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Stability Controls Against Crystallization Of Amorphous Magnesium Carbonate Analyzed Using Total Scattering Approaches

JULIANE WEBER[1], PENG YANG, PHD[1,2], DR. KE YUAN[1], ATHENA CHEN[1], SAI ADAPA[1], CHUNG DONGYOUN[1], BARBARA EVANS[1], JOERG NEUEFEIN[1], WENQUIAN XU[3], HSIU-WEN WANG[1] AND ANDREW G. STACK[1]

[1]Oak Ridge National Laboratory
[2]Brookhaven National Laboratory
[3]Argonne National Laboratory

Carbonates are one of the most prominently investigated systems forming amorphous intermediates due to their importance as paleo-proxy, their role in the global carbon cycle, during biomineralization and in engineered carbon dioxide mineralization. Despite numerous studies, the reaction mechanisms controlling stability of amorphous hydrated phases (e.g., amorphous carbonates) against crystallization are not well understood. A fundamental understanding of the reaction mechanisms controlling stability of amorphous phases is needed to enable predictions of mineral reactions in the environment. Here, we present an experimental study on the kinetics of the amorphous-to-crystalline transformation in amorphous magnesium carbonate (AMC) and calcium-doped AMC using diffraction and total scattering/pair distribution function analyses (based on both synchrotron X-ray and neutron). AMC transformed to nesquehonite below 55 ºC[1] and to hydromagnesite above 55 ºC. Based on the Johnson–Mehl–Avrami–Kolmogorov equation, we have determined that the activation energy for AMC-to-hydromagnesite transformation (116 ± 19 kJ/mol) is nearly three times higher than the AMC-to-nesquehonite (41 ± 13 kJ/mol) transformation. Kinetic isotope effects for the deuterated AMC-D_2O (d-AMC-D_2O) system are distinctly different from the protiated AMC-H_2O (p-AMC-H_2O) system: the d-AMC is two orders of magnitudes more stable than the p-AMC. This contrasts with observations of amorphous calcium carbonate, where the deuterated form was found to be less stable.[2] Our results highlight the importance of vibrational water motions for stability against crystallization for amorphous magnesium carbonate.

[1] Kloprogge, Martens, Nothdurft, Duong &Webb (2003). Journal of Materials Science 22, 825-829.

[2] King, Avaro, Peter, Hauser & Gebauer (2022). Faraday Discussions 235, 36-55.