



Inhibitory effect of coal direct liquefaction residue on lignite pulverization during co-pyrolysis



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ABSTRACT

Coal direct liquefaction residue (CDLR) inhibits lignite pulverization. In order to further understand the inhibitory effect of CDLR, Baori lignite was mixed with CDLR, and tested at different temperatures during pulverization and pyrolysis in a rotary kiln. The results showed that CDLR reduced the pulverization rate (β) of CDLR-Baori lignite co-pyrolysis char significantly, compared to Baori lignite without CDLR. The reduction in the pulverization rate ranged from 3.14% to 5.67%. Granulation occurred through CDLR and Baori lignite co-pyrolysis with granulation rate (λ) ranging from 0.38% to 4.23% at 450–550 °C, and reached saturation at around 4% after 550 °C. A rotary kiln generally provides better lignite pulverization compared to a fixed bed, as it allows more thorough mechanical mixing. Co-pyrolysis char bonding on the wall surface of the furnace was greatly reduced after 550 °C. An analysis of the pore structure and surface appearance of lignite pulverization products showed that the inhibition of pulverization through CDLR resulted from capturing fine particles with a sticky surface and reinforcing pore structure of particles.

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1. Introduction

Lignite has high moisture (20–50 wt.%) and volatile (40–55 wt.%) contents. Lignite pyrolysis technology is used as a thermo-chemical decomposition process in which lignite is converted into a carbon-rich char and volatile matter as feedstock for chemicals and liquid fuels [1–4]. Thus, coal pyrolysis is fundamental in the industrial development and utilization of lignite. However, fragmentation, and more specifically, the pulverization of lignite, seriously hinders the normal operation of the pyrolysis process. Typical lignite-processing technologies in China, include multistage rotary furnace (MRF), which was invented by China Coal Research Institute [5], liquids from coal, invented by SGI International Corporation (LFC) [6] and Da gong fast pyrolysis of solid heat carriers, invented by Dalian University of Technology (DG) [7]; all these technologies face pyrolysis-related issues. The main problems include: a) a large number of fine particles and dust are produced during lignite fragmentation and pulverization; b) dust and tar mix with each other and block the ducts. Thus, continuous pyrolysis may be hindered; and c) dust decreases the quality of tar, and makes tar and dust difficult to separate. No effective solution has been reported to solve these problems so far; therefore, a substantial technology breakthrough is needed in this regard.

The factors affecting fragmentation and pulverization mainly include water vapor behavior, heat processing parameters, and volatile release [8]. Volatile release is the most important factor for coal particle

fragmentation and pulverization during pyrolysis. A large and abrupt release of volatiles results in large inner pressure, and changes the pore structure in coal [9,10,11]. Newly generated, small pores weaken the pore wall, leading to the collapse of coal, while promoting further pulverization. This suggests that if pores are filled and reinforced, pulverization of lignite will reduce.

The key to the inhibition of pulverization is to capture fine particles, and reinforce the structure of particles. Coal direct liquefaction residue (CDLR) serves as a softening mediator with strong bonding capability and mobility at temperatures that are higher than 200 °C [12,13]. Therefore, it is possible to bond the particles to each other during the co-pyrolysis of lignite with CDLR to reduce the degree of pulverization. Furthermore, particle pores should be filled and reinforced. Interaction and co-pyrolysis of lignite and CDLR will improve the pyrolysis process and increase the yield and quality of tar [14]. Most studies related to co-pyrolysis are limited only to the yield and quality of char. Limited information is available on the method to inhibit lignite pulverization.

In this study, co-pyrolysis of lignite and CDLR at different temperatures were investigated in a rotary kiln. The work is to justify the feasibility of the inhibitory effect of CDLR on lignite pulverization during co-pyrolysis and provide a new approach to solve this problem.

2. Material and methods

2.1. Materials

Coal samples used in this study are Baori lignite from Inner Mongolia, China. Coal direct liquefaction residue (CDLR) was obtained from Direct

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Table 1
Proximate and ultimate analysis of samples.

Samples	Proximate analysis (wt.%)					Ultimate analysis (wt.%, daf)				
	M_{ar}	M_{ad}	A_d	V_{daf}	FC_{daf}	C	H	N	S	O^*
Baori lignite	32.07	24.24	13.23	46.81	53.19	57.01	4.45	0.74	0.28	37.52
CDLR	0.02	0.01	18.48	33.38	41.56	84.10	4.82	1.07	3.12	6.89

ar: as received basis; ad: air dried basis; daf: dry and ash-free basis; M: moisture; A: ash; V: volatile; FC: fixed carbon; H: hydrogen; N: nitrogen; S: sulfur; O: oxygen.

* By difference.

Coal Liquefaction Pilot Plant of the Shenhua Group Corporation, China. Proximate and ultimate analyses of samples are listed in Table 1.

2.2. Experimental apparatus

Co-pyrolysis experiments were carried out in a JXHZ-15-9 rotary kiln, manufactured by Beijing Xinzhou Technology Company, China. The nitrogen required for the experiment was generated by a PSA-20 nitrogen generator manufactured by Beijing Huagang Technology Company, China. PSA-20 separates nitrogen from clean and dry compressed air based on the principle of pressure swing absorption (PSA). The pyrolysis product size was analyzed using a $\Phi 200$ mm standard test sieve shaker, manufactured by Shanghai Shuli Technology Company, China.

2.3. Co-pyrolysis experiments

In order to evaluate the effect of CDLR on pulverization, 1000 ± 0.01 g Baori lignite and 1000 ± 0.01 g CDLR-Baori lignite (30% CDLR and 70% Baori lignite) samples were tested in the co-pyrolysis experiments. As shown in Fig. 1, the samples were pyrolyzed in the rotary kiln reactor with 99.9% pure N_2 at a flow rate of $4 \text{ m}^3 \text{ h}^{-1}$. The rotary rate was 4 rpm in all experiments. Attributes of Baori lignite, with and without CDLR, were investigated during pyrolysis at 450 °C, 500 °C, 550 °C, 600 °C and 650 °C. When the desired temperature was reached, the temperature of reactor was maintained for 30 min. After the pyrolysis process, the solid products were collected in a jar, they were then, cooled and sieved into different size grades, according to GB/T477-2008 standard in China.

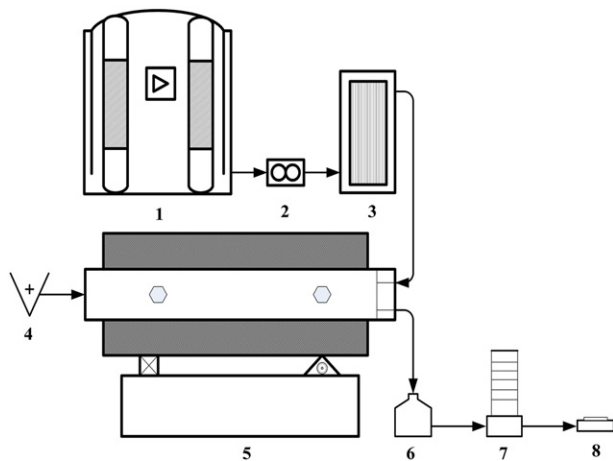


Fig. 1. Schematic view of co-pyrolysis process in rotary kiln. 1—nitrogen generator; 2—flowmeter; 3—gas heater; 4—coal preparation; 5—rotary kiln; 6—cooling; 7—screening machine; 8—electronic balance.

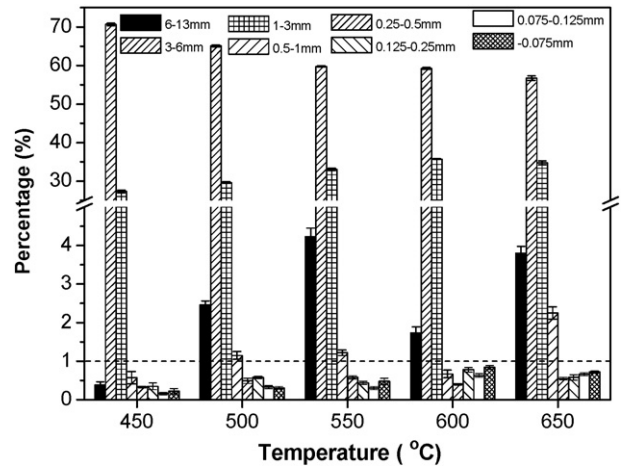


Fig. 2. Size distribution of co-pyrolysis products under different temperatures.

2.4. Calculation of pulverization rate β and granulation rate λ

Pulverization rate β was measured to characterize the degree of product pulverization, following the method used in our previous study [8], as shown in Eq. (1):

$$\beta = \frac{m_1}{m} \times 100\%. \quad (1)$$

Granulation rate λ was calculated to characterize the degree of product granulation, as shown in Eq. (2):

$$\lambda = \frac{m_2}{m} \times 100\%. \quad (2)$$

Where m_1 is the total weight of particles smaller than 1 mm, m_2 is the total weight of particles larger than 6 mm (the original size of liginites before co-pyrolysis), and m is the total weight of the sample. Higher pulverization rate indicates greater degree of pulverization with a larger proportion of fine particles. Higher granulation rate indicates higher degree of granulation with a larger proportion of large particles.

All the experiments were carried out at least three times in order to determine the variability of the results and to assess the experimental errors. In this way, the arithmetical averages were calculated for all

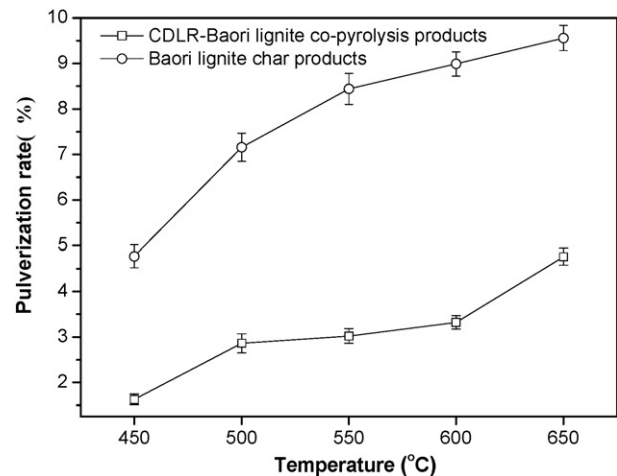


Fig. 3. The change of co-pyrolysis products and lignite pulverization rate (β) with temperature.

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