Characterisation of cronstedtite synthesized by iron clay interaction in a cooling procedure

$$\label{eq:intermediated} \begin{split} I. Pignatelli^1, E. Mugnaioli^2, J. Hybler^3, \\ R. Mosser-Ruck^1 \text{ and } M. Cathelineau^1 \end{split}$$

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The cooling of steel containers in radioactive waste storage has been simulated by an original step by step experiment from 90°C to 40°C. Among newly formed clay minerals observed in run products, cronstedtite has been undoubtedly identified by different analytical techniques (XRD, TEM and SEM). This is the first time that cronstedtite is so abundant and well-crystallized in an iron-clay interaction experiment. The formation of cronstedtite is favoured by the release of Fe and Si in solution after the dissolution of quartz, T-O-T phyllosilicates and iron metal. Cronstedtites are characterised by various morphologies: pyramidal (truncated or not) crystals have been observed from 90°C to 60°C experiments, whereas conic crystals with a rounded or hexagonal cross-section occur only in 90° and 80°C experiments. The mean formulae of pyramidal and conic crystal are very similar:

 $(Fe^{2_{+}}{}_{2.17}Fe^{3_{+}}{}_{0.80}Mg_{0.03})(Si_{1.20}Al_{0.07}Fe^{3_{+}}{}_{0.73})O_{5}(OH)_{4}$

Pyramidal crystals are, thus, more frequent and show different polytypic sequences as well as various degree of disorder. Polytypes $2M_1$, 1M and 3T have been identified thanks to TEM investigations. The two monoclinic polytypes are very rare in nature, at the contrary of 3T polytype (Hybler et al 2008). Thanks to the cooling procedure, the stability range of cronstedtite with respect to the temperature is determined. Between 90°C to 60°C cronstedtite appears to be stable and 50°C corresponds to the lower limit of cronstedtite stability as confirmed by crystal alteration. The occurrence of cronstedtite seems to be favored both by temperature range (90°-60°C) and by the progressive temperature decrease, as shown by the comparison of our results with those of previous studies carried out on the same starting claystone but with slightly different experimental conditions (duration time, liquid/rock or iron/clay ratios).

Evidence for global metasomatic enrichment in oceanic lithosphere

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Oceanic lithosphere is generally interpreted as mantle residue after MORBs extraction. However, some models [1, 2] suggest that this lithosphere could be re-enriched by metasomatic processes in the periphery of ridges. Xenoliths from OIBs could not be used to confirm or invalidate this hypothesis because the metasomatism observed in these xenoliths is likely to be related to the hot spot activity itself. Direct evidence for refertilization of oceanic lithosphere at a global scale is thus missing. Here we used Petit-spot lavas and their enclosing xenocrysts to suggest that oceanic lithosphere is likely to be metasomatized before its recycling in subduction zones.

Petit-spot lavas are interpreted as the surface expression of low degree melts extracted from the base of the lithosphere by plate flexure or crack propagation [3]. Petit-spot lavas accreted in the north of Costa Rica [4] show the presence of cpx xenocrysts similar to cpx observed in metasomatic veins wordwide. The composition of these cpx xenocrysts suggests crystallization at high pressure (15-25 km) in a differentiated liquid percolating across the lithosphere. Similar liquids are identified in Petit-spot lavas sampled on the Pacific plate in front of Japan [5]. These rocks do not contain high-pressure cpx xenocrysts, but contain low-Mg# cpx interpreted as cpx crystallized at low pressure in a liquid similar in composition to that from which cpx xenocrysts observed in Costa Rica petit-spot lavas crystallized. These different observations suggest that the lithosphere is re-enriched by metasomatic process before its subduction. Plate flexure allows low degree melts present at the base of the lithosphere to percolate and differentiate across the oceanic lithosphere, producing a refertilisation of this lithosphere, and in some cases the emission of Petit-spot lavas at the surface. Modelling of the effect of this metasomatic enrichment on the composition of the lithosphere suggests that portions of recycled oceanic lithosphere could correspond to an enriched mantle component rather then depleted mantle.

[1] Halliday et al (1995) *EPSL* **133**, 379-395; [2] Niu and O'Hara (2003), JGR 108; [3] Vallentine & Hirano (2010), *Geology* **38**, 55-58; [4] Buchs et al (2013), G3 [5]. Hirano et al (2006), *Science* **313**, 1426-1428.