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RELATIONSHIP BETWEEN COKING PROPERTIES OF LUMP COAL AND ITS PULVERIZATION IN COREX PROCESS

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Abstract

Experimental work was conducted to determine the relationships of coal char conversion with temperature and heat absorption. The results show that at an earlier stage of the coke forming process, lump coal would absorb large amounts of heat but the increase of high temperature strength of lump coal was not significant. Contrarily, the lump coal's high temperature strength was greatly improved at later stage of coke forming process. A number of residual coal was found in most tuyere coke samples illustrating that the lump coals added in the melter gasifier were not completely transformed into coke when they arrived at the tuyere region. The results indicate that the improvement of lump coal's high temperature performance is limited in the melting gasifier by the low coke forming rate which could become the core factor of fines generation of lump coal in the melting gasifier.

Introduction

Coal is used in COREX smelting reduction offering a superior environmental performance compared with the coke process[1)]. The coke's function such as heat source、reducing agent and carburization could be replaced by the coal added in COREX, but the replacement of the coke's skeleton pillar is still needed more development. Since the running of COREX C3000 in BAO-Steel, the high powder ratio of lump coal was always the most important limiting factor for COREX production. Though the measures such as changing production parameters 、coal blending ratios and size of coals were taken to improve the production, the problem of high powder ratio of lump coal was unresolved. According to the operational data from BAO-Steel, the average powder ratio of lump coal reached 60%, even the dead zone's powder ratio was more than 80%. These results made the experts of BAO-Steel to reconsider the fuel structure of COREX[1-3)].

Experience on the blast furnace, shows that the heat conduction of coke could become the limiting step for iron-making when the coke's radius is more than 60-70mm[4]. In addition, main differences between lump coal and coke can be summarized as follows:

(1) The thermal conductivity of lump coal is smaller than that of coke, so the thermal ability of coal is inferior compared with coke.

(2) Coal would absorb large amount of heat during the coke forming process in melting gasifier.

(3) The caking coal used in COREX was easy to bind together during the coke forming process and thus the heat transfer was hindered by the bigger massive coal char.

It has also been observed that lump coal tends to move towards the high temperature region over 800 ℃ immediately when it is added to the top of melting gasifier enhancing a fast pyrolysis of lump coal and precipitation of tar from the lump coal. This phenomena would lead to the blockage of gas pipeline and poor reliability of equipment[5]. The reduction process would also be influenced by the precipitated tar.

Therefore, COREX coal has been selected to investigate the transformation process of the lump coal at high temperature (1000 ℃).

Experiment

Experimental method

Table 1 shows the fundamental analysis of coal used in COREX process:

Table 1 Analysis of coals (wt.%)

Proximate analysis				Elemental analysis				Caking index
FC_d	A_d	V_d	M_{ad}	C_d	H_d	N_d	O_d	$G_{R,I}$
60.35	7.71	31.94	2.84	72.89	4.87	0.96	13	55

Lump coals (20mm) weighed about 200g used in COREX were put in a self-made stainless container which was connected to a device for tar collecting. N_2 was injected into the stainless container about 30min to exclude the air in it, then sealed the container except for remaining a gas pipeline connected to the tar collecting device.

In order to simulate the rapid heating condition prevalent in the melting gasifier, an empty muffle furnace was first heated to 1000 ℃. Then a stainless steel container equipped with coal samples was quickly introduced into the furnace for coke–making.

After a period of time, the container was taken out and placed in a drying oven to cool it down. The steps above were repeated by setting the heating time respectively at 30min, 45min, 60min, 90min, 120min, 150min, 180min, 240min. The tar precipitated from lump coal and the residual coal char were weighed by an electronic balance, and the surface texture and microstructure of the residual coal char were examined by optical microscope and SEM. Finally, the coal char's reactiveness and the strength after reaction were measured as well as the density and volume of different coal chars.

Results and Discussion

Table 2 shows the analysis of different coal chars' microstructure (minerals and impurities were not listed). Here, a new index named coal char conversion ω was defined as:

$$\omega = \frac{\sum_{i=1}^{n} \mu_i}{\sum_{i=1}^{n} \mu_i + C} \qquad (1)$$

Where $\sum_{i=1}^{n} \mu_i$ was the proportion of the coke's optical structure, and μ_i represents one of the coke's optical structure such as isotropic、anisotropic and so on. C was the proportion of the residual coal.

In Table 2, the coal char conversion ω was calculated by Eq. (1).

Table 2 Analysis results of coal char's microstructure /%

Heating time/min	Coke							The residual coal	ω/%
	Fusain	Isotropic	Anisotropic	Fine mosaics texture	Medium mosaics texture	Coarse mosaics texture	Residual carbon particle		
30	0.00	2.90	16.23	0.00	1.16	0.00	2.03	75.70	22.77
45	0.23	6.13	5.19	3.21	1.32	0.32	6.23	72.04	23.90
60	3.29	10.18	2.66	2.95	2.36	0.56	7.89	62.32	32.42
90	0.79	18.10	1.05	2.83	3.40	0.95	7.71	51.98	40.12
120	0.11	28.73	1.29	2.41	6.91	1.11	8.09	32.88	59.67
150	0.15	32.10	12.30	0.00	2.20	2.30	12.30	34.60	63.94
180	0.90	52.45	1.24	2.91	9.24	1.29	6.32	12.35	85.76
240	0.33	44.19	17.94	5.65	3.30	10.10	13.29	0.33	99.65

The coal char's reactivity and the strength after reaction were respectively defined as MCRI and MCSR, and the detection methods of MCRI and MCSR were the same as the coke's CRI and CSR. The relationships of the coal char conversion ω with MCRI, MCSR, weight loss G and tar yield are presented as in figure 1, figure 2 , figure 3 and figure 4, respectively.

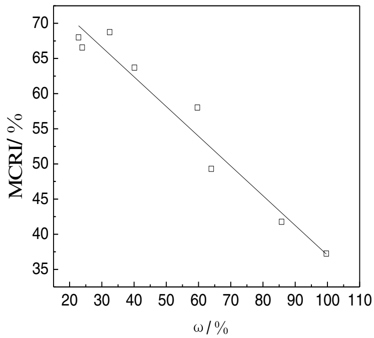

Fig.1 Relation between ω and MCRI

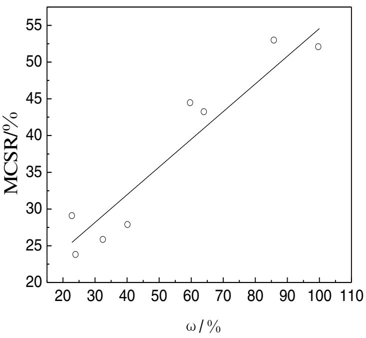

Fig.2 Relation between ω and MCSR

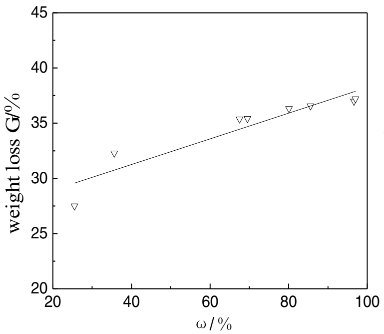

Fig.3 Relation between G and ω

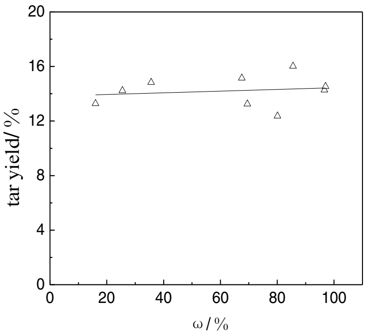

Fig.4 Relation between ω and tar yield

Relation Between Coal Char's Performance and Temperature

Calculations

A large temperature gradient would appear in the internal lump coal when suddenly added to a high temperature environment. While the coal char conversion and high temperature performance of lump coal would be affected by the variation of the internal temperature or heat absorption.

The analysis or numerical solutions of heat conduction differential equation are usually used to calculate the temperature distribution of coke oven, and the coal in coke oven is assumed to be semi-infinite plate[4]. However, the conditions in this experiment were not the same as in a coke oven, and therefore it was more appropriate to treat coal particles as spherical. The radius of lump coal is R and the initial temperature T_0 is 20℃. The lump coal was placed in a high temperature furnace whose temperature T_f is always 1000 ℃, The heat is transferred through the stainless container between the coal char and the air in the high temperature furnace. The thermal conductivity λ of stainless steel[10] is about 16 W/(m·K) which was much higher than the coal. Regardless of heat resistance of the stainless container, besides, it was assumed that the temperature of air in the high temperature furnace was kept at 1000℃.

Basic physical characteristic parameters[9-11]: when $T \le 673K$, $\lambda = 0.23$ W/(m·K), when $T > 673K$, $\lambda = 0.23 + 2.24 \times 10^{-5}(T-673)^{1.8}$ W/(m·K); when $T \le 623K$, $c_p = 1254$ J/(kg·K), when $T > 623K$, $c_p = 1254 - 1.75(T-623)$ J/(kg·K); the equivalent radius of the lump coal $R = \sqrt[3]{\frac{3m}{4\rho\pi}}$, the weight and density of the solid products were measured by experiment; the heat transfer coefficient α of air is about 5-10 W/(m^2·K) under the condition of natural convection, the test was performed in the stationary air, then the interfacial heat transfer coefficient between the air and solid products was set as α=8 W/(m^2·K).

Table 3 shows the experimentally determined densities for the obtained solid products.

Table 3 Density of different coal chars

Heating Time/min	30	45	60	90	120	150	180	240
Density ρ/kg/m^3	1056	980	863	802	745	735	710	702

For one-dimension unsteady heat conduction of sphere, the following relationship is established[11]:

$$\frac{\partial(\rho c_p T)}{\partial t} = \frac{1}{r^2}\frac{\partial}{\partial r}\left(\lambda r^2 \frac{\partial T}{\partial r}\right) + q_V \qquad (2)$$

Where ρ stands is the denstiy density, r is the sphere radius, c_p the heat capacity at the constant pressure, and q_V the strength of internal heat source, W/m^3.

Initial conditions : $T(r,0) = T_0$; Boundary conditions : $\frac{\partial T(r,t)}{\partial r}\Big|_{r=0} = 0$, $-\lambda \frac{\partial T(r,t)}{\partial r}\Big|_{r=0} = \alpha[T_f - T(R,t)]$

The first item in the infinite series of the analytical solution was only singled out for convenient calculation.

$$\frac{\theta(x,t)}{\theta_0} = 2\frac{\sin\mu_1 - \mu_1\cos\mu_1}{\mu_1 - \sin\mu_1}\exp(-\mu_1{}^2 Fo)\frac{1}{\mu_1 x}\sin(\mu_1 x) \qquad (3)$$

The curves (nomogram) of $\frac{\theta_m}{\theta_0}$(x=0) and $\frac{\theta}{\theta_m}$ change with Fo and Bi can be obtained from equation (3), and the relationship as formula (4) was adopted for each point in the sphere:

$$\frac{\theta}{\theta_0} = \frac{\theta_m}{\theta_0}\frac{\theta}{\theta_m} = \frac{T_m(t)-T_f}{T_0-T_f}\cdot\frac{T-T_f}{T_m(t)-T_f} = \frac{T-T_f}{T_0-T_f} \qquad (4)$$

Thus the temperature of different points in the sphere can be obtained by nomogram.

Figure 5 shows the distribution of internal temperature of coal char at different heating times.

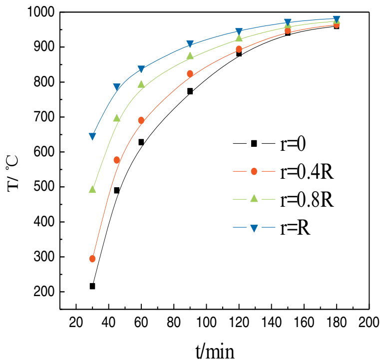

Fig.5 Distribution of internal temperature of coal

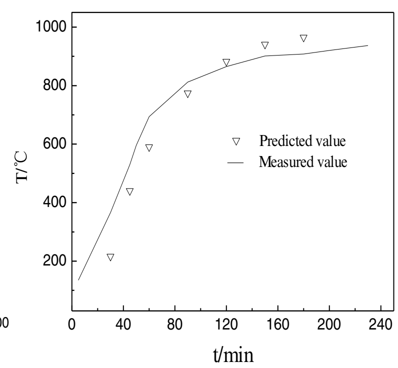

Fig.6 Measured and calculated value of coal's center temperature

Figure 6 shows the measured and estimated temperature at the centre of the coal samples. Experimental temperature was measured by a thermocouple plugged in the center of coal samples through the gas export of stainless steel container. When the empty muffle furnace was heated to 1000 ℃, the coal samples with thermocouple were quickly pushed into the furnace.

Fig 7 shows the heat absorption of coal char at different heating time.

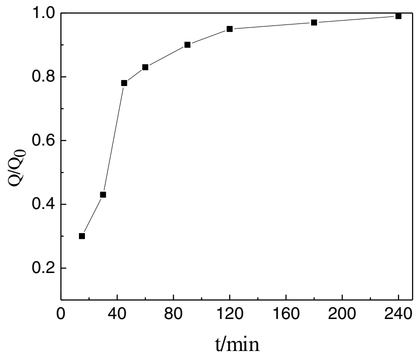

Fig.7 Heat absorption of coal char at different heating time

It can be seen that the endothermic of lump coal was concentrated on the prophase of coke forming, and the absorbed heat of the top 1/3 phase could account for 80% of total heat absorption, and the heat absorbance rate at the prophase is also higher than that at the anaphase as shown in Fig 7

Discussion

The lump coal fall from the top of COREX Melting Gasifier to the moving bed rapidly and stay in the furnace for only around three hours. The average temperature of lump coal in the furnace is about 1000 ℃[3,5]. It can be inferred from the test results that the lump coal will not transform into coke completely. Part of the lump coal may be consumed as coal in the upper-middle part of the furnace while the remaining part of lump coal will partially transform into coke and then reached front of the tuyere for combustion. In this case, the slow coke forming rate will lead to the serious chalking of the lump coal.

Conclusions

The most important restriction of COREXC3000 was the fines generation of lump coal in the melting gasifier. In this paper, the variation of high temperature performance and the cause for fines generation of lump coal were clarified through the simulation test and theoretical calculations. An important reason for the fines generation was that the low coke forming rate could not meet the requirements of coal's high temperature performance in the melting gasifier which was confirmed by detecting the tuyere coke samples. The measure of preheating the lump coal before it was added to the melting gasifier could be taken. The preheating temperature could be set to 700℃~ 800℃, with the heating rate at 20℃/min and the preheat time of 1h. Then the coal char conversion could be ensured by 30%, and the restriction of heat transfer could be dramatically reduced, even eliminated.

The internal temperature and heat changes of the lump coal were calculated by unsteady heat conduction equation. Then the relationship of the coal char conversion with average temperature, absorbed heat and high temperature performance was given by the experiment. Moreover, the effect of heat transfer process on the performance of the coke formed from lump coal was analyzed. According to the results, lump coal would absorb large amounts of heat in the early stage of coke forming process. However, the increase of high temperature strength of lump coal was not significant at early stage which leads to the fines generation of lump coal. By controlling the stage of coke forming, the coking rate could be increased and thus the high temperature performance of coal char was improved. So the fines generation of lump coal could be reduced, and the precipitation of coal tar could also be effectively controlled.

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