The Formation of Secondary Phases in Olivine Carbonation Reactions

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The natural weathering of silicate minerals like olivine via a carbonation reaction is one possible method for the sequestration of anthropogenically produced carbon dioxide. The production of secondary phases other than carbonates can decrease the reactivity and cost efficiency of this procedure as they can form passivating layers on the olivine surface. These secondary phases may also have a detrimental effect on the porosity of the host rock during in-situ carbon dioxide sequestration. Therefore, it is important to understand how reaction conditions govern the development of these secondary phases, including the effect of carbon dioxide on their precipitation. The formation of phases such as serpentines can remove the dissolved Mg from solution and, therefore, limit the production of the carbonate minerals required. Bearat et al. (2006) have also observed the formation of a passivating amorphous silica layer on olivine surfaces. We have conducted batch experiments using teflon-lined steel autoclaves over a range of temperatures, up to 200 °C, using single crystals and powdered olivine with various fluid compositions. Examination of the reaction layer using scanning electron microscopy showed that an iron oxide phase developed during reaction. This did not occur in experiments with solution of pH 12. Experiments using olivine powders which were in a carbonated saline solution for 2 weeks at 200 °C, produced a surface layer of rosette arranged plates within which well defined crystals of magnesite have grown. The platy texture and the development of red colouration which can be observed macroscopically leads us to assume that this phase is hematite, although concentration are too low to be detected using X-ray diffraction analysis. During experiments with a duration of over 2 weeks, where olivine powders reacted with a carbonated saline solution, the silica phase lizardite was observed to have formed. Neither talc nor chrysotile phase precipitation has been observed in any of our experiments. Amorphous silica has also not been identified in the reaction rim produced. Experiments observing the reaction interface using cross-sectioning of single crystals is difficult as the reaction layer is fragile and easily removed. However, with a longer experiment time the reaction layer completely covers the surface. The magnesite crystals are larger than those formed at shorter reaction times, but do not increase in number. Although reduction of the reaction temperature by 15 °C produced less angular magnesite crystals. So too did reactions conducted in solutions with lower ionic strengths.

References

Bearat H., McKelvy M., Chizmeshya A., Gormley D., Nunez R., Carpenter R., Squires K., Wolf G., (2006) Carbon Sequestration via Aqueous Olivine Mineral Carbonation: Role of Passivating Layer Formation. Environ. Sci. Technol. 40: 4802 – 4808

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