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Petrophysical characterization of Chinese coal cores with heat treatment by nuclear magnetic resonance

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HIGHLIGHTS

- NMR T_2 spectrum of water within the pore structure was influenced by coal rank.
- Heat has more significant effect on T_2 of water of LVB than that of HVB or anthracite coal.
- Porosity by NMR increases with treatment temperature, but to a different extent for each rank.

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ABSTRACT

For the past decades the nuclear magnetic resonance (NMR) technology has gained acceptance as a petrophysical tool for evaluating reservoir properties. Comprehensive reservoir evaluation requires determination of irreducible fluids, movable fluids and permeability. Although the NMR petrophysical properties of coals have been studied for decades, the impact of heat on these properties (pore size distribution, pore structures, porosity and permeability) has not yet been systematically investigated. However, these are key properties for coalbed methane (CBM) generation and production. Therefore, they may have significant implications for the effects of heat from geothermal dynamics and magma intrusion on CBM concentration and transport in coals with different ranks. Thus, NMR experiments for samples treated at different temperatures (from 25 °C to 375 °C) were designed to study the variation of petrophysical properties of three Chinese coal cores with different ranks. Results show that NMR transverse relaxation (T_2) distributions of the water saturated cores strongly relate to the coal pore structure and coal rank. Furthermore, based on T_2 cutoff time method, five models for calculating the permeability of coals to water were evaluated. The results show that the Schlumberger Doll Research (SDR) model and its improved model provided the best estimation among the five models because these two models are generally able to represent the matrix permeability of the coal, based on the comparison between the results from measured gas permeability and NMR permeability models. Further calculations indicate that the porosity of all three different rank coals have an increasing trend with exposure to temperature, but with different increments for these coals. The low-volatile bituminous coal has the largest increment (9.44%), which is an improvement of more than 200% from its original porosity (4.02%). While the permeability has no similar trend for these three rank coals after heat treatment due to the strong heterogeneity of pore structure in coals. The results may suggest complex microfractures widths change for forming/closing at different heating stages.

1. Introduction

Many coal-bearing basins or coalfields in the world have gone through contact metamorphism or thermal maturation directly or indirectly related to geothermal dynamics and magma intrusions, such as the San Juan, Raton, and Illinois basins in the USA [1,2], the Sydney and Gunnedah basins in Australia [3,4], the Highveld coalfield in South Africa [5], and the Qinshui and Fushun basins [6,7] and Huaibei coalfield [8,9] in China. The geothermal dynamics may have significant effects on the generation and concentration of coalbed methane (CBM), the petrophysical properties of the in situ coal reservoirs and even CBM production [10]. However, studies on the effects of heat from geothermal dynamics or magma intrusion on petrophysical properties of CBM reservoirs are still insufficient. Storage capacity and transport of coal reservoir fluids through coal seam are important properties to characterize coal reservoirs [11]. Coal reservoir fluids, including water

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and gas are contained in the porous coal seams, in which the liquid and/or gas and the solid materials of the coal matrix are mutually affecting each other from atomic to field scales. The storage and transport properties are determined by the petrophysical properties of coals, which are denoted as the porosity, as well as by the pore size distribution, connectivity and permeability.

Many methods have been introduced to study petrophysical properties of the coal, such as mercury intrusion, gas adsorption/desorption, scanning electron microscopy and small angle scattering techniques [12–16]. All these laboratory-based measurements are well established. However, these methods have limitations on sample preparation or data process and quality [17], due to the changes of the saturation state of the sample and/or the disintegration of the porous matrix. Some of these methods are destructive to the complex fluid/matrix system and to the interaction between the pore fluid and pore walls, influencing the measured petrophysical properties. Thus, it is important to use a nondestructive, easy and less time-consuming method to evaluate the petrophysical properties of the coal samples, and to better understand the properties of the CBM reservoir. With this respect, the nondestructive methods of nuclear magnetic resonance (NMR) and X-ray computed tomography (CT) were introduced to study the petrophysical properties [18–21]. These two methods have become indispensable tools for characterizing petrophysical properties of CBM reservoir such as pore size distribution, pore connectivity, porosity and permeability. NMR measurement involves using an external magnetic field to align hydrogen magnetic moments, and a dipole moment in the hydrogenous fluid component of the sample. The amplitude of the dipole moment is proportional to the number of hydrogen atoms present and thus, is a measure of the pore volume filled with water [22]. For fluid in a single, isolated pore and for the condition known as the fast diffusion limit, the magnetization, $m(t)$, decays exponentially with time:

$$m(t) = m_0 \exp(-t/T_2) \tag{1}$$

where $m(t)$ and m_0 represent the magnetization at time t and 0, respectively, t is time, T_2 is the transverse relaxation time. Previous research [23] has shown that the time constant, T_2, is related to the pore surface to volume ratio, $(S/V)_{\text{pore}}$, by:

$$T_2^{-1} = \rho \left(\frac{S}{V}\right)_{\text{pore}} \tag{2}$$

where ρ is a constant representing the surface relaxivity, S/V is the surface to volume ratio that relates to the pore size. Thus the pore size is corresponding to the certain relaxation time [23].

All nuclear magnetic resonance studies on water-saturated coals to date have been performed without heat treatment. However, petrophysical properties of coals usually depend very strongly on temperature history. In the present study, the proton (^{1}H) NMR signal of the hydrogen of the water saturated in coal plugs of 2.5 cm in diameter is recorded using low-field NMR spectrometers. Different rank coals were evaluated with the aim to cover a wide range of petrophysical properties. First, we characterize the pore structure of coal samples by measuring and analyzing T_2 values of water in the water saturated cores. Correlated transverse relaxation time distributions determine the extent of pore size distribution by T_2 variation. Second, NMR studies of the water re-saturated pores were used to determine the variation of petrophysical properties for Chinese coals after treatment at elevated temperatures (from 25 °C to 375 °C), which provide unique insights into the effects of heat from geothermal dynamics and magma intrusion. Finally, an optimum NMR permeability model for low permeability coal was chosen to better describe the permeability for these samples without confining stress.

2. Coal samples and experiments

2.1. Samples selection and coal analyses

Three coal samples were collected from two underground mines in the Qinshui basin, North China and one underground mine of Jixi coalfield, Northeast China. The sampling locations are shown in Table 1. The samples were collected from the depth between 500 and 1200 m using the channel method, then carefully packed and quickly transported to the laboratory for experiments. Samples including high-volatile bituminous B (LH18#), low-volatile bituminous (HSXG15#-2) and anthracite (WTP15#-2) were selected for petrophysical characteristics analysis after treatment at elevated temperatures. Two or more horizontal cylindrical cores (2.5 cm in diameter) drilled parallel to the bedding plane from each coal sample were prepared. Plugs cut from the end of the cores were used for experiments. They were divided into two sets: one set of core plugs for water-saturated porosity and air permeability and another set for NMR measurements and CT scan analyses. The air permeability was determined using a bubble flowmeter by flowing air through the core sample, which was detailedly explained in our previous work [7].

2.2. CT and NMR analyses

CT scan experiment was performed using the ACTIS-250/320PK/225FFI (US) industrial CT system. The X-ray source is a 225 kV FeinFocus focal spot, which allows for resolution of 10 μm for an object of 4.8 mm. The cylindrical core sample was placed perpendicular to the sample couch (orientated in slice plane) and was aligned in the center of the scanner's field of view. During operation, the spatial resolution was approximately 50 μm, with which most macropores and all fractures can be distinguished. The white high CT attenuation is mineral matter, and the dark low CT attenuation is macropores/fractures. Others are the coal matrix material.

After the water-saturated porosity and air permeability measurements, the NMR experiments were designed as follows: first, all the selected samples were dried with dry air flow and then they were vacuumed for 48 h till no weight change at room temperature. After that they were saturated in simulated stratum water (1.5% NaCl solution) for another 24 h. The water-saturated weights for the samples were measured again. Thus, the water-saturated porosity can be calculated. The 100% water-saturated (S_w) cores were first used to obtain one T_2 spectrum distribution. Then, the samples were progressively centrifuged to reach a perfect irreducible water (S_{ir}) condition (centrifuge pressure and time were 200 Psi and 1.5 h, respectively). The samples with irreducible water conditions were tested to obtain another T_2 spectrum distribution. When these two steps were finished, a vacuumed muffle furnace (F47920-33-80) manufactured by Thermo Scientific Company of USA was used to heat the samples under elevated temperatures (25 °C, 75 °C, 225 °C and 375 °C). Before heating the samples at the pre-set temperatures, the samples were pre-heated at 50 °C for 30 min. Then the samples were heated at the target temperatures for 3 h. After they were cooled down, they were re-saturated with water to repeat the NMR measurements and then centrifuged for more NMR measurements. The NMR measurements used a RecCore-04 instrument with a resonance frequency of 2.38 MHz. The main NMR measurement parameter set included the time interval of echoes (TE) of 0.6 ms, the waiting time (TW) of 5 s, the echo numbers of 2048, the scanning numbers of 64, and the environment temperature of 25 °C. It should be noted that there was no confining stress on the samples during air permeability, CT and NMR measurements.