UCRL-CONF-234546

LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

Isothermal Martensitic and Pressure-Induced ? to ?# Phase Transformations in a Pu-Ga Alloy

A. J. Schwartz, M. A. Wall, D. L. Farber, K. T. Moore, K. J. M. Blobaum

September 12, 2007

Plasticity 2008
Kailua/Kona, HI, United States
January 3, 2008 through January 8, 2008

UCRL-CONF-234546

ISOTHERMAL MARTENSITIC AND PRESSURE-INDUCED δ TO α′ PHASE TRANSFORMATIONS IN A PU-GA ALLOY

Adam J. Schwartz*, Mark A. Wall, Daniel L. Farber**, Kevin T. Moore**, and Kerri J.M. Blobaum****

***Physical Science Directorate, Lawrence Livermore National Laboratory, L-041, 7000 East Avenue, Livermore, CA 94550, USA, **Chemistry, Materials, and Life Sciences Directorate, Lawrence Livermore National Laboratory, 7000 East Avenue, Livermore, CA 94550, USA,**
schwartz6@llnl.gov, wall1@llnl.gov, farber2@llnl.gov, moore78@llnl.gov, blobaum1@llnl.gov

ABSTRACT: A Pu–2 at.% Ga alloy specimen is slowly compressed to ~1 GPa in a large volume moissanite anvil cell to induce the face-centered cubic δ to simple monoclinic α′ phase transformation. Optical microscopy, x-ray diffraction, and transmission electron microscopy of the specimen recovered to ambient pressure reveal that the vast majority of the microstructure consists of the α′ phase with grain sizes ranging from 10 nm to several hundred nm, with the remainder being δ phase dispersed between the α′ grains. This morphology is in contrast to the transformation product of the low-temperature isothermal martensite in which the lath-shaped α′ particles are ~ 20 μm by 2 μm.

INTRODUCTION: A well-homogenized Pu–2 at.% Ga alloy can be retained in the metastable face-centered cubic δ phase at room temperature. Ultimately, this metastable δ phase will decompose via a eutectoid transformation to the thermodynamically stable monoclinic α phase and the intermetallic compound Pu_3Ga over a period of approximately 10,000 years (Timofeeva [2000]). In addition, these low solute-containing δ-phase Pu alloys are metastable with respect to an isothermal martensitic phase transformation to the α′ phase during low temperature excursions (Orme *et al.* [1971], Hecker *et al.* [2004]) and are also metastable with respect to a δ → α′ phase transformation with increases in pressure (Faure *et al.* [2006], Hecker *et al.* [2004], and Harbur [2007]). The low temperature δ → α′ isothermal martensitic phase transformation in the Pu–2 at.% Ga alloy only goes to ~25% completion with the resultant ~20 μm long by 2 μm wide lath-shaped α′ particles dispersed within the δ matrix. In recently reported studies, Faure *et al.* [2006] have observed a δ → γ → α′ pressure-induced phase transformation sequence during a diamond anvil cell investigation and Harbur [2007] has concluded that both α′ and an amorphous phase are present in samples that were pressurized and recovered based on x-ray diffraction and density and compressibility experiments. In this work, we apply hydrostatic pressure to a Pu–2 at.% Ga alloy sample in a large volume moissanite (SiC) anvil cell and then recover to ambient pressure. The recovered specimen is characterized with optical microscopy, x-ray diffraction, transmission electron microscopy (TEM) and electron diffraction.

PROCEDURES, RESULTS AND DISCUSSION: A large volume moissanite anvil cell is constructed to permit the pressurization and recovery of specimens of a size suitable for TEM and electron diffraction studies. The cell, shown in Fig. 1, has an overall diameter of 101.6 mm, a moissanite anvil diameter of 9.00 mm, a culet size of 3 mm, and a spring steel gasket 0.5 mm thick with a hole diameter of 2.5 mm. In this study, a 2.3 mm diameter by 100 μm thick sample of δ-phase Pu–2 at.% Ga is compressed at a rate of approximately 0.05 GPa/minute to ~1 GPa to induce the phase transformation to α′. The recovered specimen is characterized with optical microscopy to evaluate the micron-sized and above features of the microstructure, x-ray diffraction to determine the crystal structure of the phase or phases, and TEM to determine the nanometer-scaled features of the microstructure. Optical microscopy reveals a very fine microstructure that appears to be single phase, although the resolution of this technique is insufficient to differentiate between single and multiple phases if the grain size is below approximately 1 μm. X-ray diffraction, using a laboratory Cu K_{α} source with wavelength of 1.542Å, shows the monoclinic reflections from the α′ phase, strong peaks from the aluminum specimen holder, and weak peaks from the face-centered cubic δ phase as shown in Fig. 2. The recovered specimen is prepared for TEM and electron diffraction studies as described in Moore *et al.* [2007]. TEM reveals small regions of δ phase with a very high dislocation density interspersed between the 10 – 100's nm α′ grains as shown in Fig 3. Electron diffraction, shown in the insert in Fig. 3, clearly reveals the presence of the δ phase. This microstructure is in contrast to the α′ particles that form as a result of the low-temperature isothermal martensite in which the α′ particles are lath-shaped and significantly larger as shown in the optical micrograph in Fig. 4 of a sample cooled to –120°C and held for 10 hours. In these preliminary results, there is no evidence of either an amorphous phase, as suggested by Harbur [2007], or the presence of a γ phase. We expected to observe an amorphous phase based on the similarity of this experiment to that of Harbur [2007]. The γ phase, as reported by Faure *et al.* [2006], does not appear to be retained to ambient pressure.

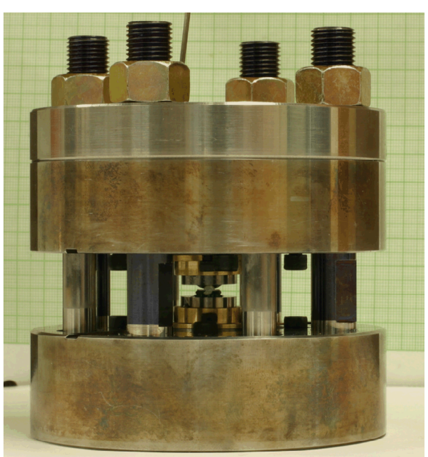

Fig. 1. Photograph of the large volume moissanite anvil cell.

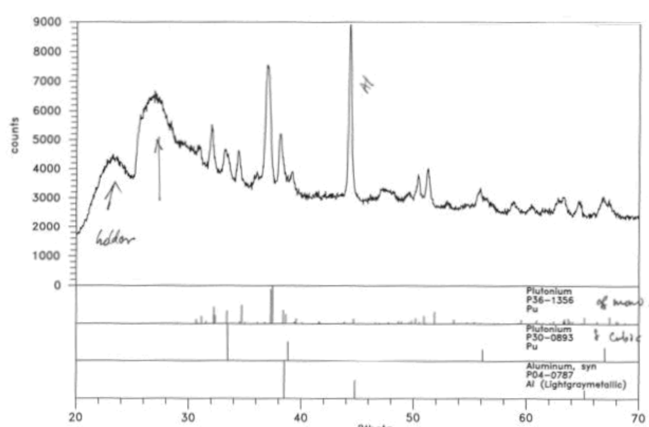

Fig. 2. X-ray diffraction scan of the specimen that was compressed to approximately 1 GPa and recovered to ambient pressure.

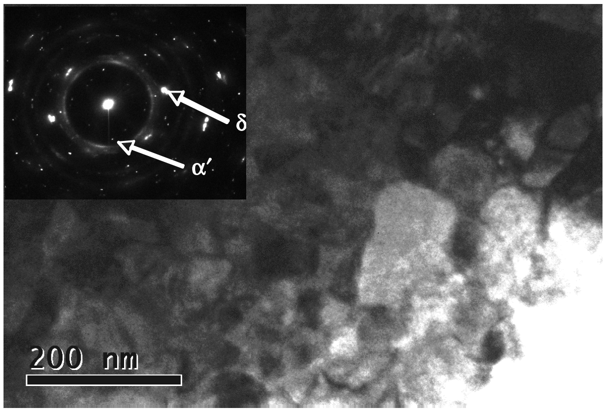

Fig. 3. Bright-field TEM micrograph and electron diffraction pattern of the Pu–2 at.% Ga specimen that was compressed to ~1 GPa and recovered to ambient pressure. Very fine grains of the α′ phase and evidence both the δ and α′ phases are observed.

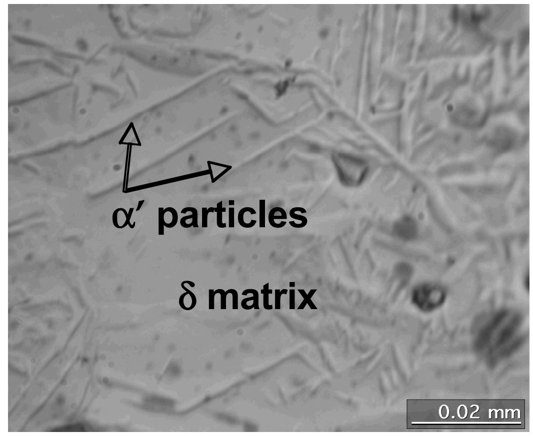

Fig. 4. Optical micrograph of the Pu–2 at.% Ga specimen that was cooled to –120°C and held for 10 hours. α′ particles are observed to be dispersed in the δ matrix.

Acknowledgment: This work was performed under the auspices of the United States Department of Energy by the University of California, Lawrence Livermore National Laboratory, under Contract No. W-7405-Eng-48.

REFERENCES:

Faure, Ph., Klosek, V., Genestier, C., Baclet, N., Heathman, S., Normile, P., and Haire, R. 2006, "Structural investigation of δ-stabilized plutonium alloys under pressure", in: J.L. Sarrao, A.J. Schwartz, M.R. Antonio, P.C. Burns, R.G. Haire, and H. Nitsche, (Eds.), Actinides 2005 – Basic Science, Applications and Technology, MRS Proceedings, **893**, pp. 223-232, Materials Research Society, Pittsburgh.

Harbur, D.R. 2007, The effect of pressure on phase-stability in the Pu-Ga alloy system", Journal of Alloys and Compounds, **444-445**, 249.

Hecker, S.S., Harbor, D.R., and Zocco, T.G. 2004, "Phase stability and phase transformation in Pu–Ga alloys", Prog. Mater. Sci. **49**, 429.

Moore, K.T., Krenn, C.R., Wall, M.A., and Schwartz, A.J. 2007, "Orientation relationship, habit plane, twin relationship, interfacial structure, and plastic deformation resulting from the δ to α′ isothermal martensitic transformation in Pu-Ga alloys", Met. and Mat. Trans., **38A**, 212.

Orme, J.T., Faiers, M.E., and Ward, B.J. 1975, "The kinetics of the delta to alpha transformation in plutonium rich Pu-Ga alloys", in: H. Blank, R. Lindner (Eds.), Plutonium 1975 and Other Actinides, North Holland Publishing, Amsterdam, 761.

Timofeeva, L.F. 2000, "Low-temperature equilibrium aging under self-irradiation in binary alloys of Pu with elements of the IIIB group", in: L. Mallison (Ed.), Proceedings of the International Congress on Ageing Studies and Lifetime Extension of Materials, pp. 191, Kluwer Academic Publishers, Dordrecht.