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Study of crystallite size distributions in ices and gas hydrates using a novel X-ray diffraction procedure

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Crystallite size distributions (CSDs) provide important insights into the formation processes of materials, in particular the balance between nucleation and growth rates. Both equilibrium and non-equilibrium physical properties of the resulting polycrystalline gas hydrate assemblages will depend on the CSDs, in particular their mechanical properties as well as their interactions with any surrounding fluid phase or other transport properties alike. We have developed over the last years a fast X-ray diffraction based procedure to determine CSDs with unprecedented precision and have applied this method to the study of ices and gas hydrates (Chaouachi et al. 2017) amongst other examples from material science. Various gas hydrates (GH) were analyzed during formation and coarsening in a sedimentary matrix. The GH crystallites have a size of a few µm when formed (confirmed also by synchrotron X-ray tomography), followed by a coarsening process which mainly takes place at the surface of GH aggregates. Important conclusions can be drawn from the time-dependent analysis of CSDs: (1) Coarsening by normal grain growth proceeds several orders of magnitude slower than in normal ice Ih at similar temperatures; this points to very slow grain boundary migration rates seemingly related to the complexity of topological reconstruction of the crystalline network across a disordered grain boundary. (2) The persisting small crystallites together with their known high resistance against deformation by dislocation motion must lead to grain size sensitive creep, most likely governed by grain boundary sliding. (3) The CSDs of GHs formed in the laboratory appear to have distinctly smaller sizes compared to natural GHs. In consequence, laboratory-based studies of GH can only be safely related to the natural situation once the mutual CSDs are characterized. Starting from crystallites of a few µm in size, obtained under common laboratory conditions from liquid water and gas, it would take hundreds to thousands of years to reach crystal sizes encountered in some NGHs, at least when assuming a normal grain growth mechanism. Anomalous grain growth may intervene and increase crystal sizes, in particular under overburden load and local stresses. Such an anomalous grain growth was indeed observed in natural air hydrates found in Greenland deep ice cores. Finally, we like to mention that our FXR-CSD method is particularly powerful for small crystallites (µm to tens of µm in size) and will allow quantitative studies of in-situ processes occurring during snow coarsening or dynamic recrystallization in ice.

M. Chaouachi, S.H. Neher, A. Falenty & W.F. Kuhs (2017) Crystal Growth & Design 17, 2458-2472

Significance statement

A new diffraction-based method adapted for in-situ study of crystal growth and coarsening phenomena in a large variety of materials including ices and gas hydrates is-presented. Normal grain growth processes of gas hydrates are found to be orders-of-magnitude slower than similar processes in ice Ih.

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Track Classification: Phases of Ice