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In-situ ion irradiation of fission products in a spent UO_2 fuel

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This study investigates the behavior of fission gas bubbles and five metal precipitates (5MPs) (Mo, Ru, Rh, Tc, Pd) in spent uranium dioxide (UO_2) fuel under various ion irradiation doses, temperatures, and flux conditions. Utilizing in-situ ion irradiation and advanced transmission electron microscopy, we analyzed the evolution of fission gas bubbles and 5MPs in UO_2 samples from the Belgium Reactor 3 (BR-3). Our findings reveal significant shrinkage of fission gas bubbles and 5MPs with increasing irradiation dose, accompanied by a decrease in pair density. We demonstrate that ion irradiation induces a homogeneous re-resolution process where individual atoms are ejected from bubbles and precipitates, leading to their dissolution and subsequent re-precipitation in the matrix. This study provides critical insights into the dynamic behavior of fission products under irradiation, facilitating the development of predictive models and contributing to the optimization of nuclear fuel performance and safety.

I. Introduction

A comprehensive understanding of the behavior of nuclear fuels under irradiation is critical for enhancing the efficiency and safety of nuclear reactors. Uranium dioxide (UO_2), the primary fuel used in commercial light water reactors, experiences significant microstructural changes during nuclear fission, driven by the generation of fission products within the UO_2 matrix. Noble gases like xenon (Xe) and krypton (Kr), accounting for about 15% of fission products, have low solubility in UO_2 [1]. As a result, they form small bubbles that coalesce, causing fuel swelling, mechanical degradation, and reduced thermal conductivity [2–6]. Additionally, metallic precipitates primarily composed of 4d group metals such as molybdenum (Mo), technetium (Tc), ruthenium (Ru), rhodium (Rh), and palladium (Pd), known as five metal precipitates (5MPs), further complicate fuel behavior under irradiation. These precipitates, also known as white inclusions [7, 8], fission-product alloy [8], epsilon particles [9, 10] and noble metal phase [11, 12], segregate in the matrix and grain boundaries, contributing to the fuel swelling. Contrarily, 5MPs may enhance fuel performance by increasing heat conduction and aiding waste storage by catalytically contributing to the reduction of actinides, promoting immobilization of the waste form, which is critical for effective nuclear waste management. This was demonstrated by Kleykamp, who showed that precipitated solid fission products may increase heat conduction of the fuel due to their higher thermal conductivity compared to the surrounding UO_2 matrix [12]. Broczkowski et al. conducted a set of electrochemical experiments to investigate the corrosion of UO_2 under nuclear waste disposal conditions, finding that the presence of 5MPs leads to a decrease in corrosion potential and inhibition of UO_2 oxidation [13]. Furthermore, as the nuclear industry progresses towards high burnup fuels [14] understanding the significant restructuring processes, including the formation of high burnup structure (HBS), becomes increasingly important. HBS, characterized by grain subdivision, redistribution of fission gases, and high concentration of intergranular porosity [15], has been linked to accelerated fission gas release, and fuel fragmentation during transient and accidental conditions [16]. Over the past decades, extensive research has been conducted on the microstructural characterization and chemical quantification of HBS regions in nuclear fuel [17–23], revealing that fission gas bubbles could exert a “pinning effect” on defect accumulation, which in turn promotes HBS formation [22].

Despite the critical role of fission gas bubbles and 5MPs in nuclear fuel behavior, detailed knowledge of their development and interaction with the microstructure at the atomic level remains incomplete. An earlier study by Baker examined five different UO_2 fuel pins to observe the effects of temperature and power density on fission gas bubble evolution [24]. Whapham studied neutron-irradiated UO_2 fuel samples at varying doses to understand fission gas bubble behavior [25]. However, these earlier studies, which relied on ex-situ irradiation capabilities, faced significant limitations. Specifically, the microstructure of irradiated fuel samples can change due to the temporal gap between irradiation and microstructural characterization. Transmission electron microscopy (TEM) with in-situ ion irradiation capabilities addresses these limitations by enabling real-time observation of fission product evolution under controlled conditions that closely simulate reactor operating conditions [26–28]. Furthermore, it facilitates accelerated evaluation of irradiation effects on materials [29]. In-situ observation is invaluable for understanding the mechanisms and driving forces behind fission product development under irradiation, thus aiding in the creation of predictive models for their dynamic evolution [30]. In this study, we utilized in-situ ion irradiation to investigate the morphological and quantitative changes of fission gas bubbles and 5MPs in spent UO_2 fuel samples from the Belgium Reactor 3 (BR-3). Meticulously chosen irradiation conditions isolated the effects of radiation and temperature on fission product evolution. Additional microstructural characterization before and after ion irradiation

was performed using ChemiSTEM™ technology to analyze the elemental distribution of relevant fission product phases [31]. This approach provides critical insights into the mechanisms and driving forces behind fission product evolution, aiding in the development of predictive models for their dynamic behavior under irradiation.

II. Experimental Details

The BR-3 spent UO_2 fuel stored at the Hot Fuel Examination Facility (HFEF), Idaho National Laboratory was used as a base material for ion irradiation experiments. The rod-averaged burnup of the fuel rod (I-334) at the end of irradiation was 39 GWd/t [32]. A fuel pin cross section, MNT52X was taken axially 4.5 cm below the centerline of I-334 BR-3 fuel rod, which was in the maximum power position. The relative gamma activity profile as a function of axial distance from the bottom of fuel rod I-334 is available in ref. [32]. Large block lift outs were taken near the fuel clad and rim region (at a location near $r/r_0 = 1$ in terms of the pellet cross section) of MNT52X using the FEI Helios Plasma Focused Ion Beam (PFIB). Multiple cross-section TEM lamellas from one of the blocks were prepared using a focused ion beam (FIB) system (Quanta 3D FEG, FEI). Each lamella was lifted out, mounted on a Mo grid and thinned to a thickness of approximately 100 nm using 30 keV Ga ions, followed by a final cleaning using 2 keV Ga ions. Locations of PFIB blocks and TEM lamella lift outs are shown in Fig. 1.

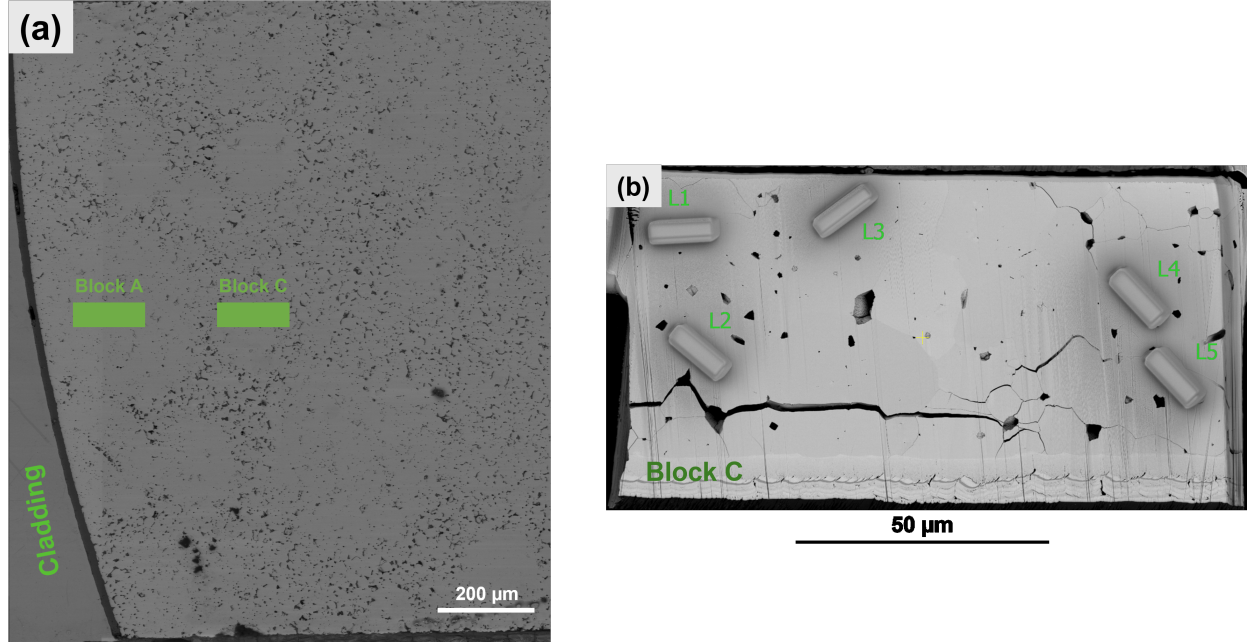


FIG. 1: Locations of PFIB lift outs near the the fuel clad and rim region of MNT52X. (a) SEM image of the fuel clad and rim region where 2 PFIB block lift outs are marked, (b) SEM image of block C where 5 FIB TEM lamella lift out locations are marked (L1-L5).

In-situ ion irradiation of these TEM lamellas was performed using a 300 keV Hitachi 9000 NAR TEM coupled with an NEC implanter, at the Intermediate Voltage Electron Microscope (IVEM) facility at Argonne National Laboratory. The angle between the ion beam and normal direction of sample surface was 15° . One of the irradiations was conducted at cryogenic temperature (-223°C) to observe the effect of radiation on the fission product evolution under athermal condition. In-situ TEM images were collected at several doses and analyzed manually by ImageJ, an image processing software [33] to measure the size of the bubbles and 5MPs and number of their co-localized pairs. The line measurement tool of ImageJ was used to estimate the diameter of bubbles and 5MPs, which, in this context, has been referenced as the size of the concerned fission product. Each reported value was the average of measurements taken from 3 areas for pair density and of approximately 20 features for the size measurements. Each size measurement was the average of long and short diameters of a bubble or 5MP. The electron energy loss spectroscopy (EELS) technique was applied to measure the thickness of the samples [34] which was used to calculate the density of pairs. Additional microstructural characterization of these samples was performed using the 200 kV FEI Titan TEM equipped with ChemiSTEM™ technology to analyze the elemental distribution of 5MPs and fission gas bubbles. Table I shows the irradiation conditions which have been used for the analysis.

The Stopping and Range of Ions in Matter (SRIM) computer code [35] was used to simulate the depth profiles of atomic displacements in UO_2 . The threshold displacement energies for oxygen and uranium were set to 20 and 40 eV , respectively, which were determined by Soullard in his electron irradiation experiments in UO_2 [36]. The incidence angles of Xe and Kr were set as 15° in the calculation. Simulations were performed in both full cascade and Kinchin-Pease (KP) damage calculation modes [37]. The results from the full cascade mode are presented in the main

TABLE I: Irradiation Conditions

Ion	Temperature °C	Dose (Up to) ions/cm ²	Ion Flux ions/cm ² .s
1 MeV Kr	20	1×10 ¹⁵	6.25×10 ¹¹
	300		
	500		
300 keV Xe	20	1×10 ¹⁵	1.25×10 ¹²
	300	4×10 ¹⁵	
	-223	5×10 ¹⁵	1.25×10 ¹²
		1×10 ¹⁶	4.13×10 ¹²

content and the outputs from the KP mode are included in the supplementary material. The average damage for the TEM lamella under 1 MeV Kr irradiation is approximately 3.3 dpa (Fig. 2a) whereas, it is approximately 5 dpa under 300 keV Xe irradiation (Fig. 2b). The ion concentration profile shows an average value of approximately 0.01 at.% for Kr irradiation and 0.12 at.% for Xe irradiation (considering a 100 nm TEM foil), suggesting that very few ions were deposited in the observation areas of the TEM foil during irradiation. The Kr peak concentration of 0.01 at.% corresponds to an addition of roughly 0.16% FIMA (fissions per initial metal atom) and the Xe peak concentration of 0.12 at.% equates to an addition of 0.65% FIMA of BU in the fuel.

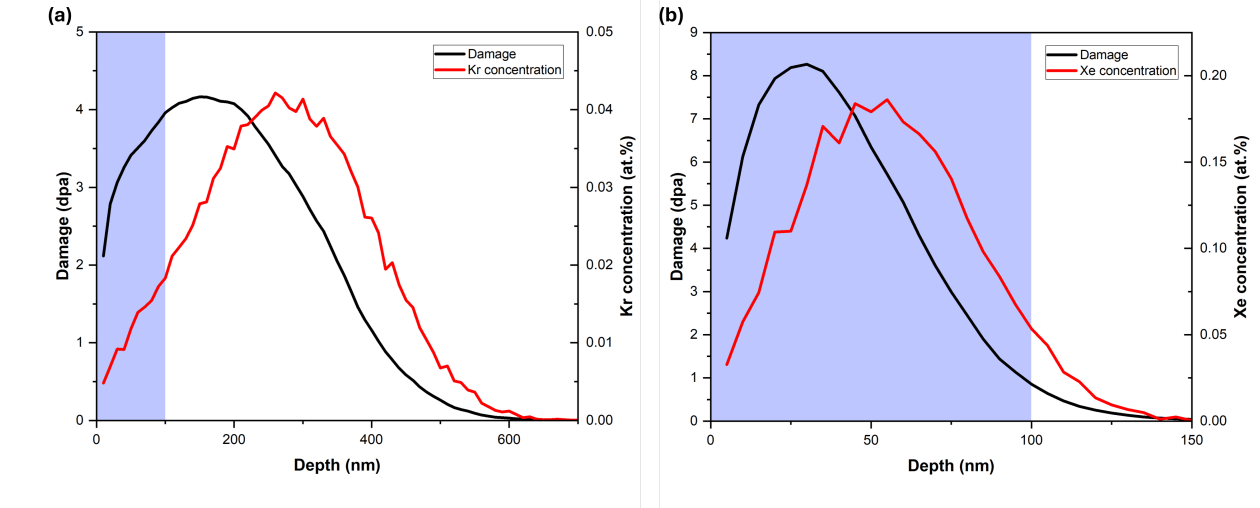


FIG. 2: The depth profiles of radiation damage in units of displacements per atom (dpa) and ion concentration in units of at.% obtained from a SRIM simulation of UO₂ subjected to (a) 1 MeV Kr ion irradiation and (b) 300 keV Xe ion irradiation, both subjected to a total fluence of 1×10^{15} ions/cm². For a TEM sample with a thickness of 100 nm, only the shadowed region needs to be considered.

III. Results

Prior to irradiation, the Xe/Kr bubbles were accompanied by 5MPs, forming pairs. Fig. 3 shows a TEM image of the sample before irradiation where, the bubbles and 5MPs are highlighted. They are approximately less than 7 nm in diameter, which has also been reported by Matzke et al. [38] and Thomas et al. [39] in their studies of fuel's rim region. In underfocused imaging condition, gas bubbles and 5MPs appear as white and black spots, respectively, due to massthickness contrast which originates from incoherent elastic scattering of electrons [40]. The contrast is reversed when the imaging condition is changed to overfocused [41]. Each reported underfocused image in the text has its focused and overfocused images. They were compared with each other to ensure correct size and density determination of the features concerned. The level of applied underfocus ranged from -0.75 μm to -1.5 μm . A specific value of underfocus was applied for every image of a set to reduce the uncertainty of bubble size determination related to the varying level of underfocus [41].

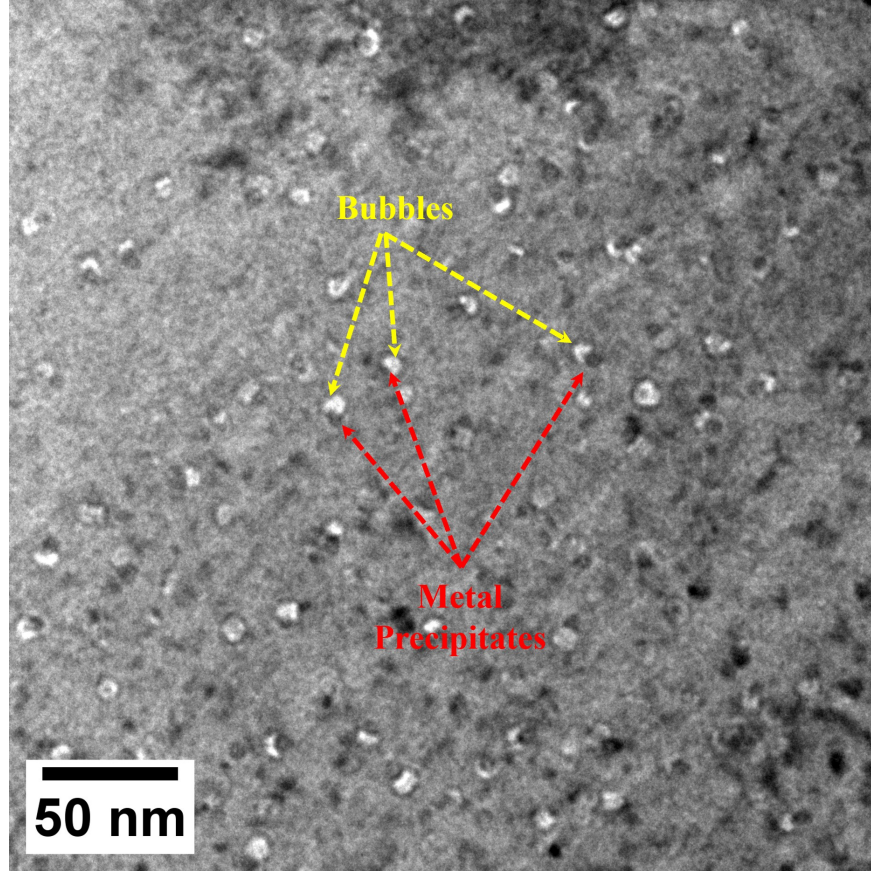


FIG. 3: TEM image of the sample before irradiation showing the pairs of nano bubbles and 5MPs. The image was captured under underfocused imaging condition, where the bubbles appear as white spots and the 5MPs appear as black spots.

The microstructural evolution of bubbles and 5MPs after 1 *MeV* Kr irradiation at room temperature (RT), 300°C and 500°C is illustrated in Fig. 4. All images shown were captured under underfocused imaging condition. These samples were relatively thinner, facilitating more detailed observations of the pairs. The shrinkage of the bubbles and 5MPs is evident with continued ion irradiation. Fig. 4(c-d) reveal the presence of extremely small bubbles, which were not clearly observed in samples at lower doses. These bubbles are likely Kr bubbles formed by implanted Kr ions. Fig. 5 shows the size of bubbles and 5MPs at different temperatures as a function of dose under 1 *MeV* Kr irradiation. The size of the bubbles and 5MPs decreased almost linearly with increasing irradiation dose at 300°C and 500°C, whereas at RT, the size decreased significantly when the ion beam was first activated and continued to decrease linearly with ongoing exposure. The change in pair density is summarized in Fig. 6. The pair density decreased linearly at RT and 300°C in response to the irradiation dose but showed an abrupt decrease at the first dose level at 500°C.

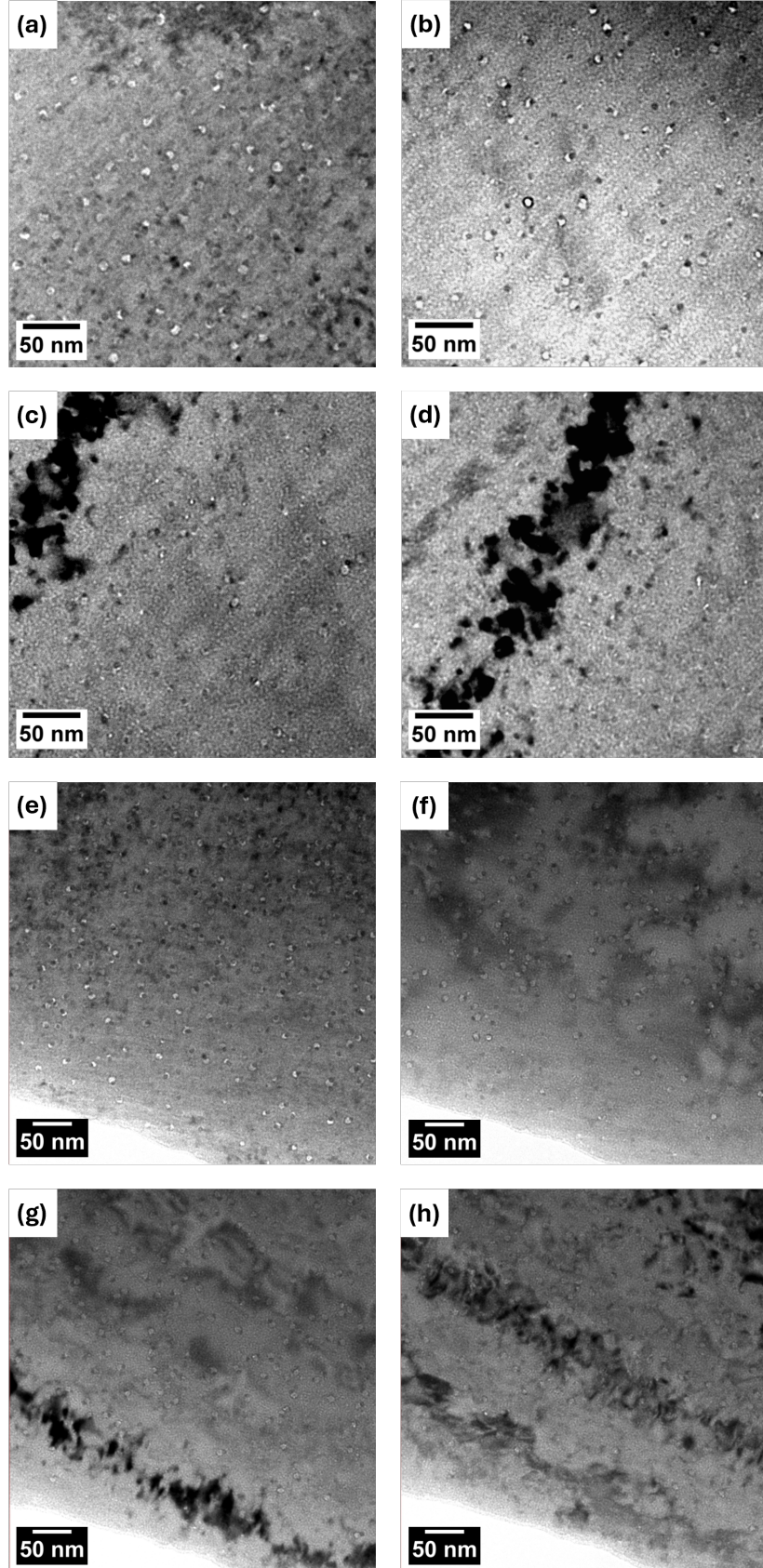


FIG. 4: TEM images of the sample irradiated by 1 MeV Kr at RT (a-d) and 300°C (e-h) at various dose levels. (a and e) Before irradiation, (b) 0.32 dpa, (c) 1.62 dpa, (d) 3.24 dpa, (f) 0.31 dpa, (g) 1.54 dpa and (h) 3.08 dpa. The images were captured under underfocused imaging condition, where the bubbles appear as white spots and the 5MPs appear as black spots. Shrinkage of bubbles and 5MPs is observed as a function

of irradiation dose.

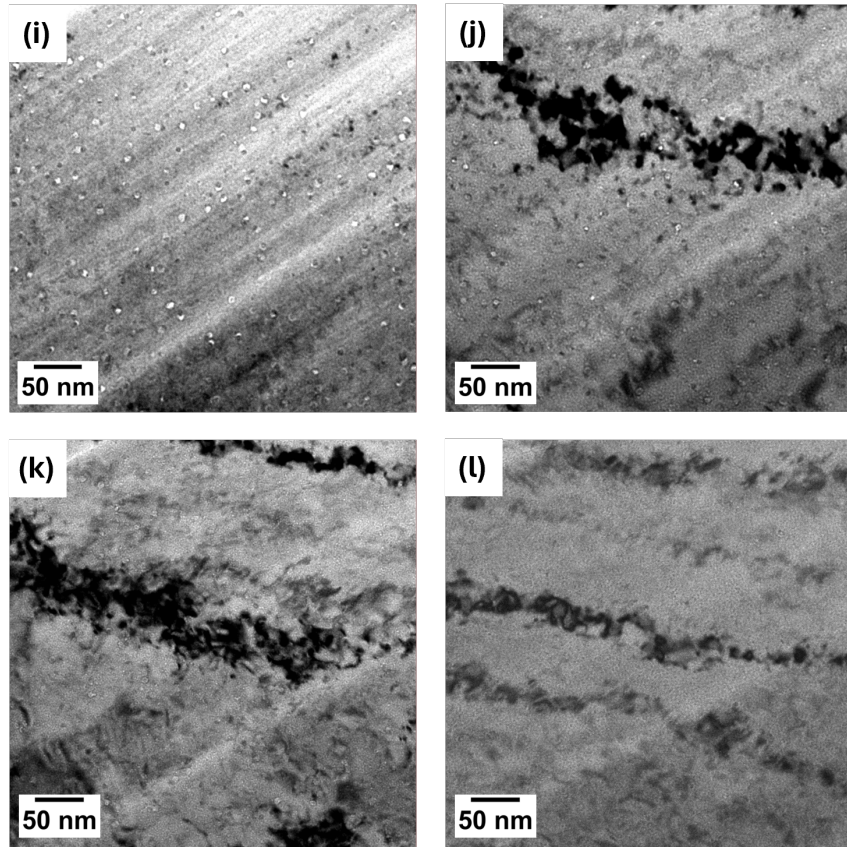


FIG. 4 (cont.): TEM images of the sample irradiated by 1 MeV Kr at 500°C (i-l) at various dose levels. (i) Before irradiation, (j) 0.35 dpa, (k) 1.74 dpa and (l) 3.48 dpa.

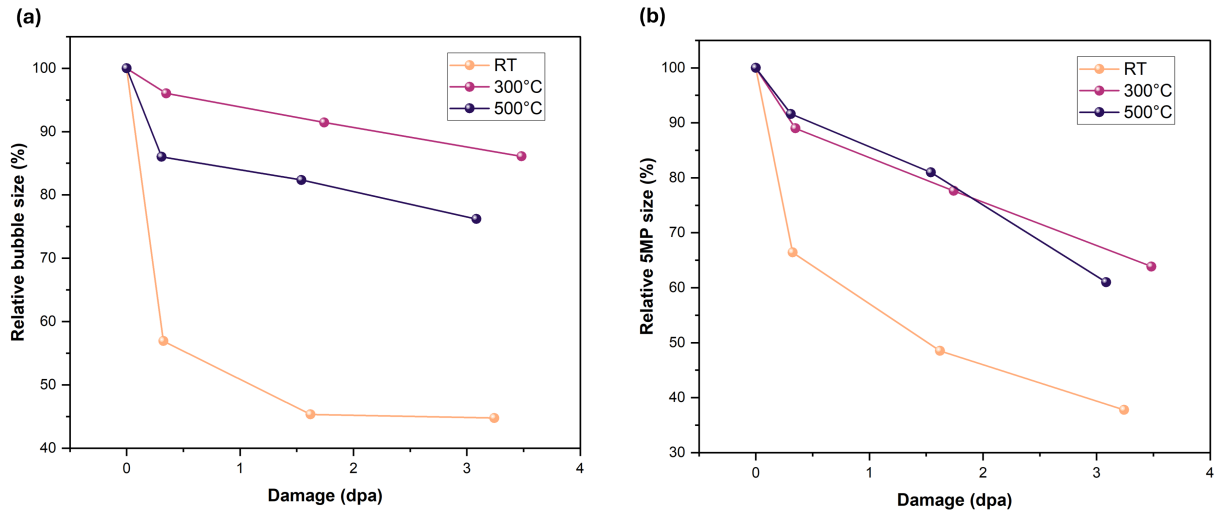


FIG. 5: Relative size of the (a) bubbles and (b) 5MPs as a function of dose at RT, 300°C and 500°C under 1 MeV Kr irradiation. Here, the size of bubbles and 5MPs at 0 dpa is 100% of the initial value and the remaining values decrease in percentage with respect to dose, indicating shrinkage of the concerned fission product phases.

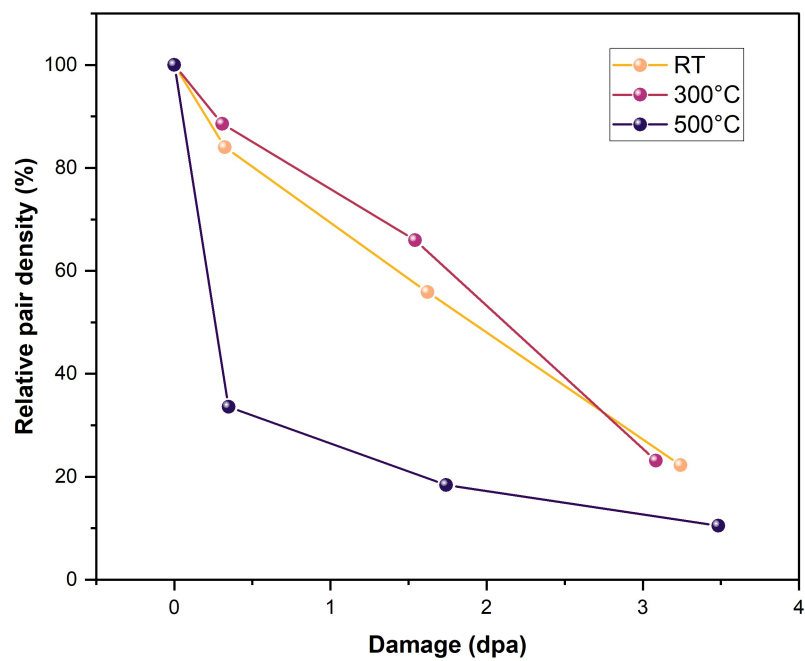


FIG. 6: Relative pair density as a function of dose at RT, 300°C and 500°C under 1 MeV Kr irradiation. Here, the pair density at 0 dpa is 100% of the initial value and the remaining values decrease in percentage with respect to dose, indicating depletion of the pairs.

The microstructural evolution of bubbles and 5MPs under 300 *keV* Xe irradiation at RT, 300°C and -223°C are shown in Fig. 7 and Fig. 8. Fig. 7(a-d) and Fig. 7(e-h) depict the trend similar to those observed in Fig. 4 with a noticeable reduction in the size of bubbles and 5MPs in response to continued exposure to radiation. At -223°C, one sample was irradiated under a high ion flux of 4.13×10^{12} *ions/cm².s* (Fig. 8(a-e)), while another one was irradiated under a low ion flux of 1.25×10^{12} *ions/cm².s* (Fig. 8(f-j)). High ion flux resulted in a more rapid reduction rate in size and pair density of the fission product phases. At high irradiation dose, pairs were scarcely present and small Xe bubbles (less than 1 *nm*) appeared due to ion implantation, as shown in Fig. 8e.

The change in the size of the fission gas bubbles and 5MPs as a function of irradiation dose and ion flux under 300 *keV* Xe irradiation is summarized in Fig. 9 and Fig. 10. Fig. 9(a-b) depict a gradual decrease of the size of bubbles and 5MPs at RT and 300°C. Similarly, the size of the bubbles and 5MPs under irradiation at -223°C also decreased with increased irradiation dose, but more remarkably at high flux compared to low flux when the ion beam was first activated (Fig. 10(a-b)). Higher ion flux resulted in a higher size reduction rate for both bubbles and 5MPs. Fig. 11 shows the change in pair density as a function of irradiation dose for 300 *keV* Xe irradiation. An abrupt reduction in pair density at early stages of irradiation was visible at high ion flux at -223°C and 300°C. Subsequently, the specimens exhibited a gradual decrease in number density as a function of irradiation dose.

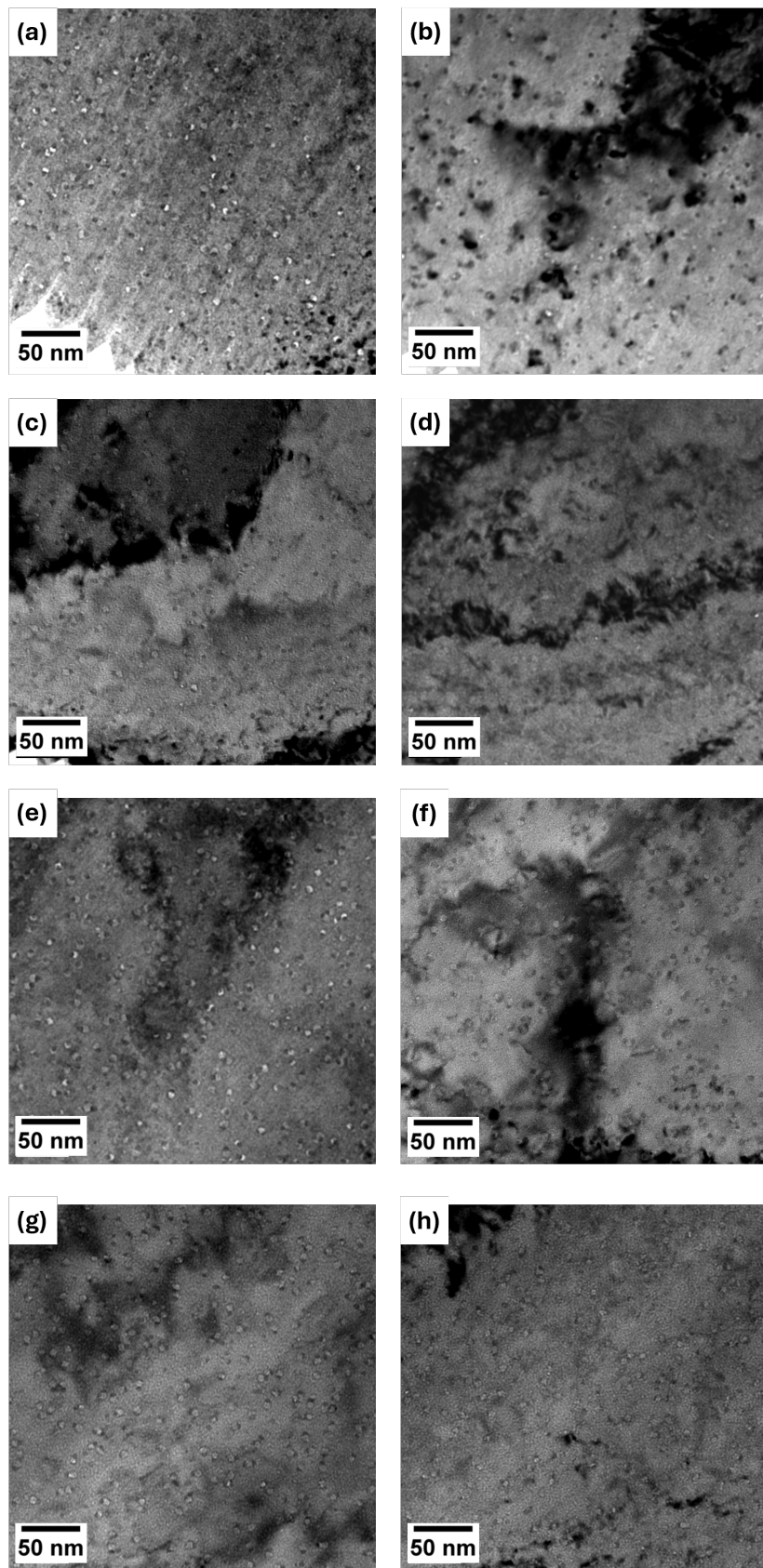


FIG. 7: TEM images of the sample irradiated by 300 *keV* Xe at RT (a-d) and 300°C (e-h) at various dose levels. (a and e) Before irradiation, (b) 1.47 *dpa*, (c) 2.95 *dpa*, (d) 5.89 *dpa*, (f) 1.61 *dpa*, (g) 3.22 *dpa* and (h) 6.44 *dpa*. The images were captured under underfocused imaging condition, where the bubbles appear as white spots and the 5MPs appear as black spots. Shrinkage of bubbles and 5MPs is observed as a function of irradiation dose.

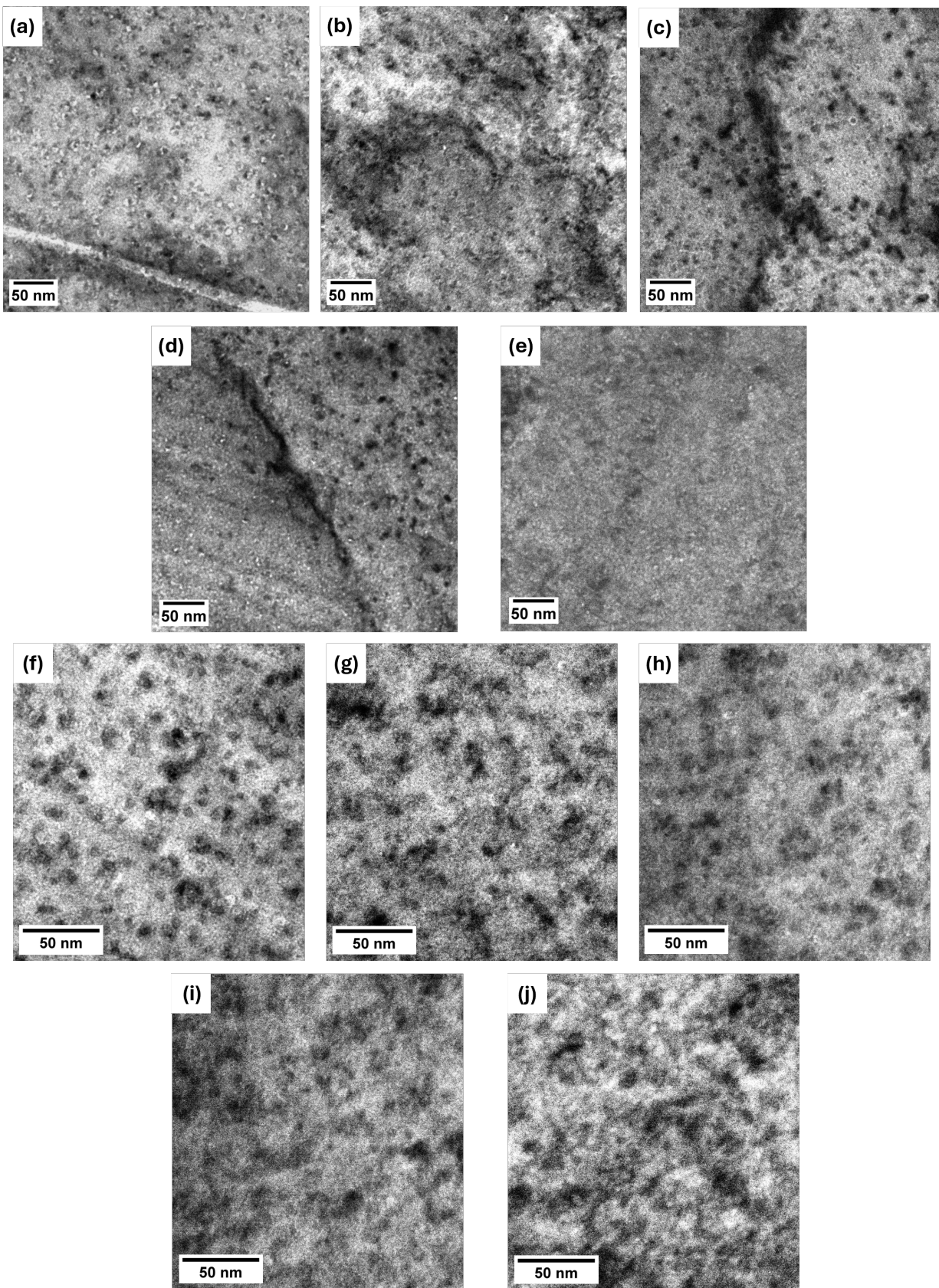


FIG. 8: TEM images of the sample irradiated by 300 *keV* Xe at -223°C (a-e) high flux and (f-j) low flux at various dose levels. (a and f) Before irradiation, (b) 0.34 *dpa*, (c) 1.71 *dpa*, (d) 3.43 *dpa*, (e) 17.16 *dpa*, (g) 0.23 *dpa*, (h) 1.17 *dpa*, (i) 2.34 *dpa* and (j) 11.72 *dpa*. The images were captured under underfocused imaging condition, where the bubbles appear as white spots and the 5MPs appear as black spots. The sample observed in (f) - (j) was relatively thicker than that of a standard TEM lamella, resulting in a more nebulous observation of the concerned fission

product phases.

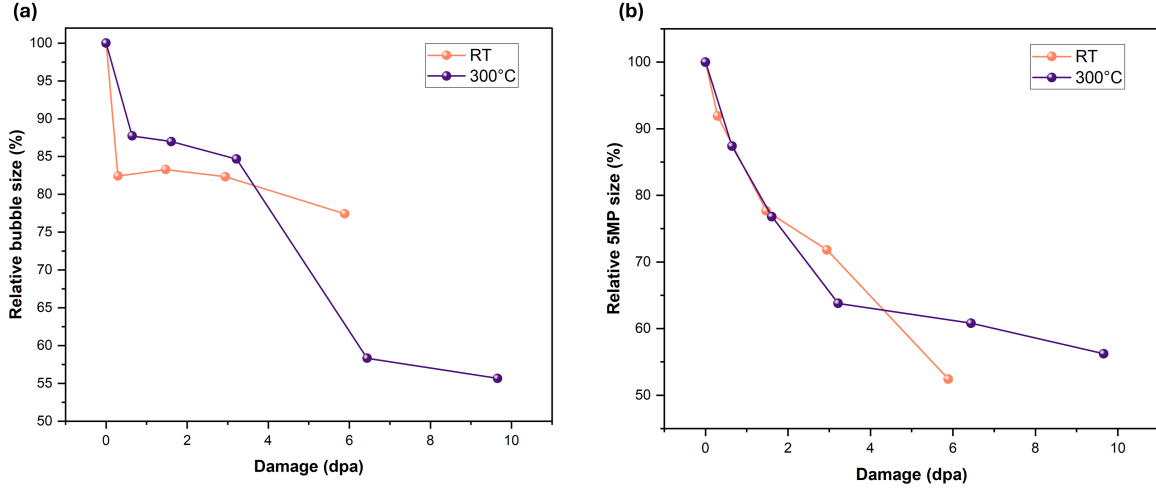


FIG. 9: Relative size of the (a) bubbles and (b) 5MPs as a function of dose at RT and 300°C under 300 keV Xe irradiation. Both temperatures show similar trends in shrinkage of the bubbles and 5MPs.

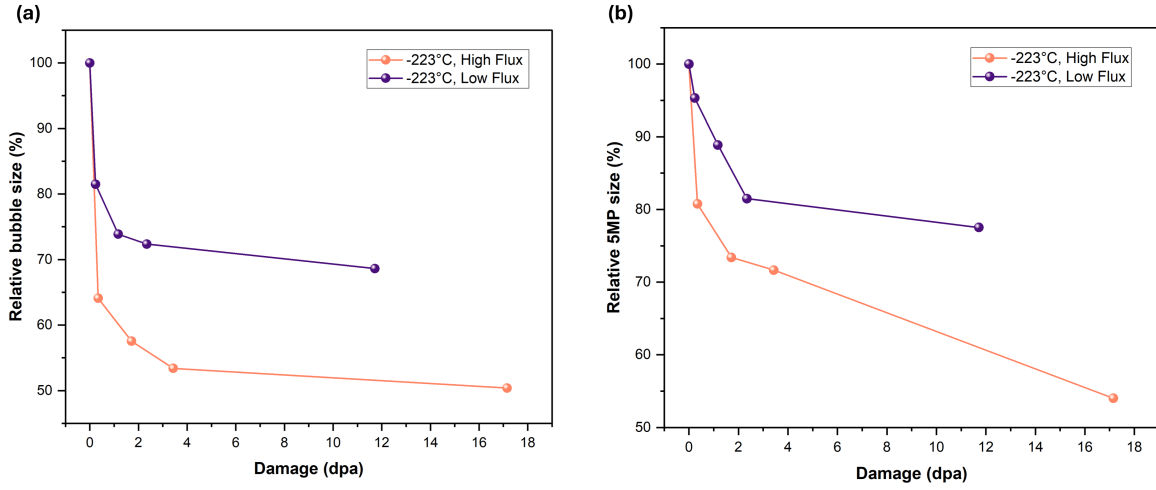


FIG. 10: Relative size of the (a) bubbles and (b) 5MPs as a function of dose at -223°C under 300 keV Xe irradiation at two different ion fluxes. Higher ion flux induced higher shrinkage rate for both fission product phases.

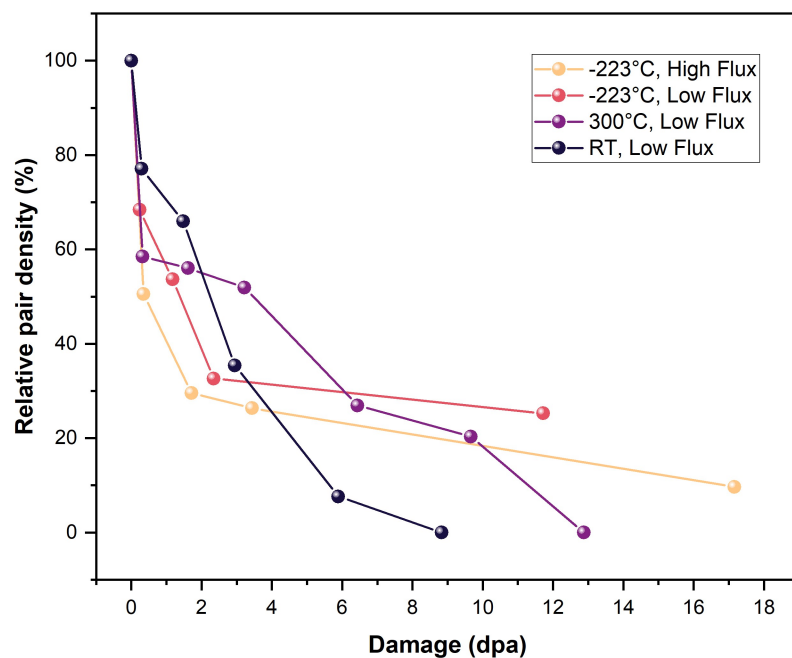


FIG. 11: Relative pair density as a function of dose at RT, 300°C and -223°C under 300 keV Xe irradiation. Pair density reduction at the first dose level is more pronounced for -223°C, high flux and 300°C, low flux irradiation conditions.

The EDS mapping of the ion irradiated sample under 1 *MeV* Kr at 300°C was compared to that of the pre-irradiated sample (Fig. 12). Before ion irradiation, the fission gas bubbles (Xe map) and 5MPs (Mo, Rh, Tc, Pd, Ru maps) were uniformly distributed throughout the sample (Fig. 12a). Post-irradiation, there was a noticeable reduction in their sizes and number of pairs (Fig. 12b). Furthermore, the relatively weaker signals of the fission products compared to Xe suggest a faster diffusion rate of the fission products under ion irradiation. The Mo signal appears relatively weaker than that of the other elements in the 5MPs shown in both Fig. 12a and Fig. 12b. This is likely because the TEM lamellae were mounted on Mo grids for high-temperature irradiation, and the strong background signal from the Mo grids obscures the Mo signal originating from the 5MPs.

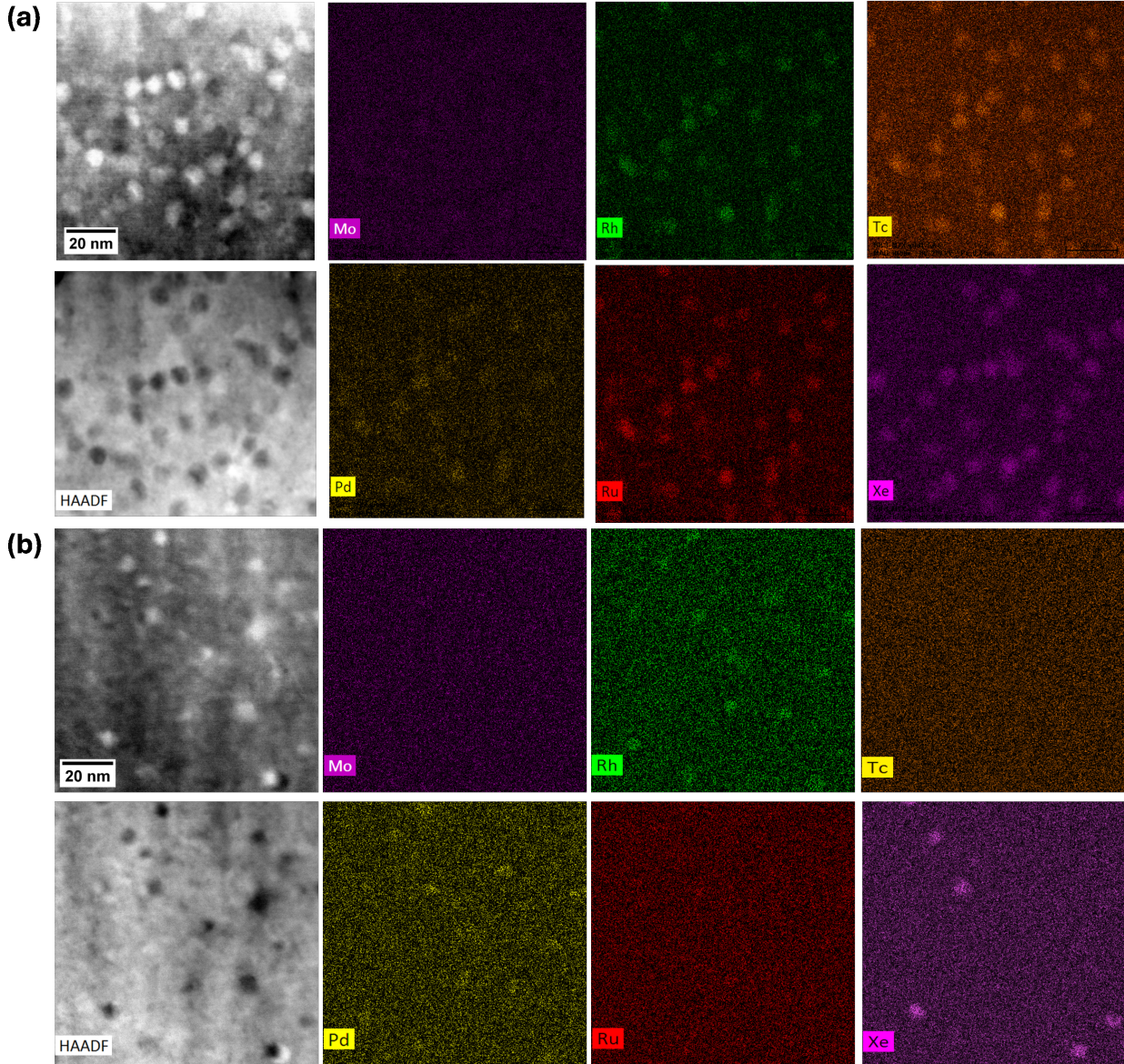


FIG. 12: EDS mappings of the BR-3 fuel sample (a) before and (b) after 1 *MeV* Kr irradiation at 300°C. The signals of 5MPs are relatively weaker compared to Xe, likely due to the faster diffusion of 5MPs under ion irradiation.

IV. Discussion

This study investigates the morphological transformation of fission gas bubbles and 5MPs in spent UO_2 fuel under various ion irradiation doses, temperatures and flux conditions. Under the irradiation conditions described in Table I, ion irradiation has induced a noticeable reduction in the size of both bubbles and 5MPs, along with a depletion in

pair number density. The possible mechanisms responsible for these effects will be discussed here.

Fission gases, such as Xe and Kr, produced because of nuclear fission, are typically trapped in bubbles distributed throughout the UO_2 fuel matrix. This trapping is not permanent, as the gas can return to the atomic solution through interactions with fission fragments or fast neutrons [42]. This process is termed as re-solution of the fission gas atoms from the bubbles in the fuel matrix. Nelson investigated how radiation affected the formation of gas bubbles in reactor fuels. Based on this study, he proposed that radiation damage of ceramic fuels caused by fission fragments might result in kinetic solubility of fission gas atoms [43]. He termed it as homogeneous re-solution process where individual fission gas atoms are ejected from the fission gas bubbles through singular binary collision recoil events, leading to a gradual reduction in size of the bubbles. The precipitate dissolution mechanism, discussed by the wellknown Nelson, Hudson and Mazey (NHM) model points to a similar path [44]. According to this model, the atoms within the precipitates return to the matrix and are uniformly distributed within it due to the dynamic collision events that arise from atomic displacement inside collision cascades. The model explains the precipitate dissolution process further by adding that uniform distribution of the precipitate atoms within the matrix may create a local supersaturation and hinder the shrinkage of precipitates [44]. This supersaturation may lead to the nucleation of new precipitates in the matrix as precipitate atoms start to backflow into the existing ones, which can be termed as “re-precipitation” [44]. Continuous dissolution of the precipitates is another possibility if they are very widely spaced, resulting in the matrix not reaching the steady-state status and impeding local supersaturation [44].

Although the energy levels of in-reactor fission fragments differ from those of the incident ions used in our ion irradiation experiments, the observed bubble and 5MP shrinkage aligns well with mechanisms proposed in previous studies [43, 44]. The in-situ irradiation and the images obtained at each dose level demonstrate that as the irradiation dose increases, the size of the bubbles and 5MPs decreases. Fig. 10(a-b) show that a higher flux of ion irradiation leads to a greater size reduction rate for the bubbles and 5MPs, which supports the theory of homogeneous re-solution of bubbles. A higher ion flux may result in a more frequent interaction between the bombarding ions and the individual atoms of fission gas bubbles and 5MPs [45], resulting in a higher size reduction rate. As the gas bubbles and 5MPs lose solute atoms and re-organize within the matrix, the pairs may get destroyed which explains the decrease of pair density as a function of irradiation dose. In the case of Xe irradiation, the size reduction of fission gas bubbles and 5MPs (Fig. 9(a-b)) shows a similar trend despite having a difference in irradiation temperature. Even at cryogenic temperature where the thermal effect is minimal on the atomic diffusion of the bubbles and 5MPs, size reduction is clear for both of the concerned fission product phases (Fig. 10(a-b)). These observations suggest that there is a weak dependence of temperature on the size of bubbles and 5MPs, therefore indicating a less temperature dependent, irradiation enhanced gas bubble/5MPs atomic diffusion co-efficient. This remark aligns with one of the outcomes of Baker’s study on fission gas bubble behavior [24] and with Matzke’s work as well where he observed that radiation enhanced diffusion co-efficient was athermal under 1000°C [46]. But there is a temperature dependence observed in the case of Kr ion irradiation. The reduction in size of both bubbles and 5MPs shows a higher decrease rate at RT (Fig. 5). However, the pair density depletes in a higher rate at 500°C (Fig. 6), which is not clearly understood at present. Fig. 13 shows sequential change in the nucleation and growth (re-precipitation) of 5MPs under ion irradiation at -223°C . These are snapshots from an in-situ TEM video recorded under underfocused imaging condition. In this condition, the dark particles in (Fig. 13(b-h)) marked by arrows are most likely 5MPs. The re-precipitation of 5MPs began at an added dose of 2.39 dpa (Fig. 13b) and the re-precipitated features increased in both size and density up to a certain degree under ion irradiation. The re-precipitation and coarsening of these 5MPs supports the NHM model [44]. The in-reactor behavior of the concerned fission product phases may differ from the above observations. Absorption at TEM lamellae surface boundary is one of the effects which is absent during in-reactor irradiation [47]. Also, the broader and continuous radiation exposure in reactors allows for ongoing damage and simultaneous recovery processes such as annealing effects [48]. These may moderate the effects of displacement cascades differently compared to the more intense but localized ion irradiation experiments. The chosen energy levels of ion irradiation (300 keV for Xe and 1 MeV for Kr) do not reflect the energy levels of fission fragments present in in-reactor conditions which may also affect the behaviors of the fission product phases in question.

The processes discussed thus far are summarized in Fig. 14 in the light of existing literature and reported results. Initially, the spent UO_2 microstructure at the rim region is characterized by the presence of pairs of nano sized fission gas bubbles and 5MPs. As the irradiation starts, the implanted ions start to interact with the atoms within Xe/Kr gas bubbles and 5MPs, resulting in their gradual loss of atoms (Fig. 14b). Some of the implanted ions may get trapped by the vacancies and voids within the matrix or, attach to existing pairs or, physically bind with detached Xe/Kr atoms. The size of the bubbles and 5MPs decreases as they lose their constituent atoms. The detached atoms from bubbles and 5MPs experience different consequences. As the bubbles lose Xe/Kr atoms due to irradiation, some of the released Xe/Kr atoms may dissolve into the surrounding matrix, contributing to a reduction in the number of detectable Xe/Kr atoms within the original bubbles. Others may migrate to inter-granular pores or escape through sample surface (Fig. 14c). As the bubbles continue to shrink, the resulting excess vacancies are filled by implanted Xe ions. Kr ions, with higher energy and lower mass, pass through the sample surface and are deposited in lower concentration compared to Xe ions, as shown by the SRIM calculations in Fig. 2. The solutes of 5MPs may distribute uniformly within the matrix until a local supersaturation is reached. At this point, the clusters of 5MPs are

difficult to detect. However, if the distance between them is sufficient, a backflow diffusion of metallic atoms to the existing metallic clusters may occur, leading to re-precipitation of 5MPs (Fig. 14d). If 5MPs are widely spaced, continuous dissolution of the 5MPs due to interaction with the implanted ions is another possibility, preventing local supersaturation. As the dose accumulates, the density of sub-nanometer bubbles containing implanted Xe ions increases.

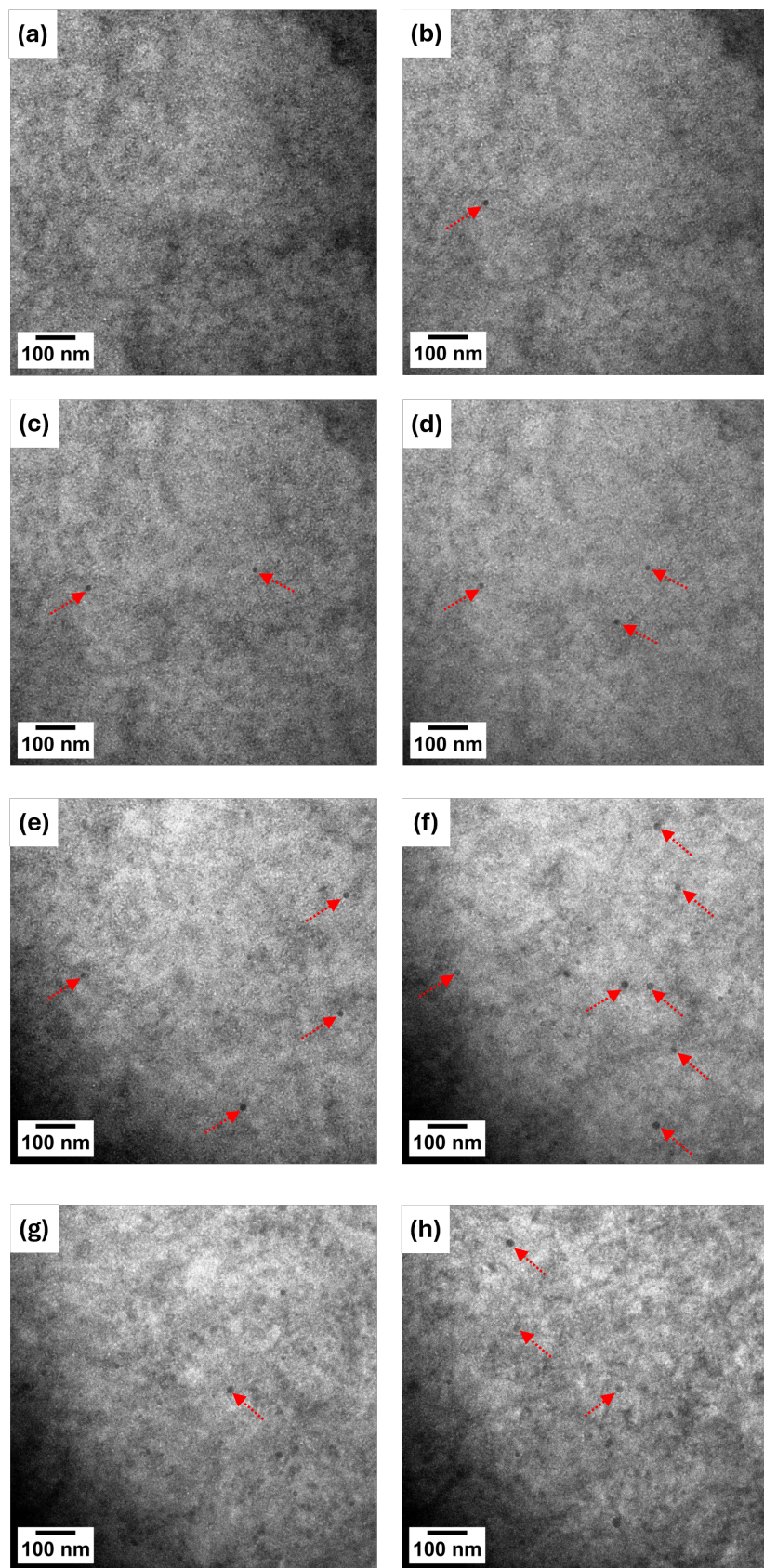


FIG. 13: In-situ TEM images of the sample irradiated to various doses at an ion flux of $1.25 \times 10^{12} \text{ ions/cm}^2 \cdot \text{s}$. These images are snapshots from an in-situ TEM video recorded under underfocused imaging condition where, 5MPs appear as black dots. The arrows mark the probable appearance of re-precipitation. (a) 2.34 dpa, (b) 2.39 dpa, (c) 2.44 dpa, (d) 2.53 dpa, (e) 3.23 dpa, (f) 4.10 dpa, (g) 4.99 dpa and (h) 5.86 dpa.

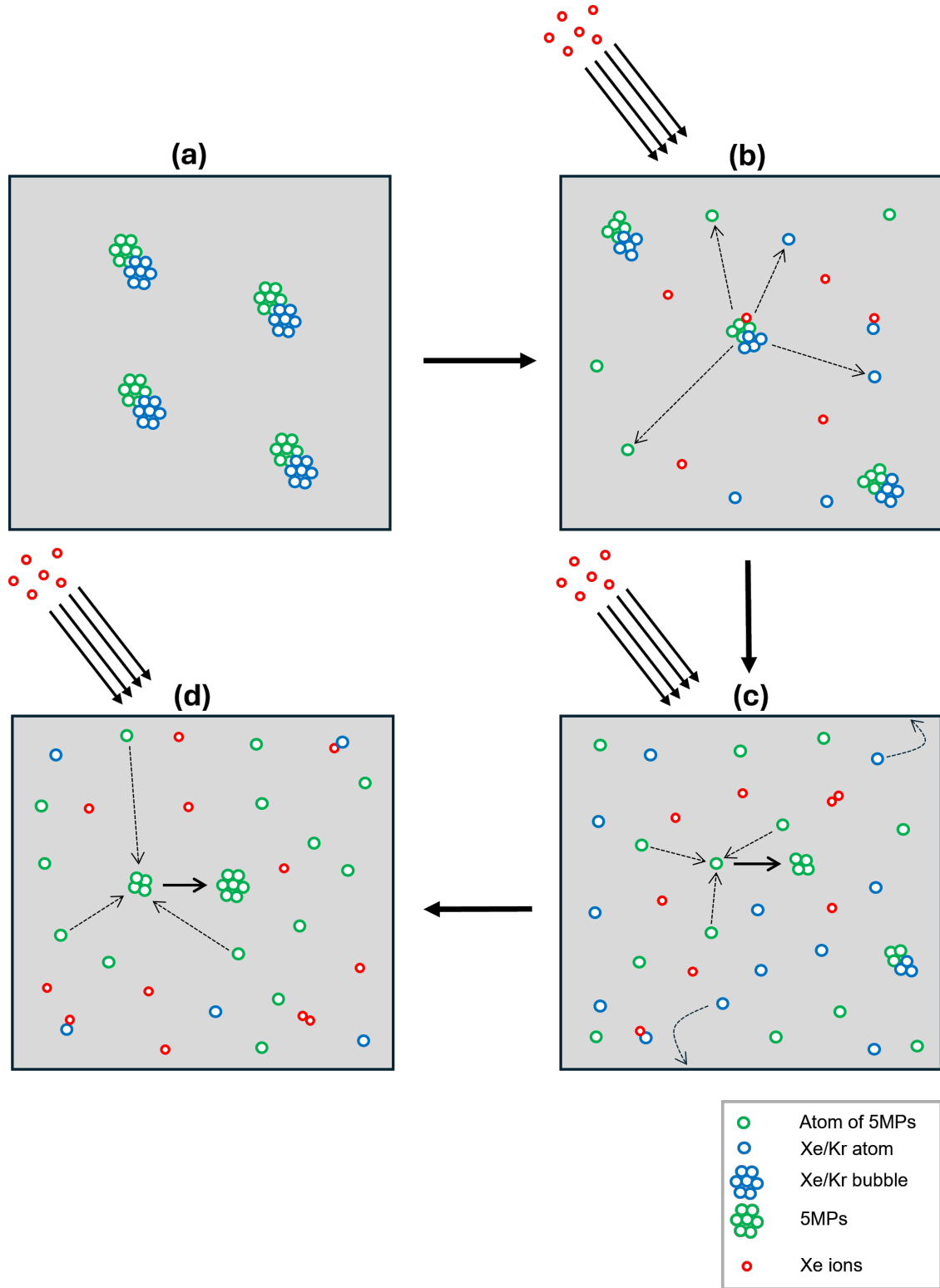


FIG. 14: Illustration of the processes involved in the shrinkage of bubbles and 5MPs and reduction in pair density.

V. Conclusion

In conclusion, this study provides significant insights into the dynamic behavior of fission gas bubbles and 5MPs in spent UO_2 fuel under various ion irradiation conditions. The in-situ TEM analyses revealed a consistent trend of size reduction and decrease in pair density for both the gas bubbles and 5MPs with increasing irradiation dose, with higher ion fluxes inducing more rapid reductions. At elevated doses, the occurrence of Xe/Kr and 5MPs pairs diminished, and sub-nanometer Xe bubbles from ion implantation were detected. Elemental mapping before and after irradiation indicated faster diffusion of 5MPs compared to Xe. These findings support existing models of homogeneous re-resolution and precipitate dissolution under ion irradiation, emphasizing the significant role of energetic collisions in ejecting atoms from fission gas bubbles and 5MPs. Additionally, the study underscores the potential for re-precipitation of 5MPs under certain conditions, consistent with the Nelson, Hudson, and Mazey (NHM) model. The selected irradiation conditions were limited to low energy irradiations and the findings may not reflect the in-reactor behavior of the fission product phases in question. The in-situ TEM video showcased the probable phenomenon of re-precipitation post-dissolution. Finally, these insights are invaluable for refining predictive models and optimizing the performance of nuclear fuels, highlighting the need for further research, particularly on the impact of temperature on atomic diffusion.

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