The kinetics of solvent-mediated trasformation of hydromagnesite into magnesite at different temperatures and pressures

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The crystallization of magnesite ($MgCO_3$), siderite ($FeCO_3$), and $(Mg,Fe)CO_3$ solid solutions has gained increasing attention in the last few years, due to the relevance of these phases in the carbonation of silicate minerals and other phenomena related to the geological storage of the anthropogenic CO_2 emissions [1]. Despite this increasing interest there are still many aspects that remain poorly known, some of them concerning the precipitation behavior of magnesite.

Although the thermodynamic stability of magnesite has been considered to be higher than the stability of other related mineral phases, the precipitation of this mineral at room temperature seems to be strongly hindered by kinetic reasons. Hydromagnesite $Mg_5(CO_3)_4{\cdot}(OH)_2{\cdot}4H_2O$, and other hydrated carbonates tend to precipitate under these conditions, in many cases as low-crystallinity phases. The subsequent solvent-mediated transformation of these precursor phases requires a extremely long induction period at temperatures < 60ºC. Here we investigate the kinectics of the coupled dissolution, nucleation, and growth processes occurring during the solvent-mediated transformation of hydromagnesite into magnesite at different temperatures (50-180 ºC) and pressures (from 1 to 10 bars). The starting hydromagnesite was obtained by mixing $MgCl_2$ and Na_2CO_3 aqueous solutions at 25°C. The precipitate was then aged in a closed reactor. The evolution of the aqueous phase was monitored by ICP-MS and alkalimetry. The mineralogical compositon of the solids was studied by powder X-ray diffraction and Rietveld analysis. From these data we have explored the transformation behaviour by analyzing the shape of the “desupersaturation profiles”, according to the protocol proposed by Cardew & Davey [2] for solvent-mediated transformations. Kinetic modelling based on the Avrami equation was also performed in order to obtain an empirical activation energy for the whole transformation process.

[1] Oelkers & Schott (2005*) Chem. Geol***.** **217**, 183-186 [2] Cardew & Davey (1985) *Proc. R. Soc. Lond. A* **398**, 415-428