

Interfacial-Reaction Study of San Carlos Olivine Carbonation

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Since the late 19th century atmospheric CO₂ levels have been steadily on the rise. Approximately one third of all human emissions come from fossil fuel power plants. As countries become more dependent on electrical energy and bring on line new power plants these CO₂ levels will continue to rise [1]. Several solutions to this problem address converting the CO₂ from the gaseous phase and storing it. Oceans, subsurface reservoirs such as depleted oil fields, and terrestrial carbon pools have all been suggested [2]. Mineral sequestration of CO₂ is a candidate technology for reducing the amount of anthropogenic CO₂ that is being released into the atmosphere.

Olivine (e.g. forsterite, Mg₂SiO₄), is a widely available mineral that reacts with CO₂ to form magnesite (MgCO₃). Magnesite is capable of immobilizing CO₂ over geological time periods.

The solid/liquid solution reaction interface provides insight to the mechanisms that govern the reactivity of the olivine. Study of these mechanisms at the atomic level is critically important to facilitate engineering new processes that will enhance the reactivity of olivine with CO₂ bearing media and to lower process costs. Field emission SEM and cross-section HRTEM are being used for initial investigation of mechanism morphology. Nanospectroscopy will be used for product chemistry identification.

Single crystal olivine samples, (~ 1cm² by 1 mm thick) were prepared by polishing two (001) sides with 1 micron diamond paste. They were then placed in the reaction chamber in a 50 mL solution of 1.0 M NaCl and 0.64 M of NaHCO₃. This is the same solution used by our collaborators at the Albany Research Center (who are working on process development), and is used to increase the activity of CO₂. The reaction chamber was then pressurized with 2200 psi CO₂ and heated to 185 C. The reaction was performed for time intervals from 1 to four hours.

Figure 1 shows an SEM image of the reaction surface area in plan view. The cracked surface is due to the volume difference between the substrate, olivine (Mg₂SiO₄), and the reaction product matrix material, SiO₂. Magnesite particles, (MgCO₃), are expected to be in the cracked reaction surface layer. Figure 2 shows a HRTEM cross section image of the reaction interface. Note the presence of the nanoparticles in the reaction layer. These particles are found throughout the reaction layer and in high density. Lattice spacing of these crystals does not match that of MgCO₃, the expected product, but rather MgO, a thermodynamically improbable product of the reaction. According to Kim et al, MgCO₃ is unstable at temperatures above ~800 K, where it decomposes to form MgO [3]. The major question now is determining whether this is a metastable intermediate reaction product or an artifact. Current research is directed at lowering the temperatures that the reacted material encounters throughout the specimen preparation process and TEM investigation to enhance the stability of MgCO₃ expected to form. The use of cold stage ion mills and low beam currents are currently being

studied, to limit specimen heating. We have also determined that bulk magnesite is quite sensitive to radiolytic decomposition, and these limits will be reported. Low dose nanospectroscopy is necessary.

REFERENCES

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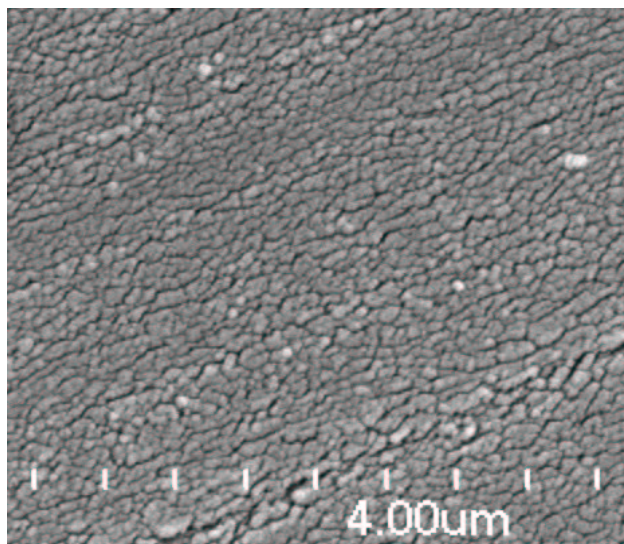


FIG. 1. The image of SEM after 4 hours of reaction.

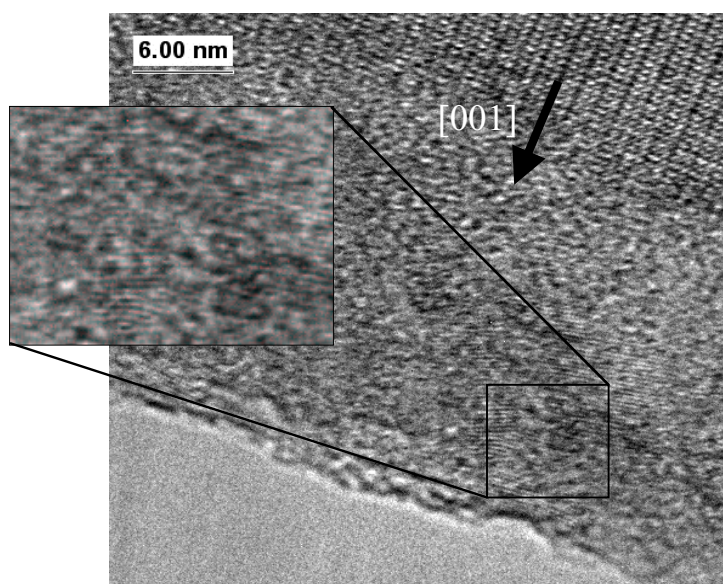


FIG. 2. Nanoparticles in amorphous reaction area(SiO_2) and Olivine structure after 4hours reaction.