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Elastic properties of glasses and glass ceramics at high pressures and high temperatures

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Figures 5.4.7, 5.4.8, 5.4.9 and 5.4.10 show the pressure variation at the three different temperatures for Young's modulus, the Lamé constant, Poisson's ratio and the bulk modulus, respectively. It is interesting to note that $(\partial B/\partial P)_T$ is positive but $(\partial C_{44}/\partial P)_T$ is negative. The implications of this will be discussed below. Table 5.4.2 is a summary of the polynomial coefficients corresponding to each of the properties at the three temperatures. The table includes also the result of the volume variation (V/V_0) of the glass versus pressure as deduced from the pressure dependence of the elastic constants. This graph is shown in figure 5.4.11.

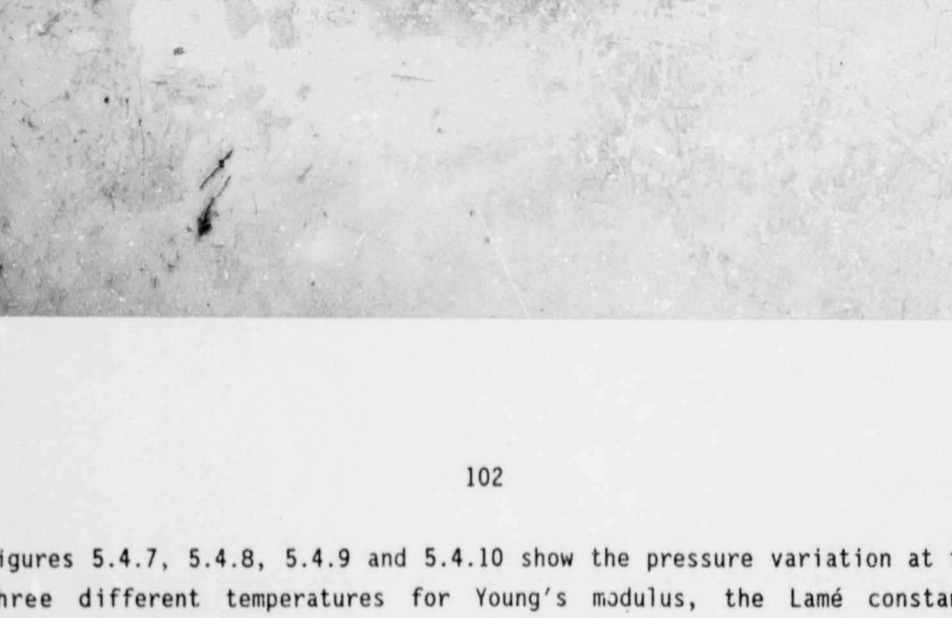


Fig. 5.4.7 119 MCY glass: Young's modulus vs pressure

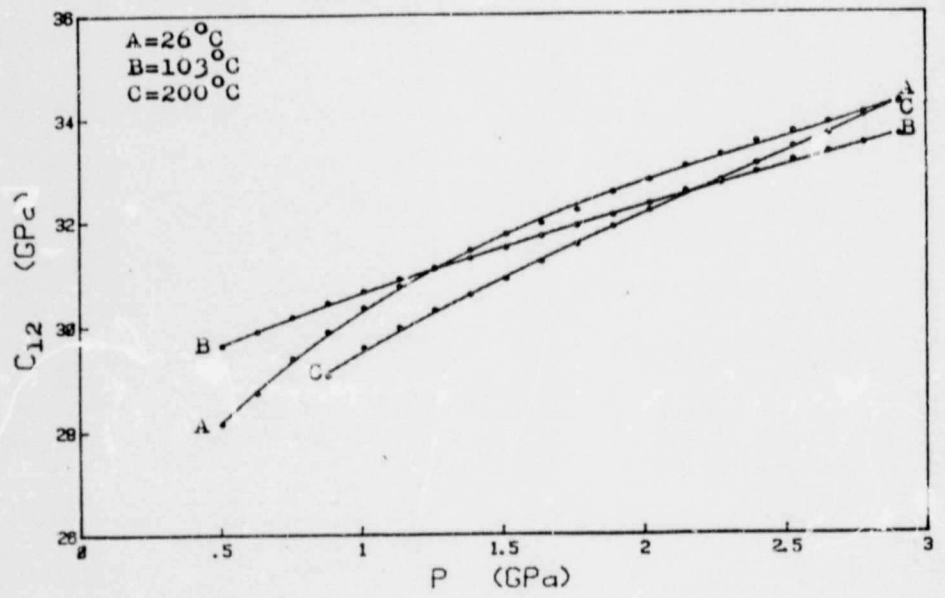


Fig. 5.4.8 119 MCY glass: Lamé constant vs pressure

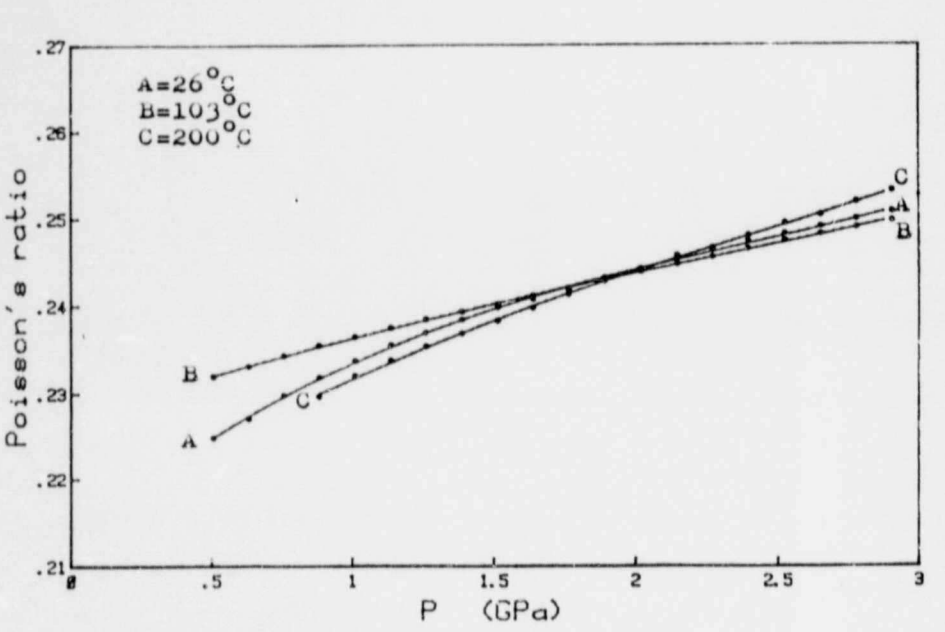


Fig. 5.4.9 119 MCY glass: Poisson's ratio vs pressure

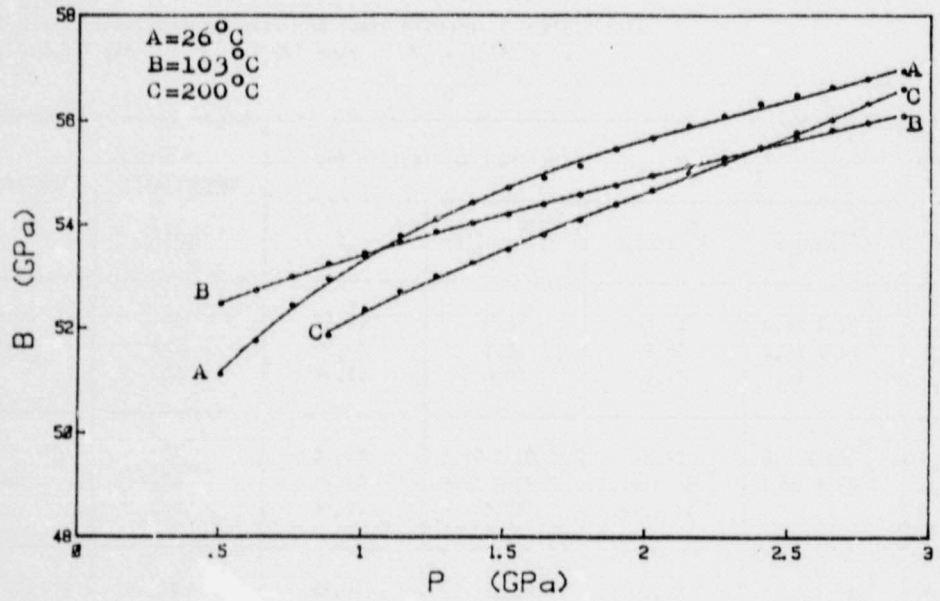


Fig. 5.4.10 119 MCY glass: Bulk modulus vs pressure

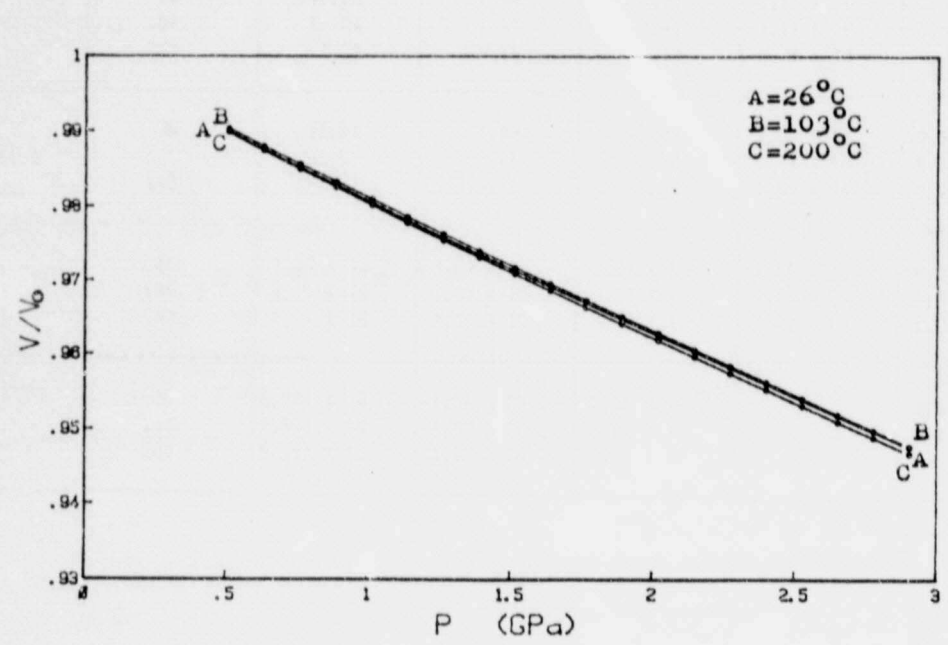


Fig. 5.4.11 119 MCY glass: Volume variation vs pressure

Table 5.4.2
119 MCY Glass: Elastic Properties and their Polynomial Coefficients
Density = 2,577 g/cc v_ℓ = 6,241 km/s v_t = 3,642 km/s

ELASTIC CONSTANT	TEMPERATURE (°C)	COEFFICIENTS OF EQUATION M = $a_0 + a_1P + a_2P^2 + a_3P^3$				X ≤ P ≤ Y(GPa)	
		a_0	$a_1(GPa^{-1})$	$a_2(GPa^{-2})$	$a_3(GPa^{-3})$	X	Y
C_{11}(GPa)	26	93,65	8,39	-3,13	$4,43 \times 10^{-1}$	0,5	3
	103	97,05	2,34	-0,48	$5,24 \times 10^{-2}$	0,5	3
	200	94,81	3,95	-1,15	0,19	0,9	3
C_{44}(GPa)	26	34,23	$6,97 \times 10^{-1}$	-0,45	$6,7 \times 10^{-2}$	0,5	3
	103	34,27	$-5,22 \times 10^{-2}$	$-5,14 \times 10^{-2}$	$5,54 \times 10^{-4}$	0,5	3
	200	34,46	-0,39	$1,66 \times 10^{-2}$		0,9	3
E (GPa)	26	83,02	3,66	-1,72	$2,47 \times 10^{-1}$	0,5	3
	103	84,11	0,60	-0,24	$1,70 \times 10^{-2}$	0,5	3
	200	83,94	0,54	-0,42	$7,83 \times 10^{-2}$	0,9	3
B (GPa)	26	48,00	7,46	-2,53	$3,53 \times 10^{-1}$	0,5	3
	103	51,36	2,41	-0,41	$5,17 \times 10^{-2}$	0,5	3
	200	48,70	4,50	-1,12	$1,73 \times 10^{-1}$	0,9	3
C_{12}(GPa)	26	25,18	6,99	-2,22	$3,09 \times 10^{-1}$	0,5	3
	103	28,51	2,44	-0,38	$5,13 \times 10^{-2}$	0,5	3
	200	25,64	4,78	-1,10	$1,65 \times 10^{-1}$	0,9	3
σ	26	$2,13 \times 10^{-1}$	$2,82 \times 10^{-2}$	$-0,87 \times 10^{-2}$	$1,20 \times 10^{-3}$	0,5	3
	103	$2,27 \times 10^{-1}$	$1,06 \times 10^{-2}$	$-0,17 \times 10^{-2}$	$2,37 \times 10^{-4}$	0,5	3
	200	$2,14 \times 10^{-1}$	$2,26 \times 10^{-2}$	$-0,05 \times 10^{-2}$	$6,79 \times 10^{-4}$	0,9	3
V/V_o	26	$9,995 \times 10^{-1}$	$-1,94 \times 10^{-2}$	$5,14 \times 10^{-4}$		0,5	3
	103	$9,999 \times 10^{-1}$	$-1,92 \times 10^{-2}$	$4,06 \times 10^{-4}$		0,5	3
	200	$9,997 \times 10^{-1}$	$-1,99 \times 10^{-2}$	$5,65 \times 10^{-4}$		0,5	3

For the 119 MCY glass, at three different increasing temperatures, the values for $(\partial B/\partial P)_{P=0}$ and $(\partial C_{44}/\partial P)_{P=0}$ are 7.46, 2.41, 4.50 and 0.7, -0.05, -0.39, respectively. It is strange that there is no trend in these derivatives but the measurements were made as accurately as possible. According to the work done by Manghnani (1970) $\partial B/\partial P$ and $\partial C_{44}/\partial P$ for a 65 % SiO_2-content glass are 3.6 and 0.64, respectively (see figure 5.3.7), compared to 7.46 and 0.7 for 119 MCY glass. Since the results compare with the soda-lime - silica and iron phosphate glasses discussed previously in chapter 1 where $(\partial B/\partial P)_T$ is strongly positive and $(\partial C_{44}/\partial P)_T$ is just negative, it is possible that this glass has an open structure.

Ota et al. (1979) reported on measurements on the SiO_2-Na_2O system which revealed that at low concentrations of sodium oxide (<15 mole %) both $\partial B/\partial P$ and $\partial C_{44}/\partial P$ are negative but at high concentration (>25 mole %) both these values are positive. At this composition, SiO_2-Na_2O glass is no more anomalous. At an alkali concentration of 15 to 25 mole % there is an "intermediate" glass in which $\partial B/\partial P$ is positive, but $\partial C_{44}/\partial P$ is still negative. Judged from the signs of $\partial B/\partial P$ and $\partial C_{44}/\partial P$ alone, one might be able to state that glasses GeO_2, SiO_2-Na_2O (Na_2O = 15 to 25 mole %) and B_2O_3-Na_2O (Na_2O = 10 to 25 mole %) are equivalent in terms of the elastic anomaly. If so, B_2O_3-Na_2O glass with a negative $\partial C_{44}/\partial P$ is in a transition state between the elastically anomalous state and the normal state. Regarding the signs of these derivatives for 119 MCY glass it might also be in this transition state, although the Na_2O is present as a concentration of only 9.9 mole %. The situation where $\partial C_{44}/\partial P$ is negative and $\partial B/\partial P$ is positive has been reported for polycrystallines ZnO and KCl as well. Although these properties look similar in glasses and in crystalline phases, this mechanism may be different between these two phases.

5.5 119 MCY glass ceramic

This material was tested at 19 °C, 103 °C and 204 °C to be close to those of the corresponding glass. Figure 5.5.1 shows the longitudinal frequency versus pressure and figure 5.5.2 shows the shear frequency variation with pressure. This glass ceramic, a partly crystalline material, behaves elastically "normal" like a crystalline material in that the pressure derivatives are positive and it is clear that the modes soften with increase in temperature.

An unusual curvature is observed for the shear mode, although it is repeatable for the different temperatures. Figures 5.5.3, 5.5.4, 5.5.5 and 5.5.6 show the longitudinal modulus, shear modulus, C_{11} and C_{44} respectively as functions of pressure.

Table 5.5.1 is a summary of the elastic properties and their polynomial coefficients. The pressure variation of Young's modulus, the Lamé constant, Poisson's ratio and the bulk modulus are shown in figures 5.5.7-10. The volume variation as derived from the bulk modulus' variation with pressure is displayed in figure 5.5.11. V/V_o versus pressure for the three isotherms produces the same graph.

The glass ceram compressible with pressure than the glass. The density, longitudinal and shear wave velocities as well as the elastic constants are higher than the values for the corresponding glass, thus the properties of the glass have been improved by producing the glass ceramic.

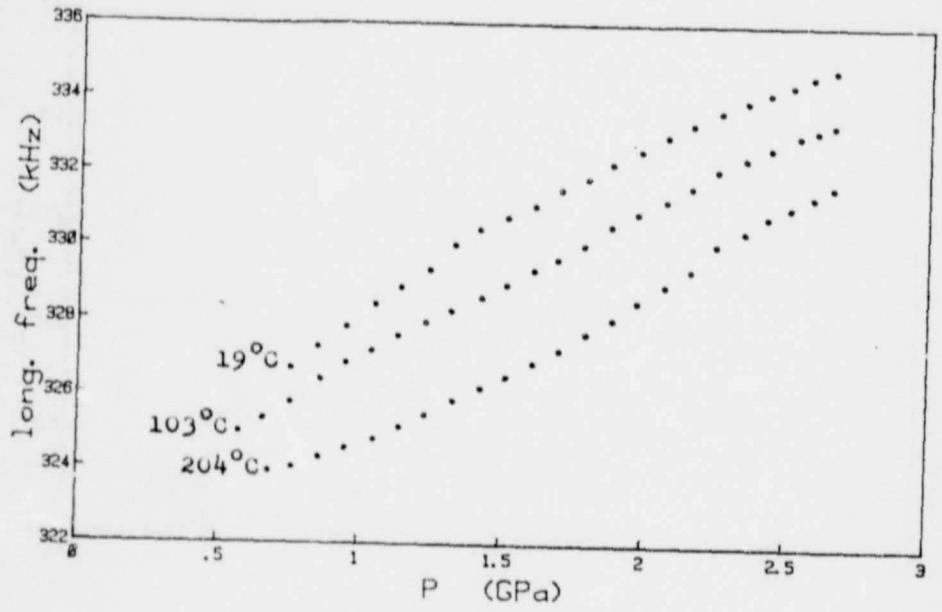


Fig. 5.5.1 119 MCY GC: Longitudinal frequency vs pressure at temperature

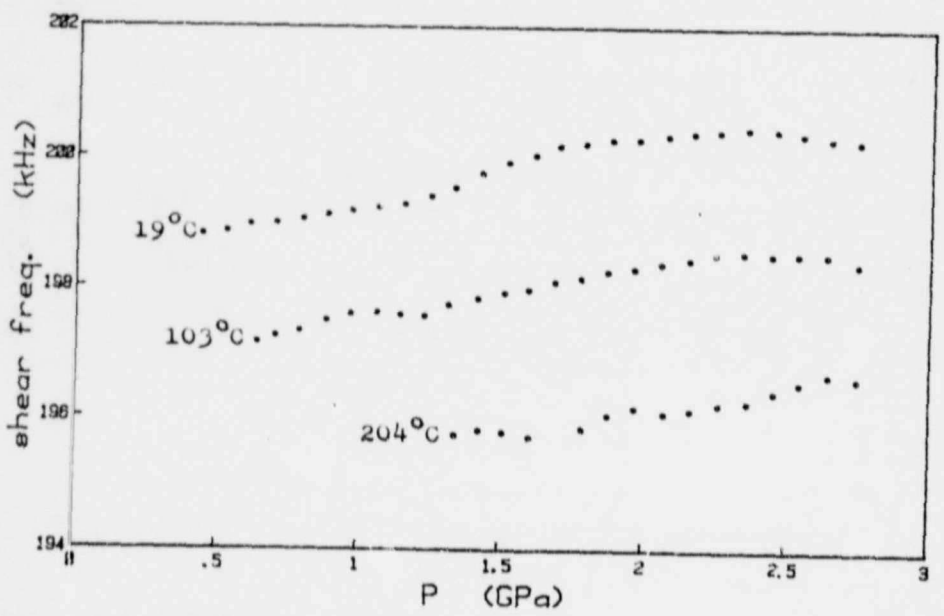


Fig. 5.5.2 119 MCY GC: Shear frequency vs pressure at temperature

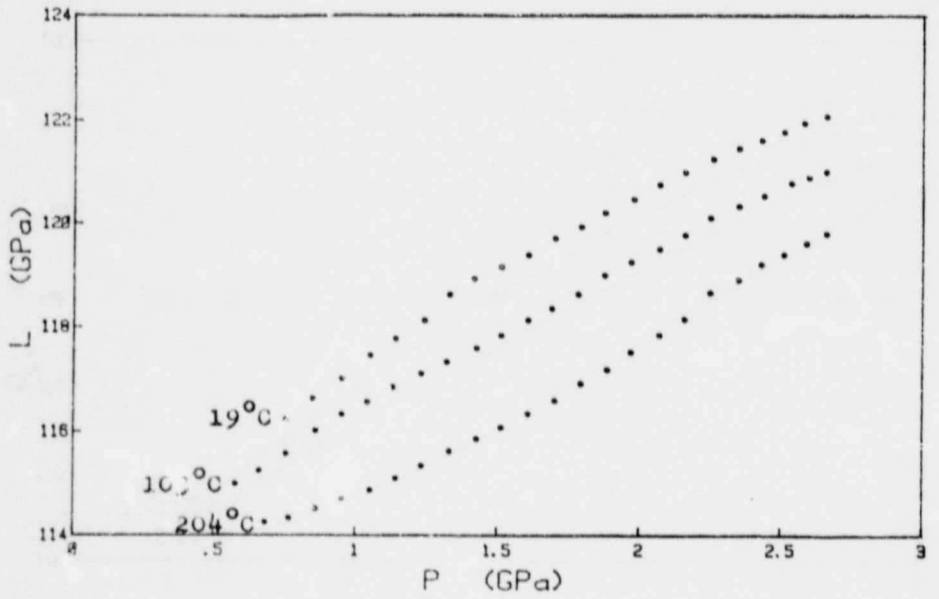


Fig. 5.5.3 119 MCY GC: Longitudinal modulus vs pressure

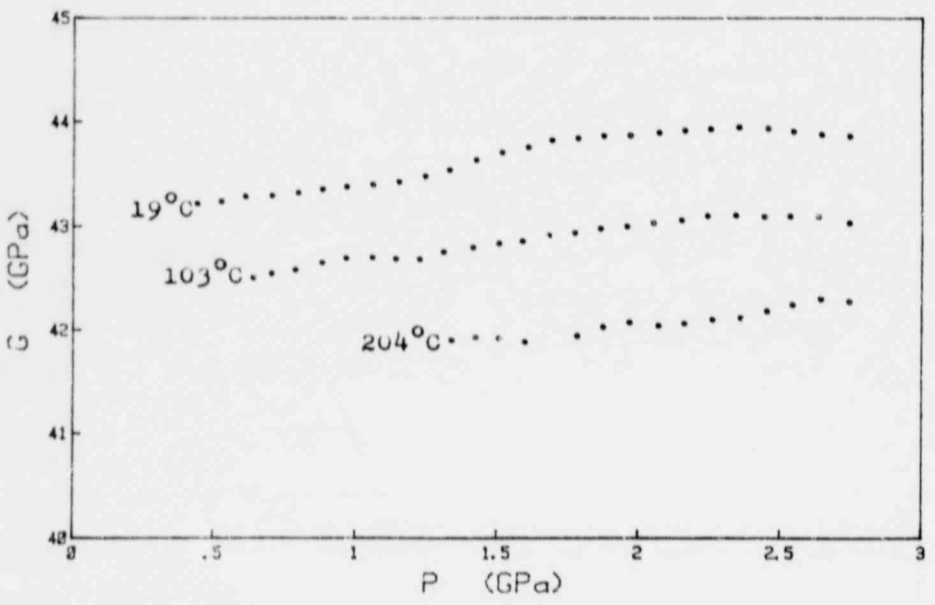


Fig. 5.5.4 119 MCY GC: Shear modulus vs pressure

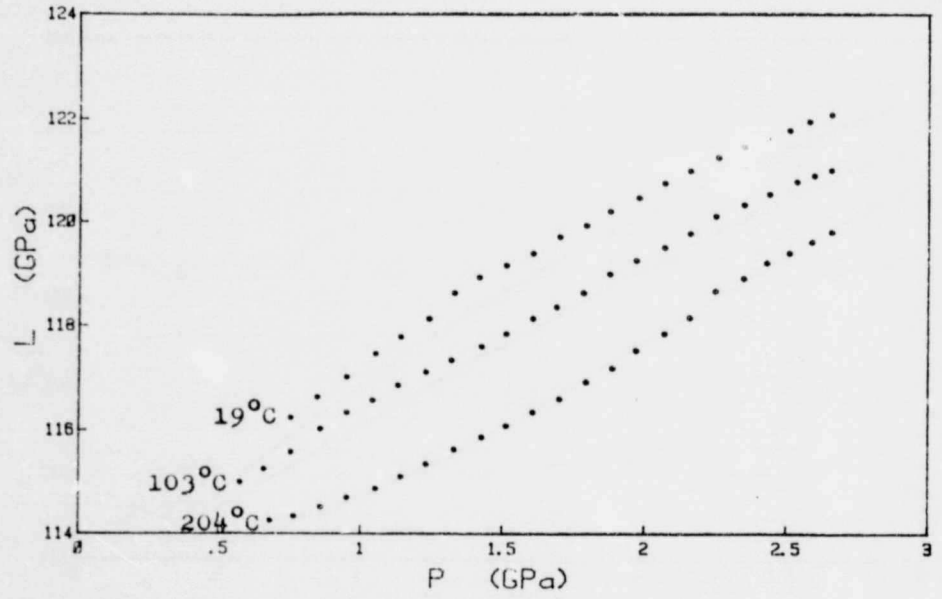


Fig. 5.5.3 119 MCY GC: Longitudinal modulus vs pressure

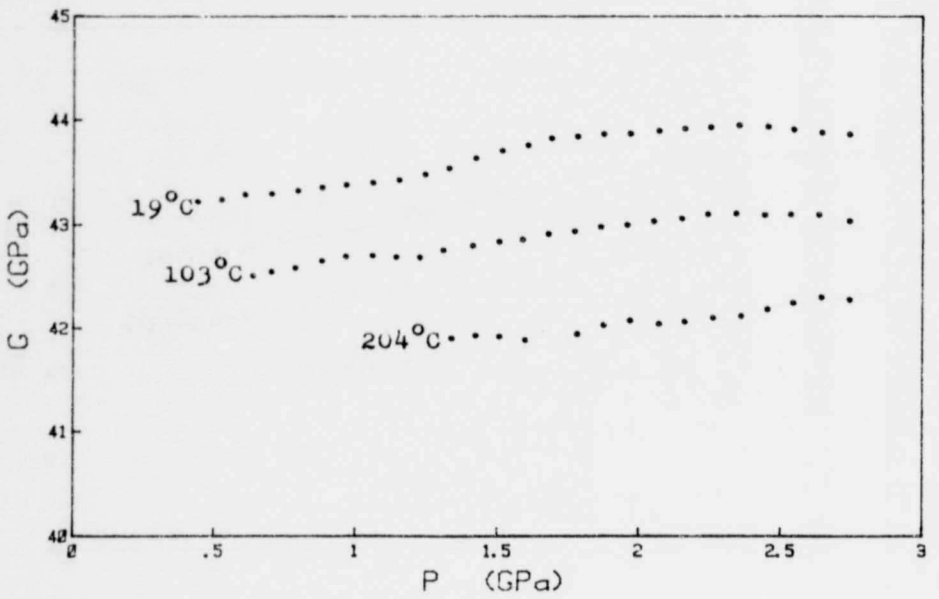


Fig. 5.5.4 119 MCY GC: Shear modulus vs pressure

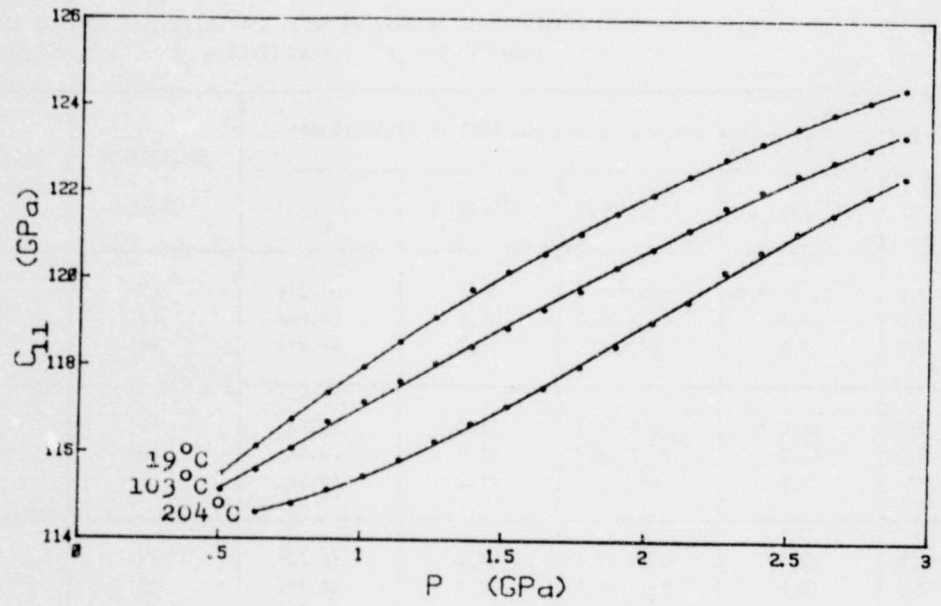


Fig. 5.5.5 119 MCY GC: C_{11} vs pressure

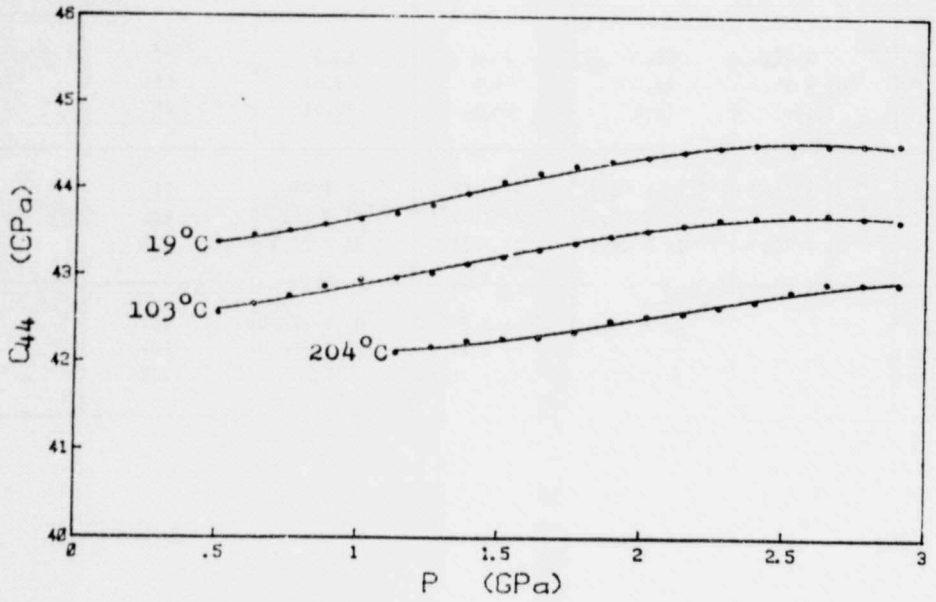


Fig. 5.5.6 119 MCY GC: C_{44} vs pressure

Table 5.5.1

119 MCY GC: Elastic Properties and their Polynomial Coefficients

Density = 2,769 g/cc v_ℓ = 6,476 km/s v_t = 3,927 km/s

ELASTIC CONSTANT	TEMPERATURE (°C)	COEFFICIENTS OF EQUATION $M = a_0 + a_1P + a_2P^2 + a_3P^3$				$X \leq P \leq Y$(GPa)	
		a_0	$a_1(GPa^{-1})$	$a_2(GPa^{-2})$	$a_3(GPa^{-3})$	X	Y
C_{11}(GPa)	19	112,68	5,84	-0,60	$-0,61 \times 10^{-2}$	0,5	3
	103	113,38	3,38	$3,61 \times 10^{-1}$	-0,12	0,5	3
	204	114,44	-1,13	2,51	-0,40	0,6	3
C_{44}(GPa)	19	43,27	-0,13	$6,77 \times 10^{-1}$	-0,17	0,5	3
	103	42,44	0,13	$4,27 \times 10^{-1}$	-0,11	0,5	3
	204	42,68	-1,35	0,93	-0,15	1	3
E (GPa)	19	102,90	1,88	$7,69 \times 10^{-1}$	-0,25	0,5	3
	103	101,95	1.34	$7,69 \times 10^{-1}$	-0,21	0,5	3
	204	102,12	-1,58	1,85	-0,31	1	3
B (GPa)	19	54,98	6,02	-1,50	$2,18 \times 10^{-1}$	0,5	3
	103	56,8	3,21	-0,21	$3,39 \times 10^{-2}$	0,5	3
	204	59,18	-1,93	2,57	-0,41	1	3
C_{12}(GPa)	19	26,13	6,11	-1,95	$3,29 \times 10^{-1}$	0,5	3
	103	28,51	3,12	-0,49	$1,09 \times 10^{-1}$	0,5	3
	204	31,09	-1,59	2,21	-0,35	1	3
σ	19	$1,89 \times 10^{-1}$	$2,57 \times 10^{-2}$	$-1,02 \times 10^{-2}$	$1,82 \times 10^{-3}$	0,5	3
	103	$2,01 \times 10^{-1}$	$1,24 \times 10^{-2}$	$-3,37 \times 10^{-3}$	$7,25 \times 10^{-4}$	0,5	3
	204	$2,12 \times 10^{-1}$	$-0,33 \times 10^{-2}$	$6,39 \times 10^{-3}$	$-1,07 \times 10^{-3}$	1	3
V/V_o	19	$9,998 \times 10^{-1}$	$-1,75 \times 10^{-2}$	$5,04 \times 10^{-4}$		0,5	3
	103	$9,999 \times 10^{-1}$	$-1,74 \times 10^{-2}$	$4,86 \times 10^{-4}$		0,5	3
	204	1,0	$-1,76 \times 10^{-2}$	$5,07 \times 10^{-4}$		0,5	3

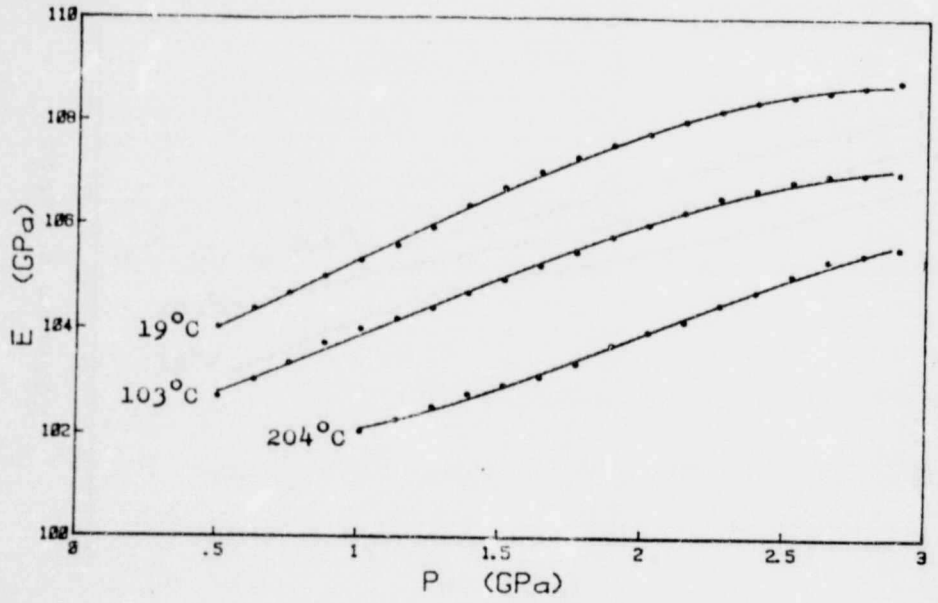


Fig. 5.5.7 119 MCY GC: Young's modulus vs pressure

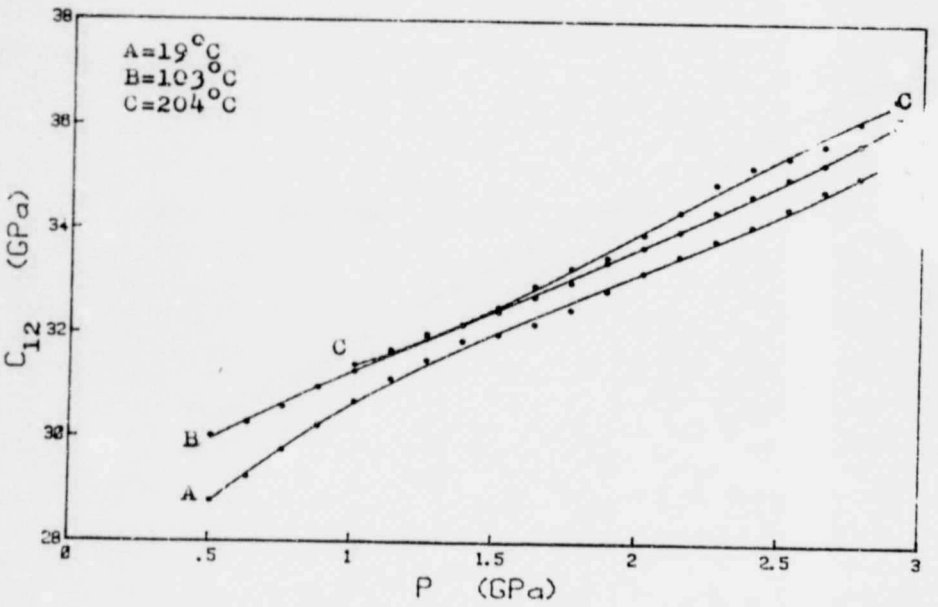


Fig. 5.5.8 119 MCY GC: Lamé constant vs pressure

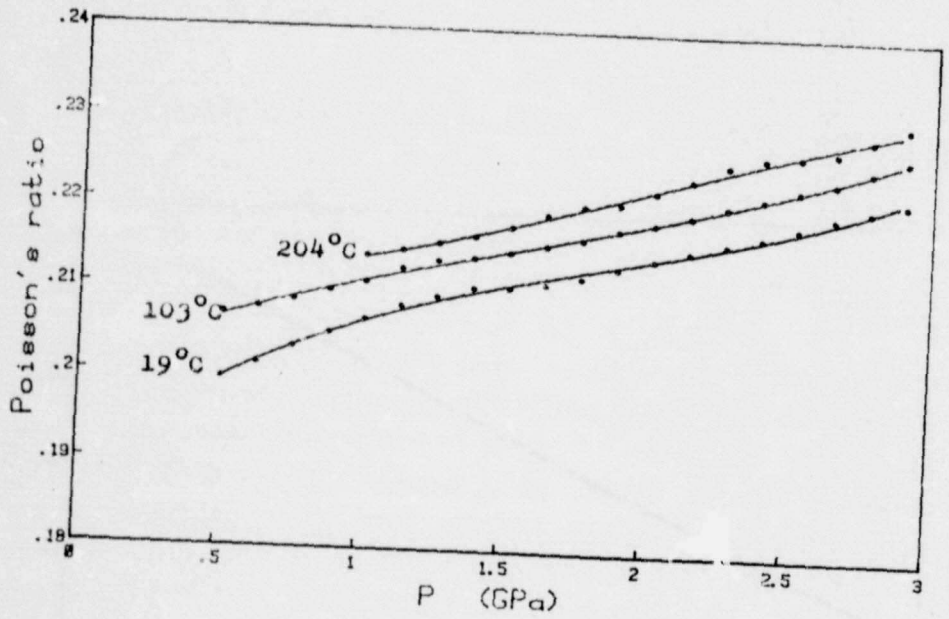


Fig. 5.5.9 119 MCY GC: Poisson's ratio vs pressure

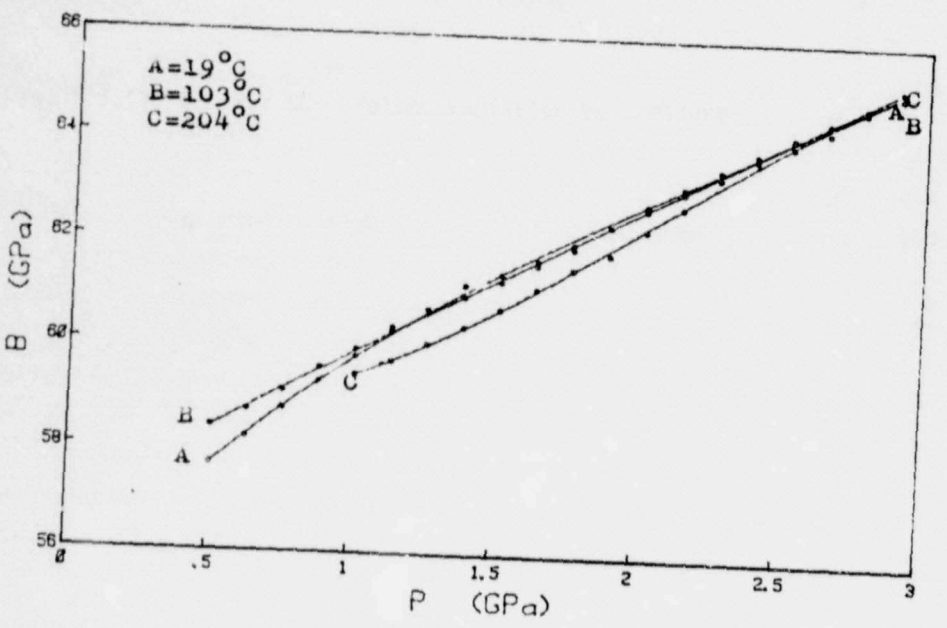


Fig. 5.5.10 119 MCY GC: Bulk modulus vs pressure

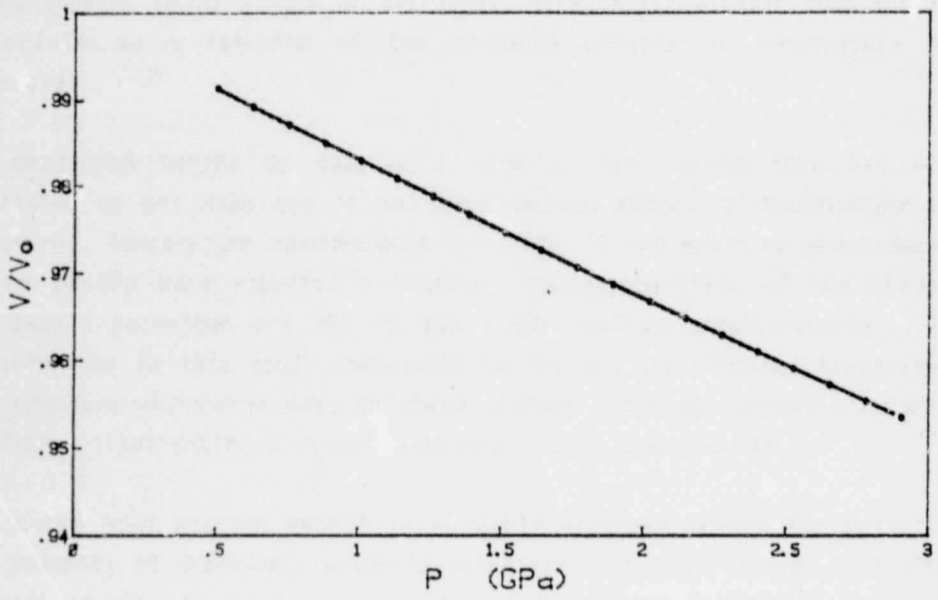


Fig. 5.5.11 119 MCY GC: Volume variation vs pressure

6. SUMMARY, CONCLUSIONS AND RECOMMENDATIONS

There appears to be a lack of ultrasonic data on the elastic constants of materials as a function of the combined effects of temperature and pressure.

As mentioned before in chapter 1 work in this field that has been published do not make use of the simultaneous effect of temperature and pressure. Temperature measurements up to 800 °C and pressure measurements up to 14 GPa were reported. However, the capabilities of the present ultrasonic technique are 300 °C and 3 GPa applied simultaneously. The measurements in this study correspond to the highest combined temperature and pressure ultrasonic data on these glasses and glass ceramics reported to date - illustrating a unique technique with unique results.

The pulse echo overlap method is a highly accurate method for measuring the velocity of ultrasonic waves in materials. It has features that other methods usually do not possess simultaneously. This method has been used during this study to measure the ultrasonic velocity for materials as a function of the combined effect of temperature and pressure.

Quasi-hydrostatic pressure was generated in a Kennedy piston-cylinder apparatus using a capsule with lead as pressure transmitting medium. A graphite resistance heater was incorporated in this capsule. Current to the heater passed into the capsule via top steel parts and a brass conductor, and generated heat in the heater. An ultrasonic wave was generated by a transducer, cemented to the base of the piston, which also acted as a buffer rod. The velocity of the ultrasonic wave travelling through the sample, was recorded as frequency as a function of temperature and pressure. Particular corrections had to be applied to the data and the pressure dependence of the elastic constants were calculated for various isotherms.

The elastic behaviour of the 119 MCY glass and the Corning 9658 glass with pressure and temperature has been established to be related to their Na_2O content and SiO_2 content, respectively. Both materials behave elastically anomalously for an amorphous material in a specific pressure range and "normal" in another pressure range between 0 GPa and 3 GPa. Therefore it appears that these glasses might be in a transition state between the elastically anomalous and the normal states.

The glass ceramics, 119 MCY and Corning 9658, show elastically normal behaviour for a crystalline material in that the pressure derivatives of the longitudinal and shear modes are positive. On the other hand results on the Zerodur glass ceramic show that both the longitudinal and shear modes decrease nonlinearly with pressure and around 1.5 GPa there is an abrupt change in the slopes. These turning points with pressure are an increasing function of temperature indicating the high-to-low quartz phase boundary in Zerodur. Consequently the phase diagram for Zerodur could have been established in the 0-3 GPa pressure range and 0-300 °C temperature range.

This technique is capable of defining a phase transition, if it occurs in a material, or else of establishing the unit volume variation of the material with pressure. In the technological field there is constantly a need for the development of materials to withstand extreme conditions of pressure and temperature. In future this technique can be applied to investigate the elastic behaviour of solid materials and a phase diagram can be drawn up.

A recommendation is that it would be useful to know the phases involved in such a phase transition. This technique should be expanded by employing a high pressure X-ray diffraction technique, such as diamond anvil cell, to define the nature of a transition.

Another recommendation is to use Du Pont 9770 conducting paste together with a Du Pont 9180 thinner (Manasreh and Pederson, 1985) for a more effective acoustic bond between the transducer and piston at high temperatures.

Combining ultrasonics with high temperature and high pressure is a difficult field to work in. However, it is real satisfaction once the results have been obtained. Might this research be a contribution towards the understanding of the elastic behaviour of materials in the expanding world of materials development.

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