

The LiCl RH₂O and LiBr RH₂O liquid glass Transition close to their eutectic point

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Abstract:

The pure H₂O liquid-glass transition cannot be studied. Inevitably, water homogeneously crystallizes before reaching 240 K. An amorphous phase can be obtained only through a rapid quench. With the addition of a pressure treatment at some MPa, a High Density Amorphous (HDA) phase, with a density approximately equal to 1.15 g/cm³ can be obtained. Conversely, as suggested more than 40 years ago by C.A. Angell et al. [1], a continuous liquid-glass transition can be obtained if salts are added, in particular, to obtain a composition close to the eutectic one. In particular, for LiCl (and LiBr) RH₂O solutions in the R=6-7.2 region, i. e. above but close to their eutectic composition, glasses can be continuously obtained with densities at T_g close to the HDA one. We have studied the corresponding supercooled liquids, between 293 and 160 K (approximately 20 K above T_g) through a HD-Transient Grating technique, supplemented by Brillouin scattering and Ultrasonic measurements.

In a first series of experiments, we have considered the R=6 solutions and shown that their alpha relaxation time, tau (sub alpha), has a typical fragile behaviour between 10⁽⁻²⁾ and 10⁽⁺³⁾ ns. Also, a second, beta, relaxation process, with an Arrhenius behaviour, starts to separate from the first one below 210 K. More interestingly, another, additional, TG signal, with an intensity increasing with R, superposes to the R=6 one in the chlorine case for 6<R<7.2 (but not in the bromine one). This signal is the signature of the formation of water rich clusters, with a few nm size, immersed in a (presumably) R=6 solution. These clusters are practically size and composition independent in the R=6-7.2 and 190-172 K ranges explored. Furthermore, their existence (at densities which increase with R) does not modify the alpha and beta relaxation processes.

Reference:

[1] C. A. Angell and E. J. Sare, J. Chem. Phys. 52, 1058 (1970).