8.2 An Atomistic Assessment of Helium Behavior in Iron — R. E. Stoller and Y. N. Osetskiy (Oak Ridge National Laboratory)

EXTENDED ABSTRACT - paper submitted to J. Nucl. Mater. after ICFRM-16

High helium generation rates in irradiated materials leads to the formation of small He-vacancy clusters that can evolve into larger bubbles and voids. In order understand and predict the behaviour of these He-vacancy defects, an accurate equation of state is required to reproduce their pressure- relationship. Previous research has employed equations of state of varying complexity, including the ideal gas, van der Waals, and hard sphere models. We recently used *ab initio* calculations to determine the energetics of helium-vacancy clusters and applied the results to develop a new three-body interatomic potential that describes the behaviour of helium in iron. This potential was employed in molecular dynamics simulations to determine the conditions for mechanical equilibrium between small helium-stabilized bubbles and an iron matrix, and to systematically map the pressure-temperature relationship for the bubbles. These atomistic results are compared to an existing equation of state and a modification is proposed for bubbles with high helium densities.

Our MD simulations with the iron interatomic potential by Ackland and co-workers [1] coupled with the ORNL three-body He-Fe potential [2] indicate that the helium-to-vacancy ratio for bubbles large enough to be visible in the transmission electron microscope is in the range of 0.3 to 1.0. Values greater than 1.0 are obtained only for bubbles with radii less than 1 nm. This result is consistent with a recent experimental measurements using electron energy loss spectroscopy to determine the helium-to-vacancy ratio for small helium bubbles in ferritic-martensitic steels. The helium density results of Wu et al. for a 1.3 nm bubble lead to a He/vacancy ratio ~0.6 [3], and earlier measurements by Frechard et al. for bubbles between 2 and 5 nm for which the He/vacancy ratio was in the range of 0.25 to 0.85 [4]. As shown in Figure 1, the helium pressure in equilibrium bubbles was in good agreement with estimates provided by the capillarity pressure of 2g/r where g is the surface energy obtained from the iron potential and r is the bubble radius. The bubble compressibilities shown in Figure 2 were found to be consistent with a computationally tractable hard-sphere equation of state [5] with a minor adjustment in the effective hard-sphere diameter used to compute the helium density. Based on previous work in which we assessed the behavior of helium vacancy clusters using three different iron potentials in conjunction with our three-body He-Fe potential, it does not appear that the choice of a different iron potential would have any significant impact on our results. In recent work by Caro, *et al*. [6] which investigated the behavior of helium in iron at higher He/vacancy ratios and employed a different equation of state, they work suggests that the equilibrium He/vacancy ratio is about 1, which is higher than we observed in our simulations. The reasons for this discrepancy are not clear at this point and may be resolved by further research.

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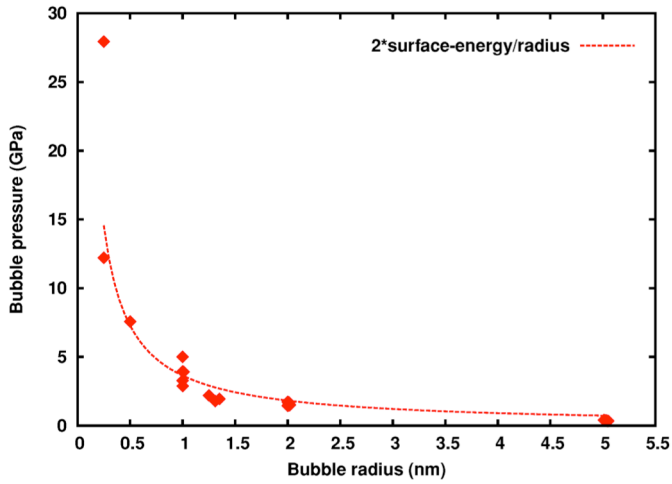

Figure 1. Bubble pressure obtained from MD simulations for bubbles in mechanical equilibrium with the iron lattice (red diamonds) compared with capillarity pressure using the surface energy obtained from MD.

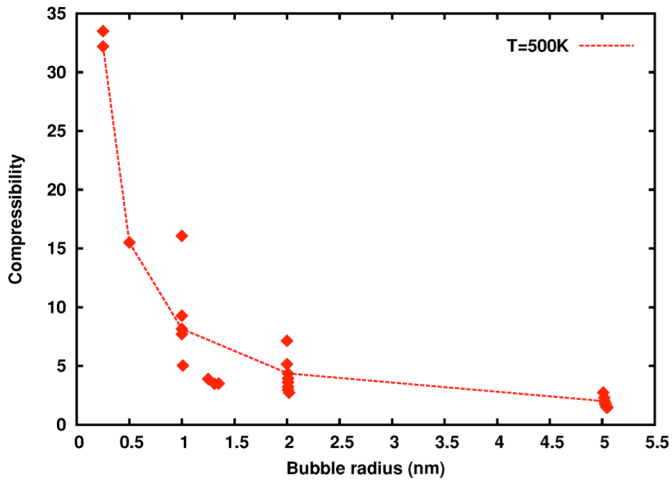

Figure 2. Compressibility of bubbles in mechanical equilibrium with the iron lattice as a function of bubble radius. Compressibility defined as ratio of pressure from MD simulations divided by ideal gas pressure.