Investigating Helium Bubble Formation in $NiFe_x$ Alloys using Electron Energy Loss Spectroscopy

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Due to its extremely low solubility in solid materials, helium (He) tends to cluster with vacancies to form pressurized bubbles [1]. The formation and accumulation of He bubbles can lead to dramatic degradation of the material's properties [2]. Electron energy loss spectroscopy (EELS) is a valuable tool to study He bubble growth processes as it can not only quantitatively characterize chemical segregation around bubbles but can also be used to measure the gas density inside individual bubbles. In this study, we applied EELS to study the effect of Fe concentration in $NiFe_x$-based single-phase concentrated solid solution alloys (SP-CSAs) on bubble formation.

Three SP-CSAs with different Fe concentrations ($NiFe_{20}$, $NiFe_{35}$, and $NiFe_{50}$) and Ni were irradiated by 200 keV He ions at 500°C to a fluence of 5×10^{16} He/cm^2. The size distribution of the He bubbles in the four samples were characterized using transmission electron microscopy (TEM); EELS spectrum images around individual bubbles were acquired using a Nion UltraSTEM operated at 100 kV. Local Fe concentrations were calculated based on the intensities of the core loss peaks for Fe (708 eV) and Ni (855 eV). Due to the overlap of wavefunctions of neighboring He atoms, the He 1s → 2p absorption peak inside a bubble will exhibit a decrease in energy when compared to a free He atom at 21.22 eV, and thus, the peak energy decrease (ΔE) is proportional to the He density within the bubble, n, i.e., $\Delta E=Cn$ [2]. To measure the ΔE, core loss signals in the range 15-55 eV on the energy loss spectrum were collected.

Our TEM analyses show the growth of bubbles is suppressed with increasing Fe concentration in the SP-CSAs. Under the same irradiation conditions, bubbles having ~10 nm diameter are widely observed in Ni and $NiFe_{20}$, while most of the bubbles in $NiFe_{50}$ are smaller than 5 nm. To understand the effect of Fe content on bubble growth, the He densities were measured in bubbles having different sizes in all three alloys. Fig. 1(a) is a typical high-angle annular dark field (HAADF)-STEM image of a bubble and Fig. 1(b) shows the EELS collected from inside (position A) and outside (position B) of the bubble. Comparing these two EELS profiles, the He absorption peak around 24 eV can be clearly identified. Fig. 1(c) summarizes the measured He densities in all three $NiFe_x$ alloys. In general, the He density decreases as the bubble size increases. This trend agrees with the Laplace-Yong law, i.e., the gas density is proportional to $2\sigma/r$, where σ is the bubble surface tension and r is the bubble radius [1]. When focusing on bubbles within a similar size range (2nm-5nm), we observe that the average He density increases from 76 nm^{-3} in $NiFe_{20}$ to 99 nm^{-3} in $NiFe_{35}$ and to 122 nm^{-3} in $NiFe_{50}$, which suggests that a higher Fe concentration can lead to a higher bubble surface tension and inhibit the bubble growth rate. EELS was also applied to characterize the chemical segregation near the bubbles. Fig. 2(a) shows a HAADF-STEM image of a He bubble in $NiFe_{50}$ and Fig 2(b) is the corresponding elemental composition map, which shows that Fe (red) is depleted while Ni (blue) is enriched around the bubble. Atomistic simulations show that in $NiFe_x$ alloys, interstitials diffuse mostly via the Ni sub-lattice while vacancies diffuse mostly via Fe atoms [3]. As bubbles are defect sinks, preferential defect diffusion will lead to segregation near the bubbles. Fig. 2(c) compares the segregation behavior for the different alloys. For bubbles having a similar size (~ 4 nm) and within a similar sample thickness (~ 0.7 inelastic mean free path), the chemical fluctuations near the bubbles become larger as the Fe concentration increases. Combining these observations with the calculated defect diffusivities [3], this segregation trend suggests that vacancy diffusion may play a dominant role during the bubble growth process in these SP-CSAs [4].

References:

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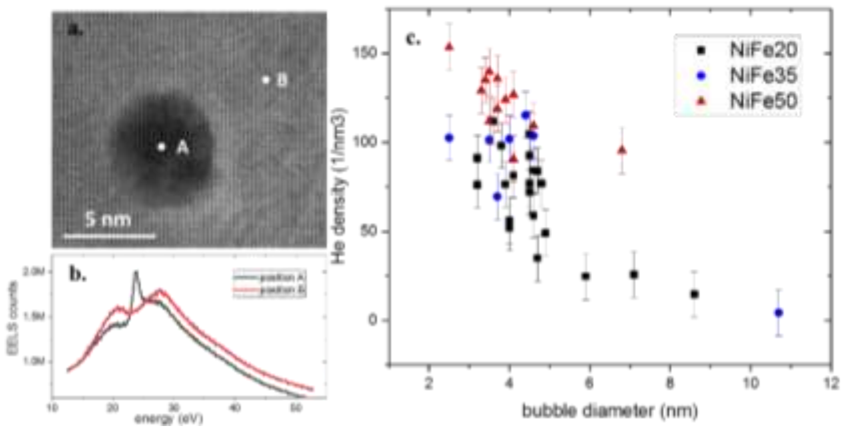

Figure 1. (a) HAADF-STEM image of a typical He bubble in $NiFe_{50}$; (2) EELS profiles acquired from positions A and B in (a); (c) He densities inside bubbles of different sizes in three SP-CSAs.

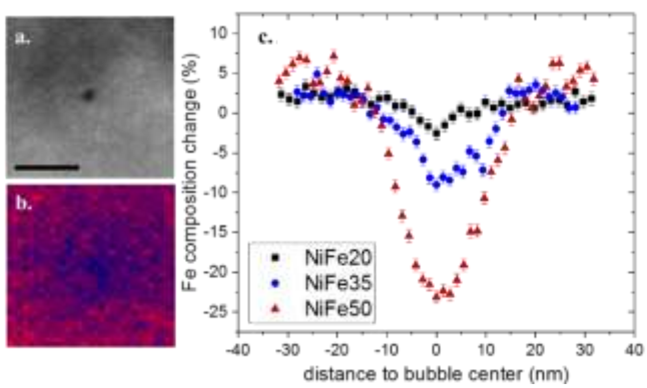

Figure 2. (a) HAADF-STEM image showing region where EELS spectrum image was taken. He bubble is low intensity region in center of image. The scale bar is 20 nm; (b) corresponding composition map (Ni is blue, Fe is red); (c) profiles of absolute Fe concentration changes near He bubbles in three $NiFe_x$ SP-CSAs.