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Heat-pulse calorimetry measurements on natural chlorite-group minerals

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Abstract

Low- and high-temperature heat capacities of five natural chlorite-group samples were measured using the heat-capacity option of the Physical Properties Measurement System (Quantum Design), which is based on the principles of heat-pulse calorimetry, and by differential scanning calorimetry. Comprehensive chemical analyses were performed on these samples by electron microprobe analysis, by inductively coupled plasma mass spectrometry, and by Karl-Fischer titration (for H_2O). The natural chlorites span a range in X_{Fe} from 0.052 to 0.885 with increasing Al-content due to the Tschermak substitution with increasing X_{Fe}.

The measured heat capacities were extrapolated to the end-member compositions of chamosite and clinochlore. Integration of heat-capacity data yields the calorimetric standard entropies of chamosite $(Fe_5Al)[Si_3AlO_{10}](OH)_8$ and clinochlore $(Mg_5Al)[Si_3AlO_{10}](OH)_8$, with values of 572.0 ± 0.2 and 425.6 ± 0.4 J/(mol·K), respectively. The C_P-polynomial for end-member chamosite is $C_P = 1151.7 - 8.4564 \times 10^3 \cdot T^{-0.5} - 13.206 \times 10^6 \cdot T^{-2} + 15.233 \times 10^8 \cdot T^{-3}$ [J/(mol·K)], valid in the temperature range of 298.15–900 K, and that for end-member clinochlore is $C_P = 1160.5 - 9.9819 \times 10^3 \cdot T^{-0.5} - 5.9534 \times 10^6 \cdot T^{-2} + 3.8677 \times 10^8 \cdot T^{-3}$ [J/(mol·K)], valid in the temperature range of 298.15–1000 K.

The Fe-rich chlorites exhibit an asymmetric distribution of the excess heat capacity in a plot of C^{ex}_P vs. T, with a maximum at about 52 K. By analogy to annite, we interpret this peak to represent the magnetic ordering temperature.

Based on our standard entropy value for chamosite, the enthalpy of formation of berthierine $(Fe_{2.5}Al_{0.5})[Si_{1.5}Al_{0.5}O_5](OH)_4$ was estimated as –3570.30 kJ/mol using a reported onset temperature of 70 °C at 16 MPa for the berthierine-chamosite polymorphic transition.

Keywords: Clinochlore, chamosite, heat pulse calorimetry, heat capacity, excess heat capacities, standard entropy, chlorite group minerals