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Characterization of Mg^{2+} dehydration in aqueous electrolyte solution: implications for CO_2 mineralization technologies

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Formed via aqueous carbonation of Mg^{2+} ions, the crystallization of Magnesite ($MgCO_3$) is a promising capture and reuse technology, albeit limited by slow precipitation rates of $MgCO_3$. Although magnesite is naturally abundant, forming at ambient pressure and low temperature (<60 °C) conditions, $MgCO_3$ production is an energy-intensive process due to the temperatures (120-185˚ C) and pressures (100-150 bar) required to prevent the formation of hydrated phases. Focus has therefore been on characterizing and optimizing the fundamental aspects of this slow precipitation under ambient conditions, towards raising efficacy and profitability of the process. The principle difficulty arises from the very strong $Mg^{2+}\cdots H_2O$ interaction, raising barriers to dehydration, while hindering nucleation and subsequent growth of Mg-carbonates. Organic ligands and/or inorganic ions (e.g. bacterial bisulfide (HS^-)) have been hypothesised to catalyse Mg^{2+} dissociation; hence, quantitatively resolving the catalytic role of composition is imperative to determine what catalyses $MgCO_3$ formation. Atomistic modelling has been used to characterize the structure, energetics, and dynamics the Mg^{2+} (de)hydration process, from the atomic through mesoscopic scales. Enhanced sampling techniques (metadynamics and umbrella sampling) have been adopted to characterize the reaction mechanism kinetics of $Mg^{2+}\cdots H_2O$ dissociation in a series of aqueous electrolyte solutions, with results showing that solution composition can modulate Mg-dehydration and, consequently, the subsequent steps of nucleation & growth, which are likewise susceptible to Mg^{2+}-(re)hydration.