MACHINING INDUCED FISSURES IN RELATION TO MICROSTRUCTURE OF U_3Si_2 FUEL PELLETS

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ABSTRACT

As uranium silicide (U_3Si_2) becomes a more desired fuel replacement for uranium dioxide in light water reactors, it is import to understand the fabrication process. Moreover, it is important to understand how the as-sintered microstructure of the fuel pellet affects the final product; uranium silicide fuel pellets are currently fabricated using a powder metallurgy process. Pellet quality and integrity is a concern as any unwanted defects may cause adverse irradiation behavior. Thus, pellets need to remain relatively free of defects (surface chipping, cracking, undesired microstructure, etc.) in order to be placed into a fuel assembly. Recently, it was observed that uranium silicide fuel pellets developed surface fissures on the machined surface after a centerless grinding process. Initial observations indicate a relation between microstructure and machining induced fissures; in essence, fissures on the machined surface are only observed in pellets containing a heterogeneous microstructure (fine grains surrounding coarse grains).

1. Introduction

Stoichiometric U_3Si_2 (92.7 wt% U, 7.3 wt% Si) fuel pellets were fabricated using a combination of arc melting and powder metallurgy techniques. These pellets were fabricated in a method similar to that presented by Harp *et al* [1] with exception of the composition and sintering environment. In the fabrication effort presented by Harp, Si did not volatize as reported by Wiencek [2], thus the use the stoichiometric composition for this fabrication effort. Due to the laboratory setup, a vacuum atmosphere was chosen to sinter these pellets to decrease the UO_2 content of the pellets. The fuel pellets had an average density of about 11.6g/cc (theoretical density (TD): 12.2g/cc), a phase purity of greater 94% U_3Si_2 [3] and a grain size ranging from about 20-110μm. Part of the fabrication process is to grind sintered pellets to the desired diameter via a centerless grinding process; the diameter of the sintered pellets in this work was decreased by roughly 1mm. During the most recent fabrication campaign at Idaho National Laboratories, surface fissures were observed on the machined surface (Figure 1b); these fissures were not observed in the as-sintered state (Figure 1a) and do not propagate an observable (via light microscopy) distance into the pellet interior. The cause of these cracks will be discussed in the following sections.

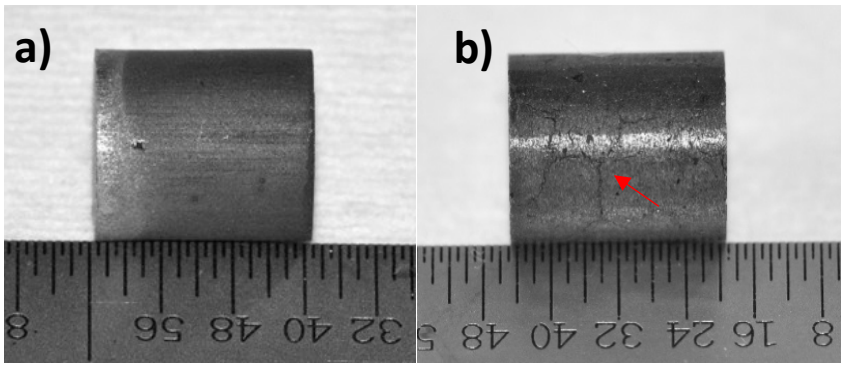

Figure 1: a) sintered pellet prior to centerless grinding, no visible surface cracks; b) pellet after centerless grinding, visible surface cracks. Pellets were sintered in vacuum at 1500°C for 5 hours. Scale bar in inches.

2. Experimental

U_3Si_2 powder was fabricated by the method described by Harp *et al.* [1], in an inert (Ar) atmosphere glovebox with exception of the composition and sintering environment. Pellets (green density: ~58% TD) were sintered in a RD Webb 69 high temperature graphite furnace, capable of vacuum or inert atmosphere cycling. The following thermal cycle was used—20°C/min to 600°C, held for 2 hours to remove binder (PolyOx WSR-301) and vacuum grease (Apiezon L); 20°C/min to 1500°C, held for 5 hours; 100°C/min to room temperature; <1e-5 Torr vacuum.

Due to the reactivity of the fine U_3Si_2 powder (<10μm particle size), and the location of the furnace outside the glovebox, it was necessary to seal green pellets in a NAC 678 graphite sintering crucible (Figure 2), in order to transfer pellets in an inert (Ar) atmosphere. Apiezon L vacuum grease was used to seal the graphite sintering crucible. Apiezon L is used as a sacrificial seal, and is burned off and pumped out of the chamber during the 600°C hold, thus exposing the pellets to the vacuum atmosphere of the furnace. Sealing the pellets into the crucible is necessary to prevent any unwanted thermal hazards, as the pellets undergo an exothermic oxidation reaction when exposed to the laboratory environment.

Pellets were machined via a centerless grinding process utilizing diamond grind wheels. This process removed roughly 1mm of material on the diameter. The initial grinding steps removed 0.25-0.5mm of material on the diameter, while subsequent steps removed .0127-.025mm on the diameter.

Samples were prepared for microscopy and XRD by sectioning with a Struers SiC high speed saw. Once sectioned, samples were mounted a Buehler Epothin 2 epoxy resin and polished to a 1μm surface finish.

A Zeiss Observer.D1m light microscope, utilizing a polarizing filter to highlight the grain structure as the tetragonal crystal structure of U_3Si_2 reflects light such that grains can be resolved under polarized light. Images from this microscope were for grain size analysis. Grain size analysis was conducted in accordance with ASTM E112-13 [4].

Figure 2: Green U_3Si_2 pellets loaded into a NAC 678 graphite sintering crucible, before sealing for transfer from the glovebox to the sintering furnace.

3. Results and Discussion

It was observed that surface fissures formed on the machined surface of the pellets. However this is not the case for every pellet. The primary difference between the two pellet types (pellets with surface fissures or pristine pellets) was the microstructure. Most interestingly, the pellets which developed the surface fissures had an unexpected microstructure—large grains (about 107 μm) surrounded by small grains (about 28 μm) (Figure 3). This observed heterogenous microstructure was not seen in previous fabrication efforts or in literature associated with powder metallurgy. Surface fissures were not observed on pellets having a homogeneous fine grain structure (about 23 μm) (Figure 4).

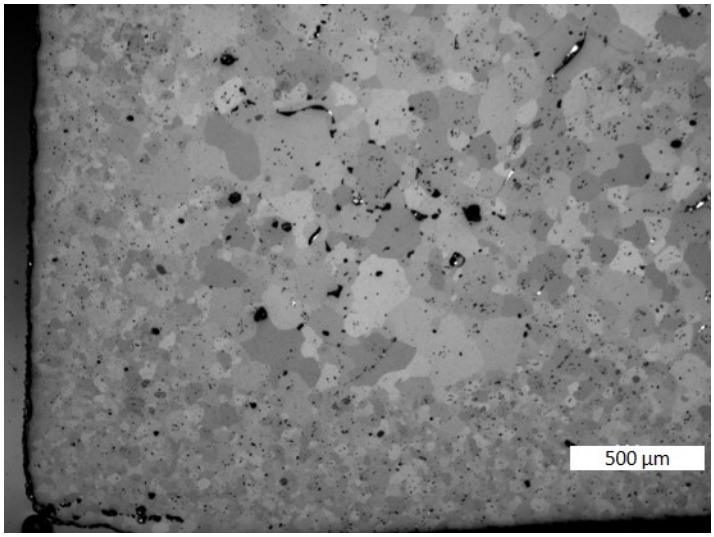

Figure 3: Polarized optical image of a pellet with heterogenous microstructure; fine grain microstructure on edge (grain size about 28 μm), coarse grain structure in center (grain size about 107 μm). Pellet sintered in vacuum at 1500°C for 5 hours, with increased thermal mass.

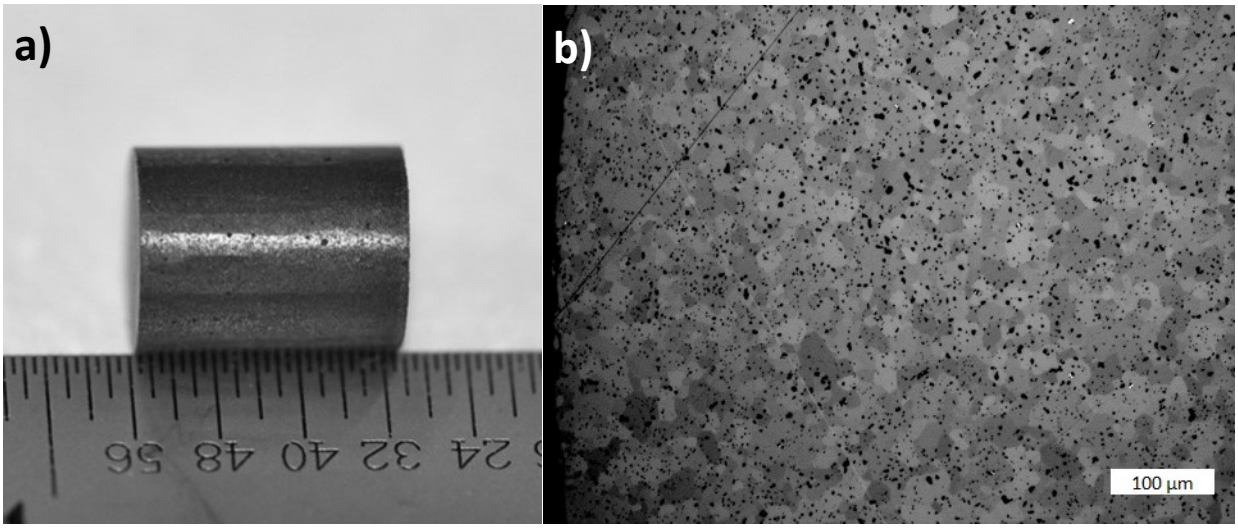

Figure 4: a) image of a centerless ground pellet—without surface fissures prior to sectioning; b) polarized image of radial cross section of pellet in (a), average grain size about 23 µm, pellet density about 95% TD (TD: 12.2g/cc). Pellet sintered in vacuum at 1500°C for 5 hours, with decreased thermal mass.

The heterogenous microstructure (Figure 3) observed in pellets with surface fissures, is due to the rapid cooling on the edge and slower cooling in the center. The observed microstructure is caused by a combination of events—sintering in vacuum (<1e-4 Torr), and placing an increased number of pellets into a graphite sintering crucible. Sintering in vacuum causes pellets to cool via a radiative cooling process, which causes the edge of the pellet to cool rapidly. A separate experiment was conducted to demonstrate the effects of cooling from peak sintering temperature (1500°C) in vacuum and in a static argon atmosphere (Figure 5). To eliminate other potential causes of the heterogenous microstructure pellets seen in figure 5 were fabricated without the use of a milling lubricant or PolyOx binder. These pellets were sintered at 1500°C for 12 hours. These pellets were sintered for 12 hours instead of 5, as mentioned earlier, was to ensure proper densification (>95%) of the pellet as the initial particle size was larger (38µm versus 10µm). An exaggerated heterogeneous microstructure was observed in the pellets sintered in vacuum, while a relatively homogenous microstructure was observed in pellets sintered in Ar. The exaggerated microstructure in figure 5 is a result of sintering/annealing for 12 hours.

The cooling rate of pellets with surface fissures is further retarded by the graphite, exacerbating grain growth in the center of the pellets. The graphite crucible is likely insulating the pellets keeping the material above the recrystallization temperature. Additionally, the observed microstructure worsened in these pellets based on an increased mass of material placed into the graphite sintering crucible, which amplified the effect of the crucible on the cooling rate, ultimately leading to microstructure observed in Figure 5a. This microstructure is not seen in pellets sintered under the same conditions, with exception of less mass in the sintering crucible (Figure 5b); i.e. less energy needing to be removed from the sintering crucible during cooling. Defects seen in figure 5 (cracks, pores, etc.) are a likely the results of the exaggerated heterogeneous microstructure (Figure 5a), and material pullout (Figure 5b) caused by the sectioning and polishing processes. It is difficult to know exactly when the transgranular cracks formed (Figure 5a), as they are indicative of formation after completion of the grain growth process. However, the visible cracks, including those extending to surface of the pellet, likely formed during the sectioning process, as a result of the residual stress present in the pellet (in upcoming discussion).

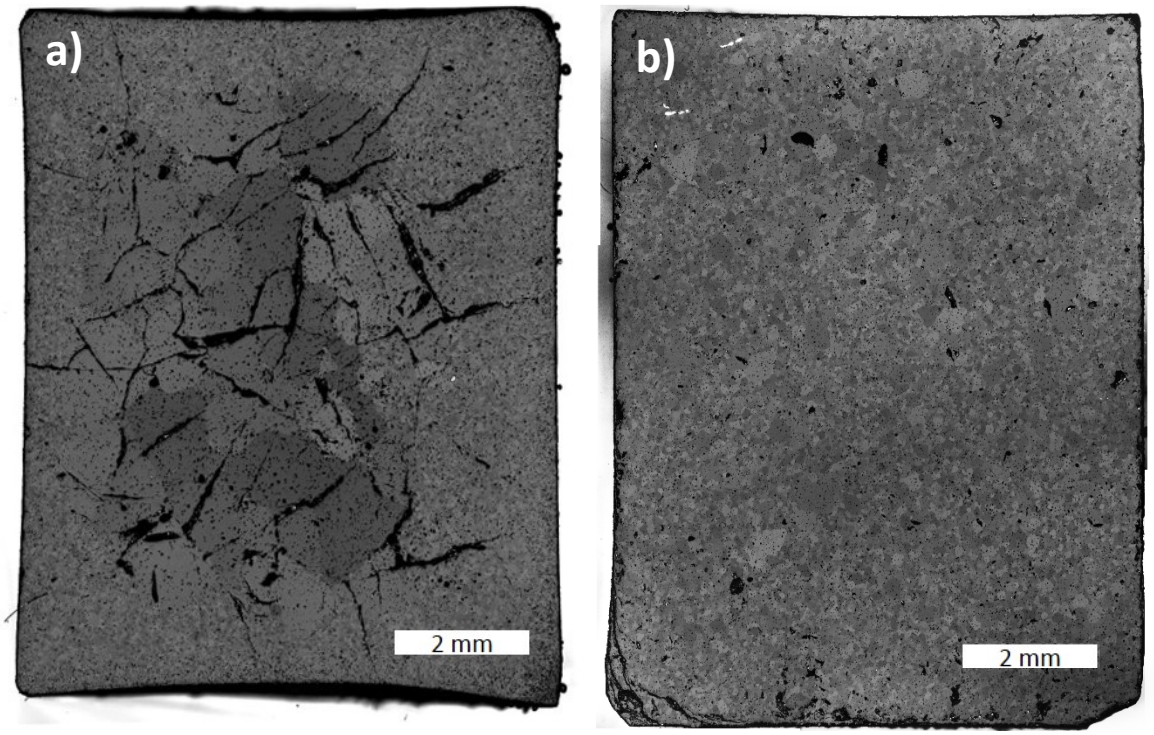

Figure 5: Pellets sintered at 1500°C, for 12 hours. a) Polarized image of longitudinally cross sectioned pellet sintered in vacuum; b) polarized image of longitudinally cross sectioned pellet sintered in static UHP argon

The microstructure of the pellets is crucial in determining whether pellets will develop surface fissures, as the difference in grain size (Figure 3) introduces residual stress in the pellet [5]. If the pellet has a homogenous microstructure any residual stress introduced is minimized. Moreover, if the pellet has a heterogenous microstructure (Figure 3), a strain mismatch is produced between the edge (fine grain) and center (coarse grain) of the pellet, and introduces residual stress. As such, it is not anticipated that phase purity will contribute to the formation of surface fissures. XRD patterns (Figure 6) for the pellet in figure 5a, show a defect structure (PDF# 04-004-1635) in the major U_3Si_2 phase (PDF# 04-007-9355) as a result of lattice strain likely due to the presence of a strain mismatch. This is evident by the observed shift in peaks at—about 27°, 34°, 35°, and 39°. The shift in peaks to the left along with the broadening of peaks at the previous mentioned angles, indicate the presence of residual stress. The major shift to the left indicates much of the lattice is under tension, while the peak broadening indicates the presence of a compressive stress on some of the lattice structure. This is supported by the differences in the observed lattice parameters—major U_3si_2 phase: a=b=7.309Å, c=4.040Å and U_3Si_2defect structure a=b=7.380Å, c=3.939Å (Figure 6). Lattice parameters for an ideal U_3Si_2 lattice (P4/mbm (127)) are—a=b=7.331Å, c=3.900Å [6]. To minimize any effects of interstitial carbon on the lattice structure, not discussed here as it is not anticipated interstitial carbon contributes to the non-uniformity of the grain structure, these pellets were fabricated without the use of organic polymer milling lubricant or binder, both of which are major sources of carbon (PEG 3350 and PolyOx WSR-301). Diffraction peaks observed around 30° are unknown FCC and HCP lattice structures. These peaks were not observed in pellets containing a similar heterogeneous microstructure thus do not contribute to the heterogeneity of the pellet in figure 5a. Further investigation is required to identify the composition of these lattices.

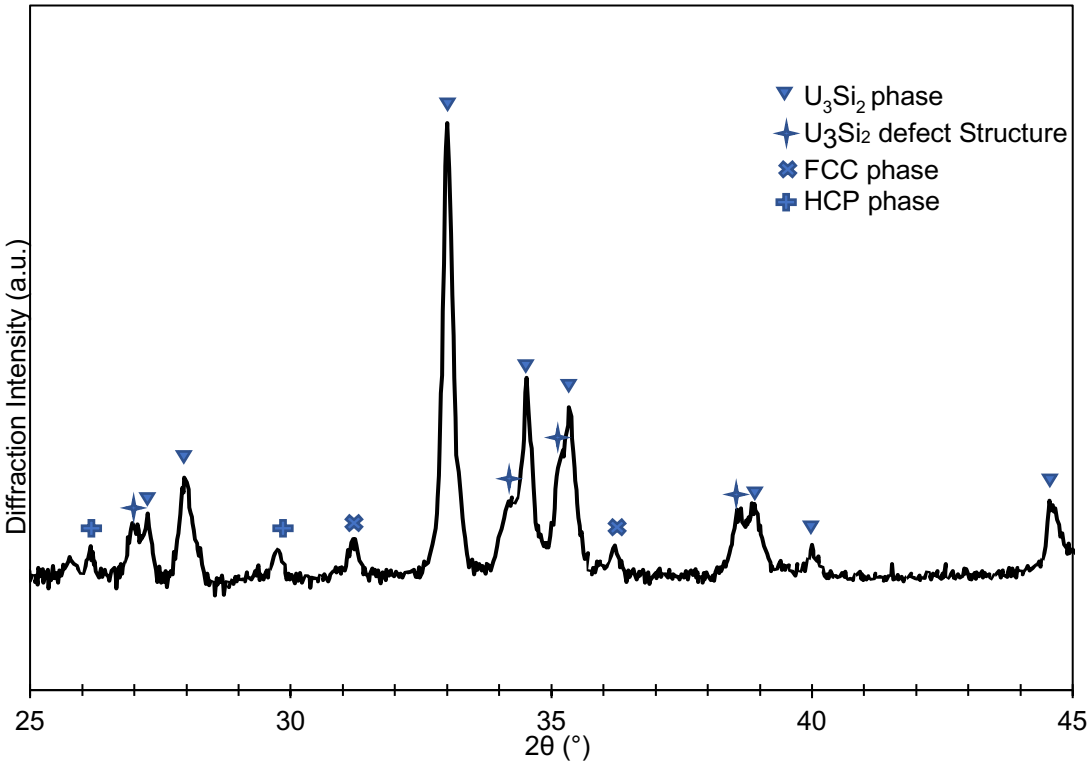

Figure 6: XRD pattern for pellets seen in figure 5a. The major U_3Si_2 phase (PDF# 04-007-9355) had the following lattice constants—a=b=7. 309Å, c=4.040Å, while the U_3Si_2 defect structure (PDF# 04-004-1635) had the following lattice constants— a=b=7.380Å, c=3.939Å.

The presence of a strain mismatch is further supported in the machining of pellets. In the as-sintered state surface fissures are not present, as pellets with a heterogeneous microstructure are in a static equilibrium as the compressive stress in the outer layer counteracts the tensile stress in the center of the pellet [5]. Upon removal of the compressive outer layer, through the centerless grinding process, the pellet returns to an equilibrium state via the formation of surface fissures (Figure 1b), likely caused by a minute expansion of the pellet. Tests were conducted to identify if the amount of material removed in the initial grinding steps influenced the formation of the surface fissures; surface fissures were observed no matter how much or little material was removed during the initial grinding steps. Surface fissures were not observed on pellets with a homogeneous microstructure (Figure 3a), as the homogeneous microstructure in the pellet does not produce a strain mismatch.

4. Conclusions

-Pellets containing a heterogeneous microstructure are observed to be free of surface fissures in the as-sintered state due to the static equilibrium produced through the interaction between the compressive and tensile layers of the pellet.
-Upon removal of the compressive outer layer, surface fissures form on the machined surface.
-The presence of residual stress caused by the grain size mismatch is supported by XRD data.
-The heterogenous microstructure is caused by a combination of a pellets being placed into a graphite sintering crucible and the radiative cooling in a vacuum environment.
-Pellets containing a homogeneous microstructure do not form fissures on the surface, as residual stresses caused by grain size differences are minimized or not present.

5. Reference

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[4] ASTM E112-13, *Standard test Methods for Determining Average Grain Size*.
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