

## CHARACTERIZATION OF SOOT ADSORPTION OF VOLATILE ORGANIC COMPOUNDS

Frederik Ossler<sup>1\*</sup>, Monika Hartl<sup>2</sup>, Luke L. Daemen<sup>3</sup>, Anibal J. Ramirez-Cuesta<sup>3</sup>,  
Charles E.A. Finney<sup>4</sup>, Jean-Christophe Bilheux<sup>3</sup>, Hassina Z. Bilheux<sup>3</sup>

<sup>1</sup>*Combustion Physics, Lund University, Lund, Sweden*

<sup>2</sup>*European Spallation Source, Lund University, Lund, Sweden*

<sup>3</sup>*Neutron Scattering Division; <sup>4</sup>Energy and Transportation Science Division  
Oak Ridge National Laboratory, Oak Ridge TN USA*

\*Presenting author's e-mail: frederik.ossler@forbrf.lth.se

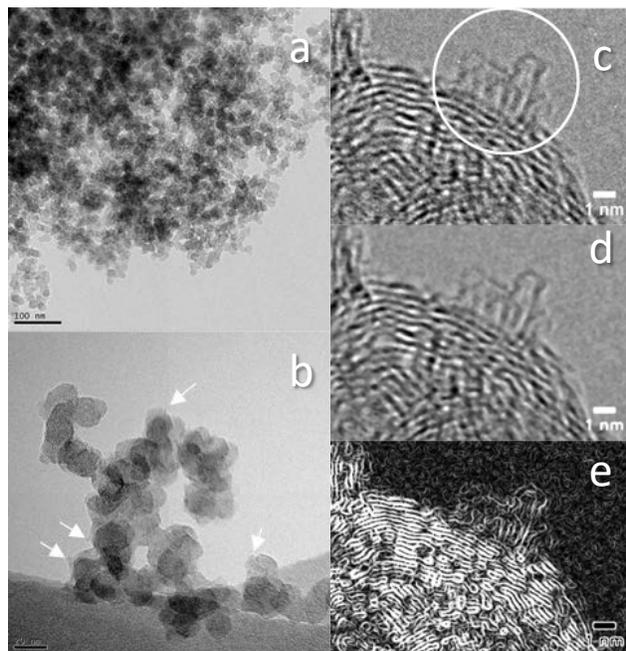
### Introduction

Soot materials have potential important benefits to society, since they, in principle, can be used as porous carbon materials for different applications. They may have a lower expected storage capacity than other porous carbon materials such as activated porous carbons, but they have considerable mechanical and thermal flexibility and greater percolation properties, which can make them interesting materials for different applications in e.g. flow dynamics and control and short-term gas storage for the energy and environmental control sectors.

We have previously performed neutron imaging based *in-situ* studies of methane and ethane in soot materials at different pressures (up to 100 bar) and at different temperatures down to cryogenic conditions at the Oak Ridge National Laboratory (ORNL) High Flux Isotope Reactor CG-1D beamline that have been presented at the Carbon 2018 conference in Madrid. Lately, we have also performed inelastic scattering (INS) measurements at the ORNL Spallation Neutron Source VISION beamline which show important new results which concern the adsorption of volatile organic compounds in soot materials. In particular, we focus on the differences between dry and wet soot materials that have not only interesting implications for technical utilization but also of fundamental science-based perspective in relation to atmospheric and environmental studies.

### Results and Discussion

Cold neutrons with a peak wavelength of 2.6 Å were used to measure the local concentrations of methane (CH<sub>4</sub>) and ethane (C<sub>2</sub>H<sub>6</sub>) in pristine soot material based on neutron radiography utilizing the method described in a previous paper<sup>1</sup>. Inelastic neutron scattering is also being employed to study the interaction between the surface of the porous carbon soot networks (PCSN), where the building blocks are the so-called soot primary particles, which are overall spherical but have varying internal and surface physical structure, often with considerable aromatic layered structure as seen from the transmission electron microscopy (TEM) images in Fig.1. Previous investigations have shown that soot materials contain mostly C, about 14% (mole fraction) H and variable amounts of O around half that of H. We obtain results that indicate that the storage capacity (SC, ratio of amount of hydrocarbon absorbed inside the network with respect to an empty space of the



*Fig. 1: TEM images of PCSN from an ethylene flame. (a) panoramic detail, (b) details of a network unit. Details of a primary particle showing (c) original-, (d) noise-reduced, and (e) edge-enhanced rendered images. The arrows in (b) indicate structures shown in the circular region in (c).*

same total volume) for  $\text{CH}_4$  and  $\text{C}_2\text{H}_6$  depend on the properties of the network, in terms of how fixed the building blocks are bound to each other. Another important aspect is the behavior of the PCSN to absorb and retain these hydrocarbons inside the network across the phase transition line of the respective hydrocarbon. At the Carbon 2018 conference, we showed that at cryogenic temperatures the network can act as a sponge, and we observed SC values as high as 10 with  $\text{CH}_4$ . New results from last year's study show that higher values can be achieved for  $\text{C}_2\text{H}_6$ , with  $\text{SC} \sim 30$  as the maximum value observed so far. For 200 K we obtained  $\text{SC} = 27$  at 2 bar, i.e., around the saturation pressure of  $\text{C}_2\text{H}_6$ , and the sublimation temperature of dry ice (194.65 K) at atmospheric pressure. These conditions are of potential interest for new technical solutions related to storage and transportation of  $\text{C}_2\text{H}_6$ , which is an important chemical and a high-value fuel. INS allows us to monitor the interaction of the condensed substances with the surface of

the network at cryogenic temperatures that are volatile at ambient conditions. Thus, INS becomes a particularly important instrument to understand the thermodynamic properties of several gaseous species inside PCSN. We have looked at the behaviour of  $\text{CH}_4$  and  $\text{C}_2\text{H}_6$  on dry and moist (water) networks as well as at para- $\text{H}_2$ ,  $\text{CO}_2$ , and acetone. We have also modified the soot surface utilizing oxidation and reduction schemes as well as subjected soot to methylation and amination. Work in progress will be presented and discussed. In addition to INS, high-resolution electron microscopy and Fourier transformation infrared spectroscopy are also being introduced to complement the characterization of the materials, and the status will be reported.

### Acknowledgment

We acknowledge Crispin Hetherington of Lund University for performing the electron microscopy measurements related to Fig.1. Author F.O. acknowledges the Generic Research for Optimized Energy Conversion Processes (GRECOP), project number 38913-2, supported by the Swedish Energy Agency. This research used resources at the High Flux Isotope Reactor and the Spallation Neutron Source, DOE Office of Science User Facilities operated by the Oak Ridge National Laboratory.

### References

1. Ossler F., Santodonato L.J., Bilheux H.Z. (2017). In-situ neutron imaging of hydrogenous fuels in combustion generated porous carbons under dynamic and steady state pressure conditions. *Carbon* 116, 766–776. doi: [10.1016/j.carbon.2017.02.025](https://doi.org/10.1016/j.carbon.2017.02.025)