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CO_2 Absorption and Precipitation in $MgCl_2$-NH_3 • H_2O Solutions: Relevance to CO_2 Sequestration

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Mineral sequestration of CO_2 is one of the safer options in the portfolio of available Carbon Storage and Sequestration (CCS) stratagems. To date, numerous approaches, such as MEA/DEA/Ammonia based post combustion scrubbing and O_2/CO_2 recycle combustion, were tested to optimize the absorption of CO_2 using various media. These methods were shown to be able to achieve CO_2 capture, but did not address the costly issue of carbon storage. Following a newly proposed pH-swing CO_2 mineralization process (Kodama *et al.* , 2008) which

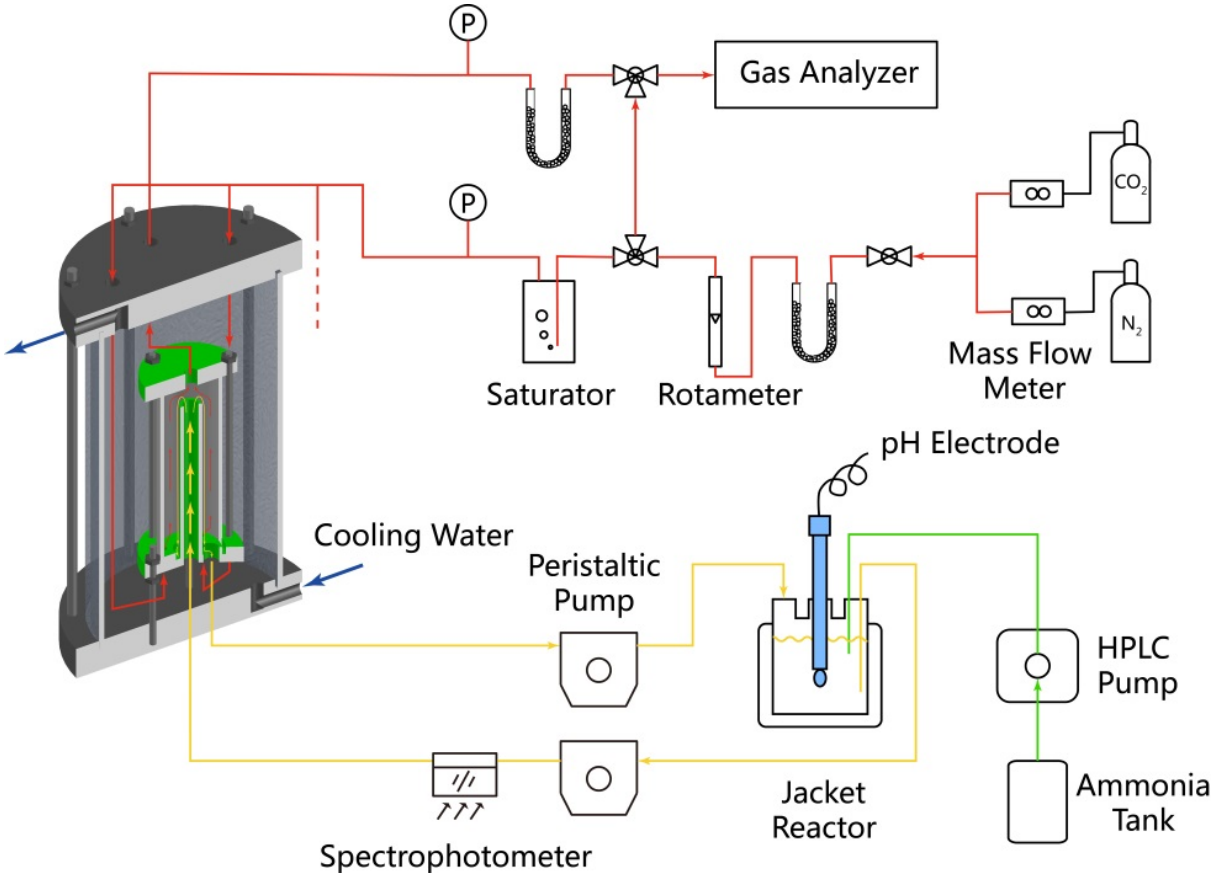

Fig. 1. Schematic diagram for the CO_2 absorption system

CO_2 absorption experiments were carried out at 298 K in solu-tions of different compositions (0.05~0.2molL-1 $MgCl_2$) to mea-sure the reaction kinetics using a wetted wall column setup simi-lar to those reported in Pacheco (1998) and Victor (2011). The solution chemistry was maintained at conditions where brucite pre-cipitation was not allowed. The absorption solution was then cycled between the column and a jacked glass reactor with its pH maintained constant, and the trans-mittance at 546 nm was monitored in real-time. Magnesium concentration was analyzed by ICP-AES with interval sampling.

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showed potential of being able to capture and store CO_2 simultaneously. we investigated the rate and kinetic controlling factors for CO_2 absorption and mineralization in $MgCl_2$-NH_3 solutions in this study to better understand the system behavior . CO_2 absorption experiments were carried out at 298 K in solutions of different compositions (0.05~0.2molL-1 $MgCl_2$) to measure the reaction kinetics using a wetted wall column setup similar to those reported in Pacheco (1998) and Victor (2011). The solution chemistry was maintained at conditions where brucite precipitation was not allowed, and magnesium concentration was analyzed by ICP-AES with interval sampling. The absorption solution was then cycled between the column and a jacked glass reactor with its pH maintained constant and the transmittance at 546 nm monitored in real-time. Preliminary results indicate that initial concentration of Mg in solution has little effect on CO_2 absorption. Although CO_2 absorption rate increased slightly over time with increasing ammonia addition, pH appeared to be the dominant controlling factor. The higher the solution pH was, the faster the absorption rate increased. Upon reaching saturation, nesquehonite precipitated as indicated by the decreased laser transmittance, leading to rapid addition of aqueous ammonia. However, precipitation of nesquehonite unexpectedly showed little influence on CO_2 absorption, suggesting that the interaction between aqueous CO_2 and OH^- or ammonium ions in liquid film may be the rate limit step during the absorption process, further indicating that the gas-liquid interaction barrier should be treated seriously in order to optimize CO_2 capture efficiency. Due to the low gas-liquid reaction area (0.005 m^2L^{-1}), extended time period (2~3 hours) was needed to dissolve enough CO_2 for nesquehonite to reach supersaturation. Measured typical absorption rates under these experimental conditions are between 0.000668 mol $s^{-1}m^{-2}$ (pH=8.74, pCO_2= 15495Pa, 298K) and 0.001997 mol $s^{-1}m^{-2}$ (pH=9.16, pCO_2= 15403Pa, 298K).

Key words: Nesquehonite, Carbon dioxide, Mineral carbonation, CO_2 sequestration

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