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# Developing a Mechanistic Understanding of $CO_2$ Mineral Sequestration Process for Power Plants

Renu Sharma, M. J. McKelvy, Hamdallah Bearat, Andrew V.G. Chizmeshya and R.W. Carpenter

Center for Solid State Science, Arizona State University, Tempe, AZ 85287-1704

Fossil fuels are one of the cheapest sources for power plants if $CO_2$ produced during the process can be disposed in a cheap and permanent way to make them environmentally safe. Mineral carbonation is one of the most benign and permanent means of carbon sequestration.[1] Brucite ($Mg(OH)_2$) is a model system being investigated as it is presumably the active species in more complex minerals e.g. serpentines, to be used as feed stock materials for carbon sequestration. The reactivity of partially dehydroxylated brucite ($Mg(OH)_2$) single crystal fragments has been observed to substantially increase at room temperature (RT). BET measurements confirm the increase in the surface area of partially dehydroxylated (90%) single-crystal $Mg(OH)_2$ fragments, which directly contributes to the increase in reactivity. Various mechanisms for the formation of MgO nanocrystals during *in situ* dehydroxylation of $Mg(OH)_2$ have been reported.[2,3] We have made in situ observation of the carbonation reaction of these freshly formed nano crystals using E-TEM in order to optimize the process for power plants.

Experiments were performed using a PHILIPS-430 electron microscope operated at 300KV, fitted with a differentially pumped environmental-cell (E-cell) and a Gatan Imaging Filter (GIF).[4] The powdered samples of natural brucite mineral ($Mg(OH)_2$) were dry loaded on holey-carbon Cu grids and heated up to 465°C in order to complete the dehydroxylation reaction. These samples were exposed 400 mTorr of humid $CO_2$ at room temperature. The HREM images were recorded in real time using a TV-rate camera (30 frames/s) connected to a video tape recorder.

The brucite crystals were observed to disintegrate into nano crystals of MgO during dehydroxylation. The intensity of hydroxide diffraction spots (marked by arrows in Figure 1a) became diffuse at 166°C and finally only $\{111\}_{MgO}$ spots with streaking could be observed at 307°C. The product was found to be very reactive and very fluid in nature. Some faint lattice fringes, belonging to $Mg(OH)_2$ (0.455 nm), observed in Figure 2a became more visible after 13 seconds (Figure 2b). After 27 seconds the lattice fringes disappeared and the edge of the fragment moved outwards and then receded back (Figure 2c-d). The $CO_2$ and $H_2O$ diffused through the grain boundaries of the nano crystals swelling the fragment and a waxing of the edge is observed. Some of the $CO_2$ is used in carbonating and rest is released back resulting in the waning of the edge. Ripples were also observed to form on the top surface of the crystal indicating the three dimensional nature of the process. Although some hydroxylation was observed but mostly the $H_2O$ acted as a solvent (psuedo catalyst) increasing the mobility of MgO molecules. All of the diffuse rings observed in SAED patterns could be indexed as MgO phase indicating non-crystalline nature of the carbonate formed. The electron energy-loss near-edge (ELNES) structure for both $C_k$ and $O_k$ -edges of the samples after reaction matched to the one obtained from $MgCO_3$ (Figure 3). Ex-situ studies have confirmed the formation of an amorphous carbonate intermediate phase during carbonation. This technique results in 30% conversion rate. Methods to improve the conversion rates at low costs using other minerals (e.g. serpentines) and their implementation in the power plants. are being investigated.

Reference

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2. McKelvy et al, Chemistry of Materials, 13 (2001) 921.
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5. DOE grants (DE-FG0395TE00068 and DE-FG26-98FT40112) and the Center for High Resolution Electron Microscopy are gratefully acknowledged.

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Figure 1 Selected area electron diffraction patterns (a) at room temperature showing $\{0001\}_b$ diffraction spots (marked by arrows) (b) fading at 166°C and (c) disappearing at 305 °C. The streaking of MgO spots indicated poly crystalline nature.

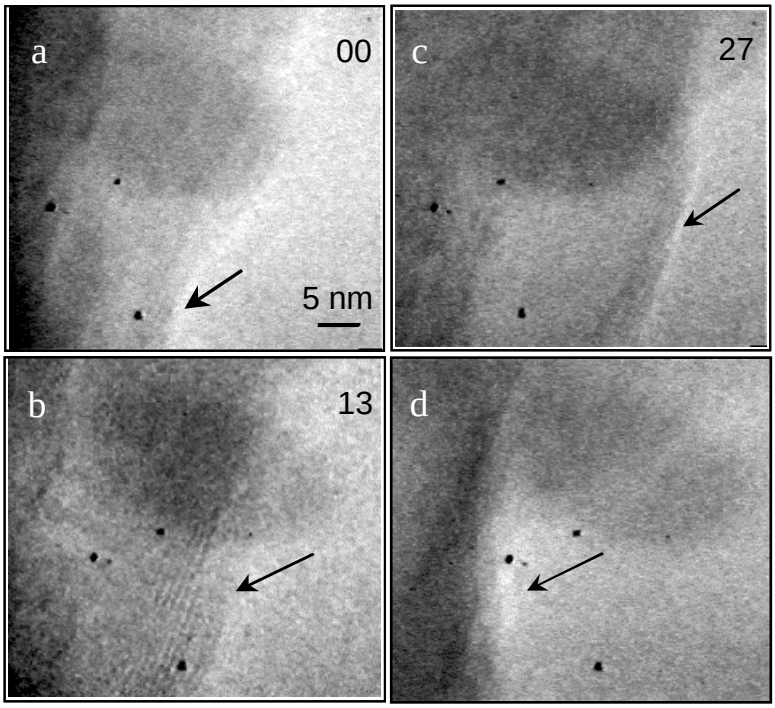


Figure 2. HREM images showing development of hydroxide fringes (marked by arrows in a and b) and waxing and waning of the edge.

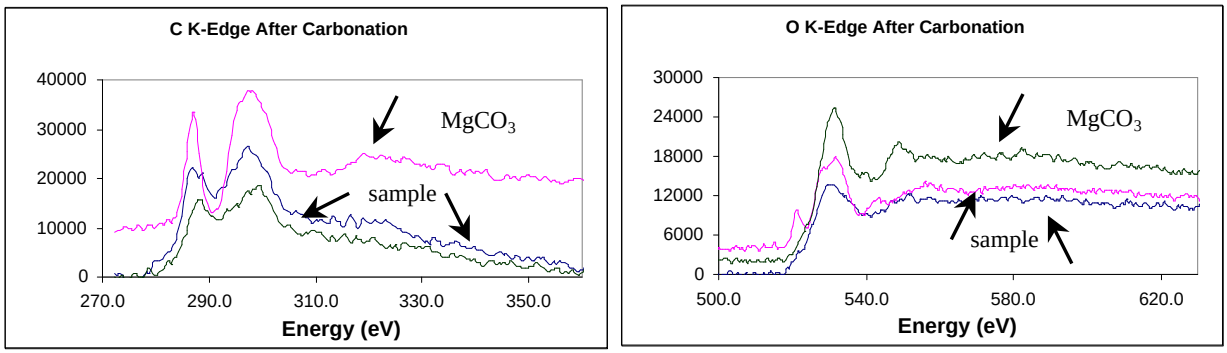


Figure 3. ELNES of $C_k$ and $O_k$ -edges recorded 70 hours after in situ observations compared to $MgCO_3$ standard sample. The spectra are shifted vertically with respect to each other for visualization purpose.

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