INL/CON-07-12208
PREPRINT

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International Topical Meeting on Research Reactor Fuel Management (RRFM)

D. E. Burkes
D. D. Keiser
D. M. Wachs
J. S. Larson
M. D. Chapple

March 2007

Transactions for the International Topical Meeting on Research Reactor Fuel Management (RRFM)
Lyon, France; March 2007

CHARACTERIZATION OF MONOLITHIC FUEL FOIL PROPERTIES AND BOND STRENGTH*

D. E. BURKES, D. D. KEISER, D. M. WACHS, J. S. LARSON AND M. D. CHAPPLE

Nuclear Fuels and Materials Division, Idaho National Laboratory
P. O. Box 1625, Idaho Falls, ID, U. S. A. 83415-6188

ABSTRACT

Understanding fuel foil mechanical properties and fuel / cladding bond quality and strength in monolithic plates is an important area of investigation and quantification. Specifically, what constitutes an acceptable monolithic fuel – cladding bond, how are the properties of the bond measured and determined, and what is the impact of fabrication process or change in parameters on the level of bonding? Currently, non-bond areas are quantified employing ultrasonic determinations that are challenging to interpret and understand in terms of irradiation impact. Thus, determining mechanical properties of the fuel foil and what constitutes fuel / cladding non-bonds is essential to successful qualification of plate-type monolithic fuel. Capabilities and tests related to determination of these properties have been implemented and are discussed, along with preliminary results.

1. Introduction

Monolithic fuel forms are necessary to convert high power nuclear reactors that could not otherwise be converted by low density dispersion fuels. Development of these fuel forms is essential to the success of the Reduced Enrichment for Research and Test Reactors (RERTR) program. Based on the success of initial irradiations of monolithic fuel plates, an aggressive campaign to further fabricate, irradiate and qualify monolithic fuel has been developed [1]. However, challenges associated with the planar interface introduced by a monolithic fuel form still remain, in particular bonding between the fuel and cladding across the interface. Therefore, understanding bond quality and strength in monolithic fuel plates is an important area of investigation and quantification. Specifically, what constitutes an acceptable monolithic fuel – cladding bond, how are the properties of the bond measured and determined, and what is the impact of the fabrication process or change in fabrication parameters on the level of bonding?

1.1 Approach

Currently, potential non-bond areas can be identified by employing ultrasonic determinations. Determination of what constitutes an unbound area and to what degree this constitution is acceptable with high confidence is challenging and somewhat unknown. This challenge creates difficulties in drawing correlations observed in post-irradiation examinations (PIE) with pre-irradiation fabrication observations. A series of tests aimed at addressing the challenges associated with acceptable bonding behaviour determination in monolithic fuel plates is underway at the Idaho National Laboratory. Understanding the bond 'quality' in monolithic fuel is essential to the successful qualification of monolithic fuel plates. Two approaches have been identified and are being investigated to determine the level and quality of bonding in the monolithic fuel plates. The first approach is through characterization of the bond layer that is fabrication technique specific, i.e. friction stir welding (FSW), transient liquid phase bonding (TLPB) and/or hot isostatic pressing (HIPing). The second approach is through determining the irradiation performance of the fuel plates, allowing correlations between fabrication processes and post-irradiation examinations to be drawn.

* Work supported by the U.S. Department of Energy, Office of National Nuclear Security Administration (NNSA), under DOE Idaho Operations Office Contract DE-AC07-05ID14517.

1.2 Characterization

Ultrasonic testing (UT) has shown some promise in determining the location and degree of non-bond areas. UT is highly desirable in the fact that the technique is non-destructive and provides information on bond quality in an efficient manner. On the other hand, mechanical testing is desirable in the fact that the technique provides quantitative information on bond strength. There are two different types of mechanical testing that can be carried out: a non-destructive technique such as a proof test, or a destructive technique such as an instrumented tensile test and/or shear test. A non-destructive proof test is coupled with UT and consists of applying a known torsional force at the ends of the fuel plate with a defined cycle, analyzing the fuel plate for non-bond areas from UT, and repeating the proof test over until a defined size of non-bond defect appears. This type of testing would demand an extremely high confidence in the UT method. A destructive instrumented tensile or shear test is carried out on both well-bonded areas and suspected non-bond areas determined by UT, and is the subject of the current paper. Changes to fabrication process parameters or conditions, e.g. addition of a secondary interface such as a diffusion barrier, lower HIPing temperatures, etc., and the impact these have on bond strength will be more easily identified and understood prior to irradiation. Microstructural characterization is carried out in a similar manner as that defined for the destructive mechanical tests. Both suspected well-bonded and non-bonded areas are sectioned creating a metallographic specimen that is mounted, prepared and examined with optical microscopy and scanning electron microscopy (SEM). Any reaction layer existing between the monolithic fuel and the cladding is clearly visible and quantified in terms of thickness and composition (employing a semi-qualitative technique such as energy dispersive spectroscopy). The microstructural method is carried out in conjunction with the ultrasonic testing and mechanical testing, creating an ensemble of information relating to bond strength and integrity.

1.3 Irradiation Performance

The general irradiation performance evaluation of bonding in monolithic fuel plates is carried out in two basic areas: modelling and post-irradiation examination (PIE). Although the modelling approach is not discussed at this time, the approach consists of finite element analysis and analytical solutions relating to both thermal and thermo-mechanical aspects of the fuel-clad interface. Specifically, these models investigate the impact of a debond on the fuel meat temperature and stress behaviour, ultimately supporting determination of an acceptable debond size and geometry. The post-irradiation examination (PIE) approach involves examination of plates previously characterized by UT scans, pull tests and/or microstructural analysis after irradiation. Specific results on PIE of the latest monolithic fuel campaign (RERTR-6) may be found in Ref. [2]. Combination of these two approaches allows observations from irradiation to be fed back into fabrication to improve subsequent irradiation experiments, utilizing characterization as an effective means to understand how and what has changed in terms of bond strength and integrity.

2. Experimental Methods

Sample plates were fabricated employing one of three processing methods, hot-isostatic pressing (HIP), transient liquid phase bonding (TLPB) or friction stir welding (FSW). An updated description of each process can be found in Ref. [3]. All of the sample plates contained a DU-10Mo (nominal wt.%) foil approximately 8.26 cm long by 1.91 cm wide with aluminium-6061-T6 used as the cladding. HIP sample plates were subjected to 580^{o}C for ninety minutes at 103 MPa pressure. TLPB sample plates were subjected to 590^{o}C for fifteen minutes at 6.89 MPa pressure. FSW sample plates were welded with an approximate load of 17.8 kN and an unknown temperature, although the processing temperature is believed to be in the range of 400-500 C. Sample plates were subjected to ultrasonic testing to determine whether or not debonds were present. Regions of interest (ROI's) were determined from the UT scans and marked. Test specimens (ROI's) were sectioned from the sample plates using a low-speed saw. Each test specimen was a square approximately 0.876 cm on edge. One test specimen for each fabrication method was bound to aluminium test platens using a high strength epoxy. Bonding of the epoxy involves a heat treatment of 165^{o}C for ninety minutes after application. The low temperature heat treatment does not affect reaction kinetics or growth of an interfacial layer in a significant manner. Pull testing was carried out on the test specimens, similar to that used in determination of bond strength between thermally sprayed coatings and a substrate [4]. An in-house test rig, shown in Fig. 1 along with a photograph of a mounted sample, was employed to carry out the pull tests. A constant crosshead rate was applied to pull the test while monitoring induced load with a tensile link load cell. A second sample from each fabrication method was cold mounted, polished using SiC paper and examined under a scanning electron microscope (SEM).

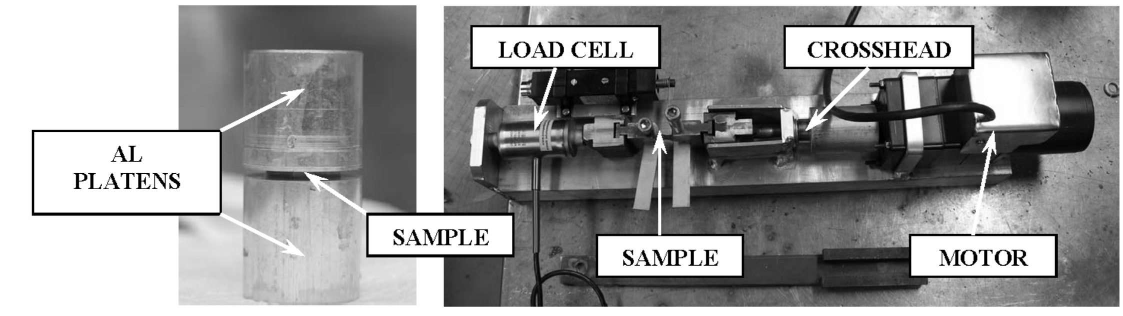

Fig. 1. Photograph of test rig employed to carry out pull tests. Pertinent features of the rig are pointed out

3. Results and Discussion

Ultrasonic testing scans of each sample plate are provided in Fig. 2. Regions of light [white] areas suggest acceptable bonding between the aluminium-aluminium cladding. Regions of light [grey] areas suggest acceptable bonding between the aluminium-fuel interfaces. Regions that appear dark in colour would suggest either a debond or inclusion/impurity in the fuel foil. However, observation of Fig. 2 reveals that this is clearly not the case for each of the sample plates fabricated using HIP, TLPB or FSW, and that each plate, based on this technique, has bonding between the fuel and cladding. Examples of SEM photomicrographs of the fuel-clad interface for each fabrication technique investigated are presented in Fig. 3. Observation of the photomicrograph for a HIP fabricated fuel plate reveals a relatively thin, uniform reaction layer, approximately 6 μm thick. The TLPB fabricated fuel plate contains a thicker (38 μm thick), non-uniform reaction layer that consists of multiple phases visible on the photomicrograph, i.e. regions A, B, C and D. Finally, the FSW fabricated fuel plate shows relatively no reaction layer at all.

Stress-time plots for each sample pull tested are provided in Fig. 4. The dashed line in the figure indicates the approximate limit (20 MPa) of the test rig, above which the crosshead is turned manually employing a wrench until failure of the interface or epoxy occurs. Observation of these plots show a steady increase in stress until catastrophic failure occurs. Samples are pulled normal to the fuel-clad interface. The HIP specimen profile reveals that the sample has bond strength of 60.3 MPa. However, failure of the epoxy occurred before that of the fuel clad interface, so that the actual bond strength, although unrealized in this plot, is greater than 60.3 MPa. An alternative test method, such as a peel test, will be used to quantify the bond strength of samples with strength greater than that of the epoxy. Currently, 60 MPa is established as acceptable bond strength, since no failures after irradiation have been observed with plates fabricated in this manner, at this time. The TLPB specimen has the second highest strength at 15.4 MPa, while the FSW specimen has the lowest bond strength at 6.42 MPa. Also observed from the stress-time plots is the significant difference in stress rate between the TLPB specimen and the HIP and FSW specimens. Since specimens are subjected to a constant rate up to the approximate limit of the test rig, variations in stress rate can provide some initial insight into the integrity of the as-fabrication reaction layer at the fuel-clad interface. The TLPB specimen has an approximate stress rate of 0.04 MPa•sec^{-1}, while the HIP and FSW specimens have approximate stress

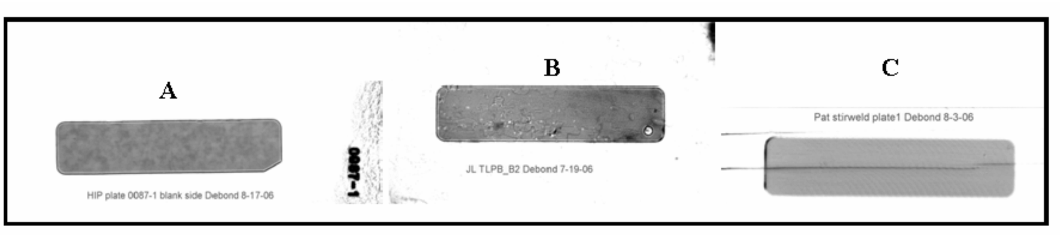

Fig. 2. Ultrasonic testing scans of (A.) HIP fabricated fuel plate, (B.) TLPB fabricated fuel plate and (C.) FSW fabricated fuel plate

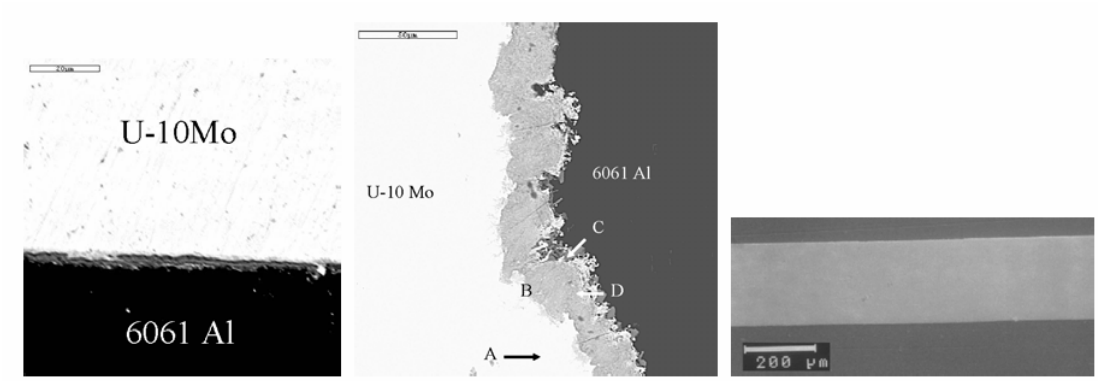

Fig. 3. SEM micrographs of the reaction layer formed from (left) HIP fabrication process, (middle) TLPB fabrication process and (right) FSW fabrication process

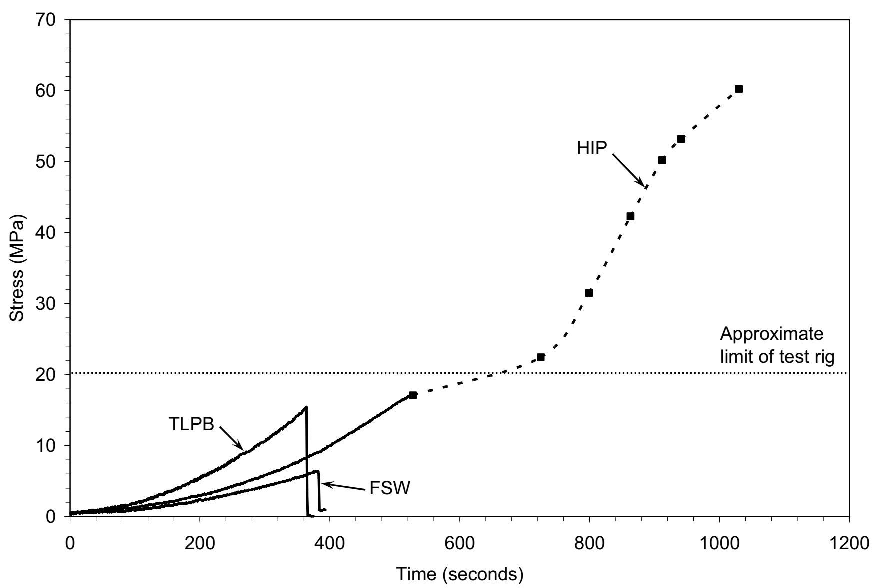

Fig. 4. Stress-time plots for test specimens obtained from pull test fabricated by each [HIP,TLPB,FSW] method

rates of 0.031 and 0.016 MPa•sec^{-1}, respectively. Hence, a hypothesis may be drawn that the eutectic formation of Al-12Si for the TLPB process behaves in a manner expected of a brittle intermetallic, i.e. high stress rate with sudden failure. In addition, this result appears to suggest that diffusion of aluminium into the foil is significant and results in a thick reaction layer, ultimately lowering the bond strength. Conversely, the FSW specimen has a low stress rate and low bond strength, suggesting that the bond is more mechanical than diffusional. This also seems intuitive since the weld tool has a low thermal conductivity compared to aluminium. Thus, as the FSW process progresses along the plate, lower loads are applied in order to compensate for the increased temperature, i.e. heat builds up in the plate and is not conducted away from the weld face. Further increases in temperature would ultimately result in increased aluminium plasticity and promote void formation or disturbance of the monolithic fuel foil. The weld surface temperature can additionally be controlled by modifying the weld tool alloy. Increasing the thermal conductivity of the tool face has been shown to significantly increase the bond strength [5]. Finally, the HIP specimen shows the ideal trade-off between fabrication temperature and pressure, promoting diffusion of atoms across the fuel-clad interface resulting in bonding, but not to a degree where the brittle intermetallic nature of the bond dominates the behaviour.

4. Conclusions

The first series of mechanical characterization tests on monolithic fuel plates fabricated by hot isostatic pressing, transient liquid phase bonding and friction stir welding has been carried out. These tests allow a greater understanding of bond strength characteristics and performance prior to irradiation, so that improved correlations between fabrication processes, foil microstructure characteristics and post-irradiation properties can be determined, enhancing the success of the RERTR fuel development campaign. Initial results show that HIPed samples provide the highest bond strength while FSW samples, fabricated in the current manner, provide the lowest bond strength.

5. References

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