

Mechanical relaxations in single and mixed alkali silicate glasses

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ABSTRACT

Isothermal mechanical spectroscopy (IMS) have been performed in single and mixed alkali silicate glasses ($(1-x)\text{Na}_2\text{O} \cdot x\text{Li}_2\text{O} \cdot 3\text{SiO}_2$ with $x=0, 0.25, 0.5, 0.75$).

Single alkali silicate glass $\text{Na}_2\text{O} \cdot 3\text{SiO}_2$ exhibits several relaxation peaks. Classically, a peak located at about 230 K at 1 Hz is assigned to the stress induced motion of alkali ions (ionic peak) and a second one (NBO peak) at about 500 K at 1 Hz is linked with a local rearrangement of the glassy structure involving non-bridging oxygen. Moreover, a small relaxation peak is evidenced for the first time at very low temperature (~ 185 K at 1 Hz). This new peak is attributed to the motion of the alkali ion inside its site.

Damping measurements carried out in the mixed alkali silicate glasses evidence the classical mixed alkali peak and point out new experimental features. Firstly, two satellite relaxation peaks are superimposed to the mixed alkali peak and correspond to the ionic and NBO peaks, respectively. Moreover, Arrhenius plot of the mixed alkali peak shows that the apparent activation energy is different below and above 340 K. These experimental results are discussed using the recent models for the alkali effect and the Meyer–Bunde model for mechanical relaxation in ionic glasses below the glass transition temperature T_g .

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1. Introduction

In single alkali silicate glasses, various peaks were previously observed: a first one, called “ionic peak” [1–4], occurring near room temperature at 1 Hz, is generally attributed to the action of the applied stress on alkali ions. At higher temperature, a second peak appears only in the presence of non-bridging oxygen and of water and is often called “non-bridging oxygen peak” (NBO peak) [2,4–7].

Mixed alkali silicate glasses exhibit a very large internal friction peak [4,5,8–12]. Some authors maintain that this peak takes the place of the ionic peak and others that it is superimposed to the peaks observed in the single silicate glasses. Another point of discussion concerns the results for mixed alkali glass with a very low ratio of a second alkali ion [5,8,11]: a small relaxation peak is described by the authors as a mixed alkali peak but in fact this peak can correspond to the NBO peak.

In order to clarify this situation, this work reports internal friction experiments performed using isothermal mechanical spectroscopy which allows very accurate measurements, for four silicate glasses of $(1-x)\text{Na}_2\text{O} \cdot x\text{Li}_2\text{O} \cdot 3\text{SiO}_2$ type with $x=0, 0.25, 0.5$ and 0.75 .

2. Experimental procedure

The apparatus used for the isothermal mechanical spectroscopy (IMS) experiments has previously been described [13]. In the case of forced vibrations, Q^{-1} is equal to $\tan \varphi$, where φ is the phase lag between the applied stress and the resulting strain. Isothermal experiments began 3 h after each temperature change, i.e. after stabilization of the sample microstructure. The measurement frequency ranged between 40 Hz and 10^{-4} Hz and ten frequencies per decade were used. Internal friction was also measured at the free frequency of the pendulum (≈ 200 Hz) by the free decay method. The raw internal friction values were corrected for taking the suspension stiffness into account [13]. The maximal strain amplitude was $\varepsilon_M = 5 \times 10^{-6}$. The dimensions of the specimens were $1.5 \text{ mm} \times 5 \text{ mm} \times 50 \text{ mm}$.

3. Experimental results

3.1. Single alkali silicate glass

Isothermal spectroscopy experiments performed on the single alkali silicate glass $\text{Na}_2\text{O} \cdot 3\text{SiO}_2$ exhibit three mechanical relaxation peaks. In fact, a new relaxation peak is observed at very low temperature (Fig. 1). The corresponding activation energy (11.8 kJ/mol) corresponds to the local motion of sodium ions inside their sites induced by to the elastic deformation of the vitreous structure.

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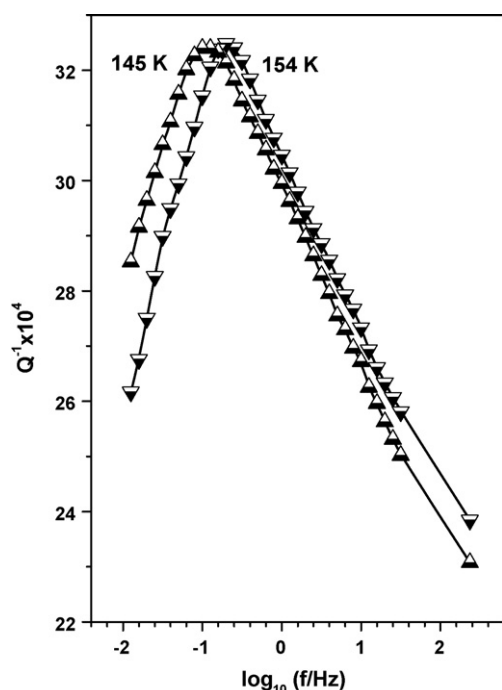


Fig. 1. Very low temperature internal friction peak in single alkali silicate glass $\text{Na}_2\text{O} \cdot 3\text{SiO}_2$ measured at 145 K (down triangles) and 154 K (up triangles).

The classical ionic peak, corresponding to the jump of sodium ions to a vacant neighbouring site, and the non-bridging oxygen peak are also observed in this sample. Fig. 2 shows the Arrhenius plots for the three relaxation peaks.

The apparent relaxation parameters are: $E_a \approx 58$ kJ/mol and $\tau_0 = 8 \times 10^{-14}$ s for the ionic peak and $E_a \approx 94$ kJ/mol and $\tau_0 = 1.5 \times 10^{-11}$ s for the non-bridging oxygen peak.

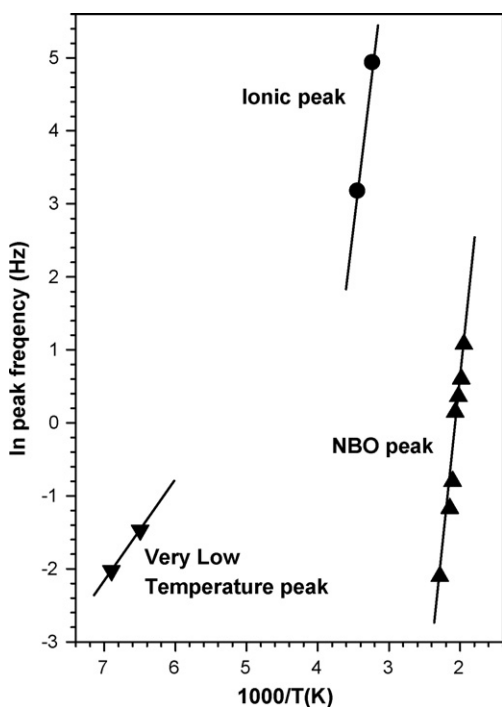


Fig. 2. Arrhenius plot of the three peaks in single alkali silicate glass $\text{Na}_2\text{O} \cdot 3\text{SiO}_2$: very low temperature peak (down triangles), ionic peak (circles) and NBO peak (up triangles).

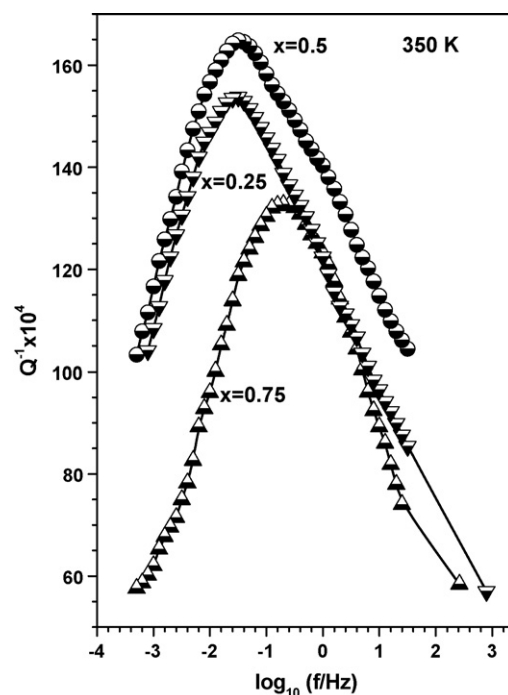


Fig. 3. Mixed alkali peak measured at 350 K in the three mixed alkali silicate glasses $(1-x)\text{Na}_2\text{O} \cdot x\text{Li}_2\text{O} \cdot 3\text{SiO}_2$ for $x=0.25$ (down triangles), 0.5 (circles) and 0.75 (up triangles).

3.2. Mixed alkali silicate glasses

For the three mixed alkali silicate glasses $(1-x)\text{Na}_2\text{O} \cdot x\text{Li}_2\text{O} \cdot 3\text{SiO}_2$, isothermal internal friction measurements exhibit an apparent peak of large amplitude. Fig. 3 shows the internal friction spectrum versus frequency for each glass ($x=0.25$, 0.5 and 0.75) at the same temperature 350 K. The height and the location of this apparent maximum depend on the glass composition.

From the isothermal experiments performed in frequency sweep, internal friction can also be plotted against the temperature at a given frequency, as illustrated in Fig. 4 at 0.0032 Hz for the $0.5\text{Na}_2\text{O} \cdot 0.5\text{Li}_2\text{O} \cdot 3\text{SiO}_2$ glass: the internal friction spectrum can be separated into a large intensity peak (mixed alkali peak) at about 330 K and, at higher temperature (≈ 410 K), a lower intensity peak (NBO peak) which appears as a shoulder at the bottom of the mixed alkali peak. Fig. 4 also shows the decrease in the modulus accompanying the relaxation peaks.

The ionic peak is also observed in mixed alkali silicate glasses. The Arrhenius plots corresponding to the ionic and NBO peaks in the single alkali glass and in the three mixed alkali glasses are reported in Fig. 5. The plots corresponding to the NBO peaks are quite similar for all the glasses. On the contrary, Fig. 5 shows that the ionic peak shifts to a lower frequency (at fixed temperature) for the mixed alkali glasses. This fact has been previously reported [4,5,8,10–12].

The Arrhenius plots corresponding to the mixed alkali peak are displayed in Fig. 6. For each sample, the experimental points cannot be aligned in a single straight line and the Arrhenius spectra correspond to two different energy activation values; the transition temperature being about 340 K. The experimental relaxation parameters are displayed in Table 1 for the three mixed alkali glasses.

The very large relaxation strength of the mixed alkali peak can be explained by recent theories about the mixed alkali effect [14–16]. According to these theories, both kinds of mobile ions have different jump paths. Mutual interception of these jump paths involves

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