Gap Analysis to Support Modeling the Degradation of Used Nuclear Fuel Canisters

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Welded stainless steel canisters are being used worldwide for dry storage of used nuclear fuel (UNF) assemblies, and the number of canisters in use is steadily increasing. In support of work currently being pursued at Pacific Northwest National Laboratory to understand atmospheric corrosion degradation processes of spent fuel dry storage systems, a gap analysis is underway to assess the state of knowledge for modeling of the long-term degradation of a UNF canister. The analysis is considering all major components of the atmosphere corrosion degradation processes, ranging from contaminant sources and climatic interactions to regional conditions of particle transport and deposition, to microscale effects leading to stress corrosion cracking. The results of this gap analysis will be used to define the R&D pathway to develop an integrated multi-scale atmospheric corrosion modeling capability for UNF in dry storage canisters that can support the safe and reliable performance of these structures for more than 100 years.

I. Background

Nuclear Power Plants (NPP) produce UNF, which is considered radioactive waste under current US policy. Even in the presence of a deep geological repository in the US, it is possible that UNF will be stored at the reactor site or interim storage facilities for periods longer than a century. The Nuclear Regulatory Commission has concluded that UNF generated by any reactor can be safely stored for 60 years beyond the licensed life of a reactor. Furthermore, it has been acknowledged that for "long term storage" of up to 160 years beyond the licensed life of the reactor "one time replacement of the Independent Spent Fuel Storage Installations (ISFSIs) and spent fuel canisters and cask" may be required (Ref 1). At present, the technical basis is insufficient to support long term storage without the replacement of the spent fuel canisters. In addition, through-wall cracking of stainless steel systems exposed to atmospheric conditions has been observed at currently operating nuclear power plants (Ref 2). It is not known if storage canisters will develop through-wall cracks which would allow air ingress and enhanced degradation of fuel cladding and canister during storage.

II. Canister Degradation

During dry storage the degradation process is expected to initiate on the outside of a UNF canister (Ref 3). The onset and progress of material degradation will depend critically on local contaminant concentration, the properties of the contaminant species and synergistic effects when multiple contaminants are present, the type of steel used to fabricate the canister, the canister temperature, ambient temperature and humidity at the site, contaminant deposition rate, residual stress, canister surface flaws, and weld defects. Reliable data, models, and mechanistic understanding are needed to develop methodologies that can predict canister degradation under a variety of weather, pollution, and external factors for up to a century into the future. Moreover, the disparity in length and time scales between degradation mechanisms at the crack tip and depositions of atmospheric contaminants has precluded an integrated understanding of the stress corrosion process.

Recently, Cole et al. (Ref 4) have developed a multiscale modeling approach, or framework, for atmospheric corrosion of metal structures in complex environments using an integrated approach that considers a range of physical scales from regional or global climatic conditions to microscale behavior associated with the surface electrochemical environment leading to local corrosion and pitting. While this effort is focused on the degradation behavior of civil structures by sea salt corrosion, such as buildings and bridges, many elements of this framework can be applied to UNF canister systems.

Figure 1 displays an adaptation of the multi-scale approach by Cole to the case of atmospheric corrosion and degradation of UNF dry storage canisters. The factors influencing the degradation processes have been separated into four major components based on the physical scale, 1) the global to regional climatic conditions, 2) the near surface conditions, 3) the surface reactions, and 4) the stress corrosion cracking process. Each of these components provides a set of parameters that drive the next lower scale. For example, the global and climatic conditions provide seasonal variations in environmental temperature, relative humidity, wind velocity and direction, and contaminant concentration in the vicinity of the storage location. This information would then be used to determine the conditions near a UNF canister,

considering the internal geometry effects on buoyancy-driven air flow within the storage system.
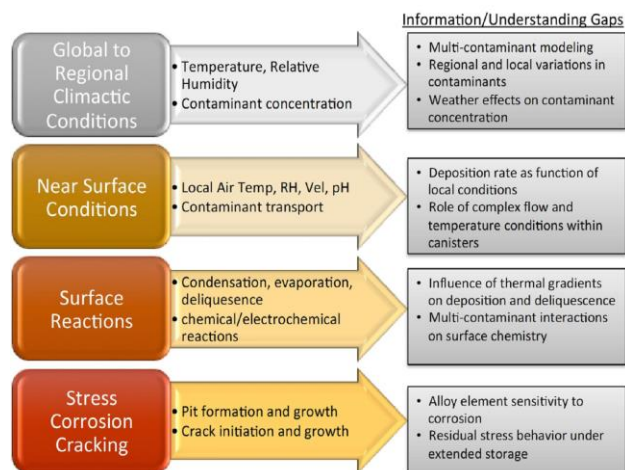

Fig. 1. Outline of Integrated Modeling

Areas that represent potential gaps in the information or understanding of the physics active in each major component are also listed in Table 1. The objectives of the gap analysis are to establish the current state of knowledge within each of these components to inform the R&D efforts to develop an integrated modeling approach.

II.A. Gaps in Knowledge Concerning Canister Degradation

The current focus of research regarding canister degradation is primarily on Chloride Induced Stress Corrosion Cracking (CISCC). This focus is logical because CISCC has historically been shown as a relevant degradation mechanism of stainless steels at NPPs (Ref 5). However, there are still many gaps in our knowledge concerning CISCC of UNF canisters. One particularly important gap concerns exactly how much atmospheric chloride will initiate CISCC within the environment of a UNF canister. Some NPPs are currently using a canister surface chloride threshold of 100 mg/m^2 for CISCC initiation. However, there is some evidence which suggests that surface thresholds of 8 mg/m^2 may initiate CISCC (Ref 6).

CISCC is also strongly dependent on the rate of deliquescence and the existing stresses in the canister. The heat affected zones around the base plate, lid, longitudinal welds, and circumferential welds are candidate locations for CISCC due to the residual stresses. Furthermore, the canister temperature distribution will greatly affect the rate of deposition and deliquescence. It is likely that the canister temperature distributions, near the heat affected zones around the base plate welds in vertical systems, and near the base plate

and lid welds in horizontal systems may be sufficiently low to cause deliquescence during the lifetime of the canister.

Another important factor is regional variation of chloride concentrations and climactic conditions. Currently, much research has focused solely on marine environments (Ref 7). While marine environments will most likely have the highest concentration of atmospheric chlorides, other interior sites may still have sufficiently high concentrations. Furthermore, the local climactic conditions (e.g. relative humidity, ambient temperature) will be a key factor in determining the likelihood of canister degradation at a site.

Contaminants other than chlorides may also contribute to canister degradation. Some of these contaminants may also act synergistically with chloride. This is a cause for concern, especially in areas near coal-fired plants. The sole focus on chlorides is a gap which needs to be addressed. Also, these secondary contaminants will most likely vary regionally, and show different deposition characteristics. Furthermore, where these contaminants deposit may vary based on the type of canister system (i.e. horizontal or vertical).

The rate of canister degradation is also highly dependent on alloy composition. There is usually a broad allowable range for alloying elements. Variations with this range (for instance, low Ni versus high Ni content) may greatly affect the rate of degradation. This factor is often overlooked.

The mechanistic understanding, for modeling purposes, of defect initiation can be determined by various methods. A single reliable method, without excess conservatism, needs to be established in order to consistently predict initiation.

Once initiation has begun, it is important to understand how these defects will grow. Several mechanisms for defect growth have been proposed. There are models that describe progression of chemical degradation over extremely long time periods and predict crack propagation in a degraded canister during transport or in the event of a handling accident. These models need to be refined and integrated so that the degradation processes at different scales are seamlessly coupled within the modeling scheme.

Finally, the lack of an integrated multiscale modeling framework that fills all of these gaps (summarized in Table 1) needs to be addressed in order to consistently predict canister degradation across all current and future storage sites. Such a framework would allow for the prediction of canister degradation based on the site characteristics (e.g. local contaminants, climactic conditions), canister surface temperature, and canister material composition.

TABLE I. Canister Degradation Gaps

Gap	Discussion
Multi-contaminant degradation	Currently, much effort is focused on Chloride Induced Stress Corrosion Cracking. However, other contaminants may also play a key role and some may act synergistically.
Degradation sensitivity due to alloy composition	Various stainless steel alloys may show superior performance in corrosive environments. Also, small variations in the alloys composition may be important.
Canister temperature distribution	The canister surface temperature distribution affects the rate at which contaminants will deposit and the location on the canister where they will deposit. Also, the onset of deliquescence will vary across the canister surface as the temperature varies.
Contaminant concentration changes due to regional characteristics	Local contaminant concentration will vary regionally. The conventional focus on the marine environment may be incorrect, especially when multiple contaminants are considered.
Deposition rate changes due to local climactic conditions	An integrated model must adequately incorporate how local changes in humidity and temperature will affect canister degradation.
Defect initiation	To accurately predict defect initiation requires detailed understanding of how the contaminants act on the surface. The stress which exists in the canister must be accounted for.
Defect growth	An understanding of whether defect growth will accelerate or decelerate with time needs to be established. Also, knowledge about how this defect will grow if an outside force is applied (i.e. during transport or handling) needs to be established.

III. Conclusion

An integrated model which builds on the knowledge base of existing models for predicting atmospheric corrosion of alloys, fuses existing field data on multiple contaminants, includes local climactic conditions, incorporates differences in temperature distribution, and creates a new capability for reliable multiscale modeling of UNF canister degradation is highly desirable. Work is underway at Pacific Northwest National Laboratory (PNNL) to develop such a model and validate it against available experimental data.

ACKNOWLEDGMENTS

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