Post Irradiation Examination of UK Advanced Gas-cooled Reactor Fuel Cladding (post-reactor and post-storage)

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ABSTRACT

The UK Nuclear Decommissioning Authority (NDA) has declared that reprocessing of spent fuel at the Thermal Oxide Reprocessing Plant (THORP) will cease in 2018. The remaining inventory of spent advanced gas-cooled reactor (AGR) fuel will be managed through long term storage pending geological disposal. The reactor operator has a strategic target to secure plant life extensions by an average of eight years, and it is estimated that stocks of AGR fuel will rise by 2040. Based on this expected rise of inventory within fuel storage ponds a programme of work , carried out at the Windscale Laboratory, was designed to characterise irradiated AGR cladding both prior to and after long term storage (>25 years). Some of this work, and the PIE techniques used, will be presented in this paper.

1. Introduction

1.1 UK Advanced Gas-cooled Reactor (CAGR)

Commercial Advanced Gas-Cooled Reactors (CAGR) were first developed in the UK and have a higher volumetric power density than the earlier designed Magnox reactors. Much of the development work for CAGR was performed at Windscale using the Windscale advanced gas-cooled reactor (WAGR). The CAGR design uses carbon dioxide as a coolant (although at a higher pressure and temperature than the previous Magnox design). Hollow bore uranium oxide (UO_2) pellets are clad with stainless steel tubes, and the fuel pellets are grouped in fuel element bundles of 36, with each tube being around 1m in length. The tubes are machined to create an external rib profile, allowing for increased heat transfer when in use. A solid graphite moderator is used and the uranium enriched to several percent U^{235} to account for the neutron absorption of the stainless steel. Due to the relatively high temperature of the reactor coolant, the steam cycle efficiencies of a CAGR are very high, at around 40% [1].

1.2 Spent CAGR fuel treatment and storage

After irradiation the fuel stringer (up to eight elements stacked axially) is dismantled and individual elements are stored before undergoing further reprocessing. Currently spent nuclear fuel is treated at the thermal oxide reprocessing plant (THORP) in the UK. However, the Nuclear Decommissioning Authority has declared that fuel reprocessing will cease in 2018, meaning that the remaining spent fuel stock within the UK will continue to rise until the AGR stations cease to operate. The reactor operator has a strategic target to secure plant life extensions by an average of eight years, and it is estimated that stocks of AGR fuel will rise by 2040 [2]. In the absence of a final geological disposal solution, these fuel stocks will be managed through long term pond (wet) storage, resulting in an increase in storage

duration to around 80 years. With this in mind, the objectives of spent fuel storage include (but are not limited to) [3]:

- Maintaining the integrity of primary containment barrier (i.e. the fuel cladding) during handling and exposure to the storage environment
- Maintaining retrievability
- Ensuring that radiation rates and doses do not exceed acceptable limits,

During wet storage, the AGR fuel cladding acts as both a primary containment barrier and allows mechanical handling of individual fuel pins during THORP operations. The fuel is stored in pond water dosed with sodium hydroxide to around pH 11.4 to inhibit cladding failure as a result of radiation induced intergranular stress corrosion cracking. This corrosion inhibitor was chosen as a result of a development programme in the 1980's, and has been in use at all AGR storage ponds at Sellafield since 1986 [3] [4].

Whilst there are over 35 years of pond storage experience at Sellafield, changes to the front end of the fuel cycle (e.g. increasing fuel burn up) and increased storage durations have necessitated further examination of the original development work. Previously defined conditions mean that the stainless steel cladding must retain around a third of the original clad wall thickness, to maintain mechanical handling integrity during future recovery operations. Although predicted corrosion rates in pond are low (<<0.5microns/year), they must be limited during wet storage. In particular, the potential for localised corrosion of sensitised fuel cladding requires attention [3].

1.3 AGR cladding sensitisation

The cladding material used in the manufacture of AGR fuel pins is 20Cr/25/Ni which is Nb-stabilised. During reactor operation, stainless steel components in the 340-520°C temperature range become sensitised, primarily as a result of radiation induced segregation [5].

Sensitisation can result in cladding failures for pins stored in pond storage environments (with high chloride ion concentrations) through intergranular stress corrosion cracking (IGSCC). In stainless steel, sensitisation is caused by free carbon reacting with the chromium, forming chromium carbides on the grain boundaries. These Cr-rich carbides deplete the chromium and the stainless steel becomes sensitive to localised grain boundary corrosion [6]. AGR cladding avoids precipitation of chromium carbides via the inclusion of niobium, which stabilises the steel due to a preferential reaction of carbon with niobium to form niobium carbides.

However, as a result of defect generation due to nonelastic displacement of atoms by fast neutrons, radiation induced segregation can occur. An atom is knocked from its normal lattice site into an interstitial location and a mobile vacancy created. Vacancies are removed from the lattice at grain boundary sinks via an exchange of position with other atoms. The diffusion coefficients of iron, chromium and nickel in austenitic steel are such that chromium diffuses fastest, with chromium moving away from the grain boundary and nickel becoming enriched. A small window of irradiation temperature exists such that these chromium depleted zones are maintained, as diffusion from within the grain to the boundary is slow and vacancies are mobile enough to reach grain boundary sinks. The nature of the chromium depletion at a grain boundary (i.e. width of depleted zone) is likely to influence intergranular corrosion [5] [7] [8].

1.4 Post Irradiation Examination

Historically, the combination of in-reactor sensitisation and the subsequent wet storage necessitated a strategy for dealing with the spent sensitised fuel. In order to do this, a number of work programmes were undertaken on irradiated materials. The goal of a number of studies was to; define a suitable in-pond corrosion inhibitor for the sensitised fuel; a general corrosion rate of AGR cladding in pond storage; examine cladding post reactor and post storage for any evidence of degredation.

An early series of tests to determine a suitable corrosion inhibitor for sensitised cladding used whole AGR pins immersed in a test solution containing chloride. Cladding perforation was detected by monitoring the activity of the solution. The tests demonstrated that at 1–100 ppm chloride, all immersed pins failed between 80 and 350 days. However, immersion in a solution containing 200 ppm hydroxide (NaOH) and 10 ppm chloride, no evidence of attack was observed after one year of testing [9].
In addition, electrochemical testing of irradiated braces predicted that at a chloride concentration of 0.5 ppm at in-pond temperatures (30 ºC), pH 11.7 would be required to inhibit intergranular attack [10] [11]. Whilst being a similar steel composition to that of the cladding, braces receive a higher dose than fuel pins for the same irradiation temperature. Historical PSE of irradiated braces had shown them to be more susceptible to corrosion than fuel pins, although some pre-treatment was required for these tests. The use of braces also avoided testing (and therefore breach of primary containment) of whole pins [11].
General corrosion rates of AGR cladding within the storage ponds were found to be limited to <0.2 microns per year, equivalent to a loss of 5% total clad thickness over the proposed 80 year storage duration. This figure was determined by weight loss experiments with irradiated braces which, as mentioned previously, are similar in composition to the AGR cladding.
An early programme of post storage examination (PSE) [4] [9] on previously intact sensitised fuel, stored in both demineralised and caustic dosed pond water, also highlighted a number of key findings.

- For sensitized fuel, irradiated to >15 GWd/teU and stored in demineralised water with a chloride concentration of >1 ppm, 50 % of pins had failed as a result of IGA.
- For sensitized fuel stored in caustic dosed pond water, no failures attributed to IGA had occurred. However, some evidence of IGA to a depth of 40 µm was observed, and on 5 out of 164 samples deeper cracking was noted (Figure 1) [9].

As described previously, changes to the front end of the fuel cycle such as increasing fuel burn up, coupled with a requirement for increased storage durations [3], have resulted in the original development work being repeated for fuels of higher burn up and increased storage term. This programme is ongoing, but has thus far tested efficacy of the chosen inhibitor on steels from much higher burn up fuels, examined long stored fuels (<25 years), and analysed representative samples of higher burn up AGR cladding pre-storage. In addition, internally funded research and development projects are exploring opportunities for deploying characterisation techniques such as atomic force microscopy, alongside development of macro and microstructural simulants for spent fuel.

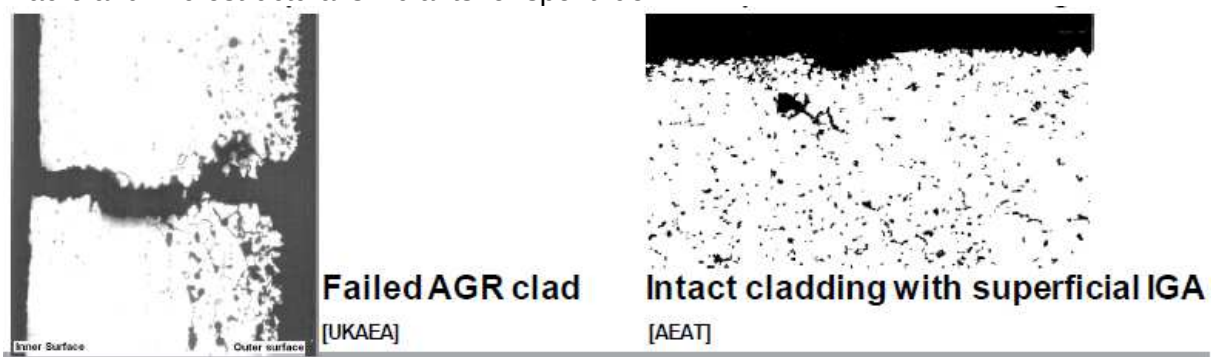


Figure 1: Example micrographs from initial PSE work programme (UKAEA and AEAT) [9]

2. Experimental Programmes

2.1 Corrosion inhibitor testing

A number of irradiated AGR braces were tested under simulated storage conditions. This was a repeat of earlier work, although there were some key differences (see Section 1.4). The irradiated braces sampled were from high burn up fuel, <38GWd/teU, and were not pre-

treated. Previously the monitoring method required some pre-treatment of the braces to detect IGA. In addition, the tests were conducted at representative temperatures of ≤ 50 ºC [11].

A number of specially designed pieces of equipment, shown in Figure 2, were required for this test, including corrosion tanks, in-cell spot welding equipment, diamond cutters and pre-assembled electrodes. After size reduction, samples were assembled into electrodes, Figure 3, and placed into one of four tanks, with a total of 48 samples being monitored simultaneously for <3 month periods. Peroxide was added to simulate the effects of solution radiolysis and carbonate was added to replicate the adsorption of atmospheric carbon dioxide. The aim was to demonstrate initiation and inhibition of corrosion using a chloride containing solution and a variety of corrosion inhibitors. Inhibitor solutions were fed into the tanks inside the hot cell, and corrosion currents were monitored using zero resistance ammetry (ZRA) [11].

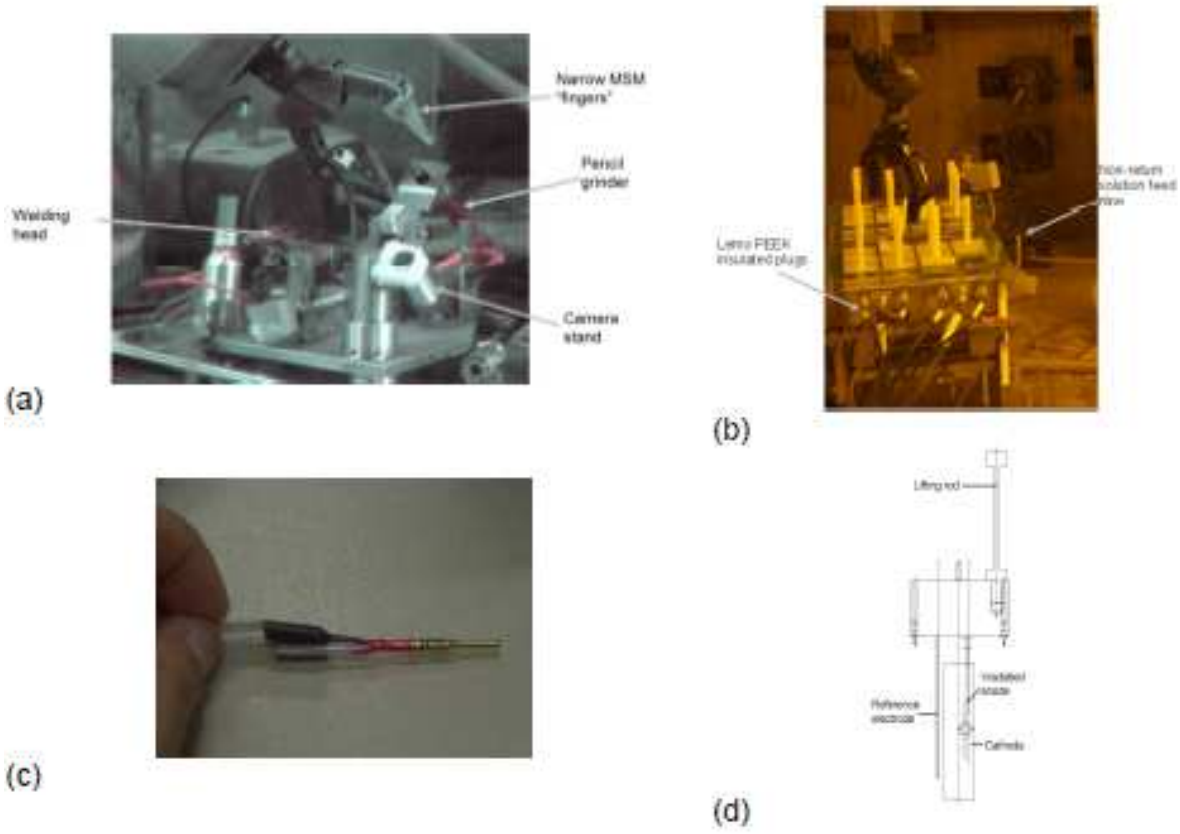


Figure 2: In-cell electrochemical corrosion testing equipment (a) spot welder (b) test tank #1 (c) anode connector (d) electrode assembly drawing

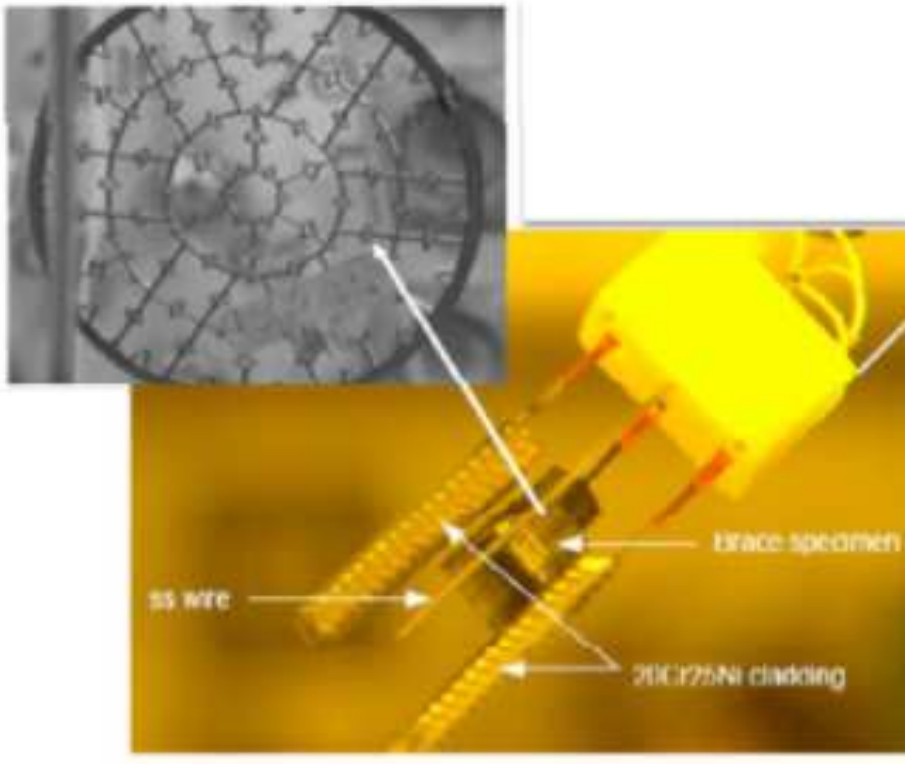


Figure 3: Assembled electrode within hot cell and sampled brace [11]

2.2 Post storage examination (PSE)

PSE of long stored, intact fuel is underway at the NNL Windscale PIE facility. This programme involves gamma scanning, visual inspection, impact (drop) testing, whole pin leak testing (on selected pins) and optical and electron microscopy. More specifically, the PSE work programme aims to determine the condition of AGR fuel cladding following extended exposure to the pond conditions planned for interim storage and to provide evidence to support current technical assessments.

Initially, the fuel was determined as intact based on the measured Cs-137 release rate from a selected, isolated container within the storage pond. The activity was monitored over a number of years. The selected container had been filled with pins known to be from several axial stringer locations i.e. sensitised and non-sensitised fuel.

The pins selected for the examination came from several axial positions within the reactor core, namely elements one, two and six. The lower elements were chosen to provide sensitised fuel cladding, and the higher element to provide a location at which no sensitisation would be expected.

Gamma scanning was initially performed in order to identify and differentiate axial positions within the fuel stringer. Once identified, pins were visually inspected to ensure they were intact, and a low power visual inspection was conducted on the selected pins for evidence of corrosion or other surface damage. Metallography and electron microscopy samples were then prepared from a batch of representative pins (see Section 2.3), and the remaining fuel rods underwent a drop test.

The drop test was specially designed to simulate the forces that fuel may experience during future handling or retrieval operations. The test rig, Figure 4, consisted of a clear tube, split into 2 halves and around 1.80m in height, into which the pin was remotely placed. Dampers ensured the pin was centralised inside the tube. A stainless steel weight was then secured at the top end of the tube, and the two halves assembled and placed upright. A quick release mechanism allowed the weight to drop onto the end of the vertical pin. After performing the test, the tube was dismantled and the pin ends assessed for signs of damage.

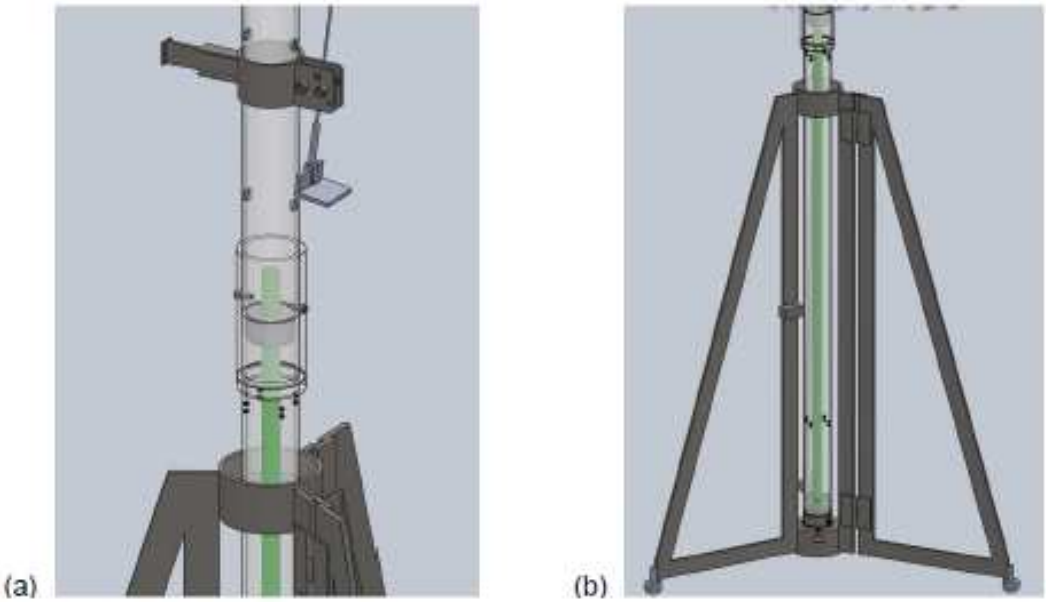


Figure 4: Impact testing rig drawing for long stored fuel (a) mid-point of assembled rig and tube holding fuel pin (b) assembled rig in vertical test position

2.3 AGR Cladding characterisation

The purpose of the clad characterisation programme was two-fold; to assess the effect of increasing burn up and reactor dwell time on the cladding, and assess the effect of extended storage on cladding.

Routine PIE was carried out on two recently discharged stringers of higher burn up fuel, with samples being taken from a series of axial positions within the stringers. Additionally, two long stored intact pins were selected for electron microscopy characterisation based on metallography characterisation.

Small strips of 3mm x 6mm, Figure 5b, cladding were removed from positions adjacent to those which had already been examined in cell. The clad strips were generally removed from what was nominally identified as the graphite sleeve facing side of the fuel pin. The cladding was hot mounted in a specially designed jig, and the samples were then thinned to remove fuel residues and heat transfer ribs, cut to form a 3mm x 3mm strip, and dissolved from the mounts. The cladding strips were exported from the hot cell and 1mm TEM discs were punched and thinned for subsequent examination. The second 3mm x 3mm strip was mounted in a standard metallographic mount and exported for SEM characterisation, Figure 5a.

The microstructures, distribution and composition of precipitates, and elemental compositions at grain boundaries were assessed using TEM and compared to available historic data.

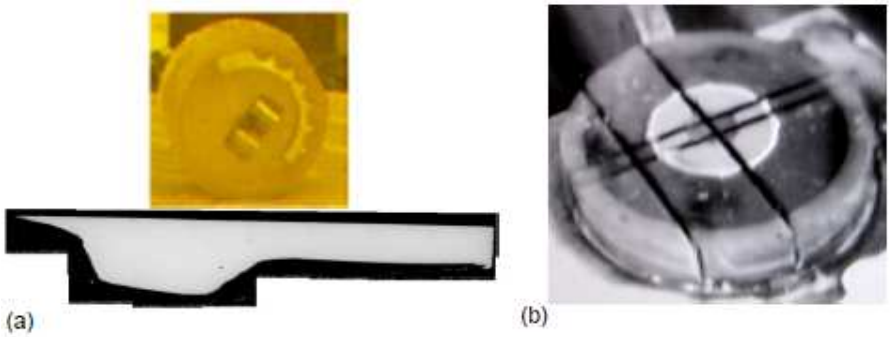


Figure 5: AGR cladding strips being prepared for electron microscopy examination (a) SEM mount and clad cross-section (b) TEM

3. Results

The electrochemical corrosion tests demonstrated corrosion occurred (and was successfully measured), inhibition of corrosion was possible and the tested inhibitors behaved as expected from previous studies. Results from the inhibitor tests using high burn up sensitized steel samples demonstrated that IGA was arrested by pH 11.0 in a solution of 2 ppm chloride at 50 ºC [11]. Further characterization of the samples (carried out at Studsvik Nuclear AB) after Strauss testing, Figure 6, demonstrated sensitization [11]. However, the grain boundary chromium depletion had not increased compared to lower burn-up cladding examined in the 1980s [9].

The inspection of the long stored intact fuel resulted in the positive identification of pins from elements one, two and six via the gamma scanning method, Figure 7. Previous routine in pond activity monitoring had shown no indication of Cs-137 release, and the pins were therefore assumed to be intact. On receipt into the hot cells further visual inspection, Figure 8, showed that the cladding was in good condition, with the exception of a few minor handling marks, and there was no evidence of corrosion or surface attack on any pins. Metallographic examination, Figure 9, further highlighted this, and no evidence of grain boundary intergranular attack was observed. The pins from element one exhibited a microstructure consistent with sensitised fuel (see Figure 9c), confirming that they would have been susceptible to attack under the required conditions. Finally, under a 15 Joule impact energy, all pins maintained mechanical integrity and further visual inspection showed no notable damage to the ends of the fuel pins.

Initial characterisation of higher burn up cladding from multiple axial locations showed results which were consistent with historic work i.e. grain boundary concentration profiles showed sensitisation where metallographic results had indicated, and cladding irradiated at higher temperatures showed no signs of grain boundary segregation, Figure 11. SEM characterisation, Figure 12, was able to identify precipitates which were consistent with earlier studies [12].

Figure 6: Irradiated AGR brace sections after strauss testing and an SEM micrograph of grain faces (SNAB) [11]

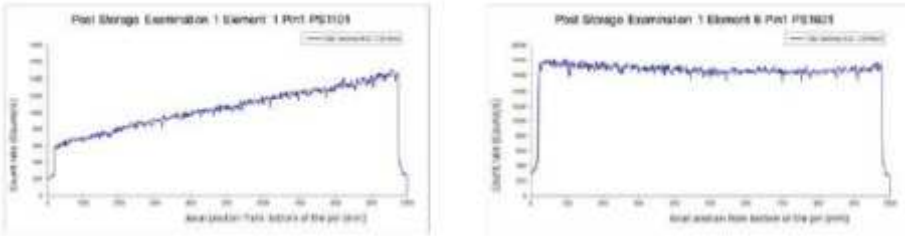


Figure 7: Example gamma scans from element one and element six pins

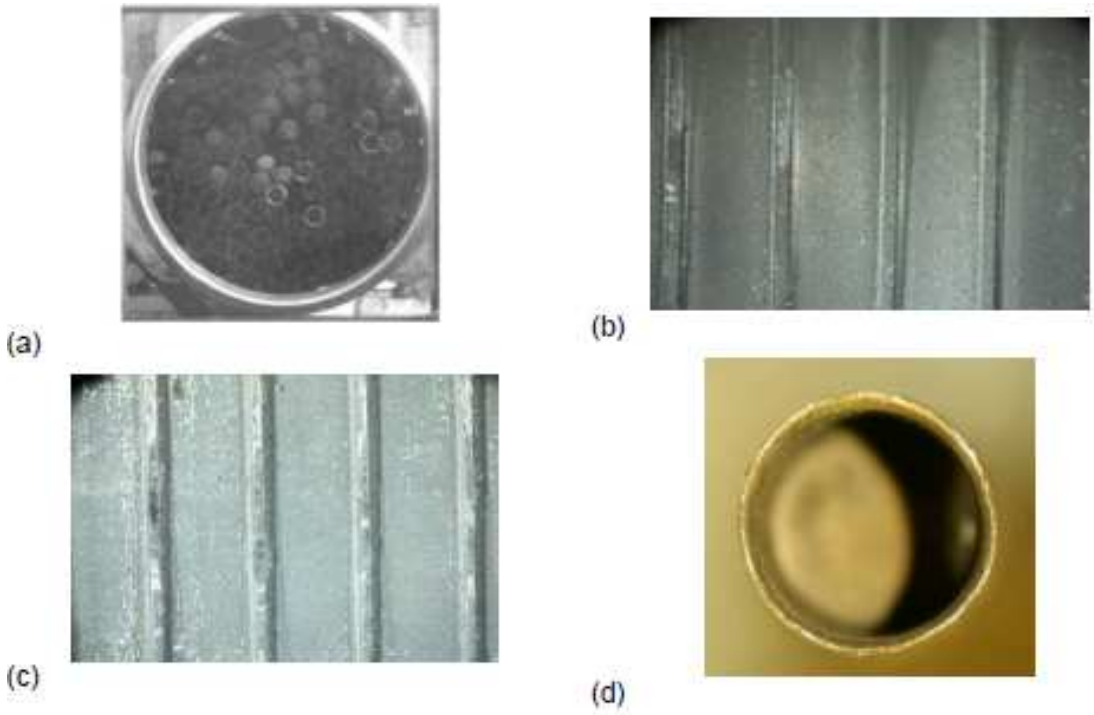


Figure 8: Visual inspection and photography of long stored pins showing (a) receipt inspection in slotted can (b) general cladding condition (c) minor handling damage on clad surface (d) end cap condition

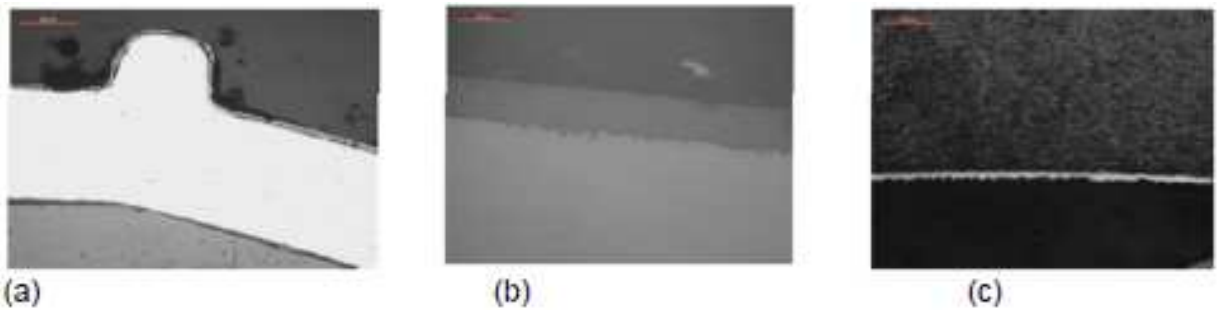


Figure 9: Metallographic examination of long stored pin cross section showing (a) clad and fuel interface as-poloshed (b) clad outer surface as-polished (c) etched cladding microstructure associated with RIS

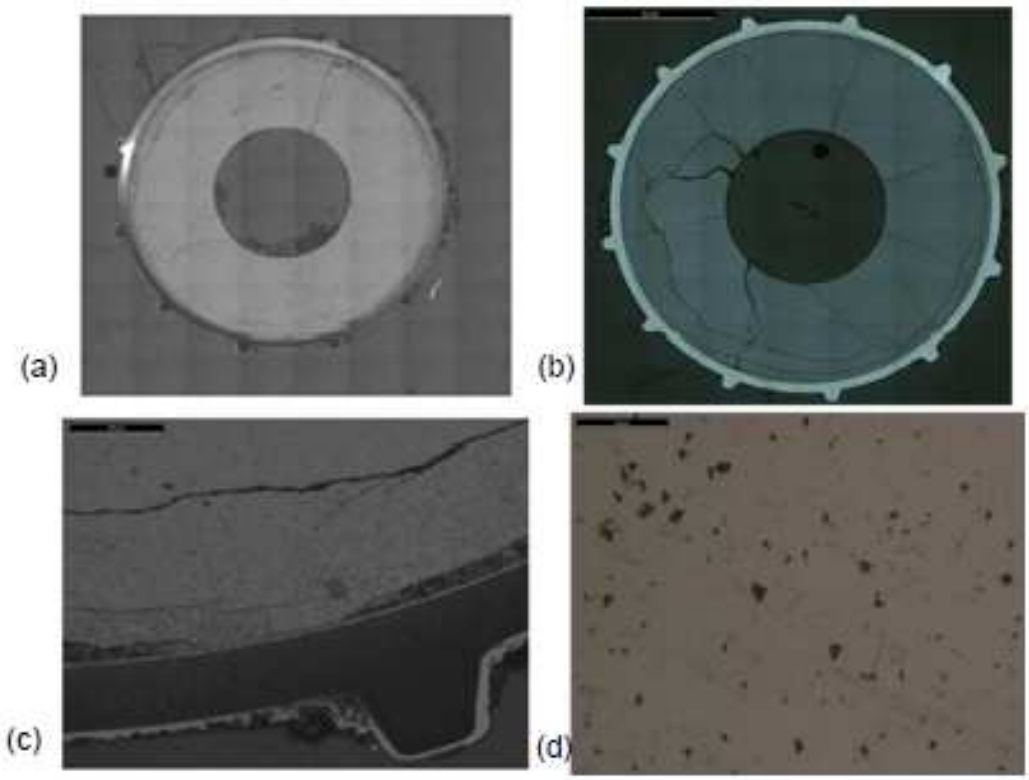


Figure 10: Metallographic images of AGR cladding microstructure from a higher burn up fuel stringer showing (a) cross section of etched fuel from a sensitised pin (b) cross section of fuel from a non-sensitised pin (c) cladding microstructure associated with RIS (d) typical clad microstructure of cladding operating at higher temperatures

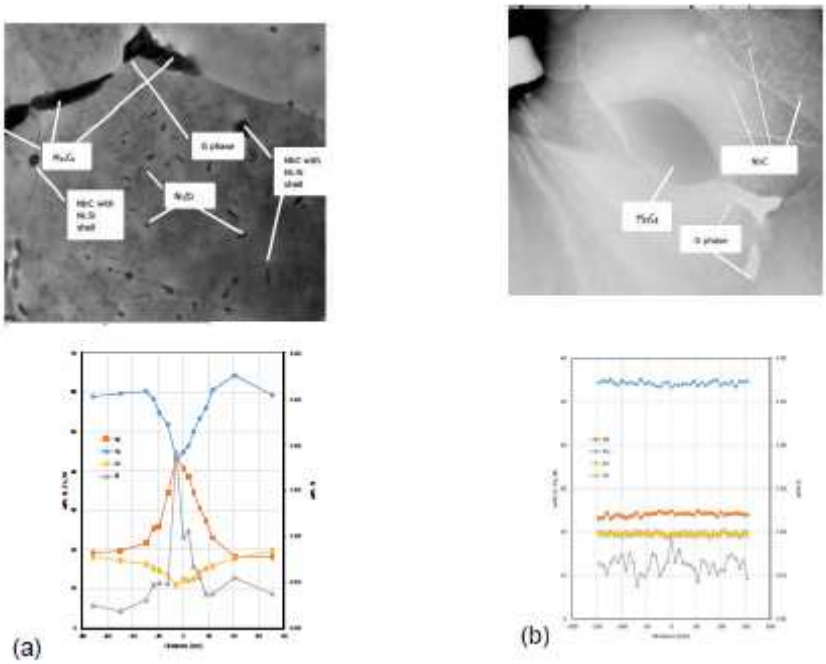
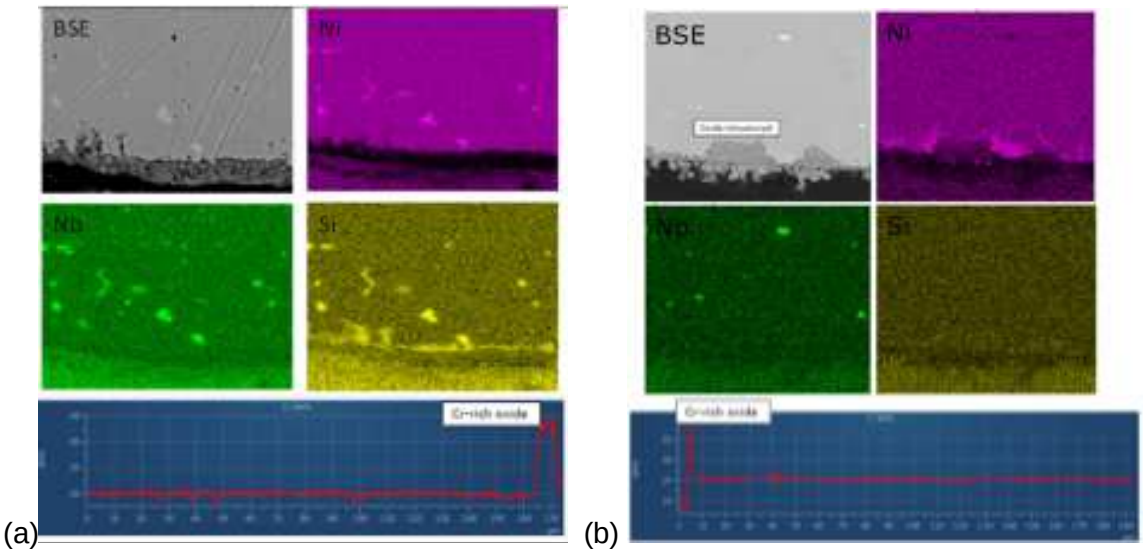


Figure 11: TEM micrographs and grain boundary compositions for high burn up AGR cladding (a) sensitized cladding (b) non-sensitised (higher temperature) cladding



(a) (b)

Figure 12:SEM micrographs, electron maps and linescans of for high burn up AGR cladding from (a) sensitised cladding (b) higher temperature cladding

4. Further Work

Further work is still to be carried out, and the results presented here (with the exception of those for corrosion inhibitor work) are initial results from work programmes in progress.[1] The PSE programme will eventually provide evidence as to whether there has been any corrosion of fuel cladding stored in a pH 11.4 caustic dosed storage pond for 25 years, and determine whether further degradation of clad condition and mechanical integrity occurs for pond failed fuel subsequently stored for a similar amount of time. Additional parts of the programme will also provide evidence of the effects of higher burn-up on the behaviour of fuel (intact and failed) pre- and post- pond storage. NNL has also commenced with an internal research project to explore the deployment of atomic force microscopy to “map” the development of corrosion sites. The same research project will also look at the feasibility of macro- and microstructural spent fuel simulants, in order to determine any available surfaces for water uptake, with respect to the final geological disposal environment.

5. Conclusions

NNL Windscale has performed many measurements on irradiated materials in support of the technical case for long term wet storage, covering a range of different PIE techniques. The current programme of work on PSE fuel has initially demonstrated that extended storage has not affected the integrity of sensitised fuel cladding, and the fuel responded well to impact testing as a means of replicating possible handling operations. Characterisation of the high burn up cladding is still in progress, but initial indications are that increased irradiation and extended dwell time do not have any adverse effects on the sensitised fuel i.e. grain boundary concentration profiles are consistent with those seen in historic studies. These studies are currently ongoing, but when combined with historic data and plant operational experience, the technical basis for long term, wet storage of AGR fuel appears to be promising.

[1] These initial results may not represent the final results and conclusions of the finished technical programme.

Acknowledgements

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