Experimental report

04/03/2021

Proposal:	**TEST-2195**	**Council:**	10/2016
Title:	Study of the water diffusion in hydrophobic confinement		
Research area:			

This proposal is a new proposal

Main proposer:	**Margarita RUSSINA**
Experimental team:	Ferenc MEZEI Margarita RUSSINA Moritz-C. SCHLEGEL
Local contacts:	Peter FOUQUET

Samples: H2O

Instrument	Requested days	Allocated days	From	To
IN11	9	9	31/07/2013	09/08/2013

Abstract:

TEST-2195 Investigation of anisotropic water dynamics in hydrophobic 1D zeolite channels.

Instrument: IN11C, 31/07/2013 - 09/08/2013

Team: M. Russina, M. Schlegel, F. Mezei

Instrument supervision: P. Fouquet

The goal of this test was a feasibility study of the microscopic mobility and transport of water confined into zeolite AlPO4-5 with oriented pore structure, which form one-dimensional channels with diameter of about 8 Å. *This approach opens up the opportunity to measure directly and model independently the characteristics of the diffusive motion as a function of direction*. Water exists in confined environments and at interfaces in a wide variety of important systems in chemistry, biology, geology, materials science and technological applications. In biology, water is found in crowded environments such as cells, in which it hydrates membranes and large biomolecules, as well as pockets in proteins. In geology, interfacial water molecules can control ion adsorption and mineral dissolution. In chemistry, water plays an important role as a polar solvent that is often in contact with interfaces. At the molecular level, hydrophobic effects are thought to drive a variety of biological and colloidal self-assembly phenomena in aqueous solution, including the folding of proteins and the formation of micelles and membranes. There were several efforts to study the relation between the dimensionality and diffusion in clays and porous carbon. However, despite of the substantial amount of the experimental work in combination with extensive numerical calculations the conclusion was that the experimental precision is insufficient to allow us to discriminate between the various possible contributions of $D_{\parallel}$ and $D_{\perp}$.

For our study we will use several *c*-oriented AlPO$_4$-5 zeolite films. The zeolite exhibit nanopores of Ø = 7.3 Å , which form one-dimensional channels. The samples have been characterized before using TEM and diffraction measurements (Fig.1). The water sorption for H_2O and D_2O has been also studied in detail at T=25°C (Fig.2) and will be used for the calibration of neutron scattering experiments. Isotherm exhibits a clear S-shape, whose strongly vertical rise points out the uniform pore size with no interconnection (Fig1). The occupation of the channels by water molecules at different humidity was also characterized using X- ray and neutron diffraction [1]. In our NSE experiment oriented ALPO4-5 single crystals have been grown in the Si substrate and have been oriented either with Q vector parallel or perpendicular to axial direction of the main channel. The time scale was roughly reaching 1 ns. We observe a pronounced anisotropy in the quasielastic spectra measured at IN11C (Fig.3). Since these differences were not observed in the spectra of dry samples, we can conclude therefore that this effect is caused by differences in the dynamics of water across and along the channels. However, for the detailed

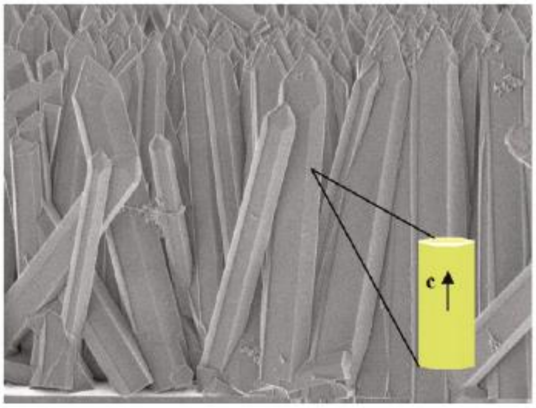

Figure 1: TEM image of the ALPO4-5 films, used in the IN11 C experiment.

assessment the study must be done in wider Q and time domains and with better characterization of the background.

Nevertheless, the results of the feasibility test were published in [2]. In the published study in addition to IN11C we used NEAT @HZB and DNA@J-PARC and investigated confined water dynamics up to 240 ps. The study reveals two-steps diffusion-based relaxation and novel state of highly cooperative water. While the first step originates from the fast spontaneous position exchange between two neighboring water molecules, the nature of the second, slow process is not quite clear with several possible explanations. To discriminate between them we need information about transport parameters in the larger time domain, information about propagation direction, activation energies and etc., therefore a continuation of the study is planned.

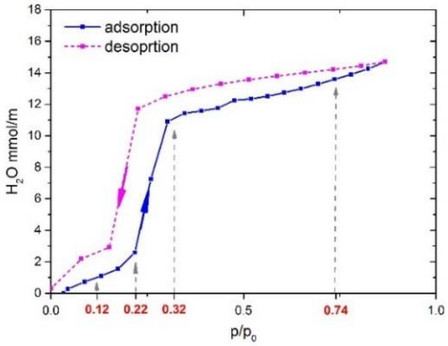

Figure 2 Water sorption isotherm in ALPO4-5 zeolite with pore size of about 1 nm. Up to p/p0 =0.22 corresponds to water occupation of positions close to the pore walls. In 0.22<p/p0 <0.32 water enters the pore central area with 90% pore filling at p/p0 =0.32. Further increase up to p/p0 =0.74 corresponds mostly to water sorption in the surface of zeolite.

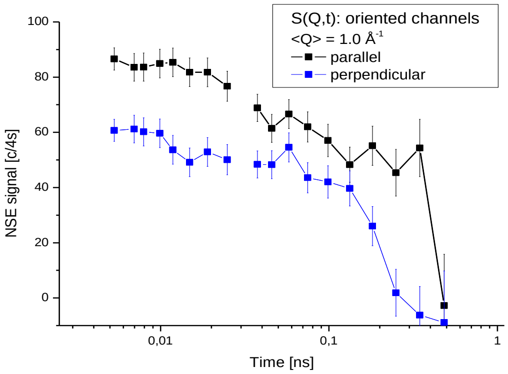

Figure 3 Neutron spin echo spectra at different orientation of the hydrated oriented zeolite sample collected at IN11C.

Publications:

[1] M. C. Schlegel, M. Russina et al *Microporous Mesoporous Mat., 2020, 109201* DOI:10.1016/j.micromeso.2018.11.025, 2018. ;

[2] M. Russina, et al, *J. Phys. Chem Lett.*, 2019, 10, 20, 6339-6344, https://doi.org/10.1021/acs.jpclett.9b02303