

A Study for Effective CO₂ Sequestration of Hydrated Cement by Direct Aqueous Carbonation

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Abstract : Global warming is a world-wide issue. Various carbon capture and storage (CCS) technologies for reducing CO₂ concentration in the atmosphere have been increasingly studied. Mineral carbonation is one of promising method for CO₂ sequestration. Waste cement generating from aggregate recycling processes of waste concrete is potentially a good raw material containing reactive components for mineral carbonation. The major goal of our long-term project is to developed effective methods for CO₂ sequestration using waste cement. In the present study, the carbonation characteristics of hydrated cement were examined by conducting two different direct aqueous carbonation experiments. We also evaluate the influence of NaCl and MgCl₂ as additives to increase mineral carbonation efficiency of hydrated cement. Cement paste was made with W:C= 6:4 and stored for 28 days in water bath. The prepared cement paste was pulverized to the size less than 0.15 mm. 15 g of pulverized cement paste and 200 ml of solutions containing additives were reacted in ambient temperature and pressure conditions. 1M NaCl and 0.25 M MgCl₂ was selected for additives after leaching test. Two different sources of CO₂ was applied for direct aqueous carbonation experiment: 0.64 M NaHCO₃ was used for CO₂ donor in method 1 and pure CO₂ gas (99.9%) was bubbling into reacting solution at the flow rate of 20 ml/min in method 2. The pH and Ca ion concentration were continuously measured with pH/ISE Multiparameter to observe carbonation behaviors. Material characterization of reacted solids was performed by TGA, XRD, SEM/EDS analyses. The carbonation characteristics of hydrated cement were significantly different with additives. Calcite was a dominant calcium carbonate mineral after the two carbonation experiments with no additive and NaCl additive. The significant amount of aragonite and vaterite as well as very fine calcite of poorer crystallinity was formed with MgCl₂ additive. CSH (calcium silicate hydrate) in hydrated cement were changed to MSH (magnesium silicate hydrate). This transformation contributed to the high carbonation efficiency. Carbonation experiment with method 1 revealed that that the carbonation of hydrated cement took relatively long time in MgCl₂ solution compared to that in NaCl solution and the contents of aragonite and vaterite were increased as increasing reaction time. In order to maximize carbonation efficiency in direct aqueous carbonation with CO₂ gas injection (method 2), the control of solution pH was important. The solution pH was decreased with injection of CO₂ gas. Therefore, the carbonation efficiency in direct aqueous carbonation was closely related to the stability of calcium carbonate minerals with pH changes. With no additive and NaCl additive, the maximum carbonation was achieved when the solution pH was greater than 11. Calcium carbonate form by mineral carbonation seemed to be re-dissolved as pH decreased below 11 with continuous CO₂ gas injection. The type of calcium carbonate mineral formed during carbonation in MgCl₂ solution was closely related to the variation of solution pH caused by CO₂ gas injection. The amount of aragonite significantly increased with decreasing solution pH, whereas the amount of calcite decreased.

Keywords : CO₂ sequestration, Mineral carbonation, Cement and concrete, MgCl₂ and NaCl

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