Experimental report

15/09/2022

Proposal: 1-10-49 **Council:** 10/2020

Title: Absolute inelastic neutron scattering cross sections of clathrate hydrate compounds for new very-cold-neutron moderators

Research area: Methods and instrumentation

This proposal is a new proposal

Main proposer: Oliver ZIMMER

Experimental team: Thomas NEULINGER
Richard WAGNER
Valentin CZAMLER
Andrew WILDES
Thomas HANSEN
Michael Marek KOZA

Local contacts: Andrew WILDES
Thomas HANSEN
Michael Marek KOZA

Samples: clathrate hydrates

Instrument	Requested days	Allocated days	From	To
D20	1	1	12/10/2021	13/10/2021
D7	2	2		
PANTHER	4	2	25/05/2021	27/05/2021

Abstract:

The general goal of this proposal is the measurement of inelastic neutron scattering cross sections (S(q,omega)) of clathrate hydrate compounds in absolute units. These materials are particularly promising candidates for new moderators for very cold neutrons to enhance intensities in the long wavelength tail of existing cold sources. The proposed experiments shall complement an experiment to be executed in spring 2021 at the cold ToF spectrometer IN5, in order to cover the needed large range of q and omega. Additional measurements at D7 shall be used to determine cage filling of a binary clathrate hydrate with molecular oxygen, and also the H/D ratio independently from diffraction experiments at D20. The results of the whole effort will provide a valuable data base for model calculations of moderation efficiencies in realistic configurations

Experimental Report 1-10-49: Absolute Inelastic Neutron Scattering Cross Sections of Clathrate Hydrate Compounds for New Very-Cold-Neutron Moderators

Experimental Team: M. Koza, O. Zimmer, R. Wagner, V. Czamler

August 2022

1 Introduction

Clathrate hydrates have been identified as a very promising candidate for a moderation material for very cold neutrons (VCN), as they possess low-energy modes that allow pre-moderated neutrons to transfer small quantities of energy to the medium at sufficiently large scattering cross sections. To some extent, the low-energy incoherent excitations of methane and tetrahydrofuran (THF, chemical formula: C_4H_8O) clathrates have already been studied [3]. Experiment 1-10-49 contributed to a measurement campaign within the HighNESS collaboration that aimed to collect comprehensive data sets on $S(q, \omega)$ of clathrate hydrate compounds in absolute units. These data collected on ILL's time-of-flight instruments serve as a benchmark for the development of new scattering kernels within the collaboration as part of the NCrystal package [2], which is open source and fully accessible to the community. Using neutron transport codes such as McStas or GEANT4 with these scattering kernels will allow the calculation of yields in realistic moderator/reflector geometries.

2 Samples

The samples with stoichiometric composition $17H_2O.C_4H_8O$ were prepared by mixing the two liquid components. The two liquid components are mixed in the correct ratio. Samples of the mixture were then filled into their respective sample containers in which they were frozen in situ. The containers defined samples in the geometry of thin-walled hollow cylinders, which limits multiple scattering, while still exposing sufficient material to the beam [4]. The (inner) sample diameters were 14 mm & 15 mm, with a sample thickness of 0.1 mm & 0.05 mm and a height of 20 mm (see figure 1). In order to achieve a contrast variation between

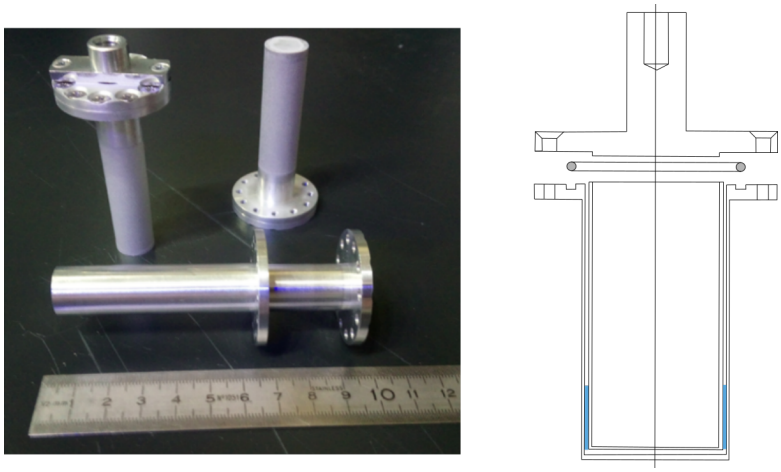

Figure 1: Photograph of sample containers used in experiment 1-10-49 (left) and drawing of these sample containers (right). The filling height of 2 cm is indicated in blue. The containers are sealed with indium.

different components of the material, samples with different partial, absent or complete deuteration of its

components were prepared: (a) D_20:THF-d, (b) H_20:THF-d, (c) D_20:THF, (d) H_20:THF. This is expected to highlight scattering contributions of the host lattice and the guest molecules to the measured spectrum. In order to calibrate the results to absolute units, measurements were also performed with empty sample holders, and with weighted vanadium samples with the same geometry as the clathrate samples.

3 Preliminary Results

Measurements were taken for two different incident energies, $E_i = 76$ meV (~ 1 Å) and $E_i = 19$ meV (~ 2 Å) of all four different samples and the respective vanadium samples for normalization. Figure 2 shows typical, still uncorrected data of the neutron scattering function $S(q, \omega)$ for a fully deuterated $17D_2O$: TDF, $E_i = 76$ meV. The strong vertical features are due to the aluminum container. Figure 3 shows a q-averaged slice through $S(q, \omega)$. The contrast variation allows to differentiate lattice modes at around 7 and 11 meV, and the guest molecule's modes.
Measurements with the empty containers served to correct for the neutron scattering by aluminum and other backgrounds. The data reduction was done in Mantid [1]. Emphasis was put on the normalization to absolute units and on correction for all instrument specific features, with the objective to create accurate and reliable data sets that serve as baseline for future simulations.

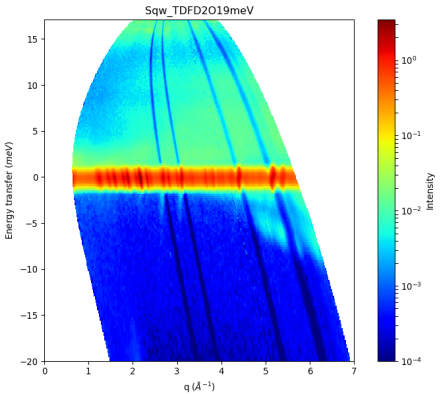

Figure 2: Measured data of the neutron scattering function $S(q, \omega)$ for a fully deuterated $17D_2O$: THF-d-clathrate, $E_i = 76$ meV (~ 1 Å).

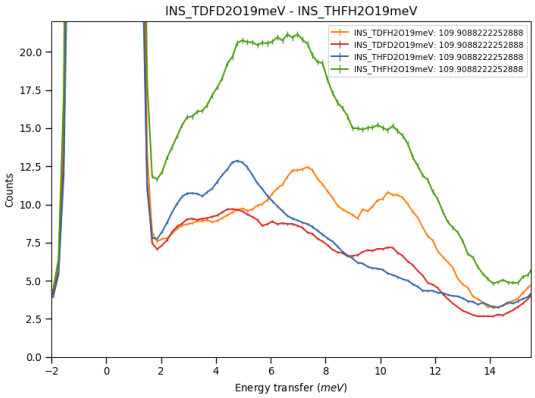

Figure 3: q-integrated slice of $S(q, \omega)$ as depicted in figure 2.

Preliminary results show good agreement with molecular dynamic simulations of our collaborators. Future experiments shall cover supplementary parts of phase space in order to complement the data sets of IN5 and Panther.

References

[1] O. Arnold, J. Bilheux, J. Borreguero, A. Buts, S. Campbell, L. Chapon, M. Doucet, N. Draper, R. Ferraz Leal, M. Gigg, V. Lynch, A. Markvardsen, D. Mikkelson, R. Mikkelson, R. Miller, K. Palmen, P. Parker, G. Passos, T. Perring, P. Peterson, S. Ren, M. Reuter, A. Savici, J. Taylor, R. Taylor, R. Tolchenov, W. Zhou, and J. Zikovsky. Mantid—Data analysis and visualization package for neutron scattering and SR experiments. *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, 764:156–166, Nov. 2014.

[2] X.-X. Cai and T. Kittelmann. NCrystal: A library for thermal neutron transport. *Computer Physics Communications*, 246:106851, 2020.

[3] H. Conrad, W. F. Kuhs, K. Nünighoff, C. Pohl, M. Prager, and W. Schweika. Inelastic scattering and spectral measurements of advanced cold moderator media. *Physica B: Condensed Matter*, 350(1, Supplement):E647–E650, July 2004.

[4] J. Wuttke. Improved sample holder for multidetector neutron spectrometers. *Physica B: Condensed Matter*, 266(1):112–114, May 1999.