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Determination of the Elastic Moduli of a Machinable Ceramic over the Range from Room Temperature to 800°C

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The ultrasonic velocities of a machinable ceramic were measured using the pulse echo overlap technique. The machinable ceramic consists of 5- to 10-μm crystallite blocks of mica in a boroaluminosilicate glass matrix. The elastic moduli are deduced from the sound velocities over the temperature range from room temperature to 800°C. Their temperature change is well described by a fourth-degree polynomial. Although the moduli decrease with increasing temperature, a plateau region appears at about 450°C. This anomalous behavior is explained by applying the simple "rule of mixtures" to constituent materials, the mica crystallites, and the glass matrix.

KEY WORDS: ceramics; elastic moduli; high temperature; pulse echo overlap technique; ultrasonic velocity.

1. INTRODUCTION

Macor is a machinable ceramic using standard working tools and techniques. This ceramic was developed by Corning Glass Works (code 9658) and contains 50–55% fluorophlogopite mica crystallites which are distributed randomly throughout the material. The remaining part is filled with a boroaluminosilicate glass matrix. Because of random orientation and interlocking of these crystals, cracks are localized during machining. As a result of this crack-arrest mechanism, Macor is easily fabricated into particular shapes. Moreover, Macor has good thermal properties, such as a maximum-use temperature of 1000°C, moderate thermal conductivity, and good thermal shock resistance. During the last 10 years, several properties

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of Macor have been investigated. The low-temperature thermal conductivity (1), the specific heat (2), and the elastic moduli (3, 4) were reported. For structural applications at high temperature, a knowledge of the elastic moduli is required, which has not yet been sufficiently investigated. The present paper reports precise measurements of the elastic moduli over the temperature range from room temperature to 800°C. The data provide useful information for high-temperature applications.

2. EXPERIMENTAL METHOD

The elastic moduli were calculated from the velocities of longitudinal and shear ultrasonic waves with a 5-MHz frequency. The echo overlapping method to measure the ultrasonic velocities (Matec 6600 RF pulse generator, 755 RF plug-in) was used. A specimen used for room-temperature measurements is disk shaped with a 30-mm diameter and 20 mm thick. A transducer of X-cut or Y-cut quartz was attached to the plane face of the specimen. A glue joint between the transducer and the specimen was made with phenyl salicylate, which is generally used for ultrasonic applications at room temperature and has a melting point of 43°C. An apparatus for high-temperature experiments is shown schematically in Fig. 1. In this case a stepped specimen [5] is used, taking advantage of its machinability. Thus, bonding problems at high temperatures can be avoided. The transducer was bonded to the room-temperature end of a 20-cm-long portion 2 cm in diameter. Screw threads were cut to prevent mode conversions at the walls. A stepped portion 2 cm long and 1 cm in diameter was held at high temperatures in a furnace. Compressed air was used to keep the transducer and the glue joint at a constant temperature. Temperature distribution at the test zone was uniform within the range of

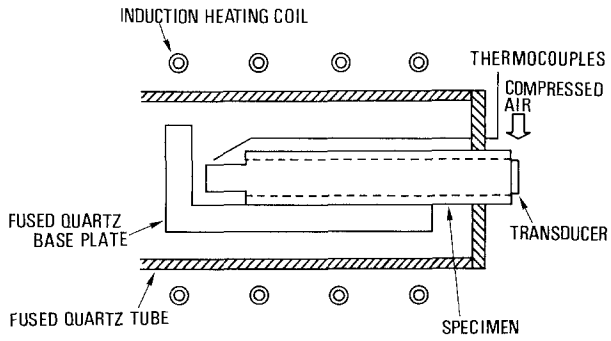

Fig. 1. Ultrasonic test specimen and furnace assembly.

$\pm 5°$ at 800°C, which was monitored with an accuracy of ± 0.2°C by two calibrated R-type thermocouples located near the specimen. Since ultrasonic waves are reflected both from the step and from the end of the specimen, measurements of the time interval between these two echoes allow calculation of the sound velocities. The velocity is expressed by

$$V = V_0 \frac{t_0}{t}(1 + \alpha \Delta T) \tag{1}$$

where V_0 is the velocity and t_0 is the time interval between two echoes at room temperature, V and t are the corresponding values at the measuring temperature, ΔT is the temperature difference, and α is the thermal expansion coefficient.

Before presenting the experimental results, the accuracy of the present measurements has been investigated. Determination of ultrasonic velocities requires measurements of the specimen length and the echo round-trip time. The specimen length was measured with a micrometer and its associated error is less than 3 parts in 10^4. The errors in measuring trip time are related to the overlap process, the bond thickness, diffraction, and instability in the electronics [6]. The error from the overlap process and instability in the electronics are averaged by repeating measurements several times and are within 5 parts in 10^4. McSkimin [7] pointed out the phase shift associated with bond thickness. According to his technique, the error due to this effect is within 4 parts in 10^4. Further, Papadakis [8] has calculated the diffraction phase shift and presented a correction curve. With this curve the error of the diffraction phase shift is estimated to be less than 3 parts in 10^5. Adding up these errors statistically as standard deviations, the present method ensures an accuracy of 0.07%.

3. RESULTS AND DISCUSSION

The ultrasonic velocities and elastic moduli at room temperature are presented in Table I, where V_l and V_t are the longitudinal and shear

Table I. The Sound Velocities and Elastic Moduli at Room Temperature

	V_l (m·s^{-1})	V_t (m·s^{-1})	E (GPa)	G (GPa)
Present work	5580	3179	64.16	25.46
Corning	—	—	64.1	25.5
Harrington et al. [3]	—	—	63.91	25.39

velocity, respectively. The quantities E and G are the Young's modulus and shear modulus, respectively. In the calculation of the elastic moduli, the density, 2.52 $g \cdot cm^{-3}$ at 4°C, as well as the thermal expansion coefficient was taken from the Corning value. The Young's modulus and shear modulus at room temperature are reported by several investigators. These values are also listed in Table I. The measured Young's modulus agrees to within 0.09% of the Corning value and 0.39% of Harrington et al. [3]. The shear modulus is also in agreement to within 0.16% of Corning and 0.27% of Harrington et al.

Figure 2 shows the Young's modulus and the shear modulus during heating and cooling from room temperature to 800°C. Each point is the mean value of determinations on two samples. The discrepancies between two samples were to be 0.5% or less. A small hysteresis is observed and this effect is discussed later. The elastic moduli at low temperatures were also calculated from Ref. 3 as shown in Fig. 2. For convenience and practical use, a least-squares fit was made of the experimental results. It was found that a fourth-degree polynomial is necessary to describe the tem-

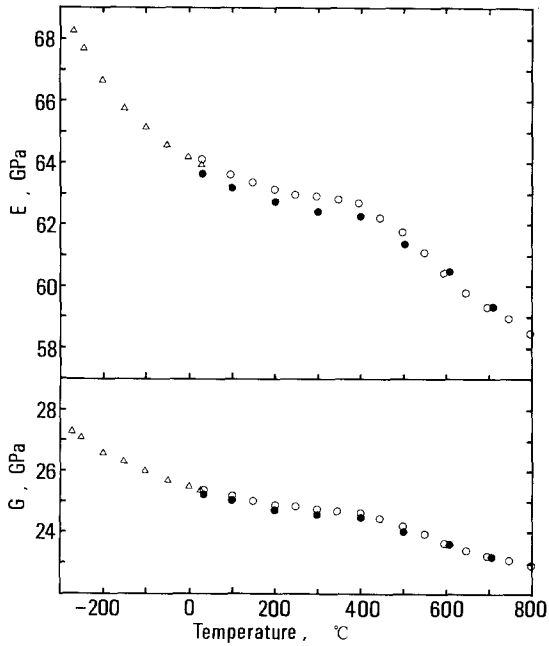

Fig. 2. Temperature dependence of the elastic moduli of Macor. (○) Present work during heating; (△) data from Ref. 3; (●) present work during cooling.

perature dependence of the averaged modulus of heating and cooling measurements.

$$E \text{ (or } G) = \sum_{n=0}^{4} a_n T^n \tag{2}$$

The coefficients of polynomials are given in Table II. The deviation of elastic modulus values from the smooth function defined by Eq. (2) is about 0.2% or less.

Having determined the elastic moduli, we now turn to a discussion of their behavior under varying temperature conditions. Usually, the elastic moduli of ceramics decrease monotonically with increasing temperature. As can be seen from Fig. 2, the elastic moduli of Macor do not follow this pattern. The absolute values of their temperature derivatives decrease with increasing temperature, reaching a minimum value at 450°C. Above 450°C, the moduli begin to decrease more rapidly. In order to understand this anomaly, it is necessary to know separately the temperature dependence of the elastic moduli of mica and glass phases. As this is practically difficult, we decided, instead, to investigate the elastic properties of Mica Ceramic. Mica Ceramic is a trademark for a machinable ceramic developed by NGK Spark Plug Corp, in Japan. The ceramic is sintered from pure, randomly oriented mica crystallites. Consequently, the material is virtually isotropic. The longitudinal and shear velocities of Mica Ceramic were measured and the elastic moduli were deduced. The Young's modulus and shear modulus of Mica Ceramic are presented in Fig. 3 as a function of temperature. The moduli of Mica Ceramic show a continuous decrease on heating and no anomalous behavior is observed up to about 450°C. The density of Mica Ceramic, $2.39\ \mathrm{g \cdot cm^{-3}}$, is about 16% less than that of mica crystal, suggesting that there exists a porous region. Although the elastic moduli are sensitive to the porosity, their relative changes with temperature may

Table II. The Coefficients of Polynomials for the Young's Modulus (E) and Shear Modulus (G)

	E (GPa)	G (GPa)
a_0 (GPa)	6.833×10^{1}	2.741×10^{1}
a_1 (GPa · K^{-1})	-2.650×10^{-2}	-1.136×10^{-2}
a_2 (GPa · K^{-2})	5.435×10^{-5}	2.069×10^{-5}
a_3 (GPa · K^{-3})	-5.406×10^{-8}	-1.982×10^{-8}
a_4 (GPa · K^{-4})	1.705×10^{-11}	6.258×10^{-12}

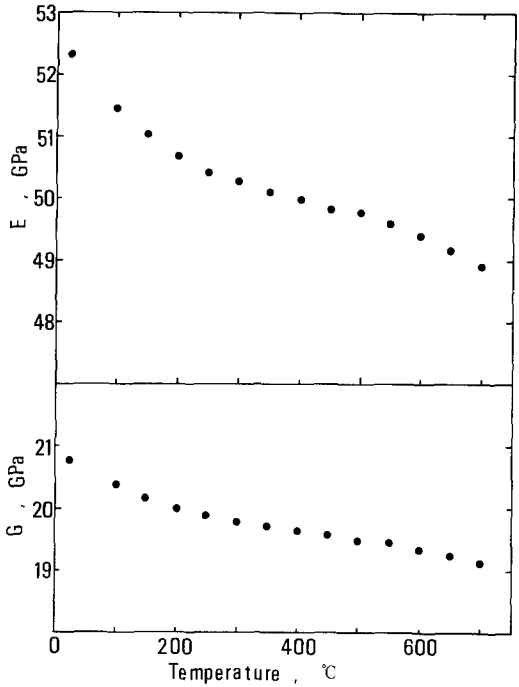

Fig. 3. Temperature dependence of the elastic moduli of Mica Ceramic.

not be affected by this factor. Therefore, it was assumed that the elastic moduli of the mica phase in Macor would change with temperature the same way as in Mica Ceramic but their absolute value is 16% higher than that.

The elastic moduli of the glass phase in Macor were determined by applying a simple rule of mixtures, where the two-phase modulus is given by the sum of the moduli weighted by the respective volume fraction as follows:

$$C = V_f C_f + (1 - V_f) C_m \tag{3}$$

where C is the elastic modulus of Macor, C_f and C_m are those of the mica and glass phases in Macor, and V_f is the volume fraction of the mica phase in Macor. In the present work, we choose 0.5 as the V_f value. This selected value is not critical to the following results. The calculated elastic moduli of the glass phase are shown in Fig. 4 and compared with those of the typical boroaluminosilicate glass, Pyrex glass (Corning code 7740) [9]. Although Eq. (3) ignores the interaction between the mica phase and the glass phase,

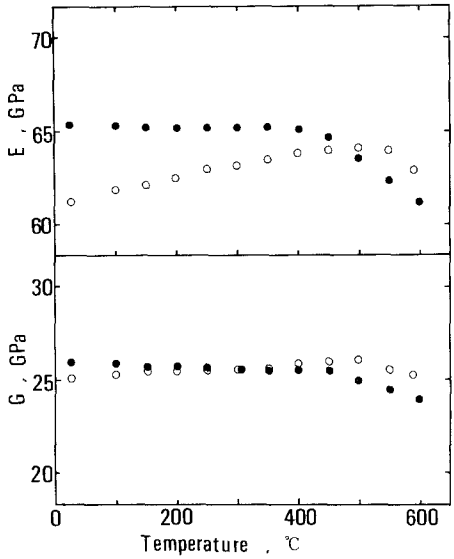

Fig. 4. Temperature dependence of the elastic moduli of the glass phase in Macor and Pyrex glass. (●) Glass phase in Macor; (○) Pyrex glass.

the calculated elastic moduli of the glass phase show behavior similar to that of Pyrex. In particular, both curves show a rapid decrease above 450–500°C. This experimental agreement supports the assumption that the simple rule of mixtures is applicable to Macor and that a rapid decrease in the elastic moduli of Macor at high temperature is due to softening of the glass phase. Since the mica and glass phases show the opposite temperature dependence, the role of the glass matrix in the thermoelastic properties increases with increasing temperature, which is in contrast to the low-temperature case, where the mica crystallites are a dominant factor [10]. Finally, the influence of microcracks on the thermoelastic properties of Macor should be noted. Some polycrystalline ceramics show an anomalous elastic behavior, i.e., the elastic moduli show a hysteresis between heating and cooling. This hysteresis effect is attributed to reversible fracturing and healing of microcracks arising from the thermal expansion anisotropy [11]. In Macor, the thermal expansion coefficient of mica crystallites is considerably different from that of a glass matrix, which might lead to nucleation of microcracks. Thus, the hysteresis effect, observed in Fig. 2, can be qualitatively explained by this process. However, the change of the moduli between heating and cooling is only 0.9%, relatively small com-

pared with that of polycrystalline ceramics such as Nb_2O_5, magnesium dititanate, and β-eucryptite [11–13]. Judging from this result, the contribution of microcracks to the thermoelastic properties may be limited.

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