Experimental report

07/09/2022

Proposal: 1-01-194 **Council:** 4/2021

Title: Absolute inelastic neutron scattering cross sections of the binary clathrate hydrate 17D2O:THF-d:2O2 fora very-cold-neutron moderator

Research area: Methods and instrumentation

This proposal is a new proposal

Main proposer: Oliver ZIMMER

Experimental team: Richard WAGNER
Valentin CZAMLER
Arnaud DESMEDT

Local contacts: Michael Marek KOZA
Jacques OLLIVIER

Samples: Water
Oxygen: O2
tetrahydrofuran

Instrument	Requested days	Allocated days	From	To
IN5	2	1	07/10/2021	08/10/2021

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 experiment shall complement an experiment executed in February 2021 at the cold ToF spectrometer IN5, using now the same typ of sample but with the abundant empty cages of the clathrate structure now filled with molecular oxygen, thus forming a binary hydrate clathrate with an exploitable intramolecular magnetic transition of only 0.4 meV.

Experimental report for experiment 1-01-194: Absolute inelastic neutron scattering cross sections of the binary clathrate hydrate 17D2O.THF-d.2O2 for a very-cold-neutron moderator

Experimental team: A. Desmedt, V. Czamler, J. Ollivier, R. Wagner, O. Zimmer

Introduction:

This experiment had the goal to determine the scattering function $S(q,\omega)$ in absolute units for the fully deuterated, stoichiometric binary clathrate hydrate $17D_2O.THF\text{-}d.2O_2$. This material has been identified as a particularly promising candidate for moderation of thermal or cold to very cold neutrons. Compared to the simpler $17D_2O.THF\text{-}d$, where the deuterated tetrahydrofuran molecules (THF-d, chemical formula C_4D_8O) occupy the large cages in the clathrate structure, in the binary clathrate the small cages (there are twice as many as large ones) are in addition occupied by O_2 molecules. These are expected to add inelastic magnetic scattering intensity, due to the magnetic triplet ground state of molecular oxygen with its zero-field splitting of 0.4 meV.

Sample:

The sample was produced in Bordeaux by exposing a stoichiometric liquid mixture of THF-d and D_2O near the freezing point for several days to gaseous oxygen at high pressure from a gas bottle. The clathrate hydrate was collected and cooled down to liquid nitrogen temperature, at which it was stored and later transported to the ILL. On the day of the experiment, the material was ground with a mortar cooled by liquid nitrogen into few-micron sized pieces, which were then filled into a sample holder of tubular geometry with a diameter of 15 mm and a gap of 0.5 mm.

Experiment:

The experiment was performed during one day of beam time at the cold-neutron Time of Flight spectrometer IN5, using a cryostat to cool the sample down to 1.5 K. As the sample preparation did not allow a quantitative determination of its mass, it was foreseen to relate the intensity due to the O_2 molecules to the excitations due to the THF-d molecules, which occur at higher energies. Prior experiments performed at IN6 with a pure O_2 gas hydrate had shown, as expected, a dispersion-free intensity at 0.4 meV with q-dependence due to the magnetic form factor of the O_2 molecules. In the present experiment, however, this sharp excitation was nearly invisible, while the intensity due to the THF-d molecules was well detectable. The most likely reason for this (non-)observation is a significant under-filling of the small clathrate cages with oxygen, which could be due to the preparation of the clathrate hydrate or a loss of O_2 during the sample preparation. It is planned to reattempt these measurements at a later stage, using a sample manufactured by a different procedure.