Thermal transformation of clay minerals probed by in situ soft-X-ray absorption spectroscopy

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ABSTRACT

For the past 20 years, the LUCIA beamline has provided the scientific community with soft-X-ray absorption spectroscopy (XAS) experiments to measure the K-edge of light elements such as Al, Si and Mg.[1] These elements are constituents of a large number of mineral phases, particularly clay minerals. A recurring question is determining how the atomic coordination in these nanostructures changes in response to external activation.

Thermal activation is one of the most commonly applied process to modify the structure, porosity and surface reactivity properties of aluminosilicate clay minerals. During thermal annealing, clay materials may proceed through intermediate and metastable ('meta') states, which are particularly suitable for use in the ceramics industry[2] or in the development of low-carbon cements.[3] Despite the strong interest in these phases, an overall description of the thermally induced atomic reorganization in clay minerals is missing.

The objective of this presentation is to provide an overview of the performance of the LUCIA beamline for in situ temperature measurements using a compact furnace capable of thermal annealing up to 1000°C while acquiring XAS fluorescence spectra in flyscan mode. This will be illustrated using natural halloysite nanotubes ($Al_2Si_2O_5(OH)_4$)[4] and synthetic imogolite-type nanotubes ($Al_2(Si,Ge)O_3(OH)_4$).[5,6] We will demonstrate that it is possible to obtain new insights on the mechanisms of structural reordering during thermal transformation through the quantitative analysis of the acquired spectra using a multivariate curve resolution alternating least squares method. The findings will highlight the influence of initial morphology (tube vs. sphere vs. platelet) or chemical composition (e.g. Si vs. Ge) on thermal reactivity of the clay minerals. From a practical point of view, the comparison of different natural and synthetic samples chemically similar shows important differences on the temperature onset at which the octahedral Al site conversion begins, which could be economically attractive in terms of energy costs for industrial applications. These results also open up perspectives for (re)examining the temperature-dependent transformation of other 'oxide' phases using in situ low-energy XAS.

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