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Kinetics and mechanisms of direct reduction of iron ore-biomass composite pellets with hydrogen gas

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ARTICLE INFO

Article history:

Received 26 December 2014

Received in revised form

13 February 2015

Accepted 14 February 2015

Available online 12 March 2015

Keywords:

Biomass

Ironmaking

Direct reduction

Kinetics

ABSTRACT

Direct reduction of iron ore pellets (DRI) with and without biomass was studied using hydrogen as the reducing agent. The influences of temperature and time on the reduction rate of pellets were investigated. X-ray diffraction (XRD), scanning electron microscopy (SEM) and BET (Bet specific surface area) tests were adopted to interpret the mechanism of Fe₂O₃ reduction with H₂, as well as explore the role of biomass in pellets during the reduction process. Results show that the biomass not only improve the reduction extent of the pellets, but also increase the reduction velocity index (RVI) of the pellets by 1.12%/min. The BET test indicates that the existence of biomass can create porosity in pellets by dehydration and combustion/pyrolysis, which increases the contact area between iron oxide and reducing gas during reduction process, and decreases the apparent activation energy of pellets from 122 kJ mol⁻¹ (without biomass) to 111 kJ mol⁻¹ (with biomass). For the DRI process, the crucial reduction step FeO → Fe is controlled by the intrinsic interfacial chemical reaction.

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Introduction

Direct reduction technology of iron ore is considered as a promising and clean production approach for ironmaking. In a typical direct reduction process, coal and natural gas are used as alternative fuels to replace metallurgical coke. However, there is no fundamental improvement in the environmental pollution control for the direct reduction technology,

especially concerning the carbon emissions, compared to the traditional ironmaking technology [1–3]. To respond to the growing energy shortage and greenhouse challenges, the requirement of incorporating renewable energy to the existing metallurgical process has become more and more urgent. Biomass is an ideal renewable energy with outstanding advantages, such as low content of harmful elements (S, P and others), natural abundant resources, as well as low level of overall net carbon emissions, which makes it a promising

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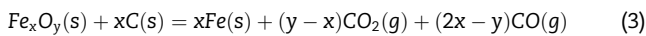
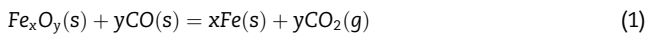
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<http://dx.doi.org/10.1016/j.ijhydene.2015.02.065>

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energy candidate in the direct reduction ironmaking to alleviate the environmental pollution.

Recently, a number of studies have been conducted to introduce biomass into metallurgical industries [4–10]. Strezov [4] investigated the fundamental mechanisms of iron ore reduction with biomass wood waste. Reduction tests were conducted using mixtures with different weight ratios of biomass and iron ore. The iron ore was successfully reduced to predominantly metallic iron phase when 30 wt.% of biomass was introduced into the mixture. Katyal et al. [5] and Emmerich et al. [6] investigated the possibility of metal smelting operations with charcoal produced from wood or other biomass materials. The results showed that the charcoal can substitute for coke successfully in the blast furnace. In these studies the reduction of metallic oxides was performed either using biomass in their natural form or char from biomass pyrolysis. The reduction process was driven by the carbonthermic Boudouard reaction:



However, the intensity of Fe–C reduction is weak. Poor kinetic conditions and low reduction efficiency limit the development of direct reduction ironmaking using biomass. To improve the reduction efficiency, Luo et al. [7] studied the possibility of introducing bio-gas produced from catalytic gasification of biomass into the direct reduction ironmaking as reducing agent. Instead of coke, biomass was mixed together with iron ore powder, water, limestone and bentonite to produce composite pellets. During the reduction process, the dehydration of the combined water and pyrolysis of the biomass make the pellets porous, leading to an increase in the contact area of iron oxide and reducing gas, as well as an improvement in heat and mass transfer. The results showed that at a reduction temperature of 1373 K and a reaction time of 60 min, the TFe content in the product reached the maximum of 94.7 wt.%. However, research about the reduction mechanism and kinetics laws in term of reducing gas and iron ore-biomass composite pellets have not been referred to and systematic studies are needed.

During the direct reduction process of ironmaking with bio-gas, the effective reducing agents are mainly CO and H₂.

The reactions of CO and H₂ with iron oxide are endothermic, which is in favor of the reduction of the pellets due to the effect of temperature field. Both CO and H₂ are ideal reducing agents for the process. However, compared to H₂, the reduction kinetics of CO is relatively poorer. Moreover, the generation of a large amount of greenhouse gas CO₂ from CO results in the oxidation of metallic iron in the porous surface to form nFeO·SiO₂ slag phase, subsequently leading to the decrease of direct reduction iron (DRI) quality.

In order to identify the optimal addition amount of biomass in pellets, a roasting process was conducted with biomass addition amount varying from 0.5% to 10% in an increment of 0.5%. It was found that the pellets with a 2%–3% weight fraction of biomass exhibited the properties that meet the industrial requirements. Therefore, all the samples used in the present study have a weight fraction of 2% biomass.

In order to explore the kinetics and mechanism of direct reduction process of ironmaking with bio-gas, the direct reduction of iron ore pellets with and without biomass to metallic iron was studied with H₂ as the reducing agent over a wide range of temperatures. The effect of temperature and time on the reduction rate of pellets was investigated. XRD, SEM and BET tests were used to analyze the reduction mechanism. The effect of biomass in pellets was also discussed in the present study.

Experiment and methods

Materials and facilities

Iron ore powder was supplied by the Wuhan Iron and Steel Corporation (Wuhan City, Hubei Province) and bentonite from the Ezhou Iron and Steel Corporation (Ezhou City, Hubei Province). The chemical compositions of the bentonite and iron ore powder are shown in Table 1. Pine sawdust, the biomass used in this study (see Table 1), was collected from the furniture factory of the Huazhong University of Science and Technology (Wuhan City, Hubei Province). The iron ore powder, bentonite, and sawdust were crushed into fine particles and sieved such that only particles less than 200 μm in diameter were accepted.

Iron ore powder, pine sawdust and bentonite (96%, 2% and 2% respectively) was rolled and pelletized with a φ1000 mm disc balling machine, and the green balls with particle size of 10–15 mm were taken as testing samples. The chemical composition of the pellets is shown in Table 2.

Table 1 – Chemical compositions of iron ore and bentonite and the proximate and elemental analyses of the biomass.

Sample	Chemical composition (%)								
	Fe	Si	Al	Mn	Ca	Mg	Ti	S	K
Iron ore	67.72	5.39	2.02	0.30	9.39	14.49	0.33	0.27	–
Bentonite	3.25	68.41	18.37	0.04	3.77	3.88	0.69	–	1.55
Biomass	Ultimate analysis (wt.%)								
	C		H	O			N	S	
	48.21		6.54	44.39			0.77	0.09	
	Proximate analysis								
	Higher heating value (MJ/kg)		Moisture content (wt.%)		Volatile matter (wt.%)		Fixed carbon (wt.%)		Ash (wt.%)
	16.83		7.78		73.06		16.33		2.83

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