7.3 Multiscale Simulation of Transmission Electron Microscopy Imaging of Helium Bubbles in Iron — B. Yao, D. J. Edwards, and R. J. Kurtz (Pacific Northwest National Laboratory), G. R. Odette and T. Yamamoto (University of California Santa Barbara)

OBJECTIVE

The objective of this task is to establish the size correlation between transmission electron microscopy (TEM) imaged helium (He) bubbles and the actual bubbles in an iron (Fe) matrix.

SUMMARY

The results of this simulation study show that the size of TEM imaged He bubbles, represented by the inner diameter of the first dark Fresnel ring under defocused condition (D_{in}), deviated from the actual bubble size (D_o). D_{in} was found to be larger than D_o when imaged with a highly incoherent electron beam, but smaller than D_o if the beam is coherent. The deviation of D_{in} from D_o increases with increasing defocus. On the other hand, the electron beam accelerating voltage, bubble size, bubble position, and TEM sample thickness do not significantly affect the value of D_0/D_o. This study also suggests that He bubbles can be differentiated from argon (Ar) bubbles by differences in Fresnel contrast.

PROGRESS AND STATUS

Introduction

Formation of nanoscale He bubbles in reduced activation ferritic/martensitic steels may lead to degradation of mechanical properties of materials [1-3]. One strategy to control the fate of He and its effect on mechanical properties of materials is through limiting the size of He bubbles with designed material composition and structure [4]. Based on established theories [4-7], a critical radius exists for the transition of He bubbles to voids. Below a critical size He bubbles are stable, while above the critical size, He bubbles transition to unstably growing voids. TEM has been commonly used for the characterization of He bubbles. Due to their small size (typically less than 5 nanometers in diameter), a defocus of 0.5 μm to 1μm is required to reveal the Fresnel rings of the bubbles. The size of Fresnel ring can be utilized to represent the bubble size. In order to accurately quantify the bubble size, one question arises: what is the relationship between the size of Fresnel rings formed during under-focused imaging and that of the actual He bubbles?

Over 40 years ago, pioneering work was conducted by Rühle and Wilkens [8-10] and Foreman et al. [11] to establish this relationship. Their approach is based on multi-beam Bloch wave calculations using column approximations. Their results indicate that the inner diameter of the first dark Fresnel ring (D_{in}) in an under-focused condition is slightly larger than the actual cavity diameter (D_0) [8]. Their calculations were based on several important assumptions: 1) the foil was relatively thick (e.g., $t_{min} \approx 6\xi_g$, where ξ_g is the extinction length); 2) the cavity was not too close to either of the foil surfaces; and 3) the microscope resolution was not better than ~ 7Å [8]. Obviously, with the improvements in microscopes and the ability to image smaller features, some of these assumptions may not be valid today.

This paper presents our study of multislice simulation of TEM imaging of He bubbles in Fe. It is known that the multislice simulation is more suitable to deal with complicated situations than

Bloch wave calculations [12]. The multislice simulation through Fast Fourier Transformation (FFT) is able to include millions of electron beams, and no special assumptions are required. Consequently, it may provide simulation results closer to the actual situation. The main objective of this study is to re-examine the size correlation between imaged Fresnel rings and the actual bubbles. Effects of a variety of imaging and specimen conditions were examined.

Methods

The program developed by Kirkland [12] was chosen for the multislice simulation of TEM images in this study. Large super-lattices containing a He bubble inside were generated according to the required interface of simulation software. Specimen variables explored in this study include sample thickness, bubble size, and bubble position along the thickness direction. Variations in imaging conditions include electron-beam incoherency, accelerating voltage (keV), and defocus. In order to accurately quantify the size of Fresnel rings from simulated images with the same standard, a post-processing program was developed to generate an average-intensity profile along the bubble radius direction. A representative simulated bubble image and its intensity profile are shown in Fig. 1(a) and Fig. 1(b), respectively. The X-axis in Fig. 1(b) indicates the distance from the bubble center with a step of one pixel, and Y-axis corresponds to an average intensity of all pixels on an annular ring. This formal analytical procedure is introduced because human vision is not sensitive to slight variations in image intensity. Through this approach, the uncertainty of determining Fresnel ring size in simulated TEM images is greatly reduced.

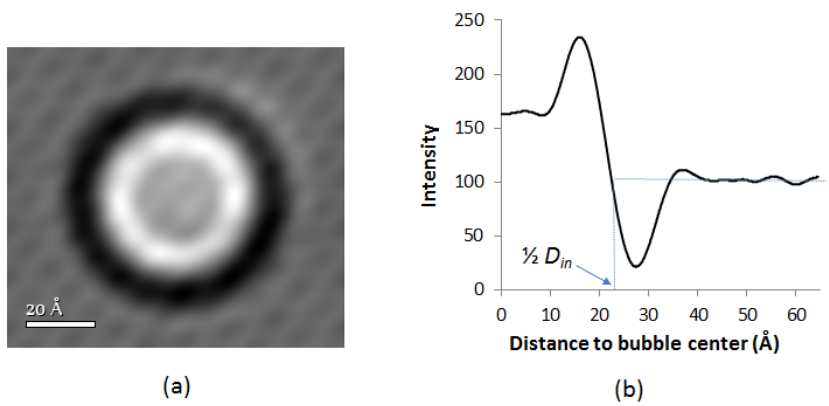

Fig. 1. (a) A representative simulated TEM image of a 5nm-He bubble with V = 200 keV, Δf = -500 nm, C_s= 1mm, and a deviation of illumination semi-angle of 0.5 mrad; (b) the corresponding average-intensity profile of the image.

Results and Discussion

Electron-beam incoherency was found to have a significant influence on the deviation of imaged bubble size from the actual one, as demonstrated by Fig. 2. The term D_{in} in the figure indicates the inner diameter of the first dark Fresnel ring in the under-focused condition, and D_0 is the actual bubble size in diameter (5 nm for the present simulations). D_{in}/D_0 represents the deviation of imaged Fresnel ring from the actual bubble size. The uncertainty of the data comes from the measurement of the size of Fresnel rings, as illustrated in Fig. 1. The coherency considered here is limited to spatial coherency. A small deviation in defocus and illumination angle was introduced to include the effects of partial coherence [12]. A small spread of defocus

(e.g., 50 Å) was found to have no detectable influence on the value of D_{in}/D_0 (results not shown). However, the effect of a deviation of illumination semi-angle ($\Delta\alpha$) is much stronger, as clearly revealed by Fig. 2. Other related parameters for the simulations include an accelerating voltage of 200 keV, a C_s of 1 mm, a super-lattice containing a 5 nm He bubble located in the center of TEM foil, a total foil thickness of 52 nm, and a defocus of 500 nm.

Fig. 2 is helpful to understand differences between results obtained in this simulation study and the previous calculations by Rühle and Wilkens [8]. Both studies confirm that the actual bubble size is close to the inner diameter of the first dark Fresnel ring. However, the Bloch wave calculations [8] predicted a value of D_{in}/D_0 greater than 1, while our simulations using coherent electron beams gave values of D_{in} less than D_0 (as will be shown later). This difference is most likely related to the incoherency of electron beams. The assumption [8] that the microscope resolution is not better than ~ 7 Å may be valid for microscopes equipped with tungsten or LaB_6 filaments, but it certainly is not applicable to FEG-TEMs.

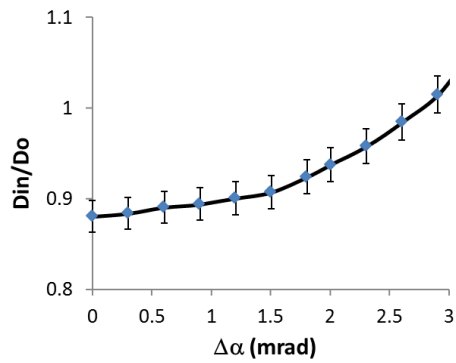

Fig. 2. Effect of D_{in}/D_0 as a function of the deviation of illumination semi-angle ($\Delta\alpha$).

Fig. 3 presents the effects of defocus and accelerating voltage of electron beam on D_{in}/D_0. The simulated samples have a thickness of 52 nm, and the bubble is located in the center of the membrane. A small beam-incoherency ($\Delta\alpha$ = 0.3 mard) was selected for the simulations. It is clearly shown in the figure that the ratio of D_{in}/D_0 generally decreases with increasing defocus. The electron beam accelerating voltage was found to have a very small influence on the change of imaged bubble size. The imaged He bubbles at a defocus of 1μm are about 85% of the actual bubble size.

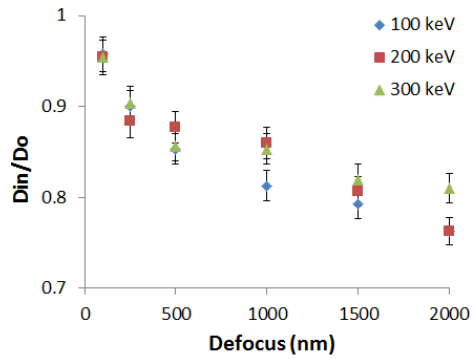

Fig. 3. A trend of D_{in}/D_0 at different electron-beam accelerating voltages as a function of defocus.

Effects of other factors, including bubble size, bubble position in the depth of membrane, and TEM sample thickness were also examined. Bubble size and position in the TEM sample thickness were found to have a negligible effect on D_{in}/D_0. The sample thickness may have a small effect. For samples thinner than 35 nm, D_{in}/D_0 slightly increases with increasing thickness, while above that, D_{in}/D_0 generally remains constant. As effects from these factors are not significant, corresponding simulation results are not presented in this report.

The ion-milling induced artifacts of Ar bubbles may cause an overestimation of He bubble density, especially in small-size range (e.g., ≤ 1 nm), or limit the TEM resolution for the examination of small He bubbles. As the atomic potential of Ar is much higher than that of He [12], it might be possible to distinguish these two types of bubbles by their image contrast. For this purpose, TEM images of a He bubble, an Ar bubble and a void with the same size (3 nm) and imaging conditions (200 eV, 1mm of C_s, 26 nm in thickness, 0.3 mrad of $\Delta\alpha$, and a defocus of 500 nm) were simulated. The results are shown in Fig. 4. Under the selected imaging conditions, the void and the He bubble give the same contrast, which suggests that the He/Vacancy ratio and standoff distance between He atoms and the Fe matrix is not a concern in TEM imaging simulations. On the other hand, the Ar bubble has a much weaker contrast under the same imaging condition. The results can be understood as Ar atoms (with an atomic number of 18) scatter the electron beam more effectively than He atoms (with an atomic number of 2). The simulation results suggest that it is possible to differentiate He bubbles from Ar bubbles during TEM examination. However, the simulation results also revealed that a larger electron-beam incoherency and defocus may greatly change the relative contrast of He and Ar bubbles. More quantitative studies are required for a more comprehensive understanding.

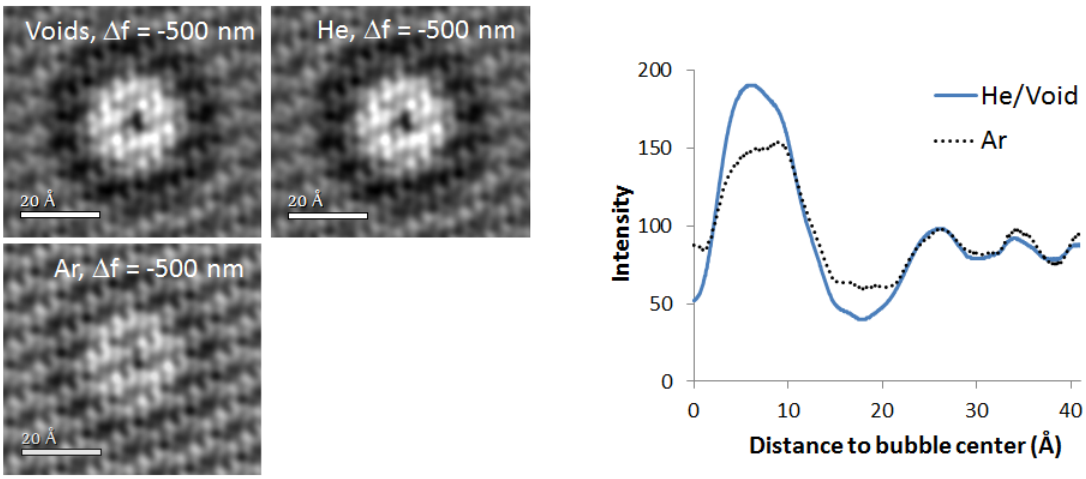

Fig. 4. (a) Simulated TEM images and (b) average-intensity profiles of a void, He bubble, and Ar bubble in Fe matrix. The defocus is 500 nm and illumination spread semi-angle is 0.3 mrad.

CONCLUSIONS

In summary, we found through multislice simulation and TEM characterization that:

1) The beam incoherency was found to have a great influence on the ratio of inner diameter of the first dark Fresnel ring to the actual bubble size, D_{in}/D_0.
2) It was also shown that D_{in}/D_0 decreases with increasing defocus. This general trend is not significantly influenced by the acceleration voltage and bubble size. With a defocus of 1 μm, D_{in} is about 85% of the actual bubble size.
3) Bubble size and position in the TEM sample thickness was found to have a negligible effect on D_{in}/D_0. The sample thickness effect was also shown to be very small.
4) Preliminary results suggest that the He bubbles and Ar bubbles can be distinguished by differences in contrast when imaged under the same conditions.

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