AD-783 680

EFFECTS OF COMPOSITION, PRESSURE, AND TEMPERATURE ON THE ELASTIC, THERMAL, AND ULTRASONIC ATTENUATION PROPERITES OF SODIUM SILICATE GLASSES

Murli G. Manghnani

Hawaii Institute of Geophysics

Prepared for:

Office of Naval Research

July 1974

DISTRIBUTED BY:

NTIS

National Technical Information Service
U. S. DEPARTMENT OF COMMERCE
5285 Port Royal Road, Springfield Va. 22151

Unclassified

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

AD 783 680

REPORT DOCUMENTATION PAGE

READ INSTRUCTIONS BEFORE COMPLETING FORM

1. REPORT NUMBER: Technical Report No. 5

2. GOVT ACCESSION NO.

3. RECIPIENT'S CATALOG NUMBER

4. TITLE (and Subtitle): EFFECTS OF COMPOSITION, PRESSURE, AND TEMPERATURE ON THE ELASTIC, THERMAL, AND ULTRASONIC ATTENUATION PROPERTIES OF SODIUM SILICATE GLASSES

5. TYPE OF REPORT & PERIOD COVERED: Technical--~~2/1/74 - 12/31/74~~

6. PERFORMING ORG. REPORT NUMBER

7. AUTHOR(s): Murli H. Manghrani

8. CONTRACT OR GRANT NUMBER(s): N00014-67-A-0387-0012

9. PERFORMING ORGANIZATION NAME AND ADDRESS: Hawaii Institute of GEophysics, University of Hawaii, Honolulu, Hawaii 96822

10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS: NR 032-527

11. CONTROLLING OFFICE NAME AND ADDRESS: Office of Naval Research, Metallurgy Program

12. REPORT DATE: July 1974

13. NUMBER OF PAGES: ~~13~~ 16

14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)

15. SECURITY CLASS. (of this report): Unclassified

15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report): Distribution unlimited.

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

Reproduced by NATIONAL TECHNICAL INFORMATION SERVICE U S Department of Commerce Springfield VA 22151

18. SUPPLEMENTARY NOTES: Paper to be published in the Proceedings, Xth International Congress on Glass, July 8-12, 1974, Kyoto, Japan.

19. KEY WORDS (Continue on reverse side if necessary and identify by block number): Sodium silicate glasses; Elastic, thermal, and ultrasonic attenuation; Composition, pressure, temperature

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Elastic moduli of sodium silicate glasses, containing 10 to 40 mole % Na_2O, have been measured by ultrasonic interferometry method to 5 kbar (at 298°K) and in the temperature range 80 to 498°K. The values of pressure and temperature derivatives of the moduli become systematically less anomalous (i.e., more positive and more negative, respectively) with increasing Na_2O content. Similarly, the mode Grüneisen parameters γ_i derived from the pressure dependence of elastic wave velocities, and the

(continued over page)

DD FORM 1 JAN 73 1473 EDITION OF 1 NOV 65 IS OBSOLETE. S/N 0102-014-6601

Unclassified 16

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

20.

thermal Grüneisen parameters γ_{th} calculated from thermal expansivity data increase with increasing Na_2O content. The variation of $\gamma(T)$ is investigated. The ultrasonic attenuation α in these glasses was studied in the temperature range of 80 to 298°K. At 298°K the value of α for sodium silicate glasses, compared to α value of < 0.1 db/cm for fused silica, systematically increases from $\sim$ 2 to $\sim$ 8 db/cm with increasing Na_2O content. At low temperatures, the attenuation-temperature curves for these glasses show reversals. The attenuation peak at $\sim$ 47°K, found in fused silica, shifts to a higher temperature in these glasses as the Na_2O is increased. Implications are discussed in terms of a network model.

TECHNICAL REPORT NUMBER 5

Contract N00014-67-A-0387-0012, NR 032-527
Office of Naval Research
Metallurgy Program
1 February 1974 - 31 December 1974

EFFECTS OF COMPOSITION, PRESSURE, AND TEMPERATURE ON THE ELASTIC, THERMAL, AND ULTRASONIC ATTENUATION PROPERTIES OF SODIUM SILICATE GLASSES

By

Murli H. Manghnani
Hawai itute of Geophysics
sity of Hawaii
Hon..lu, Hawaii 96822

D D C
AUG 26 1974

Distribution unlimited.

July 1974

EFFECTS OF COMPOSITION, PRESSURE, AND TEMPERATURE ON THE ELASTIC, THERMAL, AND ULTRASONIC ATTENUATION PROPERTIES OF SODIUM SILICATE GLASSES*

by

M. H. Manghnani and B. K. Singh

Hawaii Institute of Geophysics
University of Hawaii
Honolulu, Hawaii 96822

To be published in the Proceedings, Xth International
on Glass, July 8-12, 1974, Kyoto, Japan

*Research supported by the Office of Naval Research,
Contract N00014-67-A-0387-0012, NR 032-527
Hawaii Institute of Geophysics Contribution 598

EFFECTS OF COMPOSITION, PRESSURE, AND TEMPERATURE ON THE ELASTIC, THERMAL, AND ULTRASONIC ATTENUATION PROPERTIES OF SODIUM SILICATE GLASSES

MANGHNANI, M.H., SINGH, B.K.
Hawaii Institute of Geophysics, University of Hawaii
Honolulu, Hawaii 96822 USA

INTRODUCTION

In the course of reviewing various thermal and elastic properties of glasses, it has been clearly shown (refs. 1-2) that two types of anomalous behavior exist in the vibrational properties of inorganic glasses. The first is related to abnormally low-frequency vibrational modes for which volume dependence of frequency (i.e., $d\omega/dV$) is positive, and includes anomalous properties such as negative thermal expansion coefficient, positive temperature derivative of elastic moduli and negative pressure derivative of elastic moduli, all of which are characteristic of the tetrahedral glasses like fused silica. The second type depends solely upon the low-frequency vibrational modes and includes properties such as large acoustic loss, excess specific heat, and low-frequency Raman scattering, all of which are characteristic of glassy state, in general. This paper reports on the elastic moduli, thermal expansion and acoustic loss in fused silica, and in seven sodium silicate glasses, as the function of composition, pressure, and temperature. Although several investigations on the elastic and thermal properties of alkali silicate glasses have been carried out (e.g., refs. 1, 3-6), virtually no data exist on ultrasonic attenuation in such glasses, except for some lower frequency, high-temperature data (ref. 22).

The elastic properties of simple silicate glasses in general depend on the density of the Si-O-Si bonds in the random network of silica. The addition of a network-modifier such as Na_2O results in a breakdown of some of those bonds and the formation of relatively weaker Si-O-Na bonds in proportion to the Na_2O added. In this manner, Charles (ref. 7) has shown that Young's modulus in the Na_2O-SiO_2 glass system decreases systematically with increase in Na_2O content.

It is of interest to study the changes in the anomalous behavior of tetrahedral silica-like glasses as they are progressively modified by the addition of Na_2O. The purpose of this study is to investigate the elastic, thermal, and ultrasonic attenuation properties of the glasses in the Na_2O-SiO_2 glass system in an effort to elucidate the structural changes in the random silica-based network, and the mechanism involved.

EXPERIMENTAL TECHNIQUES

Fused silica (CGW code 7940), obtained from the Corning Glass Works, Corning, New York, and seven sodium silicate glasses, synthesized at the National Bureau of Standards, Washington, D.C., were used in this study. The chemical composition and annealing temperatures of the sodium silicate glasses are given in Table 1. Right circular cylinders, 1.25 cm long, were prepared from the glass samples for the velocity measurements as function of pressure (to 5 kbar at 298° K) and temperature (298-498° K at 1 bar); cylinders of approximately the same length but larger diameter (∿2 cm) were used for the low-temperature (80-298° K) attenuation and velocity measurements, in order to minimize the errors of the side-wall effects in attenuation work. The end-faces of cylindrical specimens were paralleled to within 1 part in 10^4 parts, and polished flat to ±1/2 wavelength of sodium light.

The pulse superposition technique (ref. 8) was employed for studying the pressure and temperature dependence of ultrasonic velocities and, hence, of the moduli. X- and Y-cut quartz transducers, 0.65 cm diameter, having 20 MHz natural resonant frequency were used to generate compressional and shear waves in the specimen; 30 MHz transducers were used for attenuation measurement. The bonding materials between the specimen and transducer were: Dow-Corning 276-V9 resin for the pressure measurements at high pressure; HT-424 epoxy (manufactured by American Cyanamid Co.) at high temperatures (298-498° K); and 'Nonaq' stopcock grease (Fisher Scientific Co.) for the low-temperature velocity and attenuation measurements. The details of the ultrasonic equipment, the high-pressure and high-temperature apparatus, and the procedure for evaluating the pressure and temperature velocity data have been described previously (ref. 9). The basic experimental measurement was the pulse repetition frequency (PRF) of the carrier rf wave applied to the transducer attached to one plane-paralleled faces of the specimen. At the 'resonant' condition, ignoring the effects of the bond, the velocity is given by $v = 2f\ell$ where f is PRF rate (i.e., reciprocal of a round-trip delay time) and ℓ is length of the specimen. For evaluating velocities and moduli at high and low temperatures (above and below 298° K, respectively), corrections were applied for path-length changes, using the measured expansivity data (in the 298-498° range). The results, to be discussed later, are shown in Fig. 4.

The "modified" pulse-echo-overlap (ref. 10) was employed for making the low-temperature measurements of ultrasonic velocities and attenuation simultaneously, and liquid-N_2 dewer and the electronic equipment similar

to that given in ref. 10. The method allowed a continuous recording of attenuation with temperature, the resolution of measured attenuation values being 0.04 dB or better. The accuracy of the measured velocity is estimated to be .03% or better.

DISCUSSION OF RESULTS

1. Elastic Moduli at Ambient Conditions. The elastic parameter measurements for fused silica and the Na_2O-SiO_2 glasses at 20 MHz are listed in Table 1. The density of the Na_2O-SiO_2 glasses continuously increases

Table 1

Composition, and elastic and thermal properties of fused silica and Na_2O-SiO_2 glasses at ambient conditions

Glass No.	Composition, mole % SiO_2	Composition, mole % Na_2O	Annealing temp., °C	$\bar{V}$, cc	ρ, g/cm³	V_s, km/sec	V_p, km/sec	θ_D, °K	C_v, cal/g.deg	β_{calc}, 10^{-6} deg⁻¹	K_s, kbar	γ_{th}	γ_{pt}
Fused silica (7940)	100	0	1,000	22.02	2.222	5.941	3.764	495.7	0.2597	0.123	381.2	0.019	-0.31
X110	90	10	575	22.08	2.252	5.569	3.478	467.0	0.2632	0.236	335.8	0.266	-1.35
X111	85	15	540	22.11	2.336	5.457	3.355	452.9	0.2643	0.339	314.6	0.365	-0.81
X112	80	20	525	22.14	2.353	5.382	3.251	443.2	0.2654	0.463	324.5	0.382	-0.35
X113	75	25	510	22.18	2.431	5.367	3.159	434.3	0.2660	0.248	372.5	0.445	0.17
X114	70	30	495	22.21	2.474	5.348	3.101	429.4	0.2664	0.612	389.6	0.511	0.49
X115	65	35	475	22.24	2.535	5.281	3.038	422.4	0.2662	0.192	415.0	0.616	0.77
X116	60	40	450	22.27	2.525	5.401	2.993	412.2	0.2656	0.359	432.5	0.676	1.27

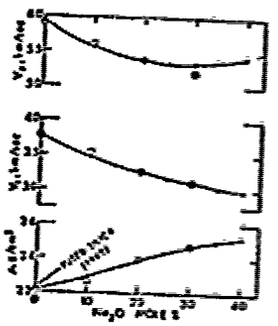

Fig. 1 Density, V_s and V_p as function of Na_2O content. Open circles represent unpublished data of Kurkjian and Krause.

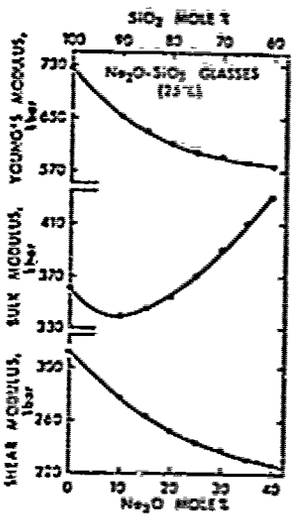

Fig. 2

Composition dependence of the elastic moduli.

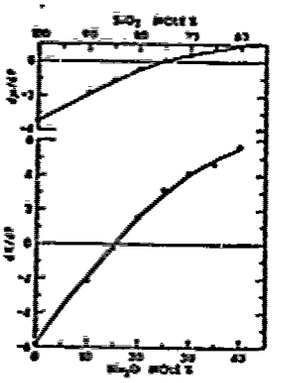

Fig. 3

dK/dP and dμ/dP versus Na_2O content.

with increasing Na_2O content, however, the velocities of both the compressional (V_p) and shear (V_s) waves decrease. Good agreement is found with the unpublished velocity data of Krause and Kurkjian (personal communication, 1974), as seen in Fig. 1. The present data indicate a minimum in V_p near 25 mole % Na_2O composition; thereafter V_p seems to increase with increasing Na_2O content. Fig. 2 shows the composition dependence of shear (μ), bulk (K), and Young's (E) moduli. Both μ and E decrease nonlinearly but systematically with increasing Na_2O content; K modulus in fused silica appears at first to decrease when Na_2O is added but, above 10-12 mole % Na_2O), it continuously increases. These results show that the role of added Na^+ is twofold: (1) the ions fill-in the void space in silica structure; and (2) they modify the Si-O-Si bonds, forming weaker Si-O-Na bonds in proportion to the amount of Na_2O added and causing moduli to decrease (ref. 7).

2. Pressure and Temperature Dependence of Elastic Moduli. The pressure dependence of the elastic moduli of Na_2O-SiO_2 glasses is found to be linear, within experimental error, in the pressure range of this study (5 kbar). Lower soda-content glasses appear more anomalous (i.e., dK/dP and dμ/dP are both more negative); the anomalous behavior systematically decreases (i.e., becomes less negative) with increase in Na_2O content (fig. 3). At lower Na_2O content ($\leq$ 20 mole %), the values of dK/dP and dμ/dP change more rapidly with composition than at higher content (> 20 mole %). From Fig. 3, dK/dP and dμ/dP are zero for glasses containing approximately 16 and 25 mole % Na_2O, respectively.

The results of the temperature dependence of fractional velocities, $(V_p)_T/(V_p)_{298}$ and $(V_s)_T/(V_s)_{298}$, and of shear modulus are shown in Figs. 4 and 5, respectively. Except for glasses K110 and K111 (containing

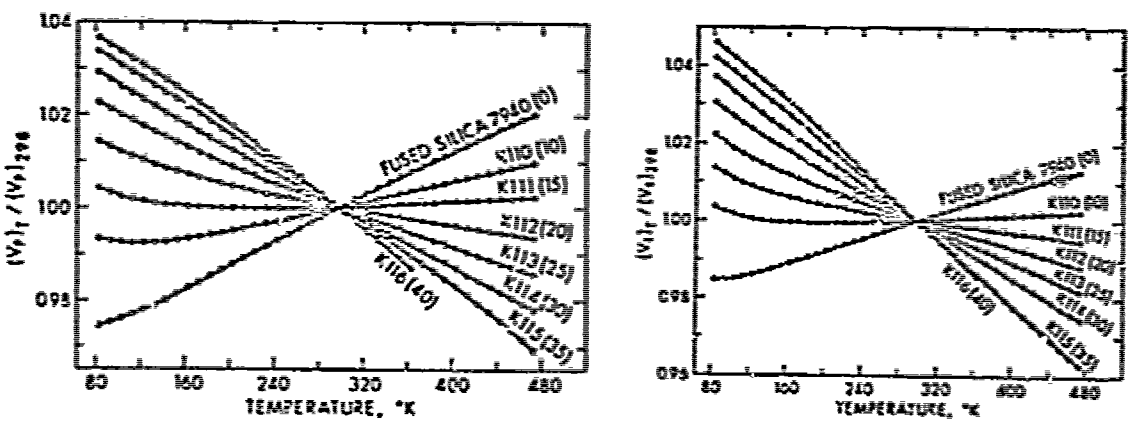

Fig. 4 Longitudinal- (left) and shear-wave (right) velocity ratio versus temperature. The values in parentheses represent Na_2O mole %.

10 and 15 mole % Na_2O, respectively), the temperature dependence of V_p (above 298° K) in all of the others is normal (i.e., negative temperature derivatives); for V_s, only K110 shows anomalous behavior (i.e., positive temperature derivative). The values of dK/dT and dμ/dT from both the high- and low-temperature measurements at 298° K are in good agreement (Fig. 6).

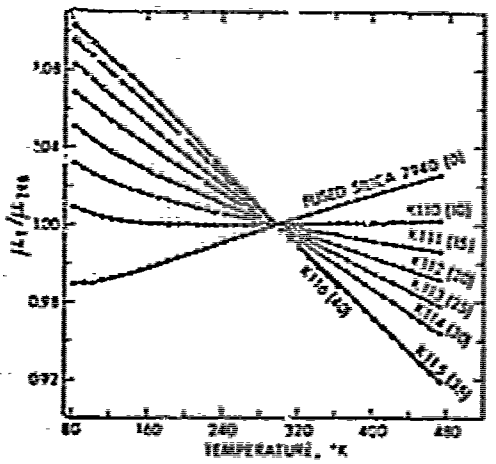

Fig. 5 Shear mod[illegible] ratio versus te[illegible]ture.

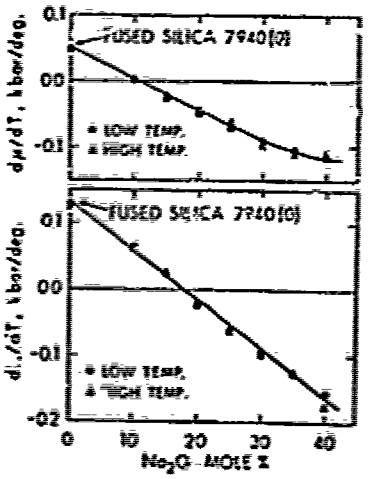

Fig. 6 dK/dT and dμ/dT versus Na_2O content.

Here it is again noted that the degree of anomalous behavior (positive dK/dT and dμ/dT) decreases with addition of Na_2O. The composition dependence of dK/dT is linear; but that of dμ/dT is somewhat nonlinear, at least for higher Na_2O content.

In the course of a study of the acoustic spectra of Na_2O-GeO_2 glasses (ref. 11), it was pointed out that a 30% Na_2O-70% GeO_2 glass shows low-temperature thermal relaxation at ~125° K. The low-temperature results in Figs. 4 and 5, showing reversals in the anomalous behavior of fused silica and the K111 and K112 glasses, also indicate such thermal relaxation. The minima in the velocity and modulus (around ~120° K), caused by the thermal relaxation, is seen to shift to higher temperatures with increase in the Na_2O content; also, the magnitude of thermal relaxation decreases, as indicated by increasing linearity of the curves, with increasing Na_2O content.

3. Grüneisen Parameters. The thermal Grüneisen parameters, $\gamma_{th} = \alpha_V K_s/\rho C_p = \alpha_V K_s/\rho C_v$, were evaluated using measured α_V, K_s and ρ values, and calculated C_v values; the latter were obtained from the Debye theory (ref. 12). Values of Debye temperature (θ_D), C_v, and γ_{th} are given in Table 1. The γ_{th} values systematically increase with increase in Na_2O

content (fig. 7) which systematic increase is also reflected in the

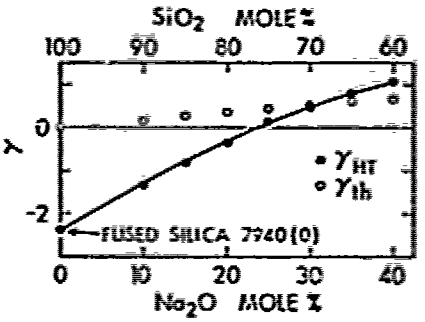

Fig. 7 γ_{th} and γ_{HT} as functions of Na_2O content.

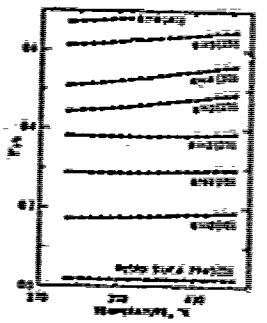

Fig. 8 γ_{th} as function of temperature.

pressure and temperature derivative values. The high-temperature limiting values of mode Grüneisen parameter, γ_{HT}, can be evaluated from pressure dependence of velocities (ref. 13). The values of γ_{HT} increase more rapidly than γ_{th} does with increasing Na_2O content (fig. 7). Although the γ_{HT} values are negative and larger than the γ_{th} values, both Grüneisen parameters increase when Na_2O is added to the silica structure.

Temperature dependence of γ_{th} in the range of 298-498° K is shown in Fig. 8. In fused silica and the lower soda-content glasses, the γ_{th} value is more or less temperature-invariant. For higher soda-content glasses, γ_{th} increases more rapidly with temperature.

4. Ultrasonic Attenuation. Large ultrasonic attenuation occurs in simple inorganic oxide glasses such as SiO_2, GeO_2, B_2O_3, and As_2O_3 at various temperatures in the range from ~50° to ~250° K (refs. 14-21). Anderson and Bömmel (ref. 14) proposed a structural relaxation mechanism to explain the attenuation spectrum for fused silica at low temperatures. According to them, thermally activated lateral shifting of an oxygen atom between two equivalent equilibrium positions, perpendicular to the Si-O-Si bond, is a source of structural relaxation which also explains the negative thermal expansion and excess specific heat anomalies at low temperatures in fused silica. An alternate mechanism, which involves the movement of the oxygen atom in the direction of the Si-O-Si bond rather than perpendicular to it, has been proposed in a two-bond-length model (refs. 15-16).

Compared to the amount of work on fused silica, very little work has been done on the study of relaxational processes in sodium silicate glasses except at low frequencies (.01 and 10 Hz) where the attenuation occurs due to ionic migration of Na^+ (refs. 17, 22) and possibly oxygen ions (refs. 22-24). The Na^+-migration relaxation is a stress-induced phenomenon, resolvable at frequencies of the order of KHz.

Figures 9 and 10 show the composition dependence of longitudinal and

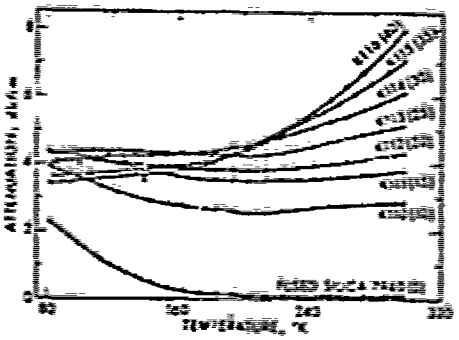

Fig. 9
Temperature dependence of attenuation (longitudinal mode).

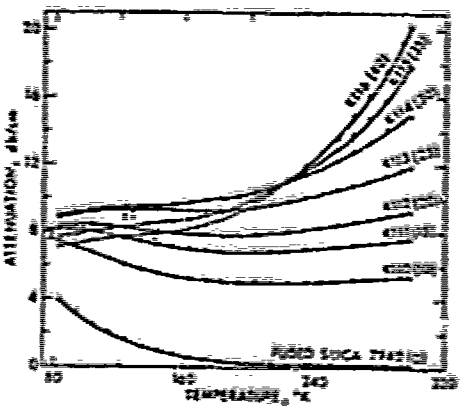

Fig. 10
Temperature dependence of attenuation (shear mode).

shear attenuation at 30 MHz, respectively, for the Na_2O-SiO_2 glasses at ambient conditions. As seen, the attenuation due to shear-wave propagation is approximately twice that due to longitudinal-wave propagation, which supports the previous conclusion (ref. 19) that the loss is in large part sensitive to shear distortion. The temperature dependence of longitudinal and shear ultrasonic attenuation are plotted in Fig. 11. The

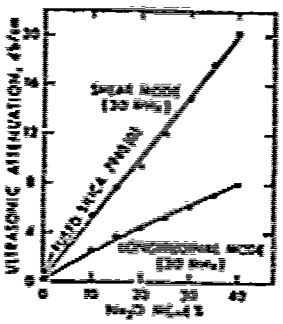

Fig. 11 Ultrasonic attenuation as function of Na_2O content.

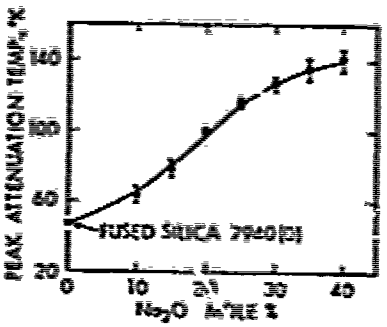

Fig. 12 Peak attenuation temperature versus Na_2O content.

present 30 MHz attenuation data for fused silica are in good qualitative agreement with previous 20 MHz data (ref. 14, 16, 21) with respect to the shape of the attenuation-temperature curve, although our attenuation values at 30 MHz are larger. Figures 9 and 10 show increasing broadening of the attenuation peaks with increasing concentration of Na_2O. This is probably due either to thermal broadening (ref. 14) or to a wider distribution of activation energy; the width of the loss peak (e.g., in fused silica; refs. 14, 16) has been interpreted in light of the distribution of activation energy. Alternately, the broadening could also be due to the superposition of the high temperature Na^+-migration relaxation on the structural relaxation, at least, for the low-silica glasses.

One of the most interesting findings of this study is the shifting of the peak attenuation temperature (in the low-temperature range) with increasing Na_2O content; the peak attenuation temperature for each glass is indicated by an arrow (figs. 9 and 10). Because of the broadness of the attenuation curves, there is significant uncertainty in determining the peak temperatures. Also, since the peak attenuation temperatures for glasses K110 and K111 would fall below 80° K--outside the temperature range of this study--the peak attenuation temperatures for these glasses were estimated by extrapolating the attenuation versus temperature curves (Figs. 9 and 10); hence the relationship shown in Fig. 12 is subject to further uncertainty.

However, compared to the peak attenuation temperature of 47° K for fused silica (refs. 14, 16), the peaks for the Na_2O-SiO_2 glasses certainly appear to be composition-dependent. Since the peak attenuation temperature increases with increase in the Na_2O content, the effect of adding Na_2O to SiO_2 is opposite that of adding Na_2O to GeO_2 (ref. 11), although there are structural similarities between fused SiO_2 and GeO_2 (both glasses have tetrahedral structures).

For the low-temperature structural relaxation mechanism, such as that present in fused silica (ref. 14) and presumably also in the Na_2O-SiO_2 glasses, it would be of interest to compute from the frequency-dependence of the peak attenuation temperatures the most probable activation energies for different concentrations of Na_2O. However, as attenuation data at different frequencies are not yet available for these glasses, no attempt can be made to calculate the activation energies for these glasses (for fused silica, activation energy is $\sim$1030 cal/mole; ref. 14). However, from the shift of the peaks to higher temperatures (Fig. 12), it is suggested that in some systematic fashion the activation energy increases with Na_2O, contrary to that found for the Na_2O-GeO_2 system (ref. 11). Although the high-temperature Na^+ relaxation (>250° K) seems to obscure the low-temperature side of the attenuation curve, at least, for higher Na_2O content glasses, it is clearly inferred from the magnitudes of attenuation curves of the Na_2O-SiO_2 glasses (figs. 9 and 10) that the magnitude of attenuation, and thereby relaxation strength, decreases with increasing Na_2O. As a phenomenon, this behavior can be interpreted on the basis of a structural model in which the Na_2O molecule modifies the existing Si-O-Si bonding sites. This modification may result in the formation of a weaker Si-O-Na bond or an unbridged Si-O- bond. The former would impede the movement of the oxygen atoms, and since movement of the oxygen atom is the cause of attenuation this would result in decrease in the attenuation, accompanied by the increase in the activation energy as revealed by the

shifting of the attenuation peak to higher temperatures. The latter modification would, however, make no contribution to attenuation. In other words, a loosening of structure occurs with the addition of soda, which is in agreement with the low frequency internal friction work (ref. 22).

In high-soda glasses, at 30 MHz, one would expect to find Na^+ relaxation at temperatures above room temperature. Near room temperature (Figs. 10 and 11), the temperature-dependence of both longitudinal and shear attenuation increases with increasing Na_2O. The effect of composition on the internal friction at much lower frequencies (.01-Hz) for Na_2O-SiO_2 glasses studied by Forry (ref. 22) shows a decrease both in intensity and peak temperature as Na_2O content increases. In light of this and our results, it may be concluded that the expected high-temperature attenuation peak is shifting toward lower temperatures with increasing Na_2O. A similar shift in Na^+ relaxation temperatures has also been observed in the Na_2O-GeO_2 glasses (ref. 11). This type of high-temperature relaxation requires large activation energy compared to low-temperature structural relaxation. It is desirable to investigate further the composition dependence of Na^+ relaxation at high temperatures and low frequencies in other types of glasses.

5. Summary

a. Elastic moduli of Na_2O-SiO_2 glasses and their pressure and temperature derivatives vary systematically with composition. Increasing Na_2O content results in decrease of μ and E moduli. K modulus first decreases ($\sim$10% Na_2O) and then increases with increase in Na_2O. Low-soda glasses exhibit anomalous behavior under pressure and temperature. The degree of anomalous behavior decreases with increasing Na_2O, i.e., the pressure derivatives of moduli increase and the temperature derivatives decrease with increase in Na_2O.

b. The Grüneisen parameters γ_{th} and γ_{HT} increase with Na_2O. The disparity between the γ_{th} and γ_{HT} values decreases as Na_2O increases; at 70 mole % Na_2O glass composition, the two values are the same. γ_{th} increases with temperature; the temperature dependence of γ_{th} increases with increase in Na_2O.

c. At room temperature, shear-wave attenuation at 30 MHz is larger ($\sim$2 times) than the longitudinal-wave attenuation; both types of losses, however, increase with Na_2O.

d. The low-temperature attenuation peak (due to structural relaxation) observed in fused silica at $\sim$47° K is also found in Na_2O-SiO_2 glasses. The peak attenuation temperature is found to increase with increase in Na_2O,

indicating that the activation energy is increasing with Na_2O, contrary to what has been previously reported for Na_2O-GeO_2 glasses (ref. 11). The attenuation-temperature curves at these low temperatures show a reversed relationship compared to that found at room temperature, i.e., the attenuation peak height decreases with increasing Na_2O. Explanation is offered in terms of the formation of Si-O-Na bonds, which are weaker than Si-O-Si bonds.

6. Acknowledgments

The authors are greatly indebted to Drs. Krause and Kurkjian of Bell Laboratories, Murray Hill, New Jersey, for their unpublished results, and for many helpful suggestions on this study. Thanks are due Dr. G.W. Cleek for the help and advice in making the glasses. The support for this research was provided by the Office of Naval Research, contract N00014-67-A-0387-0012, NR 032-527. Hawaii Institute of Geophysics Contribution 598.

REFERENCES

1. Kurkjian, C.R., Krause, J.T., J. Am. Ceram. Soc., 54, 226 (1968).
2. Kurkjian, C.R., Krause, J.T., McSkimin, H.J., Andreatch, P., Bateman, T.B., in Amorphous Materials, edited by R.W. Douglas and B. Ellis, 463-473. Wiley-Interscience, New York, 1972.
3. Shaw, R.R., Uhlmann, D.R., J. Non-Cryst. Solids, 5, 237 (1971).
4. Krüger, J., Phys. Chem. Glasses, 13, 9 (1972).
5. Manghnani, M.H., Rai, C.S., Bull. Amer. Ceram. Soc., 52, 390 (1973).
6. Ferraro, J.R., Manghnani, M.H., J. Appl. Phys., 43, 4595 (1972).
7. Charles, R.J., in Progress in Ceramic Science, 1, 1-38, edited by J.E. Burke. Pergamon Press, New York, 1961.
8. McSkimin, H.J., J. Acoust. Soc. Amer., 33, 12 (1961).
9. Manghnani, M.H., J. Amer. Ceram. Soc., 55, 360 (1972).
10. Chung, D.H., Silversmith, D.J., Chick, B.B., Rev. Sci. Instr., 40, 718 (1969).
11. Kurkjian, C.R., Krause, J.T., J. Amer. Ceram Soc., 49, 134 (1966).
12. Anderson, O.L., J. Phys. Chem. Solids, 12, 41 (1959).
13. Schuele, D.E., Smith, C.S., J. Phys. Chem. Solids, 25, 801 (1964).
14. Anderson, O.L., Bömmel, H.E., J. Amer. Ceram. Soc., 38, 125 (1955).
15. Strakna, R.E., Phys. Rev., 121, 2020 (1961).
16. Strakna, R.E., Savage, H.T., J. Appl. Phys., 35, 1445 (1964).
17. Marx, J.W., Silversten, J.M., J. Appl. Phys., 24, 81 (1953).
18. Fine, M.E., Van Duyne, H., Kenney, N.T., J. Appl. Phys., 25, 402 (1954).
19. McSkimin, H.J., J. Appl. Phys., 24, 988 (1953).

20. Krause, J.T., Kurkjian, C.R., J. Amer. Ceram. Soc., 51, 226 (1968).
21. Krause, J.T., J. Appl. Phys., 42, 3035 (1971).
22. Forry, K.E., J. Amer. Ceram. Soc., 40, 90 (1957).
23. Hopkins, I.L., Kurkjian, C.R., in Physical Acoustics, 2B, 91-163, edited by W.P. Mason. Academic Press, New York (1965).
24. Mohyuddin, I., Douglas, R.W., Phys. Chem. Glasses, 1, 71 (1960).