

Solid-state ^{29}Si MAS NMR studies of illite and illite-smectite from shale

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ABSTRACT

A new method to extract quantitative information from poorly resolved ^{29}Si magic-angle spinning (MAS) nuclear magnetic resonance (NMR) spectra of natural mixed-layer illite-smectite (I-S) clays is presented. The Si-Al distribution in layered aluminosilicates are used to link the intensities of ^{29}Si resonances from all $\text{Q}^3(n\text{Al})$ sites ($n = 0, 1, 2, 3$) to the tetrahedral layer aluminum substitution by applying Loewenstein's aluminum avoidance principle (no Al-O-Al linkages) extended to ensure a homogeneous distribution of charge. In addition, correlations between ^{29}Si chemical shifts and the Al substitution are established for illite resonances by computer fitting of well-resolved phyllosilicate spectra. Combination of these two constraints led to a general procedure for iterative fitting of ^{29}Si MAS NMR spectra of clay minerals containing high-charge (illite-like) and low-charge (smectite-like) sites. The applicability of the new method is demonstrated for two I-S samples from Cambrian black shale in the Baltic area and two I-S samples from Upper Jurassic oil-source rock in the Central Trough of the North Sea. In combination with data from XRD and chemical analysis, the results from ^{29}Si MAS NMR enables determination of the composition for the entire I-S particles.