport. The Standard Guide for Evaluation of Materials Used in Extended Service of Interim Spent Nuclear Fuel Dry Storage Systems reflects the current consensus on this topic, complimenting similar efforts in support of repository licensing.[26] However, the fuel and cladding integrity are usually the limiting considerations for drying. Storage system materials such as steel and concrete have been widely studied for general industrial applications. Some work has been done to assess their performance with prolonged exposure to gamma radiation, but in the context of drying containment material aging is not an urgent issue.

Microbially Induced Corrosion

Mostly studied with respect to otherwise inexplicable wet storage corrosion observations, microbes can hasten corrosion. Some thrive in underwater fuel storage pools, and viable biofilms have been confirmed in the high radiation fields on the cladding of used fuel.[51] In the absence of water, microbes are expected to be benign, but they could affect the drying rate or level of dryness attained, and they may be viable to enhance corrosion upon re-wetting. While dry storage is intended to be dry, water ingress events have been detailed for some facilities, and the effectiveness of any drying process can be

circumvented by human error. Additional work in this area should be considered, although perhaps it would be of greater interest with respect to repository disposal than in the context of dry storage.

Residual Water

For some cases, fuel defect models have been employed to calculate residual water after drying.[319] Such water remains in one or more of three forms: chemically adsorbed as waters of hydration associated with oxides, physically absorbed on exposed surfaces, or free liquid water. Waterlogged fuel has been addressed both by calculation and experimentally: it dries more rapidly than a credibly configured particle bed and without gross disruption to the cladding regardless of vacuum conditions applied.[99][219][220][249][253] Estimates of residual water tend to have a large uncertainty that depends heavily on knowledge of fuel condition.

Radiolysis and Pressurization Concerns

Radiolysis of residual water has been assessed by calculation (based on published experimental data) for the DOE standard canister, and was found to be a comparatively small contribution to potential canister pressure by comparison to thermal equilibration and corrosion considerations.[264][463] More recent experimental data suggests that the hydrogen yield for radiolysis may be an order of magnitude less than that assumed for the calculation.[102][174] Depending upon the cover gas, radiolysis may affect the corrosion environment in dry storage, but it does not generate quantities of gas from adsorbed moisture. Back reactions appear to limit the overall product gas pressure.[68] Likewise, decay products have been shown to be an insignificant contributor to gas pressure.[403] Corrosion reactions may consume oxygen or generate hydrogen, or both, depending on the species available, but a thermal excursion (as in an accident scenario) is the greater risk for pressurization.

Process Limitations

High burnup commercial fuel shows some additional vulnerability: to stress corrosion cracking, delayed hydride cracking, and thermal stress-related considerations in general. High burnup produces more fission products and contributes to higher hoop stress on the fuel cladding while simultaneously introducing higher levels of CsI (a source for iodine at the inside cladding surface) and higher initial decay heat.[131] High temperature drying of high burnup fuel (>45GWd/MTU where fuel temperature exceeds 400°C) appears to facilitate the hydride reorientation that can lead to embrittlement of the cladding.[242] NRC regulations have constrained fuel temperatures and thermal cycling (both during drying and in storage), but the nature of the concern is preserving the cladding integrity.[451] High burnup fuels may suffer from competing priorities: thermal limitations implemented to mitigate these cladding degradation mechanisms may lead to a problem of insufficient drying. Forced helium drying appears to address this matter, but appropriate conservatism in the predicted performance needs to be validated over extended storage for high burnup fuels.

Other fuel types must be considered on a case by case basis. Aluminum-clad plate-type UAlx fuels are also subject to certain constraints. Of the major fuel materials, aluminum is most susceptible to general and pitting corrosion, prompting early and ongoing interest in its transition to dry storage. Aluminum fuels are susceptible to creep and diffusion mechanisms at relatively low temperatures, hence drying and storage should be kept to temperatures below 200°C.[392]

Potentially Pyrophoric Materials

The transfer of corroded uranium metal fuels from wet to dry storage presents the distinct hazard of handling a potentially pyrophoric material. Study of this issue covers three areas: 1) the burgeoning understanding of the corrosion conditions favorable to the production of uranium hydride,[98][363][447] 2) the ignition and thermodynamic properties of the materials that define the magnitude of the hazard,[328][381] and 3) the development of a conditioning process to mitigate the hazard.[4][158][433] While the literature on this issue is somewhat limited, the pertinent details are adequately described.

RECOMMENDATIONS FOR FUTURE WORK

Estimates of the amount of residual water with the fuel in dry storage carry considerable uncertainty, particularly with materials degraded in service, damaged during handling, or severely corroded in wet storage. This uncertainty has been accepted, and drying procedures have evolved to improve confidence that water is reduced to a satisfactory level. The usual strategy is to address specific issues with a bounding methodology. However, residual water remains a consideration in the context of the expanding storage interim. The availability of water drives general corrosion, contributing to a complex equilibrium that does not readily lend itself to study. The study of individual contributing gas equilibrium mechanisms (such as radiolysis, corrosion, and water vapor pressure over hydrated corrosion products) enhances confidence in dry storage and the subsequent transportation of these fuels. These have been productive venues for research to support drying spent fuel for interim dry storage as well as transportation and ultimate disposition.

Also, demonstration projects have been extremely useful for validating the storage environment and subsequent fuel performance. In some cases, cover gas analysis and materials examination and testing have been used to this effect.[66][118][217] Such ongoing activities demonstrate progress supporting the increasing duration needed for interim dry storage. Wherever practical to do so, efforts at monitoring and surveillance, and periodic examination of fuel in prolonged dry storage are recommended. The need to demonstrate success or recognize and mitigate the conditions leading to failure is prompted by re-licensing efforts as the interim for dry storage expands in the absence of a firm plan for final disposition.

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- TN-24 cask family and NUHOMS modular dry storage system discussed.
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- Models UO_2 dissolution under conditions expected in a nuclear waste vault (groundwater in a gamma field).
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- Studied formation of radial hydrides in stress-relief annealed Zircaloy-4 cladding. Percent of radial hydrides increases to saturation with thermal cycles.
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- 116 Einziger, R. E., and R. Kohli, "Low Temperature Rupture Behavior of Zircaloy-Clad Pressurized Water Reactor Spent Fuel Rods under Dry Storage Conditions," Westinghouse Hanford Company, HEDL-7400 DE84 007170, (full text), Nuclear Technology, Vol. 67, October 1984, pp. 107-123 (abridged).

Spent fuel rods (Turkey Point PWR) were artificially pressurized to 150 MPa and tested at 323°C for up to 2101 hours. Hydride reorientation was observed.
- 117 Einziger, R. E., and R. V. Strain, Oxidation of Spent Fuel at Between 250 and 360°C, EPRI NP-4524, Project 2062-10, Topical Report, Electric Power Research Institute, April 1986.

Early work to investigate temperature and environment on the oxidation of UO₂ and determine whether the size of a cladding defect affects the oxidation behavior.
- 118 Einziger, R. E., H. C. Tsai, M. C. Billone, and B. A. Hilton., Examination of spent PWR fuel rods after 15 years in dry storage, NUREG/CR-6831, acc: ML032731021, Argonne National Laboratory, August 2003.

Evaluation of Surry PWR fuel after 15 years in dry storage. Peak drying temperatures over 400C, burnup up to 36 GWd/MTU. No degradation or undesirable effects observed (including unacceptable creep and hydride reorientation that could lead to embrittlement).
- 119 Einziger, R. E., H. Tsai, M.C. Billone, and B.A. Hilton, "Examination of Spent Pressurized Water Reactor Fuel Rods after 15 Years in Dry Storage," Nuclear Technology, Vol. 144, Nov. 2003, pp. 186-200.

Peak temperatures of over 400°C were attained for this benchmark test. Very little creep was observed. No additional fission gas release was detected. No evidence of hydrogen uptake or hydride reorientation was found, Little if any cladding annealing occurred during the prestorage performance or extended-storage periods.
- 120 Einziger, R. E., L. Thomas, H. C. Buchanan, R. Stout, "Oxidation of Spent Fuel in Air at 175 to 195°," Presented at the Third Annual International High-Level Radioactive Waste Management Conference, Las Vegas, Nevada, April 12-16, 1991, pp. 1449-1457, revised for publication, Journal of Nuclear Materials, Vol. 190, 1992, pp. 53-60.

Preliminary data indicates quasi-stable formation of U₄O_{9+x} at O/M = 2.4 in air at 175-195°C. Further oxidation to U₃O₈ would be very slow taking over 2000 years at 95°C.
- 121 Einziger, R. E., M. A. McKinnon, and A. J. Machiels, Extending Dry Storage of Spent LWR Fuel For Up To 100 Years, ANL/CMT/CP-96494, International Symposium on Storage of Spent Fuel From Power Reactors, Vienna, Austria, November 1998.

Points to temperature limits for stress-driven cladding degradation mechanisms with concerns for higher burnup fuel (>45GWd/MTU). Discusses relevant conditions for dry storage over 100 year period.
- 122 Einziger, R. E., S. D. Atkin, D. E. Stellbrecht, and V. Pasupathi, "High Temperature Postirradiation Materials Performance of Spent Pressurized Water Reactor Fuel Rods Under Dry Storage Conditions." Nuclear Technology, Vol. 57, April 1982, pp. 65-80.

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- 123 EINZIGER, R.E., STRAIN, R., Behavior of Breached PWR Spent-Fuel Rods in an Air Atmosphere Between 250 and 360°C, Nuclear Technology, Vol. 75, October 1986, pp. 82-95.

Defects in fuel rod segments with lower burnup propagated sooner than those with higher burnup from the same parent rod. Breached PWR fuel rods will not split open from fuel oxidation during 100 years of storage if the rod is not exposed to air until the temperature drops below 230°C. Defect shape may be more important than size in determining time-to-cladding-splitting.
- 124 Elias, E., and C. B. Johnson, Radiological Impact of Clad and Containment Failures in At-Reactors Spent-Fuel-Storage Facilities, Research Project 2062-1 Final Report, EPRI-NP-2716, DE83 900514, Electric Power Research Institute, Palo Alto, California, October 1982.

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- This companion study to [175] addresses the effects of alpha radiation on adsorbed water. The overall equilibrium mitigates pressure buildup in a closed system. Extrordinary measures are not needed to remove adsorbed moisture from the oxide.
- Examines water adsorbed/retained (in increasing quantities) with UO_2 , U_3O_8 , and UO_3 . Complete removal at 650°C is unnecessary. System is tolerant of quantities (several percent sorbed moisture) remaining after drying at 150°C . Observes only slight pressure increase in gamma field due to radiolytic production of hydrogen - and system at least partly compensated with a simultaneous loss of oxygen pressure. Back reactions were also clearly in evidence.
- Demonstrates that efforts to remove all traces of moisture from U_3O_8 are not necessary. Some H_2 was produced, but O_2 was depleted from the initial atmosphere leading to and overall pressure decrease, even for samples with 9 wt. % moisture. Back reactions were evident limiting product gas pressure in the system.
- Discusses degradation mechanisms including cladding oxidation, creep, "hydrogen pickup", delayed hydride cracking (DHC), corrosion effected cracking of steel, and expansion of UO_2 with its oxidation to U_3O_8 (at temperatures above 200°C).
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- Fuel integrity surveillance from gas sampling during and after dry storage demonstration (Castor V/21, Transnuclear TN-24P, and Westinghouse MC-10 Commercial fuel casks at INEL, expanded to include VSC-17 and REA-2023 casks, also). Gas sampling analysis indicates that fuel storage in an inert fill gas is benign. In general, handling operations are more likely to cause damage than dry storage. Only two leaking rods were identified (by Kr-85 detection) in unconsolidated storage. By comparison, about 10 of the consolidated rods began to leak after consolidation.
- Feasibility study highlights the concerns associated with the trend toward higher burnup fuel and the need to dry store (& later transport) such material.
- Possibly a precursor to [158]. BNFL subcontracted with Nuclear Electric to survey metal fuel (Magneox fuel and its transfer from wet to dry storage) experience relevant to Hanford's then-anticipated fuel treatment effort.
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- Discusses CASTOR type cask used in the Federal Republic of Germany, primarily thermal and shielding considerations. No discussion of drying.
- Develops an expression relating metal ignition temperature to specific surface area, oxidation rate, and mass of oxide on the surface. Validates with experimental data for uranium and plutonium.
- Concludes that precipitation of radially-oriented hydrides can lead to axial crack propagation.
- Air and air-argon tests on rods with artificial defect. Suggested the oxidation difference between irradiated and un-irradiated rods was due to FP gas bubble accumulation at grain boundary and FP accumulation in UO₂ matrix. The irradiated rod in argon (low oxygen) showed very little deformation.
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- Detailed description of the original Transnuclear PWR & BWR drying process with analysis.
- Compact amorphous corrosion film barrier on aluminum inhibits further corrosion. Rate varies with environment, but barrier layer thickness is independent. A second crystalline bulk corrosion layer is more permeable. The effect of alloying is on the barrier layer not the bulk.
- Uranium corrosion and fuel element ruptures (open-ended or defected elements) were examined with: temperature, pressure, steam versus liquid water, heat treatment, carbon content of uranium, zirconium content of uranium, cladding thickness, fuel geometry, annular spacings, defect geometry and size, coolant flow, hydriding of Zircaloy components, and irradiation effects.
- Assesses the as-irradiated /as stored cladding condition of available high-burnup PWR and BWR fuel in dry cask storage. In each case the general condition appears to be sound. Suggests value in additional hydrogen migration data.
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