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Uranium silicide pellet fabrication by powder metallurgy for accident tolerant fuel evaluation and irradiation

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HIGHLIGHTS

- A process to consistently produce high density U_3Si_2 pellets was developed.
- Proper milling of the U_3Si_2 to produce the correct particle sized distribution was essential for final product consistency.
- This process was used to produce pellets for irradiation in the Advanced Test Reactor at Idaho National Laboratory.

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ABSTRACT

In collaboration with industry, Idaho National Laboratory is investigating uranium silicide for use in future light water reactor fuels as a more accident resistant alternative to uranium oxide base fuels. Specifically this project was focused on producing uranium silicide (U_3Si_2) pellets by conventional powder metallurgy with a density greater than 94% of the theoretical density. This work has produced a process to consistently produce pellets with the desired density through careful optimization of the process. Milling of the U_3Si_2 has been optimized and high phase purity U_3Si_2 has been successfully produced. Results are presented from sintering studies and microstructural examinations that illustrate the need for a finely ground reproducible particle size distribution in the source powder. The optimized process was used to produce pellets for the Accident Tolerant Fuel-1 irradiation experiment. The average density of these pellets was 11.54 ± 0.06 g/cm^3. Additional characterization of the pellets by scanning electron microscopy and X-ray diffraction has also been performed. Pellets produced in this work have been encapsulated for irradiation, and irradiation in the Advanced Test Reactor is expected soon.

1. Introduction

Further improvement and development of the safety and performance of fuel for Light Water Reactors (LWR) continues to be an important area of research especially as it relates to creating Accident Tolerant Fuels (ATF). The Fuel Cycle Research & Development (FCRD) program has made investigation of ATF concepts a high priority [1]. Several accident tolerant fuel concepts are currently being evaluated by industry lead teams in collaboration with national laboratories and universities. This work is part of one such collaboration whose partners includes Westinghouse Electric Company and Idaho National Laboratory among others. The primary uranium compound used in nuclear fuel worldwide is uranium dioxide (UO_2). Alternative uranium compounds, such as uranium silicides, exist whose properties make them a potential alternative to UO_2 in nuclear fuel. In this work, samples of high density (>94% theoretical density) uranium silicide (U_3Si_2) have been fabricated by powder metallurgy techniques. The developed fabrication techniques were used to create samples for irradiation testing in the Idaho National Laboratory (INL) Advanced Test Reactor (ATR) as part of the ATF-1 irradiation [2]. Post irradiation examination of these samples will provide important information on the performance of U_3Si_2 under typical LWR conditions.

Uranium and silicon form several different stoichiometric compounds including USi_2, USi (or $U_{34}S_{34.5}$), U_3Si_2, U_3Si [3,4]. The uranium density and thermophysical properties of high uranium content uranium silicides (U_3Si_2 and U_3Si) make them an attractive

Abbreviations: ASTM, American Society for Testing and Materials; ATF, Accident Tolerant Fuel; ATR, Advanced Test Reactor; EDS, electron dispersion spectrometer/spectroscopy; FCRD, Fuel Cycle Research and Development; INL, Idaho National Laboratory; L/D, length to diameter; LWR, Light Water Reactor(s); MC-ICP-MS, Multi Collector Inductively Coupled Plasma Mass Spectrometer; OES, Optical Emission Spectrometer; PEG, polyethylene glycol; SEM, scanning electron microscope/microscopy; XRD, X-ray diffraction.

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Table 1
Key properties of UO_2 and U_3Si_2.

Property	UO_2	U_3Si_2
Theoretical density (g/cm^3)	10.96	12.2
Theoretical uranium number density (atom/cm^3)	2.44×10^{22}	2.86×10^{22}
Thermal conductivity (W/m K 400–1200 °C)	6 to 2.5	13.0 to 22.3 [10] 16.1 to 28.2 [11]
Melting point	2847	1665

material from both an economic and safety perspective as a replacement for UO_2. Experience from research reactor fuel work indicates U_3Si swells too much under irradiation for use as a nuclear fuel [5–7]; additionally it disassociates into U_3Si_2 and solid solution U above 900 °C, which is below some expected temperatures in uranium silicide fueled pins. Fortunately, U_3Si_2 has a very promising record under irradiation in research reactor fuels and maintains several advantageous properties over UO_2 [6–9]. Some of these properties are shown in Table 1. There are approximately 17% more uranium atoms in a set volume of U_3Si_2 than there are in the same volume of UO_2, given a constant percentage of theoretical density for both samples. This superior uranium loading has the potential to enable power uprates, extend cycle length in LWRs, or reduce enrichment, all of which are economically beneficial. The higher uranium loading may also allow for the practical application of advanced cladding materials such as SiC or ATF cladding materials that carry a neutronic penalty compared to zircaloy such as FeCrAl steels. The lower melting temperature of U_3Si_2 compared to UO_2 is off-set by its much higher thermal conductivity that drastically drops the anticipated centerline temperature in a fuel pin compared to UO_2 fueled pins. These properties can have positive impacts on fuel pin performance in a variety of reactor accident conditions. The high thermal conductivity of U_2SI_3 in comparison to UO_2 and thus lower operating temperatures make it attractive as an ATF concept especially when paired with SiC or FeCrAl cladding. The performance of U_3Si_2 clad in SiC and FeCrAl in postulated PWR accident conditions has been shown to be superior to that of the UO_2 Zircaloy system [12,13].

The goal of this work was to develop a process that could be used to produce U_3Si_2 pellets that was industrially scalable after the U_3Si_2 compound was produced. Once this process was demonstrated, it was then used to fabricate pellets for an irradiation experiment. The fuel fabrication process used in this work is schematically illustrated in Fig. 1. The process used to formulate U_3Si_2 is likely restricted to lab scale because it involves the production of powdered U metal which is highly pyrophoric and must be handled in an inert atmosphere glovebox. The lab scale formulation steps are segregated by the green box in Fig. 1.

Uranium silicide was formed from mixing powders of elemental uranium and silicon in near stoichiometric quantities. Uranium powder was created by a hydride/dehydride process, and the necessary silicon was procured as a powder. The mixture was then pressed into a compact. The compacts were agglomerated in a furnace at 1450 °C, and then sent to an arc melter to completely react the uranium and silicon. In the arc melter, a tungsten electrode passed current through the uranium and silicon sample into a water cooled copper or graphite hearth. The arc melting process was repeated two additional times to ensure complete reaction. This process produces uranium silicide that is more U_3Si_2 phase pure (97% U_3Si_2) than what was typically produced by arc melting bulk uranium and silicon pieces which usually had about 10% U_3Si [3,14].

The formulated U_3Si_2 was then sent through a standard powder metallurgical process that could be scaled up to an industrial scale. These steps are depicted inside the (in web version) red box in Fig. 1. The ingots from arc melting are comminuted or turned into a powder by planetary ball milling. The resulting powder is pressed into green (formed but not sintered) pellets and then sintered. Various different sintering parameters were explored to achieve the desired theoretical density. A programmatic goal density of greater than 95.5% theoretical density (11.65 g/cm^3) was established based on the current criteria used for UO_2 in commercial fuel fabrication. The sintered pellets were machined using centerless grinding which had not been previously demonstrated on U_3Si_2.

2. Uranium silicide formulation

As was discussed briefly in the Introduction, U_3Si_2 was produced using a lab scale process. Separate work is underway that seeks to find a pathway to produce industrial quantities of U_3Si_2 from standard nuclear industry uranium sources (i.e., UF_6). U_3Si_2 formulation is carried out by a unique technique for arc melting uranium metal powder with elemental silicon powder. The formulation process took place inside inert atmosphere (Ar) gloveboxes with tightly controlled O_2 partial pressures. Typically, the O_2 content of the glovebox is kept below 2 ppm. The reactive nature

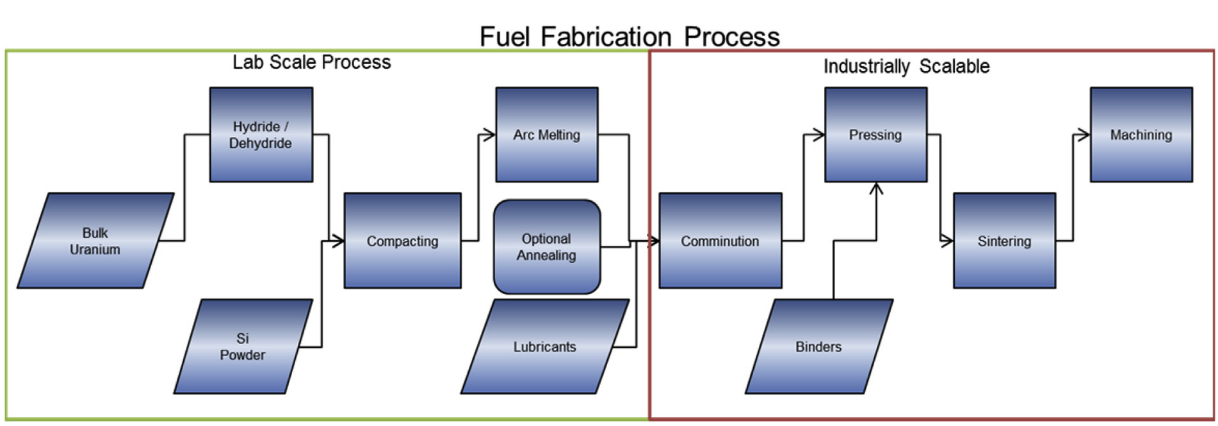

Fig. 1. Fabrication Flow chart for uranium silicide pellet production in this work.