SPENT FUEL MANAGEMENT CONSIDERATIONS FOR ACCIDENT TOLERANT FUELS

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Abstract

The accident at the Fukushima-Daiichi plant in Japan in 2011 highlighted vulnerabilities in the current zirconium (Zr) alloy clad uranium dioxide (UO_2) fuel to an extended loss of cooling. Improving the resilience of the fuel and cladding is considered a high priority for the nuclear industry and has resulted in significant research into the development of so-called Accident Tolerant Fuels (ATF). ATF are widely expected to be deployed in the near future in existing and future Light Water Reactors (LWRs). Post discharge management and dispositioning of spent ATF is a topic that must be addressed in order to demonstrate responsible management of the fuel cycle and yet has received little attention to date. In this review the spent fuel management considerations of several leading ATF fuel and cladding concepts are assessed against current LWR fuels. The concepts include coated Zr alloys, advanced iron alloys and silicon carbide composite claddings and advanced UO_2 and high uranium density fuels. Technical challenges regarding each different material are highlighted; particularly focusing on reactivity and durability in water. Recommendations are made where variations of current storage procedures are likely to be required.

1. INTRODUCTION

Accident Tolerant Fuels (ATF) are being developed to improve the resilience of LWR fuels under accident conditions. This is being achieved largely through the development of more oxidation resistant cladding materials, although improvements in the thermal conductivity of the fuel can also reduce pellet centreline temperatures and increase the power-to-melt. Such considerations have also led to interest in higher density fuel materials, which despite their higher reactivity to water compared to UO_2, compensate for the increased costs associated with the more robust claddings materials. Whilst the performance of ATF during reactor operations is of primary importance during early stage development, as these concepts mature it becomes important to understand potential impacts on the whole fuel cycle to avoid creating unexpected back end costs and/or leaving the industry exposed to public challenges on issues of responsible management and sustainability [1]. This paper provides an initial overview of the likely performance of ATF during post-irradiation storage and disposal.

The candidate concepts for ATF cladding include a near term option of coating the existing Zr alloy cladding as well as longer term options of Fe based alloys, (drawing on past experience of austenitic stainless steels) and ceramic silicon carbide composites. Candidate fuels include advanced (higher thermal conductivity) UO_2 and higher density fuels including uranium nitride (UN) and uranium silicide (U_3Si_2)

After discharge from reactor, all spent LWR fuel is stored for an initial cooling period in station ponds. The pond water is typically kept at a temperature of below 50°C in order to maintain adequate cooling of the fuel and minimise evaporation. The pond water chemistry is closely controlled to minimise corrosion and is constantly monitored.

There are two principle options for management of spent fuel: disposal or reprocessing. Both options involve interim storage and transport. Fuel can be shipped from station cooling ponds once the heat generation rate has fallen to an acceptable level for transport. This can be as short as a few months to a few years, depending on the fuel type, irradiation and the transport cask design.

LWR fuel can be reprocessed a few years after discharge from reactor. Fuel is typically transferred to storage ponds at reprocessing plants prior to reprocessing as this allows the flow of materials to be buffered and allows optimisation of the materials entering reprocessing. Reprocessing of fuels is not considered in detail in this paper as it entails dismemberment and dissolution of the fuel assemblies. Disposal is, however, considered because it involves emplacement of the fuel assemblies into a disposal facility and therefore the characteristics of the fuel cladding and fuel matrix have an impact on subsequent behaviour and activity release.

Compared with reprocessing, spent fuel needs to be stored for much longer periods of time before the heat generation rate falls to a level considered acceptable for disposal. The minimum cooling period varies with fuel irradiation and disposal concept but is typically many decades. The quantity of fuel being stored and the duration of storage are therefore greater for this option. Long term storage of spent nuclear fuel can be conducted in either in ponds (wet storage) or in dry stores and a range of storage options are available for this [1]. Typically, wet storage is undertaken in similar conditions to those used at power stations. Dry storage

typically involves drying of the fuel and sealing into a container with an inert gas. This leads to higher fuel temperatures during the drying process and during the initial dry storage period, than are experienced in pond storage. Most dry storage systems employ passive cooling and therefore require less infrastructure than pond storage to maintain cooling and hence containment. There are a wide range of potential degradation mechanisms that can affect fuel storage systems and a number of reviews of fuel degradation mechanisms have been conducted in the past 10 years (e.g. [3-5]). For current Zr alloy clad UO_2 fuel the following degradation mechanisms are considered most important in wet storage:

— Uniform (aqueous) corrosion;
— Localized corrosion (Pitting, galvanic, microbial induced corrosion (MIC));
— Hydriding;

whereas the dominant degradation mechanisms in dry storage are [1]:

— Air oxidation;
— Thermal creep;
— Stress corrosion cracking (SCC);
— Delayed hydride cracking (DHC);
— Hydride reorientation; and
— Hydrogen migration and redistribution.

Although operating conditions in LWRs are more severe than in storage the potential exposure times are much longer (and more so in disposal). Testing of newly developed ATF at low temperatures relevant to storage and disposal conditions will be important for evaluating the impact on back-end fuel cycle options.

In geological disposal concepts, fuel is dried and encased in high integrity canisters that provide containment for the fuel and prevent contact between the fuel and groundwater for several thousand years. The disposal container designs associated with well-developed disposal systems are, in general, smaller than those most commonly being loaded with LWR fuel for dry storage.

2. CLADDING

2.1. Coated Zr Alloys

The addition of a protective coating on the outer surface of the Zr-alloy cladding is a relatively simple but effective method of reducing the rate of oxidation of the Zr alloy in high temperature steam that would be encountered during a Loss of Coolant Accident (LOCA). Whilst these coatings are not expected to be able to protect the fuel indefinitely, they are seen as a means of increasing the coping time during which operators may be able to restore cooling and save the reactor from core melt. A wide range of coating materials have been examined, particularly those which form protective chromia, alumina or silica layers when exposed to high temperature steam. Although alumina provides the best high temperature oxidation resistance, it is less stable in reactor operating conditions and therefore most focus is currently on chromium [6]. Cr coated Zr alloys have demonstrated an approximately 10-fold reduction in oxidation due to the production of a Cr_2O_3 protective layer, with commercial irradiation of test rods already in progress [7]. Reduced corrosion also leads to less hydrogen production compared to standard cladding. This has potentially beneficial consequences because hydrogen generated by fuel corrosion during irradiation leads to the formation of zirconium hydride platelets in the cladding. During drying prior to dry storage, the temperature can increase to between 250°C and 400°C. When temperatures are at the higher end of this range, hydrogen dissolution can occur as the solubility of the hydrogen is increased. When cooled, the hydrides will re-precipitate and favour a radial orientation due to the internal pressure of the fuel pins caused by azimuthal stresses. These radial hydrides significantly increase the probability of stress related failure of the fuel pins. In order to mitigate this, the amount of hydrogen allowed in the fabricated Zr alloy is very low (~1.5 ppm) [8] and limits are applied to the maximum cladding temperature during drying and the number of drying cycles that can be used.

Corrosion during pond storage is also expected to be decreased when compared to uncoated Zr alloys. Eventual dissolution of the cladding could lead to localised loss of Cr which could subsequently lead to localised corrosion of the Zr alloy. For a disposal system this is unlikely to be of concern as current performance assessments do not claim credit for cladding integrity.

Dissolution of Cr can lead to the formation of CrO_4^{2-} which is a strongly oxidising species, which could affect the environmental conditions experienced by the fuel matrix if it were to accumulate within a repository. In water CrO_4^{2-}formation is thermodynamically stable at electrode potentials of around 100 mV(SHE) at pH12 to around 600 mV(SHE) at pH7 [9] in the absence of hydrogen peroxide. Therefore, its formation in repository

is unlikely [10], but cannot be precluded without an understanding of expected groundwater chemistry and radiolysis effects.

2.2. Advanced Fe Alloys

Austenitic stainless steels were used as cladding materials in LWRs until the 1980's prior to the widespread adoption of Zr alloys. The preference for Zr alloys is largely an economic one, due to its low neutron capture cross-section. Estimates suggest a cost penalty of 15–35% for steel cladding compared to Zr alloys even with a reduction in cladding thickness to around 300µm [11]. However, Fe based alloys containing Al exhibit excellent oxidation resistance due to the formation of an alumina protective layer, up to temperatures close to the melting point of the alloy (1475°C). Incidentally, under normal operating conditions chromia layers are formed preferentially, providing adequate protection. There are commercial alloys such as Kanthal APMT with the nominal composition of Fe-21wt.%Cr-5wt%Al-3wt%Mo, but other alloy compositions are under development [13].

Spent fuel storage experience of steels (types 304, 304L, 34 and 348H) from five US LWRs, totalling >2000 fuel assemblies was summarised in an EPRI report in 1996 [14]. No unexpected behaviour was noted for these spent fuel assemblies during pond storage over a period of up to 25 years. Uniform corrosion rates during wet storage (<50°C) were predicted to result in a reduction in wall thickness of 15µm over a 50-year period. The behaviour of these materials, and claddings of similar composition, is expected to be similar to other stainless steels (such as those used for UK Advanced Gas cooled Reactor (AGR) fuel cladding). Experience of storing AGR fuel in the UK indicates no discernable loss of wall thickness after several decades of storage in the presence of corrosion inhibitor which provides further confidence in the long term wet storage of stainless-steel clad fuels.

Compared to Zr alloys, iron alloys do not readily absorb hydrogen so embrittlement is not a significant issue. In general, steel cladding has a higher acceptable temperature during drying than Zr alloy cladding as it is not affected by hydride reorientation.

However, iron alloys, especially those with higher Cr content, are susceptible to sensitisation mechanisms (both thermal and irradiation assisted). Thermal sensitisation, characterised by the formation of chromium carbide precipitates and depletion of Cr in solution is not expected to affect LWR fuels since temperatures in excess of 427°C (800°F) are required and the alloy can be manufactured to be largely resistance to thermal sensitisation. Neutron irradiation leads to Radiation Induced Segregation (RIS) as a result of a dynamic equilibrium in atomic migration of vacancies and atoms at grain boundaries. Under a limited range of temperatures, including those encountered by AGR fuel, this can lead to significant depletion of Cr at grain boundaries [15–17], leaving the material susceptible to localised corrosion, particularly stress corrosion cracking (SCC) [18]. However, there appears to be mechanistic differences between SCC in gaseous environments and in water that result in more severe and extensive corrosion is low humidity 'dry' storage conditions. Currently dry storage of AGR fuel is not underpinned and further work would be required to underpin dry storage of other RIS affected fuel claddings in dry conditions in order to avoid failures.

Unlike Zr alloys, which are effective hydrogen getters, Fe based alloys will allow hydrogen (and similarly tritium) to permeate. The consequence of this both in reactor as well as for long term storage will need to be evaluated. A liner material could potentially be used as a barrier to tritium release.

2.3. Silicon Carbide Composites (SiC/SiC)

SiC/SiC composites consist of high purity SiC fibres wound and braided into tubular forms and then impregnated with a matrix of SiC. The oxidation resistance of these materials in high temperature steam is superior to both the coated Zr alloys and Fe based alloy ATF options. However, the lack of any prior experience in LWRs and the fact that the material behaves more like a ceramic than a metallic cladding, means that its introduction as an ATF concept is particularly challenging. One area of concern is hydrothermal corrosion under normal operating conditions due to the difficulty in forming a stable oxide layer. This is particularly true of the oxidising conditions present in Boiling Water Reactor (BWR) water chemistries. One potential solution is to dose with hydrogen as is typical for Pressurised Water Reactors (PWRs), however if this is not sufficiently effective an additional protective coating may be required.

SiC is very corrosion resistant at low temperatures in water and air as a result of the formation of a protective SiO_2 surface layer, although irradiation damage has been found increase the rate of dissolution. Experimental work on TRISO particles has been carried out (both irradiated and unirradiated) which suggested that the SiC layer has long term durability in repository conditions [19,20].

Since SiC/SiC cladding contains a source of carbon, there is potential for the formation of organic molecules or gaseous species. SiC/SiC cladding may therefore add to the source term for carbon in a repository and the form of carbon and its rate of generation will be of interest for the performance assessment. Whilst it is

not currently possible to assume that the SiC/SiC cladding is fully corrosion resistant under repository conditions it is clear that it has a sufficiently high durability that any additional source of carbon will not form an acute release and there is no reason to consider that it would be unacceptable within the current Geological Disposal Facility (GDF) concepts. The evolution of C from the fuel cladding would nevertheless require work to identify the potential corrosion products and the rate of production under relevant disposal conditions at an appropriate point prior to a disposability assessment.

A UC_2 kernel fuel coated with a layer of pyrolytic carbon encased in loosely sintered SiC within a SiC cladding tube were tested in the UK Windscale Advanced Gas-cooled Reactor (WAGR) on an experimental basis in the late 1960s. During testing failure in the cladding was detected at low burn up. The pins were placed straight into pond storage and have recently been migrated to dry storage. No further signs of corrosion were identified during repackaging. A few fuel pins did fracture but these are thought to have been caused during the packing process rather than the irradiation or storage.

Although this WAGR testing provides a good start point for exploring the suitability of SiC/SiC in storage, it was carried out on a small scale for monolithic SiC materials, so testing of modern composite materials irradiated to levels commensurate with modern fuel is recommended to provide confidence in the effects of different materials processing, irradiation conditions and burn-up.

3. FUEL

3.1. Uranium dioxide (UO_2) and doped UO_2

UO_2, with or without minor dopant additions is the fuel of choice for current LWRs. It was chosen because it has good resistance to oxidation in water and a high melting point. The main drawback is its low thermal conductivity which further decreases with temperature and irradiation and results in high thermal gradients and fragmentation of the fuel within the reactor. The behaviour of UO_2 in storage and disposal is relatively benign, in that it oxidises/corrodes slowly in air at low temperatures and in common groundwaters. When oxidised, UO_2 expands initially by accommodation of interstitial oxygen up to a stoichiometry of U_4O_9 then to U_3O_8 with a change of atomic structure. The volume expansion leads to a friable (dispersible) oxide and can cause progressive failure of cladding. Therefore, inert gas is preferred for dry storage. Irradiated fuel initially oxidises more rapidly than UO_2, however the presence of fission products tends to stabilise the U_4O_9 structure leading to a delay on subsequent oxidation [21]. Accelerated testing of UO_2 and MOX fuels has shown that volume expansion due to helium accumulation in the fuel matrix saturates at around 2%. At this level there is no concern over the integrity of Zr alloy or steel-clad fuels during long term storage. Some testing to assess the long term swelling behaviour of other fuel matrices would be recommended if such fuels are selected for deployment to confirm both the rate of accumulation and saturation levels.

The addition of small quantities of dopants to the UO_2 matrix can assist densification and grain growth, with consequential lower fission gas release and better resistance to fuel wash out [22]. Fuels of this type are already commercial products, such as the Westinghouse ADOPT™ fuel. Further work on doped fuels is underway as part of an EU funded Horizon 2020 project, Modern Spent Fuel Dissolution and Chemistry in Failed Container Conditions (DisCo) [23]. There is also interest in more advanced UO_2 fuels where the goal is to increase the thermal conductivity, for example by creating a network of higher thermal conductivity material within the microstructure [24]. These fuels, which have benefits of reducing peak pellet centreline temperatures enabling more efficient heat removal, require higher quantities of dopant materials (typically 10 wt%). Spent fuel management of these advanced fuels has yet to be assessed but is not expected to be significantly inferior to UO_2.

3.2. Uranium Nitride (UN)

Uranium Nitride (UN) is being considered as a candidate ATF due to its superior thermal properties and higher uranium density when compared to UO_2. Reactivity with water and oxygen is the main concern for UN since it is known to be significantly inferior to UO_2, which could have implications for spent fuel storage and disposal. A number of studies of the oxidation and hydrolysis of UN have been undertaken on both solid and powdered materials [25,26]. The onset of rapid oxidation (ignition) of powders typically occurs around 300°C in air and 340°C in water vapour, but depends greatly on the particle size. Solid materials are relatively stable at room temperature due to the formation of epitaxial layers of U_2N_3 and UO_2 which afford protection [26]. Within the temperature range encountered during fuel drying and storage, inert atmosphere conditions would need to be maintained. It is worth noting that the reactivity of UN to water is also a concern during reactor operation, due to the potential for fuel pin failures. Efforts are underway to improve the water tolerance of UN, for example by forming a composite with another material [27]. Hence there is a reasonable expectation that any fuel so

developed will also perform adequately in the conditions encountered during wet or dry storage. Testing of these modified fuel forms will need to be undertaken in representative conditions over sufficient time periods to underpin this assumption.

Carbon-14 is a known fission product when using standard nitrogen (99.63% N-14). This can be volatised which would cause the release of a radioactive gas. This could be a significant challenge for storage and disposal facilities. The proposed solution is to enrich the nitrogen in N-15, however it is not yet clear the level of enrichment that would be required to keep C-14 production to acceptable levels and enrichment processes are not yet economic on a large scale.

3.3. Uranium Silicide (U_3Si_2)

Uranium Silicide (U_3Si_2) is also being considered as an ATF candidate fuel for the same reason as UN (improved thermal conductivity and uranium density). Although the increase in uranium density is lower than for UN (17% compared to 40%), there are no isotopic concerns. Like UN, U_3Si_2 is known to oxidise in both water and oxygen, although testing under pressurised water conditions shows notably better performance than UN [28]. Ignition of bulk U_3Si_2 in air typically occurs at around 400°C, and in steam rapid pulverisation occurs at around 460-480°C [29]. The pulverisation in steam is thought to be associated with the formation of a uranium silicide hydride ($U_3Si_2H_{1.8}$) phase, with similarities to the way that hydrogen can pulverise bulk uranium [29]. At lower temperatures in water, cast bars of U_3Si_2 exhibited little evidence of reaction in water over 16 days at 100°and 4 days at 200°C [30]. Longer term testing has not yet been performed. As with UN there is currently on-going research aimed at improving the water tolerance of U_3Si_2 in reactor conditions, one example being the addition of dopants [31]. Promising candidate compositions will also need to undergo long term testing at low temperatures of both unirradiated and irradiated materials to underpin their safe storage and disposal.

4. CONCLUSIONS

Significant international research is being undertaken to develop ATF for LWRs that could provide greater resilience from the fuel in the event of a severe accident. A range of different fuel and cladding concepts are under development which might be described as evolutionary or revolutionary by the extent to which they deviate from the current fuel. This paper has provided an initial assessment of spent fuel management considerations for a number of these concepts that are being actively researched. Spent fuel management has received little consideration to date, but this is expected to change as concepts progress to licensing. Of the concepts assessed the following conclusions can be made:

- Evolutionary concepts, such as coated Zr alloys and doped UO_2 fuels, which are closest to commercial deployment, will benefit from current experience with standard UO_2/Zr alloy fuel, and although some testing is likely to be required, their behaviour is expected to be bounded by the current fuel;
- Advanced iron alloy claddings can be expected to exhibit lower general corrosion rates than Zr alloys. These materials are however prone to localised corrosion resulting from radiation induced sensitisation. Evidence from the early use of austenitic stainless steels in LWRs indicates that these effects should not be an issue, although corrosion inhibitors have been successfully used during pond storage and could be adopted if required;
- SiC/SiC composites are expected to be exceedingly durable cladding materials in long term storage, although data underpinning this assumption will be required. Differences in the quality of SiC/SiC composites with processing and joining technologies will need to be accounted for and the potential for the evolution of carbon containing species assessed;
- Both UN and U_3Si_2 fuels show a greater propensity for corrosion than UO_2 in both air and water. Improvements in water tolerance are being sought to ensure their suitability for use in LWRs and it is these composite or doped fuels that will require testing at low temperatures relevant to storage and disposal conditions.

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