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SOUND VELOCITY AND ELASTIC MODULI IN α-PLUTONIUM AT PRESSURES TO 50 KBAR*

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The elastic moduli - bulk modulus and shear modulus - were measured in polycrystalline α-plutonium at pressures to 50 Kbar. An ultrasonic technique enabled measurement of both longitudinal and shear wave velocities in a girdled-piston high pressure cell. The average pressure derivatives were 14.3 and 4.3 m/sec/Kbar for the longitudinal and shear velocities, respectively. Bulk and shear moduli were calculated from the sound velocity data. The average pressure derivatives for the moduli were 14 and 3.5, respectively. Poisson's ratio was calculated directly from the ratio of longitudinal and shear velocities and increased from 0.17 at atmospheric pressure to 0.28 at 50 Kbar.

*This paper is based on work performed under United States Energy Research and Development Administration Contract AT(45-1)-1830.

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Longitudinal and shear sound velocities were measured in α-plutonium at pressures to 50 Kbar. The pressure derivatives were 14.3 and 4.3 m/sec/Kbar, respectively. The elastic bulk and shear moduli were calculated and the average pressure derivatives were 14 and 3.5, respectively. Poisson's ratio increased from 0.17 at atmospheric pressure to 0.28 at 50 Kbar.

INTRODUCTION

Alpha plutonium has enough unusual mechanical and physical properties that attempts to predict an unknown property based on behavior or trends in other metals are likely to be unsuccessful. A review of the elastic properties of the actinides by Fisher [1] indicates little available data on plutonium and neptunium under pressure. The increasing availability of single crystals of α-plutonium will hopefully enable a measurement of elastic constants in the near future. Alpha neptunium is not yet available in sufficient quantity to warrant such an ambitious study, though the fabrication of single crystals should be easier for neptunium because of the fewer allotropic transformations compared to plutonium. The present work was done on high purity electrorefined polycrystalline α-plutonium to give for the first time the pressure variation of both shear and bulk moduli.

Experimental Technique

High purity electrorefined metal with approximately 600 ppm total impurities including 10 ppm Al, 15 ppm Fe, 20 ppm Mo, 25 ppm O, 5 ppm Ta, 4 ppm Th, 69 ppm W, 150 ppm Am, and 20 ppm U was obtained for the study. The initial density was 19.72 g/cm^3 but increased to 19.80 g/cm^3 after pressurization to 50 Kbar. Thus these measurements were on metal with very nearly theoretical x-ray density (19.83 g/cm^3). Sound velocities were measured at ambient pressure on the densified metal. The metal was extruded in the α-phase to refine the grain size and $\alpha \rightarrow \beta \rightarrow \alpha$ cycled once to remove the texture caused by extrusion. The grain size after these treatments was equiaxed and 2-4 μm. X-ray diffraction indicated no appreciable texture in the specimens.

A high pressure cell of the girdled-piston type was used for the sound velocity measurements with a technique used by Gilmore [2]. The maximum attainable pressure was approximately 60 Kbar. Transducers were positioned on the back surfaces of the carbide pistons above and below the sample. This arrangement permitted measurement of both shear and longitudinal wave transit times during the same pressurization cycle.

The measurement of transit times was conveniently done with a pulse-echo overlap technique [3]. This technique was especially appropriate because of the high attenuation of the wave in plutonium which made an interference technique difficult to use (though of potentially higher accuracy). The transit times were measured to within about 0.1%, which was more than adequate when the error involved in assigning specimen length is considered.

The specimen length was calculated by the difference between the LVDT displacement measurement and cell deflection. The cell deflection calibration (deflection vs

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load) was obtained by measuring transit times in aluminum (for various loads and corresponding pressures) and calculating the specimen length from published values for sound velocity in aluminum under pressure [2,4,5]. The calibration was checked by sound velocity measurements on copper which agreed well with values taken from published data.

To minimize the effects of dimensional changes, the specimens were cycled to 50 Kbar twice before the transit time measurements. The transit times were measured on the loading part of the third cycle. Specimen length was measured after unloading to give a reference length for calculating the length under pressure. About 2 Kbar was required to establish acoustic coupling between the carbide piston and the specimen. Transit times measured at the acoustic coupling point upon unloading and at the beginning of the third cycle pressurization were used to calculate specimen length at the beginning of the third pressurization cycle. This calculation was based on equality of velocities at the loading and unloading acoustic coupling points. The small specimen length change between the acoustic coupling pressure (∿2 Kbar) and complete unloading was negligible as determined by cycling a specimen between these two points several times. With the length determined at the beginning of the third cycle, the length at higher pressures was calculated from the LVDT displacements and the cell deflection calibration. The accuracy of length calculated was estimated at ±0.6%.

The pressure cell was calibrated with the documented phase transitions of bismuth (I - II, 25.4 Kbar; III - IV, 55 Kbar) and thallium (35.6 Kbar). Small wires of these metals were positioned axially in the cell within a silver chloride sleeve and electrical resistance was monitored to detect the phase transitions. Pressure calibrations were done on a third pressure cycle since the pressure-load relation depended on the amount of irreversible axial deformation, which increased with each pressure cycle.

RESULTS

Sound Velocity

The longitudinal and shear velocities to 50 Kbar are listed in table 1 and plotted in fig. 1. The longitudinal velocity had an unusually large pressure dependence. A best linear fit gave $dV_l/dp \approx 14.3$ m/sec/Kbar (longitudinal) and $dV_s/dp \approx 4.3$ m/sec/Kbar (shear). The irregularity of the longitudinal velocity data points around 15 to 25 Kbar was caused by an interfering reflection from the pyrophyllite gasket that altered the shape of the pulse from the plutonium.

Table 1
Sound Velocities and Derived Moduli and Densities

Press (Kbar)	V_l (m/sec)	V_s (m/sec)	B_T (10^{11} dyne/cm^2)	B_s (10^{11} dyne/cm^2)	G (10^{11} dyne/cm^2)	Density (g/cm^3)
0.0	2415	1525	4.76	5.41	4.60	19.80
5.9	2478	1526	5.36	6.08	4.67	20.04
8.8	2532	1534	5.81	6.59	4.74	20.13
11.5	2594	1549	6.30	7.14	4.85	20.22
14.2	2641	1563	6.66	7.54	4.96	20.30
16.8	2695	1579	7.08	8.03	5.08	20.38
20.6	2762	1599	7.63	8.64	5.24	20.48
24.2	2805	1616	7.97	9.02	5.37	20.57
27.6	2859	1632	8.44	9.54	5.50	20.66
30.9	2899	1646	8.78	9.93	5.62	20.73
33.9	2937	1659	9.13	10.31	5.72	20.80
36.9	2972	1671	9.44	10.66	5.83	20.87
39.6	3001	1682	9.70	10.96	5.92	20.93
42.1	3033	1691	10.01	11.30	6.00	20.98
44.5	3059	1699	10.26	11.58	6.07	21.03
46.7	3085	1706	10.52	11.87	6.13	21.07
49.4	3113	1715	10.81	12.19	6.21	21.12
50.7	3129	1718	10.98	12.38	6.24	21.15

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39.6	3001	1682	9.70	10.96	5.92	20.93
42.1	3033	1691	10.01	11.30	6.00	20.98
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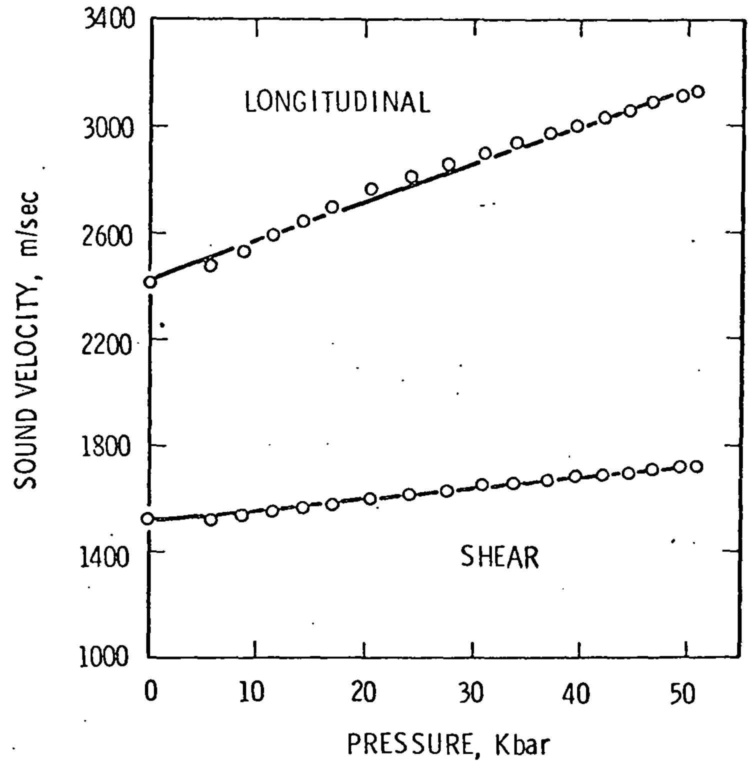

Figure 1. Longitudinal and Shear Sound Velocities in α-Plutonium versus Pressure.

Calculation of Elastic Moduli

The adiabatic bulk modulus, B_S, and shear modulus, G, are given by

$$B_S = \rho \left[V_l^2 - (4/3) V_s^2 \right] \quad (1)$$

$$G = \rho V_s^2 \quad (2)$$

where ρ is the density and V_l and V_s are the longitudinal and shear velocities.

The isothermal bulk modulus, B_T, is given by

$$B_T = B_s/(1 + \alpha\gamma T) \quad (3)$$

where α is the volume thermal expansion = $1.62 \times 10^{-4}/°K$,
γ is the Gruneisen constant = 2.87 and
T is the temperature = 296°K.

For the calculations, γ/V (V = volume) was assumed constant with increasing pressure [6].

The moduli were calculated after an iterative procedure was used to calculate the density at a given pressure. The isothermal modulus was calculated at pressure P_1 (atmospheric for first point). The change in density due to pressurization to P_2 was then calculated by $\rho_2 = \rho_1\ (1 + (P_2 - P_1)/3B_T)$. With the density at P_2, equations (1) and (3) gave B_S and G. This procedure was repeated for data points to the highest pressure, table 1, fig. 2. The pressure-volume relation was plotted together with Bridgeman's [7] values from isothermal compressibility measurements, fig. 3. The isothermal bulk moduli at high pressure agree quite well with the earlier work, e.g., we obtained B_T = 11 x 10^{11} dyne/cm^2 at 50 Kbar compared to 10.3 x 10^{11} dyne/cm^2 by Bridgeman. The average pressure derivatives for adiabatic bulk modulus and shear modulus were 14 and 3.5 (pressure and moduli expressed in same units).

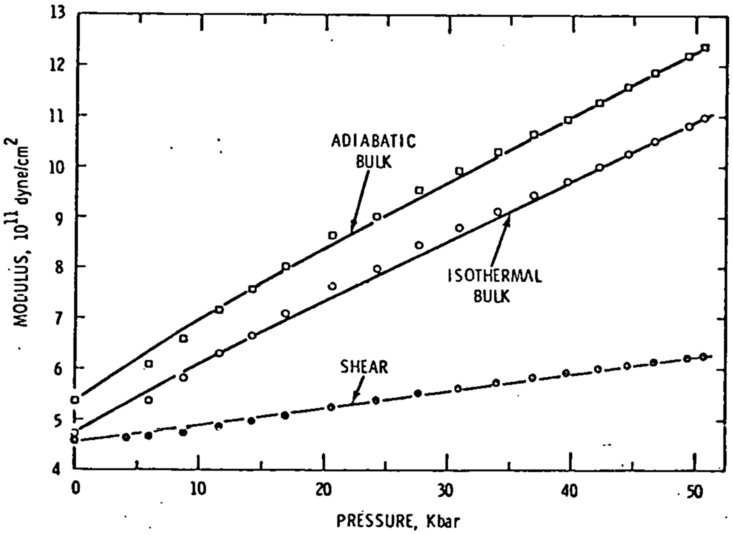

Figure 2. Bulk and Shear Moduli for α-Plutonium versus Pressure

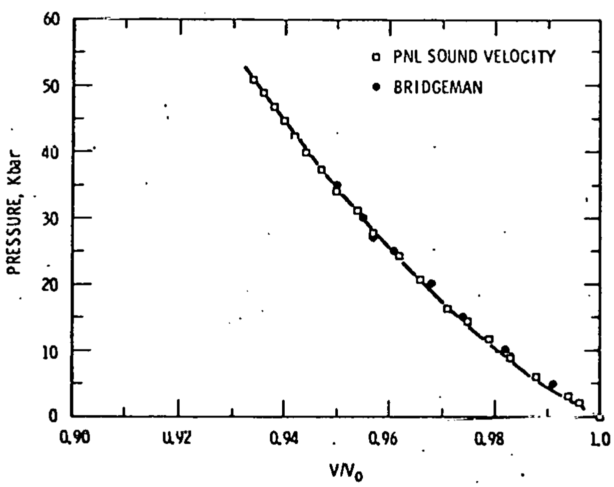

Figure 3. Pressure-Reduced Volume Relation for α-Plutonium

In addition to the values of B and G, the related pressure variation of Poisson's ratio, σ, was calculated according to

$$\sigma = 1/2(A - 2)/(A - 1) \quad (4)$$

where $A = (V_l/V_s)^2$

Poisson's ratio increased from 0.168 at atmospheric pressure to 0.28 at 50 Kbar. This is an enormous increase compared to that observed in cubic metals by Voronov [5] where σ increases typically 0.5 to 3% for a similar pressure increase. Thus the capacity for volumetric deformation is greatly decreased in α-plutonium upon application of pressure.

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