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ABSTRACT

The need to provide more accurate property information on U-Mo fuel alloys to reactor operators, modelers, researchers, fabricators, and regulators increases as success of the RERTR program continues. This presentation will provide an overview of fresh fuel U-Mo characterization activities on monolithic fuel occurring at the Idaho National Laboratory. The overview will particularly be focused on properties available through current and previous research, and also on the type of information still needed. The presentation will deal with mechanical, physical, and microstructural properties in terms of both integrated and separate effects. Appropriate discussion in terms of fabrication characteristics, impurities, thermodynamic response, and the effects on the topic areas will be provided, along with a brief background on the characterization techniques being used or being developed to obtain the information. Furthermore, efforts to measure similar characteristics as a function of irradiation conditions and determine end-of-life observations with beginning-of-life behavior will be discussed.

1. Introduction

Characterization of fresh U-Mo fuel alloys is a key element of the RERTR 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 behaviors for additional fabrication processes and the best possible irradiation performance. The classical materials science tetrahedron, provided in Figure 1,

shows the need for characterization to unite structure, processing, properties, and performance with one another. Different processing methods can be employed, depending on the scale of experiment or production. For example, the INL has traditionally employed arc melting for mini-plate experiments while Y-12 has employed induction melting for full-size plate experiments. Two different fabrication 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.

Separate and integrated properties must be carefully considered, in terms of the U-Mo fuel alloy, Aluminum Alloy 6061 (AA6061) cladding, and composite behavior. Processing, microstructure, and impurities can significantly influence the mechanical properties of the alloy and/or the fuel foil. Inadequate homogenization, such as that obtained from arc melting, can result in off-normal property measurements. Fuel to cladding interface behavior can also be affected by process parameters and is a key component in the success of monolithic fuel.

Different structures can result based on the alloy composition, both in terms of crystallographic nature and grain size. Structure is important in terms of desired phase(s) present, fission product retention, release, and/or migration, which can all significantly influence fuel and composite swelling.

Finally, expected and desired performance must be clearly defined in terms of target burn-ups, normal and off-normal events, and requirements for fuel qualification. For example, the fuel will undergo significant changes in time, temperature, and irradiation conditions. Furthermore, normal events could include vibration and off-normal events could involve thermal shock. All these effects must be well understood through characterization, so that a change at one end of the tetrahedron does not translate to an unexpected event at another end.

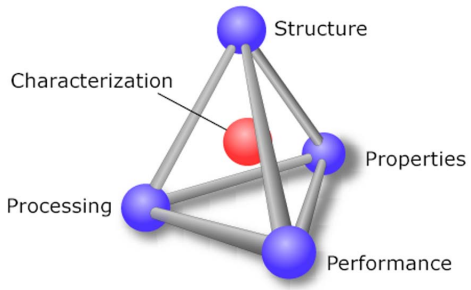

Fig. 1. Materials Science Tetrahedron showing the role of characterization in the relationship between processing, performance, properties, and structure.

2. Current Fresh Fuel Characterization Methods

Physical Metallurgy

Optical microscopy is most often used to evaluate the influence of the foil fabrication method on the microstructure of the alloy. Arc melting, employed at the INL for small scale experiments, can result in inadequate homogenization of Mo, creating Mo-rich and Mo-lean zones termed chemical banding. An example of chemical banding in a U-10wt% Mo alloy is provided in Figure 2. Conversely, given the increased electro-magnetic mixing time and cooling time with induction melting, used at Y-12 for large scale experiments, improved homogenization can be achieved. An example of a U-10wt% Mo alloy cast by induction melting and hot rolled is also provided in Figure 2. Note the lack of chemical banding in induction cast alloy.

Mechanical Testing

Mechanical properties dominate during fabrication, given the magnitude of stresses associated with some of the fabrication methods, the presence of a planar interface, and fuel dominating the overall composite behavior. In addition, during irradiation, the response of the fuel plate to loads (vibratory or shock) is central for configuration control. These particular properties can be influenced by microstructure as a result of either chemical banding or selected alloy composition. An example of the influence of chemical banding on microhardness measurements is provided in Figure 3, where the nominal DU-12wt% Mo alloy had a grain size the same order of magnitude as the indenter diagonal. Since this particular alloy had a significant degree of chemical banding (Mo-rich zones), the microhardness value obtained was much greater than expected for the nominal composition. The DU-11wt% Mo alloy had very little chemical banding and as a result had "expected" linear behavior. Figure 4 shows an example of the influence of alloy content on ductility, i.e. increase in Mo content results in increased ductility. Also, Figure 4 shows the

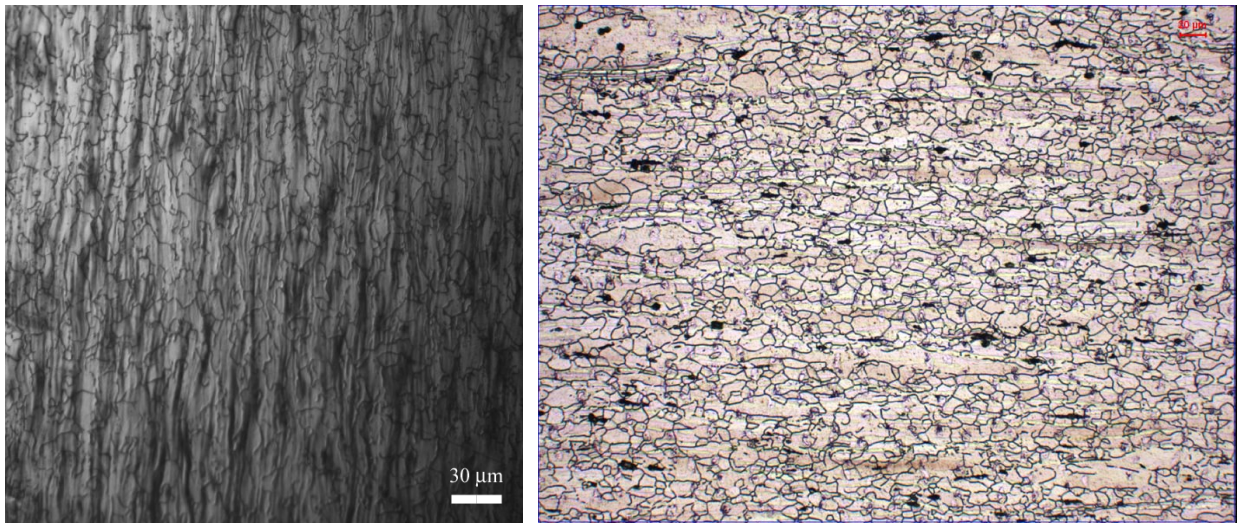

Fig. 2. Example metallograph of an arc-melt and cast DU-10wt% Mo alloy (left) and an induction cast U-10wt% Mo alloy (right). Both alloys were hot rolled. Note the significant differences in terms of chemical banding.

influence of annealing temperature and duration on DU-10wt% Mo foils fabricated at INL. Specifically, an increase in both temperature and annealing duration results in a significant increase in yield strength. Furthermore, it is important to note that rolled foils, which did not undergo a homogenization treatment, resulted in lower yield strength values than an induction melted as-cast sample that inevitably had better homogenization.

Microscopy

Scanning electron microscopy (SEM) and transmission electron microscopy (TEM) are very important tools in determination of interface properties, failure analysis, and impurity effects. For example, SEM combined with energy and wavelength dispersive spectroscopy (EDS and WDS, respectively) are vital to determination of Si and Zr modifications made to

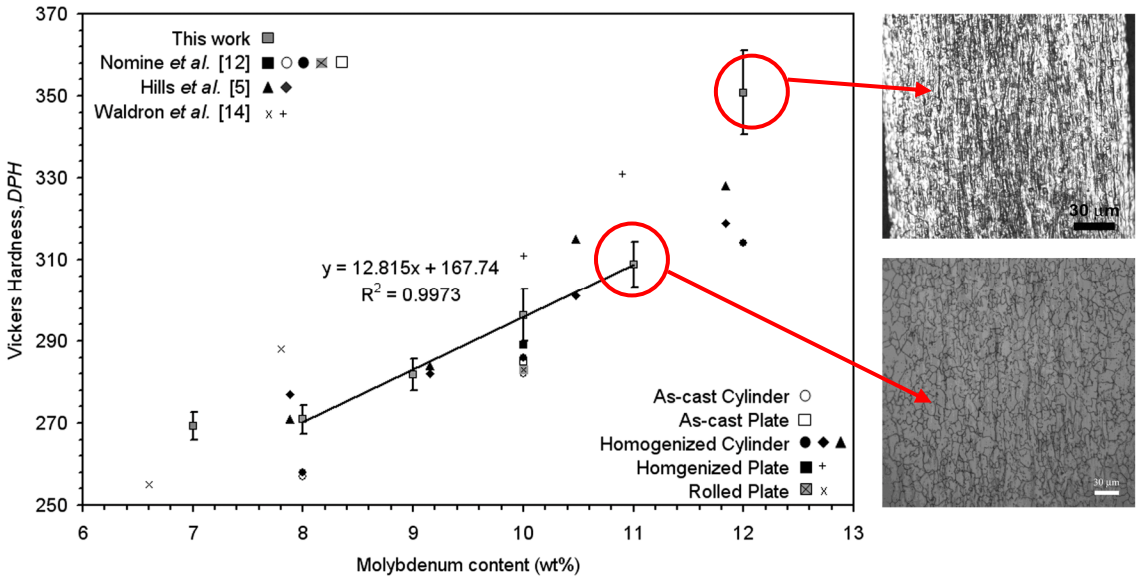

Fig. 3. Alloy microhardness as a function of Mo content. Note the significant increase in hardness for the nominal 12wt% Mo alloy, resulting from the significant chemical banding compared to the near linear behavior of the nominal 11wt% Mo alloy with minimal chemical banding.

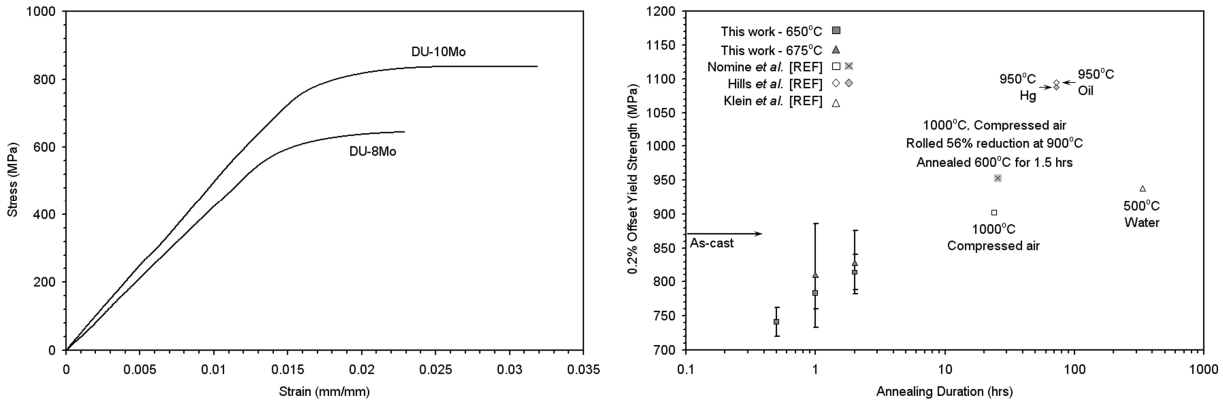

Fig. 4. Example of the influence of Mo on the alloy ductility (left), and the influence of annealing temperature and duration on yield strength of a DU-10wt% Mo alloy.

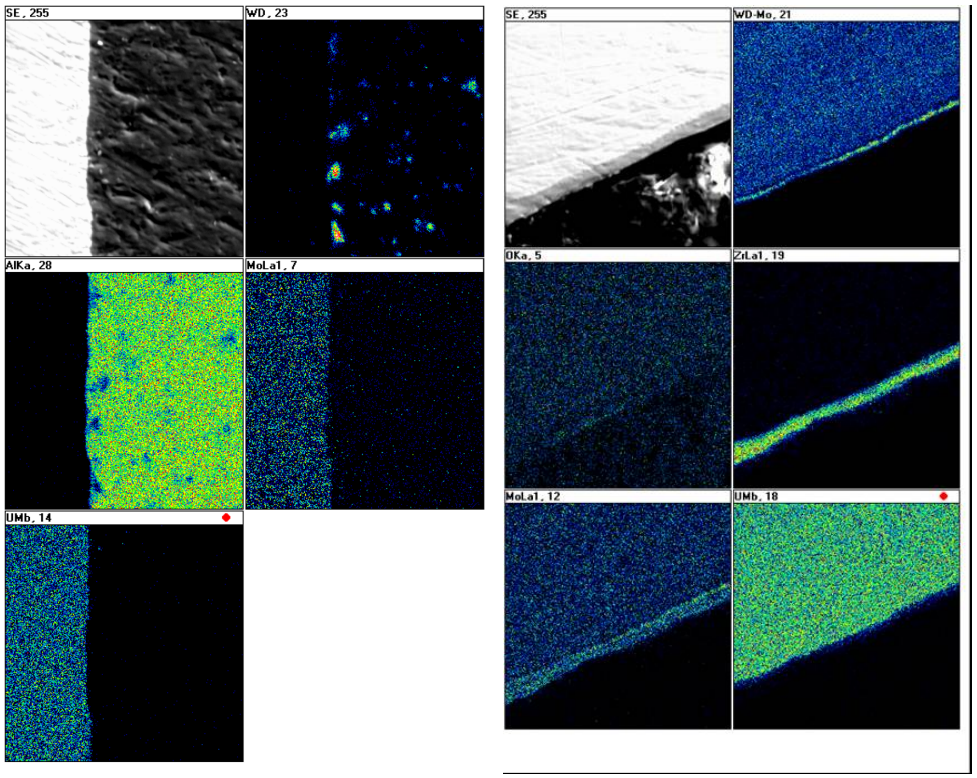

Fig. 5. Examples of SEM and WDS images of a U-10wt% Mo fuel foil clad in AA6061 by Friction Bonding with elemental Si (left) and Zr (right) at the interface.

the monolithic fuel interface. Impacts of plate processing method can also be investigated. Examples of microscopy performed on Si and Zr interface modifications are provided in Figure 5. Note the significant dispersion of Si at the planar interface as a result of friction bonding, i.e. the process breaks the Si coating up and flow of the aluminum moves the material away from the interface. Without this effect, bonding between aluminum and U-Mo might not be achieved. Also note the enrichment of Mo at the Zr interface in Figure 6 that results during hot rolling and post-process annealing, given the high solubility between Mo and Zr.

Failure mode can provide significant insight on the impurity concentration of the alloy. Carbon results in inclusions that decrease the strength of the alloy, hydrogen can result in stress corrosion cracking, and oxygen can embrittle the alloy. Examples of failure modes caused by oxygen and carbon impurities are provided in Figure 6. Low carbon concentrations result in purely decohesive grain rupture, while high carbon concentrations result in ductile dimple fracture. Similarly, high oxygen concentrations result in grain embrittlement and decohesive rupture.

Carbon impurities are mainly introduced either as carryover from the feedstock material or during the casting operation, i.e. by using graphite crucibles. Fabrication processes often set the limit of carbon in the alloy, typically defined at 250 ppm or less. TEM photomicrographs of typical carbon inclusions in a U-10wt% Mo alloy are provided in Figure 7. The carbon inclusions manifest themselves both in the grain interior and at the grain

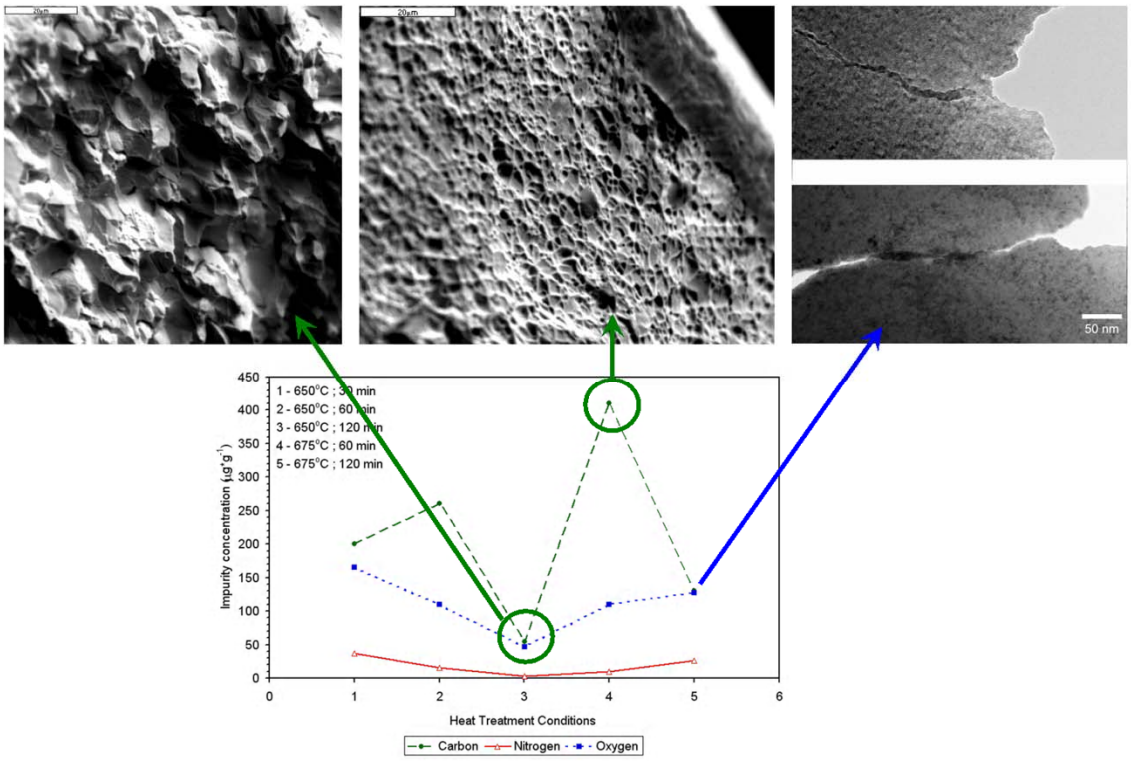

Fig. 6. Example of decohesive rupture and ductile dimple rupture failure modes resulting from carbon (center) and oxygen (right) impurities in a DU-10wt% Mo alloy.

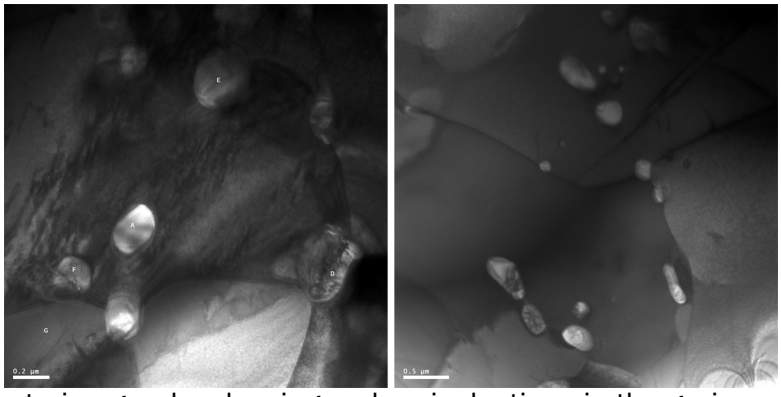

Fig. 7. TEM photmicrographs showing carbon inclustions in the grains and at the grain boundaries of a U-10wt% Mo alloy.

boundary. Ultimately, it is these inclusions that create regions of localized strain discontinuity that nucleate microvoids, which grow and coalesce as the foil is deformed, and eventually create a continuous fracture surface and the cuplike dimples observed in Figure 6.

Thermophysical properties

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. Often, thermal conductivity is determined experimentally employing three different measurement methods: calorimetry for heat capacity, dilatometry for density, and laser flash for thermal diffusivity. Errors associated with each of these measurements can be compounded in determination of thermal conductivity, and must be appropriately understood. Determination of separate effects such as burn-up and fuel temperature is another key requirement for thermal analysis of U-Mo fuel alloys, as is the overall integrated behavior of the fuel plate, including any modification made to the interface, e.g. Si or Zr.

Non-destructive analysis

Currently, ultrasonic testing (UT) is employed as a non-destructive technique for both monolithic and dispersion type fuels. The technique is based on the transmission of sound through the integral fuel plate to determine areas of poor or incomplete bonding. Furthermore, the technique can determine the thickness of cladding material over the fuel zone based on measuring reflection of sound to the interface. The technique, however, is somewhat sensitive to surface defects, as illustrated in Figure 8, and to edge effects, where an interface might be deflected at a very sharp angle. These sensitivities can be reasonably accommodated, but must be well understood through characterization efforts.

3. Specific Areas of Technique Development

Instrumented indentation

One of the most important aspects of monolithic fuel forms is the behavior 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. An example of an F_x-t curve illustrating this behavior is provided in Figure 9 [1]. A FEA model can be developed to calculate cohesive strength of the interface from the normal load.

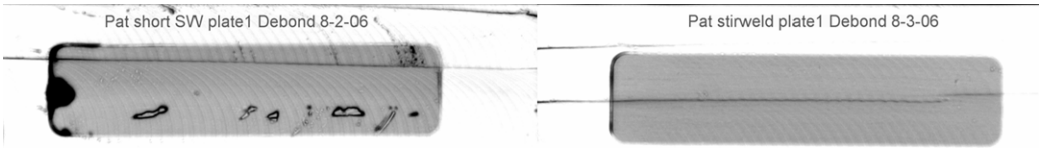

Fig. 8. Ultrasonic testing scans of an as-fabricated plate with surface defects (right), and a scan of the same plate after surface finishing (left).

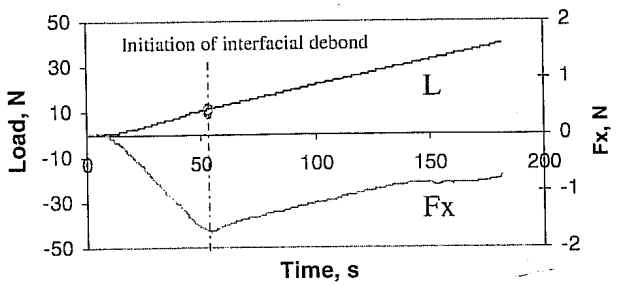

Fig. 9. Example of a lateral force as a function of time plot showing decohesion of the interface.

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 behavior 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 [2]. 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). An example of a typical shear punch test set-up is provided in Figure 10 [3].

Positron Annihilation Spectroscopy

Positron Annihilation Spectroscopy (PAS) is a non-destructive characterization technique that is sensitive to open-volume defects. A new, Doppler broadening PAS system has recently been developed at INL. The new system is unique in that measurements are spatially resolved, rather than just single point measurements. The PAS technique provides a good indication of a materials residual stress and is currently being developed for cladding materials. The Doppler broadening technique is based on the shape factor created by the g-ray energy of 511 keV characteristic of positrons annihilating electrons in the outer shell. The greater the shape factor, the greater the amount of material damage, or residual stress.

Hot Disk Technique

The hot disk technique is currently being developed for both out-of-pile and in-cell Integrated and separate effect thermal behavior measurements. The technique offers the unique advantage of understanding deterioration in thermal properties of a fuel without the significant cost and time required to carry-out the experiment in-reactor. Both integrated and separate effects such as burn-up and temperature, influence of micro-cracks, fission products, porosity, Frenkel defects, on thermal performance can be evaluated. The hot disk technique is based on modified transient plane source technique. A one-sided, interfacial, heat reflectance sensor applies constant heat source to the sample where the known current provides a small amount of heat. The concomitant heat results in a slight interfacial

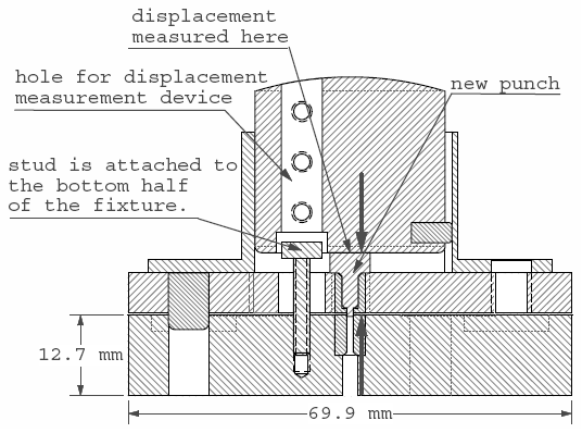

Fig. 10. Typical set-up of the die and punch for a shear punch test.

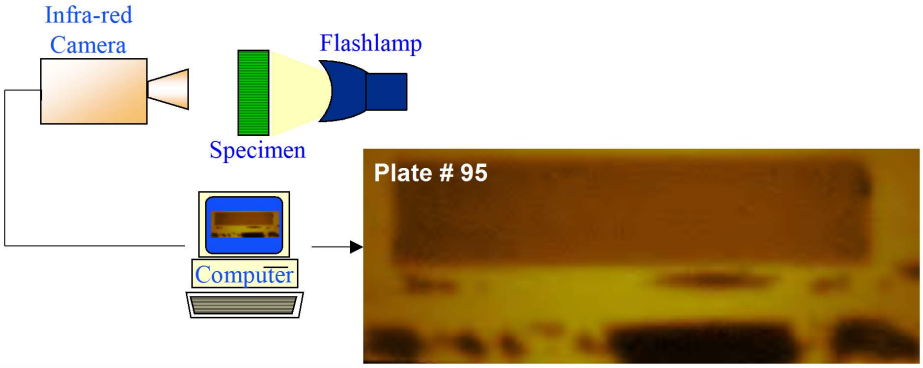

Fig. 11. Example schematic identifying pertinent features of the through-thickness flash thermography system.

temperature rise, resulting in subsequent voltage drop of the sensor. The thermo-physical properties of the sample are inversely proportional to the rate of change in sensor voltage (more insulating = steeper voltage rise). Thermal conductivity and effusivity are measured directly from the sample – non-destructively.

*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 with minimized errors, 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 currently being evaluated for application to both out-of-pile and in-cell measurements. A sample schematic of the through-thickness flash thermography method is provided in Figure 11.

4. Conclusions

Fresh fuel characterization is a key element in understanding and predicting fuel behavior, 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 RERTR. 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.

5. References

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[3] M.B. Toloczko al., JNM 307-311, (2002).

6. Acknowledgements

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