



Intensification mechanism of oxidized coal flotation by using oxygen-containing collector α -furanacrylic acid



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ABSTRACT

A traditional hydrocarbon collector dodecane and an oxygen-containing collector α -furanacrylic acid were selected for oxidized coal flotation in the present study, respectively. The changes of the surface functional groups and zeta potential of oxidized coal were measured before and after α -furanacrylic acid absorption and the electro-negativity of functional groups in α -furanacrylic acid was calculated to indicate its absorption mechanism on oxidized surface. Surface free energy and interfacial interaction energy were calculated to identify the interaction mechanism between coal and α -furanacrylic acid from the perspective of thermodynamics. Results show that 53.80% flotation yield with 14.03% ash content was obtained when 7000 g/t dodecane was used. In contrast, 69.17% yield with 11.98% ash content could be obtained only using 931 g/t α -furanacrylic acid. The high flotation recovery was attributed to the hydrogen bonding function between α -furanacrylic acid ($-\text{COOH}$ and oxygen-containing five-membered rings) and oxidized coal surface. The interfacial interaction energy is $21.20 \text{ mN}\cdot\text{m}^{-1}$ for coal-water- α -furanacrylic acid system while $-23.09 \text{ mN}\cdot\text{m}^{-1}$ for coal-water-dodecane system. A repulsive hydration force was found between oxidized coal and α -furanacrylic acid via water phase. α -Furanacrylic acid could not form hydrogen bond directly with the oxygen-containing functional groups on coal surface. Instead, the water molecules in hydration film may act as the bridging role of hydrogen bond for α -furanacrylic acid absorption on oxidized coal surface.

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1. Introduction

Oxidized coal is generally classified into weathered coal and spontaneous combustible coal. Weathered coal refers to that exposed on the ground surface under natural weathering conditions such as wind, sunshine, and precipitation. It undergoes a gradually slowing weathering process, and the coal is oxidized from outside to inside. As oxidation progresses to the inner parts of the coal, humic acid eventually forms [1,2]. Spontaneous combustible coal refers to the coal undergoing oxidation under hypoxic and high temperatures, such as partial cleavage and combustion. Spontaneous combustible coal is also referred to as high-temperature oxidized coal [3]. During oxidation, the physical and chemical properties of coal are degraded. The most prominent characteristic change is an evident in floatability. The extremely low floatation recovery of oxidized coal results in a significant loss of low-density clean coal, causing a serious waste of coal resources [4,5].

Froth flotation is an effective separation method for fine coal cleaning and upgrading that is based on the differences in surface hydrophobicity between organic matter and mineral matter [6–9]. Common oily collectors (diesel and kerosene) are added to the flotation

pulp to make coal particles more hydrophobic and separable from the pulp by attachment to rising air bubbles that form a particle-rich froth on the suspension surface. However, the beneficiation of oxidized coal is difficult to achieve by conventional flotation with oily collectors because of the presence of high surface oxygen functional groups. The firm hydration film formed at the hydrophilic sites of oxidized coal surfaces because of hydrogen bonding leading to the drainage rate of the thin liquid films between bubbles and oxidized coal particles is slow. As a result, bubble–particle adhesion is denied. The fundamental factors that affect the floatability of oxidized coal slimes are its intrinsic properties. It is well accepted that the addition of surfactants or oxygenated functional groups to oily collectors can significantly enhance the oxidized coal flotation performance [10–13]. Numbers of works have been conducted to investigate the interaction between oxidized coal particles and the reagents. The most representative work was done by Jia et al. [14]. A series of non-ionic surfactants containing oxygenated and aromatic functional groups, i.e., tetrahydrofurfuryl esters (THF), as collectors were used to enhance low-rank/oxidized-coal flotation and the interaction mechanisms between collectors and coal surface were proposed by Jia et al., as shown in Fig. 1: Hydrogen bonding of oxygen atoms in the THF polar part of the molecule with oxygenated surface sites on the coal; Hydrophobic bonding of the aliphatic hydrocarbon chain with the hydrophobic sites on the coal surface; Pi-bonding of

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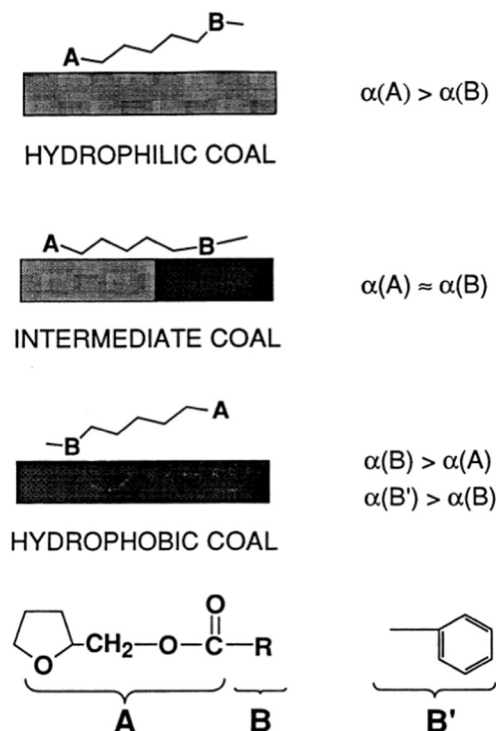


Fig. 1. The interaction mechanisms between collectors and coal surface (The affinity of individual functional group of reagents for the coal surface is represented as α and the polar head and hydrocarbon chain in collectors are represented as A and B, respectively) [14].

the benzene ring on the hydrocarbon chain of the collector with aromatic sites on the coal surface. High oxidized coal flotation recovery was attributed to the hydrogen bonding function. The adsorption/spreading mechanisms of reagents for direct contact flotation procedure and normal flotation procedure for oxidized coal were also studied by Ahmed and Drzymala [15]. Xia et al. found that oxidized coal flotation can be achieved by using oxidized diesel oil also due to the hydrogen bonding with the polar part of oxidized coal surface and oxidized diesel oil indicated by Fourier transform infrared spectroscopy [16]. The other enhancement methods for oxidized coal flotation, i.e., grinding, premixing/preconditioning, ultrasound, thermal, microwave and direct contact mixing of the reagents with dry coal before wetting, have been reviewed by Xia et al. [17].

Although the hydrogen bonding between oxidized coal surface and polar oxygen-containing collector is widely accepted to be the intensification mechanism for oxidized coal flotation, most of the results were obtained through qualitative speculation. The intensification mechanism and the interaction between oxygen-containing group collector molecules and oxidized coal surface are complex and require to be explained quantitatively. In the present study, a traditional hydrocarbon collector dodecane and oxygen-containing collector α -furanacrylic acid were selected for oxidized coal flotation, respectively. The changes of surface functional groups and zeta potential of oxidized coal were measured before and after α -furanacrylic acid absorption to indicate the absorption mechanism of α -furanacrylic acid on oxidized surface. The electro-negativity of α -furanacrylic acid was calculated to explain the zeta potential results. Finally, the surface free energy and interfacial interaction energy were calculated to identify the interaction mechanism quantitatively between coal and dodecane or α -furanacrylic acid from the thermodynamics perspective. The results of this research are expected to provide a basic understanding of intensification mechanism of oxidized coal used oxygen-containing collector and guidance for the industrial application of oxidized coal flotation.

2. Experimental materials and methods

2.1. Materials

2.1.1. Coal samples

Fine oxidized coal samples were collected from Ningxia Taixi Anthracite coal plant, China. The coal samples were first placed outdoors (average temperature 32 °C) for two months for natural oxidization and then placed in a 150 °C for 10 days oven for high-temperature oxidation. The proportion of the -0.045 mm particle size fraction was 49.31%, with a total ash content of 21.26%. The proportion of the -1.6 g/cm³ density fraction was 43.70% with an ash content of 31.17%. In order to gain more information regarding the properties of the coal samples, scanning electronic microscopy (SEM, QuantaTM 250, FEI, USA), X-ray diffraction (XRD, D8 ADVANCE, Bruker, Germany) and X-ray photoelectron spectroscopy (XPS, ESCALAB 250Xi, Thermo Scientific, Waltham, MA, USA) experiments before and after oxidation were conducted.

SEM results of the coal samples before and after oxidation are shown in Fig. 2. The surface of the oxidized coal was clearly more rugged with massive pores/gaps compared with that of fresh coal. Mineral impurities, which always appeared to be grey in SEM photographs, were found on the oxidized coal surface while not observed directly on fresh coal surface. It indicates that during the oxidation process, organic compounds on the coal surface decomposed, whereas minerals did not. Organic shedding and relative minerals enrichment directly resulted in the formation of gullies and pores. The oxidation process significantly changed coal surface physical-chemical properties. XRD analysis of the coal samples before and after oxidation is shown in Fig. 3. Quartz and clinocllore are the main mineral impurities that further increase the hydrophilicity of the coal samples. All of these factors make coal particles are difficult to be captured by air bubbles in flotation process. It should be noted that the phase composition do not show great difference between fresh coal and oxidized coal.

XPS experiments were conducted to obtain more comprehensive information about the oxidized coal surface properties before and after oxidation. An XPS system with Al K α radiation ($h\nu = 1486.6$ eV) and a 900 μ m light spot size was used to characterise the surface chemistry composition of oxidized coal. Each XPS spectra data set was fitted using the XPS Peakfit software. Binding energy calibration was performed by setting the C1s hydrocarbon peak to 284.6 eV. The XPS wide energy spectrums of the coal samples before and after oxidation are shown in Fig. 4. Semi-quantitative results for the surface chemical compositions of fresh coal and oxidized coal, based on the XPS wide energy spectrums, are shown in Table 1. The content of element C decreased from 49.12% to 40.18% after oxidation, while oxygen content increased from 32.63% to 39.89%. It indicates that a fraction of the organic carbon was indeed attacked by the oxygen in air to inorganic carbon dioxide during the oxidation stage. A large amount of oxygen-containing functional groups existed on the surface of oxidized coal. The contents of inorganic mineral elements (Si, Al, and Na) on oxidized coal surface were also higher than that of fresh coal, which is consistent with the SEM results. The organic fractions on the coal surface were attacked by oxygen, and fresh minerals were fully exposed to the surface.

2.1.2. Collectors

Based on the analysis of coal sample characteristics, a large number of oxygen-containing functional groups such as $-OH$ and $-COOH$ were found on oxidized coal surface, which significantly decreases the surface hydrophobicity. In the regular coal flotation process, hydrocarbon oil collectors can effectively adsorb at hydrophobic sites on the coal sample surface, increasing the hydrophobicity and the floatability. However, the beneficiation of oxidized coal can be achieved by using polar oxygen-containing group collector. Thus, a traditional hydrocarbon collector dodecane and oxygen-containing collector α -furanacrylic acid

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