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Inhibiting effect of imidazolium-based ionic liquids on the spontaneous combustion characteristics of lignite



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ABSTRACT

In this study, three imidazolium-based ionic liquids (ILs) and distilled water flushing were used to treat lignite subsamples to ascertain the inhibiting effect of ILs on the spontaneous combustion characteristics of lignite. The combustion characteristics, gas generation results during decomposition, and changes in the functional groups in these IL- and water-treated coal subsamples were compared with those of untreated coal through thermogravimetry and Fourier transform infrared spectroscopy analysis. The results showed that 1,3-dimethylimidazolium iodide ([Mmim][I]) was the most capable of reducing the – OH groups in lignite and inhibiting the spontaneous combustion of lignite. Furthermore, 1-ethyl-3-methylimidazolium iodide ([Emim][I]) was the most capable of increasing the - COOH groups in lignite and reducing the maximum mass loss rate of lignite. Analysis of the recovered ILs revealed that the ILs did not substantially change after the treatment, and recovery rates higher than 92% were achieved.

1. Introduction

Coal is the most abundant fossil fuel in the world and is the principal energy source in many countries [1]. For instance, coal is China's primary energy source, comprising approximately 64% of China's current primary energy consumption structure. This structure implies that coal still plays a central role in China's energy sector. However, coal production is still faced with numerous challenges. Spontaneous combustion is one of the main disasters potentially producing a large amount of poisonous and hazardous gases, which present enormous threats to workers' lives and undermines the safety of coal industry. According to the relevant literature, the spontaneous combustion of coal starts with the oxidation of coal surface-active structures at low temperatures [2,3]. Because of the accumulation of heat from oxidation, coal temperatures reach the ignition point, ultimately causing spontaneous combustion [4]. The low-temperature oxidation of coal is substantially affected by the number of active functional groups. For example, some studies have confirmed that the presence of more O-containing functional groups in coal may result in a higher tendency for spontaneous combustion [5,6]. Additionally, these O-containing functional groups can readily cross-link with the generated CO₂ and H₂O gases [7]. This process implies that reducing the number of active functional groups is

also likely to reduce the harmfulness of coal's spontaneous combustion.

Ionic liquids (ILs) are salts that have a melting point below 100 °C [8,9]. They have been identified as green solvents with low melting point, negligible vapor pressure, excellent thermal stability, and high solubility [10-12]. Because of these advantages, ILs have been used in chemical and coal industries. Painter et al. [13] used various ILs to treat Illinois No. 6 coal and found that the ILs dispersed and fragmented around the coal. Zhang et al. [14] observed that ILs could inhibit the spontaneous combustion ability of coal by breaking the active functional groups in coal. Wang et al. [15] used various ILs to break the functional groups in bituminous coal and determined that the ILs used could partially change the functional groups in bituminous coal. Regarding the oxidation inhibiting effect, Zhang et al. [16,17] reported that ILs can retard coal oxygenolysis. Cummings et al. [18] applied four ILs for treating two lignite samples and confirmed that the IL treatment process altered the thermal properties of both lignite samples and increased the aliphatic/aromatic ratios in both lignite samples. Wang et al. [19] revealed that [HOEmim][BF₄] and [Amim][Cl] can inhibit the cross-linking reaction between the O-containing functional groups in coal, thereby lessening the harmfulness of coal's spontaneous combustion.

In this study, three ILs with the same anion and different cations

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Fig. 1. Structural formula of [Pmim][I], [Emim][I],



were deliberately selected to examine the interactions between ILs and lignite and recovered ILs for each run to assess the viability of IL treatment.

2. Experimental

2.1. Materials

2.1.1. Coal sample preparation

The lignite sample used in this study was obtained from Selian, Inner Mongolia, China. It was ground to a size of approximately 200 mesh.

2.1.2. IL preparation

Three ILs, namely 1-propyl-3-methylimidazolium iodide ([Pmim] [I]), 1-ethyl-3-methylimidazolium iodide ([Emim][I]), and 1,3-dimethylimidazolium iodide ([Mmim][I]), were purchased from Aolike Chemical Industry Co., Lanzhou, GanSu, China, at 99 mass% purity and were used in this study. Fig. 1 illustrates the chemical structures of these ILs [20–22].

2.2. IL treatment of coal subsamples

Because [Emim][I] and [Mmim][I] are solid at ambient temperature, they were heated in water until melting. Subsequently, all ILs were intensively and similarly mixed with lignite subsamples. After melting, they were vibrated until their temperatures were the same as ambient temperature. The lignite subsamples were dried in a vacuum dryer for 24.0 h at ambient temperature. The IL and lignite mixtures were then prepared at a 1:2 mass ratio of lignite to ILs. The IL and lignite mixtures were sealed in jars that were placed in an ultrasonic vibration generator for 3.0 h. Thereafter, the mixtures were washed using distilled water to remove ILs until the pH value of the filtrate was neutral, and the mixtures were filtered using filter paper. Subsequently, the distilled water and IL mixtures were placed in an oven at 80 °C to recover the ILs, and the lignite subsamples were placed in a vacuum dryer for 60.0 h at ambient temperature to dry. To mitigate the effects of distilled water flushing on the lignite subsamples, a dry lignite subsample was flushed with distilled water and dried under the same conditions. The lignite subsample that was not pretreated with liquids, including ILs and distilled water, was named untreated coal (Utc). The lignite subsample that was only flushed with distilled water was named water-treated coal (Water-tc). The IL-treated coal (IL-tc) subsamples, [Pmim][I]-, [Emim] [I]-, and [Mmim][I]-treated coal, were named [Pmim][I]-tc, [Emim] [I]-tc, and [Mmim][I]-tc, respectively. To guarantee the consistency of the procedure and experimental results, three subsamples were assigned to each IL treatment group.

2.3. Experimental procedure

Thermogravimetry coupled with Fourier transform infrared spectroscopy (TG-FTIR) is an effective technique used to investigate oxygenolysis characteristics and the gases produced at different time points [23–26]. In this study, the functional groups in the Utc, Water-tc, IL-tcs, ILs, and recovered ILs were detected using the PerkinElmer Spectrum 100 FTIR. Each subsample was scanned four times, and the detection range was from 650 to 4000 cm^{-1} with a resolution of 4 cm^{-1} .

PerkinElmer Pyris 1 TG was used to perform decomposition experiments. Five species lignite subsamples, each with an initial mass of approximately 9.0 mg, were prepared for these series of experiments. The heating rate was set at 5 $^{\circ}$ C min⁻¹, and the temperature ranged from 30 to 800 $^{\circ}$ C. Air flow was set at 20 mL min⁻¹, and the gases produced through the oxygenolysis of the lignite subsamples were analyzed through FTIR to test the variety of the gases.

and [Mmim][I] [19].

3. Results and discussion

3.1. Thermogravimetry-differential thermogravimetry analysis of lignite subsamples before and after treatment

The thermal stability of the subsamples before and after treatment with the ILs and distilled water was determined through TG. Because TG was the main experiment in this study, the three subsamples in each IL treatment group were analyzed through TG to guarantee consistency. The typical TG and differential thermogravimetry (DTG) curves of the subsamples before and after treatment are shown in Fig. 2. The results of ultimate analysis of the studied coal sample are shown in Table 1. In theory, the apparent exothermic onset temperature of the lignite subsamples (T_0) was defined as the temperature at the intersection point of two lines on the TG curve. One is the horizontal line at the beginning of mass loss. The other is the tangent of the point at the temperature of the fastest mass loss rate. The peak temperature (T_p) was defined as the temperature at which the highest rate of conversion occurs on the DTG curves.

3.1.1. Analysis of TG curves

As depicted in Fig. 2, the Utc and Water-tc subsamples exhibited similar curves. The curves of the IL-tc subsamples exhibited different degrees of change compared with those of the Water-tc subsample. The decomposition properties of lignite did not prominently change after flushing with distilled water. The ILs could inhibit the decomposition of lignite. Moreover, the average values and standard errors of T_0 for the Utc, Water-tc, [Pmim][I]-tc, [Emim][I]-tc, and [Mmim][I]-tc subsamples were 398 ± 1.3 , 401 ± 0.6 , 406 ± 1.9 , 414 ± 1.2 , and 426 ± 2.0 °C, respectively. They were in the order [Mmim][I]-tc > [Emim][I]-tc > [Pmim][I]-tc > Water-tc > Utc. This novel finding indicated that distilled water flushing could slightly increase the apparent exothermic onset temperature of lignite. After excluding the effect of distilled water flushing, the results showed that the ILs were more capable of increasing the apparent exothermic onset temperature of lignite than did distilled water flushing.

Among the ILs, [Mmim][I] possessed the strongest capacity to increase the apparent exothermic onset temperature. [Pmim][I] exerted a weak effect on the apparent exothermic onset temperature. Furthermore, the mass losses of the five species lignite subsamples differed during decomposition. The mass losses of the Utc, Water-tc, [Pmim][I]-tc, [Emim][I]-tc, and [Mmim][I]-tc subsamples were $96.05 \pm 0.08\%$, $96.39 \pm 0.06\%$, $97.03 \pm 0.03\%$, $96.95 \pm 0.03\%$, and $96.84 \pm 0.02\%$, respectively. The order of mass loss was [Pmim][I]-tc > [Emim][I]-tc > [Mmim][I]-tc > Water-tc > Utc. The mass loss result indicated that flushing with distilled water can remove the mineral matter in the lignite. Moreover, the ILs can break the internal

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