Structure and Thermal Transformations of Imogolite Studied by ^{29}Si and ^{27}Al High-Resolution Solid-State Nuclear Magnetic Resonance

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Abstract: Solid-state nuclear magnetic resonance (NMR) spectroscopy, thermal analysis, and X-ray powder diffraction data on the tubular, hydrous aluminosilicate imogolite were found to be fully consistent with a previously proposed crystal structure consisting of a rolled-up, 6-coordinate Al-O(OH) sheet, bonded to isolated orthosilicate groups. The calculated ^{29}Si chemical shift of this structure agreed with the observed shift within 3 ppm. Thermal dehydroxylation of the Al-O(OH) sheet produced predominantly NMR-transparent 5-coordinate Al, but a few 4- and 6-coordinate sites and some residual hydroxyl groups may also have formed, as shown by NMR spectroscopy. Changes in the ^{29}Si NMR spectrum on dehydroxylation suggest a condensation of the orthosilicate groups, but steric considerations rule out bonding between adjacent silicons. To account for these observations, an alternative mechanism to orthosilicate condensation has been proposed, involving the fracture and unrolling of the tubes, followed by the condensation of fragments to form a layer structure. The layer structure has a calculated ^{29}Si chemical shift of -95.6 ppm, in good agreement with the observed value of -93 ppm.

Key Words: Aluminosilicate gel • Crystal structure • Dehydroxylation • Imogolite • Nuclear magnetic resonance • Thermal treatment • X-ray powder diffraction

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