TEM study of mullite growth after muscovite breakdown

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ABSTRACT

Mullite (Mul) formation after high-T muscovite (Ms) breakdown has been studied in phyllosilicaterich bricks. At $T \ge 900$ °C Ms dehydroxylation is followed by partial melting that triggers the nucleation and growth of Mul acicular crystals. An analytical electron microscopy study reveals that the Mul is a 3:2-type with a ${}^{[6]}(Al_{1.686}Ti_{0.031}Fe_{0.159}Mg_{0.134}){}^{[4]}(Al_{2.360}Si_{1.649})O_{9.82}$ formula and an O atom vacancy of x = 0.18. This is consistent with X-ray diffraction results [i.e., unit-cell parameters: a =7.553(7), b = 7.694(7), and c = 2.881(1) Å, V = 167.45 Å³]. The initial stage of the process resulting in Mul growth followed the balanced reaction Ms $\rightarrow 0.275$ Mul + 0.667Melt + 0.244K₂O + 0.01Na₂O + 0.125H₂O, yielding an alkali-poor peraluminous melt. H₂O with K (and Na), which are lost along the (001) planes of dehydroxylated Ms, play a significant role as melting agents. The c-axes of the Mul crystals are oriented parallel to $[010]_{ms}$ or to the symmetrically equivalent $\langle 310 \rangle_{ms}$ zone axis, while the $(120)_{mul}$ or $(210)_{mul}$ planes are subparallel to $(001)_{ms}$ (TEM results). These systematic orientations point to epitaxial Mul nucleation and growth on the remaining Ms substrate, which acts as a template for Mul heterogeneous nucleation. Randomly oriented Mul growth is also observed during the late stages of the process (i.e., melt cooling). The epitaxial nature of Mul growth after dehydroxylated Ms melting minimizes the energy requirement for nucleation. In addition, the water released after Ms breakdown and the multicomponent nature of the melt enable this high-T aluminum silicate to grow at $T \sim 900$ °C, almost 100 °C below the SiO₂-Al₂O₃-K₂O ternary system eutectic (after a melt with an end-member Ms composition).