

Transmission electron microscopy and differential thermal studies of lazurite polymorphs

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ABSTRACT

Transmission electron microscopy (TEM) and thermal (DTA-TG) analyses of lazurite, $(\text{Na,Ca})_8[\text{Al}_6\text{Si}_6\text{O}_{24}](\text{SO}_4,\text{S})_2$, indicate three forms: cubic, modulated, and orthorhombic. Cubic lazurite contains mainly $[\text{Na}_3\text{CaSO}_4]^{3+}$ clusters that are disordered together with the O1 and O2 oxygen atoms, and no satellite reflections occur. In the modulated lazurite structure, $[\text{Na}_3\text{CaSO}_4]^{3+}$ and $[\text{Na}_3\text{CaS}]^{3+}$ clusters are ordered and possibly cause ordering of the framework oxygen atoms on the O1 and O2 positions and produce satellite reflections. Different ordering of these clusters gives an orthorhombic supercell with parameters of $2d_{110} \times 6d_{1\bar{1}0} \times c$ with respect to the cubic subcell. The DTA-TG study indicates that the orthorhombic to the modulated phase transition occurs at a peak temperature of 489 °C. A single-crystal fragment of lazurite may contain all three phases, as was observed by TEM in this study. Such intergrowths indicate a continuous framework with different regions of the crystal containing different ordering and chemistries. Two new lazurite superstructures were observed with dimensions of $6d_{110} \times 3d_{001}$, and $3d_{112} \times 3d_{\bar{1}11}$ with respect to the cubic subcell.