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UPDATE ON FRESH FUEL CHARACTERIZATION OF U-MO ALLOYS

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ABSTRACT

The need to provide more accurate property information on U-Mo fuel alloys to operators, modellers, researchers, fabricators, and government increases as success of the GTRI Reactor Convert program continues. This presentation provides an update on fresh fuel characterization activities that have occurred at the INL since the RERTR 2008 conference in Washington, D.C. The update is particularly focused on properties recently obtained and on the development progress of new measurement techniques. Furthermore, areas where useful and necessary information is still lacking is discussed. The update deals with mechanical, physical, and microstructural properties for both integrated and separate effects. Appropriate discussion of fabrication characteristics, impurities, thermodynamic response, and effects on the topic areas are provided, along with a background on the characterization techniques used and developed to obtain the information. Efforts to measure similar characteristics on irradiated fuel plates are discussed.

1. Introduction

Characterization of fresh U-Mo fuel alloys is a key element of the GTRI Reactor Convert fuel development program for three main reasons: (i) effectively document available information for current and future fuel and material researcher use, (ii) establish a link between fresh fuel properties and irradiated fuel observations so that the effect(s) of irradiation can be confidently deduced, and (iii) provide a feedback loop into the fuel plate preparation methods to optimize behaviours for additional fabrication processes and the best possible irradiation performance. As the fuel development campaign progresses, the necessity of bridging the gap between lab-scale process demonstration and commercial scale development intensifies. Characterization of fabrication processes and material properties is at the forefront of bridging this gap, so that continuity between lab-scale and bulk production is maintained. For example, the INL employs arc melting (lab-scale) for mini-plate experiments while Y-12 has employed induction melting (bulk production) for full-size plate experiments. Two different encapsulation methods are being investigated: friction bonding at the INL for mini- and full-size plate experiments and hot isostatic pressing at the INL for mini-plate experiments and Babcock & Wilcox for full-size plate experiments. Forming operations required for some reactor designs also introduce considerations for processing. The continuity of these two scales is of the utmost importance to ensure that desired fuel performance behaviour and attributes are maintained.

For the most part, this paper will serve as a progress report of the characterization techniques being employed and developed at the Idaho National Laboratory (INL) since the RERTR 2008 conference in Washington D.C. Specific progress has been made performing needed material property measurements and further identifying areas where improved material property information is needed to support fuel fabrication and qualification. Furthermore, progress has been made on characterization technique development in terms of equipment procurement and installation.

2. Thermophysical Property Measurements

Thermal properties of nuclear fuel, specifically thermal conductivity, play a vital role in fuel performance. However, evaluation of these properties is both challenging and very expensive to determine experimentally. Furthermore, fuel performance codes must assume

extrapolated thermal property values or values calculated employing the Neumann-Kopp approximation, even though there is insufficient data for pure elements in some cases at elevated temperatures. Thermal conductivity is most commonly determined experimentally employing three different measurement methods: differential scanning calorimetry (DSC) for specific heat capacity, dilatometry for density, and laser flash for thermal diffusivity (LFTD). Errors associated with each of these measurements can be compounded in determination of thermal conductivity, and must be appropriately understood.

Measurements have been conducted on alloys encompassing the proposed range of fuels, i.e. U alloyed with 7 to 12 wt% Mo, with each of these techniques. Combined data plots for each of these alloys and techniques are provided in Fig. 1. For the alloy containing 10 wt% Mo, the specific heat capacity, density, and thermal diffusivity behaviour is as expected, from near room temperature up to 800°C. For the alloys containing 7 and 12 wt% Mo, a change in behaviour is observed between 400 and 600°C. In both cases, the behavioural change is attributed to decomposition of the desired high temperature γ-U(Mo) phase into α-U(Mo) and/or U_2Mo. Based on time-temperature-transformation diagrams (TTT) on the U-Mo system, the rate at which the quenched γ-U(Mo) phase will decompose into α-U(Mo) phase in a homogeneous alloy is strongly dependent upon the isothermal treatment temperature, being more pronounced when the alloy is near the eutectoid temperature (550°C) [1,2]. The α-U phase forms continuously by a cellular matrix decomposition reaction as a function of time, nucleating primarily at sub-grain boundaries and leading to precipitation at the grain boundaries [3].

For the 7 wt% Mo alloy, decomposition of γ-U(Mo) into α-U(Mo) is relatively rapid as defined by the TTT diagram, with the kinetics becoming much more sluggish with increased wt% Mo. For the 12 wt% Mo alloy, decomposition would not necessarily be expected. However, the foil sample used for DSC measurements contained a significant amount of chemical banding that resulted from inadequate homogenization during the arc-melting fabrication process. Conversely, the cast samples used for dilatometry and LFTD did not exhibit the banding. Chemical banding is the result of Mo-rich and Mo-lean zones, and XRD and

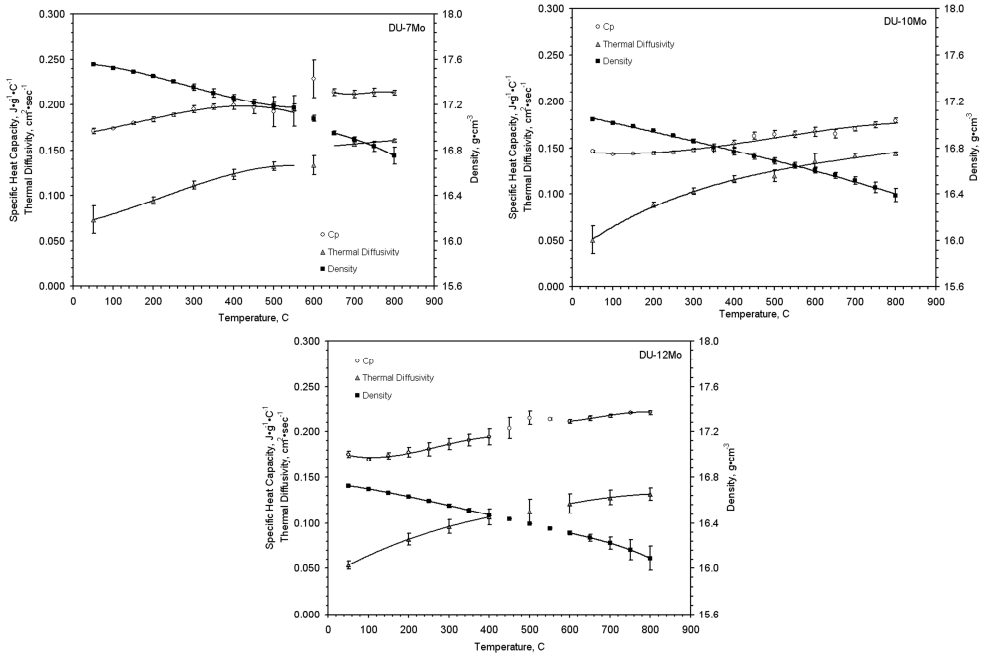

Fig. 1. Combined specific heat capacity, density, and thermal diffusivity data plots for representative DU-7, -10, and -12 wt% Mo alloys

microhardness measurements performed on the foil sample more accurately placed the Mo content in the 13-14 wt% range, rather than the nominal 12 wt% range [4,5]. The kinetics

associated with the decomposition of γ-U(Mo) into α-U(Mo) and U_2Mo begin to accelerate rapidly beyond the 12 wt% content. Thus, thermal cycling of the samples during measurements on these alloys will result in a mixed microstructure of γ-(Mo), α-U(Mo), and U_2Mo for the 7 and 12 wt% Mo alloys, rather than a single phase as was the case for the 10 wt% Mo alloy. The thermophysical properties of these phases are different, and therefore result in unexpected alloy behaviour. This is demonstrated by the thermal conductivity determined by the combined data plots presented in Fig. 1. The results of these determinations are provided graphically in Fig. 2. These measurements illustrate the importance of solid characterization and understanding of alloy behaviour on desired material attributes, such as thermal conductivity. Furthermore, values available from literature were determined by electrical conductivity measurements and converted to thermal conductivity employing the Wiedemann-Frantz law. Thermal conductivity measurements obtained from electrical conductivity are lower than those measured in a direct or semi-direct manner, since electrical conductivity only considers the electronic contribution to thermal conductivity and phonon-phonon scattering is not taken into account.

Sound knowledge of the fresh fuel thermal conductivity is necessary in order to determine separate effects such as burn-up and fuel temperature. In addition, in order to understand overall integrated behaviour of the fuel plate, one must be able to effectively deconvolute the effects of modifications made to the fuel alloy, including any modification made to the interface, e.g. Si or Zr.

3. Instrumented indentation

One of the most important aspects of monolithic fuel forms is the behaviour of the interface. Integrity of the bond between the U-Mo monolith and cladding material must be measured as a function of processing method, processing parameters, and interface modifications, e.g. Si or Zr. Instrumented indentation offers a unique capability to measure the normal and lateral forces simultaneously in order to determine the cohesive strength of the interface. Basically, a conical indenter is placed near the interface of interest and a load applied. As the normal load increases, lateral force changes accordingly as the tip is pushed away from the cohesive interface. As the interface delaminates, the indenter moves backward towards the interface as the normal load continues to increase. Thus, a change in slope for the F_x-t curve corresponds to delamination or cracking, and the decohesive force is taken as the normal load at that point. A FEA model can be developed to calculate cohesive strength of

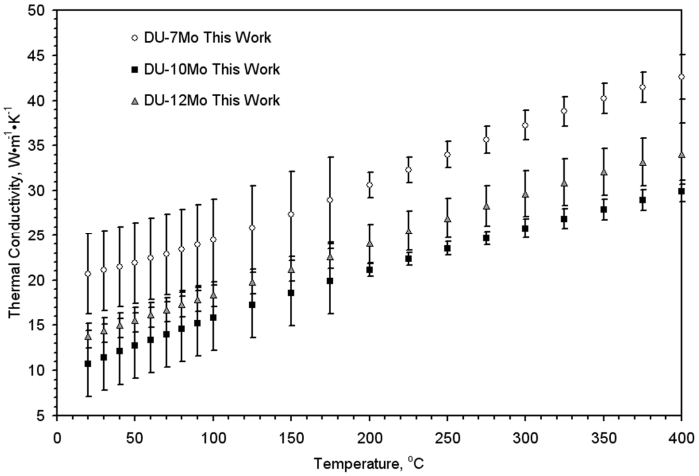

Fig. 2. Thermal conductivity of the DU-7, -10, and -12 Mo alloys determined from the combined data plots in Fig. 1.

the interface from the normal load. A tribometer has been procured from CETR (California) to perform this function at the INL. The instrument is capable of both vertical and lateral displacement measurement, modular load capacity (up to 1kN), and comes equipped with an

optical microscope, acoustic emission detector, and an environmental chamber capable of temperatures up to 1000°C.

4. Shear Punch Testing

Small scale specimen testing is desirable to determine mechanical properties of fuel alloys. Specifically, such techniques are desirable to correlate beginning of life mechanical behavior with end of life behaviour in terms of burn-up and temperature, while also investigating the effects of microstructure, impurities, and the interface layer. Shear (or small) punch testing is ideal for this purpose and is based on blanking a circular disk from a fixed sheet specimen. The technique is relatively simple and straightforward in the fact that a cylindrical punch with a flat end is utilized to punch a hole in the circular disk. The technique is currently being developed for both out-of-pile and in-cell measurements. Deformation and failure processes that occur during shear punch testing are analogous to those that occur in uniaxial tension. The load displacement data can be interpreted in terms of and correlated with uniaxial mechanical properties data (flow properties such as yield stress, UTS, work hardening exponent and ductility parameters).

Verification and validation of the SPT fixture and procedure is continuing out-of-pile. Stainless steel 304 samples are being employed as a calibration standard. SPT measurements performed on these calibration samples are in good agreement with uniaxial tension measurements. Furthermore, the technique is being validated on fresh U-Mo alloy samples. Examples of SPT measurements performed on a DU-10 wt% Mo alloy are provided in Fig. 3. In-cell SPT development continues, with an Instron being prepared for insertion into the INL Hot Fuel Examination Facility in mid March. A photograph of the instrument is provided in Fig. 3. The first SPT on irradiated fuel alloys will begin in mid May on the AFIP-2 experiment.

5. Through-thickness Flash Thermography

Through-thickness flash thermography is an alternative non-destructive evaluation tool currently being investigated at the INL for use on plate-type fuels. Theoretically, the technique should allow the minimization of errors, increase product yield due to reduced measurement time, increase fuel performance confidence, and offer additional information to better understand potential effects that may result in performance deterioration. The technique can be qualitative or quantitative. Qualitative analysis uses differences in contrast to identify areas with flaws or material differences. Quantitative analysis determines the specimen's thermal diffusivity, α, as a function of location. The technique is

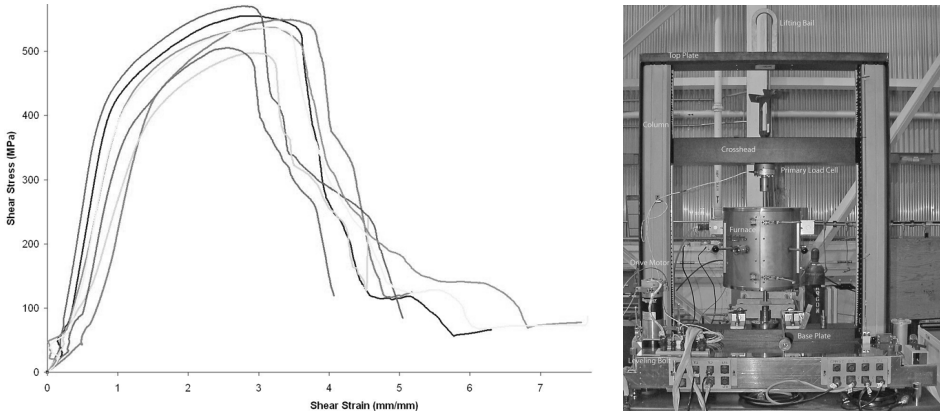

Fig. 3. Examples of SPT measurements performed on a fresh, DU-10Mo alloy (left) and a photograph of the in-cell Instron at INL (right)

Fig. 4. Simple schematic of the through-thickness flash thermography technique illustrating qualitative analysis capability (left); Example thermal diffusivity profile obtained from through-thickness flash thermography illustrating quantitative analysis capability (right).

currently being evaluated for application to both out-of-pile and in-cell measurements. A simple schematic of the through-thickness flash thermography method and an example thermal diffusivity profile is provided in Fig. 4.

6. Conclusions

Fresh fuel characterization is a key element in understanding and predicting fuel behaviour, optimization of fabrication processes to tailor properties to desired performance, and vital to transfer of knowledge and technologies to current and future researchers and developers. A number of characterization techniques are being utilized at the INL to accomplish the mission of qualifying usable U-Mo dispersion and monolithic fuel forms. This has culminated in vast educations and experiences to the benefit of the GTRI Reactor Convert Program. Specific areas of technique development to provide links to use out-of-pile characterization to better understand post-irradiation observations are also essential. These techniques must therefore be developed for both beginning of life and end of life measurement.

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