Study of the transformation mechanism of kaolinite to illite at high pressure by ^{29}Si and ^{27}Al MAS NMR

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Experimental illitization of kaolinite in aqueous solutions containing K^+ has been the subject of study for a number of years since Velde,[1] in 1965, synthesised muscovite from kaolinite and KOH solutions at 300 ºC. Most of the studies deal with the kinetics of kaolinite illitization using low solid/liquid ratios up to a maximum of 200 mg/mL.[2,3,4] The authors accept a dissolution – precipitation mechanism, which is clearly favoured by low solid/liquid ratios. High values of the solid/liquid ratio would be expected to favour a solid state-like reaction. The aim of the present study is to analyse the reaction mechanism of kaolinite to illite in KOH solution using a high solid/liquid ratio. Special emphasis has been made on the first reaction stages, when incipient illite nuclei are being formed from kaolinite layers. The methodology followed consists in the hydrothermal treatment, at 500 bars, of a powdered kaolinite in a KOH solution, using a solid/liquid ratio = 1000 and increasing reaction times (from 1 to 24 hours). The transformation of the Si and Al environments has been followed by ^{29}Si and ^{27}Al MAS NMR spectroscopy and X-ray diffraction. The results indicate that the first stage in the illitization of kaolinite is the diffusion of Al from the octahedral to the tetrahedral sheet of the kaolinite layers (Figure 1). The Al diffusion gives rise to partially transformed kaolinite layers which must be the precursors of the illite/muscovite nuclei. These nuclei require the condensation of an additional Si-Al tetrahedral sheet to be formed. Illite/muscovite coherent diffraction domains only form after 13 hours treatment; at shorter reaction times, proper illite/muscovite layers, with a basal spacing of 10 Å characteristic of TOT layers with interlayer K^+, are not observed as indicated by the absence of any reflection at 8.6 º2θ on the XRD diagrams.

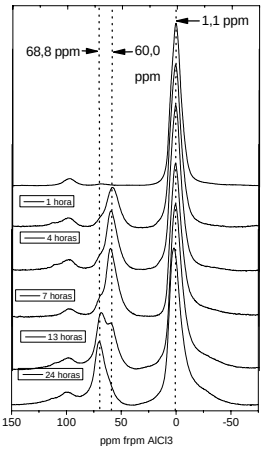

Figure 1: ^{27}Al MAS NMR spectra of kaolinite samples submitted to HT treatment in KOH solution at increasing times

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[4] Bauer, A., Velde, B., Berger G. *Appl. Geochem*. **1998**, 13, 619-629.

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