ISIS Experimental Report

Rutherford Appleton Laboratory

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Title of Experiment:	The structure, equation of state, and phase transitions of ammonia dihydrate polymorphs.	**Local Contact:**	W. G. Marshall
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INTRODUCTION

Ammonia dihydrate (ADH) is very likely to be the most significant mineral after water ice in the mantles of ice rich moons orbiting Saturn, Uranus and Neptune [1]. It allows for low temperature melting, and is invoked to explain a wide variety of geological processes observed on these bodies. The physical properties and phase relations of ADH are poorly constrained [2]. There are two currently known phases (I and II) at low pressures [3-5]. ADH I (stable at ambient presure) has a cubic unit cell but the structure of ADH II (stable above 4.5 kbar) is unknown. We have recently measured the incompressibility and thermal expansivity of ADH I [6]. However, there is a clear need to study the phase relations at higher pressures.

EXPERIMENTAL

Stoichiometric liquid $ND_3.2D_2O$ was loaded into the Paris-Edinburgh cell, in encapsulated TiZr gaskets, along with a small wad of silica wool and a lead disc (3mm diameter, 1mm thick). The glass wool acts as a nucleator, and the lead serves as a pressure calibrant.

Four loadings were carried out during this experiment; the first and fourth were successfully compressed to an applied load of ~80 tons and decompressed; the second loading* ended with a burst gasket under ~66 tons load; the third loading leaked under a compression of 7 tons.

*Loading two was more water rich than the others as a result of forming a blue nickel-ammonia complex in the needle used to fill the gasket space.

For each of the successful loadings, the sample was first compressed under a ram pressure of 12-13 tons at room temperature and then cooled to ~170K by drizzling liquid N_2 over the sides of the P-E cell. At these temperatures crystallization usually takes several hours. Following crystallization, the ram pressure was increased in 5-10 ton increments up to around 80 tons (~8.6 GPa).

RESULTS

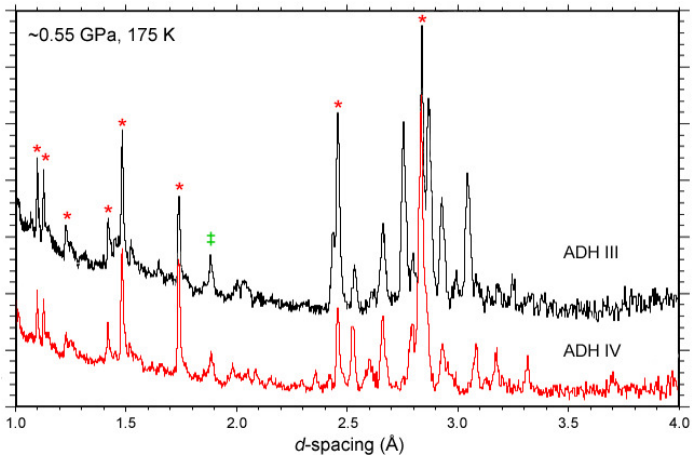
We did not observe any phase that matched the diffraction pattern of ADH II previously seen by Loveday et al. [5] or by ourselves [6]. At low pressure (~0.6 GPa) we saw two new phases. The first, ADH III, formed from a supercooled liquid, or a glass, at ~0.6 GPa and ~ 170K. The second, ADH IV, was first seen coexisting with ice VI (in the off-stoichiometry loading) under the same P-T conditions as phase III. We later made ice-free ADH IV by pressure freezing the stoichiometric liquid at ~200K.

When compressed, ADH III amorphized at ~ 2.5 GPa. Although three rather broad humps developed from this amorphous phase, we do not believe we observed a well crystallized solid. ADH IV underwent a similar transformation to a poorly crystalline phase at ~ 2.5 GPa. These two solids are referred to as Intermediate structures A and B for the time being.

At the maximum compression, two phases were observed at nearly identical P-T conditions. ADH V is characterised by two large broad peaks, and ADH VI by only one. Indeed ADH VI appears to have an identical diffraction pattern to AMH VI [7] supporting the idea that a solid solution series exists from AMH VI to ice VII with the same bcc structure. ADH V, on the other hand, appears to be a tetragonal distortion of the phase VI structure. ADH VI was successfully decompressed to 0.5 GPa (at 190 K) whereupon it reverted to phase IV.

The degree of metastability we have observed, and indeed the fact that we have now seen three different structures at ~0.5 GPa at 170K, is not surprising, and indeed is no different from the monohydrate system or pure ice at low temperatures.

Figure 1. Diffraction patterns of ADH III and ADH IV (roughly two hours of integrated data collection each). The strongest peaks from the lead pressure marker are highlighted with a red star, and the strongest peak arising from the tungsten carbide anvils is marked with a green double dagger.



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