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A Robust Processing Approach for Producing Highly Loaded Dispersion Fuels

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ABSTRACT

A robust fabrication method, resulting in higher yields, to produce highly loaded U_3Si_2-Al dispersion fuels for converting research reactors from a high enriched to a low enriched uranium fuel needs to be developed. To reliably produce a highly loaded dispersion fuel, process changes need to be implemented where the traditional approach has experienced challenges and poor yields. The present work describes the key changes needed. Parts of the work were done with uranium silicide and parts were completed with a representative silicide surrogate. The major deviations from traditional fabrication methods are associated with a refinement in particle size distribution, method for compacting to achieve complex shapes, and welding of the aluminum picture frame used to encapsulate the fuel compact. Methods for rolling and arc melting are also discussed. By using the methods described within, fabricating a highly loaded dispersion fuel that can meet stringent fuel homogeneity and geometry requirements at higher yields and lower costs may be possible.

1 Introduction

There is a global effort to reduce the enrichment of fuels used in the research and test reactors around the world from highly enriched uranium (HEU) to high assay low enriched uranium (HALEU; <20 wt% U-235). Many medium and low power research reactors have been converted to HALEU fuel and do not require any increased fuel particle loading to achieve an acceptable performance level. However, the high performance research and test reactors (HPRR) require increased particle loading to achieve the same performance with lower U enrichment. Highly loaded dispersion fuels have been developed to achieve the necessary high volume percent U loadings.

Currently, consistently producing highly loaded dispersion fuels that meet the tight specifications required for HPRR is problematic. Issues with geometry defects described as “fish tail” and “dog-bone” are ubiquitous, fuel particles are rarely homogeneously distributed throughout the entirety of the fuel zone, fuel oxidation is a concern, and stray fuel particles are often found during inspection. These issues result in large numbers of nonconformance reports (NCR) for reactor fuel plates and an associated higher cost of quality. The non-conformances result in either additional

labor costs to analyze specific plates to determine if they can still be used, or the plate being deemed unusable meaning all the costs of fabrication are lost. Furthermore, when a plate is unusable, there are additional costs to recover the fuel which involves chemical and labor-intensive processing.
A robust fabrication process that is less prone to the deficiencies seen in traditional manufacturing methods is needed. To this end, Pacific Northwest National Laboratory (PNNL) is engaged in research and development to create a more robust process that resolves many or all the associated issues with fabricating highly loaded dispersion fuels. The processing scheme was developed for a HALEU U_3Si_2 fuel with plans to fabricate complex fuel shapes for use in the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory. The methods developed and detailed within are not just specific to HFIR or U_3Si_2, but rather to all highly loaded dispersion fuels of varying types (UAl_x, UO_x, USi_x, etc).
The general fabrication techniques used to produce the dispersion fuel composites reported within this document have been used for many years in industrial-scale composite and powder metallurgy fabrication. While the generalized fabrication approach is widely used and reported [1], the required tolerances and level of homogeneity can be far more stringent than in other applications. This has required many modifications and specialized approaches for individual parts of the fabrication which are not part of the general fabrication techniques used in traditional composite and powder metallurgy fabrication.

2 Experimental Details:

2.1 Arc-melting and powder production:

The U_3Si_2 was fabricated by alloying elemental U and Si in an Edmund Buehler MAM-1 arc-melter. Depleted uranium plate (99.95% Manufacturing Sciences Corp.) and silicon lump (99.9999% Alfa-Aesar) were used as the starting material. The DU feedstock was cut into an approximately 70g rectangle and etched prior to being alloyed. During the etching process, the DU feedstock was placed in an 8M nitric acid bath for 10 minutes, rinsed with deionized water, rinsed with ethanol, and left to dry. A hyperstoichiometric amount of Si was added (7.4 wt%) in the arc melter to the U. Powder x-ray diffraction indicated stoichiometric purity of the U_3Si_2 sample.
The U_3Si_2 buttons were transferred to an inert glovebox that is maintained below 0.1 ppm O_2 where powder was fabricated by ball milling. The particles were captured on 5 sieves ranging between 90μm and 45μm. Each particle size fraction was stored separately. The U_3Si_2 powder size distribution (PSD) is displayed in Figure 1.
Surrogate silicide powders (WSi_2 and $MoSi_2$) were used to carry out forming and rolling studies in a non-radiological environment and validate finite element method (FEM) models. The surrogate work average PSD was smaller and had a larger distribution. The PSD for surrogate powders is also shown in Figure 1.
The surrogate and U_3Si_2 powders were mixed with pure atomized Al (single digit micron sizes) at loadings between 42 and 48 volume percent, which equates to 4.8 and 5.3 grams of uranium per cubic centimeter of fuel meat (gU/cc). The mixing was done in a v-shell blender for the U_3Si_2-Al mixes, and a low speed rotating ball mill with steel ball media for the surrogate-Al mixes.

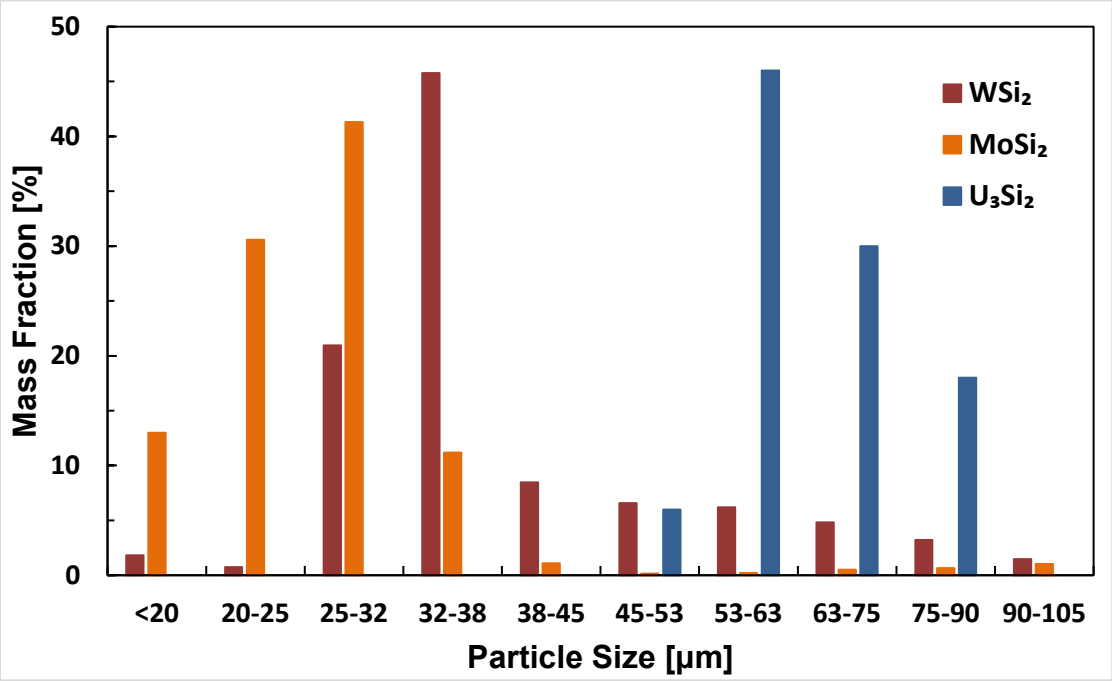

Figure 1: PSD for the surrogate and uranium silicide powders.

2.2 Powder compaction

The U_3Si_2-Al powders were compacted using a 6mm square die set under uniaxial loading with a Carver hydraulic press at approximately 690 MPa. The compact was held at maximum pressure for one minute and then ejected from the die. The surrogate powder mixes were compacted using a cold isostatic press (CIP) from American Isostatic Press Co. and held at 345 MPa. The CIP part was made using a silicone mold cavity with the shape that was expected to yield a final rolled HFIR fuel form found from [2,3]. The molds were filled with powder and then contained in vacuum sealed bags when set into the CIP to prevent the liquid pressurization media from interacting with the powders. The CIP pressurization was stopped immediately upon reaching target pressure and depressurized slowly over approximately 30 minutes.

To accomplish rolling studies in parallel to the CIP development, additional fuel meats were purchased from DWA-USA containing 45 vol% SiC in an Al matrix. These fuel meats were chosen to simulate the behavior of U_3Si_2-Al fuel during rolling. Burnable absorber surrogates (hereafter referred to as filler) with 10-20 vol% SiC in an Al were also purchased from DWA Aluminum Composites.

2.3 Rolling Pack Fabrication:

The rolling pack consists of the SiC surrogate fuel meat, two surrogate fillers encapsulating the fuel meat, and a Al6061 picture frame and cover plates. The fuel meat and filler are inserted into the cutout of the picture frame and then encapsulated on both sides by cover plates. The rolling pack is then sealed using a friction stir weld (FSW) to contain the fuel and filler. The FSW tool has a 12.7mm shoulder diameter and a 9.2mm long pin. It is used to lap weld all three layers in a one-sided pass at a welding speed of 0.3m/min and 500 rpm. The FSW centerline was 12mm from the fuel meat to the picture frame interface on all sides.

2.4 Hot Rolling

Hot rolling was done in a Waterbury Farrel Foundry and Machine Co. rolling mill at 400°C. The plates were heated in a furnace for 1 hour initially and for 15 minutes between each pass and immediately transferred to the rolling mill. The rolling mill has 356 mm diameter rolls and is

operated at 19 meters per minute. The reduction ratio is a constant 20% per pass for the work shown here. The plates are rolled to a nominal thickness of 1.27mm.

3 Discussion:

The basic steps for fabricating highly loaded U_3Si_2 dispersion fuel are shown in Figure 2. The procedure consists of arc melting elemental Si and U at a slightly hyperstoichiometric ratio (~7.4% Si), grinding and sieving the powder, weighing and blending the matrix and fuel powder, forming a compact using CIP, assembling and FSW the picture frame around the fuel and filler, then hot rolling to final fuel size. These steps when done in tandem and as discussed can result in a homogenous fuel with complex geometries.

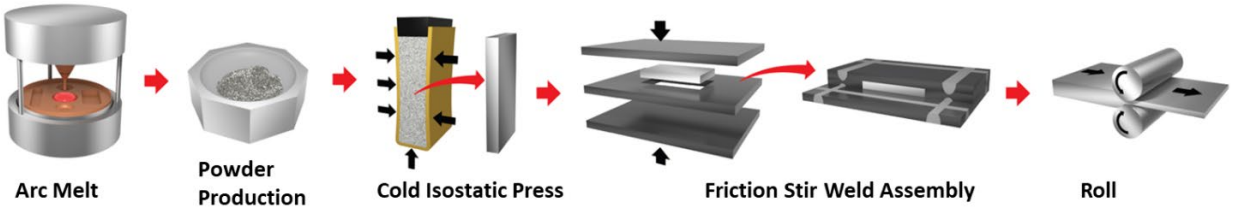

Figure 2: Process steps for fabrication

3.1 Arc Melting:

Many variables can affect the phase purity and minor impurities present in the button during arc melting. The U_3Si_2 phase, while technically a line compound, can be made phase pure at different stoichiometric and hyperstoichiometric quantities ranging from 7.3 wt% to 7.7 wt% [4-8]. In general, important parameters in achieving phase purity are flipping the button, arc current, and melt time. Additionally, the diligent melting of a Zr getter in the chamber and the use of a static ultra-high purity Ar atmosphere can greatly reduce the introduction of minor impurities. Flow through systems and Ti getters, while commonly used, are not as effective at limiting impurity pickup.

3.2 Powder Production:

Homogenous dispersions of U_3Si_2-Al were achieved by controlling PSD via sieving to those seen in Figure 1, and constituent mixing for 4 hours using a V-shell blender with atomized 5 μm Al powder. Tight control of the PSD is essential in forming thin fuel zones and homogeneous dispersion of the fuel is shown in Figure 3. Additionally, the use of a V-shell blender and then diligence in powder handling after it is mixed prevents segregation of the U_3Si_2 and Al due to density differences. Acts like shaking a vial with mixed powder can quickly segregate the powders.

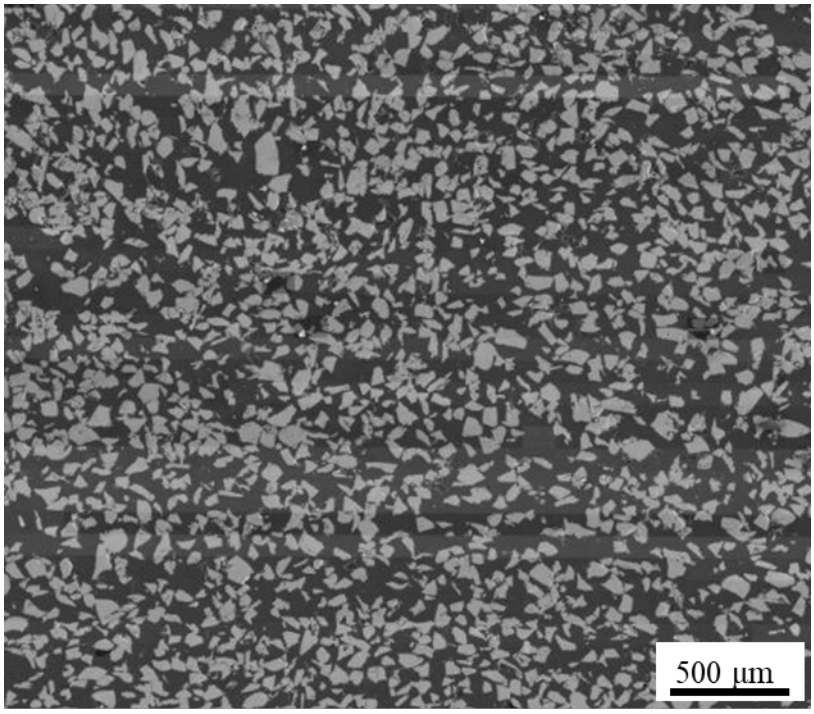

Figure 3: Homogeneous distribution of U3Si2 in an Al matrix.

3.3 Powder Compaction:

Methodologies of loading multiple powder charges into the compacting die then sweeping the powder layers to the desired contour prove challenging and result in unevenness in the top fuel layer due to the powders being dragged during sweeping. Thin individual powder layers contribute significantly to particle streaking and inhomogeneity. Moreover, the particle size and morphology of U_3Si_2 and mismatch of mechanical properties between the filler and fuel can greatly affect homogeneity in thin sections.

For very thin parts, where traditional methods such as sweeping proved difficult even with the correct particle choice, specific dies have been designed for use with CIP. The dies utilize a core and shell configuration with engineered reliefs to allow for uniform compaction and a the formation of complex shapes with contoured edges as thin as 0.6mm. CIP molds and resulting parts are shown in Figure 4. Common die set issues such as die wall friction and non-uniform density are alleviated by hydrostatic pressing with malleable molds in the CIP process. Uniform densities and very complex geometries are achievable with correct die designs.

Moreover, the geometric features employed in the powder compaction step drive the final fuel shape after rolling. Through complex finite element modeling and experimental CIP work done with $MoSi_2$ and WSi_2 surrogates, initial compaction geometries were mapped to desired features such as an axial toe on the fuel and complex contours across transverse sections while minimizing undesirable features like defects called dogbone. A dogbone occurs when the fuel core thickens at the ends during roll bonding. This results in higher fuel concentration that could lead to excessive temperatures during irradiation and thinner cladding that might not meet the minimum cladding thickness required for safe operation. Previously utilized highly enriched UAl_x or UO_x dispersion plate type fuels were not affected by dogbone because of lower total uranium loadings. Modeling informed two very important parameters affecting the formation of dogbone: 1. Mismatch of filler and fuel flow stress, 2. Compact taper length. The effects of each of these is shown in Figure 5 where the dogbone factor is defined as the fuel thickness in the dogbone region divided by the target fuel thickness. When the filler and fuel flow stresses are more closely matched, the dogbone is virtually eliminated. Additionally, increased length of the compact taper along the rolling direction reduces dogbone. Taper length can only be increased to a certain extent without substantially increasing the axial toe length and therefore is dependent upon the HFIR fuel

geometries defined axial length characteristic. Therefore, to control dogbone, a filler with SiC particles as second phase particle strengtheners is used. Careful control of the SiC volume fraction in the Al1100 filler matrix is vital to increase the filler strength to a value not exceeding the Al6061 clad strength so as not to move the dogbone from the fuel-filler interface to the filler-clad interface. Experimental work with surrogates has shown that a volume fraction of 10-20% SiC particles with a FEPA 220 size distribution is the optimum range to control dogbone. When the flow stress of the U_3Si_2-Al composite is determined, it is anticipated the value will still lie within this range but may not be the exact same as with the $MoSi_2$-Al and WSi_2-Al surrogate composites.

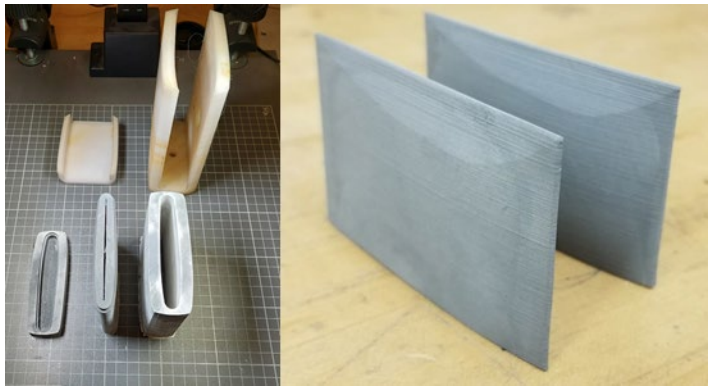

Figure 4: (Left) CIP double shell molds in the front and behind is a brace to keep the mold from deforming during powder loading. (Right) A CIP compact made from $MoSi_2$-Al with tapers in the axial direction and transverse contours.

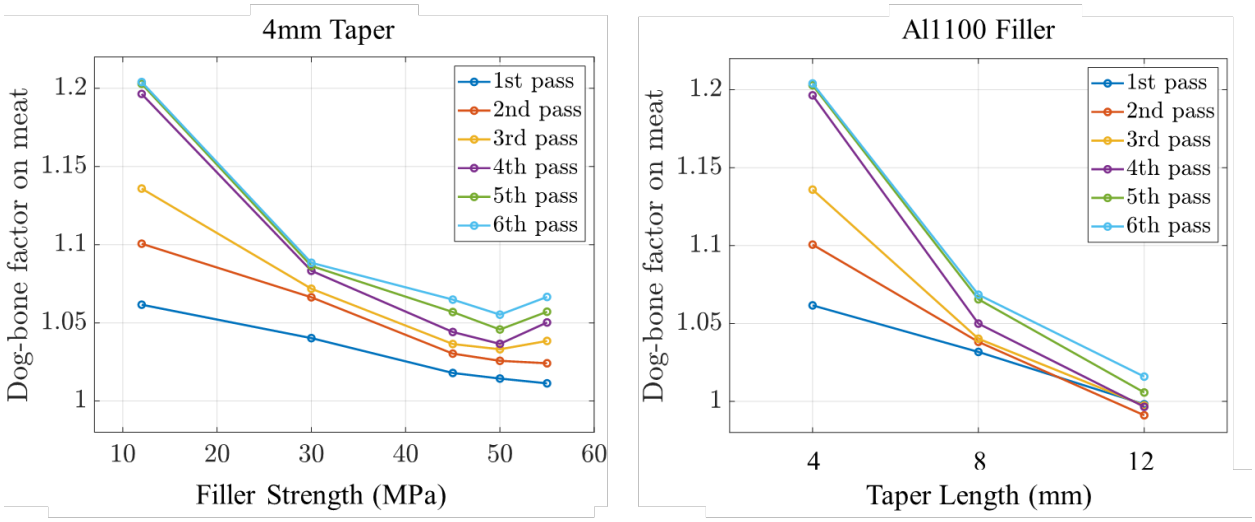

Figure 5: Comparison of dogbone size as a function of (left) filler flow stress and (right) axial taper length. A dogbone factor of one means there is no localized thickening.

3.4 Pack Assembly:

Stray particles outside the fuel zone can occur during rolling operations. This is the result of fuel particles being dragged through the gap between the cover plates and picture frame. Traditionally, the cover plates are welded along the outermost edges, leaving the rolling operation to diffusion bond the cover plates with the picture frame. The diffusion bond takes multiple rolling passes to seal the fuel meat, and fuel particles often escape the fuel meat before bonding. A FSW is utilized to create perimeter welds very close to the fuel meat to seal it with little to no porosity for stray particles to escape before the traditional diffusion bonding would occur. While a traditional fusion weld could potentially do this as well, U_3Si_2 is readily oxidized. To prevent oxidation, it is critical to identify a bonding process that can preserve the oxidation state of the U_3Si_2 in the fuel. FSW is a solid-state, severe plastic deformation process, that has been effectively used to bond materials and requires no external heat source. As FSW is solid-state, the processing temperatures during

FSW are significantly lower than fusion welding processes, which have been demonstrated to oxidize U_3Si_2 even when used at the far ends of the cover plates. The FSW tool can traverse at a high speed (typical range of 0.3 m/min to 2m/min), thus the time at peak temperatures is short, resulting in a smaller heated zone as compared to fusion welding. The temperatures at the fuel zone, about 12mm away from the FSW, are less than 200°C which is well below the ~300°C temperature where U_3Si_2 rapidly oxidizes. A FSW picture frame with fuel and filler encapsulated is shown in Figure 6.

Additionally, FSW has the potential to realize other important benefits during pack fabrication and rolling such as full bonding of the clad-filler-fuel interface prior to rolling, lower distortion and warpage of the fuel plate during and after rolling, and a more efficient, robust automated process. Lastly, if needed, a FSW process can be employed that leaves the fuel region in a vacuum atmosphere to mitigate oxidation during hot rolling. While an electron beam (EB) weld would also produce lower fuel zone temperatures than fusion welding and could be employed under vacuum, EB welds are problematic in 6xxx series Al due to Mg volatilization and brittle Si-Mg precipitate formation [9].

Figure 6: A FSW picture frame assembly with an encapsulated fuel and filler in the center region. The FSW centerline is approximately 12mm away from the edge of the fuel zone.

3.5 Rolling:

Traditionally, during metal fuel rolling, the majority of deformation occurs while the metal is at high temperatures, so it is ductile enough to deform without cracking. Dispersion fuels with Al cover plates are typically rolled at 400-500ºC. U_3Si_2 readily oxidizes in air at temperatures above 300ºC. Significant oxidation may occur during hot rolling if the fuel compact is not evacuated of air prior to hot rolling. Such oxidation is undesirable for multiple reasons: 1. UO_x has a lower density than U_3Si_2. The volume change from oxidation causes localized spalling or cracking; 2. Oxides at the fuel edges cannot be readily differentiated from delamination in the cover plate during ultrasonic testing and, therefore, give defect indications during the non-destructive inspections required of all fuel plates bound for nuclear reactors; 3. UO_x has different swelling and thermal properties under irradiation which can cause localized cracking and high local strains before the fuel end of life; 4. Defects such as fish tails, particle agglomeration, and inconsistent fuel zone thicknesses are problematic in the highly loaded dispersion fuel systems.

To reduce these defects, an optimized hot and cold rolling temperature and reduction per pass (referred to as rolling schedule) is employed to ensure uniform deformation throughout the full thickness of the fuel meat while maintaining the bonding that must occur between the Al cover plates. If a vacuum FSW fuel encapsulation process is not used, it is still possible to minimize oxidation through controlled rolling schedules. The first rolling passes, until bonding occurs, can be done below the oxidation temperature using large reductions with special roll geometries. This allows for negligible oxidation and an evacuated and sealed fuel compact. Then, after the large

deformation, low temperature rolling fully bonds the cladding around the fuel region, rolling temperature is increased to traditional values and the reductions are adjusted to minimize defects such as particle agglomeration and fish tail.
As shown in Figure 7, a tapered compact design, close matching of fuel and filler strengths, and controlled reduction ratios result in axially tapered fuel meats without dogbone in the axial direction and complex contours in the transverse direction. The shark mouth feature exists in the filler region, but not in the fuel region where it is more relevant. Different optical imaging microscopes were used to capture the transverse and axial direction; therefore, the colors are different. The material is the same in both sets of images. The filler design used in this work has a thin lip encapsulating the fuel on all edges. The lip helps to center the fuel meat and hold it in position while the picture frame is assembled. Due to the lip, the filler extrudes past the fuel meat in both directions. If this feature of the filler proves undesirable, the lip can be removed.

Figure 7: Optical imaging of rolled surrogate fuel plates after sectioning. Different optical imaging equipment were used; therefore, the colors appear different. (A) The contoured transverse shape similar to HFIR designs in [2,3]. (B) The axial leading edge of the rolled plate. There is no obvious dogbone and an axially tapered toe feature is seen.

4 Conclusion

A more robust fabrication method is needed to consistently fabricate highly loaded dispersion fuels for HPRR that meet tight nuclear fuel tolerancing requirements and result in higher yields and lower cost of quality. PNNL has developed fabrication methodologies that result in highly loaded homogeneous fuels designed with complex fuel shapes (i.e., transverse and axial contouring) and thin layers containing burnable absorbers. The present work specifically investigated the U_3Si_2-Al dispersion fuels proposed to support conversion of the HFIR reactor, but these processing approaches can be applied to any highly loaded dispersion fuel.

5 References

[1] Knight et. al.,1968 “Fabrication Procedures for Manufacturing High flux Isotope Reactor Fuel Elements.” No. ORNL-4242. Oak Ridge National Lab., Tenn.
[2] Betzler et. al., 2021. “High Flux Isotope Reactor Low Enriched Uranium Low Density Silicide Fuel Design Parameters.” ORNL/TM-2020/1798.
[3] Bae et. al., 2021. “High Flux Isotope Reactor Low Enriched Uranium High Density Silicide Fuel Design Parameters.” ORNL/TM-2020/1799.
[4] Harp, et.al., 2015. “Uranium silicide pellet fabrication by powder metallurgy for accident tolerant fuel evaluation and irradiation.” Journal of Nuclear Materials, 466, pp.728-738.
[5] Havette, et al., 2020. “From arc-melted ingot to MTR fuel plate: A SEM/EBSD microstructural study of U3Si2.” Journal of Nuclear Materials, 537, pp.152224.

[6] Wagner, et al., 2019. “Fabrication of stoichiometric U3Si2 fuel pellets.” MethodsX, 6, pp.1252-1260.
[7] White, et al., 2015. “Thermophysical properties of U3Si2 to 1773 K.” Journal of Nuclear materials, 464, pp.275-280.
[8] Wiencek, T.C., 1995. “Summary report on fuel development and miniplate fabrication for the RERTR program 1978 to 1990.” No. ANL--RERTR/TM-15. Argonne National Lab.
[9] Zhou, Li, et al. 2017"Study on the burning loss of magnesium in fiber laser welding of an Al-Mg alloy by optical emission spectroscopy." The International Journal of Advanced Manufacturing Technology 88, pp.1373-1381