BEHAVIOUR OF SPENT FUEL DURING STORAGE

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Continued storage operations are reliant on being able to demonstrate, and provide confidence to key stakeholders, that spent fuel can be safely stored, retrieved and transferred or transported to the next stage in the fuel cycle. Since 1981, the International Atomic Energy Agency (IAEA) has assisted in this process by organizing Coordinated Research Projects (CRPs) on the behaviour of power reactor spent fuel during long-term (or extended) storage. The projects were initially called BEFAST-I to -III (BEhaviour of spent Fuel Assemblies in STorage), and later SPAR-I to -III (Spent fuel Performance Assessment and Research), respectively. Upon completion of each CRP, a summary of the results was published in an IAEA technical document (TECDOC). The technical documents catalogue the evolution of spent fuel storage over the past 33 years and technical underpinnings.

In 2014, the process of consolidating the findings from the BEFAST and SPAR projects was initiated. The current paper provides an over-view of these findings, paying particular attention to those on spent fuel degradation mechanisms and behaviour in wet and dry storage.

I. INTRODUCTION

In 1979, the International Atomic Energy Agency (IAEA) initiated a survey of world experience of water reactor spent fuel storage. The results of this survey were later published in a Technical Report Series document [1]. The main findings from the survey were:

"Experience exists with wet storage of light water reactor (LWR) and heavy water reactor (HWR) fuel for periods up to 20 years with low burnup fuel. No significant difficulties are expected in projecting spent fuel behaviour in wet storage for longer times and higher burnups. Nevertheless, observation and investigation should be continued to evaluate the behaviour of high burnup spent fuel assemblies during prolonged storage periods and to confirm the present and positive experience".

The recommendation became the basis for the initiation of the IAEA's first Coordinated Research Project (CRP) on the behaviour of spent fuel during long-term (or extended) storage. The CRP initiated in 1981 was called BEFAST (BEhaviour of spent Fuel Assemblies in Storage) and the main objective of the CRP was to provide a significant database on cladding integrity of spent fuel after extended storage periods. The scope of the project also included storage equipment and reporting on-going research and development in support of spent fuel storage.

On the completion of BEFAST I, because of the evolving situation associated with fuel cycle efficiencies, the introduction of new fuels and the development of new storage systems (particularly dry storage systems for HWR and LWR fuels), it was recognized that the task of building the technical knowledge base had only just begun. The BEFAST project continued until 1996, after the third phase had been completed. In 1997, the project was reinitiated under the new name SPAR (Spent fuel Performance Assessment and Research) but the main objective and scope essentially remained the same:

To develop a technical knowledge base on the long-term behaviour of power reactor spent fuel and storage system materials through the evaluation of operating experience and research by participating Member States.

Some 33 years on, with the completion of SPAR III, 6 technical documents have been produced [2–7]. The technical documents catalogue both the evolution of spent fuel storage over the past 33 years and technical underpinning. In the current paper it is not possible to summarise all the information reported in the BEFAST and SPAR documents. This activity is currently in progress and we look forward to issuing a consolidated technical document in 2015. Although BEFAST and SPAR covered all fuel types, the present paper will focus on how our understanding of spent zirconium-based alloy fuel behaviour has developed since 1981.

II. SPENT FUEL DEGRADATION MECHANISMS IN WET AND DRY STORAGE

At the beginning of storage, the spent fuel has undergone a number of changes as a result of reactor service. Examples include: Irradiation which causes

modifications to clad and fuel pellet microstructures; fission gas production leading to the fuel rods being pressurized; internal attack of the fuel clad by fission gas products; thermal changes, impacts from mechanical interactions; and corrosion/pick-up of chemical elements from the reactor coolant. These beginning of storage properties can influence how the fuel will behave in extended storage.

In BEFAST I, the potential degradation mechanisms for zirconium clad LWR fuels in wet and dry storage were represented by 3-D pie diagrams of a fuel pin section; for example Fig. 1.
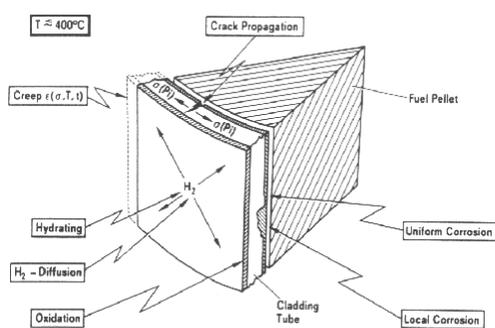

Fig. 1. Mechanisms affecting spent fuel cladding during dry storage [8].

As stated in [2], the degradation mechanisms in wet and dry storage are in principle the same but the order of severity differs. For example, the application of the ideal gas law tells us that rod internal gas pressure (RIP) is not significant at wet storage temperatures in comparison to dry storage.

II.A. Wet Storage

At the time of BEFAST I, the potential degradation mechanisms that may affect cladding integrity in wet storage were identified as:

- Uniform (aqueous) corrosion;
- Pitting, galvanic, and microbiologically-influenced corrosion (MIC); and
- Hydriding.

The current understanding is that there are neither observed nor theoretical mechanisms that have been identified which would limit the time zirconium alloy clad fuel could remain in pool storage provided pool chemistry is maintained within specified limits.

For uniform corrosion, the corrosion behaviour of zirconium alloys in pressurised water and water vapour is well known for temperatures above 200°C. Because the corrosion rate follows the Arrhenius law in the linear, post-transition corrosion regime, corrosion rates in pool water <50°C are a factor of ~10^{10} lower than those under operational reactor conditions. Recently, immersion tests

have been undertaken on irradiated zircaloy-4 clad specimens at 50°C in a variety of potential pool chemistries for up to 1 year [7]. The results of this study support that zirconium-based alloys are virtually immune to uniform corrosion during wet storage, as observed corrosion rates were below the resolution limit of the measurement techniques being used; i.e. weight loss and scanning electron microscopy.

In terms of pitting, galvanic and microbiologically-influenced corrosion (MIC), a combination of zirconium-based alloys being near the noble end of the galvanic series, material selection for storage racks etc., control of pool water chemistry, and monitoring for the presence of harmful biological species has avoided spent fuel degradation from these mechanisms in wet storage. Even where we might expect galvanic corrosion, for example where zircaloy is in contact with aluminum (near the active end of the galvanic series), this has not occurred due to the passivating effect of the oxide layer formed on zircaloy surfaces in reactor.

Whilst hydrogen behaviour in zirconium-based alloys has become a major topic of research over the past 10 years for spent fuel being transferred into dry storage, the position in wet storage is entirely different. When the fuel is cooled to the storage pool temperature, hydrogen in solid solution in the alloy precipitates in the form of hydride platelets primarily oriented in the circumferential direction, except possibly for the hydrogen that is associated with (or trapped in) irradiation defects, such as vacancy dislocation loops. Circumferential hydrides, in combination with hardening due to irradiation effects, decrease cladding ductility in response to axial and hoop loads, but the cladding retains enough residual ductility not to be impaired by fuel handling operations.

Due to the heat transfer characteristics of water, hydrogen redistribution by thermal diffusion and formation of so-called pits by an Oswald ripening can be ruled out during long-term wet storage at the pool temperatures of practical interest. Any further loss of clad ductility during wet storage therefore, can be ruled out.

The dataset that supports on-going spent fuel clad integrity in wet storage is extensive. At the time of BEFAST I, there were a number of benchmarked spent fuels in monitoring or surveillance programmes that had been underway for 10+ years. These continued into the 1990s but, by the end of the 1990s further periodic inspections were mostly postponed as no changes had been observed neither were any expected; by this time some of the fuel had been in storage for 40 years. On-going spent fuel integrity is now confirmed through routine pool water sampling (an indicator of bulk or generic clad failure) and visual inspection when fuel is transferred to either dry storage or reprocessing. With the exception of a number of reported PWR top nozzle separations during spent fuel handling, as a result of stress corrosion cracking in the structural joints between the top

nozzle and fuel bundle in some older PWR fuels [4], there has been no reported degradation issues for zirconium-based spent fuels in wet storage for >50 years. Changes to PWR fuel design and specific limits on pool water impurities addressed the issue related to top nozzle separation in some PWR fuels and no further problems have been reported since.

II.B. Dry Storage

In 1986 the only licensed power reactor dry storage facilities in operation were those at the Wylfa nuclear power plant (NPP) in the United Kingdom (UK). The dry storage facilities, for MAGNOX fuel, comprise three carbon dioxide cooled vaults (1971) and two air cooled vaults (1979). The air cooled stores have since been defueled and Wylfa NPP is partly shut-down with the remaining reactor programmed to be shut-down in 2015 and defueled to Sellafield (UK) for reprocessing.

Despite this, there was significant interest in the dry storage of zirconium-based alloy fuels and BEFAST I reported on a combination of active demonstrations and laboratory based research programmes from Austria, Canada, Germany, Japan and United States of America (USA). The technical questions being asked at this time were:

- What are suitable storage media?
- What is the maximum cladding temperature below which a significant amount of cladding failures will not occur?
- How well can the cladding temperature be predicted to ensure that the temperature limit will not be exceeded?

The potential degradation mechanisms that have since been identified which could affect cladding integrity of LWR fuels during dry storage and subsequent handling and transportation operations are:
1) Air Oxidation
2) Thermal Creep
3) Stress Corrosion Cracking (SCC)
4) Delayed Hydrogen Cracking (DHC)
5) Hydride Reorientation
6) Hydrogen Migration and Re-distribution

Each mechanism is discussed briefly.

Under normal operating conditions, air oxidation can be discounted. Air oxidation would, however, be the most important degradation mechanism in dry storage if defected fuel, where UO_2 is exposed, was present. At temperatures >250°C the UO_2 can convert to U_3O_8 and the resultant volume increase can lead to enlargement of the defect and potential loss of fuel material. BEFAST 1 studies [2] suggested that the defect had to be sufficiently large, for example 0.8mm.

In BEFAST I, thermal creep was considered as one of the most limiting degradation mechanisms in dry storage. Spent fuel rods in dry storage undergo creep deformations

at a rate determined by the internal fuel rod pressure, cladding temperature, and composition and metallurgical conditions of the rod cladding. Under certain conditions, this deformation can lead to fracture of the fuel cladding (creep rupture). To protect against the possibility of creep rupture during dry storage a limit of a 1% end-of-storage-life clad creep strain was proposed in Germany [4] and was later adopted by licensing authorities. The limit was based on an observation that post-pile burst tests of irradiated cladding will reach a 1% uniform strain before rupture.

Since these early investigations, additional studies have been undertaken in Canada, France, Japan, Spain and USA. The studies have significantly improved the understanding of thermal creep behaviour [6] through: (1) increasing the experimental creep dataset, including creep rupture criterion; and (2) the development of analytical models capable of simulating the cladding response under prescribed temperature and internal pressure conditions representative of dry storage applications.

This body of information led the US Nuclear Regulatory Commission in 2003 to draw the conclusion that thermal creep would not result in gross rupture of the cladding and that the geometric configuration of the spent fuel will be preserved, provided that the maximum cladding temperature does not exceed 400°C [9]. In terms of regulatory requirements for dry storage, the current position is that we now have a two tier system with those working to the original 1% creep strain limit and an alternative position adopted in the USA of no limit.

The potential for iodine-induced stress corrosion cracking (SCC) is discussed in detail by Peehs [10]. For SCC to occur, a combination of high temperature, chemically active iodine and stress are required. At temperatures typical in interim storage conditions, the fission products are practically immobile in the fuel matrix and iodine is not present in a form that could trigger SCC. It can, therefore, be concluded that cladding failure via this mechanism is not expected to occur. Consequently all fission-product-driven defect mechanisms can be discounted.

DHC is a very specific mechanistic process requiring tri-axial stressing of the zirconium to dilate the crystal lattice. Tri-axial stresses dilate the Zr lattice tetrahedral sites that are the ones occupied by hydrogen atoms in solution. This allows the hydrogen atoms to diffuse up the stress gradient and precipitate in the peak stress region. Hydrides grow in this region by dissolving hydrogen in adjacent hydrides in unstressed regions. The hydrides grow until big enough to crack. Although DHC has been observed in Zircaloy specimens that are sufficiently thick, DHC is not expected to be an active degradation mechanism in cladding tubes, given that the latter do not appear to have enough wall thickness to generate much triaxial stress. Rashid et al. [11] present an analysis defining five conditions that should exist for DHC to be

an active mechanism in LWR fuel during dry storage. The analysis concludes that DHC is not an operative mechanism in dry storage. It shows that stress intensity factors in LWR fuel cladding are below the critical stress intensity factors for either crack initiation or sustaining crack growth.

DHC, however, is considered to be the most critical mechanism that could affect the CANDU bundle integrity (endcaps and endplate welds) during dry storage [6].

The behaviour of hydrogen in zirconium-based alloys has received a great deal of attention over the past two phases of SPAR. The current understanding is that hydride reorientation, hydrogen migration and re-distribution are unlikely to impact fuel clad integrity during storage. However, they have the potential to impair the ability of the cladding to effectively withstand potentially adverse mechanical challenges resulting from handling or transportation accidents. This is through a reduction in fuel clad ductility mainly as a result of hydrogen being dissolved and re-precipitated in the radial direction and can occur under a given set of conditions.

It has been established that only hydride that is dissolved by raising the cladding temperatures during the act of transferring spent fuel from a wet environment to a dry environment, and then re-precipitated during subsequent cooling, can potentially re-orient. The amount of hydrogen which goes into solution is a function of the clad temperature reached during the drying process and the hydrogen solubility limit in zirconium (210 ppm). For example, there is around a three-fold increase in the amount of hydrogen in solution at 400°C compared to 300°C. Hence, we may simply conclude that it is better to transfer long cooled fuel. This, however, is only part of the story and in SPAR III Argentina, France, Japan, Republic of Korea, Spain, Switzerland and USA were investigating the influence of the following parameters on subsequent fuel clad properties:

1. Material type (composition and initial microstructure);
2. Hydrogen content and distribution;
3. Fluence;
4. Temperature;
5. Applied stress;
6. Cooling rate; and
7) Extent of temperature history, such as cycling and annealing.

The experimental set-ups, results obtained and a discussion of the results is reported extensively in the SPAR III technical document [7] and will not be repeated here.

Based upon the findings from the studies on unirradiated and irradiated clad tube specimens, we may expect that fuel rapidly cooled will be more ductile than fuel that is allowed to cool down slowly. This conclusion tends to be supported, for normal fuel handling operations, by the experiences gained in dry transferring

high burnup (HBU) spent fuel from French NPPs to the reprocessing facilities at Cap La Hague, as no fuel failures on receipt at the reprocessing facilities have been reported. Other findings suggest that the presence of a zirconium liner appears to be beneficial and some clad materials appear to be better than others. There are still unknowns and the the properties associated with the presence of fuel may be different. A gap in this area is in establishing experiments under actual storage conditions.

Although there is > 30 years' operating experience in dry storing power reactor fuel, in terms of the performance of spent fuel that has been in dry storage, this is limited in comparison to wet storage. The main reason is most spent fuel loaded to a dry storage system has not been retrieved and that majority of dry storage systems were not designed for direct inspection or monitoring of the spent fuel. On-going spent fuel integrity is mostly inferred through confirmation that the inert conditions inside the storage system have been maintained. The main exception is MAGNOX fuel stored at Wylfa NPP (UK) which has been routinely retrieved for reprocessing; the modular vault dry store at Paks NPP (Hungary) where the storage tube cover gas can be periodically monitored; Japan where two casks have been opened as part of license requirements; Japan where one cask of spent fuel was inspected in support of transfer operations to the new temporary storage facility at Fukushima Daiichi NPP and R&D programmes for example in Canada and the USA. New demonstration programmes on HBU spent LWR fuels are planned in Japan and USA.

III. DEGRADATION MECHANISM IN THE VERY LONG-TERM

In the very long-term, α-decay damage and the accumulation of helium are the key processes affecting the evolution of properties and behaviour of spent fuel in extended storage. Cladding stress during storage may be affected by fuel swelling, helium generation due to α-decay of actinides and fuel fragmentation.

The potential for this to be a significant degradation mechanism was first evaluated during SPAR I [5]. More recently, SPAR III, a multi-year study programme into the impact of α-decay damage and the accumulation of helium was initiated by the Joint Research Centre Institute for Transuranium Elements (JRC-ITU). The initial findings from the studies on α-doped UO_2 materials are reported in SPAR III [7].

These are summarized as, although there remains unknowns in relation to gas retention by the fuel matrix, bulk pellet swelling or release mechanisms to the free volume, even where all the gas generated in the first 300 years of UO_2 spent fuel storage is released, there is a negligible effect on cladding stress during storage. More attention has to be devoted to spent MOX fuel behaviour.

IV. SPAR IV?

Continued spent fuel storage and future transitions from one phase of the back-end of the nuclear fuel cycle to the next require that the operational experience and research results be reported and disseminated to those engaged in spent fuel storage for input to continued operations safety assessments and the licensing of new facilities. In this respect, field experience needs to be collected and reported, especially from the deployment of new technology.

For these reasons, it is proposed that the IAEA continues to develop its technical knowledge base on the long-term behaviour of power reactor spent fuel and storage system materials.

V. CONCLUSIONS

There is >50 years' experience of storing zirconium-based fuels in pools. Performance in storage remains excellent with no generic failure mechanism identified or experienced.

There is > 30 years' operating experience in storing power reactor fuel dry. Our understanding of the potential degradation mechanisms in dry storage has improved considerably since 1981 and we would not expect spent fuel to degrade during dry storage.

The transfer of short cooled HBU zirconium-based alloy spent fuel from wet to dry storage or reprocessing has the potential to lead to an alteration of the clad properties. There has been a significant amount of studies in this area. Whilst the studies using empty unirradiated and irradiated clad tubes have improved our understanding of hydrogen behaviour in zirconium-based alloy tube specimens, there are still unknowns and the properties associated with the presence of fuel may be different. A gap in this area in establishing experiments under actual storage conditions has been identified.

In the very long- term, α-decay damage and the accumulation of helium are the key processes which could impact on spent fuel integrity. However, initial studies have concluded that there is negligible effect in the first 300 years of storage for UO_2 fuels.

ACKNOWLEDGMENTS

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