SUPPLEMENTARY MATERIAL

Exploring high-pressure transformations in low-Z (H_2, Ne) hydrates at low temperatures

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The main text is supported by an extensive collection of experiments, which are described in more details in this material. Three hydrogen hydrate (HH) sample batches (A, B and C) and one neon hydrate (NeH) sample (c) were prepared. Five HH (*a*1, *a*2, *a*3, *b*, *c**) and one NeH experiments (*c*) were run on the SNAP high-pressure diffractometer at the Spallation Neutron Source in the Oak Ridge National Laboratory, USA.

A. IPTS-21975 (HH)

*a*1. Loading 1

Crystalline CS-II HH, 17.096(6) Å at 0.0(1) GPa, 95 K. Compressed at 90 bar[1]/h to 1.3(2) GPa, with phase transition (CS-II → C_1) at 1.1(2) GPa. Upon recovery to atmospheric pressure at 95 K, C_1 transformed back to CS-II with no other phases observed in the diffractogram.

*a*2. Loading 2

Crystalline CS-II HH, 17.091(6) Å at 0.0(2) GPa, 95 K. Compressed at 50 bar/h to 4.8(4) GPa. CS-II → C_1 transition identified at 1.12(5) GPa. Partial decomposition of C_1 observed at 3.5(2) GPa, in which a mixture of C_2 and ice VIII' (~50% v/v C_1, ~25% C_2, ~25% ice VIII') formed. The sample was heated but the high-pressure caused parasitic peaks from the anvil materials to cover most of the diffractograms, and no further data could be identified.

*a*3. Loading 3

Crystalline CS-II HH, 17.104(8) Å at 0.0(5) GPa, 95 K. Compressed at 50 bar/h to 0.97(8) GPa. Heated at 10 K/h to 200 K. At 1.19(4) GPa, 165 K, CS-II fully transformed into C_1. From 200 K, 1.56(5) GPa, the sample was continuously compressed to 7.8(2) GPa. Partial decomposition of C_1 observed at 3.6(1) GPa (composition could not be determined). Decompression at 200 K to atmospheric pressure, with recovery of empty C_1, ice II.

B. IPTS-26245 (HH)

Crystalline CS-II HH, 17.094(1) Å at 0.1(1) GPa, 95 K. Compressed at 12.5 bar/h to 2.2(1) GPa. CS-II could be kept metastably to the highest pressure at 95 K, in which C_1 reflections started to peak. Heated directly to 200 K leading to the complete conversion of CS-II into C_1. Compressed at 200 K, with partial decomposition of C_1 observed at 3.15(2) GPa. After further compressing at 200 K to 5.0(2) GPa (and after about 18 hours), the sample constituted 20% C_1 and 20% C_2/60% ice VIII'. Cooled to 95 K and slowly decompressed (~100 bar/h). After complete removal of the load from the press the residual pressure remained at 0.1(2) GPa, where C_1 and ice VIII' could be identified. By completely opening the press and releasing all load from the sample gasket, the sample was recovered to atmospheric pressure. A mixture of ices II (~20%), I_h (~60%) and I_c (~20%) could be identified.

C. IPTS-24413

c. Loading 1 (NeH)

Crystalline CS-II NeH, 17.1161(8) Å at 0.09(5) GPa at 90 K. Compression to 3.0(1) GPa, with pressure-induced amorphization identified at 1.86(3) GPa. Recrystallization of the amorph a mixture of C_2, ice VIII' and a small fraction of (metastable) ice I_c.

*c**. Loading 2 (HH)

Crystalline CS-II HH, 17.094(1) Å at 95 K.

[1] Oil pressure in the Paris-Edinburgh press hydraulic system.

Table S1. Refinement details from neutron powder diffraction data for C_2 Neon hydrate. Data collected at the high-pressure diffractometer SNAP (beamline 3) at the Spallation Neutron Source, USA

Parameter (units)	**Fit result (deviation)**
Formula	$H_2 \cdot H_2O$
Space Group	$Fd\bar{3}m$
Temperature	90(1) K
Pressure	3.6(1) GPa
Lattice Parameter (Å)	a = 6.402(5)
Radiation	Spallation Neutrons
Diffractometer	SNAP (BL-3), SNS, ORNL
R_w (%)	1.68
χ_2	0.92