



Sticking mechanism of low grade iron ore-coal composite in rotary kiln reduction

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ABSTRACT

A rotary kiln ($\Phi 1.5 \text{ m} \times 15 \text{ m}$) was designed to verify feasibility of the reduction technology of a low grade iron ore with low temperature via ore-coal composites. One of the troublesome problems was the sticking of kiln burden during the reduction process. Reduction behavior and sticking mechanism of composite pellets in the rotary kiln were investigated in this paper. Technology of X-ray diffraction (XRD) and scanning electron microscope-energy dispersive spectroscopy (SEM-EDS) were applied to reveal the sticking mechanism. Products with metallization degree of 84.83% were obtained after reduced in the rotary kiln. Reduction of Fe_2O_3 to FeO and reduction of FeO to Fe were mainly occurred at location of 9 m–13.5 m and 5.5 m–11.5 m in the rotary kiln, respectively. Pellets or pellets powders were bonded by low melting point substances formed in coal ash: hedenbergite and fayalite. These low melting point substances were mainly generated from coal ash and formed at location of 5.5 m–11.5 m in the rotary kiln. Hedenbergite occurred firstly, followed by bonding of pellets or pellet powders and formation of fayalite. Melt of fayalite intensified agglomerating behavior.

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1. Introduction

Beneficiation of low grade hematite ore resource (with high SiO_2 content) has puzzled the mineral trade for several decades for its fine disseminated grain size but complex association relationship [1,2]. The total reserve of such iron ore in China is up to 30million tons. Effective utilization of this resource may bring significant benefits to the ironmaking industry in China. Our research group has investigated such iron ore for many years, and the method of low temperature reduction approach via ore-coal composite pellets followed by magnetic separation was proposed [3–5]. Reduction products with metallization degree higher than 70% was obtained when reduced at 950°C – 990°C for 35 min–60 min. Based on the success of laboratory investigation, a rotary kiln ($\Phi 1.5 \text{ m} \times 15 \text{ m}$) was specially designed to verify feasibility of this low temperature reduction technology.

Sticking of iron ore pellets happened frequently during reduction process. When reduced by CO, formation of fibrous metallic iron and fresh metallic iron with high activities might be the reason for the sticking of iron ore pellets during reduction process. While sticking extent decreased with addition of H_2 [6]. Sticking of iron ore-coal composite pellets reduction may attribute to the intergrowth of metallic iron or ferrous oxide crystals between boundaries, and high melting point substances formed by MgO and Fe_xO_y can effectively prevent such

intergrowth [7]. Coal-fired rotary kiln is widely applied to iron ore pellets and direct reduction iron (DRI) production in China [8]. However, one of the limits of this technology is inevitable ring formation or deposit formation in the kiln. It makes the quality of pellet declining, production efficiency reducing, and production cost increasing [9,10]. Ring formation or deposit formation in the rotary kiln could be consider as the large scale sticking of materials (mainly iron ore pellets and coal) in the rotary kiln. Mechanism of ring formation in oxidized pellet produce rotary kiln has been intensively researched [11–15]. Iron-ore pellet powder and coal ash which was generated by coal combustion were thought to be the raw materials in ring formation process [11]. Recrystallization of Fe_2O_3 between particles may cause bonding of pellet powders, and coal ash could facilitate the bonding behavior for the formation of liquation phase (aluminosilicate) or liquid phases (low melting substance) [12–14]. The deposit formation process in a pulverized coal fired grate-kiln may attribute to hematite pellet powders and residual carbon in coal ash, among which hematite was reduced to wustite by the residual carbon and lead to low melting point phases formed. Those low melting point substance transferred into liquid phases and therefore caused deposit formation [16,17]. The phases of deposition formation are composed of Si, Al, Fe, Ca and O [9,18,19]. Even though considerable researches were focused on reveal sticking or ring formation behavior of pellets during reduction process, mechanisms are still not completely understand for the complexity of reactions and variability of raw materials, especial for coal-fired kiln. Otherwise, investigation obtained from a rotary kiln scale rather than laboratory scale can reveal the sticking behavior more effectively.

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Table 1
Chemical analysis of iron ore/wt%.

T.Fe	SiO ₂	Al ₂ O ₃	CaO	MgO	P	S	K ₂ O
33.86	40.61	4.36	1.57	1.05	0.38	0.082	1.58

In this paper, commercial experiment of iron ore-coal composites reduction in a rotary kiln ($\Phi 1.5 \text{ m} \times 15 \text{ m}$) was conducted to utilize a typical low iron grade but high SiO₂ content iron ore. Reduction behavior of composites in the rotary kiln was investigated. Chemical component and mineral phase of agglomerations were analyzed. Technology of XRD (X-ray diffraction) and SEM-EDS (scanning electron microscope-energy dispersive spectroscopy) and thermodynamic software (FactSage7.0) were applied to observe the mechanism of sticking behavior.

2. Materials and methods

2.1. Materials

The iron ore with a low iron grade but high SiO₂ was obtained from Qidong area (Hunan, China). As shown in Table 1, the total iron content (T.Fe) was 33.86 wt%, while the content of SiO₂ was 40.61 wt%. Other constituents included 4.36 wt% Al₂O₃, 1.57 wt% of CaO, and 1.05 wt% of MgO. XRD pattern (Fig. 1) shows that hematite, quartz, chlorite and sericite constituted the main phases of this ore. The microstructure image of the ore (Fig. 2) shows that hematite grains presented extremely complex embedded characters with gangue substrate. Reducant coal used in this research was a bituminous, produced in Shanxi province China. Proximate analysis of the coal and chemical analysis of coal ash are shown in Table 2. Results show as follows: 58.53 wt% fixed carbon, 32.01 wt% of volatile matter, 11.32 wt% of ash.

Coal ash was mainly composed of 24.52 wt% Fe₂O₃, 36.82 wt% SiO₂, 13.08 wt% CaO and 6.74 wt% Al₂O₃. XRD analysis (Fig. 3) reveals that mineral phases of ash were mainly hematite, quartz, lime and corundum.

2.2. Methods

Both the raw ore and coal were crushed (PE60 $\times$ 100 mm crusher, Zhengzhou Yiju Poly Machinery Equipment Co., Ltd., Henan, China) to -1 mm and then milled (ZQM-150 $\times$ 50mm ball milling, Zhengzhou Yiju Poly Machinery Equipment Co., Ltd., Henan, China) into powders for the pelletizing procedure.

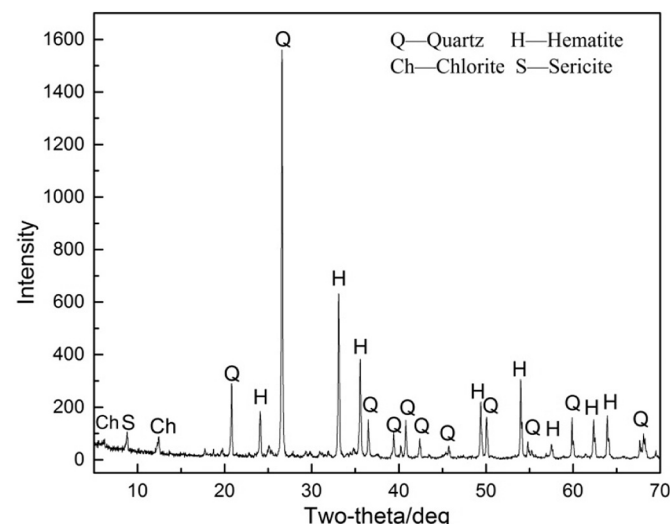


Fig. 1. X-ray diffraction pattern of the iron ore.

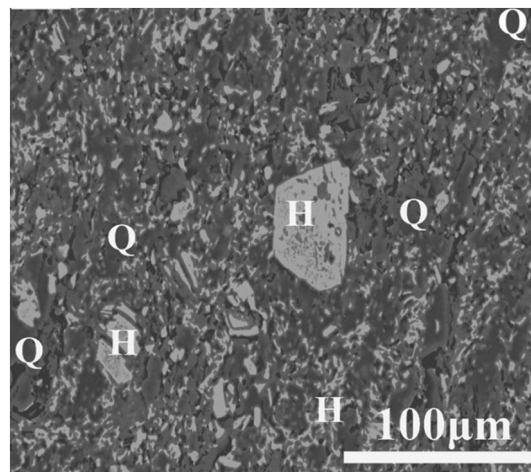


Fig. 2. Micrograph of iron ore.

Iron ore-coal composite pellets were made for the rotary kiln reduction purpose. Mixtures of raw iron ore, coal, NaCl (used as additive) [3] and bentonite (used as a binder) were pelletized into a size of 5–8 mm composite pellets. A rotary kiln ($\Phi 1.5 \text{ m} \times 15 \text{ m}$) was built for the reduction process, as shown in Fig. 4. Composite pellets were fed into the rotary kiln from the rotary kiln tail and reduced pellets were discharged from the rotary kiln head. Coal powders were injected into the rotary kiln by an injection gun fixed at the kiln head. Combustion-supporting secondary air was blow into the rotary kiln through five secondary air pipeline set in the rotary kiln. There were seven thermocouples to measure the temperature of the rotary kiln, which were distributed from the rotary kiln head to the rotary kiln tail. Temperature distribution of the rotary kiln is shown as Table 3. It is indicated that high temperature zone distributed in 3# thermocouple and 4# thermocouple. Duration of stay for pellets in the rotary kiln was 100 min.

Reduced pellets discharged from the rotary kiln head were selected to analysis reduction result. Besides, to investigate the reduction process of composite pellets in the rotary kiln, rotary kiln was stopped at normal operation conditions without discharging materials. And then, plenty of N₂ was inlet to the rotary kiln to prevent oxidation of reduced pellets. As rotary kiln was cooled down to environmental temperature, sample pellets were collected every 0.5 m along the rotary kiln.

Among products discharged from the rotary kiln head, agglomerations consist of pellets or pellet powders and other materials were found. Therefore, several agglomerations discharged at the same time with normal reduced pellets were collected to research the formation mechanism.

X-ray source X-ray diffraction (XRD, D/max 2550PC, Rigaku Co., Ltd., Tokyo, Japan), was used for mineral phase identification by configurations of the step of 0.02° at 10° min⁻¹ ranging from 10° to 80°. Morphological analysis and micro-zone chemical composition of the samples were analyzed by scanning electron microscope equipped with an energy diffraction spectrometer (SEM-EDS, JSM-6510A, JEOL, Tokyo, Japan). The acceleration voltage of the SEM was 30kv. Besides, a thermodynamic software FactSage7.0 (Thermfact/CRCT (Montreal, Canada) and GTT-Technologies (Aachen, Germany)) was applied to obtain the phase diagrams [19]. Thermodynamic results were used to analyze the formation mechanism of the deposits in the grate.

3. Results and discussions

3.1. Reduction of composite pellets in rotary kiln

Chemical composition and XRD analysis of reduced pellets are shown in Table 4 and Fig. 5, respectively. Reduced pellets were mainly composed of metallic iron, quartz and residual carbon. Metallization

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