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An Empirical Study on Laboratory Coke Oven-based Coal Blending for Coking with Tar Residue

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Tar residue is the solid waste produced in coking plants or gas generators. Utilizing tar residue as additive in coal blending for coking is an effective approach for coal coking in our country, not only improving the yield and nature of coking products, but also realizing the recycling of tar residue. In this paper, tar residue is used as additive in coal blending for coking experiments, and combined with experiments on the crucible coke, small coke oven and industrial coke oven, the influences of the addition amount of tar residue on coke yield, coke reactivity and coke strength after reaction are further analyzed. As the experiment results reveal, compared to the experiment results of crucible coke and small coke ovens, the coke yield, coke reactivity and coke strength after reaction of industrial coke ovens are all bigger, leading to potential industrialization of coal blending for coking. Moreover, the influence of the amount of tar residue on the coal tar yield rate and gas yield is the same as that of coke reactivity and coke strength after reaction.

1. Introduction

With the rapid development of economy, the fast growth of iron and steel industry has led China's coke industry to accomplish extraordinary achievements (Si et al., 2017). According to incomplete statistics, China's total coke production capacity exceeded 500 million tons, making China the largest country in coke production, consumption and export (Shen et al., 2016; Long et al., 2011). Based on China's energy structure, we should make full use of coal resources, give full play to the advantages of coal resources, and develop them with new technologies and new methods (Du et al., 2018). Because of the different periods and depths of coal strata, there are various types of coal resources in China, with coking coal accounting for about 20% of the total coal storage and only around 32% of coal for coking, and therefore, there is a lack of high-quality coking coal resources (Wang et al., 2014; Tiwari et al., 2014). Hence, it is urgent to develop a new and improved coking process to fully cope with the lack of high-quality coal resources in China (Wang et al., 2016). The iron and steel industry sets an increasingly higher standard for coke quality, and traditional methods of coal blending for coking can no longer meet the requirements of metallurgical furnaces and blast furnaces in terms of coke quality (Wang et al., 2018; Yang and Wu, 2018). Some factories have improved the coke quality by increasing the allocation of high-quality coke and fat coal, not only greatly increasing the cost, but also aggravating the shortage of high-quality coking coal resources (Das et al., 2016). Meanwhile, the large amount of waste residue generated in the coking process of tar residue not only pollutes the environment, but causes waste of resources. Currently, it is mainly used in producing protective coatings (Jenkins and Mahoney, 2015). In certain ratio, tar residue and coal can be converted into coke, tar, and coke oven gas at high temperature, both utilizing the tar residue waste and producing high-quality coke. And the three mature principles of coal blending for coking include the co-carbonization, interchangeability, and the glial layer overlapping (Diez and Borrego, 2013). In this paper, tar residue was used as additive to carry out the experiments of coal blending for coking, which dramatically propelled the industrialization and application process.

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2. The status quo of coking coal and tar residue

2.1 The supply and demand of coking coal

Coking coal is the metamorphic bituminous coal generated in producing coke and other coking products and is the general name of all types of coking coal (Barnakov et al., 2016; Zhang et al., 2015). There is a huge demand for coking coal. In China, most of the existing coking coal is located primarily in North China, and is heavily unbalanced (Flores et al., 2017). Usually, various kinds of coal samples are used in coking. Bituminous coals can all be used for coal blending for coking according to different proportions. For example, gas coal, fat coal and coking coal all have different coking characteristics and functions.

Coal blending requires low ash content and sulfur content, proper volatile matter content and good adhesion. With the increasing demand for high quality coke, an average annual growth rate of 7%, the price of coke is rising steadily. Also, in the process of iron smelting, the growing of blast furnace scale and the improvement of blast furnace coal injection technology also put increasingly higher requirements on coke quality. Table 1 displays the coke quality index of China and several other countries, showcasing that the grade I metallurgical coke quality of our country is the same with foreign countries, and even more demanding, and the grade II coke quality index are overall lower than those of other countries.

Table 1: Coke quality index of many countries

Index	China		United States	Germany	France	Japan	Russia
	I	II					
St/%	≤0.61	0.61-1					
Block degree/mm	20-70	20-70	20-70	20-65	20-65	20-65	20-55
M_{40}	> 80%	> 76%	> 80%	> 80%	> 75%	—	> 90%
M_{10}	< 9%	< 10%	< 11%	< 8%	< 7%	8%	< 6%
Ad	≤13%	13-16%	8%	8%	9%	10%	11%

2.2 The status quo of tar residue utilization

Tar residue is the solid waste generated from coking plant or gas generators, and the secondary use of tar residue can produce coke and tar (Kim et al., 2016). Coal tar is a liquid product produced through decomposition at high temperature, consisting three types according to different temperatures. They are low temperature tar, the medium temperature tar and high temperature tar, with high temperature coal tar accounting for over 75% of the total. Because of the different generation techniques and raw materials, there are significant differences in the nature of different coal tar. Table 2 shows the nature of the tar, which reveals clearly that hot coal tar has significantly lower yield, bigger relative density and lower phenol content than those of the low temperature tar. By increasing the bulk density and the temperature, improving the nature of coal, the coking process and the coke post-treatment process, the quality of coke could be dramatically improved and better tar residue obtained.

Table2: Quality of tar

Tar types	Yield/%	Relative density	Moisture/%	Phenol/%	Naphthalene/%	Anthracene/%
Low temperature tar	12	0.99	—	15.6	2.3	1.5-1.8
High temperature tar	4	1.19-1.23	5-7	1.6-3	6-9	4-6

3. The experiment of coal blending for Coking with tar residue

3.1 Experiment equipment and procedures

In this experiment, tar residue produced by CRPC was used in the coal blending for coking experiment, and the main intangible data of the residue tar is shown in Table 3. The 600g crucible coke and 70kg small coke oven were selected as the experiment equipment and all the basic coal for experiments remained the same. Figure 1 shows the effect of tar residue addition on the mixed coals bond index, and it can be seen that with the increase of tar residue addition ratio, the mixed coals bond index first increases and then decreases. The coal samples were converted into coal powders and mixed to carry out the coal blending for coking process, and after the drying process and being converted into powder, the tar residue was blended into coking coal according to the ratio of experiment design. The coke oven body and the iron parts with the central control system were selected as experiment device, and the riser and cross pipe were used as smoke exhaust device. During the experiment, the small coke oven was first preheated to 700 ℃, and the coal and coke samples were put into the small coke oven, and then increase the center temperature to 950±10℃ through electric heating, and make the coking time to be 24 hours. After the experiment was finished, wet coke quenching method was used to quench coking. In the crucible coke experiment, Muffle furnace was used to control temperature, while all other operations remained the same.

Table3: The main intangible data of the residue tar

Analysis indicators	Mad/%	Aad/%	Vad/%	FCad/%	Sad/%
Tar residue	30.16	22.27	22.28	24.85	0.63

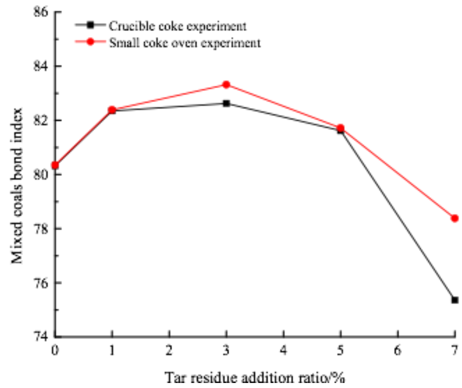


Figure 1: The index with tar residue addition on coal caking

3.2 Experiment results and discussion

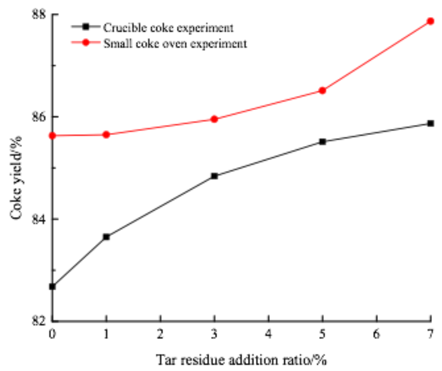


Figure 2: The comparison of coke yield

Fig. 2 shows the influence of tar residue addition ratio on coke yield. With larger tar residue addition ratio, coke yield increases. By comparing the small coke oven experiment and crucible coke experiment, it is shown that both experiments shared the same tendency of change. They had little influence on the coke yield and the nature of coking coal. Meanwhile, the small crucible coke experiment and small coke oven experiment shared the same result and pattern, with the coke yield of small coke oven slightly higher, indicating that it is feasible to scale up the experiment. Table 4 shows the test results of coking coal blending tar residue, revealing the change of moisture, ash, volatile, fixed carbon and abrasive wear resistance in coking coal as the tar residue addition ratio varied in the small coke oven experiment. Figure 3 displays the comparison of coke reactivity. With a 3% of tar residue addition ratio, the coke reactivity in both experiments reached the biggest, and with a 2%-6% of addition ratio, the coke reactivity in small coke oven experiment became bigger than that of the crucible coke experiments. Figure 4 shows the comparison of coke strength after reaction. As the addition ratio of tar residue increases, the cook strength after reaction first increases and then decreases. With 3% of addition ratio, it reaches the maximum, and the coke strength after reaction of the small coke oven experiment is greater than that of the crucible coke oven. In conclusion, compared with the crucible coke experiment, the coke yield, reactivity and coke strength after reaction of the small coke oven experiment all improved in some degree.

Table 4: The test results of coking coal blending tar residue

Project	Tar residue contents				
	0	1	3	5	7
Mt,ar/%	4.84	5.12	5.01	5.34	5.86
Aad/%	11.57	12.25	12.34	12.87	13.24
Vdaf/%	1.35	1.41	1.44	1.76	2.02
Std/%	0.70	0.71	0.70	0.72	0.73
FCad/%	86.45	85.38	86.52	86.32	86.53
M40/%	84.83	85.20	87.20	83.01	81.25
M10/%	7.01	7.06	6.31	5.70	6.14
＜25mm/%	5.64	5.13	5.31	4.44	4.51
＞80mm/%	2.01	1.58	1.44	1.67	1.32

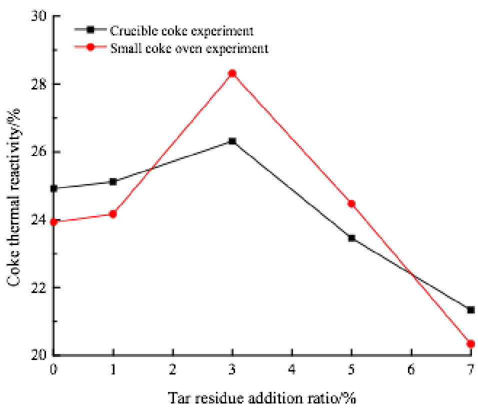


Figure 3: Compare of the coke reactivity

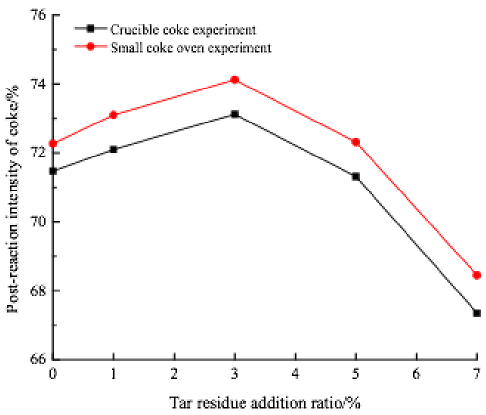


Figure 4: Comparison of coke strength after reaction

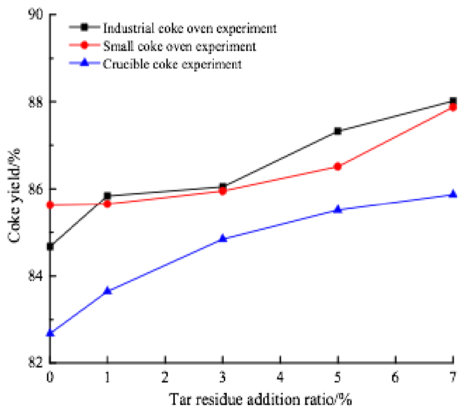
4. The coal blending for coking experiment of industrial coke oven

4.1 Experiment procedures

It is the basic principle that during the process of coal blending for coking, the bond index should not be too small and tar residue remains a certain level of matter feature. According to Section 3, we have realized the experiment of coal blending for coking in the laboratory, and we will apply the approach to the experiment of industrial coke oven generation. In the coking plant, there are 55 coke oven holes in total, and the experimental coke oven is 6 meters tall, with 20 hours for carbonization. The major components of the coking plant include the carbonization chamber, the combustion chamber, the regenerative chamber and the flue pipe, etc., controlled by the computer central control system. After the experiment is completed, the coke dry quenching method is used to quench the coke from the oven, and then the coke is transported to the coke storage field through the transmission system. The industrial production of coke oven includes coal blending, tar residue blending, coking production and subsequent assessment of coke, etc. The production of tar residue coal blending follows the same procedures as those in Section 3.1.

4.2 Experiment results and discussion

Figure 5 shows the influence of tar residue addition on coke yield, and it can be seen that adding an amount of tar residue in a certain range is beneficial to the raise of coke yield. With a comparative analysis of crucible coke and small coke oven experiment results, the coke yield of industrial coke ovens is relatively bigger. Figure 6 and Figure 7 respectively reveal the impact of of adding tar residue on coke reactivity and post-reaction strength, and it is evident that a 3% of tar residue addition in coal blending for coking brings the best coke reactivity and coke strength after reaction, and the experiment results of industrial coke ovens are even better. In the process of coal coking, the yield rate of coal tar and gas cannot be overlooked, which is relevant to the nature of coke. The higher the coal tar and gas yield rate, the higher the coke quality is, and thus could better satisfy current needs for high-quality coke. Figure 8 displays the impact of the adding amount of tar residue on coal tar yield rate, which shares the same pattern with the change of coke reactivity and post-reaction coke intensity, meaning that a 3% of tar residue addition brings the biggest coal tar yield rate. Since the coal tar has a high industrial value, and therefore, it could bring greater economic benefits to the enterprises. Besides, Figure 9 shows the influence of the tar residue addition on gas yield rate, adding tar residue brings little impact on gas yield rate, which again proves the feasibility of coal blending for coking.



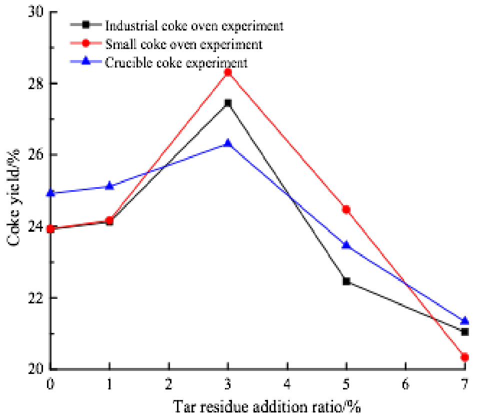
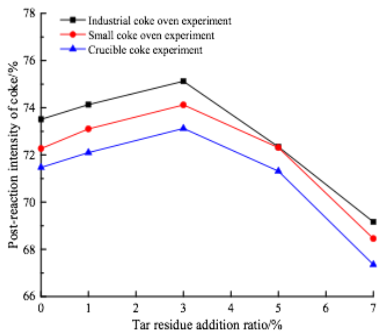
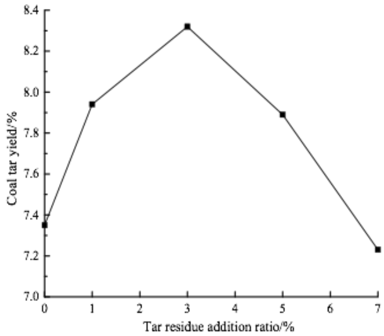


Figure 5: Effect of adding tar residue on coke yield *Figure 6: The effects on coke reactivity add tar residue*





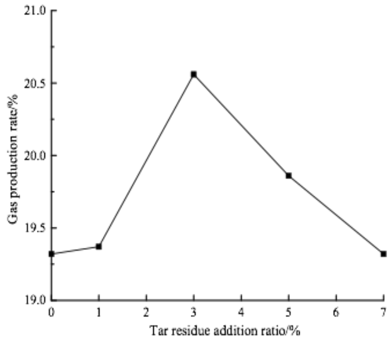


Figure 7: The impact of strength after reaction of coke add in tar residue; Figure 8: The effect of the amount of adding coal tar on tar residue yield 69; Figure 9: The effect of adding the amount of coal tar residue yield

5. Conclusion

In this paper, tar residue is used as additive in the coal blending for coking experiment, with the crucible coke, small coke oven, and industrial coke oven experiments, the influences of the addition amount of tar residue on coke yield, coke reactivity and post-reaction coke intensity are further analyzed, which lead to the following conclusions:

(1) The mixed coal should have low ash content and sulfur content, proper volatile matter content, as well as a good bond index. By increasing the bulk density and the temperature, improving the coal properties, the coking process and coke post-treatment process of the coke oven, coke quality could be significantly improved and better tar residue obtained.

(2) The tar residue added within a certain amount is conducive to the improvement of coke production rate. Compared with the experimental results of crucible coke and small coke oven, the coke production rate of industrial coke oven is much larger.

(3) A 3% of tar residue addition in coal blending for coking results in the best coke reactivity and post-reaction coke intensity, and the experimental effect of industrial coke oven is even better, and therefore the industrial production of coal blending for coking can be realized.

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