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Short Communication on "*In-situ* TEM ion irradiation investigations on U₃Si₂ at LWR temperatures"



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ABSTRACT

The radiation-induced amorphization of U_3Si_2 was investigated by *in-situ* transmission electron microscopy using 1 MeV Kr ion irradiation. Both arc-melted and sintered U_3Si_2 specimens were irradiated at room temperature to confirm the similarity in their responses to radiation. The sintered specimens were then irradiated at 350 °C and 550 °C up to 7.2×10^{15} ions/cm² to examine their amorphization behavior under light water reactor (LWR) conditions. U_3Si_2 remains crystalline under irradiation at LWR temperatures. Oxidation of the material was observed at high irradiation doses.

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1. Introduction

The tsunami and subsequent nuclear accident in Fukushima stimulated global efforts to search for nuclear fuels with enhanced accident tolerance. The accident tolerant fuel (ATF) program aims to replace the conventional UO₂-zirconium alloy solution with a novel fuel design that can withstand a loss of coolant accident (LOCA) for a significantly longer period of time [1]. Having superior thermal conductivity and uranium density compared to UO₂ [2], U₃Si₂ has been widely regarded as a promising ATF candidate [3,4]. To evaluate the qualification of U₃Si₂ as a LWR fuel, its fuel performance, especially fission gas behavior, must be experimentally examined. In addition, development of advanced fuel performance codes that can precisely predict the fuel behavior of U₃Si₂ in LWRs calls for experimental microstructural references and validations of fission gas behavior. Previous studies on arc-melted U₃Si₂ at research reactor temperatures (<250 °C) show that ion irradiation completely amorphizes U₃Si₂ at a very low dose level (approximately 0.3 dpa) [5]. As amorphization significantly alters the microstructure of the material, and therefore interferes with the evolution of fission gas bubbles [6,7], the radiation-induced amorphization behavior of U₃Si₂ at LWR temperatures (from approximately 350 °C to 800 °C) must be well characterized. In addition, when cold pressing and sintering technique is used in fabrication, the microstructure of produced U₃Si₂ fuel pellets features reduced grain size, and precipitation of secondary phases such as USi and UO₂ [8]. This difference in microstructure may also lead to amorphization behaviors different from those in the arcmelted fuel material. Therefore, it is important to understand and characterize any difference in irradiation behavior of the U₃Si₂ samples fabricated by the aforementioned methods. Although inpile irradiation tests are essential to provide reliable experimental data describing material behavior in nuclear reactors, ion irradiation provides an inexpensive and time-saving option to obtain valuable qualitative information about irradiation effects on nuclear materials. In this regard, the combination of transmission electron microscopy (TEM) and in-situ ion irradiation makes it possible to capture the kinetics of radiation-induced microstructural modifications [9], including amorphization [5,10-12], in materials. Therefore, an in-situ TEM ion irradiation technique was utilized in this study to characterize the amorphization behavior of U₃Si₂ at LWR conditions.

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2. Experiments

Fine uranium powder (92.5 wt%) and silicon powder (7.5 wt%) were mixed and pressed at 225 MPa. The powder mixture was then arc melted to produce U_3Si_2 ingots. To fabricate U_3Si_2 pellets, the arc-melted ingots were further comminuted into fine powder. The

U₃Si₂ powder was then cold pressed and sintered in an Ar atmosphere to form fuel pellets using the same technique as for samples irradiated in-pile for the ATF-1 campaign. The details of this fabrication can be found in Ref. [8]. The arc-melted ingots are relatively pure, containing only 1.5 vol% U₃Si precipitates, whereas the sintering technique introduced USi and UO₂ secondary phases

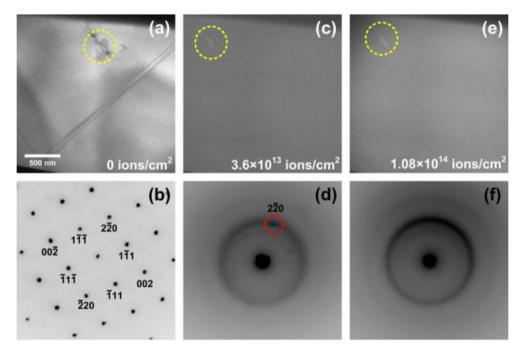


Fig. 1. Amorphization of arc-melted U_3Si_2 irradiated by 1 MeV Kr at room temperature: (a)/(b) Prior to ion irradiation, the arc-melted specimen contains stacking faults and shows clear U_3Si_2 diffraction pattern; (c)/(d) the majority of the U_3Si_2 phase becomes amorphous at 3.6×10^{13} ions/cm² (0.1 SRIM dpa), eliminating the preexistent stacking faults; (e)/(f) full amorphization occurs at 1.08×10^{14} ions/cm² (0.3 SRIM dpa).

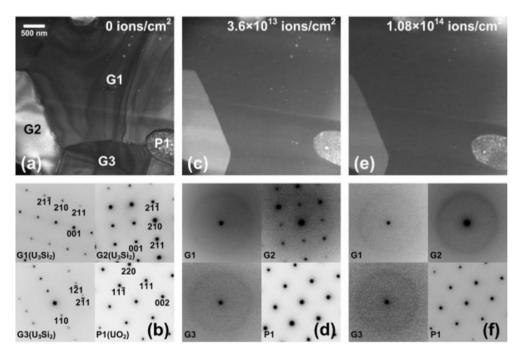


Fig. 2. Amorphization of cold pressed and sintered U_3Si_2 irradiated by 1 MeV Kr at room temperature: (a)/(b) prior to ion irradiation, three crystalline U_3Si_2 grains (G1, G2, and G3) and one UO_2 precipitate (P1) were identified in the specimen; (c)/(d) at 3.6×10^{13} ions/cm² (0.1 SRIM dpa), G1 and G3 become completely amorphous, whereas G2 has distinguishable diffraction pattern, showing various amorphization rates depending on crystallographic orientation; (e)/(f) At 1.08×10^{14} ions/cm² (0.3 SRIM dpa), all three grains become completely amorphous, while the UO_2 precipitate remains stable and crystalline during the ion irradiation at room temperature.

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