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# A microfluidic framework for studying relative permeability in coal

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ABSTRACT

A significant unconventional energy resource is methane gas stored in shallow coal beds, known as coal seam gas. The flow and transport of fluid in coal beds occur in a well-developed system of natural fractures, called cleats. In this study, we developed an efficient workflow for the fabrication of microfluidic chips based on X-ray micro-Computed Tomography (micro-CT) images of coal. A dry and wet micro-CT imaging technique is utilized to image coal cleats that would be otherwise non-resolvable. The obtained image of the cleat network is then etched into silicon wafers and used to fabricate poly dimethyl siloxane (PDMS) microfluidic devices. Fluid transport and displacement efficiency are visualized and quantified in real time by injecting water with a flow rate of 1 μl $min^{-1}$ into the fabricated cleat structure initially saturated with air. A microfluidic approach is used to measure the relative permeability of a realistic coal cleat system by monitoring the liquid recovery at recorded saturations after the breakthrough. Relative permeability curves show the cross and end point values for the water and gas flow, and predict a maximum relative permeability of 0.15 for the water phase. Understanding the relationship between coal cleat structure and the resulting relative permeability is crucial for the optimization of methane gas extraction and to reduce the environmental concerns of excess surface water production. Also, pore network modelling based on the Maximal Ball (MB) concept is applied to predict relative permeability curves numerically. Our experimental results are in good agreement with pore network modelling outcomes and provide consistent and smooth macro-scale relationships along with direct observation of the pore-scale physics. Therefore not only can the microfluidic approach be used as a validation tool for multiphase flow numerical models but it can also provide direct visualization of transport properties unique to coals. Overall, our developed system provides a better understanding of fluid flow behaviour in coal and presents novel relative permeability data for coal seam gas reservoirs under identical conditions and cleat sizes.



## 1. Introduction

Coal has been a viable source of energy for many centuries. Gas production from coal however is a recent and welcoming change (Hamawand et al., 2013). Decline in conventional resources, along with the environmental benefits of utilizing natural gas rather than coal has inclined the global interest to this alternative source of energy (Dabbous et al., 1974; Williams et al., 2012). Commercial extraction of methane from coal seam gas (CSG) reservoirs began in the United States in the 1970s. Exploration of CSG in Australia started in 1976, and the first commercial production occurred in 1996 in Queensland (Geoscience Australia, 2009). Since then, the CSG industry in Australia has developed rapidly, production was 2.8 billion $m^3$ for 2007 and 2008, and has risen to 6.2 billion $m^3$ in 2013 (Hamawand et al., 2013). Currently, CSG is an important and well-considered unconventional resource of energy in the United States, Australia, Canada, China, India and United Kingdom, with an estimation of around 256 $Tm^3$ of resources in these top-producing countries (Geoscience Australia, 2009; de Haas et al., 2013; Hamawand et al., 2013; Squarek and Dawson, 2006; Xu et al., 2014).

Methane gas is mostly stored in the coal matrix by adsorption, or as free gas in the larger pores or micro-fractures. As reservoir pore pressure reduces, the methane desorbs from the coal matrix surface and propagates towards the naturally developed micro-fractures, called 'cleats'. The cleats are originally saturated with water and thus as the gas is released a multiphase displacement process occurs (Harpalani and Schraufnagel, 1990). The rate of recovery from a coal seam reservoir is highly dependent on the porosity of the network, water saturation, and relative permeability of the system (Purl et al., 1991). Besides, a major environmental concern for CSG developments is surface water production, which is largely controlled by water phase relative permeability. The relative permeability of water in coal however is far from understood and available literature data varies significantly. Literature values for water phase coal relative permeability are displayed in

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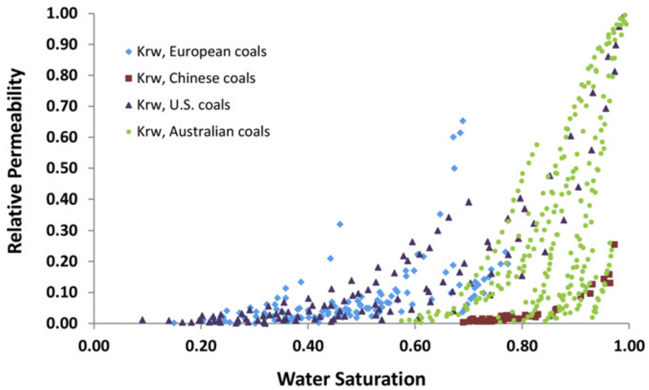


**Fig. 1.** Experimental data showing the water relative permeability for coals of various countries obtained from laboratory measurements and field production (Chen et al., 2014).

Fig. 1, which illustrates the wide range of relative permeability relationships that are possible. Typical techniques for collecting such data include laboratory core-flooding experiments and/or history matching to production data (Pan and Connell, 2012). Likewise a limited number of numerical and experimental studies have been reported for measuring and analysing absolute and relative permeability in coal (Ataie-Ashtiani et al., 2002; Blunt, 2000; Chen et al., 2013; Gash, 1991; Harpalani and Chen, 1995; Nguyen et al., 2013; Pan et al., 2010; Pant et al., 2012; Purl et al., 1991; Robertson and Christiansen, 2006; Wang et al., 2009; Zou et al., 2015).

Accurate measurement of relative permeability is of utmost importance for prediction of production rates (Blunt et al., 2013). It is well known that the relative permeability of a reservoir rock is highly dependent on the rocks underlying pore-scale structure, i.e. topology and geometry of the pore space. In a recent review (Zhang et al., 2014) it was highlighted that the relative permeability function for coal is far from understood. New methodologies for the measurement and evaluation of relative permeability results are required due to the unique structure of coal with cleat apertures ranging from 3 to 40 μm, small porosities in the range of 0.1% to 16%, and very low permeability of 0.1 to 100 md (Harpalani and Chen, 1997; Lamei Ramandi et al., 2015; Laubach et al., 1998; Meaney and Paterson, 1996; Radlinski et al., 2004). Additionally, there are the inherent difficulties in dealing with a brittle and deformable reservoir rock. In general, the relative permeability ($K_{ri}$) of phase $i$ is defined as $K_{ri} = K_i / K$, where $K_i$ is the effective permeability of phase $i$, and $K$ is the absolute permeability. The corresponding effective permeability of phase $i$ for a given saturation is calculated using the 2-phase extension of Darcy's Law (Dake, 1983)

$$K_i = \frac{q_i \mu_i L}{A \Delta P} \tag{1}$$

where $\mu$ is the viscosity of phase $i$, $L$ is system length, $A$ is cross sectional area normal to flow direction, $\Delta p$ is the pressure difference, and $q_i$ indicates the flow rate of phase $i$. This is a purely phenomenological approach in which the underlying physics are lumped into a saturation dependent relative permeability function, which provides no means to relate the underlying pore structure to the effective permeability. To select reservoirs with low water and high gas production rates, it is critical to understand the structural parameters in the coal cleat system that provide the wide range of relative permeability realizations (Fig. 1).

With conventional laboratory measurements however we are not able to quantitatively study the movement of fluids through the coal cleat system and correlate bulk measurements of relative permeability to the pore-scale topology and geometry of the cleat system.

Microfluidic studies are emerging in many biomedical and energy applications and can help to significantly reduce the cost of experiments while facilitating better process control and faster analysis in a dynamic fashion. In recent years, various types of microfluidic systems have been developed to study fluid flow behaviour inside a porous media for energy applications and/or oil recovery (Avraam and Payatakes, 1999; Joseph et al., 2013; Karadimitriou et al., 2012; Xu et al., 2014). For instance, Joseph et al. (2013) measured permeability in a microfluidic porous medium and studied the relationship between permeability and effective porosity. They designed a network, based on information obtained from sandstone rocks, and employed a computational network generation method called Delaunay triangulation. Xu et al. (2014) measured permeability in a periodic and random designed network. They found that the measured permeability in homogeneous pore networks is in reasonable agreement with the values estimated from the Carman-Kozeny equation. In another study, Avraam and Payatakes (1999) studied relative permeability for steady state flow in a network designed with a matrix of squares. None of these studies however attempted to make microfluidic chips with actual pore structures similar to features of real reservoir rock. More recent studies are devoted towards fabrication of microfluidic chips using geo-materials. For example, a new approach for fabrication of micro-channels inside geomaterials using laser etching and reactive ion etching (RIE) was developed and the rank and lithotype effect on coal wettability was explored (Mahoney et al., 2015a, 2015b). Porter et al. (2015) reported a direct visualization of flow behaviour inside natural fracture patterns. In another study, oil recovery in water flooding processes in periodic and random networks are compared, and the effect of pore size distribution and matrix geometry on displacement efficiency was explored (Xu et al., 2014). Gunda et al. (2011) fabricated a microfluidic chip with a network of reconstructed reservoir porous medium derived from scanning electron microscopy (SEM) images of actual samples. Delaunay triangulation was used, and the usual log distribution of pore sizes was adopted to extract a simplified network of pores and throats. They measured oil recovery in the micro-scale setup, which mimics the standard water flooding process. Thermal oil recovery has also been studied using microfluidic experiments (de Haas et al., 2013), and the effect of alkaline additive in oil recovery in two different grain size matrices was measured. Researchers also studied Bitumen oil production using $CO_2$ injection. They suggested microfluidic experiments to be easier, cheaper and safer to operate in comparison to conventional approaches (Fadaei et al., 2011; Lele et al., 2014). Song et al. (2014) used a real rock sample to fabricate a calcite microfluidic chip. They simulated an acid injection process, which is a secondary and tertiary oil recovery method, and visualized interactions between the matrix and acidified fluid.

Visualizing fluid flow in a 3D cleat system in real time is difficult, mainly due to the fast displacement and the complexity of the pore structure. A 2D microfluidic chip based on the cleat system representing the actual size and geometry of a real cleat structure, can easily display the multiphase flow behaviour and interactions within those complex structures. As can be seen in literature, 2D microfluidic studies in porous medium are being used and developed for several applications. However, multiphase flow in coal seams and its relative permeability have never been considered in these studies. Here we develop a novel workflow based on micro-computed tomography (micro-CT) imaging, Scanning Electron Microscopy (SEM) technology, and advanced image segmentation algorithms, to model exact coal cleat networks for the fabrication of a microfluidic device. We visualize flow in the coal cleat pattern, analyse contact angle and wettability effects, and simulate the gas recovery process. We measure relative permeability of a realistic coal structure network using microfluidic methodology, and validate our methodology by comparison of the results to literature values