

Study of the Al coordination in mullites with varying Al:Si ratio by ^{27}Al NMR spectroscopy and X-ray diffraction

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ABSTRACT

Mullite is an aluminosilicate of the composition $\text{Al}_2(\text{Al}_{2+2x}\text{Si}_{2-2x})\text{O}_{10-x}$ with x generally ranging between 0.2 and 0.5. XRD and ^{29}Si and ^{27}Al nuclear magnetic resonance (NMR) have been used to investigate the structure of various mullite compositions ($x = 0.26, 0.36, 0.42,$ and 0.69) as well as sillimanite ($x = 0$) and $\gamma\text{-Al}_2\text{O}_3$ ($x = 1$). ^{27}Al magic angle spinning (MAS) NMR of the central and satellite transitions have been used to determine the chemical shifts and quadrupole interaction parameters for the various AlO_4 and AlO_6 units. The isotropic chemical shifts of the various units are 5.9 ppm for AlO_6 , 69.1 ppm for $\text{AlO}_4(\text{T})$, 55.0 ppm for $\text{AlO}_4(\text{T}')$, and 45.7 ppm for $\text{AlO}_4(\text{T}^*)$ where $\text{AlO}_4(\text{T})$ denotes the aluminum tetrahedra in the double chains, and $\text{AlO}_4(\text{T}')$ and $\text{AlO}_4(\text{T}^*)$ those next to the oxygen Oc^* . Quantitative numbers of the aluminum occupancy of these sites have been determined for the various powder samples. These results are in good agreement with the average structure model of mullites. NMR and XRD proved the presence of impurities of kyanite in natural sillimanite and of $\gamma\text{-Al}_2\text{O}_3$ in the mullites with $x = 0.42$ and 0.69 .