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Probing the high-pressure behaviour of H₂SO₄ and MgSO₄ hydrates with neutrons

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Hydrates of sulfuric acid, and of magnesium sulfate, have been reported on the surfaces of the Galilean satellites of Jupiter [1], and models suggest that these hydrates will be abundant in their deep interiors [2], consequently experiencing modest hydrostatic pressures. Investigation of their high-pressure behaviour is important since there are likely to be changes in both the hydrogen bond network as well as possible changes in ion speciation, and pressure-induced dehydration. Such phase changes may influence heat transport inside icy planetary bodies, and hence control their overall structure and evolution. Using neutron powder diffraction, we have carried out studies upon a range of hydrates relevant to the internal structure and dynamics of icy satellites, including deuterated isotopologues of sulfuric acid 8-, 6½-, and 4-hydrate, and magnesium sulfate 11-, and 7-hydrate, at pressures up to 4 GPa in the Paris-Edinburgh press. Much of our earlier work is summarised in reference [3]. In sulfuric acid tetrahydrate, we have identified two monoclinic high-pressure polymorphs, SAT-II and SAT-III, in addition to the low-pressure tetragonal phase. SAT-II is formed by warming SAT-I above 235 K at 550 MPa, and this has been successfully recovered to atmospheric pressure at 50 K. SAT-III has been observed over the range 1.6–3.9 GPa, melting at 380 K at 3.9 GPa. However, sulfuric acid tetrahydrate is extremely difficult to crystallise at high-pressure, requiring deep undercooling. In the MgSO₄-hydrates we have now observed a reproducible sequence of phase transitions, in agreement with ultrasonic wave-velocity observations reported elsewhere [4], in which pressure-induced dehydration occurs. At 295 K, MgSO₄·7D₂O (synthetic epsomite) undergoes its first transition at 1.2 GPa to a lower hydrate + aqueous solution, this phase being stable over only a narrow pressure range (0.2 GPa), before the onset of the next phase transition and the growth of ice VII. Similarly, MgSO₄·11D₂O (synthetic meridianiite), when compressed at 240 K, breaks down to a lower hydrate + ice VI at ~ 0.9 GPa. We report the status of our work to interpret the high-pressure behaviour of these materials, including recent attempts to recover the products of various phase transitions to ambient pressure for better characterisation.

References

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