INL/CON-15-37380
PREPRINT

# U.S. Progress on Property Characterization to Support LEU U-10 Mo Monolithic Fuel Development

## European Research Reactor Conference (RRFM/IGORR 2016)

James I. Cole, Barry H. Rabin, James A. Smith, Clark L. Scott, Bradley C. Benefiel, Eric D. Larsen, Paul R. Lind, David A. Sell

March 2016

# US PROGRESS ON PROPERTY CHARACTERIZATION TO SUPPORT LEU U-10 MO MONOLITHIC FUEL DEVELOPMENT

JAMES I. COLE, BARRY H. RABIN, JAMES A. SMITH, CLARK L. SCOTT, BRADLEY C. BENEFIEL, ERIC D. LARSEN, PAUL R. LIND, DAVID A. SELL
*Nuclear Fuels and Materials Division, Idaho National Laboratory*
*PO box 1625 MS 3818, Idaho Falls, ID 83415-3818, USA*

ABSTRACT

The US High Performance Research Reactor program is pursuing development and qualification of a new high-density monolithic LEU fuel to facilitate conversion of five higher power research reactors (ATR, HFIR, NBSR, MIT and MURR) and one critical facility (ATR-C) located in the US. In order to support fabrication development and fuel performance evaluations, new testing capabilities are being developed to evaluate the properties of fuel specimens. Residual stress and fuel-cladding bond strength are two characteristics related to fuel performance that are being investigated. In this overview, new measurement capabilities being developed to assess these characteristics in both fresh and irradiated fuel are described. Progress on fresh fuel testing is summarized and on-going hot-cell implementation efforts to support future PIE campaigns are detailed. It is anticipated that benchmarking of as-fabricated fuel characteristics will be critical to establishing technical bases for specifications that optimize fuel fabrication and ensure acceptable in-reactor fuel performance.

## 1. Introduction

To support the U.S Department of Energy (DOE), National Nuclear Security Administration (NNSA) mission to reduce the threat posed by the civilian use of highly enriched uranium (HEU) worldwide, the US High Performance Research Reactor (USHPRR) fuel development (FD) program is pursuing development and qualification of a new high-density monolithic U-Mo fuel to facilitate conversion of five higher power research reactors (ATR, HFIR, NBSR, MIT and MURR) and one critical facility (ATR-C) located in the U.S. from HEU to low enriched uranium (LEU). These reactors require higher uranium fuel density than is achievable with dispersion fuel systems used to convert lower power research reactors. The down selected fuel system consists of U-10Mo alloy fuel foils having a thin Zr diffusion barrier interlayer, clad in 6061 Al alloy by hot isostatic pressing (HIP) [1]. Fabrication process development activities are assessing methods to produce the U-Mo foil, apply the Zr barrier foil and optimize HIP strategies to maximize uranium utilization and minimize overall cost. Baseline knowledge of the stresses developed in the fuel plate constituents as well as the strength of the bond interfaces will be important to understanding how fabrication parameters influence fuel performance.

In order to support fabrication development, fuel performance evaluations, generic fuel qualification through the Nuclear Regulatory Commission (NRC), and eventual reactor licensing, a series of irradiation experiments and testing campaigns are being planned. The tests are designed to provide data needed for fabrication process down selection, to establish acceptable fuel performance under normal operating conditions and anticipated transients, and to demonstrate successful scale-up to prototypic reactor-specific fuel element designs. Materials property characterization data considered in the scope of this fuel development effort include mechanical strength of the individual fuel plate constituents, strength and quality of interfacial bonds between the plate layers and residual stresses developed during fabrication. The same data is needed for the as-irradiated fuel plates to confirm adequate fuel performance and make adjustments to fabrication methods if necessary. This paper describes efforts to develop two specific testing techniques to evaluate the relationship between fabrication variables and fuel performance, and provide needed materials properties data to improve the fidelity of fuel performance models being developed for the US-HPRR FD program. Following a brief overview of the specific techniques for residual stress and bond strength and their implementation in the hot-cell environment, a discussion on the implications the collected data will have on meeting fuel performance requirements is presented.

## 2. Materials Properties Testing Techniques

### 2.1 Residual Stress

Monolithic fuel plates are effectively a layered composite system composed of materials with differing mechanical and thermal properties and constrained interfaces. Residual stresses can form as a result of thermo-mechanical processing due to these differing properties, in particular, as a result of cooling from the HIP processing temperature [2]. Tensile and compressive stresses through the thickness of the part ultimately have to balance, so large compressive stresses in one layer will, by necessity, induce large tensile stresses in another. If these stresses exceed the ultimate tensile strength of the material, component failure can result.

Fuel performance modeling results suggest pre-irradiation residual stresses from fabrication do not significantly influence irradiation performance because these stresses are relaxed very quickly during initial irradiation. However, post-irradiation residual stresses (developed during reactor shutdown) are believed to play an important role in causing fuel failures at high burnup [3]. It is also important to understand if the proposed alternate fabrication processes, i.e. application of Zr by electroplating or plasma spraying, have an effect on the post-irradiation stress state.

A variety of techniques can be employed to measure residual stresses in as-fabricated, unirradiated fuel plates and many of these techniques are non-destructive. For example, monolithic fuel plates have been examined using diffraction techniques [4]. In general, these methods cannot be readily implemented in a hot-cell environment for use on irradiated fuel specimens.

The US-HPRR FD program has explored alternate methods of measuring residual stresses. Several destructive measurement techniques are available and, of these, the incremental slitting or crack compliance technique developed at Los Alamos National Laboratory seemed the most amenable to hot-cell adaptation [5]. This technique uses incremental slitting of the plate and measures the deflection to calculate the residual stress. Typically, in non nuclear applications, the slits are made with electric discharge machining [6] and deflections are measured using strain gauges. However, due to the need to operate the system remotely, the method has been adapted to facilitate hot cell deployment. The EDM slitting has been replaced by a small milling tool, and the strain gauges been replaced by non-contacting displacement transducers. The schematic shown in Figure 1 shows a simplified version of the setup with all of the essential elements.

The measurement is made by clamping the fuel plate on one end vertically, then milling a slit across the width of the plate. As the residual stress is relaxed, the end of the plate opposite the clamp will deflect, and the extent of deflection is measured by the transducer. The direction of the deflection will be dictated by the nature of the residual stress, tensile or compressive. The milling tool depth is incremented (~10 microns for this application) and another slit is made. In this manner a 1-D through plate profile of residual stress can be mapped. Of particular interest is the extent of the stress that develop at the interfaces between the cladding, Zr barrier layer and the U-Mo fuel, and how these stresses relate to bond strength.

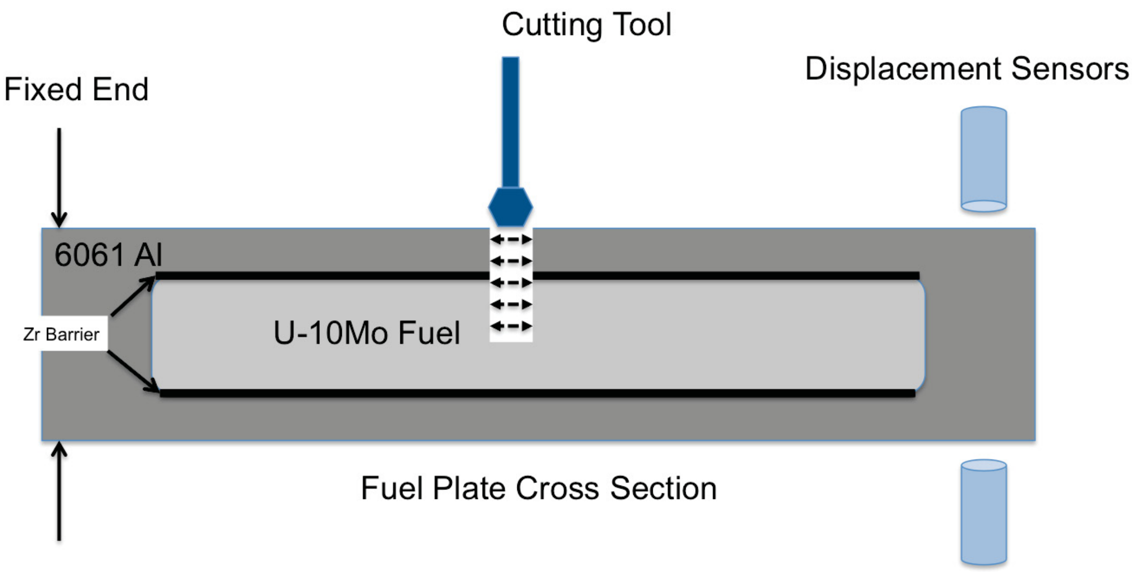


Figure 1  Schematic of slitting residual stress measurement system.

In order to be effectively utilized for testing of highly irradiated fuel plates, the equipment has to be adapted from its conventional configuration, to a configuration that can be readily used inside of a hot-cell with remote manipulators. The customized system being developed incorporates a heavy baseplate to reduce vibration, a clamping jig to hold the sample, displacement sensors and a cutting tool. The sensors and the cutting tool are fully motion controlled to allow precise positioning and calibration. Eddy current sensors were chosen for the displacement measurements, rather than capacitive displacement sensors, as they were deemed to be more robust in a high radiation environment.

Both this system and the bond strength system (discussed below) require installation of new feed-throughs into the Hot Fuels Examination Facility (HFEF) hot cell in order to provide instrument control and minimize the amount of materials that have to be introduced into the hot-cell to conduct the testing. The residual stress-system will have a footprint that can be accommodated within the HFEF containment box, which is an area separated from the main HFEF cell and kept at negative pressure. Operations that generate larger quantities of fines and debris such as cutting, grinding and polishing are frequently conducted in the containment box to minimize contamination levels in the rest of the cell.

## 2.2 Bond Strength

Acceptable interfacial bond strength between fuel and cladding will be critical to maintaining fuel integrity during irradiation. A variety of techniques can be used to measure bond strength (peel test, double cantilever test, bulge test, etc.) that require significant set-up and hands on sample manipulation. However, for the monolithic fuel plates being tested in a hot-cell, a customized laser shock/ultrasonic testing system has been developed by the INL, in partnership with the National Research Council of Canada Corporation (NRC). This system employs a non-contact method using a high power laser pulse to induce an acoustic shockwave in the test piece. The generated wave is a compressive shockwave and when it is reflected off the back surface of the piece it becomes a tensile wave that can de-bond internal interfaces [7-8]. The velocity of the shockwave is detected on the back side of the plate and can be related to the internal stress. By incrementally increasing the laser power and using laser-UT scanning between shots to detect the occurrence of the de-bond, the tensile stresses to induce the de-bond can be qualitatively estimated.  The magnitude of the measured de-bond stress can be compared, for example, for different fuel plate fabrication conditions, however, it's important to note that these stresses are not directly comparable to, for example, the stress that would be measured in a tensile test, because of the very high strain-rates associated with laser-induced shockwaves.

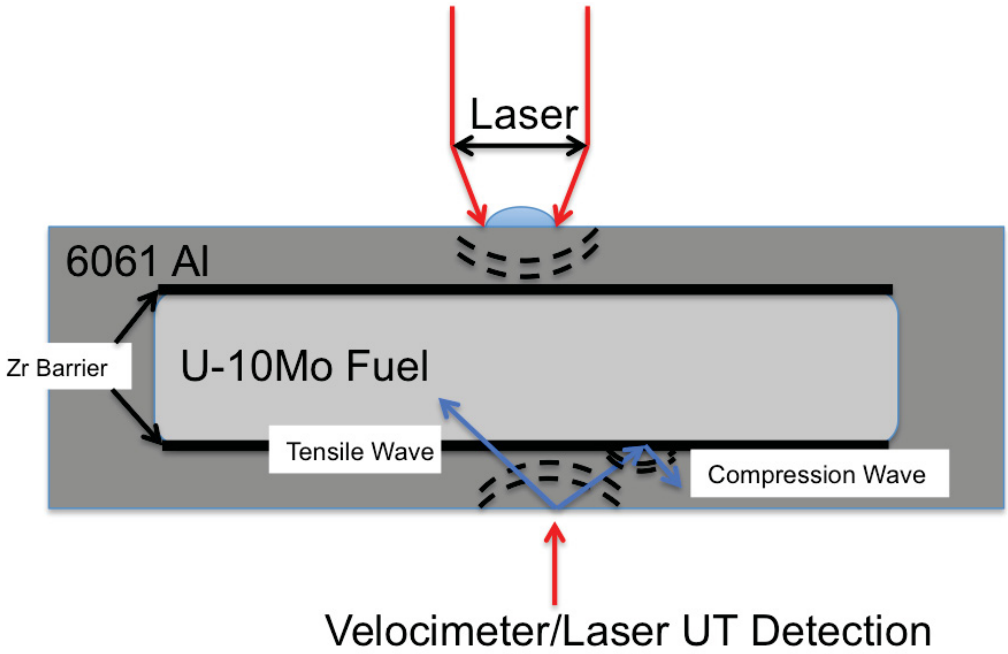


Figure 2 Schematic of laser shock bond strength measurement.

The system is able to determine at which fuel-cladding interface within the fuel plate the debond occurs, however,, the resolution is not adequate to identify whether the failure occurs at the Zr-cladding or fuel-Zr interface. Further evaluations of the debonds created can be made using metallographic examinations. For post-irradiation bond strength testing, localized bond-strength evaluation as a function of fission density within a fuel plate will provide valuable data on the evolution of bond strength during irradiation and the resistance of fuel plates to delamination.

The laser shock system is currently undergoing modification for hot-cell use. The rather complex optical paths of the laser shock and laser-UT scanning system has required fabrication of a large diameter hermetically sealed feed through to accommodate the optical fibers. Qualification of the system for in-cell use has entailed ensuring the laser optical paths can be run through the feed through and maintain signal integrity. Initial testing of this modified system is on-going.

## 3. Initial Testing Results and Fuel Performance Considerations

As mentioned previously, a prototype (proof of principle) residual stress system was developed and tested at LANL. Based on modifications of this design, a new system has been assembled for hot-cell implementation and is currently undergoing qualification testing at INL. Initial results for a surrogate fuel sample that consists of an aluminum clad stainless steel foil bonded using friction stir welding are shown in Figure 3. Deflections as a function of slit depth illustrate a change in the sign of the stresses at the internal interfaces. These initial results show promise in being able to measure residual stresses in-cell. Further analysis is being conducted to calculate the residual stresses in the surrogate plate from the measured deflections. Following further system refinement, tests will be conducted on HIP processed plates to ascertain baseline residual stress states prior to irradiation.

It is important to understand the baseline residual stress state of the as fabricated fuel plates to evaluate whether the fabrication processes have introduced stresses into the fuel plate that might enhance chances for debonding during irradiation. As the plate is composed of three disparate materials, the thermal history will introduce differential thermal expansions and result in stress gradients through the plate thickness. Most of these stresses are predicted to be relaxed during the initial stages of irradiation, but additional stresses will be imposed during irradiation as the fuel swells, cladding deformation occurs, and as the fuel plate cools during reactor shut down. Post-irradiation residual stress examinations will aid in the determination of likely stress concentrators and with additional property measurements (e.g. hardness, bend testing, bond strength) give a good indication if any of the fuel plate constituents are near the failure limits.

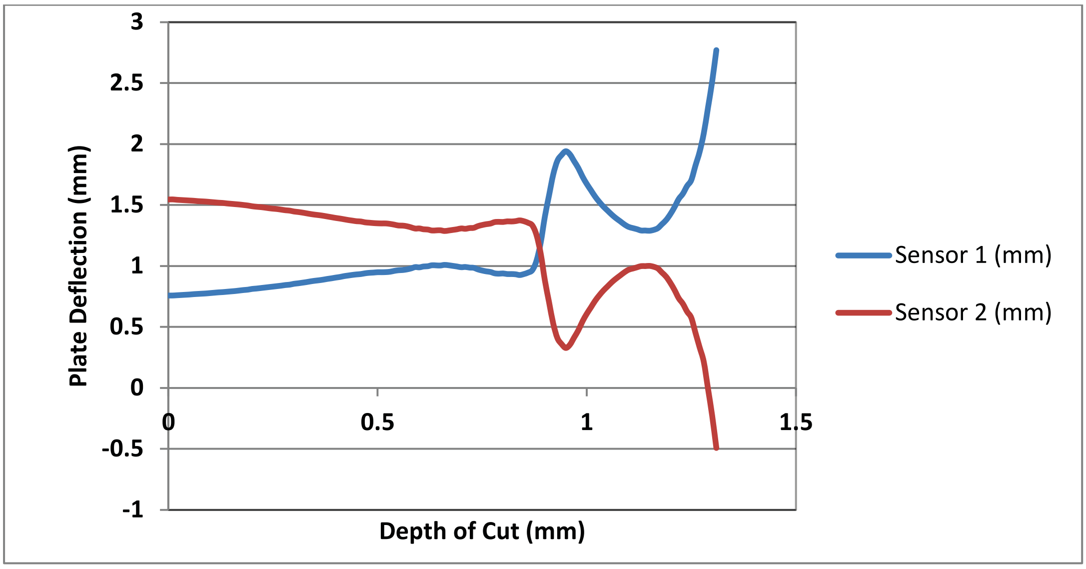


Figure 3 Sample deflection vs slit depth for surrogate fuel plate generated during initial system qualification tests.

Initial testing of the laser shock/UT system was conducted on surrogate fuel plates similar to those used for residual stress (Al cladding with SS foil) to determine if bond quality could be effectively discerned. In this case the fuel plate was HIP bonded and the circular features in the image provided in Figure 4 indicate where the laser shock tests were conducted. Signal analysis indicates that tests conducted in the clad-clad region did not de-bond the interface (locations 1 and 2), while the larger dark circles in the surrogate fuel region (locations 3 and 4) indicate fuel-clad de-bonding. In this particular sample, it is anticipated that bonding should be weaker as the samples were intentionally contaminated with a parting agent.

In a further series of tests actual HIP'ed fuel plates containing U-Mo fuel were examined. Surface velocity at the back side of the plate was measured as a function of shock laser energy to discern the threshold for de-bonding. In some cases, as the graph in figure 5 shows, de-bonding occurred at a lower measured back surface velocity than a previous measurement. Thus, this measurement could not provide an accurate de-bond threshold. Efforts are on-going to refine correlations between measured surface velocity and actual de-bond stress by comparing with results from other tests [10] as well as thermo-mechanical modeling. Overall, the initial testing of samples that had known regions of good bonding and poor bonding could be discerned with the system, and the system is now undergoing qualification for in-cell implementation.

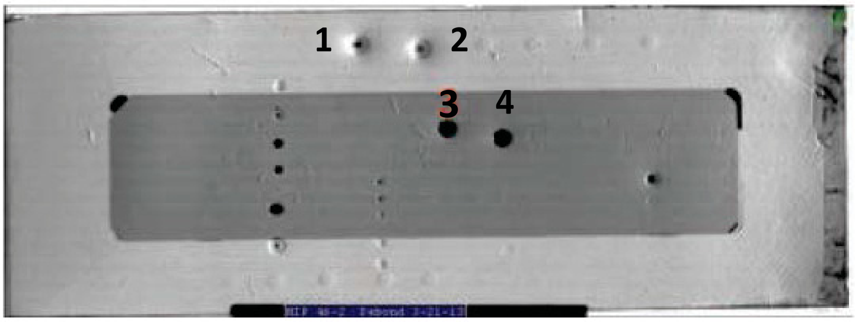


Figure 4  Image of fuel plate following laser shock testing showing regions of good bonding and poor bonding [REF].

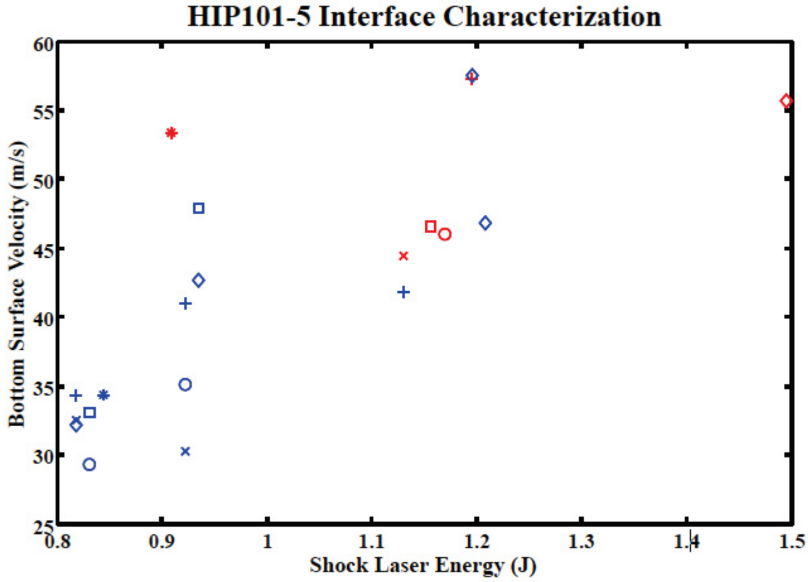


Figure 5 Measured bottom surface velocity as a function of shock laser energy for an RERTR-12 HEU mini-plate.  Each symbol represents a separate test locations with red symbols indicating a de-bond occurred.

The primary functional requirements for monolithic fuel are mechanical integrity, geometric stability, and stable and predictable behavior. Residual stresses both prior to and developed during irradiation are important relative to possible fuel plate delamination failures, in-reactor pillowing, or cladding breach. It is important to understand if the peak stresses developed in the fuel on reactor shutdown can exceed the ability of the fuel to accommodate the stresses. It is also important to understand the relationship between the post-irradiation residual stress state and the fuel-cladding interfacial bond-strength, since local failure at these interfaces could potentially lead to fuel plate delamination.

## 4. Conclusions

The US HPPR program is undertaking a comprehensive effort to develop and qualify a new fuel form.  As part of this effort, new testing techniques to measure fuel-cladding bond strength and residual stresses are being developed and adapted for hot-cell use that will ensure fuel properties are well understood and potential failure precursors are identified to the extent possible.  In combination with fuel performance modeling, behavior of this new fuel type should be known with a high degree of confidence.  A significant outcome of this program will be the qualification and eventual licensing of a new plate-type fuel system for use in research reactors through the NRC for the first time in nearly 3 decades.

## 5. Acknowledgements

The U.S. Department of Energy, Office of Material Management and Minimization, National Nuclear Security Administration, under DOE-NE Idaho Operations Office Contract DE-AC07-05ID14517, supported this work. A contractor for the U.S. Government authored this summary. The publisher, by accepting the article for publication, acknowledges that the U.S. Government retains a nonexclusive, paid-up, irrevocable, worldwide license to publish or reproduce the published form of this manuscript, or allow others to do so, for U.S. Government purposes.

## 6. References

1. A.B. Robinson, et al., Irradiation Performance of U–Mo Alloy Based "Monolithic Plate-Type Design Selection Update", Idaho National Laboratory, INL/EXT-09-16807, Rev.1, July 2013.
2. Hakan Ozaltun, "The Effects of Fabrication Induced Stress Strain-States on the Irradiation Performance of Monolithic Mini-Plates", Proceedings of the ASME 2015 International Mechanical Engineering Congress & Exposition, IMECE2015, November 13-19, 2015, Houston, Texas, paper 53050.
3. H. Ozaltun, P. G. Medvedev, A. B. Robinson, B. H. Rabin, "Shutdown-Induced Tensile Stress in Monolithic Miniplates as a Possible Cause of Plate Pillowing at Very High Burnup" , RRFM2014, Paper A0101.
4. D.W. Brown, D.J. Alexander, K.D. Clarke, B. Clausen, M.A. Okuniewski and T.A. Sisnerosa, "Elastic properties of rolled uranium–10 wt.% molybdenum nuclear fuel foils", Scripta Materialia 69 (2013) 666–669.
5. Prime, Michael B., Lovato, Manuel L., Alexander, David J., Beard, Timothy V., Clarke, Kester D., Folks, Bo S., "Incremental Slitting Residual Stress Measurements for a Hot Cell", Los Alamos National Laboratory, LA-UR-14-23273.
6. Hill, M. R., 2013, "The Slitting Method," Practical Residual Stress Measurement Methods, G. S. Schajer, ed., John Wiley & Sons, Ltd, pp. 89-108.
7. Perton Mathieu, Lord Martin, Jean-Pierre Monchalin, and Guy Rousseau, Laser Shock/ultrasonic Testing System Instructions Manual and Documentation, National Research Council of Canada.
8. Smith, James A.; Rabin, Barry H., Perton, Mathieu, Lévesque, Daniel, Monchalin, Jean-Pierre, Lord, Martin, "Laser Shockwave Technique For Characterization Of Nuclear Fuel Plate Interfaces," RERTR 2012 — 34th International Meeting On Reduced Enrichment For Research And Test Reactors, Warsaw Marriott Hotel Warsaw, Poland, October 14-17, 2012.
9. D.E. Dombrowski, C. Liu, M.L. Lovato, D.J. Alexander, K.D. Clarke, N.A. Mara, W.M. Mook, M.B. Prime, D.W. Brown, B. Clausen, "Experimental Investigation Of Bonding Strength And Residual Stresses In Hip Clad Fuel Plates", Advances in Powder Metallurgy and Particulate Materials - 2013, Proceedings of the 2013 International Conference on Powder Metallurgy and Particulate Materials, PowderMet 2013, p 1134-1148, 2013.