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Maceral controls on porosity characteristics of lithotypes of Pennsylvanian high volatile bituminous coal: Example from the Illinois Basin

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ABSTRACT

Porosity characteristics of vitrain, clarain, durain, and fusain lithotypes of the Springfield Coal Member of the Petersburg Formation and the Danville and Hymera Coal Members of the Dugger Formation from the Illinois Basin were investigated with a special emphasis on the control of coal macerals on pore-size distribution. These Pennsylvanian coals are of high volatile bituminous rank and have vitrinite reflectance ranging from 0.51 to 0.60%. The lithotypes studied show decreasing values of Brunauer-Emmett-Teller surface area, and micro- and mesopore volumes from vitrain through clarain and durain to fusain. Within the mesopore size range, vitrain and clarain are dominated by pore-size widths of 4 to 10 nm, whereas durain and fusain have more volume for pores larger than 20 nm. In contrast to mesopores, micropore sizes are very similar for all lithotypes, averaging 1.37 to 1.39 nm. In addition to differences among lithotypes, there are significant differences in pore characteristics among the three coals studied, with largest surface areas and pore volumes documented for the Hymera, followed by the Danville and the Springfield. A strong relationship exists between surface area, mesoporosity, and microporosity and maceral composition, with vitrinite having a very strong positive correlation, liptinite having a weak positive correlation, and inertinite having a strong negative correlation. Negative correlations of total porosity with vitrinite and liptinite and positive correlations with inertinite suggest that among the maceral groups, pores in inertinite contribute most to the total porosity. The Fourier transform infrared spectrometry technique demonstrates that fusains from the three coals studied have higher aromaticity and a higher degree of aromatic ring condensation and lower hydrocarbon potential than the other lithotypes, whereas chemical differences between vitrain, clarain, and durain are less distinct. In addition, there is a relationship between aromaticity of the lithotypes and surface area and mesopore and micropore volumes.

1. Introduction

Coal lithotypes, heterogeneous and complex mixtures of organic matter and minerals, are megascopically recognizable bands of different maceral compositions. Lithotype variations in coal seams reflect changes in organic input and depositional conditions in the mire, modified later by diagenesis. Classification of brown coal lithotypes is based dominantly on color, texture, and degree of gelification (Hagemann and Hollerbach, 1980; Styan and Bustin, 1983), and for coals of sub-bituminous rank and higher, a proportion of bright versus dull bands is the main distinguishing criterion, although texture, luster, and mineral matter content are also important identifying parameters. In bituminous coals, lithotypes are classified into vitrain (bright coal), bright clarain (banded bright coal), clarain (banded coal), dull clarain (banded dull coal), durain (dull coal), and fusain (fibrous coal) (Stopes, 1919; Diessel, 1992). The classification system uses a minimum lithotype thickness of 3 or 5 mm; consequently, thinner bands become part of a thicker lithotype unit.

It has been well documented that coal lithotypes possess different physical and chemical properties, a reflection of their different depositional and postdepositional histories. Vitrain, formed by coalification of peat-forming woody material, is composed of homogeneous bright bands that are rich in vitrinite, especially collotelinite (Ting, 1982). Clarain originates from coalified plant fragments and detrital plant material and includes thin bright and dull bands. Durain, developed from detrital material, usually reflects more open-water conditions with increased mineral matter influx. Megascopically, durain is grayish to brownish black and is usually the hardest and most resistant lithotype. Fusain is a soft component that blackens hands upon touch; it has a fire origin (e.g., Scott and Glasspool, 2007) or can form as a result of degradation of peat-forming material (Stach et al., 1982). Usually it forms thin layers between other lithotypes (Marchioni and Kalkreuth, 1991; Mukhopadhyay and Hatcher, 1993).

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One of the parameters differentiating coal lithotypes is their maceral composition (Crelling et al., 1988; Marchioni and Kalkreuth, 1991; Lamberson et al., 1991; Lamberson and Bustin, 1993). Typically, there is a progressive increase of inertinite and a decrease in vitrinite content from vitrain to fusain (Crelling, 1982; Lamberson et al., 1991; Mastalerz et al., 2008a, 2010). This difference in maceral composition also results in differing chemical properties of lithotypes. For example, fusain is more aromatic than other lithotypes, whereas the chemistry of vitrain, clarain, and durain can vary depending on the proportion of liptinite, vitrinite, and inertinite macerals (e.g., Cao et al., 2011). Several studies documented significant differences in porosity characteristics among lithotypes. For coals of high volatile bituminous rank, fusains typically have the smallest micro- and mesopore volumes and vitrains have the largest (Clarkson and Bustin, 1996, 1999; Mastalerz et al., 2008a). The different porosity characteristics of lithotypes result in different gas (methane and CO_2) adsorption capacities, with vitrain usually having the largest adsorption capacity and fusain the smallest (Lamberson and Bustin, 1993; Clarkson, 1994; Mastalerz et al., 2008a, 2012a). It has also been reported that fusains have the fastest gas desorption rates, while vitrains have the slowest desorption rates (Crosdale et al., 1998; Karacan and Mitchell, 2003; Mastalerz et al., 2012a). Vitrain, exhibiting more fractures (cleats) than other lithotypes (Ting, 1982), is most permeable; clarain has medium permeability, which increases with increasing vitrinite content, and durain and fusain usually have low permeability because of high mineral and inertinite content, respectively (Clarkson and Bustin, 1997). Permeability of durain and fusain may, however, increase with mineral removal due to an increase in cleat density and connectivity of cleat system (Ramandi et al., 2016). These lithotype-derived differences in adsorption capacity and permeability are important to the understanding of coalbed methane storage and producibility and, therefore, for CO_2 injection into coal beds (Karacan and Mitchell, 2003; Mastalerz et al., 2008a).

Despite the abundant information on coal pore characteristics and lithotypes, much less is known about how maceral composition controls porosity characteristics in lithotypes of specific coal ranks. This paper investigates porosity characteristics in three Pennsylvanian high volatile bituminous coals from the Illinois Basin: the Danville and Hymera Coal Members of the Dugger Formation and the Springfield Coal Member of the Petersburg Formation (Table 1). All three coals come from one location, Bear Run Mine in Sullivan County, Indiana. The same coalification history of these three coals eliminates the difference in coal rank as a factor controlling porosity characteristics and provides an excellent opportunity to look at maceral influence in isorank coals. Our main objective is to document the range of variations in porosity and maceral characteristics of vitrain, clarain, durain, and fusain and to understand how and why these lithotypes can differ between the isorank coal seams. To address these issues, petrographic composition, total porosity, meso- and micropore characteristics, and functional group distribution were documented and compared among the three coals studied.

Table 1
Stratigraphic chart of the Pennsylvanian System in the Illinois Basin, showing major coals. (Modified from Mastalerz and Harper, 1998). The coal beds studied are marked in italics.

Pennsylvanian	Upper	McLeansboro Gp.	Mattoon Fm.	
			Bond Fm.	
			Patoka Fm.	
	Middle		Shelburn Fm.	
		Carbondale Gp.	Dugger Fm.	*Danville* *Hymera* Herrin
			Petersburg Fm.	*Springfield* Houchin Creek
			Linton Fm.	Survant Colchester
		Raccoon Creek Gp.	Staunton Fm.	Seelyville
			Brazil Fm.	Minshall/Buffaloville Upper Block Lower Block
	Lower		Mansfield Fm.	Shady Lane Mariah Hill Blue Creek Pinnick St. Meinrad

2. Methodology

Three coal beds, the Springfield, Danville, and Hymera Coal Members from Bear Run Mine in Indiana, were selected for this study. Fresh coal blocks were transported to the laboratory where four lithotypes—vitrain, clarain, durain, and fusain—were hand-picked for further analysis. Well established criteria (e.g., Hower et al., 1990) such as texture, luster, and the proportion of bright versus dull bands were used to differentiate lithotypes. To obtain sufficient material for each lithotype, we combined multiple layers of the same lithotype from a large coal seam sample. This way, each lithotype sample represents truly average composition of each lithotype in this location.

2.1. Petrographic analysis

For petrographic analysis, coal preparation techniques followed standard organic petrography procedures (ICCP, 1963). For vitrinite reflectance analysis (R_o random), 75 measurements were averaged for each lithotype, while for maceral analysis, 500 points were counted. The reflectance of fusinite in fusain was measured in addition to vitrinite reflectance. A Leica DM 2500P reflected-light microscope linked to a TIDAS PMT IV photometric system was used for these analyses.

2.2. Porosimetry

Pore classification into micropores (pores having a diameter < 2 nm), mesopores (2–50 nm diameter), and macropores (diameter > 50 nm) follows that of the International Union of Pure and Applied Chemistry (Orr, 1977). Micro- and mesopore characteristics were evaluated based on low-pressure gas adsorption using a Micromeritics ASAP-2020 apparatus. Samples of ~60 mesh (~250 μm) in size and weighing 1.5 to 2 g were analyzed with nitrogen (N_2) adsorption to evaluate surface area and mesopore characteristics, and carbon dioxide (CO_2) adsorption to analyze micropore characteristics. Before the analysis, lithotype samples were degassed at about 110 °C in a vacuum for about 14 h to remove adsorbed moisture and volatile matter. For N_2 adsorption, the samples were exposed to liquid nitrogen at the temperature of 77.35 K. Both adsorption and desorption analyses were conducted, of which the adsorption branch was used to determine mesopore surface areas and volumes. The surface area, mesopore volume, and average pore size were calculated according to the adsorption theories of Brunauer-Emmett-Teller (BET) and Barrett-Joyner-Halenda (BJH). CO_2 adsorption occurred at the temperature of 273.1 K. Micropore surface area and monolayer capacity were calculated using Dubinin-Radushkevich (D-R), and micropore volume using Dubinin-Astakhov (D-A) methods (Webb and Orr, 1997; Clarkson and Bustin, 1999; Mastalerz et al., 2012b, 2013). The precision of the measurements is as follows: BET surface area $\pm$ 0.04 m^2/g, BJH mesopore volume $\pm$ 0.0002 cm^3/g, D-R micropore surface area $-$ 0.34 m^2/g, and D-A micropore volume $-$ 0.0006 cm^3/g.

Total porosity was determined on 7 mesh (2.83 mm) splits by subtracting grain volume determined by a ULTRAPYC 1200e helium pycnometer from the sample volume determined by GeoPyc 1360 and dividing the difference by the sample volume. Each analysis was done in duplicate to obtain the average values expressed in percent.