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Changes in the petrophysical properties of coal subjected to liquid nitrogen freeze-thaw – A nuclear magnetic resonance investigation

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HIGHLIGHTS

- NMR was used to explore the petrophysical properties of frozen-thawed coal.
- Freeze-thaw cycles had the most significant effect on petrophysical properties.
- The modification on different coal ranks was affected by coal's initial porosity.

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ABSTRACT

Liquid nitrogen (LN_2), a non-aqueous medium, has attracted attention in recent years as a fluid for fracturing in the petroleum/energy industry. This study proposes a freeze-thaw method using LN_2 to improve coal permeability for the production of coal bed methane. Experiments were conducted using nuclear magnetic resonance (NMR) to explore the physical properties of frozen-thawed coal. The coal samples were subjected to different LN_2 freezing times and to freeze-thaw cycles and coals of different rank and with different moisture contents were tested. Changes in these four freeze-thaw variables changed the petrophysical properties of the frozen-thawed coal samples; the pore structure, porosity, and permeability of the coals were modified. Of these variables, the number of freeze-thaw cycles had the most substantial effect on modifying the coal's petrophysical properties. The degree of modification on the coals of different rank was affected by the coal's initial porosity. In general, lignites were modified the most, anthracite coal was modified less, and bituminous coal was modified the least. The study analyzed three of the classic NMR transforms for determining permeability and found that the Schlumberger-Doll Research (SDR) model matched the measured gas permeabilities most consistently. Based on this SDR permeability model, equations suitable for predicting the permeability of frozen-thawed low-rank coals were derived. In addition, results from scanning electron microscope studies showed that a fracture network with fracture widths of as much as 32.3 μm was formed in the coal after 30 freeze-thaw cycles. Additionally, micron-size particles falling from the coal surface gradually increased as the number of freeze-thaw cycles increased, indicating that freeze-thaw using LN_2 materially modified the physical properties of the coal.

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1. Introduction

Coal bed methane (CBM) is an unconventional energy resource with enormous reserves around the world [1–3]. Draining CBM before coal mining can not only provide energy but can also have a positive influence on greenhouse gas emission [4,5]. In addition, CBM drainage can help to prevent gas disasters in coal mines as well as facilitate economic coal mining [6,7].

Coal has a porous reticular structure and most CBM is adsorbed onto the micropores. Owing to coal's low permeability, extracting CBM needs to augment the following processes. The CBM must be desorbed from nanometer scale pores, diffuse between nanometer scale pores, migrate through micrometer scale flow channels, and finally be extracted [8]. Many methods, such as hydraulic fracturing and water jet slotting, have been used to improve the permeability of coal seams [9,10]. However, the hydraulic measures

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can cause water pollution and reservoir damage and these methods cannot guarantee sustainable and large-scale increases in permeability [11–13]. Further studies on water-free fracturing technologies for CBM extraction have developed techniques using liquid nitrogen (LN_2) and liquid carbon dioxide as fracturing fluids [14–18].

LN_2, is an environmentally friendly low-temperature medium; its temperature is −196 °C at atmospheric pressure (0.1 MPa). One cubic meter of LN_2 vaporizes into 696 m^3 of nitrogen gas at 21 °C and the latent heat of vaporization is 5.56 kJ/mol. Because all coal cleats contain water, water in the cleats rapidly freezes as LN_2 is vaporized and the H_2O expands when the water in the coal turns into ice. In addition, because of the expansion caused by the water-ice phase change, a frost heaving force of 207 MPa is produced in fissures in the coal [19]. Based on the properties of LN_2 and the behavior of coal upon freezing, Zhai et al. [20] proposed a freeze-thaw cycling method using LN_2 to fracture coal and improve CBM production. In nature, in cold climates, natural freeze-thaw cycles caused by changes in the weather damage buildings and decrease the stability of rock masses. In cold areas, numerous studies have been carried out on freeze-thaw variables and the failure of rocks and soil masses [21–24]. However, studies on coal frozen using LN_2 are rare.

Because it is an innovative method, fracturing of coal reservoirs using LN_2 has not been intensively studied and is not widely used. However, in the 1990s, this fracturing method was tried in the San Juan CBM field, New Mexico, USA [25]. The results showed that LN_2 can efficiently increase the permeability of coal seams. Grundmann et al. [26] fractured shale by using LN_2 and improved gas production rate by 8% compared with the production obtained using traditional fracturing methods. Cai et al. [15] studied the changes in coal pores and the coal's mechanical strength after single LN_2 freezing events. Li et al. [17] proposed a fracturing technique suitable for shale reservoirs based on the vaporization of LN_2. Coetzee et al. [27] found that using LN_2 as a fracturing fluid can effectively promote the extension of fractures.

At present, no in-depth studies on how the physical properties of coal are modified by freeze-thaw variables have been published. The study presented here systematically analyzes the changes in the properties of coal before and after freeze-thaw treatment using nuclear magnetic resonance (NMR) spectroscopy. Four aspects of the process are considered: LN_2 freeze-thaw time, number of freeze-thaw cycles, coal moisture content, and coal rank. In this way, this study attempts to determine how these four freeze-thaw variables affect several coal properties such as pore structure and permeability. The study also aims to provide data to support programs to fracture coal reservoirs using LN_2 freeze-thaw cycling.

2. Experimental

2.1. Nuclear magnetic resonance principles and theory

Petrophysical properties of coal involve the pore size, distribution, and connectivity of pores and fissures [28]. The methods for determining pore size can be divided into qualitative and quantitative methods. The former methods include those using different kinds of microscopes (optical, scanning electron, and transmission electron microscopes), whereas the latter include the mercury intrusion method (MIP), nitrogen and carbon dioxide adsorption methods, small angle X-ray scattering/small angle neutron scattering (SAXS/SANS) methods, and micro-X-ray computed tomography. Some testing methods for pore size have limitations such as low testing efficiency, limited testing ranges, and damage to the original pore structure [29]. Fig. 1 lists the testing ranges for some pore size measurement methods including MIP (100 nm–100 μm), nitrogen adsorption (2–100 nm), carbon dioxide adsorption (0.4–2 nm), SAXS/SANS (1–100 nm), and NMR (0.1–100,000 nm) [29].

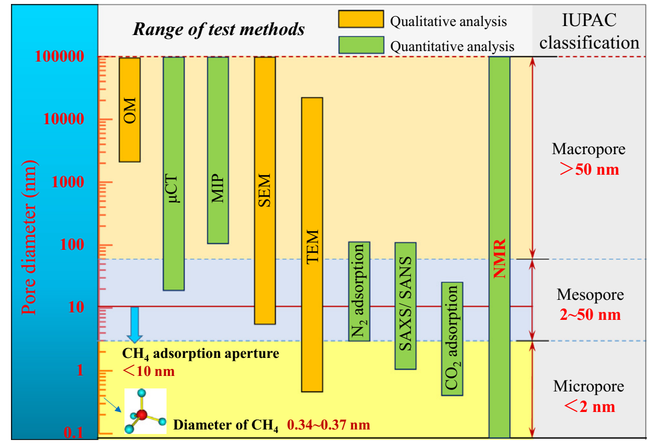

Fig. 1. Comparisons of the measurable ranges of testing methods for coal pore sizes (nm). Modified from Fu et al. [29].

The diameters of methane molecules ranges from 0.34 to 0.37 nm and most methane molecules in coal are adsorbed in pores with diameters of less than 10 nm. The ranges for the methods in Fig. 1 show that NMR can test the largest range of pore sizes and NMR is a nondestructive and highly efficient test method [30]. NMR spectroscopy can accurately characterize methane adsorption and flow in coal.

By employing NMR spectroscopy to test Carr-Purcell-Meiboom-Gill (CPMG) pulse sequences on rock cores completely saturated with water, the attenuated signals of spin-echo strings are obtained. The attenuated signals are the superposition of hydraulic signals from pores with different sizes. By fitting a curve to the signals, a distribution curve of transverse relaxation time, T_2, is obtained [31]. Thus, the distribution and connection of pores in the coal samples and the physical parameters of the coal can be acquired. The relationship between the NMR transverse relaxation time T_2 and the pore size can be expressed by Eq. (1) [32–34]:

$$\frac{1}{T_2} = \rho \times \frac{S}{V} = F_S \times \frac{\rho}{r} \tag{1}$$

where T_2 is the transverse relaxation time (ms), ρ is a factor for the intensity of transverse surface relaxation; S and V are the surface area (cm^2) and volume (cm^3) of the pores. The F_s factor is a shape factor (the F_s of spherical pores, cylindrical pores, and fractures are 3, 2, and 1, respectively) and r is the pore size.

The amplitudes and peak areas of the T_2 distribution curve mirror the number of pores in the coal and the continuity of the curve reflects the connectivity of the pores [35]. A factor called $T_{2cutoff}$ can be used to distinguish between open and closed pores in coal [30].

2.2. Sample preparation

For the study, lignite and bituminous coal samples were collected from coalfields in the Inner Mongolia autonomous region, China, the Shengli and Jungar coalfields, respectively. Anthracite samples were collected from the Xinbei coalfield in Gansu province, China. Lignite, bituminous and anthracite respectively represent low-rank, middle-rank and high-rank coal. In which, the lignite coal samples collected from Shengli coalfield were used to analyze the influences of freeze-thaw variables, including freezing time, freeze cycles and coal moisture content. All coal samples were fresh and collected from mines. The same variables were studied using the samples taken from the same block of coal. The

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