Experimental report

09/10/2023

Proposal: 6-07-108 **Council:** 10/2022

Title: Study of H2 mass transport and dynamics in ordered microporous zeolite-templated carbons

Research area: Materials

This proposal is a new proposal

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Experimental team: Rasmus PALM

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Samples: zeolite-templated carbon (pure carbon)

Instrument	Requested days	Allocated days	From	To
IN5	5	4	04/04/2023	08/04/2023

Abstract:

Development of high efficiency and safe hydrogen storage materials is vital for the move towards a hydrogen-based greenhouse gas emission free and sustainable economy. Zeolite-templated carbons (ZTCs) are a class of highly potential hydrogen storage materials. In addition, the highly ordered microporous structure of ZTCs is excellent as a model material to investigate the hydrogen-carbon interactions in pores with specific sizes, geometries, and with chemical dopants. In this study we propose to investigate the influence of the ordered microporous ZTC structures and N-functionalisation of these structures on the hydrogen mass transport properties. The hydrogen mass transport and dynamic properties will be determined from the quasi-elastic broadening of H2 neutron scattering. From this the influence of N-functionalise groups on H2 confinement in the porous structure will be determined. The H2 transport properties will be collaborated with the porous structure characterisation applied for on SANS instrument D33 and with lab-scale characterisation.

Study of H_2 mass transport and dynamics in ordered microporous zeolite-templated carbons

Scientific background and aim of this experiment

Adsorption of H_2 in high surface area microporous carbon materials for hydrogen storage applications has been extensively investigated [1–4]. An important aspect to fully understand H_2 adsorption in microporous carbon materials is understanding the mass-transfer and mobility of adsorbed H_2 at low temperatures ($\leq$ 77 K) in the confining influence of pore walls and the influence of doped functionalised groups [4–7]. The formation of highly dense adsorbed H_2 phases in micropores has been shown previously, where the adsorbed phase was shown to be denser than liquid H_2 [7–9]. Thus, in this experiment, we investigated the role of N-functionalisation and ordered microporous structures on the self-diffusional properties H_2 phase. In addition, changes in the carbon structure caused by the adsorbed H2 are determined from the elastic scattering part of the measured data. For this, zeolite-templated carbons (ZTCs) were used as robust model materials for their highly ordered pore structure with well-defined pore widths and high specific surface areas (up to 3100 $m^2\ g^{-1}$) and the possibility to control N-heteroatom functionalization.

Experimental routines

Neutron scattering measurements were performed on three ZTC materials: Bulk carbon ZTC, ZTC1, and two N-heteroatom active sites containing ZTCs, ZTC2 and ZTC3. All ZTCs were degassed before measurement, and neutrons with wavelengths of 2 and 4.8 Å were used. The used experimental routines are brought in Table 1.

Table 1. Used experimental routine

ZTC1	ZTC2	ZTC3	Empty sample holder
Degassing at RT	Degassing at RT	Degassing at RT	Degassing at RT
Cooling to 10 K	Cooling to 10 K	Cooling to 10 K	Cooling to 10 K
$S(Q,E)$ measurements from 10 K to 80 K with 10 K temperature steps with neutron wavelength 4.8 Å	$S(Q,E)$ measurements from 10 K to 80 K with 10 K temperature steps with neutron wavelength 4.8 Å	$S(Q,E)$ measurements from 10 K to 80 K with 10 K temperature steps with neutron wavelength 4.8 Å	$S(Q,E)$ measurements from 10 K to 80 K with 10 K temperature steps with neutron wavelength 4.8 and 2 Å
Dosing with 0.323 bar of H_2 at 77 K	Dosing with 0.584 bar of H_2 at 77 K	Dosing with 0.74 bar of H_2 at 77 K	
Cooling to 10 K	Cooling to 10 K	Cooling to 10 K	
$S(Q,E)$ measurements from 10 K to 80 K with 10 K temperature steps with neutron wavelength 4.8 and 2 Å	$S(Q,E)$ measurements from 10 K to 80 K with 10 K temperature steps with neutron wavelength 4.8 and 2 Å	$S(Q,E)$ measurements from 10 K to 80 K with 10 K temperature steps with neutron wavelength 4.8 and 2 Å	

Experimental results

The degassed ZTC S(Q,E) were determined to be temperature dependant, where a slight increase in backgorund scattering intensity was detected in case of increased temperature. With the applied H_2 loading QE broadening was detected already at 40 K (Figure 1), which corresponds to the mobile adsorbed H_2 in the nanometer scale pores of the ZTC. In addition, in ZTC2 and ZTC3, i.e., ZTCs with N-heteroatom additions and with the presence of smaller pores, the formation of scattering peaks corresponding to ordered domains were measured at 40 and 50 K. After heating to 60K, the scattering peaks dissapeared (Figure 2).

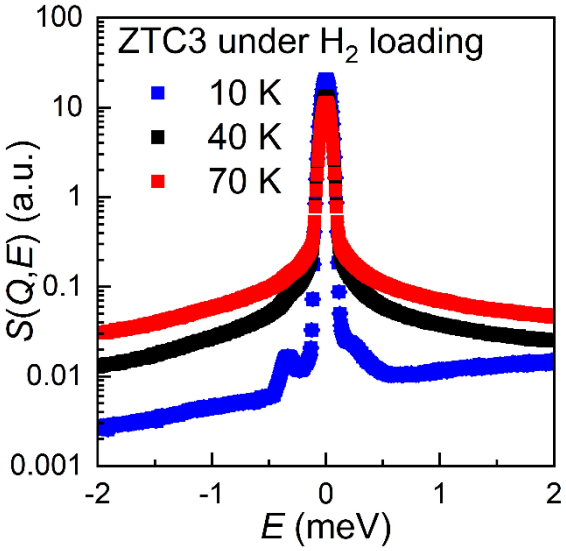

Figure 1. $S(Q,E)$ of ZTC3 under $p_{H2,load}$ of 0.74 bar, applied at 77 K.

Further plans

Deconvolution of the S(Q,E) data will be performed to separate the fractions corresponding to mobile H_2. The areas and FWHM of the mobile fractions will be analysed and translational jump diffusion models will be used to determine the diffusion coefficients, jump length distributions and activation energies of the H_2 translational jumps. A manuscript will be prepared based on the obtained results after the paper outlining the synthesis of the used ZTC has been published (is currently in the works). Based on the results from ZTC3, experiments utilizing polarized neutron analysis are planned to separate the coherent and incoherent part of scattering from H_2 to be able to determine exactly which part of the formation of the three scattering peaks at 40 and 50 K is caused by the adsorbed H_2 phase or changes in the structure of the ZTC3.

References

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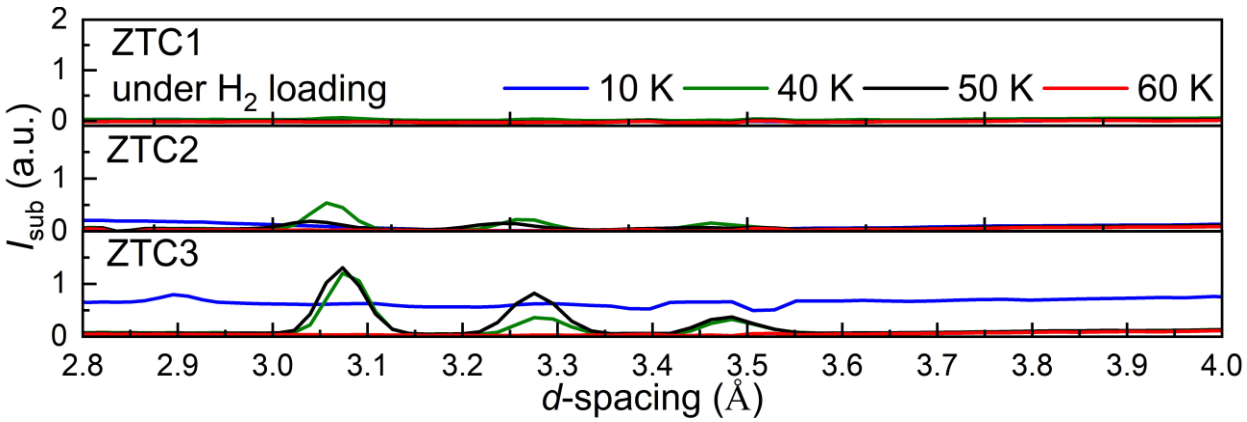

Figure 2. $S(Q)$ of all three ZTCs integrated in energy transfer from −0.1 to 0.1 meV and from which the corresponding $S(Q)$ from degassed ZTCs has been substracted.