Deuterated Clathrate Hydrates as a Novel Moderator Material for Very Cold Neutrons. Project Aperçu and First Results.

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Abstract. Clathrate hydrates are water-based solids with large crystallographic unit cells that show promise as potential moderators for use in new, more intense sources of very cold neutrons (VCN), which would enhance neutron scattering techniques and increase sensitivity of particle physics experiments. These so-called inclusion compounds seem particularly suitable for this application due to the low-energy modes of guest molecules engaged in nano-voids formed by a crystalline network of hydrogen bonded water molecules. In this article we present first results of an ongoing, extensive experimental campaign with the aim of characterizing the scattering properties of deuterated clathrate hydrates relevant for moderator applications. Experiments include neutron diffraction (carried out at Institut Laue-Langevin (ILL) using the instrument D20), as well as measurements of the temperature-dependent dynamical structure factor \( S(q, \omega) \) in absolute units (carried out on ILL’s Panther and IN5). These measurements will serve as a benchmark for development of new scattering kernels.

1 Introduction

Very cold neutrons (VCN) cover a wide spectral range within the long-wavelength tail of typical sources for cold neutrons \(^1\), with energies below, say, 1 meV (9 Å) down to few hundreds of neV (> several 100 Å), the domain of ultracold neutrons (UCN). Notably wavelengths of up to several tens of Å are in the focus of current interest of several research communities. In condensed matter physics, they serve as a sensitive probe and could improve multiple neutron scattering techniques in terms of spatial resolution as e.g. for small angle neutron scattering (SANS), or energy resolution as e.g. in time of flight (TOF) or neutron spin echo (NSE) spectrometers \(^1\). In particle physics, higher VCN intensities would increase the sensitivity of experiments that employ beams of slow neutrons. Examples are the search for neutron anti-neutron oscillations \(^2\), where the figure of merit (FOM) \( \propto \lambda^2 \), in-beam searches for a static neutron electric dipole moment (EDM), and further searches for new fundamental forces where FOM \( \propto 1 \) \(^3\) \(^4\). Furthermore, an enhancement of the flux of neutrons with wavelengths around 9 Å would be a valuable asset for in-beam UCN-sources based on super-fluid helium \(^5\), including variants with in-situ UCN production and detection approaches, as proposed in \(^6\). Despite the scientific potentials of VCN beams, at present the only existing VCN beamline is the instrument PF2 located at the Institut Laue-Langevin (ILL) in Grenoble, France. At this instrument, the vertical extraction by a strongly curved neutron guide from one of ILL’s cold sources transmits only the long-wavelength tail of the liquid-deuterium moderator spectrum. In order to realize higher fluxes of VCN, additional moderation to lower temperatures seems inevitable, which strongly motivates the search for appropriate, new moderator materials.

1.1 Required properties of novel cold and very cold moderator materials

The moderating properties of a material are practically determined by its excitation spectrum and its coupling strength to the neutron field via the cross section. Thus, promising materials for moderation to VCN need to possess low-energy modes that enable the incremental deposition of small quantities of the neutron’s energy. This can hardly be provided by collective excitations such as phonons since their cross section, for a neutron of energies \( E < k_B T_D \), in a cold medium with the Debye temperature \( T_D \) is proportional to \( (E/k_B T) \) \(^3\) \(^7\) (for single phonon emission) \(^7\). Localized, dispersion-free excitations, such as displacement of confined molecules, often referred to as Einstein modes, molecular rotations, librations or paramagnetic excitations, on the other hand, do not suffer from this limitation and allow for an efficient neutron slowdown even at lowest neutron temperatures. Besides suitable localized excitations, the material needs to exhibit low absorption and large scattering cross sections in the corresponding energy and temperature region. In addition, a moderator material must also fulfill more technical requirements. If its thermal conductivity is low, the material density to a moderator material must also fulfill more technical requirements. If its thermal conductivity is low, the material density...
must be arranged in a manner that enables efficient cooling. On top of that it must withstand radiational damage to a degree that prevents the formation of radiolytic hydrogen as well as the accumulation of free radicals. This could result in a self-sustaining reaction of their recombination leading to a fast heating of the moderator, often referred to as ”burp” [8] [9] [10]. In this article we present parts of an extensive inelastic neutron scattering study on different clathrate hydrate samples carried out on various neutron scattering instruments at the ILL.

1.2 A promising material class: Fully deuterated clathrate hydrates

Promising moderator materials investigated so far are different hydrocarbons, such as triphenylmethane [11], mesitylene and toluene [12], as well as clathrate hydrates [13] hosting methane or tetrahydrofuran (THF) [14]. Mesitylene is already in routine usage in form of a pelletized cold moderator at the IBR-2M reactor in Dubna [15]. This article focuses on the latter material class, where the enclosed guest molecules provides a range of localized, low-energy excitations that can be exploited for neutron slowdown. Given their very large crystallographic unit cells (as in the case of the clathrate structure II (CS-II) 17.1 Å – 17.33 Å, see table 1), which allows for coherent scattering up to neutron wavelengths of \( \lambda \approx 20 \) Å, these materials promise extraordinarily large albedos for the entire cold neutron range. This allows neutrons that did not scatter down to the desired energy range to be ”trapped” within the moderator material by diffuse transport, thus largely increasing the chance for further, energy decreasing interactions.

In this work, we present a study of clathrate hydrates hosting tetrahydrofuran as a guest molecule (sum formula: \( 17H_2O\cdotC_4H_8O \)), from here on referred to as THF-hydrates. A great advantage of this type of hydrate is its very reliable and reproducible manufacturing from a stoichiometric mixture under ambient pressure (see section 2.1). This would even allow for an in-situ annealing, thus re-establishing the clathrate structure within the moderator volume, in order to prevent a ”burp” of the moderator due to radiolysis. To some extent, the low-energy excitations of THF-hydrates have already been studied [14] [15]. We have extended those studies to include deuterated materials, and to cover \( S(q,\omega) \) in a large energy-momentum phase space in absolute units, which is essential for a quantitative analysis of the moderation properties in realistic scenarios.

While fully deuterated THF-hydrates (\( 17D_2O\cdotC_4D_8O \)) by themselves show very promising properties as a moderator material for VCN, they are also a good starting material for the formation of binary clathrate hydrates with both THF and molecular oxygen as guest molecules (\( 17D_2O\cdotC_4D_8O\cdot2O_2 \)).[14] Therein, \( O_2 \) occupies the small cages, which are twice as abundant as the bigger cages occupied by the TDF. The magnetic triplet ground state of molecular oxygen with its zero-field splitting of 0.4 meV allows for moderation via a cooling cascade mechanism in fully deuterated \( O_2 \)-clathrate hydrates, as described in [17]. It is this low excitation energy (as opposed to modes of several meV of the lattice and the THF) that will significantly enhance the moderation efficiency towards VCN, with the inelastic magnetic scattering providing a final cooling decrement.

1.3 Development of new scattering kernels in the HighNESS project

The study of novel VCN moderator materials is part of a collaborative effort within the European project HighNESS [18], committed to expanding the scientific potential of the European Spallation Source (ESS), by exploring new possible sources of slow neutrons. One of the goals is to establish a database of the dynamic structure factor \( S(q,\omega) \) for various promising clathrate hydrates and to develop novel scattering kernels that allow computation of neutron spectra in realistic moderator and reflector geometries in Monte Carlo (MC) simulations (see also Ref. [19]). To this end, the experimental campaign is accompanied by the development of simulation software to describe neutron interactions in materials that we investigate. The approach is to benchmark the excitation spectrum from ab initio molecular dynamics (MD) calculations, with the experimental data presented in this article. In combination with processing codes, such as NJOY [20] and NCrystal [21], this allows the creation of scattering kernels that are callable from McStas [22], OpenMC [23], MCNP [24] or PHITS [25]. This provides the tools to simulate novel moderator and reflector materials in realistic geometries, and compare simulations with results of experiments to be performed on a later stage using a large bulk of material as a demonstrator of a VCN moderator.

2 Experimental Program

A starting point of the experiments described here is establishing a reliable and scalable technique, that allows production of relatively large quantities of hydrates with minimal amounts of residual ice. This technique is described for THF-hydrates in section 2.1. The obtained structures were studied by neutron diffraction at the high-intensity two-axis diffractometer D20. An exemplary powder pattern and details of the associated Rietveld-refinement are given in section 2.2. The structure analysis is followed by a study of the low-energy dynamics of different THF-hydrate samples, as described in section 2.3. The data presented in this article were collected during cycles N° 189, N° 190 and N° 191 at the ILL.

2.1 Manufacturing procedures

A great advantage of THF-hydrates, compared to most other clathrate hydrate compounds, is that they form at ambient conditions from a stoichiometric mixture of its two liquid components, water (\( H_2O \)) and THF (\( C_4H_8O \)), [14] which allows for coherent scattering up to neutron wavelengths of \( \lambda \approx 20 \) Å, these materials promise extraordinarily large albedos for the entire cold neutron range. This allows neutrons that did not scatter down to the desired energy range to be ”trapped” within the moderator material by diffuse transport, thus largely increasing the chance for further, energy decreasing interactions.

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[1] The oxygen abundance is, unlike the \( C_2D_2O \) (TDF), non-stoichiometric and strongly depends on the sample preparation. This is represented by "\~0."
in a ratio of 17 to 1. After carefully weighing and mixing the two liquids with a teflon-coated magnetic stirrer, a cool-down of the solution results in solidification in the CS-II structure (see table 1). The CS-II structure is the most common among clathrate hydrates and its ideal unit cell contains 136 (H$_2$O) molecules forming 16 small and 8 large cages. This structure has been extensively studied before (see e.g. [26]). The study presented here focuses on the yields of the CS-II structure and quantifying the residual ice, for two different methods to cool down the sample. A great advantage of the manufacturing technique is, that it allows to perform a contrast variation between the host and the guest contribution in spectroscopy experiments. By simply substituting one of the liquid components with its deuterated counterpart (water (H$_2$O) with heavy water (D$_2$O) and THF (C$_2$H$_5$O) with TDF (C$_3$D$_5$O)) one obtains four different samples which are summarized in table 2.

### Table 1: Characteristics of the CS-II hydrate crystal cell structure

<table>
<thead>
<tr>
<th>Crystal system</th>
<th>Cubic</th>
</tr>
</thead>
<tbody>
<tr>
<td>Space group</td>
<td>Fd3m (N$^*$227)</td>
</tr>
<tr>
<td>Lattice description</td>
<td>Face centered</td>
</tr>
<tr>
<td>F Lattice parameters</td>
<td>$a = 17.1$ Å-17.33 Å</td>
</tr>
<tr>
<td>Number of cages</td>
<td>8 large ($5^{12}$ $6^4$), 16 small ($5^{12}$)</td>
</tr>
<tr>
<td>Ideal unit cell formula</td>
<td>8($5^{12}$ $6^4$) · 16($5^{12}$) · 136H$_2$O</td>
</tr>
</tbody>
</table>

The diffractometer was operated using the (115) reflection of a Germanium(113) monochromator at a wavelength of $\lambda = 1.546$ Å, at a take off angle of 90° resulting in a resolution of $\Delta d/d = 3 \cdot 10^{-3}$. The cylindrical sample container made from vanadium (with a diameter of 6 mm and a wall thickness of 0.1 mm) was filled with the liquid stoichiometric mixture and quenched in liquid nitrogen before being loaded into the cryogenic sample environment. After equilibration to a set temperature value, it took about 30 minutes to obtain a diffraction pattern within a scattering angle range of 4° to 150° with satisfactory statistics. This allowed measurements within a temperature range of 2 K - 230 K. Afterwards, the sample was heated up above the melting point ($\sim$ 277.5 K) and slowly (about 7 K per minute) cooled down again below 200 K. We observed that the CS-II structure is formed through both rapid quenching and slower cooling of the liquid sample. Effects of texture of the polycrystalline samples were observed as a dependence of diffraction patterns on sample orientation (which for an ideal powder would be absent). They are inherent for an uncontrolled in-situ crystallization, which can lead to a small number of crystallites and ultimately to a deviation from continuous powder lines (towards single points on the Debye-Scherer cones). Mitigation of this effect was attempted by rotating the sample in increments of 20° and adding up the resulting 9 diffraction patterns, for better approximation of a powder average. Figure 1 shows an exemplary diffraction pattern obtained with this procedure at a temperature of 230 K.

### Table 2: Differently deuterated and protonated samples prepared for spectroscopy experiments. The deuteration of either the guest molecule or the host lattice allows to highlight the different parts of the sample in the scattering signal. Note that the fully or partial deuteration is able to slightly change the dynamics of the sample, while the structure remains the one described in table 1.

<table>
<thead>
<tr>
<th>Abbrev.</th>
<th>Host</th>
<th>Guest</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fully protonated</td>
<td>THF · H$_2$O</td>
<td>136H$_2$O</td>
</tr>
<tr>
<td>Deuterated guest</td>
<td>TDF · H$_2$O</td>
<td>136H$_2$O</td>
</tr>
<tr>
<td>Deuterated cage</td>
<td>THF · D$_2$O</td>
<td>136D$_2$O</td>
</tr>
<tr>
<td>Fully deuterated</td>
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</tbody>
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### 2.2 Neutron diffraction on fully deuterated THF-hydrates

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### 2.2.1 Structural Details

Data analysis and structure refinement were performed in the program Fullprof [29] [30]. A multi-phase Rietveld-refinement (see e.g. [31]) yields the lattice constants, the positional and thermal parameters, as well as the weight
The TDF molecule was treated as a rigid body, with its position and the orientation within the large cage as free parameters. Aside from the phase under investigation (TDF-hydrates), the refinement includes the aluminum of the sample environment and residual hexagonal ice. The weight percentage \( W_j \) for the phase \( j \) can be calculated as in \([32]\):

\[
W_j = \sum_i S_i Z_i M_i V_{ij} / \sum_i S_i Z_i M_i V_{ij}
\]

with \( S, Z, M \) and \( V \) being the scale factor, the number of formula units per unit cell, the mass of one formula unit and the unit-cell volume, for each phase and \( i \), respectively. Fullprof also accounts for the multiplicities of each site, for occupation numbers unequal to 1, via the factor \( f_j \), and the micro absorption of neutrons (Brindley factor \( t_j \)).

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### 2.3 Time of flight spectroscopy

After having confirmed the CS-II structure of the samples solidified by quenching or slower cooldown within the cryostat we proceeded to study their low-energy excitations. To this end, experiments on the ILL time of flight (TOF) spectrometers IN5 \([34]\) and Panther \([35]\) were conducted. The goal of these experiments was to measure the dynamic structure function \( S(q, \omega) \) over a large fraction of \((q, \omega)\)-space, which is made possible by the wide and complementary kinematic range of those two instruments. The analyzed data of these measurements will finally allow benchmarking density functional theory (DFT) and molecular dynamics (MD) simulations (elaborated in Ref. \([19]\)).

The four samples described in Table 3 were measured under the configurations summarized in Table 4. Additional measurements of each empty sample holder were conducted in order to subtract their contribution from the respective scattering signal. Vanadium standards were used to normalize our data to absolute units. Particular emphasis was put on selecting the geometries of the sample and the standard as identical as possible. By choosing thin hollow cylinders (with a wall thickness of \( d = 0.05 \text{ mm} \) for the partly or fully protonated samples and \( d = 0.1 \text{ mm} \) for the fully deuterated one) as sample geometry the multiple scattering was reduced to a minimum \([36]\). Note that unlike at D20, the sample containers are made from aluminum. The respective vanadium standard was cut from a sheet of the same thickness and placed inside the sample container without overlap. The vertical intensity profile of the beam was mapped out prior to the measurements. This allowed us to place the samples and the vanadium standards in the intensity distribution such as to minimize potential systematic effects due to possible small differences in their vertical extensions.

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f = \frac{N_v \sigma_v}{N_S}
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purity and geometry of both the sample and the vanadium standard.

2.3.1 Low-energy excitations of THF-hydrates

Figure 2 and Figure 3 show two spectra for different THF hydrate samples measured at IN5 and Panther. These spectra are obtained by integration over a given $q$-range, providing an average of the coherent signal in this range. In the investigated energy region the dynamics of the host structure are dominated by the translational modes of the H$_2$O or D$_2$O molecule. This leads to two characteristic peaks at about 7 meV and 10.5 meV of every CS-II hydrate structure (see e.g. Ref. [41],[42],[14],[16]), which can also be observed in Figure 3 for both the protonated and the deuterated host structures. The most pronounced peaks are visible in the TDF · H$_2$O sample (orange), as the contribution of the deuterated guest molecule is suppressed. The excitations of the host molecule are located at lower energy and show very distinct peaks at about 2.9 meV and 4.7 meV, best visible in the THF · D$_2$O sample (green) in figure 3. These localized excitations are particularly promising for moderation to the VCN range. As expected, they are shifted to slightly lower energies when substituting hydrogen with deuterium due to the increased mass and thus moment of inertia. This is the case for both translational modes of the host lattice and the excitations of the THF molecule, and consistent with phonon density of states (PDOS) computed by our collaborators [19].

3 Conclusions

VCN sources that deliver higher fluxes of very slow neutrons than current "standard" liquid deuterium sources could provide a powerful new tool for the neutron scattering community in the investigation of mesoscopic structures (e.g. in SANS), as well as tiny energy transfers (e.g. in NSE). This holds potential for bringing valuable advantages to the fields of soft condensed matter and structural biology. Furthermore they could play a significant role in increasing the sensitivity of particle physics experiments that employ beams of slow neutrons.

This work presented preliminary experimental results necessary for the development of VCN moderators using fully deuterated clathrate hydrates as a moderation material. In particular, a characterization of the structure and low-energy dynamics of THF-hydrates is elaborated. We have demonstrated that producing these hydrates is straightforward, and we were able to investigate localized, low-energy excitations using a method that involves contrast variation through partial or complete deuteration of the sample’s constituents. The experimental determination of their dynamic structure factor $S(q, \omega)$ in absolute units provides an important benchmark for scattering kernels developed within the HighNESS project. Together, these works shall enable reliable simulations of future neutron moderators using fully deuterated clathrate hydrates as a moderating medium. Additionally, these experiments serve as a baseline for future experiments on binary clathrate hydrates with both THF and molecular oxygen as guest molecules (17D$_2$O:C$_4$D$_8$O:~2O$_2$).

4 Acknowledgement

We would like to thank the HighNESS collaboration, particularly Shuqi Xu from the European Spallation Source and Sara Isaline Laporte from the University of Milano-Bicocca for many inspiring discussions. This work was funded by the HighNESS project within the European Framework for Research and Innovation Horizon 2020, under grant agreement 951782.