The Troubles with Bubbles – Modeling the Material Degradation Caused By Helium Bubble Growth

The certification of our weapons components and subsystems remains a critical requirement. The elimination of underground testing and reduction in test facilities and capabilities has prompted us to develop high fidelity models based on knowledge of physical processes and materials behavior. Among many issues that need to be understood is material degradation associated with tritium aging. Tritium decay produces helium atoms that cluster into nano-scale bubbles. The nucleation, growth and interaction of these bubbles alter the material's mechanical properties and creates microstructural defects such as dislocations. A key phenomenon tied to the presence of bubbles is low level helium gas release from the metal-tritide alloy which increases cataclysmically in time. This has been observed in the most-studied system of palladium-tritide and in erbium-tritide used in MC4277 and MC4300 neutron tubes. Release of helium from aging tritide targets produces a buildup of gas within the vacuum tube envelope, thereby limiting component performance and lifetime.

The Physical and Engineering Sciences Center 8700, and Materials and Process Sciences Center 1800, are collaborating to use analytical and computational models to study the material defects created during helium bubble growth and to understand the mechanical interaction between these defects and the bubbles themselves, as well as predict the physical mechanism responsible for the release of helium gas. These models operate over various length scales, down to the level of inter-atomic spacing.

Using Sandia's Embedded Atom Method, an inter-atomic potential model, simulations of molecular dynamics have been performed by Jonathan Zimmerman and Jeffrey Hoyt to examine material defects from spherical helium bubbles within a palladium lattice. Figure 1 shows a model system

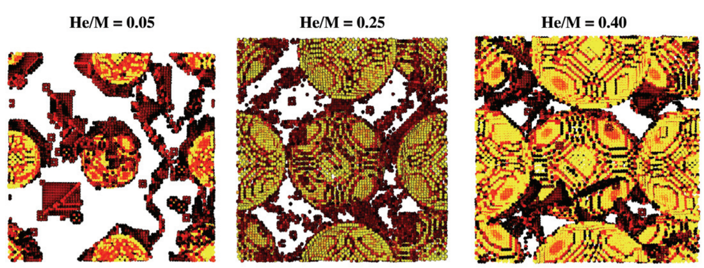

Figure 1. Molecular dynamics simulations of dislocations, surfaces and other material defects created during the growth of helium (He) bubbles within a bulk lattice. Three different concentrations of helium to metal (M) atoms are shown. Atoms are colored according to their value of centrosymmetry parameter; hence, only atoms bordering defects are visible.

of four bubbles growing within a bulk lattice. Atoms bordering defects such as dislocation cores and bubble surfaces are visually isolated by using the centrosymmetry parameter, a measure of radial symmetry of distributed near-neighbors for a given atom. As the bubbles grow, numerous defects are generated such as dislocation threads that interconnect bubbles and stacking fault tetrahedra. These results are quite different from those of previous models that depicted prismatic loops of dislocations that emerged from a bubble's periphery. Similar defects were also observed in simulations of bubbles grown near a free surface, as shown in Figure 2. In these simulations, some of the threading dislocations connect the bubbles to the free surface, resulting in roughening, a phenomenon that has been observed in experiments of helium-implanted palladium samples.

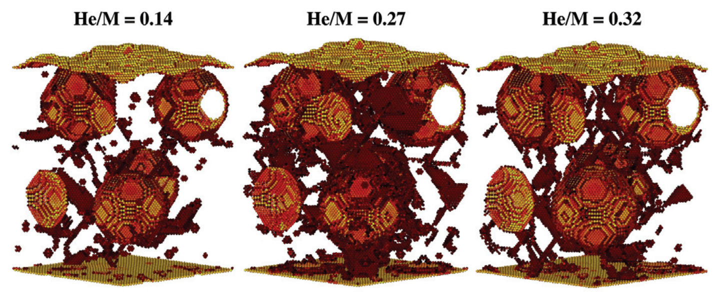

Figure 2. Molecular dynamics simulations of defects created helium (He) bubble growth near a free surface. Three He/M ratios are shown, and only atoms bording defects are visible.

Further development is underway to develop a physically realistic inter-atomic potential for the metal-hydride alloy, and to use it in future simulations of bubble growth. It is anticipated that these simulations will display additional mechanisms to explain the accelerated release of helium gas that occurs late in the aging process.

Work is also being done by Don Cowgill to develop larger-scale, analytical models. These models predict bubble size and spacing distribution, how stress fields originating from bubbles and dislocations interact and affect bubble growth, and how that interaction leads to failure of the material through either early or late release of helium gas. Fundamental knowledge obtained through these models has shown how material properties affect the shape of the bubbles formed, which in turn dictates what types of defects are emitted during the growth process. Figure 3 schematically shows two types of bubble shapes and the resulting defects created during bubble growth, that of spherical bubbles that grow via dislocation loop punching and dislocation thread emission and that of platelet-shaped bubbles that grow by the expansion of dislocation dipoles.

Micromechanical models developed by Don show that lower pressures are expected when the bubbles assume a preferred shape (due to the mechanical properties of surface energy, shear modulus and Burgers' vector). The preferred shape is spherical in palladium-tritide, and resembles platelets in erbium-tritide. These predictions were confirmed through micrographs for tritide samples of a sufficient age. The model also predicts that at a very early age, i.e. for very small bubbles, palladium-tritide should prefer the platelet bubble shape for clustering helium. To validate these models, collaborators at Lawrence Livermore National Laboratory and Sandia will perform experiments on palladium-tritide aging whereby imaging techniques will be used to clarify bubble shape. Further model development is also in progress to determine how these unique defects affect early release of helium gas in erbium-tritide systems.

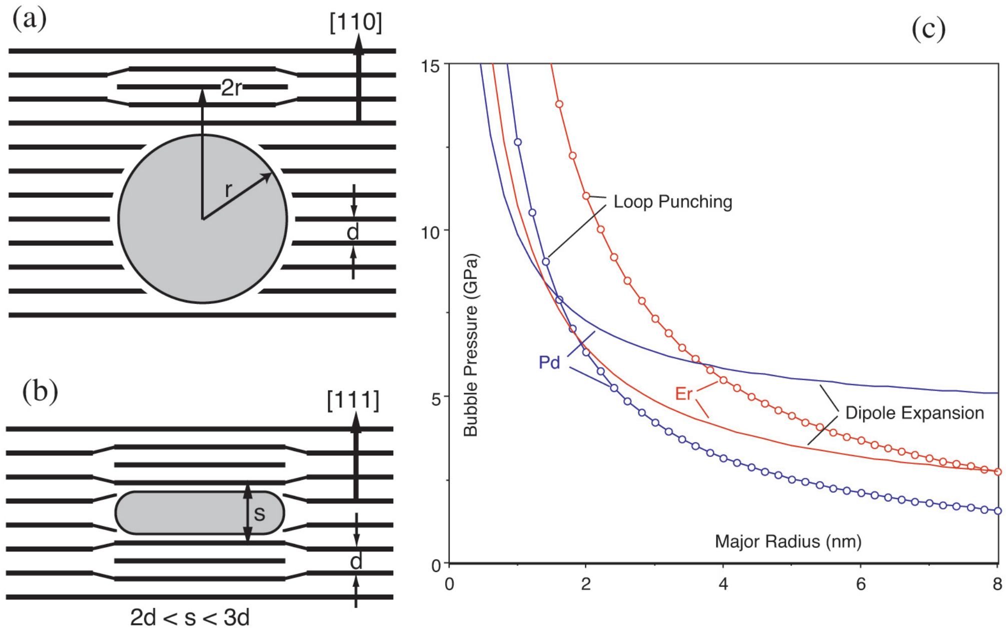

Figure 3. Schematics of (a) helium (He) bubble growth via dislocation loop punching and (b) He platelet growth via dislocation dipole expansion. (c) Models of how bubble pressure changes with defect size show that lower pressures are predicted when He platelets grow in erbium and when He bubble grow in palladium. Note that for very small bubbles, platelet growth is preferred by both materials.

The ultimate goal of these modeling efforts is to construct continuum-scale constitutive models that depend on initial defect content, stress-strain history, temperature, radiation, target film processing and film microstructure. By incorporating relevant details of atomic-scale physical mechanisms operating during helium bubble growth, these models benefit the Laboratory's stockpile stewardship mission.

Jonathan Zimmerman (standing) joined the Science-Based Materials Modeling Dept. 8763 in 1999. To the far left is New Mexico-based team member Jeff Hoyt, and seated is Don Cowgill. Jonathan's research interests include the mechanics of materials, atomistic simulation, fractures, dislocation and effect mechanisms, and multiscale modeling methods. He obtained his Ph.D. from Stanford University in mechanical engineering in 1999, where he also received a master's degree in the same subject. He also holds a B.A. in physics from the State University of New York at Binghamton and a B.S. from that campus in mechanical engineering. Outside of work he enjoys movies and television and is a comic book aficionado with a collection of superhero statues in his Sandia office.