



# Desulfurization of low rank coal co-pyrolysis with reduced iron powder followed by dry magnetic separation

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## ABSTRACT

Organic sulfur in coal is usually integrated with macromolecular skeleton structure of organic components and hence difficult to remove through physical separation. Chemical leaching and biological treatments are considered as the effective methods for the removal of organic sulfur from coal, however these treatment processes require huge investment and long treatment period. In this paper, low rank coal co-pyrolysis with reduced iron powder (Fe powder) and then followed by dry magnetic separation was used to reduce the sulfur content of low rank coal. After dry magnetic separation, the organic sulfur content can be reduced from 2.00% for low rank coal before the pyrolysis to 0.33% for the coal after 700 °C co-pyrolysis with Fe powder at the coal/Fe-powder mass ratio of 4:1. However, the organic sulfur content can be only reduced to 1.28% for the coal after 700 °C pyrolysis without Fe powder. The Fe powder can promote/enhance the decomposition of organic sulfur and the released sulfur can be bonded with Fe powder to form the sulfocompounds which is separated along with the Fe powder during dry magnetic separation. X-ray photoelectron spectroscopy was used to indicate the sulfur forms in the separated Fe powder (namely magnetic tailings) and showed the magnetic tailings contained more FeS and FeS<sub>2</sub> compounds than the Fe powder before the pyrolysis. The magnetic tailings contained about 4.6% sulfur content at the co-pyrolysis temperature of 700 °C while the sulfur content of the Fe powder before the pyrolysis was nearly zero. A large proportion of organic sulfur was transferred to the Fe powder and then was separated along with the magnetic tailings by dry magnetic separation.

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## 1. Introduction

During the coal combustion and utilization, the sulfur, such as pyrite and organic sulfur in coal is a key issue for the environmental protection (Restrepo et al., 2015). The sulfur in coal is removed or controlled by three major methods, i.e. removal of sulfur by coal preparation or cleaning techniques prior to utilization (Eliot, 1978), sulfur-fixing technology by adding sulfur-fixing agent during the burning of coal (Cheng et al., 2003), and desulfurization treatments of sulfur-containing flue gas after the burning (Calkins, 1994; Wang et al., 2008). For the coke making industry, coal desulfurization is essential because the sulfur has negative effects on coke quality and production. Generally, coal preparation or cleaning prior to the utilization or coke production is considered as the most economic

and effective approach. Even though the removal of both inorganic and organic sulfur compounds during coal combustion is also effective, high costs are required for the investment in equipment and operation. Therefore, researchers pay relative more attention to the coal desulfurization and cleaning prior to the utilization (Demir, 2017).

For the removal of inorganic sulfur from coal, such as pyrite which is usually integrated with high ash gangue minerals. If the coal is crushed or ground to pass a specific size and the high ash gangue particles are not integrated with organic components of coal, these high ash gangue particles can be easily separated by physical separation technique. For example, the gravity-based separation (Honaker et al., 2000; Özbakir et al., 2017) and magnetic separation (Maxwell and Kelland, 1978; Yavuz et al., 2009) have been proved to be of feasibility in coal desulfurization. Additionally, flotation is applied in the separation of fine high ash gangue particles from coal (Demirbaş, 2002; Xia et al., 2017b, 2018). In other words, the inorganic sulfur, to some extent, can be effectively removed prior to the utilization by the gravity-based

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separation or flotation.

In the past decades, microwave energy was widely used in material treatments. For coal desulfurization, Zhang et al., 2015, 2018 claimed that the magnetism of pyrite has been proved to be significantly enhanced using microwave irradiation, and then the pyritic sulfur content of coal can be reduced by the following magnetic separation (Jorjani et al., 2004; Uslu and Atalay, 2004). The microwave is also used by Yang et al. (2016) to enhance the chemical desulfurization of coal with the desulfurization additive and acid/alkaline reagents.

However, the removal of organic sulfur from coal seems invalid using physical separation or flotation methods because the organic sulfur is usually integrated with macromolecular skeleton structure of organic components in coal. Therefore, the chemical leaching desulfurization, chemical oxidation desulfurization (Shen et al., 2012) as well as the bio-desulfurization (Monticello and Finnerty, 1985) are used for the removal of the sulfur from coal in laboratory scale, but they are difficult for the quick application in the clean coal industry.

The pyrolysis of coal is a majority for coke production. For low rank coal utilization, coal pyrolysis is now considered as a clean coal utilization way to obtain gas/liquid components for raw chemical materials and coal tar for the combustion or other applications (Niu et al., 2017; Xia et al., 2017a). Because low rank coal is usually high ash, high sulfur and high moisture, the upgrading approaches, such as desulfurization, drying and deashing processes are very important for low rank coal prior to the utilization. Tang et al. (2018) found that the pyrolysis of coal have positive effects on the removal of organic sulfur from coal because of the decomposition of organic sulfur from coal structure. Telfer and Zhang (2001) found the alkylsulphides, thiophenes, arylsulphides compounds in coal can be decomposed and the sulfur-containing gas will be released when the pyrolysis temperature reaches 700–900 °C. The decomposition of pyrite is happened when the pyrolysis temperature is only more than 300 °C (Khan, 1989). In addition, Gryglewicz et al. (1996) found that the magnetism of pyrite is also enhanced during the pyrolysis processes, and hence the pyrolysis process of coal was combined with dry magnetic separation to effectively remove the pyritic sulfur from coal (Celik and Yildirim, 2000; Koca et al., 2000; Renda et al., 2001). The application of dry magnetic separation of coal benefits both the economic and technical targets for coal cleaning because dry separation can save water resource. However, the combination of pyrolysis and dry magnetic separation is still invalid for the organic sulfur removal from coal.

In our previous study, a novel low rank coal desulfurization method was proposed (Xia, 2018) by using the Fe powder to promote the coal desulfurization. It was found that the organic sulfur can be transferred to the Fe powder. After the pyrolysis of coal and Fe powder mixture at the temperature of 400–550 °C, plenty of Fe-

related magnetic compounds were formed and then can be separated by magnetic separation. The sulfur content of coal after the pyrolysis and dry magnetic separation is significantly reduced. However, the mechanism of what are the Fe-related magnetic compounds and the optimized pyrolysis temperature and the coal/Fe-powder mixture mass ratio have not been well investigated. This paper will give a comprehensive study about this novel desulfurization of low rank coal.

## 2. Experimental method and procedure

### 2.1. Analysis of coal sample properties

The lump low rank coal sample was provided by Xinglinhaote of Inner Mongolia of China. The lump low rank coal was crushed to 1–6 mm diameter and the sink-float tests were done for obtaining low density coal particles with low ash content. Then, the 1–6 mm low density low rank coal particles were further crushed and ground to pass 0.074 mm sieve and finally the <0.074 mm fine low rank coal was used as raw coal sample, named low rank coal sample in this investigation. The proximate and sulfur forms analysis of low rank coal sample were presented in Table 1. The organic sulfur content of low rank coal sample was 2.00% which is much higher than that of pyrite sulfur content (0.07%) and sulfate sulfur content (0.25%). It indicates the sulfur in this low rank coal sample primarily consists of organic sulfur.

### 2.2. Analysis of Fe powder properties

The Fe powder size is below 0.045 mm diameter. In this paper, the Fe Powder was analyzed by XRF (Bruker S8 Tiger, Germany) to see the elements composition. The element content of Fe powder is presented in Table 2. The content of element Fe is about 98%, which shows this Fe powder sample owns high purity. Then, the Fe powder was also analyzed using Scanning electron microscopy (SEM) with energy dispersive spectrometry (EDS) and the results are presented in Fig. 1. The weight of element Fe is about 94% from EDS results. The two analysis results are close and both indicate that the Fe powder owns high purity. In addition, there is only few S element in the Fe powder which will be further analyzed using X-ray photoelectron spectroscopy (XPS) measurement.

### 2.3. Pyrolysis treatment, dry magnetic separation and sulfur analysis

The pyrolysis treatment of low rank coal was conducted under nitrogen atmosphere and the pyrolysis treatment period was 1 h. The pyrolysis of coal or coal/Fe powder mixture was finished in the

**Table 1**  
Proximate and sulfur forms analysis of low rank coal sample.

Proximate analysis/%					Sulfur forms analysis/%		
<i>S(t,d)</i>	<i>Ad</i>	<i>Vdaf</i>	<i>FCdaf</i>	<i>Mad</i>	<i>S(p,d)</i>	<i>S(s,d)</i>	<i>S(o,d)</i>
2.32	13.21	65.19	34.81	14.79	0.07	0.25	2.00

*Mad* and *Ad* represents the moisture based on the air-dry basis and the ash based on the dry basis. *FCdaf* and *Vdaf* represent fixed carbon and volatile matter contents based on the dry-ash free basis. *S(t,d)*, *S(p,d)*, *S(s,d)*, and *S(o,d)* represent the total sulfur, pyrite sulfur, sulfate sulfur, and organic sulfur contents based a dry basis.

**Table 2**  
Element content analysis of the Fe powder by XRF.

Element name	<i>Fe</i>	<i>K</i>	<i>Mn</i>	<i>Ti</i>	<i>Gd</i>	<i>Ca</i>	<i>Al</i>	<i>Mg</i>	<i>P</i>	<i>Si</i>
Relative content/%	98.19	0.02	0.04	0.05	0.13	0.24	0.25	0.29	0.34	0.45

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