



## Multi scale characterization of coal structure for mass transport



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### ABSTRACT

The porous structure of coal is analyzed to understand the complexities involved in the process of coal bio-conversion to methane. Computed X-ray tomography (CT) imaging is used to obtain the three-dimensional porous microstructure of coal in a non destructive manner. Novel image processing techniques combined with stochastic analysis are used to analyze the 3D images. A mercury intrusion porosimetry study is done to analyze the pore sizes present in the coal structure. Finally, permeability measurements are done using a conventional core experiment. The porosimetry studies show that coal has a bimodal porous structure with a primary mode in the range of 5–10 nm and a secondary mode in the range of 2–10  $\mu\text{m}$ . The imaging analysis showed that almost half of the pores in 2–10  $\mu\text{m}$  range are percolating. However the porous volume accessible to microbes and nutrients is quite small due to low porosity. The permeability analysis at core scales shows that coal is an extremely low permeability medium due to its low porosity and small pore sizes. Overall, the study shows that one must improve the permeability and porosity of the coal matrix to make the coal bio-conversion process faster and economically viable.

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### 1. Introduction

Conversion of coal to methane provides a way to reduce the green house gas emissions and other environmental impacts related to coal burning such as SO<sub>x</sub> emissions (leading to acid rain), particulate matter emission, and heavy metal emission [1]. Coal can be converted into methane by either thermogenic or biogenic conversion. Thermogenic methanation happens in mature coal seams at large depths where the pressure and temperature are sufficiently high to enable cracking reactions to occur [2]. This process occurs naturally over geological timescales. Microbial bio-conversion, on the other hand occurs by the production of methane by coal consuming bacteria and associated archaea. This process can be manipulated to increase conversion of the organic components of coal to methane [3–10], and therefore it is the focus of current work. Fermentative bacteria colonies have been identified in several coal basins [3,10]. In general, while the details are still not well understood, these bacteria convert carbon within the amenable organic matter in the coal through a sequence of degradation steps including: (a) defragmentation of coal organic matter into organic acids, ketones, and aromatics; (b) anaerobic fermentation of these com-

ponents into hydrogen, carbon dioxide, and acetate; and (c) methanogenesis yielding methane [7,3]. This bio-conversion process however is slow and methane recovery at natural rates of conversion is not economically viable [5]. The energy recovery process is further complicated by the type and bio-activity of the indigenous microbes, the methods used to accelerate the bacteria metabolic activity, and mass transfer limitations associated with nutrient supply.

The bio-conversion of coal to methane needs to be primarily understood at two scales: (a) micro scale, where most of the bio-conversion takes place; and (b) millimeter scale, where the nutrients and reactants to the microbes are provided and methane is extracted. Coal has a hierarchical porous nature with cleats, macro pores, and micro pores as shown in Fig. 1 [11]. The cleats are in the size range of a few millimeters in length with apertures of the order of few hundred nanometers to a few microns in extent. The macro porous blocks between cleats are of length scale of the order of 10–100 mm. The macro-porous blocks themselves consist of macro-pores and micro-porous matrix, where the macro-pores are in the size range of 10–100  $\mu\text{m}$ . The micro-porous matrix between macro-pores consists of pores with sizes from a few nanometers to a few microns [11].

The primary interest thus far has been on the cleat structure shown in Fig. 1(a). Most of the microbes however reside in the

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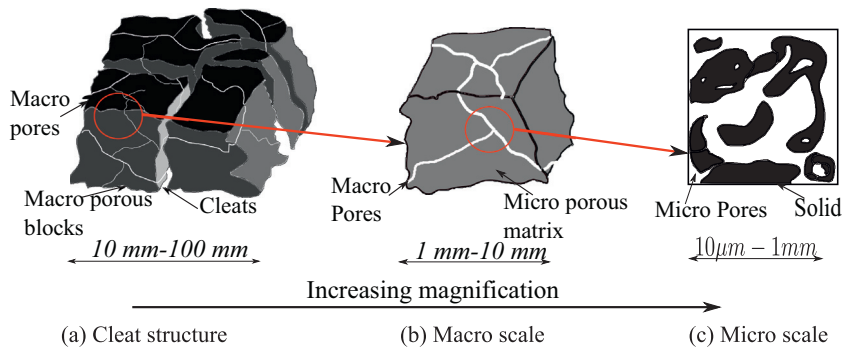


Fig. 1. Illustration of internal structure of coal at different length scales [11].

macro pores and micro-porous matrix shown in Fig. 1(b) and (c). The microbes with an average size of around  $1\ \mu\text{m}$ , can only access pores larger than  $0.2\ \mu\text{m}$ . Pore connectivity will play a critical role in the micro-porous matrix as the matrix has a size in the range of a few millimeters but the micro-pores are in the range of a few microns. Only the micro-pores which are connected to the macro-pores can receive the nutrients and microbes. Further, an understanding of interfacial area will be helpful to estimate microbe adsorption and population. A three-dimensional (3D) reconstruction of the coal geometrical structure would be extremely helpful to understand where microbes live and what limitations exist with respect to their growth and distribution within the media and with respect to mass transfer.

Several studies have been presented in the literature on estimating coal microstructure properties and coal microstructure visualization [12–14,11,15–33]. Small angle X-ray scattering has been used to estimate pore size distribution, porosity and interface area of coal [17,24,31]. This technique even though informative, does not give complete details about the internal structure of coal. Scanning electron microscopy (SEM) has been used to visualize the internal structure of coal, especially to understand its porosity and pore structure. SEM imaging has shown three types of porosity: fracture, phytal and micro porosity [11]. The micro pores appeared to be continuous for a long range and contributed significantly to overall permeability [13]. Due to the micro pores, coal has a large internal surface area (i.e. within the micro-porous blocks) which determines the adsorbed gas content [12].

X-ray computed tomography (CT) has recently shown good prospects for analyzing geological structures due to its ability to differentiate between different phases [14,16,34–37]. CT has been extensively used to study the effects of gas adsorption on the coal structure and also for characterizing transport characteristics [22,26,21,23]. However, very few of the studies using CT have attempted to visualize the entire coal structure using 3D representations [25,30,32]. Among these few studies, only a recent study by Wang et al. [32] has focused on visualizing the microstructure while the rest have focused on visualizing millimeter scale features such as cleats and fractures. Given the importance of the coal microstructure for the bio-conversion of methane, studies should also be focused on understanding the coal microstructure. Micro-CT has been used to obtain 3D reconstructions of several porous media [38–40]. Apart from qualitative visualization, micro-CT based 3D reconstructions of porous media can also be used to perform quantitative measurements. The 3D images provide estimates of pore sizes, pore connectivity and micro scale porosity. The accuracy of these estimates is however dependent on the image resolution, as structural features which are smaller than the image resolution will not be detected. The images can also be used to obtain stochastic correlation functions such as two point correlation function (TPCF), cluster function and pore size distribution (PSD). These stochastic correlation functions carry geometric

information which could be useful for porous media characterization. For example, the TPCF can give information about interface area, the cluster function is useful in determining pore connectivity and the PSD is useful to understand the pore sizes [41–43]. Detailed explanation about these correlation function and their measurement methods are given in the literature [41–43]. Image analysis methods have not been used to analyze coal structure thus far. The micro-scale 3D reconstructions combined with image processing can provide highly accurate information depending on image resolution in a short amount of time, making it an attractive method for porous media characterization.

Apart from the imaging techniques, the microstructure can also be analyzed using mercury intrusion porosimetry (MIP) or adsorption techniques. Using MIP and nuclear magnetic resonance, Zou et al. [44] determined the porosity of coal to be between 5.5% and 10.5% and the fractal dimension was found to be between 2.5 and 3. A bimodal PSD was observed in most of the samples with peaks in the ranges of 10–100 nm and 500 nm to  $5\ \mu\text{m}$ . Due to the high mercury pressure used in MIP, even the pores in nanometer range can be explored. However, mercury can not intrude the closed pores and therefore can not account for the non-connected void volume.

At the macro scale, both coal-bed methane production and in situ coal bio-conversion to methane rely on an understanding of the transport mechanisms of fluids (water and gas) and/or microbes through the coal reservoirs. An effective estimate of pore connectivity and efficiency of species transport within a porous block is its permeability. The pore and especially fracture permeability is a key factor that controls fluid flow in coal seams and access of microbial consortia to the coal matrix [3]. Coal permeability is also important for estimating the gas drainage rate or gas recovery ratio from coal seams [45,46]. A network of cleats and natural fractures contributes towards the permeability of coal. The permeability is typically anisotropic because of differences between face and butt cleat properties and in situ anisotropic stress conditions. The cleat aperture is sensitive to the effective stress, with increased effective stress acting to decrease the cleat aperture and thus net permeability [47,48]. In addition to laboratory measurements, permeability variation can be verified from field testing [49,50]. Based on field and laboratory experimental results, several permeability models have been developed for coal seams [51–56].

There is a lack of understanding of internal structure of coal, especially at micro-scale. CT imaging has immense potential for visualizing and understanding the internal structure of porous media. Imaging combined with stochastic characterization methods provide an easy and fast technique to estimate several porous media properties. This article presents one of the first CT based 3D visualizations of the coal structure at micrometer resolution. Furthermore, novel stochastic image processing techniques are used to estimate porous media properties. Mercury porosimetry

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