

## Water and ionic dynamics in amorphous carbonates probed by neutron and X-ray scattering

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Amorphous calcium carbonate (ACC) is a metastable material formed during the formation of calcium carbonate biominerals. These organisms take advantage of the moldable character of the ACC structure and of its kinetic persistence against crystallization to form their shells and skeletons, with intricate shapes and functionalities. Different authors have suggested that the water structure and hydrogen bonding are important factors enhancing the kinetic stability of these materials. In addition, some additives—such as  $Mg^{2+}$ —are known to increase their stability, retarding their crystallization.

In this study, a multi-technique approach, including state-of-the-art coherent X-ray techniques and inelastic neutron scattering, are used to probe the nanoscopic dynamics in  $Mg^{2+}$ -bearing ACC. Specifically, our study shows the mechanism by which  $Mg^{2+}$  enhance the strength of the hydrogen bond network, increasing the kinetic persistence of the amorphous material. Moreover, and unexpectedly, synchrotron X-ray photon correlation spectroscopy (XPCS) experiments have shown that the presence of  $Mg^{2+}$  enhances the atomic diffusivity within the solids, creating also a strained structure. This strained and stiffer hydrogen bond network is proposed as the main reason behind the increased persistence against crystallization of these Mg-doped materials. The results are analyzed here within the framework of the classical nucleation theory, providing a unique way to develop a mechanistic understanding of crystallization processes.