Carbonate Formation and Distribution during Carbon Dioxide Sequestration by Accelerated Concrete Carbonation

Sormeh Kashef-Haghighi, Subhasis Ghoshal
McGill University, Department of Civil Engineering, Montreal, Canada
sormeh.kashefhaghighi@mail.mcgill.ca

and

Subhasis Ghoshal
McGill University, Department of Civil Engineering, Montreal, Canada
subhasis.ghoshal@mcgill.ca

A range of carbon capture and storage (CCS) technologies are required to mitigate greenhouse gas emissions. Storage of CO_2 in reactive minerals involves the chemical reaction of CO_2 with reactive metal oxide feedstocks. Calcium and magnesium are two metals that are usually found in naturally abundant silicate minerals such as olivine, serpentinite and wollastonite. Freshly mixed, compact concrete is a calcium-rich porous media, composed of mainly calcium silicates, which have the potential to react with CO_2-rich gas streams from industrial flue gas. Therefore, CO_2 accelerated concrete curing is considered as a new carbon dioxide mitigation technology which has the potential to permanently and safely sequester carbon dioxide while producing non-reinforced concrete with improved physical properties and lesser curing time. In this research, the rate and extent of concrete carbonation is studied experimentally using two flow-through reactors: a completely mixed flow-through reactor for loose grout sample as well as a 1-D flow-through reactor for the compacted concrete. In order to simulate as-captured flue gas of cement industry, the gas mixture for carbonation was 20% CO_2 in nitrogen balance.

The maximum theoretical CO_2 uptake by concrete is calculated based on the reacting oxides present in the cement while the uptake by quartz aggregates is considered to be negligible. The elemental oxide composition of cement was defined through X-ray fluorescence analysis. The major oxide was CaO and represented 63.1% of cement followed by SiO_2, Al_2O_3, SO_3, MgO, and Fe_2O_3. The theoretical limit of uptake was then calculated based on the Steinour formula and was found to be approximately 50 wt% of cement for type 10, Portland cement (i.e. CO_2 can be reacted up to half of cement's weight in concrete mixture). The CO_2 uptake in concrete was calculated based on the inline measurement of CO_2 concentration in the effluent gas by a non-dispersive infrared sensor.

The porosity of carbonated concrete decreased as a result of formation of solid calcium carbonate in the pores. The mercury intrusion porosimetry results showed an approximately 15% decrease in the porosity after an hour of carbonation (and 3 hours of initial hydration) compared to non-carbonated concrete. The non-carbonated concrete sample was hydrated for 24 hours in order to reach an initial strength required for mercury intrusion. The loss of pore volume is mainly attributed to the expansion of solid matrix and pore blockage due to the formation of solid products mainly calcium carbonate. The X-ray diffraction analysis of non-carbonated and carbonated cement also confirmed the formation of calcite as the main calcium carbonate polymorph. The CO_2 uptake by concrete in the flow-through reactor, however, did not reach the theoretical maximum value. The average carbonation efficiency obtained in compact concrete was approximately 18% compared to 100% maximum theoretical efficiency. The decrease in the reactive surface area of cement during carbonation due to the formation of solid

$CaCO_3$, and its effect on the dissolution of unreacted cement compounds was determined as a critical process affecting the CO_2 uptake. The formation of solid calcium carbonate layer in the carbonated sample was also confirmed using an electron microprobe imaging technique. A thick layer of $CaCO_3$ with heterogeneous thickness was seen in the carbonated samples, while it was not present in the non-carbonated concrete samples.