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Nanometre-sized pores in coal: Variations between coal basins and coal origin

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ABSTRACT

We have used small angle neutron scattering (SANS) to investigate the differences in methane and hexane penetration in pores in bituminous coal samples from the U.S., Canada, South Africa, and China, and maceral concentrates from Australian coals. This work is an extension of previous work that showed consistent differences between the extent of penetration by methane into 10–20 nm size pores in inertinite in bituminous coals from Australia, North America and Poland.

In this study we have confirmed that there are differences in the response of inertinite to methane and hexane penetration in coals sourced from different coal basins. Inertinite in Permian Australian coals generally has relatively high numbers of pores in the 2.5–250 nm size range and the pores are highly penetrable by methane and hexane; coals sourced from Western Canada had similar penetrability to these Australian coals. However, the penetrability of methane and hexane into inertinite from the Australian Illawarra Coal Measures (also Permian) is substantially less than that of the other Australian coals; there are about 80% fewer 12 nm pores in Illawarra inertinite compared to the other Australian coals examined. The inertinite in coals sourced from South Africa and China had accessibility intermediate between the Illawarra coals and the other Australian coals.

The extent of hexane penetration was 10–20% less than CD₄ penetration into the same coal and this difference was most pronounced in the 5–50 nm pore size range. Hexane and methane penetrability into the coals showed similar trends with inertinite content.

The observed variations in inertinite porosity between coals from different coal regions and coal basins may explain why previous studies differ in their observations of the relationships between gas sorption behavior, permeability, porosity, and maceral composition. These variations are not simply a demarcation between Northern and Southern Hemisphere coals.

1. Introduction

The micro-, meso- and macro-sized pores in coal determine the movement of gases and liquids within coals. Coal has a strong propensity to sorb gas and possesses substantial porosity. However, previous studies of gas sorption and pore volume of coal do not show consistent trends in gas sorption, porosity or permeability measurements with maceral composition. For example, Teng et al. (2017) concluded that vitrinite was more porous than inertinite in Illinois Basin, U.S. coal, while Shan et al. (2015) and Berbesi et al. (2009)

concluded that vitrinite was less porous than inertinite in coals from the Sichuan Basin, China and the Guasare Coalfield, Venezuela, respectively. Adeboye and Bustin (2013) and Keshavarz et al. (2017) showed permeability increased with increasing inertinite in coal from the western Canadian sedimentary basin and the northern Sydney Basin respectively. Unsworth et al. (1989) found that total porosity increased with increasing inertinite content, but that vitrinite had more accessible pores in the < 2 nm size range than the inertinite in a suite of globally sourced coals. Using mercury porosimetry, Stanmore et al. (1997) found that the inertinite in a suite of coals from Queensland contains

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more total porosity than the corresponding vitrinite: the porosity in vitrinite was similar in all of the coals examined and porosity in inertinite varied widely between the coals. Other studies found no correlation between maceral composition and total porosity (Gürdal and Yalçın, 2001; Kędzior and Jelonek, 2013). Weishauptová and Sýkorová (2011) indicated that CO₂ sorption capacity increased with increasing vitrinite content for coals from one Polish coal basin but not another. These conflicting results are, in part, due to the different techniques that were used to measure porosity within the coal samples and that the coals were sourced from different coal basins and coal regions. The depositional and thermal history of a coal basin may play a role in the relationships between porosity, permeability, gas flow, and maceral composition. To test this, we have examined a suite of coal samples from different coal basins and coal regions with a single technique, small angle neutron scattering (SANS).

The SANS technique can be used to determine the fraction of open pores as a function of pore size. In a SANS experiment the scattering intensity from a sample is recorded as a function of a momentum transfer Q , which is defined as.

$$Q = 4\pi \sin(\theta/2)/\lambda \quad (1)$$

where θ is the scattering angle (Fig. 1) and λ is the wavelength.

The indicative pore radius can be obtained from Q using the approximate relationship $R \sim 2.5/Q$, where most of the contribution to the scattering at a given Q comes from pores of this radius $\pm 50\%$ (Radlinski et al., 2000).

Information on the fraction of open pores is obtained using the technique of contrast matching. The scattering intensity from a two-component system such as the solid coal matrix and the pore-invading fluid is proportional to the square of the difference in the scattering length density (SLD) of the two phases and to the number of pores. When a fluid is added, the difference in SLD between accessible pores and the matrix is reduced. If the SLD of the fluid and matrix are equal (the zero average contrast or contrast-matching condition), the scattering of all pores accessible to the fluid is eliminated and any remaining scattering is arising from the inaccessible pores (Melnichenko et al., 2012). The scattering intensity from the pores in the coal with the added fluid, $I(Q)_{PCF}$, expressed as a fraction of the scattering intensity from the pores in the coal in its absence, $I(Q)_{PC}$, is the fraction of closed pores ($F_c(Q)$) within a sample, which can also vary with Q .

$$F_c(Q) = I(Q)_{PCF}/I(Q)_{PC} \quad (2)$$

Fig. 1 shows a schematic of how SANS data are generated and processed.

Sakurovs et al. (2012) used SANS to identify fundamental, physical differences among the inertinite in bituminous coal from the Sydney

Basin, Australia, and those from North America, and Poland. They used deuterated methane (CD₄) as the penetrant gas and showed that the fraction of closed nanometre sized pores (defined as pores of $< 1 \mu\text{m}$ radius in this study) within the samples from Australia decreased linearly with increasing inertinite content. This was clearly observed in 8 nm radius pores; and extrapolating the results to a theoretical bituminous Australian coal containing 100% inertinite indicated that there would be no closed pores $> 8 \text{ nm}$ radius (all $> 8 \text{ nm}$ pores would be open). In contrast, in the examined Polish and U.S. coals, even in inertinite-rich specimens, $> 50\%$ of the 8 nm pores were closed (though this percentage decreased with increasing pore size). Liptinite levels were below 7% in all but one case and ash yields were below 10% in all but three Australian coals, thus the differences were not due to liptinite or mineral matter influences. The combined results (Sakurovs et al., 2012) suggest that there are measurable differences in the closed porosity in inertinite macerals in bituminous coals sourced from different coal basins, and these differences may explain why researchers report conflicting correlations between porosity, permeability, and gas sorption behavior with maceral composition (Stammore et al., 1997; Unsworth et al., 1989; Gürdal and Yalçın, 2001; Weishauptová and Sýkorová, 2011; Shan et al., 2015; Berbesi et al., 2009; Adeboye and Bustin, 2013; Kędzior and Jelonek, 2013; Keshavarz et al., 2017; Teng et al., 2017).

In this study, additional coal samples have been investigated to see if the closed porosity in inertinite from bituminous coals varies between other coal basins and if any patterns in these variations can be discerned.

2. Experimental

2.1. Coal samples

The samples examined were from Australia, U.S., South Africa, Canada, China, and Eastern Europe and were sourced from commercial coal mines. Due to confidentiality constraints, limited source information was supplied in most cases. Table 1 lists the analytical data for the coals used in this study and their source, where available. Note that Table 1 includes results for coals from the U.S. and Poland that were reported previously by Sakurovs et al. (2012) (denoted as ^a) and Grigore et al. (2012) (denoted as ^b). Coals examined in this study and their sources include: Qld coals–Queensland, Australia; ILL–Illawarra Coal Measures, New South Wales (NSW), Australia; and NSW–Sydney Basin, NSW, Australia. The South African coals were sourced from: TKD–Tshipise-Pafuri coal field; FOZ–Witbank coalfield; and DEN and OGS–Highveld coalfield (Okolo et al., 2015). The South African

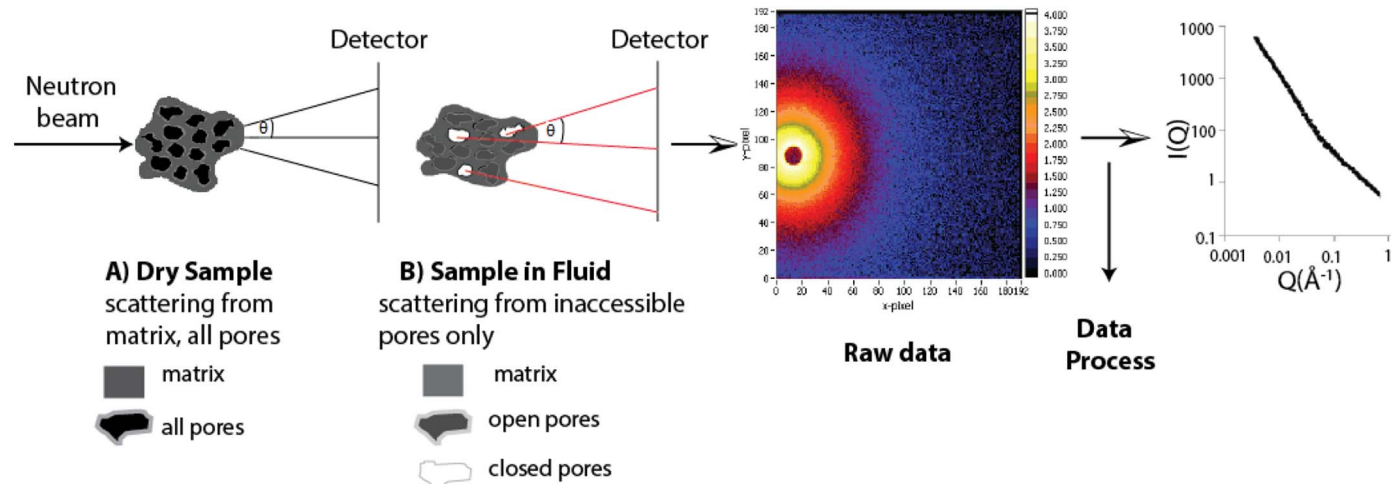


Fig. 1. Schematic of a SANS experiment where θ is the scattering angle, Q is the momentum of transfer, and $I(Q)$ is scattering intensity.

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