

## Saponite crystallization via an amorphous intermediate

ROGIER BESSELINK,<sup>1</sup> TOMASZ M. STAWSKI,<sup>1</sup> HELEN M. FREEMAN,<sup>1</sup> JÖRN HÖVELMANN,<sup>1</sup> DOMINIQUE J. TOBLER,<sup>2</sup> LIANE G. BENNING.<sup>1,3,4</sup>

<sup>1</sup>German Research Center for Geosciences, GFZ, Telegrafenberg, 14473, Potsdam, Germany, [rogier@gfz-potsdam.de](mailto:rogier@gfz-potsdam.de), [stawski@gfz-potsdam.de](mailto:stawski@gfz-potsdam.de).

<sup>2</sup>Nano-Science Center, Department of Chemistry, University of Copenhagen, Copenhagen, Denmark

<sup>3</sup>Department of Earth Sciences, Free University of Berlin, 12249 Berlin, Germany. [benning@gfz-potsdam.de](mailto:benning@gfz-potsdam.de)

<sup>4</sup>School of Earth and Environment, University of Leeds, Leeds, United Kingdom.

Clays are the most abundant minerals on the Earth's surface and are one of the main sources of iron and magnesium in soils. They are typically formed through weathering of primary rock-forming minerals. Such weathering reactions are driven by fluid-rock interactions and they often occur at Earth surface conditions. However, the formation pathways and mechanisms of how clays form at ambient conditions are not well understood and so far most synthetic procedures attempting to give insights into the plausible formation mechanisms rely on synthesis methods at hydrothermal conditions.

Here, we present data on a 'green' synthesis method for magnesium-saponite, a clay belonging to the smectite family. We successfully synthesized saponite at atmospheric pressures and moderate temperatures (~ 25 to 95°C) and characterized the resulting materials at various stages of the reaction by synchrotron-based high-energy X-ray diffraction combined with pair distribution analyses (HEXD and PDF), as well as Fourier transform infrared (FTIR) spectroscopy and transmission electron microscopy (TEM).

Our results reveal that saponite formed via a two stage process: 1) a rapid nucleation and growth (within 5 minutes), of a Mg-rich amorphous intermediate in which ~20 % of the magnesium became incorporated within a boundary layer of the aluminosilicate network, followed by 2) a much slower crystallization mechanism (> 4h at 95 °C), where the remaining magnesium became incorporated into the developing nanocrystalline, and sheet-like saponite structure. We show that stage one - this rapid incorporation of magnesium within a boundary layer of the amorphous aluminosilicate precursor phase plays a key role in formation of saponite and other Mg-rich clays.