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Pressure and Temperature Dependences of the Isotropic Elastic Moduli of Polycrystalline Alumina

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The isotropic elastic moduli of polycrystalline alumina have been determined as a function of hydrostatic pressure up to 10 kbar and also as a function of temperature over the range 4.2° to about 1300°K. The pressure dependence of the elastic moduli is linear over this pressure range. The low-temperature limit of the elastic Debye temperature, 1044 (± 3)°K, compares very well with thermal Debye temperature. Values of various pressure derivatives evaluated at 298°K are as follows:

Pressure derivatives	dL/dp	dG/dp	dB/dp
$(\partial M^*/\partial p)_T$	6.57 (6.58)	1.79 (1.73)	4.19 (4.27)
$(\partial M^T/\partial p)_T$	6.62	1.79	4.23
$(\partial M^*/\partial p)_s$	6.52	1.73	4.16

The quantities in the parentheses are averaged values calculated from the single-crystal second-order elastic constants and their first pressure derivatives. The experimental data are interpreted with respect to (a) the polycrystalline data calculated from the corresponding single-crystal data, (b) the temperature dependence of the isotropic elastic moduli, (c) the acoustic Grüneisen parameters and their comparison with the corresponding quantities evaluated from thermodynamic properties, (d) the equation of state for alumina, and (e) the Debye temperature as a function of temperature and pressure.

1. INTRODUCTION AND THE SCOPE OF THE PRESENT WORK

Precise data for the equation of state for oxides and silicates are important not only because of the direct interest in the basic properties of these materials, but also because such data furnish guidelines for the extrapolation in both temperature and pressure of data for more complex solids (like rocks). As part of a continuing study of the thermodynamic properties of oxides and silicates at high pressure and high temperature, we report in this paper our experimental results on the isotropic elastic parameters of high-purity, high-

density polycrystalline alumina (α -Al₂O₃). The importance of work of this kind has been indicated earlier.^{1,2}

Much experimental work on the thermal and elastic properties of alumina has already been reported: such thermal properties as expansivity^{3,4} and specific heat⁵ are available for high-purity alumina over a wide range of temperature; the single-crystal second-order elastic

¹ G. Simmons, Proc. IEEE **53**, 1337 (1965).

² D. H. Chung, J. Appl. Phys. **38**, 5104 (1967).

³ J. B. Wachtman, Jr. *et al.*, J. Am. Ceram. Soc. **45**, 319 (1962).

⁴ A. Schauer, Can. J. Phys. **43**, 523 (1965).

⁵ G. R. Furukawa *et al.*, J. Res. Natl. Bur. Std. **57**, 67 (1956).

constants^{6,7} and the corresponding polycrystalline isotropic elastic moduli⁸ have been studied carefully over a limited range of temperature (80°–900°K); Bridgman⁹ measured the isothermal volume compressibility and Hart and Drickamer¹⁰ recently extended his values to higher pressure; shock-wave compressions to about 1500 kbar were made by McQueen and Marsh¹¹ on both single-crystal and sintered polycrystalline aluminas; the dependence of sound velocities on hydrostatic pressure¹² to 4 kbar and that on temperature (300°–1400°K)¹³ on a rather "impure" Lucalox alumina¹⁴ was recently reported.

The determination of the third-order elastic constants of this material in both single-crystal¹⁵ and polycrystalline¹⁶ forms has just begun. Our purpose in presenting this paper is to report the isotropic elastic parameters of polycrystalline alumina as a function of hydrostatic pressure to 10 kbar and also as a function of temperature over the range 4.2°–1300°K, and to (1) compare the polycrystalline data with the corresponding single-crystal data, (2) analyze the explicit temperature dependence of the elastic moduli, (3) calculate the acoustic Grüneisen parameters (γ_G and ψ_G) and compare them with the corresponding thermal Grüneisen parameters, (4) give an equation of state for alumina, and (5) calculate the elastic Debye temperature as a function of temperature and pressure.

2. EXPERIMENTAL PROCEDURE

2.1. Specimens and Material Characterization

Several samples of polycrystalline alumina were fabricated by the hot-pressing procedure described by Crandall *et al.*¹⁷ from 99.95% pure α -Al₂O₃ powder. These same specimens were used in earlier measurements⁸ of the isotropic elastic moduli. In the present

study, we used one of the typical samples. It was cut into two pieces—one cube and one rod. The faces of the cube were ground and polished to optically flat and parallel surfaces. The final size of the specimen was 1.28245×1.28143×1.28194 cm. The rod was commercially ground to the shape of a long cylinder to a final size of 0.63501 cm diam × 8.89250 cm in length. The measured density of the specimens was 3.974 g/cm³ at 300°K, which may be compared with the single-crystal density of corundum (3.986 g/cm³). X-ray studies indicate the specimens were made of corundum crystals and inspection of the electron micrograph shows that the grain diameters range from about one to 15 μ .

The elastic isotropy was checked by measuring both the longitudinal and transverse sound velocities in the cube specimen at room conditions for different directions of wave propagation. The specimen was found to be isotropic. The maximum and minimum velocities for the longitudinal wave were 10.848 and 10.842 km/sec. For the transverse wave, the maximum and minimum velocities were 6.379 and 6.375 km/sec, respectively.

The isotropic elastic moduli were determined with McSkimin's pulse-superposition method.¹⁸ X-cut and Y-cut quartz transducers, $\frac{1}{2}$ in. in diameter with fundamental resonance frequencies near 20 MHz, were used.

In general, it is desirable for a bonding material to adhere to both transducer and specimen over the largest temperature and pressure range and accommodate a differential thermal expansivity between transducer and specimen. For this reason, several bonding materials were used as follows: a phenyl-salicylate at room conditions, Dow Corning resin 276-V9 for measurements under hydrostatic pressure, and between 4.2° and 300°K in the ultrasonic method, Fisher's Nonaq stopcock grease. Variations of the isotropic elastic moduli with temperature were determined by a bar-resonance method¹⁹ (above 77°K) and also by the ultrasonic pulse-superposition method¹⁸ (below 300°K).

2.3. Cryostat and High-Temperature Furnace

In the study of temperature dependence using the ultrasonic method, the specimen was mounted on a flat copper plate which was placed in a copper can covered with Styrofoam insulation. A heater coil, a Honeywell germanium resistance thermometer, and a copper-constantan thermocouple were attached to the plate. Three thin brass rods served as supports for the plate and provided a slight thermal contact with the bath of either liquid nitrogen or liquid helium. A Brown electronic recorder was used to control and measure the temperature of the copper plate and the specimen. To ensure that the system had attained thermal equilibrium, a pause of 20–30 min was necessary whenever the

⁶ J. B. Wachtman, Jr. *et al.*, J. Res. Natl. Bur. Std. **64A**, 213 (1960).

⁷ W. E. Tefft, J. Res. Natl. Bur. Std. **70A**, 277 (1966); and also J. B. Wachtman, Jr. *et al.*, Phys. Rev. **122**, 1754 (1961).

⁸ D. H. Chung, Bull. Ceram. Res. **26**, (297), 1 (1961); J. Appl. Phys. **39**, 2777 (1968).

⁹ P. W. Bridgman, Proc. Am. Acad. Arts Sci. **77**, 187 (1949).

¹⁰ H. V. Hart and H. G. Drickamer, J. Chem. Phys. **43**, 2265 (1965).

¹¹ R. G. McQueen, F. Birch, and S. P. Marsh, in *Handbook of Physical Constants*, S. P. Clark, Jr., Ed. (The Geological Society of America, Inc., New York, 1966), p. 154. More recent data were obtained through personal communications (1968).

¹² E. Schreiber and O. L. Anderson, J. Am. Ceram. Soc. **49**, 184 (1966).

¹³ N. Soga, E. Schreiber, and O. L. Anderson, J. Geophys. Res. **71**, 5315 (1966); see also, N. Soga and O. L. Anderson, J. Am. Ceram. Soc. **49**, 355 (1966).

¹⁴ The nature of Lucalox alumina was described by R. L. Coble in U.S. Patent 3,026,210 (Mar. 1962). As described also by S. K. Roy and R. L. Coble, J. Am. Ceram. Soc. **51**, 1 (1968), the Lucalox alumina contains small amounts of magnesium-aluminate spinel as a major secondary phase.

¹⁵ J. H. Gieske (personal communications, 1967).

¹⁶ D. H. Chung (unpublished).

¹⁷ W. B. Crandall *et al.*, in *Mechanical Properties of Engineering Ceramics*, W. Krieger and H. Palmour, Eds. (Interscience Publishers, Inc., New York, 1961).

¹⁸ H. J. McSkimin, J. Acoust. Soc. Am. **37**, 864 (1965).

¹⁹ S. Spinner and W. E. Tefft, Proc. ASTM **61**, 1221 (1961).