7.1 Progress on Developing an Atomistic Equation of State for Helium in Iron —

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OBJECTIVE

The objective of this research is to develop the modeling and simulation tools necessary to improve our understanding of how helium interacts with other defects in iron-based alloys to produce changes in the microstructure and mechanical properties.

SUMMARY

A large series of molecular dynamics simulations have been completed to map out the pressure-temperature relationship for helium in iron to provide the basis for fitting of a new equation of state for helium bubbles. The behavior of helium is described by the three-body Fe-He potential developed at ORNL under the fusion program.

PROGRESS AND STATUS

Introduction

Because of the high helium levels generated by nuclear transmutation reactions, predictions of structural material performance under DT fusion conditions must account for how the helium influences microstructural evolution and the related mechanical properties. One of the primary issues that must be addressed is the behavior of He-vacancy clusters, which evolve, into finite-sized bubbles with the potential for growing into voids. In this case, a 'bubble' is defined as a cavity, which is stabilized by the helium pressure it contains. If helium is removed a bubble will shrink. In contrast, a 'void' is a cavity, which has grown large enough that growth can be maintained by the absorption of a net vacancy flux. The gas pressure plays essentially no role in voids stability. The properties of small He-vacancy clusters, such as the helium and vacancy binding energy are critical to predicting bubble stability. These processes can only be accurately described by atomistic simulations such as molecular dynamics (MD), which requires an interatomic potential to describe the behavior of both the metal and helium atoms. In order to improve our predictive capability for the Fe-based materials of most interest to fusion structural applications, a new Fe-He interatomic potential was developed based on ab initio calculations of the interactions between He and point defects in Fe [1-2]. Additional work using this potential has characterized the properties of small He-vacancy clusters and parameters such as the He-to-vacancy ratio as a function if cluster/bubble size and temperature [3-4]. The observations of that work are being extended to develop an atomistic equation of state for helium in iron, which can then be applied in a broad range of mesoscale to predict microstructural evolution [5].

Current Progress

In order to provide a basis for fitting the pressure-temperature relationship required for the equation of state (EOS), a series of MD simulations have been carried out covering the temperature range of 200 to 100K with bubble radii from 0.25 to 5 nm. For each of the bubble size, the helum-to-vacancy ratio was varied to produce a broad range of pressures, from highly over-pressurized to a void-like under-pressurized state. The pressure at mechanical equilibrium was also being determined.

As discussed in Ref. [5], the He-Fe repulsive interaction leads to a region of the bubble adjacent to the iron matrix that is effectively void-like. Thus the volume occupied by the helium is less than the bubble volume based on the number of vacancies that comprise the bubble. This effect is shown in Fig. 1 in which equilibrium bubble pressure is shown for a range of bubble sizes at 300K. Three different equations of state were used to calculate the pressures

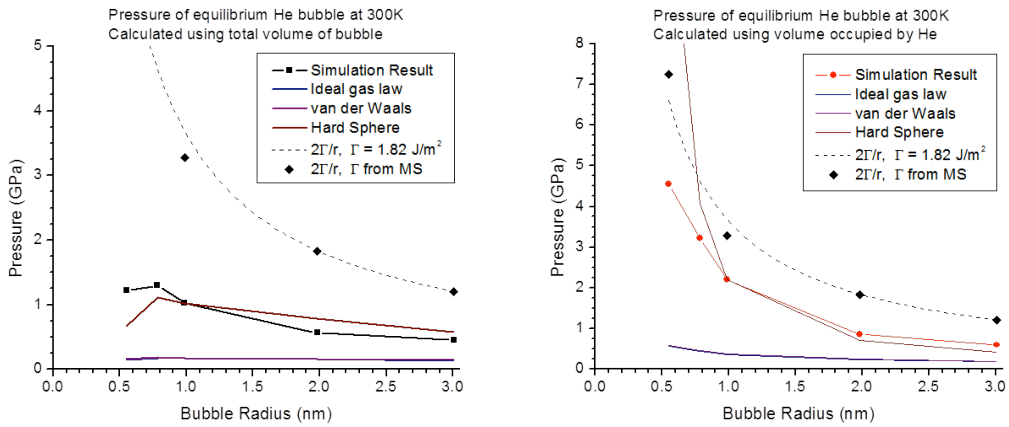

Fig. 1. Influence of gas-free region near the bubble surface on the equilibrium pressure calculated for helium bubbles in iron.

and are compared with the pressure obtained in the MD simulations. For part (a) the pressure was calculated using the total bubble volume and in part (b) using only the volume occupied by helium. The difference is dramatic for the smaller bubble sizes in which the helium-free volume is a significant fraction of the total volume. Note that the equilibrium pressure predicted by the capillarity model (2G/r) is generally much higher than that obtained in the MD simulations. The hard sphere EOS of Brearley and MacInnes [6] is reasonably consistent with the MD results and will be used as the basis for developing the MD-based EOS.

The influence of gas pressure or He/vacancy ratio is illustrated in Fig. 2, which compares the He atom, pair correlation functions obtained at different pressures at 600K. More liquid or solid-like behavior observed for at the higher He/vacancy ratios, i.e. there is clear evidence of the formation of a He atom lattice in the curve for a He/vacancy ratio of 1.5. The helium atoms are more gas-like and randomly distributed at lower He-vacancy ratios.

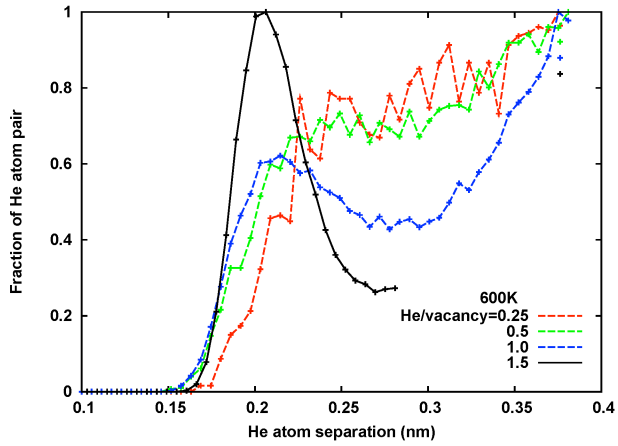

Fig. 2. He atom pair correlation function in bubbles at 600K for different pressures (He/vacancy ratios).

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