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Bubble morphology in U_3Si_2 implanted by high-energy Xe ions at 300 °C

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ABSTRACT

The microstructure modifications of a high-energy Xe implanted U_3Si_2, a promising accident tolerant fuel candidate, were characterized and are reported upon. The U_3Si_2 pellet was irradiated at Argonne Tandem Linac Accelerator System (ATLAS) by an 84 MeV Xe ion beam at 300 °C. The irradiated specimen was then investigated using a series of transmission electron microscopy (TEM) techniques. A dense distribution of bubbles were observed near the range of the 84 MeV Xe ions. Xe gas was also found to accumulate at multiple types of sinks, such as dislocations and grain boundaries. Bubbles aggregated at those sinks are slightly larger than intragranular bubbles in lattice. At 300 °C, the gaseous swelling strain is limited as all the bubbles are below 10 nm, implying the promising fission gas behavior of U_3Si_2 under normal operating conditions in light water reactors (LWRs).

1. Introduction

Development of new fuel-cladding solutions with enhanced accident tolerance so as to replace currently dominant UO_2-zirconium solution in light water reactors (LWRs) has been a focus of nuclear materials community since the tragic nuclear accident in Fukushima Daiichi Nuclear Power Plant [1,2]. The fuel materials satisfying the qualifications of accident tolerant fuels (ATFs) are expected to be compounds or alloys of uranium that have two advantages over UO_2: higher thermal conductivity and higher heavy metal density. The former advantage ensures a lower stored energy during normal operation as well as fast removal of decay heat under accident conditions, whereas the latter one leads to extra neutronic benefits that may either provide more flexibility in the selection of ATF cladding materials or increase economic profits for utilities. Therefore, with both advantageous features discussed above [3], U_3Si_2 has been regarded as a promising ATF candidate and thus attracting intense attention from the nuclear materials community [4–8].

In order to validate U_3Si_2 as a qualified LWR fuel material, its fuel performance must be comprehensively understood through systematic investigations. However, as a fuel material that has been successfully applied in research reactors to reduce uranium enrichment, previous experimental and modeling efforts on U_3Si_2 have been concentrated on low-temperature research reactor conditions [9–13]. Namely, there only exist a limited number of references that involves fuel performance of U_3Si_2 under LWR conditions [4,7]. At typical research reactor temperatures ($<$ 250 °C), U_3Si_2 loses its crystalline structure at merely 0.3 dpa and keeps amorphous throughout the remainders of fuel life [14]. On the other hand, at LWR temperatures ($\geq$ 300 °C), U_3Si_2 tends to maintain its tetragonal lattice structure under irradiation [7]. The first results from the U_3Si_2 irradiation test discussed in Ref. [5] are currently being generated. Initial neutron radiography indicates that the pellets are largely intact and no run-away swelling was observed at a fission density of approximately 6×10^{20} fissions/cm^3 [15]. Therefore, the U_3Si_2 materials in research reactors and in LWRs are literally in two different phases (amorphous phase versus tetragonal crystalline phase), and hence subject to have significant dissimilarity in fuel behavior. In this regard, it is insufficient to solely rely on research reactor data of U_3Si_2 to evaluate its qualifications as a LWR fuel material.

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Gaseous fission products are generated during fission reactions, and accumulate to form intragranular and intergranular bubbles in nuclear fuels. By causing gaseous swelling and originating fission gas release, those fission gas bubbles significantly contribute to the degradation of fuel performance. Thus, establishing comprehensive understanding of fission gas behavior in U_3Si_2 under LWR conditions is crucial to the determination of this material as an ATF. Unfortunately, this effort is obstructed by the absence of experimental investigations of bubble evolution in U_3Si_2 at LWR temperatures. Although the in-pile irradiation experiments of U_3Si_2 have been in progress as a part of the ATF-1 irradiation campaign in Advanced Test Reactor (ATR) [5,16], the detailed post-irradiation experiment (PIE) data will not be available to the ATF community for a while considering the time-consuming and costly nature of PIE on in-pile irradiated fuel materials. Hence, it is of great value to utilize ion irradiation as an inexpensive and timely alternative to study fission gas behavior in U_3Si_2. Being capable of creating various neutron-induced microstructure modifications, ion irradiation has been extensively used to study radiation effects in materials [17–19]. More importantly, the ~100 MeV fission fragments, which cause the majority of microstructure modifications in nuclear fuels, can be replicated by the high-energy ion irradiation technique [20]. In this study, the high-energy ion acceleration capability of Argonne Tandem Linac Accelerator System (ATLAS) [21] was utilized to implant Xe, the most important and representative gaseous fission product, into U_3Si_2 material at a LWR temperature so that the fission gas behavior in U_3Si_2 under normal LWR operating conditions can be unveiled prior to the availability of in-pile irradiation PIE data.

2. Experiments

2.1. Sample preparation

The U_3Si_2 pellet used in this study was manufactured at Idaho National Laboratory [5]. The mixture of 92.5 wt% fine uranium powder and 7.5 wt% fine silicon powder was pressed at 225 MPa before being melted to produce ingots of U_3Si_2 compound. Those arc-melted ingots were then comminuted into fine powder. The U_3Si_2 fine powder was cold pressed and sintered into fuel pellets in an Ar protection atmosphere. The U_3Si_2 pellet used in this study was fabricated using the same procedures as for the pellets irradiated in ATR for the ATF-1 campaign [16]. More details about the pellet fabrication are described in Ref. [5]. Previous investigations of as-fabricated U_3Si_2 pellets indicate that the cold pressing and sintering technique introduces USi and UO_2 precipitates that comprise approximately 14% volume fraction. The pellet was cut into 3 mm thick discs with an 8.3 mm diameter for the ion irradiation experiment. The surface exposed to ion irradiation was first mechanically polished to 0.05 micron surface roughness and then vibratory polished to reach its final surface finishing.

2.2. Ion implantation

The high-energy ion irradiation experiment was performed at ATLAS facility, Argonne National Laboratory. An irradiation chamber was designed and established between the PII Linac and the Booster Linac of ATLAS. At this position, ATLAS is capable of accelerating heavy ions up to approximately 1 MeV per nucleon [21]. The disc specimen was adhered to a copper sample holder using PELCO high performance silver paste. A HeatWave Labs TB-175 cartridge heater that can heat the sample up to 1200 °C was mounted on the back of the copper sample holder. The heater was powered by a DC power supply controlled by a proportional-integral-derivative (PID) controller. The temperature of the specimen was increased to 300 °C and irradiated by 84 MeV Xe ions. This irradiation temperature is within LWR fuel temperature range, which is similar to the fuel surface temperature in boiling water reactors (BWRs). According to the two K-type thermocouples located approximately 5 mm away from the specimen on the sample stage made of oxygen-free high conductivity (OFHC) copper, the temperature of the specimen slightly fluctuated several times when the ion beam was interrupted, whereas the PID controller managed to limit the temperature fluctuation, and maintain the specimen temperature within a range between approximately 290 °C and 330 °C. The energy of Xe ions is close to that of fission products and is therefore expected to produce similar microstructure modifications. The Xe beam profile was measured and centered by a Faraday cup. The beam current was maintained at approximately 100 particle nA for 20 h. Assuming a 2D Gaussian beam shape, the peak ion fluence is approximately 1.38×10^{17} ions/cm^2. According to the quick damage mode SRIM simulation [22] following Stoller et al.'s method [23] ($E_d^U = 61$ eV [24,25]; $E_d^{Si} = 15$ eV), near the center of the specimen, the peak irradiation dose is 499 dpa, which occurs in a depth of ~6 μm to the surface (see Fig. 1(b)). Meanwhile, the average Xe fraction from 5 micron to 8 micron from the surface is approximately 0.92%, which is equivalent to a 6.36%FIMA (fissions per initial metal atom).

2.3. Characterization of the irradiated specimen

The PIE characterization of the Xe-implanted U_3Si_2 specimen was carried out at the Materials Characterization Suite (MaCS) at Center for Advanced Energy Studies (CAES). A transmission electron microscopy (TEM) foil was lifted out (see Fig. 1(a)) from the center of the beamspot on the irradiated specimen and thinned to approximately 75 nm (see Fig. 1(b)) using an FEI Quanta 3D FEG focused ion beam (FIB). Then the TEM foil was investigated by an FEI Tecnai TF30-FEG STwin STEM working at 300 kV. Both TEM bright field (BF) and scanning transmission electron microscopy (STEM) high-angle annual dark field (HAADF) imaging techniques were utilized to examine the microstructures in the specimen, especially the Xe bubbles. Additionally, STEM-based energy dispersive X-ray spectroscopy (EDS or EDX) was used to analyze the element compositions of various phases in the specimen.

3. Results

3.1. Phases identification

As previously reported by the manufacturers of the U_3Si_2 [5], two intrinsic secondary phases, USi and UO_2, coexist with the U_3Si_2 matrix in the fuel pellet in this study. After Xe implantation, those secondary phases were observed to remain in the specimen as indicated by Marks 3 and 4 in Fig. 2(a). In comparison to the U_3Si_2 matrix (Fig. 2(c)), the EDS analysis of the Mark 3 precipitate (Fig. 2(g)) clearly shows the existence of extra Si, which is consistent with the element composition of USi. On the other hand, the EDS analysis of the Mark 4 precipitate (Fig. 2(h)) illustrates the enrichment of O and depletion of Si, which implies the element composition of UO_2. The electron diffraction pattern (DP) of the Mark 4 precipitate further shows the characteristic [001] zone DP of UO_2 (Fig. 2(b)).

Aside from the intrinsic secondary phases, high-temperature ion implantation also facilitated the formation of an approximately 1 micron thick oxidation layer (see Mark 1 in Fig. 2(a)). As previously observed in the *in situ* TEM ion irradiation investigation of U_3Si_2 [7] and reported in the *ex situ* ion irradiation experiments of Ce_3Si_2 [26], a non-radioactive surrogate of U_3Si_2, U_3Si_2 is readily to be oxidized under irradiation, even in a ~10^{-5} Pa vacuum in TEM