



Accelerating methane desorption in lump anthracite modified by electrochemical treatment



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ABSTRACT

The slow rate of methane desorption in coal can seriously affect the effectiveness of coalbed methane production. An electrochemical method was proposed to modify coal and accelerate methane desorption. The desorption kinetics of methane from lump anthracite, before and after modification, were studied at four adsorption equilibrium pressures using our own manufactured devices, electrochemical modification equipment and lump coal adsorption–desorption apparatus. Based on mercury intrusion tests and SEM-EDS analyses, the possible modification process of accelerating methane desorption in lump anthracite was further analyzed. The results showed that (1) the average desorption rate of methane from anthracite after modification, at the low end of the pressure ranges from 1 to 4 MPa, increased by 68% over the period of reaching desorption equilibration; (2) with increasing adsorption equilibrium pressure, the methane desorption time constant for lump anthracite changed according to negative exponential rules; and (3) in the anodic zone, the mineral-filled fractures of coal samples were unblocked by electrolytic reactions. In the cathodic zone, electrophoresis and electro-osmosis caused charged particles (clay minerals and coal particles) and the electrolyte in the coal matrix and cleats to move, which led to the initiation, development and connection of pores or fractures. These changes increased the number of Darcy flow channels and shortened gas diffusion distance and desorption time.

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1. Introduction

Methane is primarily adsorbed to the internal coal surface, and its capacity is reasonably correlated with coal surface area (Levy et al., 1997). Anthracite has a strongly developed inner surface and contains potentially high levels of adsorbed methane. However, because of the low permeability of anthracite, methane desorption rates are very slow. Thus, the effectiveness of coalbed gas production can be seriously affected, which restricts commercial coalbed methane development (Alexeev et al., 2007; Bodden and Ehrlich, 1998; Harris et al., 1996). Therefore, there is an urgent need to find a method for effectively accelerating methane desorption from low permeability coal reservoirs.

Recently a number of approaches have been considered for accelerating methane desorption. Based on the effects of CO₂–CH₄ competitive sorption and counter-diffusion, Shi et al. (2008) reported a core flooding test where supercritical CO₂ gas was injected into a large coal core to promote methane desorption and to improve the efficiency of coalbed methane recovery. Many studies (Faiz et al., 2007; He and Zhang, 2000; Jiang et al., 2010) have found that the methane desorption rate can be considerably enhanced due to activated diffusion under the

action of a physical field, such as a temperature field, sound field, electromagnetic field. However, these methods significantly change the gas adsorptivity or diffusivity of coals but do not solve the key problem that influences the rate of methane desorption: low permeability. In addition, the stress-relief methods, such as hydro-fracturing (Colmenares and Zoback, 2007), microwave-induced fracturing (Kumar et al., 2011) and blasting-induced fracturing (Guo et al., 2008), were developed to increase coal seam permeability and significantly accelerate the desorption rate. However, the penetration depth and overall impact of these approaches are limited. Therefore, an exploratory study on accelerating methane desorption in coal modified by electrochemical treatment was proposed.

At present, the electrochemical method has been applied to improving coal quality and increasing coal efficiency, which is at an early stage of exploration. Lalvani et al. (1983) reported that electrochemical coal cleaning can remove practically all of the inorganic sulfur and a portion of the organic sulfur with very little coal loss. Fuerstenau et al. (1988) found that electrical charges are developed on its surface when coal is immersed in a liquid environment, indicating that coal has electrokinetic behavior. They also investigated the surface charge's generation mechanism and influential factors using electrokinetic measurements. Lockhart (1982) noted that the electro-osmosis phenomenon that water can be moved through a charged capillary in coal by the action of an electric field will occur, and founded that two coal flotation

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concentrates were dewatered by electro-osmosis in small-scale laboratory tests to 84 and 88 wt.% solids. There is enough evidence to warrant consideration for accelerating methane desorption by electrochemical method. In this paper, we carried out a series of methane desorption kinetic experiments with lump anthracite at four adsorption equilibrium pressures, studied desorption time of methane from coal samples before and after the electrochemical modification, and discussed the possible modification process coupled with mercury intrusion tests and SEM-EDS analysis.

2. Experimental

2.1. Coal samples

The experiments were conducted on the anthracite with a vitrinite maceral content of 86.3% and inertinite content of 13.7% (vitrinite reflectance of 2.86%). The large coal blocks were directly collected from the No.15 coal seam in the Southeast Qinshui Basin from Sihe coal mine (Shanxi province, China). Cleats in coal are mostly filled with calcite and clay. The ash yield (weight %, dry-basis) of coal is 12.83%, and its chemical components using X-ray fluorescence spectrometry (XRF) are mainly CaO 26.27%, SiO₂ 24.83%, Al₂O₃ 22.14%, Fe₂O₃ 11.87% and SO₃ 10.81%. The coal samples from the same location were processed into six cylindrical samples with a diameter of 130 mm and a length of 150 mm and two small cubes of 6 mm to a side. The cylindrical samples were randomly divided into two groups, one for desorption kinetic experiments on unmodified samples and one for experiments on modified samples. Before testing, the coal samples were dried in an oven at 378–383 K until a constant weight was achieved, at which time the samples were weighed and recorded. The cube sample surfaces were polished and sputter-coated with gold–palladium for studying in scanning electron mode.

2.2. Electrochemical modification

The coal samples were modified using the electrochemical modification equipment shown schematically in Fig. 1, which mainly consists of an electrolyzer, electrode, coal samples, electrolyte, direct current (DC) power supply, current meter and wire. The electrode is a square graphite plate that is 100 mm long and 5 mm thick. The three cylindrical samples were side by side in the middle of electrolyzer, and the two cube samples were placed near the anode and cathode, respectively. The electrolyte is Na₂SO₄ solution with a concentration of 0.05 mol · L⁻¹. The electric potential gradient is 4 V · cm⁻¹. Wang et al. (2011) found that the electrochemical method can change the tensile strength and pore structure of mudstone and set the modified time to 120 h. The action time in this study is also 120 h. After the modification, the coal samples were removed and their surfaces were cleaned with distilled water.

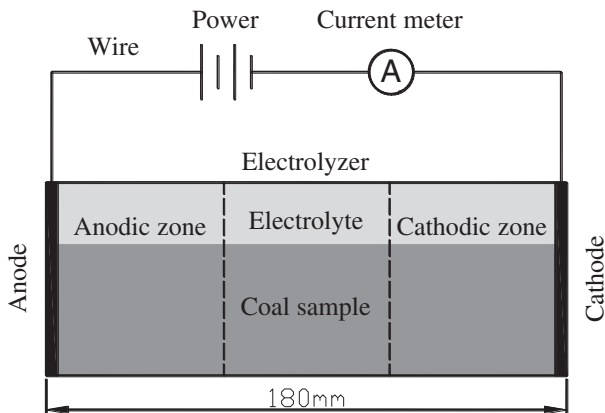


Fig. 1. Diagram of electrochemical modification equipment.

Then, the modified samples were dried in an oven at 378–383 K until a constant weight was achieved, at which time the samples were weighed and recorded.

2.3. Experimental apparatus and process

The experimental apparatus used for the desorption kinetic experiment of lump anthracite is shown schematically in Fig. 2. It mainly consists of an adsorption canister, reference canister, thermostatic water bath, methane cylinder, vacuum pump and measuring cylinder. The adsorption canister is 150 mm in diameter and 200 mm tall.

Based on the volumetric method, methane desorption kinetic experiments were carried out with samples before and after modification according to the following steps. (1) A sample was sealed into an adsorption canister and degassed at 333 K to get a static vacuum of 1 Pa within 3 days. (2) The temperature of the water bath was adjusted to 298 K, and then the adsorption canister was filled with methane. The methane was then absorbed by the sample at constant temperature and constant pressure (298 K, 1 MPa). (3) When the pressure difference in the adsorption canister was less than 0.001 MPa · d⁻¹, the coal methane emission was released under atmospheric pressure and bled into an inverted graduated cylinder filled with water. The gas volume was carefully measured in a series of readings over a period of weeks or months, until little or no more gas was produced. (4) The adsorption equilibrium pressure was changed (in order, 298 K, 2 MPa; 298 K, 3 MPa; 298 K, 4 MPa), and the above steps were repeated.

After the experiment, one small piece of sample was selected on each side of the cylinder samples, before and after modification, and ground with abrasive papers to a cube with 6 mm edge lengths for pore size distribution with mercury injection. The mercury porosimetry experiments were performed using an Autopore IV 9500 Instrument (Micro-meritics, US), which permits the mercury filling at as low as 3.5×10^{-3} MPa, up to 207 MPa. The dry sample was evacuated from the instrument to <50 μmm Hg. The volume of mercury penetration into the sample was measured with increasing applied pressure. A contact angle of 130° and surface tension of 485 dyn/cm were used in the Washburn equation to determine the pore size distribution (Clarkson and Bustin, 1999). Friesen and Mikula (1987) found that coal compressibility has an effect on mercury porosimetry results especially when pressure is greater than 10 MPa. Therefore, the pore volume at pore sizes larger than 100 nm is the injected mercury volume corresponding to the pressure of about 10 MPa. In addition, the surface components of the polished cube samples, both before and after modification and in the anodic and cathodic zones, were measured by SEM-EDS analysis. The experiments for SEM-EDS analysis were performed using a TM-1000

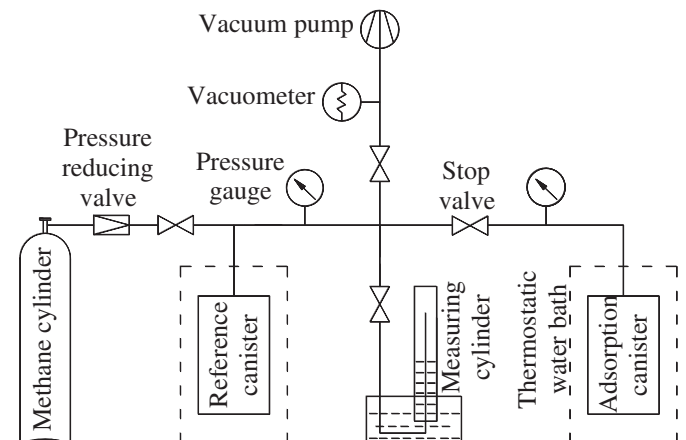


Fig. 2. Diagram of lump coal adsorption-desorption apparatus.

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