

## Heterogeneous carbonation in the MgO-H<sub>2</sub>O-CO<sub>2</sub> System

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Carbonation of natural brucite is carried out in aqueous environments at room temperature and moderate CO<sub>2</sub> pressure to examine the product mineral phases and reaction kinetics. Two sets of initial conditions are examined: brucite in pure H<sub>2</sub>O v. s. in diluted (1%) HCl. Time-dependent XRD analysis shows that carbon fixation process begins within 30 min of the experiments irrespective of the original makeup of the slurry. XRD and FT-IR reveal that nesquehonite is by far the dominant C-bearing species in the mineral product assembly with minor components being basic Mg-carbonate hydromagnesite and dypingite in water and chloride-bearing artinite in HCl. However, thermodynamic calculation suggests that such assembly is most likely kinetically favored because the experimental conditions are more saturated with respect to hydromagnesite and magnesite than to nesquehonite. A pseudo first-order rate law is found to best describe the time-dependent measurements for both settings. Moreover, fitting the rate expression yields a higher rate constant for the experiments performed in HCl solutions. The faster kinetics implies that the carbonation reaction may be a multi-stepped process, involving first the dissolution of brucite and CO<sub>2</sub> to generate Mg<sup>2+</sup> and CO<sub>3</sub><sup>2-</sup>, followed by precipitation of magnesium carbonate phases from aqueous solutions. This leads to our proposition that direct heterogeneous reaction between CO<sub>2</sub> and solid phase Mg(OH)<sub>2</sub> is probably not the pathway for the overall carbonation process. Assuming the upper limit of carbon content C<sub>max</sub> = 8.7% (based upon that of nesquehonite), measured total carbon in the product C<sub>tot</sub> show a carbonation rate of 83.9% and 94.3% for brucite in HCl and DDW at the end of 2.5 hr experiments. However, significant amount of brucite (~ 30 to 40%) remains unreacted in HCl, sharply contrasting to < 5% in DDW. Solution chemistry analysis indicates that most CO<sub>2</sub> stays in aqueous phase in both media; however, the concentration of HCO<sub>3</sub><sup>-</sup> and pH in HCl dip below those in H<sub>2</sub>O after about 2 hr, suggesting lower saturation may be responsible for the inferior carbonation extent in HCl.

## Origin of diagenetic fluid: Constraints from *in situ* analyses of stable isotopes and trace elements in carbonate veinlet and wallrock from the Ediacaran system in South China

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To retrieve the original signatures of sedimentary geochemistry in carbonates requires chemical and isotopic separation of precipitation fluid from diagenetic fluid. However, it is a challenge to trace the origin of diagenetic fluid in sedimentary limestone and dolostone that suffered the influence of freshwater and terrigenous weathering. In order to constrain the origin of diagenetic fluid in postglacial carbonates, *in situ* analyses of C-O isotopes and trace elements were carried out on carbonate veinlet and wallrock from the Upper and Lower Units of the Lantian Formation in South China, which is equivalent to the Doushantuo Formation elsewhere in South China and to the upper part of the Ediacaran system elsewhere in the world.

In the Upper Unit, δ<sup>13</sup>C-δ<sup>18</sup>O values and REE+Y patterns for veinlet are similar to those for wallrock, indicating the internal origin of diagenetic fluid from pore water and thus precipitation water. In the Lower Unit, however, the geochemical features of veinlet are highly different from those of wallrock, suggesting an external origin of diagenetic fluid. The REE+Y patterns for both veinlet and wallrock in the two units differ from those for normal marine carbonates, suggesting incorporation of terrigenous weathering by freshwater into the depositional basin.

There are negative and positive correlations between the δ<sup>13</sup>C and δ<sup>18</sup>O values for the Upper and Lower Units, respectively. This indicates that the isotopic compositions of wallrocks in the two units were influenced by different origins of diagenetic fluid. Both veinlet and wallrock in the Upper Unit are similar to each other with unusual depletion of <sup>18</sup>O, indicating their deposition from continental deglacial meltwater. The variation in δ<sup>18</sup>O values are probably caused by different proportions of mixing between meltwater and seawater during carbonate precipitation in addition to precipitation temperature. On the other hand, the high δ<sup>13</sup>C and δ<sup>18</sup>O values for the wallrock of the Lower Unit suggest that the precipitation water is similar to the seawater, whereas the low δ<sup>13</sup>C and δ<sup>18</sup>O values for the veinlet that are consistent with those for the Upper Unit indicate the action of diagenetic fluid derived from the continental deglacial meltwater.