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Implications for the Structural Origin of the Increase in Configurational Heat Capacity with Increasing B_2O_3 Content in Borosilicate Glasses

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In this work, we explore the structural origin of the compositional evolution of the jump in isobaric heat capacity (ΔC_p) during the glass transition in a series of $(75q)B_2O_3$-$(75(1-q))SiO_2$-$15Na_2O$-10CaO glasses. ΔC_p represents the difference between the liquid C_{pl} and the glass C_{pg} and is usually approximated as the configurational heat capacity ($C_{p,conf}$), particularly for relative strong inorganic system. As determined by differential scanning calorimetry (DSC), ΔC_p is found to increase non-linearly when substituting B_2O_3 for SiO_2. By using Raman spectroscopy we investigate whether and how the increase in ΔC_p is associated with changes in glass structure, particularly in the intermediate range order (IRO) structure. Raman bands in the medium-frequency region of 550-810 cm^{-1} can be assigned to the vibrational modes of various superstructural units, reflecting the IRO. Our results demonstrate a possible link between the evolution of IRO structures and ΔC_p, which reflects the number of minima in a multidimensional potential energy surface. In the silica-rich compositions, two types of borosilicate units exist, leading to the rapid increase of ΔC_p with B_2O_3 content. In the compositions with similar concentration of B_2O_3 and SiO_2, the concentration of borosilicate units and borate superstructures change in an opposite way, leading to an approximate constant value of ΔC_p as a function of composition. In the boron-rich compositions, the presence of six-membered borate rings with one and two $[BO_{4/2}]^-$ groups are responsible for the increase of ΔC_p with B_2O_3 content.