

Excess heat capacity and entropy of mixing in high structural state plagioclase

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ABSTRACT

Low- and high-temperature heat capacities for a series of synthetic high structural state plagioclase crystals (Ab-An) were measured using both a relaxation calorimeter and a differential scanning calorimeter. The measurements were performed at temperatures between 5 and 800 K on milligram-sized polycrystalline samples that had been characterized in a previous study. The data show positive excess heat capacities of mixing at temperatures below 300 K with a maximum value of ~ 2 J/(mol·K). Below ~ 70 K, the excess heat capacities exceed two standard deviations and are thus significant. Above 300 K, the measurements indicate negative excess heat capacities with a maximum of ca. -1.5 J/(mol·K) at about 400 K, and do not exceed two standard deviations. The excess vibrational entropies of mixing are positive with an asymmetric variation. At $T = 298.15$ K, the largest deviation from ideal behavior occurs at Ab₂₀An₈₀ amounting to $\Delta S_{\text{vib}}^{\text{ex}} = 2.8 \pm 2.4$ J/(mol·K). An asymmetric Margules mixing model was found to adequately describe the vibrational entropy-composition behavior, yielding $W_{\text{AbAn}}^{\text{Svib}} = 16.4$ J/(mol·K) and $= W_{\text{AnAb}}^{\text{Svib}} 4.7$ J/(mol·K).

Keywords: Feldspar, plagioclase solid solution, low- and high-temperature heat capacity, excess heat capacity of mixing, vibrational entropy of mixing