The structural relaxation properties of the amorphous ices

C. Salzmann$^1$, J. Shephard$^{1,2}$, J. Evans$^2$

$^1$University College London, Department of Chemistry, London, UK, $^2$Durham University, Department of Chemistry, Durham, UK

Despite the importance of low-density amorphous ice (LDA) in critical cosmological processes and its prominence as one of the polyamorphs of water there is still an incomplete picture of the processes that take place upon thermal annealing. We show that a gradual structural relaxation process takes place upon heating vapor-deposited LDA, also called amorphous solid water, and LDAs obtained from several different states of high-density amorphous ice. The structural relaxation leads to an increase in structural order on local and more extended length scales as the average O-O distance shortens and the O-O distance distribution narrows. The relaxation process is separate from crystallization and it does not seem to reach completion before crystallization sets in. Our findings are therefore difficult to reconcile with the postulated glass transition of LDA to the supercooled and highly viscous liquid prior to crystallization. On the basis of a comparison of the calorimetric data of LDA with those of some of the crystalline phases of ice we propose that the calorimetric feature of LDA prior to crystallization may in fact be connected to the kinetic unfreezing of defect-migration mediated reorientation dynamics. We finally discuss the relaxation properties of the various kinds of high-density amorphous ice in the context of these new findings.


Keywords: amorphous ice, structural relaxation, spectroscopy