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Study of mixed ions effect in Li-Cs-Sr metaphosphate glasses by solid state NMR and impedance spectroscopy

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With the addition of a mobile ion species, ionic conductivity is introduced in a glass matrix, which makes these materials interesting for applications in batteries and sensors. However, when a second mobile ion species is added to the system, strong anomalies in the conductivity might be observed, reducing the values by orders of magnitude below the expected additive behavior. (1)This phenomenon is known as the mixed ion effect (MIE). The extent of the deviation is usually correlated with the size mismatch between the mixed ions, attaining several orders of magnitude for pairs such as Li-Na or Li-Rb. Other properties of the material dependent on ion diffusion also exhibit nonlinear behavior. A model that tries to explain the MIE is the Random Ion Distribution Model (RIDM) (2) whose hypothesis is the random mixture of mobile species and the presence of structurally different sites of occupation for each ion. To study the validity of the model, a vitreous system with intense MIE (Li-Cs metaphosphate) was disturbed by the addition of a third non-mobile specie (Sr), looking for conditions to inhibit MIE associated with the mixture of alkaline ions, according to the composition $(50 - x - y)\ Cs\_2O - x\ Li\_2O - y\ SrO \check{}50P\_2O\_5$, varying x from 0 to $37.5mol\%$ and y from 0 to $40\ mol\%$. Solid-state NMR, DSC and impedance spectroscopy (IE) techniques were used. With solid-state NMR, we observed linearity in the spectral line FWHM of the nucleus of ^{7}Li and ^{133}Cs as a function of their ionic density. For all Li-Cs ratios, we measured linear behaviors, indicating that variations of any of the ions do not affect the dilution of the others, indicative of the random mixture of ions, in accordance with the RIDM. In addition, through IE, we show that MIE is still present after the addition of Sr, which acts as blocking ion in the diffusion of Li and Cs, decreasing the conductivity globally.

Palavras-chave: Mixed Ions effect. Phosphate glass. Nuclear magnetic ressonance. Impedance sprectrocopy.

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