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Cryosalt Formation in Delaminated Clays

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Hydrohalite ($NaCl{\cdot}2H_2O$) forms by evaporation, sublimation and freezing of aqueous solutions of NaCl. Although this process is traditionally deemed to occur in aqueous solutions little attention has been paid on whether this is possible with minerals. Smectite minerals are particularly interesting in this regard for their ability to accommodate water between aluminosilicate sheets, allowing them to swell and even delaminate. In particular, montmorillonite possesses high affinities for water and can play important roles in water retention and ice formation in nature, as well as in strategies for nuclear waste storage and even for technological applications. [1,2]

For this study, we aimed to develop insight into the molecular-level nature of hydrohalite formation at surfaces of montmorillonite particles as well as in their interlayers. Thin films of Na^+ and Ca^{2+} exchanged montmorillonites deposited on a diamond-based Attenuated Total Reflectance (ATR) cell were interacted with (0.01, 0.1, 1 and 5M) NaCl solutions, and then frozen to -10˚C. The resulting frozen montmorillonites pastes were then probed by ATR Fourier Transform Infrared (FTIR) spectroscopy, which is a highly sensitive technique for probing hydrogen bonding in minerals and water. Our results on Na-montmorillonite showed that hydrohalite, with its characteristic O-H stretching ($\nu_{OH} \sim$ 3245-3265, 3408, 3462, 3555 cm^{-1}) and bending ($\delta_{OH} \sim$ 1614 and 1641 cm^{-1}) bands, formed from solutions of at least 0.1 M NaCl, yet well below the typical homogeneous crystallization of this phase from pure aqueous solutions.[3] Further analysis of the O-H stretching and silicate ($\nu_{Si-O} \sim$1000 cm^{-1}) regions of frozen paste of montmorillonites revealed that hydrohalite formed within interlayers and at surfaces of Na-montmorillonite. Ca-montmorillonite did not, on the other hand, promote hydrohalite formation but did undergo Ca^{2+}/Na^+ ion exchange due to exposure of the NaCl solutions.

Given the inability of Ca-montmorillonite at hosting hydrohalite, the results of this study can be used to suggest that delaminated Na-montmorillonite sheets encapsulate the salt solutions and by preventing sublimation of water, promotes crystallization of hydrohalite. As delamination is not possible in Ca-montmorillonite, water more readily sublimated from the system, leaving behind a dry Ca-montmorillonite/NaCl assemblage. As such, this work identified processes through which clay minerals can affect the formation of cryosalts that are not only of importance to terrestrial environments of the cryosphere but also to atmospheric processes involving dust aerosols.

[1] Yeşilbaş, M. and Boily, J.-F. (2016), Scientific Reports. 6, 32136.

[2] Yeşilbaş, M. and Boily, J.-F. (2016), J. Phys. Chem. Lett. 7, 2849-2855.

[3] Wagner, R., Möhler O., Schnaiter, M. (2012), 33, 8557-8571.