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Ultrasonic properties of fluorosilicate glass-ceramics at cryogenic temperatures

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The longitudinal ultrasonic wave velocity and attenuation, and shear wave velocity for machinable fluorosilicate glass-ceramic (MACOR) samples (regular shelf stock, annealed to 900°C, and 1000°C for 10 hrs) were measured as the functions of frequency and temperature with the pulse-echo method. The frequency and temperature range used were 5 MHz to 50 MHz, and 100 K to 300 K respectively. Both velocities for three samples decrease linearly with temperature, and the ultrasonic attenuation increases linearly with the frequency square, and decreases monotonically with increasing temperature. These data were used to calculate the elastic moduli, Lame parameter, and Poisson's ratio for the materials.

1. Introduction

MACOR is machinable with ordinary steel or carbide tools, it is a good thermal and electrical insulator and it has a density of 2.52 g/cm^3 [1]. It has been extensively used in aerospace and other commercial applications. Since the mechanical performance of a material as a structural element is often critical to the success of a design, it is essential to fully characterize the response of the material to vibration and stress-wave propagation. There is no complete set of mechanical properties data for MACOR in the literature. This study addresses this absence and examines likely physical/mechanical processes operating within MACOR.

The ultrasonic properties of MACOR were studied in this work to determine the elastic and anelastic parameters of the material.

Mechanical properties of glass-ceramics depend upon both composition and microstructure [2]. The bulk-oxide composition of stock MACOR is 46% SiO_2, 16% Al_2O_3, 17% MgO, 10% K_2O, 7% B_2O and 4% F [1]. We annealed MACOR at high temperature to modify its texture. Ultrasonic wave velocities and attenuation were then measured as functions of frequency and temperature (100 K–300 K) and related to the microstructural effects of annealing.

Reported here are the longitudinal-wave attenuation and velocity and the shear-wave velocity for the MACOR samples as functions of frequency and temperature. The elastic moduli, Lame constant (C_{12} in elastic constant matrix for isotropic body), and Poisson's ratio [3, 4] were calculated from the velocities.

2. Experimental procedure

The pulse-echo technique was used to acquire the ultrasonic data [5]. The schematic diagram of the experimental setup is shown in Fig. 1. A Matec MBS-8000 system was employed to produce and detect the pulses. The phase coherent pulse is generated using a continuous wave signal generator and a gated amplifier. It is applied to the ultrasonic transducer through an impedance-matching circuit. The transducer generates and receives the ultrasonic signals that pass through the specimen. The received ultrasonic signals are detected and recorded with an oscilloscope and the echo envelope is matched to an exponential curve. The amplitude loss and time interval between echoes were measured to determine the attenuation coefficient (db/cm) and velocity of the ultrasonic waves. Procedural details of the experiment can be found elsewhere [6].

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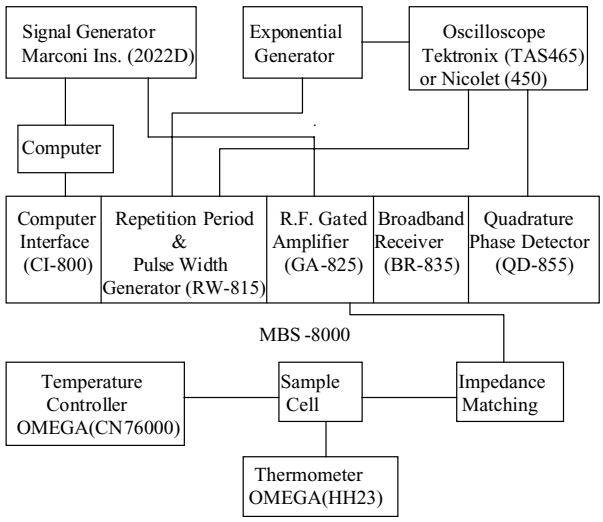

Figure 1 Schematic diagram of experimental setup.

The samples were 2.5 cm in diameter and approximately 1 cm thick. Final polishing was done in 0.05 micron alumina. Lithium niobate transducers (for both longitudinal and shear modes), having 5 MHz and 10 MHz fundamental frequencies, were bonded on the sample surface using nonaqueous stopcock grease. Diffraction corrections to wave amplitude were applied to the 5 MHz to 30 MHz. Diffraction effects were insignificant at frequencies above 30 MHz in this study [7, 8]. All data presented here are the result of averaging several data values.

During the annealing process, the temperature was increased at a rate of approximately 7 K/min and decreased at 15 K/min. The original samples (sample S1) were annealed to 900°C (sample S2) and 1000°C (sample S3) for 10 hrs, respectively. Cryostat temperature was controlled to ± 0.1 K. The error in the velocity is at most 0.5%, and that in attenuation is less than 8%.

Samples of the test materials were prepared for analysis with scanning electron microscopy (SEM). After grinding, samples were polished with 5-micron paste. Light etching in HF was applied to bring out the contrast between the grains and glass pockets and to highlight textural morphology. A JEOL 1200 SEM was used with a 30 kV beam to produce 1000× micrographs. Grain size was determined from the micrographs via a transect method [9]. Each grain size represents an average of over a 100 transect/grain crossings.

3. Results

Scanning electron micrographs for three samples are shown in Fig. 2. These micrographs indicate the grain boundary and grain size alteration that occurred with different degrees of annealing. A higher annealing temperature yields larger average grain sizes. Such "ripening" is common in silicates and is due to surface energy effects [10]. The average grain sizes were estimated at 5.7 micron for S1, 9.8 micron for S2, and 11.1 micron for S3. Clearly noticeable in the micrographs are the high porosity and surface area in these samples.

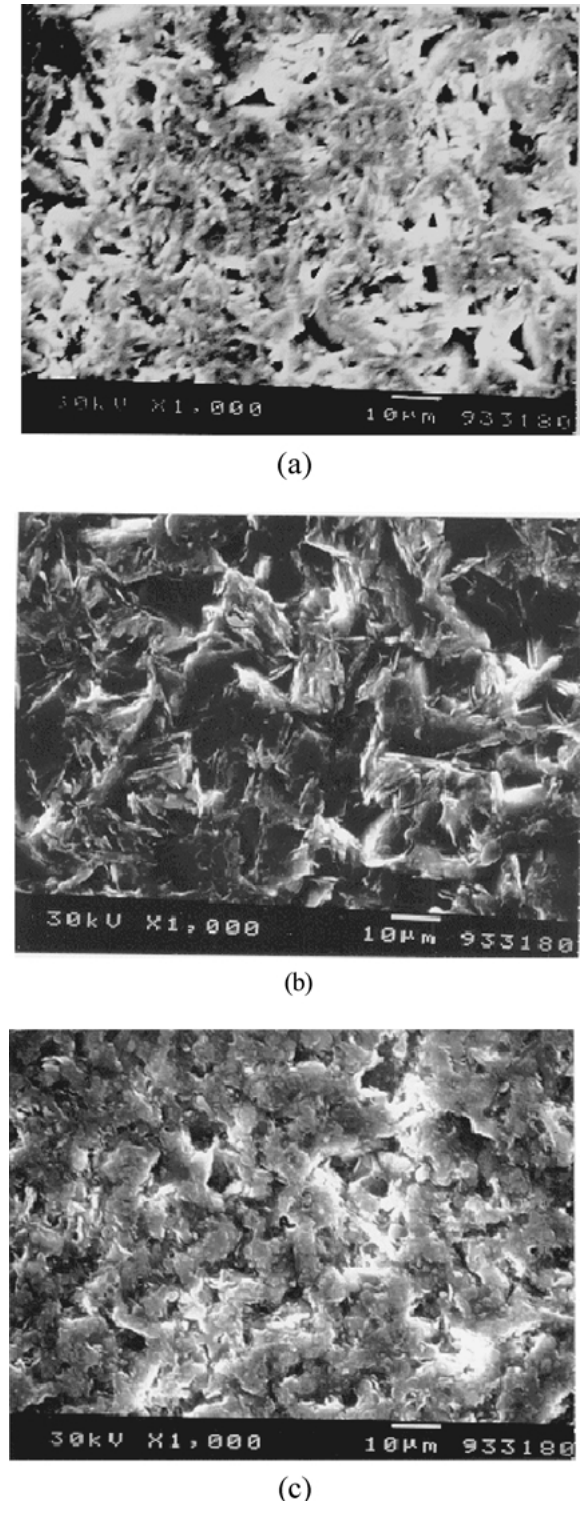

Figure 2 Scanning electron micrograph for three samples: (a) original sample without annealing (S1), (b) sample annealed at 900°C for 10 hrs (S2), and (c) sample annealed at 1000°C for 10 hrs (S3).

The cryogenic temperature dependence of the longitudinal and shear-wave velocities at 10 MHz are shown in Fig. 3. The error bars indicate the maximum estimated measurements errors calculated from the several measurements of the data points. Both velocities for three samples decrease linearly with increasing temperature. The temperature dependence of velocity between 100 K and 300 K for fluorosilicate glass-ceramics were quite small: about 1% for longitudinal waves and 2% for shear waves. This indicates that elastic properties

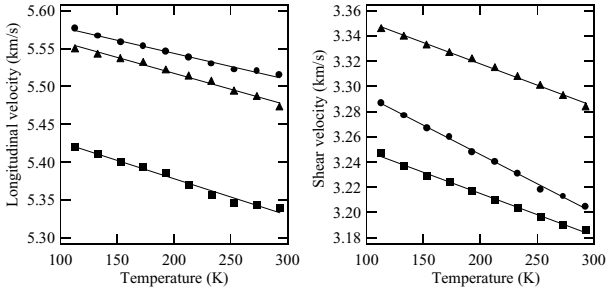

Figure 3 Temperature dependence of longitudinal and shear wave velocities at 10 MHz where ●, ■ and ▲ indicate S1, S2 and S3 throughout this paper.

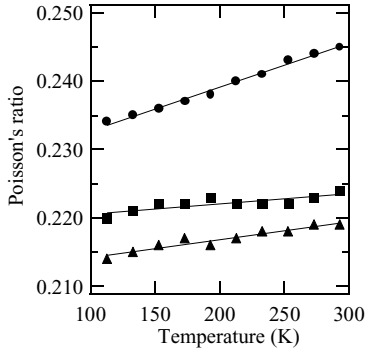

Figure 4 Poisson's ratio as a function of temperature.

do not change appreciably for this temperature. For the longitudinal wave velocity, S2 has approximately 3% lower velocity compared to S1 and S3. For the shear wave velocity, it is observed that S3 have the largest velocity and that the velocity difference between S2 and S3 was about 3%. Overall there is little variation in the temperature dependence of the longitudinal and shear wave velocities over the temperature range studied.

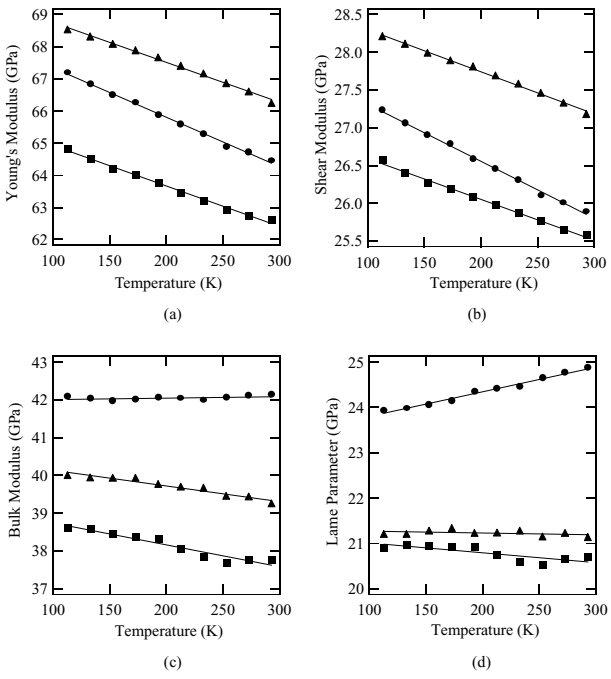

Figure 5 Elastic moduli determined from longitudinal and shear velocity data as a function of temperature.

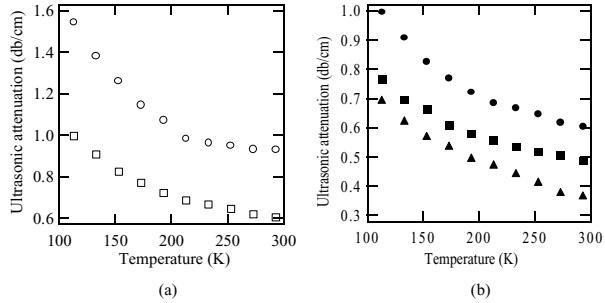

Figure 6 Temperature dependence of ultrasonic attenuation: (a) for S1 at 10 MHz (□) and 15 MHz (○) and (b) for three samples at 10 MHz.

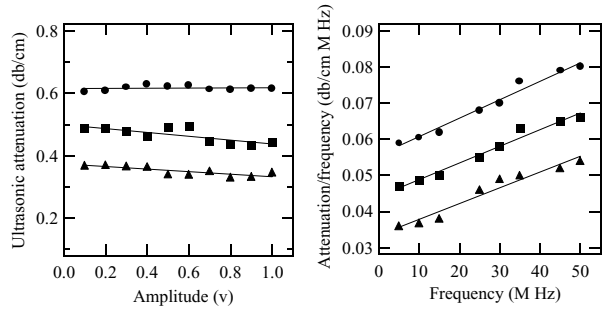

Figure 7 Ultrasonic attenuation as a function of pulse amplitude and attenuation coefficient/frequency as a function of frequency.

In the temperature range of 100 K to 300 K, Poisson's ratios for all samples are between 0.21 and 0.25. Elastic moduli determined from the longitudinal and shear velocity data as a function of temperature are shown in Fig. 5. It is observed that elastic moduli for all annealed samples decrease linearly with increasing temperature. The effect of annealing is even more striking in the Lame parameter. Only for the unannealed sample (S1) is there a significant increase in it with measurement temperature.

The temperature dependence of ultrasonic attenuation for S1 at 10 MHz and 15 MHz, and for three samples at 10 MHz is shown in Fig. 6. These results show that as the temperature increases the attenuation coefficients for three samples decrease monotonically. Furthermore, the attenuation coefficient for all samples varies with the square of frequency and is independent of pulse amplitude, implying that the attenuation is due to a linear loss mechanism.

4. Discussion and conclusions

When annealed at 900°C, the temperature dependence of the bulk modulus is no longer constant, but decreases as the temperature is increased. This decrease is also observed for the two moduli-Young's and shear moduli. Therefore, the heating process in this annealed material lowers all elastic moduli: Young's, shear, and bulk moduli. For both annealed and unannealed samples, the shear modulus is the most strongly influenced and the bulk modulus is the least affected by temperature changes. Annealing to 900°C lowers elastic parameters at the higher temperature environment. However, higher annealing (at 1000°C for 10 hrs) raises both velocities. Although the exact micro-mechanical

processes responsible for this behavior are not known, these results are consistent with thermal microcracking associated with the annealing and some crack "healing" (sintering) at the higher anneal temperature. The higher temperature annealing seems to rearrange the ceramic microstructure to increase Young's and shear moduli, but not the bulk modulus.

The monotonic decrease in the longitudinal attenuation coefficient between 100 K and 300 K was approximately 65% for S1, 60% for S2 and 90% for S3 at 10 MHz. In addition, it is observed that the ultrasonic attenuation at higher frequency has more temperature dependence at low temperature. The longitudinal wave attenuation coefficient divided by frequency increases linearly with increasing frequency and the frequency dependent slope decreases with increasing the annealing temperature. It suggests that the ultrasonic attenuation decreases with increasing grain sizes because the average grain sizes distribution increases as the annealing temperature increases. Since the attenuation coefficient is proportional to the square of frequency and it is independent of pulse amplitude, a possible cause of ultrasonic attenuation may be a linear structural relaxation.

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