



Effects of iron compounds on pyrolysis behavior of coals and metallurgical properties of resultant cokes

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ABSTRACT

The utilization of highly reactive and high-strength coke can enhance the efficiency of blast furnace by promoting indirect reduction of iron oxides. Iron compounds, as the main constituent in iron-bearing minerals, have aroused wide interest in preparation of highly reactive iron coke. However, the effects of iron compounds on pyrolysis behavior of coal and metallurgical properties of resultant cokes are still unclear. Thus, three iron compounds, i. e., Fe_3O_4 , Fe_2O_3 and $\text{FeC}_2\text{O}_4 \cdot 2\text{H}_2\text{O}$, were adopted to investigate their effects on coal pyrolysis behavior and metallurgical properties of the resultant cokes. The results show that iron compounds have slight effects on the thermal behavior of coal blend originated from thermogravimetric and differential thermogravimetric curves. The apparent activation energy varies with different iron compounds ranging from 94.85 to 110.11 kJ/mol in the primary pyrolysis process, while lower apparent activation energy is required for the secondary pyrolysis process. Iron compounds have an adverse influence on the mechanical properties and carbon structure of cokes. Strong correlations exist among coke reactivity, coke strength after reaction, and the content of metallic iron in cokes or the values of crystallite stacking height, which reflect the dependency of thermal property on metallic iron content and carbon structure of cokes.

1. Introduction

The sustainable development of iron and steel industry is now facing some challenges from severe environmental pressure and depletion of high quality coking coal reserves in some developing countries, especially in China. The blast furnace (BF) iron-making process, as the largest energy consumer, accounts for more than 60% of total energy consumption in iron and steel industry. Therefore, reducing fuel consumption and CO_2 emission in BF process has become an important task. Increasing the utilization rate of CO in furnace by using highly reactive coke can be an effective way to achieve this goal. Some researchers have confirmed that the utilization of the highly reactive coke can obviously promote the shaft efficiency and decrease fuel consumption owing to its lower initial reaction temperatures with CO_2 , which also provides favorable thermodynamics and kinetic conditions for reduction reactions between reductants and iron oxides in BF^[1-4].

For preparation of highly reactive cokes, previous

studies indicated that the alkaline-earth metals^[1], alkali metals^[2] and transition metals^[5] present a strongly positive catalysis effect on the coke gasification with CO_2 . However, alkali metals and alkaline-earth metals generally lead to the erosion of furnace lining and hearth^[6]. Different from that, the iron compounds have received more attention because they do not have negative impacts on the BF smelting process and can be easily obtained from iron-bearing minerals. Application of the iron coke containing 43% iron was found to lead to an increase of 6.8% in shaft efficiency of BFs^[7]. Furthermore, while no direct relation between the coke reactivity and total iron (TFe) content was found, the positive influence of iron compounds on the gasification of coke was manifested using more detailed phase analysis including metallic iron, Fe_3O_4 and Fe_2O_3 ^[8-10]. However, the relationship between iron compounds and coal carbonization has been seldom reported. Coal pyrolysis is a fundamental process in the carbonization and has a significant influence on the structure, physical and chemical properties of the

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resultant coke^[11,12]. Many factors, such as rank of coal, particle size and composition, temperature, time and heating rate, were found to affect the pyrolysis and further the structures and physiochemical properties of resultant coke^[13–18]. Many literatures reported the effects of iron compounds on coal pyrolysis. Cypres and Soudan-Moinet^[19,20] concluded that Fe₃O₄ and Fe₂O₃ have a positive influence on coal pyrolysis by decreasing coal reactivity. Yet, the relationship between iron compounds and kinetics of coal pyrolysis also requires further investigation.

In this study, the pyrolysis of coal blend was investigated experimentally using three iron compounds by means of thermogravimetric (TG) analysis and differential thermogravimetry (DTG). The effects of iron compounds on thermal behavior of coal blend were revealed and the kinetic parameters of pyrolysis including the apparent activation energy and pre-exponential factor were calculated by applying a non-isothermal kinetic model. Furthermore, the effects of iron compounds on the properties of resultant cokes and their mutual relationship were also studied.

Table 1

Main characteristics of individual types of coal used in experiment (%)

Coal	Proximate analysis					Ultimate analysis				\bar{R}_0
	M _{ad}	VM _d	A _d	FC _d	C _d	H _d	N _d	S _d	O _d	
YH	2.38	26.90	10.33	62.77	75.01	4.52	1.76	2.01	6.38	0.95
NT	0.89	21.31	12.03	66.66	76.82	4.16	1.21	0.51	5.27	1.03
SM	2.85	18.22	11.98	69.80	76.13	3.78	0.93	0.54	6.65	1.80

Note: M—Moisture; VM—Volatile matter; A—Ash; FC—Fixed carbon; Subscript ad—Air dry; Subscript d—Dry; \bar{R}_0 —Mean reflectance of vitrinite.

BC-FeC₂O₄ coke, BC-Fe₃O₄ coke and BC-Fe₂O₃ coke, respectively.

2.2. Pyrolysis tests and non-isothermal kinetics analysis

The pyrolysis tests of the coal blend with different iron compound additives were carried out using a thermogravimetric analyzer (SETARAM SETSYS Evolution 16/18, France). Typically, approximately (7 ± 2) mg sample was placed in a ceramic crucible and heated from room temperature to 1050 °C with a heating rate of 10 °C/min. To prevent oxidation, the tests were conducted in N₂ atmosphere at a flow rate of 50 mL/min. The weight of samples was recorded automatically.

In this work, several specific temperatures related to the thermal degradation of coal were defined, as shown in Fig. 1 and listed below^[24]: T_{\max} is the temperature of the maximum volatile evolution obtained from the strongest peak of DTG curve, while T_i and T_f are the initial and final temperatures of primary pyrolysis reaction resulted from the point of inter-

2. Experimental

2.1. Preparation of samples

A typical coal blend composed of three kinds of coal samples, 50% Yonghun coal (YH), 40% Nantong coal (NT) and 10% Shoumei coal (SM), was used and homogenized in a laboratory-scale mixer for subsequent tests. The particle sizes of the selected coal samples ranged from 0.5 to 3.0 mm. The proximate and ultimate analyses of the three types of coals are presented in Table 1. Three chemical pure reagents of iron compounds (purity: approximately 98%), FeC₂O₄ · 2H₂O, Fe₃O₄, and Fe₂O₃, were chosen as additives. The iron compounds can be ranked as Fe₃O₄ > Fe₂O₃ > FeC₂O₄ · 2H₂O according to their TFe content. The iron compounds and coal blend were uniformly mixed in the ratio of 3 to 100 as reported in previous studies^[21–23]. In this study, the pyrolysis tests of the coal blend and different additives were labeled as BC-RAW, BC-FeC₂O₄, BC-Fe₃O₄, and BC-Fe₂O₃, and the relevant resultant coke samples were described as BC-RAW coke,

section between the tangent line which passes the point of T_{\max} and the slight weight loss at the beginning (T_A) and end (T_B) of coal pyrolysis. The beginning and end of coal pyrolysis can be confirmed by the method of least significant difference and described as the following equations:

$$G_A = \min\left(\frac{G_n - G_{n+1}}{T_{n+1} - T_n}\right) \quad 1 \leq n \leq 166, n \in N^*, 200^\circ\text{C} \leq T_n \leq 478^\circ\text{C} \quad (1)$$

$$G_B = \min\left(\frac{G_n - G_{n+1}}{T_{n+1} - T_n}\right) \quad 166 \leq n \leq 510, n \in N^*, 478^\circ\text{C} \leq T_n \leq 1050^\circ\text{C} \quad (2)$$

where, G_A and G_B are the minimum values of the weight loss per unit temperature at the beginning and end of coal pyrolysis, respectively; T_n is the temperature with the counting interval of 10 s; and G_n is the weight loss per unit temperature at counting interval of 10 s. Measurements started at 200 °C to avoid the influence of water (from moisture) on the results.

The conversion rate X resulting from the weight loss of the coal samples pyrolysis is shown as follows^[25]:

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