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Acoustic Velocities in Polycrystalline NaCl at 300°K Measured at Static Pressures From 25 to 270 kbar

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The room temperature longitudinal and shear acoustic velocities in polycrystalline NaCl have been measured at static pressures in the range of 25–270 kbar (2.5–27.0 GPa). The measurements were made by ultrasonic interferometry in a variable lateral support Bridgeman anvil device. The velocity data are developed with the aid of the Decker equation of state. Previous measurements of acoustic velocities made in the range of 0–100 kbar are in agreement with the data. At higher pressures the shear mode velocity is approximately constant or slowly increasing with increasing pressure. This is in agreement with predictions made with next nearest neighbor interatomic force calculations. Nearest neighbor only and fourth-order finite strain theory calculations which use the assumption that the elastic parameter c_{44} goes to zero at the 292-kbar phase transition are determined to be inaccurate near the transition. It is suggested that the ratio of the acoustic velocities of polycrystalline NaCl can be used in the future as a parameter for calibration of ultrahigh-pressure devices.

INTRODUCTION

The equation of state of NaCl is used as a primary pressure calibration standard for X ray measurements [*Decker et al.*, 1972]. However, knowledge of the bulk modulus versus pressure for NaCl or any other material is often insufficient for the solution of problems in high-pressure physics. For example, the bulk modulus of rock materials may be insensitive to many phase changes occurring in the earth's interior. Knowledge of the acoustic velocities must be used, but predictions of the acoustic velocities, especially the shear velocities, are fraught with risk [*Anderson*, 1972].

In this paper, measurements of the acoustic velocities of polycrystalline NaCl to 270 kbar (27.0 GPa) are reported. The highest pressure to which the acoustic velocities of NaCl were previously reported is 100 kbar by *Voronov and Grigorèv* [1971] and *Voronov et al.* [1971]. The measurement of acoustic velocities in isotropic materials at very high pressures provides information for verifying the predictions of finite strain continuum and lattice dynamics theories. For example, *Anderson and Demarest* [1971] and *Thomsen* [1972] have predicted that the shear velocity of NaCl decreases or tends to zero near the 292-kbar phase transition.

In order to measure the acoustic velocities of any material under static pressures of more than 100 kbar, it is necessary to use a solid pressure device with a large specimen in comparison with the specimen size used in diamond anvil devices. In a solid system the question of whether the measured physical properties are affected by shear stresses always arises. This problem is minimized in this experiment by the use of a pressure device which balances lateral and horizontal forces actively.

DESCRIPTION OF THE EXPERIMENT

The pressure device used is a variable lateral support Bridgeman anvil device capable of exerting static pressures of up to 400 kbar and has been described by *Kendall et al.* [1975]. The advantage of this design is that an active lateral, as well as vertical, force is exerted upon the specimen. This should result in a much more uniform stress than is achieved when lateral support is provided only by a passive gasket.

The pressure device has been calibrated on the basis of the observation of resistometric changes at phase transitions such as those of bismuth at 25, 74, and 320 kbar [*Kendall et al.*, 1975; *Homan et al.*, 1975*a*] and those of GaP at 220 kbar [*Homan et al.*, 1975*b*] and NaCl near 290 kbar. From these observations a relationship between the specimen pressure and the hydraulic pressure upon the rams of the press (referred to as the 'load') has been developed. In general, the use of the load to specimen pressure relation has an error determined to be less than 15% of the pressure.

All data taken with this pressure device are referred to the 25-kbar pressure level. This is because both resistometric and ultrasonic observations indicate that the change in pressure with respect to the change in load is more erratic from run to run in the range of 0–25 kbar than the data at higher pressures.

The specimen is prepared from reagent grade NaCl. It is ground to a fine powder, baked at 100°C and 140×10^3 dyn/cm^2 air pressure for 3 hours, pressed into a pellet 1.2 mm in height and 4.35 mm in diameter, and stored in a desiccator. Prior to a run the specimen is placed in the pressure device, and a pressure of at least 35 kbar is exerted for several minutes. This insures that the specimen is fully compacted and that the gasket material is set. In addition, all data from the first run or two are disregarded. To check on specimen compaction, the density of a specimen after a series of runs was determined gravimetrically in absolute benzene. It was equal to the theoretical density within the accuracy of the measurement.

It is observed that the zero-pressure thickness of a specimen decreases from one run to the next by a few percent. This is due to extrusion of a small amount of specimen material. Most of this extrusion has been determined to occur between the 0- and 25-kbar pressure levels on the increasing pressure stroke of a run. Thus the assumption of constant specimen mass can be made from 25 kbar to any higher pressure.

For the results of this paper, nine runs are utilized. Only the data obtained on the increasing pressure portion of a run are used. Most runs were terminated at pressures of 130 kbar or less to establish a reliable base line for higher-pressure runs. Of the two highest maximum pressure runs, one run reached a pressure of more than 300 kbar, and the 292-kbar phase change was observed with simultaneous resistance measurements at approximately 290 kbar. The reported ultrasonic data are terminated at 270 kbar because of the onset of a noticeable degradation of the ultrasonic signal at this pressure level. The

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degradation could be interpreted as dispersion in the sample material, perhaps due to individual grains transforming prior to the specimen as a whole.

The apparatus for making the acoustic velocity measurements is shown in Figure 1. A transducer is attached to the force-free surface of the anvil. The anvil acts as a buffer rod between the transducer and the specimen. The ultrasonic signal can be continuously varied from 4 to 16 MHz for data collection. By switching between the longitudinal transducer on one anvil and the shear transducer on the other, effectively simultaneous measurements of both acoustic modes can be made as the load is continuously increased.

Since the specimen thickness is of the order of magnitude of the acoustic wavelength, the most practical method of measuring the acoustic velocities is the technique of ultrasonic interferometry [*Ahrens and Katz*, 1962; *Papadakis*, 1971]. It consists of observing minima in the echoes returning from the specimen-anvil interfaces. Because the acoustic impedance of the anvil material is greater than the impedance in the specimen, acoustic waves traveling in the anvil and reflected off the specimen receive a phase shift of π. Destructive interference in the echo returning to the transducer occurs when there are an integral number of acoustic wavelengths in the specimen. This condition is given as

$$F_n = nv/2d \tag{1}$$

where n is an integer, d is the specimen thickness, v is the acoustic velocity of the specimen material, and F_n is the frequency at which the destructive interference minimum is observed. For an isotropic material there are two acoustic velocities, the longitudinal velocity v_p and the isotropic shear velocity v_s. For each mode there is a set of destructive interference minima. For a nondispersive medium the frequency interval $\Delta f = F_{n+1} - F_n$ is independent of n in each set. The result is a measurable frequency interval for each acoustic mode:

$$\Delta f_p = v_p/2d \qquad \Delta f_s = v_s/2d \tag{2}$$

Use of (2) assumes that the anvil faces are flat and parallel. The lack of 'dishing' or plastic deformation of the anvil faces of less than 2.5×10^{-3} mm after ultrahigh-pressure runs and the clear minima that we obtained indicated that those conditions were adequately maintained.

Method of Data Analysis

In (2) there are three unknowns in the two equations: the velocities v_p and v_s and the specimen thickness d. To solve this system of equations, a third equation relating the bulk modulus to the velocities is used. The relationship for any isotropic material under hydrostatic conditions is [*Truesdell and Noll*, 1965]

$$(1 + \Delta)B_T = B_S = \rho(\partial P/\partial \rho)_S = \rho(v_p^2 - 4v_s^2/3) \tag{3}$$

where B_T and B_S are the isothermal and adiabatic bulk moduli, respectively, and $\Delta = 9\alpha^2 TB_S/\rho C_P$ [*Zemansky*, 1957], where ρ is the density, α is the linear thermal expansion coefficient, T is the temperature (in degrees Kelvin), and C_P is the specific heat at constant pressure.

It is necessary to be concerned with both the isothermal and the adiabatic bulk modulus because static compression is approximately isothermal and the acoustic velocities are the result of infinitesimal adiabatic compressions. The ratio of the bulk moduli is pressure dependent. *Gilmore* [1968] has derived an approximate relationship for the pressure variation of Δ:

$$\Delta/\Delta_0 \simeq (B_S\rho)_0/(B_S\rho) \tag{4}$$

where the subscript 0 denotes evaluation at zero pressure. For NaCl, Δ_0 equals 0.054.

Substitution of (2) into (3) yields a useful form of the bulk modulus velocity relationship:

$$(1 + \Delta)B_T/\rho = 4d^2(\Delta f_p^2 - 4\Delta f_s^2/3) \tag{5}$$

Since the equation of state of NaCl is considered to be well determined [*Decker et al.*, 1972], known values for B_T/ρ can be used in (5) to determine the specimen thickness and the acoustic velocities. Such an analysis of the ultrasonic data has been done, and the results are given in Table 1 and shown in Figures 3 and 4.

For a material whose equation of state is unknown, *Katz and Ahrens* [1963] showed that an equation of state can be solved for by assuming that the geometry of the specimen changes under pressure such that

$$\rho = \rho_0 X^n \tag{6}$$

where

$$X = d_0/d \tag{7}$$

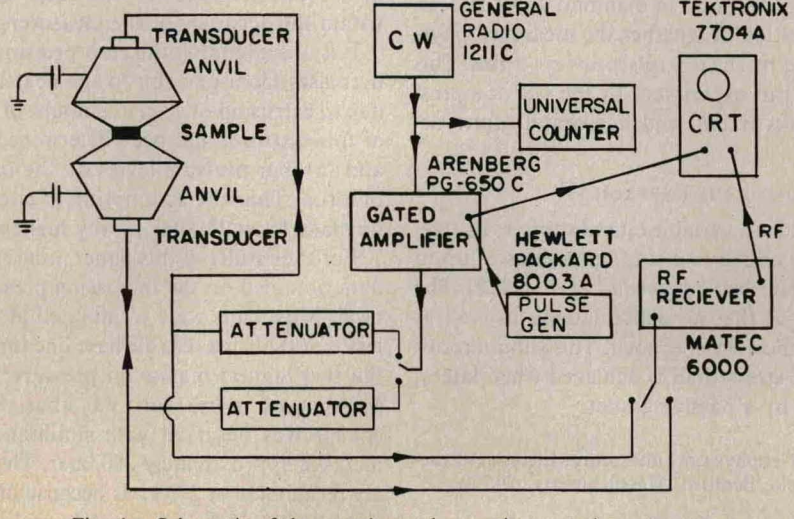

Fig. 1. Schematic of the experimental acoustic measuring apparatus.