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neutron irradiation induced amorphization of uranium silicides

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Abstract

Time-of-flight neutron powder diffraction studies of incrementally neutron-irradiated U_3Si and U_3Si_2 have revealed details of progressive amorphization of bulk materials, crystalline transformation prior to amorphization, elastic strain and diffuse scattering resulting from scattering interference between crystalline and amorphous fractions. Density differences between amorphous and crystalline fractions give rise, respectively, to tensile and compressive strain in U_3Si and U_3Si_2 . Diffuse scattering associated with each Bragg peak shows crystallographic direction-dependent variation both in magnitude and displacement (relative to the Bragg position). A theoretical model describing this behavior relates the size of the amorphous zones, and the magnitude and displacement of the diffuse scattering contribution. After complete amorphization of U_3Si and U_3Si_2 , anneals to progressively higher temperatures generate gradual evolution of the short- to intermediate-range amorphous structures prior to re-crystallization.

Keywords - neutron irradiation, amorphization, powder diffraction

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1. Introduction

High density intermetallic alloys of uranium and silicon have been considered for use in high power-density nuclear applications and lower power-density, reduced enrichment applications. U_3Si and U_3Si_2 have received the most attention as reactor fuels. A fundamental stumbling block to the use of U_3Si in such applications is catastrophic swelling, occurring at high fuel loadings, due to plastic flow of amorphous U_3Si during irradiation [1,2].

Because of this swelling in U_3Si , emphasis shifted to U_3Si_2 . Although scant data regarding the irradiation behavior of U_3Si_2 exist [see references in ref. 4], it also becomes amorphous under ion and neutron irradiation [3]. Such amorphization raises concern about potential mechanical instability in U_3Si_2 and motivates this work. This study of the irradiation behavior of U_3Si and U_3Si_2 , synopsised here and reported in detail elsewhere [4,5], was carried out as part of the reactor fuel development program for the Advanced Neutron Source (ANS).

2. Discussion

This work utilized high resolution neutron powder diffraction (NPD), on the General Purpose Powder Diffractometer (GPPD) [6] at IPNS, following neutron irradiation [7] and *in situ* transmission electron microscopy with electron diffraction during Kr ion irradiation, using Argonne's HVEM facility [8]. Ion irradiation provides rapid damage accumulation, in thin specimens, where atomic scale structures of specimens can readily be determined by electron diffraction. Quantitative details of the amorphization, though, are not easily obtained by electron diffraction. High resolution NPD, on the other hand, yields precise crystallographic and phase information. NPD experiments are especially valuable because they measure bulk behavior. Precisely controlled neutron irradiation doses (at IPNS), coupled with quantitative diffraction measurements allowed, accurate recording of the amorphization process, via Rietveld profile refinement [9] with Fourier filtering analysis [10] (for modeling of amorphous fraction).

Neutron irradiation results in uranium fission into energetic fragments producing amorphous zones. The volume fraction of amorphous material increases exponentially with dose, and amor-

phization is complete by a dose of 0.38 displacements per U atom (dpa) for both alloys (shown for U_3Si in Figure 1). In each case, a density change from undamaged to amorphous material generates lattice strain in the undamaged fraction. Initially, the lattice strain (tensile for U_3Si and compressive for U_3Si_2) increases linearly with amorphous volume fraction (see Figure 2). At higher irradiation levels, however, plastic flow in the amorphous volume fraction of U_3Si relieves strain in the remaining crystalline volume fraction, following an appropriate rate law (see legend in Fig. 2).

NPD patterns of partially amorphized U_3Si and U_3Si_2 are quite complex. In addition to Bragg scattering from remaining crystalline portion, and oscillatory scattering characteristic of amorphous material, there is significant diffuse scattering associated with each Bragg peak, due to scattering interference between the crystalline and amorphous materials. For U_3Si this contribution appears to be symmetric about the Bragg position, while in U_3Si_2 it is distinctly asymmetric.

Amorphous zones (roughly spherical regions of amorphized material) produced by neutron irradiation generate displacement fields which shift atoms from their ideal positions. These displacements need not have a direct correlation with the crystalline lattice. This phenomenon results in an additional scattering profile whose maxima are closely, but not directly, correlated with Bragg positions. The positions of Bragg peaks at intermediate doses are controlled by the elastic constants for crystalline U_3Si and U_3Si_2 , while positions for diffuse scattering maxima are more subtly controlled. Through the application of standard theories for diffuse scattering [11], we have developed a means for quantifying such factors as the size of the amorphous zones, the magnitude of the diffuse scattering contribution, and crystallographic direction-dependent variations in the displacement of the diffuse scattering component from associated Bragg peaks (Figure 3).

Currently underway is a study of the re-crystallization of U_3Si and U_3Si_2 by vacuum anneals at progressively higher temperatures. These data illustrate gradual evolution of the short- to intermediate-range amorphous structure prior to re-crystallization [12].

Acknowledgements

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Figure Captions

Figure 1. Small portion of the neutron diffraction pattern for U_3Si at different levels of irradiation. This illustrates the anisotropic strain induced by amorphization, and the fully amorphous silicide produced after 0.38 dpa damage.

Figure 2. Lattice strain in U_3Si resulting from neutron irradiation. Strain follows new rate law as plastic flow of amorphous fraction reduces the impact of the density change on amorphization.

Figure 3. Illustration of the quality of fit achieved with a defect model accounting for diffuse scattering associated with each of the crystalline Bragg peaks in U_3Si_2 . This fit is for the (311) reflection.

Estimation of the length of this paper

a) Title, authors, affiliations and 157-word abstract	177 words
b) Three figures, fitting a single column	450 words
c) Manuscript text	655 words
d) References (12)	201 words

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Fig. 1