

The rare gas ion irradiation effects on the critical size of solidification of Xe precipitate

K. Mitsuishi, M. Song and K. Furuya

National Research Institute for Metals, Sakura, Tsukuba 305, Japan
Fax: 81-298-59-5054, e-mail: mitsuishi@nrim.go.jp

The irradiation effects of rare-gas ions toward solid Xe precipitates embedded in Al matrix were observed using dual ion beam interfaced High voltage transmission electron microscopy (HVTEM). The Xe precipitate is made in an Al specimen by ion-irradiation and is successively irradiated by He or Ar ions. The critical size change of Xe precipitate by He irradiation is observed at the dose over 1×10^{17} ion/cm². For Ar irradiation case, critical size of Xe precipitate is not changed until a dose of 2×10^{16} ion/cm². In diffraction patterns, blurred spots among the Xe and Ar position are observed at 2×10^{16} ion/cm² indicating that the solid Ar is grown in the specimen independent from Xe precipitate. This is also confirmed by HRTEM image of a precipitate, which has a moiré fringe of Al and Ar. Although the change in critical size of Xe precipitate is not observed, precipitates having imperfect shapes are often observed above the dose of 5×10^{15} ion/cm². These precipitates are considered as partially Xe precipitates.

Keyword: irradiation, precipitate, critical size, Xe, HRTEM

1. INTRODUCTION

The behavior of nanometer size precipitates created by implantation of inert gases in materials has been studied extensively for more than 35 years because of problems associated with the development of fusion and fission reactors^{1,2)}. By using high-resolution transmission electron microscopy (HRTEM)³⁻¹¹⁾, the shape and topotactical alignment of the precipitates have been investigated. Rare gas Xe forms a nanocrystals having a diameter less than 4nm and is mesotactically aligned with the surrounding matrix when it is implanted at room temperature. The phases of these rare-gas precipitate are important to understand the initial stage of radiation damage relevant to the hardening of material.

This solidification of rare-gas nanocrystals in metal is explained by surface tension of host matrix metal. The rare-gas is pressurized by the pressure which inversely proportional to its radius. Therefore, some critical size exists and below the size, the rare-gas is pressurized high enough to be solidified even at room temperature. Especially in fission reactor, various kinds of rare-gases are produced in the structure materials or fuel elements, therefore the mixing of rare-gases in materials is needed to be taken into account. Depending on a solubility of rare-gas toward other rare-gases, the critical size may change or in some case, partial solidification of precipitate is expected to be observed. Singleton and Halsey¹²⁾ showed that Ar and Xe were insoluble in the solid phase at 78K and the solubility of solid Xe in liquid Ar is studied by Yunker and Halsey¹³⁾ and reported that the solubility is less than 0.5% with no detectable temperature dependence. However, there is no work on solubility of Xe and Ar at room temperature, and at a high-pressure region. For the case of He-Xe system, no work has been done even at low temperature region. In this paper, we use the dual ion beam interfaced High

Voltage Transmission Electron Microscope (HVTEM) to observe the effect of mixing of rare-gases by means of successive irradiation of Ar or He gases toward the solid Xe precipitate in Al matrix.

2. EXPERIMENTAL

The Al specimens were cut from 5-9's starting material and electronically polished using an electrolyte of HNO₃: CH₃OH= 1:2 at 253 K and implanted with 50 keV Xe at room temperature to a dose of 1×10^{16} ion/cm² by the HVTEM. Specimens were then annealed at 725 K for 0.5 h in a vacuum to remove radiation damage in the Al matrix and to consolidate the Xe within the precipitates. Templier et al.¹⁴⁾ have reported that such annealing results in sharper precipitate image and more intense electron diffraction from the solid Xe in Al.

The He irradiation is performed at 4keV at room temperature to a dose of 1×10^{17} ion/cm² and Ar irradiation is made at 25keV at room temperature to a dose of 2×10^{16} ion/cm². The specimens were implanted at a flux of 2.1×10^{13} ions/cm²s, HRTEM observations were performed with a JEM ARM-1000 high-voltage TEM, operated at a voltage of 1 MV with a spatial resolution of 0.13nm, with the incident electron beam along a $\langle 110 \rangle_{Al}$ direction. All observations were performed at room temperature. The electron beam current density was 1.2×10^5 A/m². Displacement per atom per second for Al at this electron flux is $3 \times 10^{-3.5}$.

3. RESULT AND DISCUSSION

He irradiation

He ion was irradiated up to 1×10^{17} ion/cm². Figure 1 shows HRTEM images of Xe precipitates in Al with the

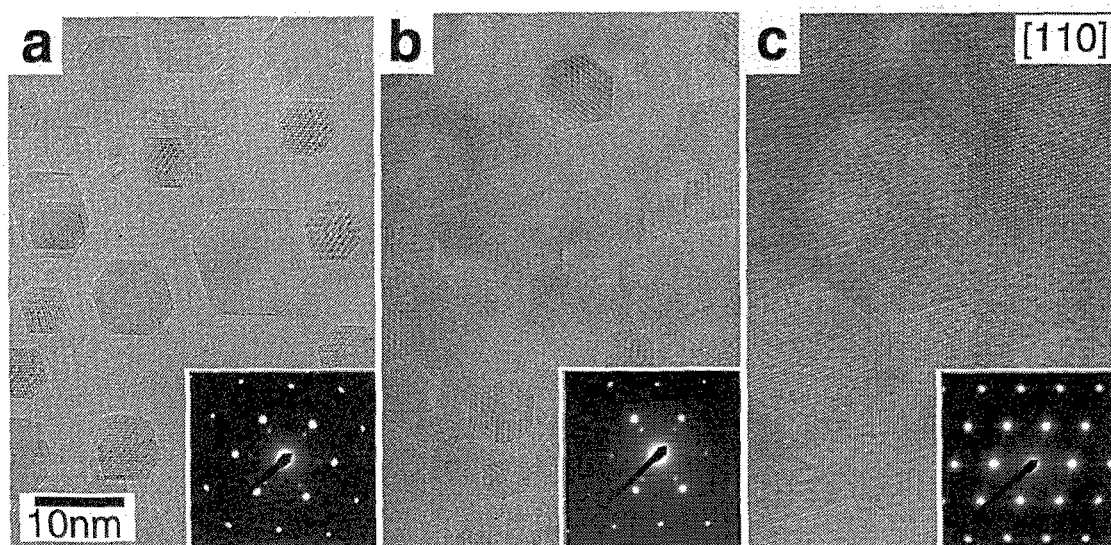


Fig. 1. HRTEM image with corresponding selected area diffraction pattern; (a) before irradiation, and after (b) 8×10^{16} ion/cm² and (c) 1×10^{17} ion/cm² of He irradiation.

selected area diffraction pattern up to a dose of 1×10^{17} ion/cm². The incident electron beam is parallel to $\langle 110 \rangle$. To extract the Xe precipitate image from that of the Al matrix, a specific off-Bragg condition¹⁵⁾ was used choosing the optimum illumination angle and defocus value. In fig. 1 (a), the maximum size of solid Xe precipitate before He irradiation was approximately 10nm. In the selected area diffraction pattern, the Xe spots located at 2/3 of Al spots was clearly observed meaning many Xe precipitates were in the solid phase as observed by HRTEM image. At the dose of to 1×10^{16} ion/cm², no change of the critical size was observed although the image was getting worse because of the radiation damage. Even at to 8×10^{16} ion/cm² the maximum size of solid Xe precipitate did not change. At this dose, the bright field image was also taken as shown in fig.2. Many of small white contrast besides the Xe precipitates were observed in this figure meaning that the He indeed was implanted

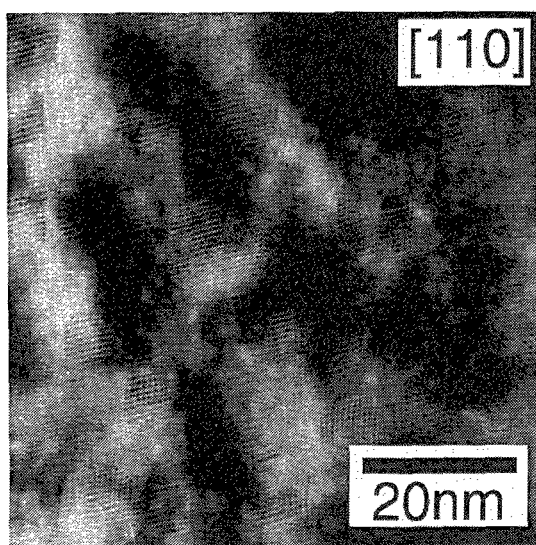


Fig. 2. Bright field image after 8×10^{16} ion/cm² He irradiation.

in the specimen. The situation drastically changed when the dose reached to 1×10^{17} ion/cm² (fig.1 (c)). At this dose, no solid Xe precipitate was observed although the faceted shapes of precipitates in various sizes were still observed. The fringes seen in the precipitate has a width of Al [210] lattice fringes. Some of the precipitates were seen as small as 5nm. This is the size of the precipitate, which was observed in solid phase in fig.1 (a). Therefore this change is not due to the growth of bubbles beyond the critical size, but the change of the critical size itself by the He irradiation. This change was also seen in the diffraction pattern where no Xe spots are observed anymore.

Ar irradiation

Fig. 3 is a HRTEM image of Xe precipitates in Al with the selected area diffraction pattern. The incident electron beam is parallel to $\langle 110 \rangle$. In fig. 3 (a), the maximum size of solid Xe precipitate before He irradiation was 11nm which was almost same as in fig.1 (a) and the Xe spots were seen in the diffraction pattern. For Ar irradiation, although no significant change of the critical size was not observed same as the He case, up to the dose of to 1×10^{16} ion/cm², the number of solid Xe precipitate was decreased not because the change of the critical size, but because the growth of precipitate. The image was getting worse again because of the radiation damage. In the diffraction patterns, however, blurred spots among the Xe and Ar position were observed at to 2×10^{16} ion/cm². This means that Ar ions indeed were implanted in the specimen and solid Ar was also grown in the specimen independent from Xe precipitate the same manner as the He irradiation case. In fig. 4, a precipitate, which has a moiré fringe of Ar and Al are shown at a dose of 2×10^{16} ion/cm². Off Bragg condition for solid Ar precipitate is much harder compare to Xe precipitate case because of closer lattice constant and scattering factor toward Al. Therefore here in the image, shape of the precipitate and the individual atom image of Ar is not clear compare to Xe case.

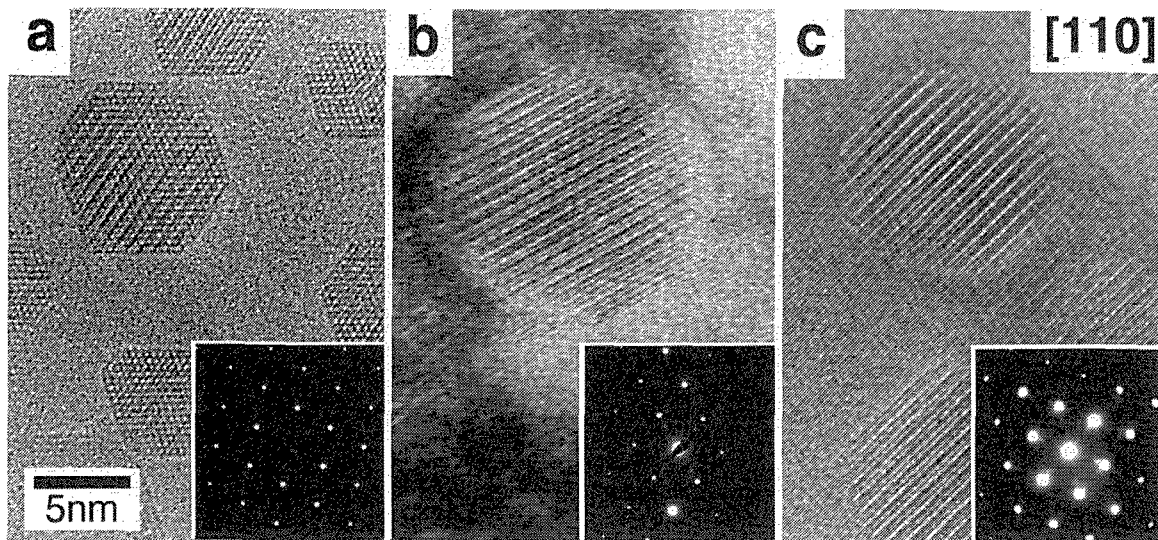


Fig. 3. HRTEM image with corresponding selected area diffraction pattern; (a) before irradiation, and after (b) 1×10^{16} ion/cm² and (c) 2×10^{16} ion/cm² of Ar irradiation.

Although the no significant changes are observed in critical size of Xe precipitate, precipitates having imperfect shapes were often observed above the dose of to 5×10^{15} ion/cm². In fig. 5, examples of these precipitate are shown in through focus image. In any of these defocus values, no overlapping of other precipitates or defects are not visible. This can be considered as a partially solid precipitate that consists of solid Xe part and liquid Ar part. Since solubility of Xe and Ar is low, the Xe may solidify separately from Ar. Since the required pressure of solidification is much higher than Xe, partially solid is expected to be observed. Further study is required to determine the nature of this precipitate.

4. CONCLUSION

The irradiation effects of rare-gas ions toward solid Xe

precipitates embedded in Al matrix were observed using high-resolution transmission electron microscopy (HRTEM). Al TEM thinned specimens was implanted with 50keV Xe at room temperature to a dose of 1×10^{16} ion/cm² and successively implanted with He or Ar ions of 4 and 25keV, to a dose of 1×10^{17} and 2×10^{16} ion/cm², respectively.

By He irradiation toward solid Xe precipitate, no significant change in precipitate shape or the critical size was observed up to the dose of 8×10^{16} ion/cm². Nevertheless, all of the precipitates were observed in liquid phase when the dose over 1×10^{17} ion/cm². At this dose, there were some small precipitates remains and were observed in liquid phase, indicating that the liquefaction occurs not because the growth of the precipitates but because the change of the critical size.

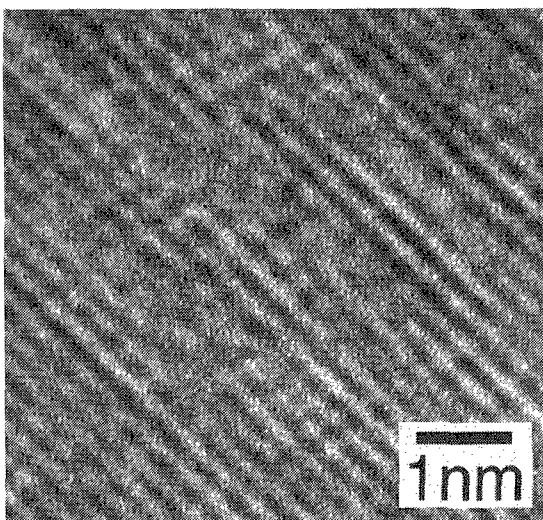


Fig. 4. HRTEM image after 2×10^{16} ion/cm² of Ar irradiation. A moiré fringe of Al and Ar is seen in the image.

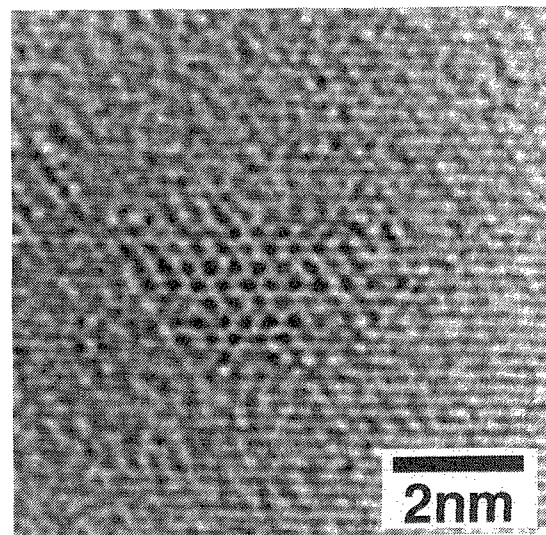


Fig. 5. HRTEM image of imperfect Xe precipitate

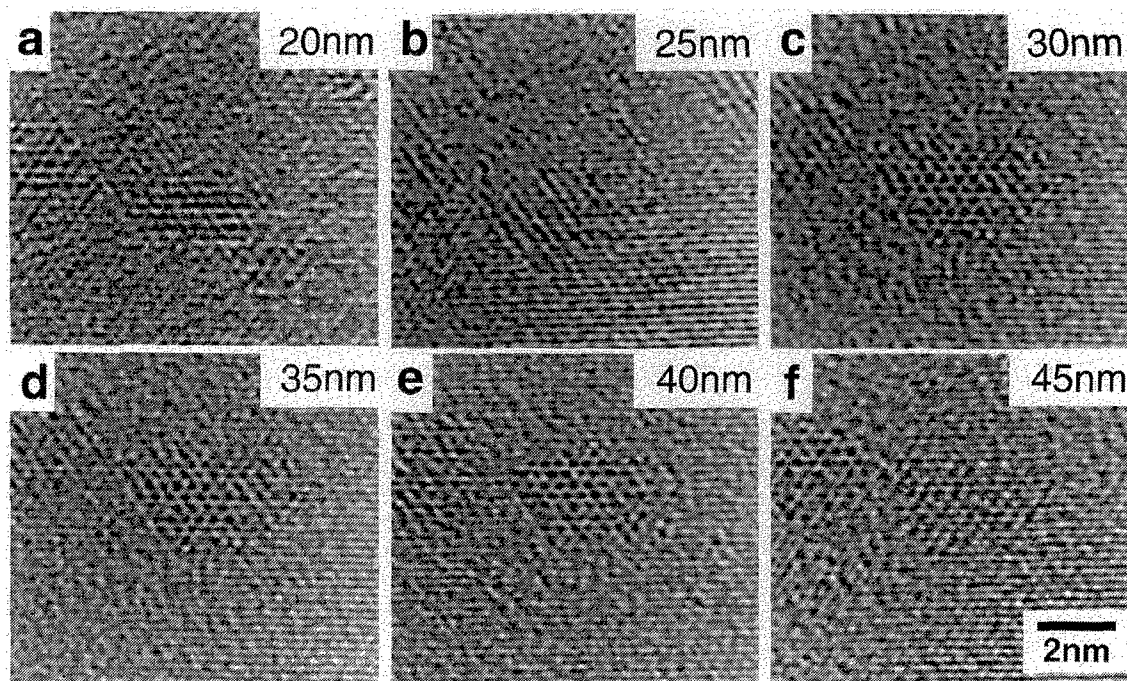


Fig.6 Through focus image of imperfect Xe precipitate observed at 2×10^{16} ion/cm²; (a) 20nm, (b) 25nm, (c) 30nm, (d) 35nm, (e) 40nm, (f) 45nm.

For Ar irradiation case, critical size of Xe precipitate is not changed until a dose of 2×10^{16} ion/cm². In diffraction patterns, blurred spots among the Xe and Ar position are observed at 2×10^{16} ion/cm² indicating that the solid Ar is also grown in the specimen independent from Xe precipitate as same manner in He case. This is also confirmed by HRTEM image of a precipitate, which has a moiré fringe of Al and Ar. Although the no changes are observed in critical size of Xe precipitate, precipitates having imperfect shapes are often observed above the dose of 5×10^{15} ion/cm². These precipitates are considered as partially Xe precipitates. Further study is required to determine the nature of this precipitate.

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