Radiation-induced Grain Subdivision and Bubble Formation in U_3Si_2 at LWR Temperature

Tiankai Yao¹, Bowen Gong¹, Lingfeng He², Jason Harp², Michael Tonks³, Jie Lian^{1*}

¹. Department of Mechanical, Aerospace & Nuclear Engineering, Rensselaer Polytechnic

Institute, Troy, NY 12180, United States

². Idaho National Laboratory, Idaho Falls, ID 83415, United States

³. Mechanical and Nuclear Engineering Department, Pennsylvania State University,

University Park, PA 16802, United States

* Corresponding author

Abstract:

U_3Si_2 , an advanced fuel form proposed for light water reactors (LWR), has excellent thermal conductivity and a higher fissile element density. However, limited understanding of the radiation performance and fission gas behavior of U_3Si_2 is available at LWR conditions. This study explores the irradiation behavior of U_3Si_2 by 300 KeV Xe^+ ion beam bombardment combining with *in-situ* transmission electron microscopy (TEM) observation. The crystal structure of U_3Si_2 is stable against radiation-induced amorphization at 350 °C even up to a very high dose of 64 displacements per atom (dpa). Grain subdivision of U_3Si_2 occurs at a relatively low dose of 0.8 dpa and continues to above 48 dpa, leading to the formation of high-density nanoparticles. Nano-sized Xe gas bubbles prevail at a dose of 24 dpa, and Xe bubble coalescence was identified with an increase of irradiation dose. The volumetric swelling resulting from Xe gas bubble formation and coalescence was estimated with respect to radiation dose, and a 2.2% volumetric swelling was observed for U_3Si_2 irradiated at 64 dpa considering fission gas only.

Due to extremely high susceptibility to oxidation, the nano-sized U_3Si_2 grains upon radiation-induced grain subdivision were oxidized to nanocrystalline UO_2 in a high vacuum chamber for TEM observation, eventually leading to the formation of UO_2 nanocrystallites up to 80 dpa.

Keywords: U_3Si_2 fuel; ion irradiation; gas bubble; polygonization; oxidation;

I. Introduction

U_3Si_2 , combining with advanced claddings, is being considered as a promising ATF concept under active development by Westinghouse Electrical Company LLC, collaborating with multiple national labs and universities. U_3Si_2 displays a high fissile element density up to 11.3 g U/cm^3 , 17% more uranium loading than UO_2 fuels. U_3Si_2 also displays high thermal conductivity at elevated temperature (*e.g.*, $\sim 25 \text{ W/m}\cdot\text{K}$ at 1673 K [1]) and a melting temperature of 1665°C . The superior uranium loadings and thermal properties of U_3Si_2 can potentially reduce the operation temperature, enable power uprates, extend fuel cycle, and improve safety margin of LWR reactor operation.

In order to support the validation of U_3Si_2 as a qualified fuel form for LWRs. U_3Si_2 fuel pellets have been fabricated in large quantity through arc melting, atomization, and subsequently sintering [2]. BISON code simulations [3] suggests a significant decrease of fuel temperature compared with standard UO_2 fuel at all the investigated burnup conditions. However, simulated temperatures for U_3Si_2 at LWR conditions are higher than 300°C , necessitating a reevaluation of the available knowledge regarding the irradiation behavior of U_3Si_2 since all the available radiation data for U_3Si_2 is lower than 250°C [4]. Historically, U_3Si_2 has been used as a dispersed phase in aluminum plate-type reactor fuel, and the knowledge of radiation behavior under relatively low temperature ($< 150^\circ\text{C}$) to very high fission density is available [5]. $\text{U}_3\text{Si}_2/\text{Al}$

plate-type dispersed fuel after a burn up of 5.2×10^{21} f/cm³ showed uniformly-distributed separate small fission gas bubbles and a stable swelling behavior in U₃Si₂ [6]. However, a temperature dependence of bubble morphology was found in U₃Si₂/Al plate fuel irradiated at Advanced Test Reactor (ATR) [4]. At a local life average fuel temperature of 160 °C, there is an appearance of interconnected abnormally-enlarged fission gas bubbles up to 40 μm after a comparable fission density of 6.1×10^{21} f/cm³ [4]. Limited ion beam bombardment experiments were performed on U₃Si₂ at low-temperature regimes [7] with key observations including follows: (1) U₃Si₂ is sensitive to radiation-induced amorphization at low temperature, and the critical amorphization dose is 0.38 dpa (1.5 MeV Kr⁺ irradiations) or 1.1×10^{23} fissions/cm³ (neutron irradiation) at 30 °C; (2) A 2.2% volume contraction observed for U₃Si₂ upon radiation-induced amorphization, potentially beneficial for suppressing fission gas mobility; (3) a threshold temperature of 250 °C existing at a dose rate of 1.5×10^{-3} dpa/s, above which no amorphization can occur. Very recently, Miao [8] studied irradiation of U₃Si₂ using 1 MeV Kr²⁺ at 350 °C and 550 °C, temperatures predicted by BISON code [9] respectively for U₃Si₂ pellet rim and center in LWR condition at a burnup of 30-50 MWD/MTU.

In this study, the irradiation behavior of U₃Si₂ was investigated by 300 keV Xe⁺ ion beam at 350 °C and the microstructure evolution was studied by *in-situ* TEM observation. U₃Si₂ experienced a radiation-induced grain subdivision initiating at a relatively low dose of 0.8 dpa. A competition between irradiation induced grain polygonization and oxidation of U₃Si₂ was observed due to the high sensitivity of the nano-sized uranium silicides to oxidation. Xe-gas bubbles formed and grow with increased Xe concentration and radiation damage level. The volumetric swelling of U₃Si₂ was estimated based purely on Xe gas bubble formation and

coalesce. Results reported here demonstrate that U_3Si_2 fuel may experience grain subdivision, similar to the rim structure of UO_2 oxide fuels in LWR conditions.

II. Experiments

The bulk uranium silicide pellets were prepared through a powder metallurgy method following arc melting of constitutive uranium metal and pure silicon and consolidation by conventional high-temperature sintering. The lamella sample for ion beam irradiation was prepared by focused ion beam (FIB) with a uniform thickness of 100 nm. 300 keV Xe ion irradiation was performed using IVEM-Tandem facility at Argonne National Laboratory, and ion flux is 6.25×10^{11} ions/cm²·s. Irradiation temperature was carefully controlled at 350 °C through a GATAN double-tilt heating stage. After the irradiation temperature was reached, detail microstructure characterization took ~ 30 mins before the Xe beam started bombarding the FIB lamella. Microstructure evolution, structural transformation and bubble formation of uranium silicides upon irradiation were closely monitored through bright field transmission electron microscopy (BFTEM) imaging and selected area electron diffraction (SAED) on clean area with little FIB damage in Hitachi H900 NAR intermediate voltage electron microscope (IVEM) operated at 300 KeV. Electron beam was turned off during irradiation experiment.

Radiation damage profile and Xe distribution were calculated in quick Kinchin-Pease model SRIM 2013[10] (Fig. 1), and 61 eV and 15 eV were chosen as the atom displacement energy for uranium atoms as reported in references ⁵ and silicon atoms, respectively. An incidence angle of 15°, the same as the charged heavy ion incident angle with respect to the normal direction of TEM FIB lamella, was used for the SRIM damage calculation. The calculated damage rate on U_3Si_2 irradiated by 300 keV Xe^+ ions is 5.03×10^{-3} dpa/s. Xe atomic concentration has a peak

value of $\sim 2.78\%$ at a depth of 50 nm for 300 keV Xe^+ irradiation in U_3Si_2 at an ion flux of 6.25×10^{11} ions/cm²·s.

The chemical composition of the Xe-irradiated sample was analyzed using a Cameca SX-100 electron probe micro-analyzer (EPMA). Interaction of electron beam with U_3Si_2 FIB lamella is simulated with Casino [11], a software package based on Monte Carlo simulation of electron trajectories in solids. An operation voltage of 10 keV was selected based on simulation results as electrons generated by the selected accelerating voltage have sufficient energy to penetrate the 100 nm thick FIB U_3Si_2 lamella with limited interaction with the sample holder underneath. Energy dispersive spectrum (EDS) in a large area was also obtained by the EPMA to verify that there are no counts from the holder materials. Five locations were randomly selected through the $10 \times 20\ \mu\text{m}$ sample area, and the elemental distribution was averaged to reveal the chemical composition of the sample. The standard deviation of the measured values from 5 locations was estimated as the experimental uncertainty. The standard used for U element was UO_2 ; while ThSiO_4 was used for O and Si elements.

Post-irradiation TEM images were acquired to further reveal the size and distribution of Xe gas bubbles in the Xe-irradiated U_3Si_2 , in complementary to Fresnel contrast imaging from the same area, under-focused and over-focused TEM images were imported into MATLAB[®] and converted to RGB values at each pixel and consequently converted to a grayscale value at each pixel. Since gas bubbles show the white contrast in under-focused images and black contrast in over-focused images, the generated images enable us to reveal the location and size of gas bubbles by subtracting gray values at each pixel. Despite this, when analyzing these images, gas bubbles were found to be close to each other, making it hard for image-processing software to analyze them automatically. Therefore, we manually counted the numbers of bubbles and

measured bubble sizes in several randomly-selected regions using ImageJ. Volumetric swelling induced by Xe bubbles was calculated based on the number of Xe bubbles and size measured in 2-dimensional TEM images. We also assume that the Xe gas bubbles are the only source for volumetric swelling and no radiation-induced dimensional change was considered when reporting the volumetric swelling [12].

III. Results and Discussion

Due to extreme conditions of high temperature and intensive radiation, it is expected that nuclear fuels experience drastic microstructure evolution and restructuring induced by radiation damage from energetic neutrons (up to 5 MeV) and highly ionizing fission fragments (with energy up to 100 MeV). Materials subjected to intensive displacive damage, e.g., by charged heavy particles, will also experience radiation-induced amorphization and structure transformation under relatively low temperatures, and radiation-induced defects can be recovered at higher temperatures due to a dynamic annealing process. A critical value in determining materials radiation resistance against amorphization is the critical amorphization temperature, above which all of the defects can be recovered and no amorphization can occur anymore. LWR fuel UO_2 and its isostructural oxides with a simple fluorite structure, such as CeO_2 , and ZrO_2 , exhibit excellent structural stability and resistance against radiation-induced amorphization [13]. Different from oxide fuels, U_3Si_2 , an intermetallic compound with a tetragonal crystal structure, is sensitive to radiation-damage-induced amorphization at low temperature. Both heavy ion and neutron irradiations confirm that U_3Si_2 lose its crystal structure and is readily amorphized at a low dose of ~ 0.3 dpa at room temperature. However, the critical amorphization temperature is as low as ~ 240 °C due to the thermal recovery of irradiation damage [14].

Since the relevant LWR operation temperature (both inlet and outlet temperatures) is high enough to ensure the temperature of the fuel rim above the critical amorphization temperature of U_3Si_2 , it is expected that U_3Si_2 fuel will be stable against radiation-induced amorphization. This is confirmed by *in-situ* TEM observation of the 300 keV Xe-irradiated U_3Si_2 at 350 °C. Before irradiation, a SAED pattern showing single crystalline features for the FIB lamella was obtained (Fig. 2a), which can be well indexed by U_3Si_2 with tetragonal structure along $[0\bar{4}8]$ zone axis. No diffuse halos were observed in the SAED patterns with increased radiation damage levels up to 64 dpa (Fig. 2e), 200 times of the critical dose required to fully amorphize U_3Si_2 at room temperature. These results indicate that complete amorphization does not occur in U_3Si_2 upon intensive displacive damage, and U_3Si_2 as the leading ATF concept is likely stable against radiation-induced amorphization upon energetic neutron damage at reactor conditions.

A distinct feature observed in the SAED for unirradiated U_3Si_2 is the existence of weaker diffraction spots of (100), (200) and (300) as indexed in Fig. 2a which can be attributed to the structural modulation and existence of superstructure. The superstructure is typically observed in materials with complex compositions (e.g., minerals) with multiple cations in which a chemical ordering exists due to the distribution of cations on equivalent crystallographic sites. Such an ordered structure is possibly associated with non-stoichiometry of U_3Si_2 recently suggested by a DFT study [15]. It still remains open for the structural modulation and chemical ordering of the superstructure observed.

Upon radiation, uranium silicide experiences a rapid order-disorder structure transition as evidenced by the disappearance of the superstructure diffraction spots in SAED patterns at a relatively low dose (~ 0.8 dpa). In fact, Al_3Zr , which share same crystal structure prototype (Cu_3Au) with U_3Si_2 , readily experience the similar order-to-disorder transition under the

exposure to beam radiation of neutrons and charged heavy ions at low radiation level ~ 1 dpa [16]. and therefore, the order-to-disorder transition is considered posing no appreciable effects on the radiation response of U_3Si_2 fuels [17].

In addition to the superstructure, additional subsets of diffractions were also identified that can be indexed to different zone axis $[\bar{1}\bar{2}\bar{2}]$ (Fig. 2b) and $[\bar{1}\bar{2}\bar{1}]$ (Fig. 2c). The additional subset diffractions may be due to the existence of the second grain within the FIB lamella or the sample bending from Xe^+ irradiation satisfying additional diffraction conditions. Sample bending and appearance of additional subset diffractions were also observed by *in-situ* TEM for CeO_2 upon Kr^{2+} irradiation [18]. A close-up look of the SAED pattern for U_3Si_2 irradiated to 0.8 dpa shows diffuse spots distinctively on (210) and (011) planes (Fig. 3a), indicating the appearance of sub-grains within the starting U_3Si_2 grains. A narrow distribution of extra spots on the arching indicated that those newly formed sub-grains may share small-angle grain boundaries. Similar features on SAED patterns have been reported for HBS in spent UO_2 fuel [19], and the variation of sub-grains orientations is very small.

Radiation-induced grain subdivision and polygonization were also observed in Xe-irradiated U_3Si_2 at 350 °C, and the grain subdivision initiated a very low radiation level of ~ 0.8 dpa as evidenced by the diffused SAED spots (Fig. 3a), the characteristics of nanostructured materials. The grain subdivision as indicated by the appearance of polycrystalline diffraction rings is clearly revealed in the sample irradiated by 24 dpa (Fig. 2c and Fig. 3b). The polycrystalline rings were indexed by the tetragonal structure of the U_3Si_2 as labeled in Fig. 3b. Accompanying with radiation-induced grain subdivision and polygonization, oxidation of nano-sized U_3Si_2 also occurred during ion beam irradiation and *in-situ* TEM observation, resulting in the appearance of the polycrystalline diffraction rings that can be indexed to UO_2 nanocrystals (Fig. 3b). Due to the

similarity of some inter-plane spacing between these two compounds, overlaps of the polycrystalline rings from both U_3Si_2 and UO_2 nanocrystals were observed. Such small lattice mismatch of specific d-spacings between U_3Si_2 and UO_2 is consistent with the susceptibility of U_3Si_2 against oxidation. However, characteristic diffraction rings exist exclusively from U_3Si_2 , e.g., the (221) plane (see Fig. 2d-e and Fig. 3b) which cannot be indexed by structural parameters of UO_2 or other uranium silicide compounds (e.g., U_3Si_5). This enables the exclusive determination of the nano-sized U_3Si_2 and radiation-induced grain subdivision process. The radiation-induced grain subdivision continues with the radiation damage level beyond 64 dpa as evidenced by the fact that the diffraction pattern (Fig. 2e) consists of residual diffractions (e.g., (421) spot and (221) ring) of U_3Si_2 .

Microstructural evolution of the Xe-irradiated U_3Si_2 was also closely monitored by BFTEM imaging as a function of irradiation doses (Fig. 4). Throughout the investigated irradiation dose range up to 80 dpa, there is a constant evolution of band contour with irradiation due to irradiation-induced strain. The high density of nano-sized grains showing dark diffraction contrast in the BFTEM image formed upon irradiation dissociated from the original large-sized grains, which become more noticeable with the dose above 24 dpa (Fig. 2c). The formation of nano-sized grains can be attributed to the radiation-induced grain subdivision of U_3Si_2 and oxidation of the nanocrystalline silicides to oxides. It is difficult to differentiate silicide nanocrystals from oxide nanocrystals due to similar diffraction contrast, consistent with the observation of both silicide and oxides coexisting as indexed by SAED patterns (Fig. 3). These nano-sized grains experience a moderate radiation-induced grain coarsening with the size increasing up to 20 nm at the highest dose of 80 dpa (as shown in Fig. 4f). Radiation-induced grain subdivision and polygonization are well documented in UO_2 oxide fuels at the rim region

of the fuel pellets with higher burn-up[20]. The rim structure or high burn-up structure (HBS) is typically characterized by a mesoporous material in which micron-sized pores embedded into the nanocrystalline UO_2 matrix with an average grain size of several hundred nm. The formation mechanism of the HBS is generally considered as a competition between irradiation-induced grain polygonization and grain recrystallization/growth at reactor working temperatures. Fission gas bubbles may also play an important role in which an optimized ratio of grain size/pore size may exist for the formation and stabilization of the HBS. Radiation-induced grain subdivision and polygonization observed here suggest a possible formation of rim structure in U_3Si_2 as the leading ATF fuel during reactor operation. Similar to oxide fuels, the nanostructure remains stable without dissociation or drastic size increase upon high dose radiation (up to 64 dpa) at 350 °C.

The 300 keV Xe^+ irradiation-induced grain subdivision at 350 °C observed in this study is consistent with an earlier study of radiation-induced polygonization of U_3Si_2 by 1.5 MeV Kr^{2+} at 350 °C [14]. In their study, the starting microstructure was totally replaced by refined subdivided grains at a radiation dose of ~10 dpa, however, the dose to initiate the grain subdivision was not provided. We also performed 1 MeV Kr^{2+} ion beam irradiation at the same temperature, and the radiation damage-induced grain subdivision also occurs in U_3Si_2 with the initial dose for grain subdivision between 3~4 dpa (data not shown here), which is substantially higher than the 0.8 dpa reported here for 300 keV Xe^+ . The difference of required dose levels for the onset of grain subdivision could be attributed to the ion trajectory in U_3Si_2 of different ion type and energy. For 1.5 MeV Kr ion irradiation, all of the Kr ions penetrated through the TEM samples, and the radiation-induced microstructure evolution is mainly due to the point defect accumulation. However, the 300 keV Xe^+ beam used in this study will be implanted inside the TEM samples,

leading to the formation of high density nano-sized bubbles (as discussed later). The coupled effects from both Xe bubbles and radiation defect accumulation may lead to a lower initiate dose for grain subdivision.

At higher dose irradiation of 80 dpa, only weak diffraction spots with dim contrast exist for U_3Si_2 , and the dominant diffraction ring patterns can only be indexed to nano-crystalline UO_2 (Fig. 3c), suggesting that almost all of the nano-sized silicides were oxidized to nano-sized UO_2 . Using electron microprobe analysis, a substantial amount of oxygen was found in the Xe-irradiated U_3Si_2 at a dose of 80 dpa at 350 °C (Fig. 5), and the average U/Si ratio is close to stoichiometric U_3Si_2 , confirming the oxidation of the silicide under current experimental conditions. In dry oxidation condition, U_3O_8 and UO_2 are major oxidation product [21]. While in the appearance of water, typical hypothesis for accident scenario, hydrogen gas is proposed to form [22]. In low oxygen partial pressure, i.e. high vacuum or protecting environment such as helium gas, the oxidation behavior remains unclear so far with limited oxidation behavior of the silicides.

The transition of micron-sized U_3Si_2 to nano-sized UO_2 was also observed upon different ion irradiations (*e.g.*, 1 MeV Kr^{2+}) in the same facility at the same temperature [8]. In their experiment, the formation of nano-sized UO_2 from U_3Si_2 upon irradiation was attributed to irradiation enhanced/induced oxidation due to the fact that part of the sample which was shielded from irradiation by sample grid remains crystalline U_3Si_2 . Surrogate Ce_3Si_2 [2] was also oxidized by proton irradiation with oxides built up on the sample surface at a comparable vacuum level as the TEM utilized here; whereas bulk CeSi_2 is not oxidized at the same experimental condition. These results suggest that irradiation enhanced/induced oxidation as proposed in Ref [7] may not be the dominant mechanism for the formation of high-density nano-sized UO_2 . Oxidation may

occur in U_3Si_2 at temperature of 350 °C on the timescale of minutes albeit in a low oxygen environment (e.g., 0.2 atm oxygen) [21]. Additionally, radiation-induced grain subdivision may also contribute to the oxidation process. U_3Si_2 nanocrystalline maybe more susceptible to oxidation due to the greatly increased density of surfaces and grain boundaries and maybe easily be oxidized even in a high vacuum TEM chamber.

In addition to radiation-induced microstructure evolution, fission gas behavior and gaseous swelling of nuclear fuel are also critical for evaluating fuel performance. Fission gas behavior was extensively studied on $\text{U}_3\text{Si}_2/\text{Al}$ dispersion fuels with the focus on bubble formation and volumetric fuel swelling. At low temperatures, stable swelling occurs in U_3Si_2 even at a very high fission density (i.e. burn-up), in contrast to the breakaway swelling observed in many other intermetallic phases including U_3Si . Visible fission gas bubbles first start to form on grain boundaries and grow slowly as more fission gas atoms are produced. This stable swelling behavior was explained as a result of relatively small and uniform fission gas bubbles that do not have a tendency to coalesce under these conditions. Fission gas behavior in U_3Si_2 is affected significantly by fission rate, uranium-to-silicon ratio, and temperature. At a very high fission density, the population of bubbles increases and becomes bimodal. At a temperature of 160 °C, large fission gas bubbles with size up to ~40 microns were observed at a fission density of $6.1 \times 10^{21} \text{ f/cm}^3$ as a result of bubble growth [4]. Bubble interconnection also occurred, leading to the breakaway swelling of the fuel particles. However, fission gas behavior in monolithic silicide fuels at relevant reactor operation temperatures has not been reported.

The formation and redistribution of Xe gas bubbles in the 300 keV-irradiated U_3Si_2 at 350 °C were also investigated and the bubble size/density with respect to irradiation doses was quantitatively analyzed. High-density nano-sized bubbles can be seen by through-focused bright

field TEM images (*e.g.*, Fig. 6a) showing the bright contrast of cavities in the under-focused image (over-focused BFTEM images were not shown here). Xe bubbles with several-nm diameter become pervasive (Fig. 6) at a high dose regime above 24 dpa. However, strong diffraction contrasts from high-density nano-sized nanoparticles upon radiation-induced polygonization make it difficult in determining exact bubble morphologies. To clearly reveal bubble morphology and determine bubble sizes, through-focused BFTEM images were processed to align the under-focused and over-focused images at the exact location by MATLAB[®]. As stated in the experiment section, a matrix containing grayscale values for each pixel can be created in MATLAB[®]. By simply subtracting the matrix generated from the under-focused image and the over-focused image, a new matrix can be obtained, which contains the difference between these two images. This new matrix was then converted back to an image, which reflects the difference between the aforementioned two images. In the generated image, gas bubbles are in dark color while other regions are in bright color, which makes it much easier to distinguish gas bubbles. The diffraction contrasts resulting from nanoparticles are removed such that the bubbles can be clearly revealed (see enhanced contrast images in Figs. 6d-f). Statistical analysis was performed by manually counting the number of bubbles and measuring its size from different areas of the enhanced contrast images, and uncertainties of the bubble size and density were derived based on the standard deviation of the distribution.

Fig. 7a shows a histogram of the numbers of bubbles per unit volume at different bubble sizes estimated from the measured value of bubble areal density from 2D TEM images and assuming 100 nm as the thickness of the FIB lamella. A single mode distribution of the bubble size can be identified for samples irradiated at different damage levels. It should be recognized the uncertainties are induced in discerning exact bubble morphologies due to small bubble size.

In general, there is an appreciable shift of the peak size for Xe bubbles from ~2.0 nm to ~3.5 nm with increased irradiation doses, suggesting a minor bubble growth. In addition to bubble size increase, a decrease of number density at higher irradiation doses is observed due to radiation-induced bubble coalescence. Note that the bubble size studied here is generally smaller than 5 nm, well below the resolution limit of SEM (40 nm) used before for PIE observation of $\text{U}_3\text{Si}_2/\text{Al}$ dispersed plate fuels shown in previous studies [23]. Interestingly, the sluggish bubble growth observed here with intense radiation at 350 °C is probably due to the low mobility of Xe in silicides. However, this is in contrast to the large size bubble formation/growth up to 50 microns and the breakaway of $\text{U}_3\text{Si}_2/\text{Al}$ dispersed fuels observed at 160 °C [4]. A surface effect may occur leading to a minimized bubble growth in the 100 nm-thick TEM lamella.

The sluggish bubble growth behavior upon low energy 300 keV Xe^+ irradiation in this study is consistent with a recent irradiation study on bulk U_3Si_2 by high energy 84 MeV Xe^+ to 500 dpa at 300 °C [24] showing the formation and a uniform distribution of dense 10 nm Xe gas bubble. The formation of high density nano-sized bubbles with a uniform distribution suggests a favorable fuel gaseous swelling behavior of monolithic U_3Si_2 fuel in LWR conditions. Post irradiation examination of the ATF-1 U_3Si_2 fuel burn to $\sim 6 \times 10^{20}$ fissions/cm³ also shows no run-away swelling and the clear gap between U_3Si_2 and cladding [24]. These results all together by high energy and low energy Xe irradiations imply a favorable fission gas behavior. The volumetric fuel swelling in ATF U_3Si_2 fuel might be different from the previously reported one in a low-temperature research reactor, necessitating a systematically revisit of the fuel swelling behavior at a boarder range of radiation conditions at different doses, rates, and temperature.

Estimated volumetric expansion due to Xe gas bubbles indicates that there is a peak volumetric expansion with radiation damage levels (see Fig. 7b in which the radiation-induced

coarsening of nanocrystals is also included). The estimated volumetric swelling increases with irradiation dose up to 64 dpa (0.8% for 24 dpa, and 2.2 % for 64 dpa), and then slightly decreases to 80 dpa (1.3 %). Note that the volumetric swelling is estimated only based on the measured bubble density and size from 2D images and extended to 3D geometry. The uncertainty in determining bubble size and density will propagate into the estimation of the volumetric swelling. In addition, radiation-induced volumetric change is not included, in which a 2.2 % volumetric contraction was also reported by Ref. [25] due to full amorphization of U_3Si_2 . Volumetric change is also expected to result from the phase transformation from crystalline U_3Si_2 to nano-sized U_3Si_2 and eventually, amorphous UO_2 since the unit volume of UO_2 is 0.162 nm^3 while that for U_3Si_2 with the same U content is 0.209 nm^3 . Such a reduced free volume is considered beneficial to fission gas retention as it suppresses the fission gas mobility. On the other hand, the incorporation of additional oxygen and other oxides will lead to volumetric expansion. Despite the estimated swelling is less quantitative with uncertainties, the relatively small volumetric swelling (~a few vol.%) resulting from fission gas accumulation only is encouraging. Further experiments performed at higher temperatures and in bulk samples mitigating surface effects are necessary to gain a complete understanding of the fission gas behavior (bubble formation, migration, and growth) in silicide fuels.

IV. Conclusions:

In this paper, radiation-induced microstructure evolution and fission gas behavior of U_3Si_2 irradiated by 300 keV Xe^+ at 350 °C are studied by *in-situ* TEM. U_3Si_2 is resistant against amorphization at reactor operation temperatures even up to a very high dose of 60 dpa. Radiation-induced polygonization and grain subdivision occur in U_3Si_2 irradiated by 300 keV Xe at 350 °C. The polygonization begins at a low dose ~0.8 dpa and continues beyond 48 dpa. The

radiation-induced polygonization leads to the formation of a high density of nanoparticles (~20 nm) which experience a grain coarsening with radiation dose up to 80 dpa. Xe bubbles form in Xe irradiated U_3Si_2 and a single mode size distribution was observed. A radiation-induced bubble coalescence is also identified, and the estimated volumetric swelling from gas bubbles only is low. Uranium silicide nanoparticles upon radiation-induced grain subdivision are easily oxidized even during *in-situ* TEM observation, eventually leading to complete oxidation to UO_2 nanocrystals stable up to 80 dpa. These results indicated that at relevant reactor operation conditions, uranium silicide as the leading candidate of ATFs is stable against radiation-induced amorphization, but likely experiences a grain subdivision and possibly a rim structure, similar to UO_2 fuels.

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Figure Captions:

Fig. 1 Damage profile and Xe ion distribution calculated from SRIM simulation of U_3Si_2 subject to 300 keV Xe^+ irradiation with a dose of 1×10^{16} ions/cm²

Fig. 2. Selected area electron diffraction (SAED) patterns of U_3Si_2 after irradiation by 300 keV Xe^+ at 350 °C at various dose of (a) as received; (b) 0.8 dpa; (c) 24 dpa; (d) 48 dpa; (e) 64 dpa; and (f) 80 dpa

Fig. 3 SAED patterns of U_3Si_2 after irradiation by 300 keV Xe^+ at 350 °C at various doses of (a) 0.8 dpa; (b) 24 dpa; and (c) 80 dpa. Irradiation induced grain polygonization of U_3Si_2 occurs at 0.8 dpa; At 24 dpa, rings for nano UO_2 grains (red colored) coexist with one for sub grains of U_3Si_2 (green colored). At 80 dpa, nano UO_2 is the dominant phase whereas tracks for sub grains of U_3Si_2 highlighted by white pseudo-hexagonal frame is significantly weak

Fig. 4 In-situ bright field TEM images of U_3Si_2 after irradiation by 300 keV Xe^+ at 350 °C at various dose of (a) as received; (b) 0.8 dpa; (c) 24 dpa; (d) 48 dpa; (e) 64 dpa; and (f) 80 dpa

Fig. 5 EPMA results for the elementary composition in U_3Si_2 irradiated by 300 keV Xe^+ at 350 °C to 80 dpa

Fig. 6 Xe gas bubble formation and evolution with irradiation doses of (a, d) 24 dpa; (b, e) 64 dpa; and (c, f) 80 dpa, where a, b, and c are under-focused BF TEM images; and d,e,f are processed images to show Xe bubbles size and distribution.

Fig. 7 Xe gas bubble size distribution (a) and size of nano-crystalline (b, open square) and estimated volumetric expansion (b, solid square) at various irradiation doses.

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5

Fig. 6

Fig. 7