Supporting Information

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SI Text

1. Effect of Dissolved Helium in Water. We note that among the gases commonly used for pressure experiments, helium is the one with the lowest solubility in water (1). For this reason, helium has been widely used as the standard pressurizing gas in previous measurements on high-pressure low-temperature water. From the data reported in ref. 1 and using Henry's law, it is possible to estimate the amount of helium dissolved in the water at the different pressures investigated. According to our calculations, the direct effect of the presence of helium will only negligibly change both the mass and the scattering length density of the sample (<0.07% at 3,000 bars, 0 °C). Although this number will increase as the temperature is lowered, it will still be an insignificant correction (the solubility increases almost linearly from 0.0013 g helium/kg water to 0.0017 as the temperature is lowered from 60 °C to 0 °C. http://www.engineeringtoolbox.com/gases-solubility-water-d_1148.html). Of course, the effect of helium atoms on the structure of water is more difficult to estimate, but it is beyond the scope of the present work.

2. SANS Model Considering the Nonuniform Distribution of Water in the Pores. In general, the small-angle neutron scattering intensity distribution can be written as

$$I(Q) = n V_p^2 (\bar{\rho} - \rho_s)^2 \cdot \bar{P}(Q) S(Q), \qquad \text{[S1]}$$

where n is the number density of the scatterers, V_p is the volume of each scatterer, $\bar{\rho}$ is the average scattering length density (SLD) of the scatterer $\bar{\rho} = \frac{1}{V_p} \int_{V_p} \rho(r) dr^3$, ρ_s is the SLD of the environment, $\bar{P}(Q)$ is the normalized form factor of each scatterer, and $S(Q)$ is the structure factor representing the spatial arrangement of the scatterers. In the case of hydrated MCM-41-S, each scatterer is a column of water confined in a cylindrical pore. If we assume that the water molecules distribute nonuniformly only along the radial direction of the cylindrical pore as $\rho(r)$, the normalized form factor $\bar{P}(Q)$ can be written as

$$\bar{P}(Q) = \int_0^1 d\mu \left[\frac{1}{\pi R^2} \cdot \frac{\sin(Q\mu L/2)}{Q\mu L/2} \right.$$
$$\left. \cdot 2\pi \int_0^R r \frac{\rho(r) - \rho_s}{\bar{\rho} - \rho_s} J_0(Qr\sqrt{1-\mu^2}) dr \right]^2, \qquad \text{[S2]}$$

where R is the radius of the water column, and L is the length of the column. ρ_s is the SLD of the silica matrix. Because the sample is comprised of different crystallites oriented randomly, the scattering profile is obtained by averaging over all the possible orientation of the pores with respect to the incoming beam (integrated over $\mu = \cos\theta$). $\bar{\rho}$ is the average SLD of the water in the pore (which can always be defined, and may or may not have physical relevance).

1. Wilhelm E, Battino R, Wilcock RJ (1977) Low-pressure solubility of gases in liquid water. *Chem Rev* 77:219–262.