MAR16-2015-002101

Abstract for an Invited Paper
for the MAR16 Meeting of
the American Physical Society

Real time studies of Elastic Moduli Pu Aging using Resonant Ultrasound Spectroscopy

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Elastic moduli are fundamental thermodynamic susceptibilities that connect directly to thermodynamics, electronic structure and give important information about mechanical properties. To determine the time evolution of the elastic properties in ^{239}Pu and it Ga alloys, is imperative to study its phase stability and self-irradiation damage process. The most-likely sources of these changes include a) ingrowth of radioactive decay products like He and U, b) the introduction of radiation damage, c) δ-phase instabilities towards α-Pu or to Pu_3Ga. The measurement of mechanical resonance frequencies can be made with extreme precision and used to compute the elastic moduli without corrections giving important insight in this problem. Using Resonant Ultrasound Spectroscopy, we measured the time dependence of the mechanical resonance frequencies of fine-grained polycrystalline δ-phase ^{239}Pu, from 300K up to 480K. At room temperature, the shear modulus shows an increase in time (stiffening), but the bulk modulus decreases (softening). These are the first real-time measurements of room temperature aging of the elastic moduli, and the changes are consistent with elastic moduli measurements performed on 44 year old δ-Pu. As the temperature is increased, the rate of change increases exponentially, with both moduli becoming stiffer with time. For T>420K an abrupt change in the time dependence is observed indicating that the bulk and shear moduli have opposite rates of change. Our measurements provide a basis for ruling out the decomposition of δ-Pu towards α-Pu or Pu_3Ga, and indicate a complex defect-related scenario from which we are gathering important clues.