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Internal Friction in L.A.S. Type Glass and Glass-Ceramics

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Abstract: Internal friction measurements have been performed on glass and glass-ceramics of the $\text{Li}_2\text{O}-\text{Al}_2\text{O}_3-\text{SiO}_2$ type by isothermal mechanical spectroscopy. Experiments were carried out over a large frequency range (10^{-4} Hz - 31.6 Hz) for various temperatures between 260K and 850K. For the glass, a relaxation peak is observed at low temperature (276K for 1Hz). This peak does not appear in the glass-ceramics; however, for each of them, two other peaks were observed: the first one at about 343K (1Hz) and the second one at higher temperature, between 600K et 700K (1Hz). Our dielectric loss investigation data obtained on the same materials have allowed to assume that the low-temperature peaks are due to the stress-induced diffusion of Li^+ ions, in the vitreous phase for the glass and in the crystal phase for the glass-ceramics respectively. The peaks observed at higher temperature in glass-ceramics depend on the crystal phase but the relaxation mechanisms responsible of them are not well defined.

1. INTRODUCTION

Contrary to binary silicate glasses which have been largely studied [1-3], only few internal friction investigations have been performed on more complex glasses which can be converted into glass-ceramics and on the glass-ceramics themselves [4-6]. This work is about the investigation, by isothermal mechanical spectrometry, of L. A. S. type glass and glass-ceramics, with identical chemical composition, between 77 K and 800 K. It seems interesting to study and compare the internal friction of these materials in order to observe changes in microstructure due to crystallization and to estimate the contribution of the glassy and crystalline phases. In order to obtain further information on the relaxation phenomena, dielectric measurements have been performed on these materials.

2. EXPERIMENTAL PROCEDURE

2.1. Materials

The materials investigated are a glass (A) and two glass-ceramics (B and C) of the $\text{Li}_2\text{O}-\text{Al}_2\text{O}_3-\text{SiO}_2$ system provided by Corning Europe Inc. The three materials have the same composition (%wt): 70 SiO_2 - 20 Al_2O_3 - 6 ($\text{Li}_2\text{O}+\text{MgO}+\text{ZnO}$) - 4 ($\text{TiO}_2+\text{ZrO}_2$).

Glass A, after submitting to two different controlled heat treatments, was converted in glass-ceramic B and C respectively. Glass-ceramic B is transparent and is primarily composed of β -quartz solid solution crystals ($\text{Li}_2\text{O}-\text{Al}_2\text{O}_3-n\text{SiO}_2$). Glass-ceramic C is opaque and its main crystalline phase is β -spodumene ($\text{Li}_2\text{O}-\text{Al}_2\text{O}_3-4\text{SiO}_2$). The crystallinity rate of these glass-ceramics is about 90%.

2.2 Experimental techniques

Internal friction measurements were performed with an inverted torsional pendulum with a high natural frequency subjected to subresonant forced oscillations. With this method the internal friction Q^{-1} is equal to $\tan \phi$, where ϕ is the phase lag angle between applied stress and resulting strain.

The experimental system was described in details elsewhere [7]. Internal friction measurements were carried out in isothermal conditions as a function of frequency on a wide range [10^{-4} Hz -30Hz], between

77 K and 800 K, with strain amplitude ranged between 5.10^{-6} and 20.10^{-6} . The Q^{-1} accuracy is about 2.10^{-4} and the sample were cut in flat bars ($50 \times 5 \times 1.5 \text{ mm}^3$).

The dielectric loss measurements were carried out at various stabilized temperatures between room temperature and 500K under partial pressure of N_2 in the frequency range $[20 \text{ Hz}-10^6 \text{ Hz}]$ with a HP 4284 bridge which allows to measure the values of the equivalent capacitance C_p and resistance R_p of each sample. The specimens were flat parallelepipeds (thickness:1mm) covered by a circular silver film (diameter:10mm) in order to improve the electrical contact with the electrodes. The values of ϵ'' (imaginary part of the complex permittivity ϵ^*), is obtained from the values of R_p and C_o (capacitance in vacuum).

3.RESULTS

For glass A, the internal friction spectrum in the temperature range investigated [200 K-850 K] can be separated in the relaxation peak P_{1A} below room temperature and a high temperature increasing background (Figure 1). The peak P_{1A} occurs at 276K for 1Hz; it is asymmetrical and is too wide to be entirely described on a spectrum against frequency.

Figure 1

Figure 2

Figure 1: Glass A: internal friction versus $1000/T$ spectrum - measurement frequency: 1 Hz -

Figure 2: Glass-ceramics B and C: internal friction versus temperature spectrum - measurement frequency: 3.16×10^{-2} Hz -

For glass-ceramic B, two internal friction peaks (P_{1B} and P_{2B}) are observed from the isothermal Q^{-1} measurements performed on a large temperature range (between room temperature and 700 K). Like material B, two internal friction peaks (P_{1C} and P_{2C}) are observed for glass-ceramic C for the same investigated temperature range. The Q^{-1} data are plotted against measurement temperature for a given frequency (3.16×10^{-2} Hz) in Figure 2. For this frequency, P_{1B} and P_{2B} occur at 325 K and 556 K while P_{1C} and P_{2C} are located at 310 K and 579 K respectively.

The features of all these peaks (height: h , peak temperature at 1 Hz: T_p and the width at half maximum expressed in decade of frequency: δ) are listed in Table 1.

For dielectric measurements carried out on each material A, B and C, between room temperature and 500 K, the loss factor ϵ'' exhibits a high exponential low frequency background. After subtraction of this background a dielectric relaxation peak is revealed.

4.DISCUSSION

Arrhenius plots are drawn in Figure 3 for the low temperature internal friction and dielectric peaks of the three materials A, B and C. There are three distinct straight lines, each of them corresponding to one of the three materials; for each material (A, B or C), the points arising from mechanical and dielectric relaxation peaks are on the same straight line. That means that for every particular material (A, B, C) at stake, the relaxation mechanisms, responsible for the mechanical and dielectric peaks at relatively low temperature, are the same. Thus, the relaxation units responsible for the peaks P_{1A} , P_{1B} and P_{1C} are electrically charged. It should be mentioned that the straight line concerning P_{1A} is well separated from the two others. The Arrhenius plots relative to the internal friction peaks P_{2B} and P_{2C} are shown in Figure 4. The apparent activation energy (E_a) and the limit relaxation time (τ_0), deduced from the Arrhenius plots are summed up in Table 1. The peaks P_{1B} and P_{1C} have close features but the relaxation parameters of P_{1A} are different from those of P_{1B} and P_{1C} . The peaks P_{2B} and P_{2C} are different and it seems obvious that they are due to two distinct mechanisms.

Figure 3

Figure 4

Figure 3: Arrhenius plots of mechanical (•, *, *) and dielectric (o, ◊, +) relaxation phenomena observed at low temperature for sample A, B, C respectively.

Figure 4: Arrhenius plots of internal friction peaks P_{2B} and P_{2C} .

Table 1: Relaxation parameters of peaks P_{1A} , P_{1B} , P_{1C} , P_{2B} and P_{2C} .

	P_{1A}	P_{1B}	P_{1C}	P_{2B}	P_{2C}
E_a (eV)	0.63	1.0	0.88	1.8	1.3
τ_0 (s)	4.5×10^{-13}	2.4×10^{-16}	2×10^{-14}	4×10^{-16}	2.6×10^{-11}
T_p (K)	276	340	345	615	665
δ	> 4.8	2.8	2.8	2.2	2.6
$h \times 10^4$	40	37	29	27	30

Several facts suggest that the low-temperature internal friction peak P_{1A} observed in glass A is due to the stress-induced diffusion of lithium ions. P_{1A} is characterized by an activation energy of 0.63 eV close to those found by some authors for alkali ions relaxation. According to many studies, the low-temperature internal friction peak, ascribed to the stress controlled movement of alkali ions, appears below 273 K (about 238 K for 1 Hz) [1], but the addition of Al_2O_3 [8] and alkali-earth oxides (such as MgO , CaO) [9]

cause the shift of this peak to higher temperature (above 273 K). As glass A belongs to a complex L.A.S system which contains, among other components, alkali earth oxide (MgO), the position of P_{1A} slightly above 273 K is in agreement with the previous observations. Furthermore, according to the studies of Li *et al.* on the influence of the crystallization process in some oxide glasses, such as $K_2O-Li_2O-Al_2O_3-SiO_2$ [5,6] and $MgO-Al_2O_3-SiO_2-TiO_2$ [5], it seems obvious that the peak observed at low temperature in $K_2O-Li_2O-Al_2O_3-SiO_2$ glass is linked with the presence of alkali ions. The peak P_{1A} observed in glass does not appear in the two glass-ceramics B and C. Day and Rindone, in the study of the partly crystallized $Li_2O-2.75 SiO_2 (+0.0005 \% Pt)$ glass [4], noticed that the low-temperature peak linked with alkali ion diffusion markedly decreased with the increasing crystallinity and also disappeared when the amount in the crystal is very high (~78%). The fact that the peaks P_{1B} , P_{1C} , P_{2B} and P_{2C} , which occur in the glass-ceramics are not observed in glass A, is in agreement with the study of Li *et al.* They observed that for the $K_2O-Li_2O-Al_2O_3-SiO_2$ system glass-ceramic after crystallization, the peak observed in the glass has disappeared, however, two other internal friction peaks are present at higher temperature. Thus the peaks P_{1B} , P_{1C} , P_{2B} and P_{2C} are certainly linked with relaxation mechanisms occurring in the crystalline phase. There is a great similarity for the peak position and activation energy between the peaks P_{1B} , P_{1C} and the first peak observed in the $K_2O-Li_2O-Al_2O_3-SiO_2$ glass-ceramic, which appears at 343 K at 1Hz and has an activation energy of 0.96 eV [5]. Furthermore, as previously deduced, P_{1B} and P_{1C} are due to electrically charged entities. So, it is likely that the relaxation phenomena of P_{1B} and P_{1C} involve the lithium ions of the crystalline phase. However, as shown in Table 1, P_{1B} has a higher magnitude than P_{1C} and the relaxation parameters (E_a , τ_0) of these peaks are slightly different. These divergences could be explained by a different environment for the Li^+ ions of the crystal phase because B and C have different crystalline structure (hexagonal system for B and tetragonal for C).

As shown in Figures 2 and 4, it seems obvious that the relaxation phenomena responsible for P_{2B} and P_{2C} respectively are different. Furthermore, the disparity between the E_a and τ_0 values suggests that the relaxation units involved are not the same. As it was not possible to perform dielectric measurements in the appropriate temperature range, no information is available about the nature of the relaxation entities.

5. CONCLUSION

The investigated glass, which derived from the L.A.S. system, shows an internal friction peak close to 273 K, attributed to the stress-induced diffusion of lithium ions. After crystallization heat treatment, this peak does not appear any more but for each glass-ceramic two other peaks are observed. The first one, at low temperature (about 343K for 1Hz), is linked with Li^+ ions of the crystalline phase and its characteristics depend on the microstructural environment of lithium ions. The second one, which appears at higher temperature (between 600 and 700K) is largely different according to the considered glass-ceramic. This peak is due to entities which relax in the crystalline phase but the relaxation units and mechanisms implied are not identified.

References

- [1] Zdaniewski W.A., Rindone G. E., Day D. E., J. Mater. Sci., **14** (1974) 763-775.
- [2] Forry K.E., J. Am. Ceram. Soc., **45** (1957) 90-94.
- [3] Taylor T. D., Rindone G. E., Relax. Proc. in Glasses, (Day D. E. Ed., North Hol. Pub. Co, 1974) 157
- [4] Day D.E , Rindone G.E., J. Am. Ceram. Soc., **44** (1961) 161-167.
- [5] Li J., J. Non-Cryst. Solids, **80** (1986) 1-10.
- [6] Li J., Luo J., Chen H., Scienta Sinica, **13** (1964) 1888-1894.
- [7] Woigard J., Sarrazin Y., Chauvet H., Rev. Sci. Instrum., **48** (1977) 1322-1325.
- [8] Day D.E , Rindone G.E., J. Am. Ceram. Soc., **45** (1962) 496-504.
- [9] Ryder R.J., Rindone G.E., J. Am. Ceram. Soc., **43** (1960) 662-669.