

# Experimental report

09/10/2023

**Proposal:** 6-07-106

**Council:** 10/2022

**Title:** Study of the porous structure of pure and N-functionalised zeolite-templated carbons

**Research area:** Materials

**This proposal is a new proposal**

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**Samples:** zeolite-templated carbon (pure carbon)

Instrument	Requested days	Allocated days	From	To
D33	1	1	17/04/2023	18/04/2023

## 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 are a class of highly potential hydrogen storage materials. In addition, the highly ordered microporous structure of zeolite-templated carbons 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 apply to characterise the porosity of three high surface area microporous zeolite-templated carbons. Two of the zeolite-templated carbons have been functionalised with N-groups to investigate the hydrogen interactions with N-functionalised ordered microporous carbon structures in complementary lab-scale and SANS and ND measurements. Deuterated toluene contrast-matched SANS will be used to subtract the signal from closed porosity and density fluctuations from inside the carbon material and to determine the prevalent structure of partially filled porosity. This measurement will yield highly valuable characterisation of the porous structure of materials of high interest.

# Study of the porous structure of pure and N-functionalised zeolite-templated carbons

## Scientific background and aim of this experiment

Adsorption of H<sub>2</sub> in high surface area microporous carbon materials for hydrogen storage applications has been extensively investigated [1,2,3,4]. In addition to the pore size, the pore geometry and the corrugation of the pore walls have been shown to strongly influence the confinement of H<sub>2</sub>, where prevalently spherical pores can confine H<sub>2</sub> at much higher temperatures [4,5]. N-functionalisation has been shown to yield varying results regarding the enhancement of hydrogen storage capability, where increases in H<sub>2</sub> adsorption capability and no effect have been reported by different sources [6,7,8]. Thus, in this experiment, contrast matched small angle neutron scattering (SANS) was used to characterise the porous structure of zeolite-templated carbons (ZTCs), where the same ZTCs were used in two additional experiments at ILL to measure formation of an ordered adsorbed D<sub>2</sub> phase and self-diffusion of adsorbed H<sub>2</sub>. Contrast matching with toluene-d<sub>8</sub> (C<sub>7</sub>D<sub>8</sub>) was performed to subtract the scattering caused by the density fluctuations inside the adsorbent phase and to probe the structure of partially filled porosity [9,10,11]. The three used ZTCs are robust model materials for their highly ordered pore structure with well-defined pore widths and high specific surface areas (up to 3100 m<sup>2</sup> g<sup>-1</sup>) and with controlled N-heteroatom functionalization.

## Experimental routines

SANS curves were measured 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 all ZTCs were measured as dry material, submerged in C<sub>7</sub>D<sub>8</sub>, and with wetted C<sub>7</sub>D<sub>8</sub>, where C<sub>7</sub>D<sub>8</sub> vapor phase was used for partial pore filling. Measurements were performed with incident neutron wavelength of 4.62 Å. In addition, SANS curves of bulk C<sub>7</sub>D<sub>8</sub> and empty cuvettes were obtained for background subtraction and scattering data processing routines.

## Experimental results

All scattering curves were obtained successfully and the scattering from carbon density fluctuations was subtracted, yielding only scattering from the porous structure (Figure 1). Differences in the unfilled pore structure and

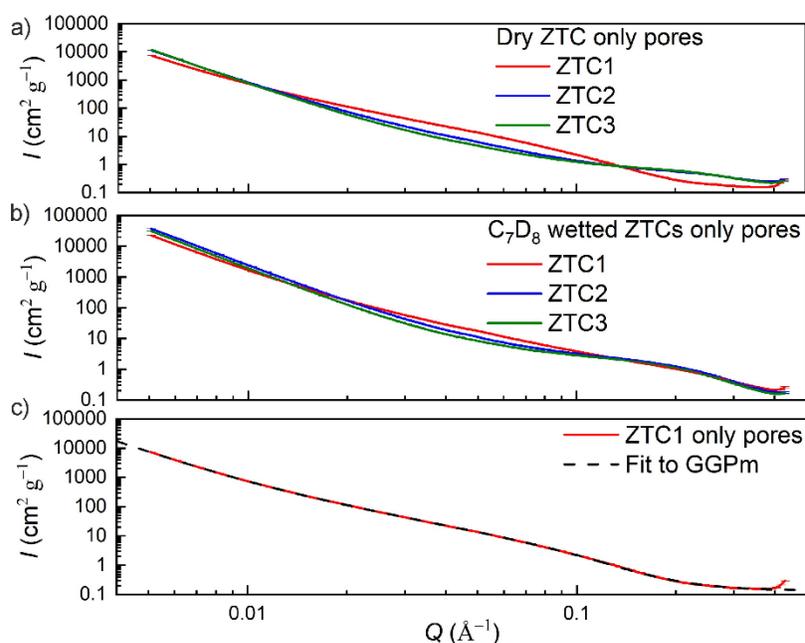


Figure 1. SANS scattering curves of only pores in a) dry ZTCs, b) ZTCs partially wetted with C<sub>7</sub>D<sub>8</sub>, and c) fitting of the dry ZTC1 porosity to generalized Guinier-Porod model.

pore structure partially filled with C<sub>7</sub>D<sub>8</sub> were measured and determined (Figure 1a and 1b). In addition, generalized Guinier-Porod model was applied to determine the average radius of gyration and the average dimensionality of the pores.

### Further plans

Fitting with generalized Guinier-Porod model and other methods, e.g., chord length distribution, will be used to characterise the porosity and its accessibility to C<sub>7</sub>D<sub>8</sub>. The obtained characterisation results will be used to support the discussion of the H<sub>2</sub> self-diffusional properties obtained from QENS measurements and the formation of ordered D<sub>2</sub> adsorbed phase from NPD measurements. These results will be used in a paper dealing with the beforementioned H<sub>2</sub> and D<sub>2</sub> adsorbed phase investigations.

### References

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