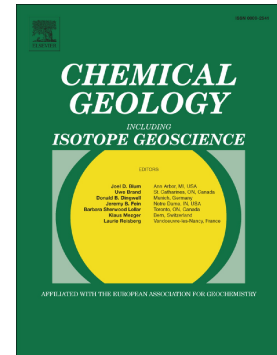


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Theoretical analysis of the kinetics of precipitation of lizardite and magnesite from olivine alteration.

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The present theoretical study analyzes the kinetic characteristics of precipitation of magnesite and lizardite as a result of hydrothermal alteration of olivine under high carbonate alkalinity. It makes use of a simulation strategy in which, both the chemical evolution of the aqueous solution and the solid phases are considered. The simulation is performed with the help of the NANOKIN code. It includes a full treatment of speciation processes in the aqueous solution, a rate equation for the dissolution of olivine, and a full account of nucleation and growth processes during the formation of secondary particles. The comparison between experimental (Lafay et al., 2014) and simulation results puts strong constraints on the simulation parameters, in particular those related to the nucleation and growth of the particles. Modeling points to an incongruent olivine dissolution and reveals a transitory precipitation of SiO₂, not detected experimentally, showing that the fate of Si is more complex than a mere incorporation in lizardite. It highlights how lizardite, magnesite and SiO₂ compete for the incorporation of silicon and magnesium released by the olivine dissolution.

Keywords: hydrothermal alteration, CO₂, nucleation and growth, computer simulation, NANOKIN code, nanoparticles, olivine, magnesite, lizardite

PACS numbers:

I. INTRODUCTION

In the context of the increasing global warming of the Earth atmosphere, thought to be essentially due to anthropogenic CO₂ emissions, artificial weathering of magnesium bearing silicates, widespread in the Earth crust, has been proposed as a promising solution for geological storage of CO₂. During such an industrial process, gaseous CO₂ would be dissolved into an aqueous solution subsequently injected into the geological reservoir. The drop in pH in the solution due to CO₂ would induce the silicate dissolution before its carbonation. Understanding the reaction mechanisms, the kinetics and the role of pressure, temperature and pH has stimulated numerous experimental studies of the serpentinization or carbonation of various silicates (Luce et al., 1972; Chen and Brantley, 2000; Oelkers, 2001; James et al., 2003; Giammar et al., 2005; Bearat et al., 2006; Hänchen et al., 2006; Prigiobbe et al., 2009; Andreani et al., 2009; Garcia et al., 2010; King et al., 2010; Daval et al., 2011; Malvoisin et al., 2012; Gadikota et al., 2014).

These studies mainly focused on the determination of kinetic rate laws of dissolution as those control the availability of chemical elements from the altered minerals. However experimental approaches generally show coexisting dissolution and precipitation, which opens the question of feed-back effects and complex coupling between secondary mineral precipitation and the dissolution process that initiates the whole alteration process. This was, in particular, the case for Lafay et al. (2014),

who recently studied the simultaneous precipitation of lizardite and magnesite from hydrothermal alteration of olivine under high-carbonate alkalinity in the lab. This work raised interesting questions on the competitive incorporation of magnesium and silicon released by the dissolution of olivine, into the two secondary mineral phases.

In the present work, we complement these lab experiments by a simulation strategy allowing a better understanding of these complex processes which coexist in natural hydrothermal systems. Our kinetic modeling treats on an equal footing dissolution and precipitation processes. It makes use of the NANOKIN home made simulation code (Noguera et al., 2006a,b; Fritz et al., 2009), which allows an account of ion speciation in the aqueous solution, dissolution, nucleation and growth processes. It has been recently applied to decipher the mechanism of calcite formation by hydrothermal carbonation of portlandite (Fritz et al., 2013), as well as to analyze the characteristics of amorphous silica nanoparticles precipitation under various thermodynamic conditions (Noguera et al., 2015).

The paper is organized as follows. Section II provides information on the synthesis route and a thorough interpretation of thermogravimetric analysis results (TGA). The numerical method is presented in Section III and the choice of the parameter values is discussed. The main results on the alteration of olivine are presented in Section IV. In the last section (Section V), the incongruent olivine dissolution and the existence of a transitory SiO₂ secondary phase are discussed. A preliminary attempt to

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