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Thermal and structural properties of $20Li_2O$-$80TeO_2$ glasses

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Abstract – Tellurite $20Li_2O$-$80TeO_2$ glasses were prepared with identical nominal composition but with different glass-forming histories. Structural and thermal properties of these glasses were studied by using X-Ray Diffraction Analysis (XRD) and Differential Scanning Calorimetry (DSC) techniques to understand and control the crystallization process on this glass matrix. The γ-TeO_2, α-TeO_2 and α-$Li_2Te_2O_5$ phases were identified during the isothermal induced crystallization in this glass, where the γ-TeO_2, phase is a metastable structure. The measured activation energies showed that γ-TeO_2 and α-TeO_2 crystallizes at very close temperatures but distinct for the $Li_2Te_2O_5$ crystallization found in this glass. This result suggests that both γ-TeO_2 and α-TeO_2 phases first crystallizes in the glass matrix.

Physical properties of glasses depend strongly on the glass-forming histories. The adequate knowledge of the correlation between physical properties and the glass-forming history is indispensable for a correct interpretation of several processes in glasses, in particular the crystallization kinetic. In recent years, TeO_2-based glasses were extensively studied to understand different properties of these important glass matrixes and improve some specific characteristics of technological interest. Depending on the composition, some TeO_2 glasses present very interesting non-linear properties such as a third harmonic (χ^3) generation almost one order greater than some other important oxide glasses. However, the technological applications of these glasses require a profound comprehension of fundamental characteristics such as structural, thermal, optical and electrical properties. The purpose of the present work was to study the effects of the glass-forming history on structural and thermal properties on $20Li_2O$-$80TeO_2$ glasses by using the XRD and DSC techniques as investigative tools.

Glasses studied in the present work were prepared with nominal compositions $20Li_2O$-$80TeO_2$ but with different glass-forming histories. Therefore, stress free and stressed glasses were obtained. Powdered glasses with particle size 63-75 μm, 45-63 μm, 38-45 μm and < 38 μm were used for studies reported in this work. The structure of the crystallized glasses was analyzed by using XRD. DSC measurements were performed to study the thermal properties of the studied glasses. Activation energies were evaluated considering that observed DSC crystallization peak is a superposition of three Gaussian functions, associated to three distinct crystallization phases in the glass devitrification. This assumption was supported by XRD results.

The XRD results confirm the amorphous state for all as-quenched glasses used in the present investigation and that different thermal histories were not sufficient to produce any crystalline phase in these glasses. XRD results show the γ-TeO_2, α-TeO_2 and $Li_2Te_2O_5$ phase crystallization in the glass matrix during devitrification, where the γ-TeO_2 phase is considered a metastable structure. The structure of the α-TeO_2 are formed by three-dimensional network TeO_4 units sharing oxygen corners by symmetric Te-O-Te bridges while the γ-TeO_2 structure can be considered as a chain system, where TeO_4 units are alternatively linked by nearby and high symmetric Te-O-Te bridges. Based only on XRD it was not possible to determine whether α-TeO_2, γ-TeO_2 and $Li_2Te_2O_5$ phases crystallize simultaneously or at distinct onset crystallization temperatures. From DSC results, the obtained glass transition temperature (T_g) values were between 263-267ºC and the onset crystallisation temperature between 328-347ºC. The DSC heating curves of the stress-free glasses shows endothermic peaks close to T_g while DSC curves of the stressed glasses demonstrate a typical glass transition steps. This result provide a clear evidence that the observed endothermic peak close to T_g have no direct correlation with residual stress in the tellurite glasses studied in the present work. In accordance with XRD results, the asymmetry on DSC crystallization peaks suggests a multiphase crystallization in the glass. Under these considerations, each DSC crystallization peak was treated as a convolution of three individual peaks with distinct maxima temperature (T_p) to determine the activation energies. Results shows that $E_3 > E_2 \approx E_1$ (327 > 298 ≈ 296 kJ.mol^{-1}) for glass with particle size 63-75 μm, while $E_1 < E_2 \approx E_3$ (246 < 289 ≈ 293 kJ.mol^{-1}) is for glass with particle size smaller than 38 μm. Therefore, the energies E_1, E_2 and E_3 were associated to the following γ-TeO_2, α-TeO_2 and $Li_2Te_2O_5$ crystallization in the $20Li_2O$-$80TeO_2$ glass, respectively.