

Energy, Resources and Environmental Technology

## Integration of coal pyrolysis process with iron ore reduction: Reduction behaviors of iron ore with benzene-containing coal pyrolysis gas as a reducing agent☆

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## ABSTRACT

An integrated coal pyrolysis process with iron ore reduction is proposed in this article. As the first step, iron oxide reduction is studied in a fixed bed reactor using simulated coal pyrolysis gas with benzene as a model tar compound. Variables such as reduction temperature, reduction time and benzene concentration are studied. The carbon deposition of benzene results in the retarded iron reduction at low temperatures. At high temperatures over 800 °C, the presence of benzene in the gas can promote iron reduction. The metallization can reach up to 99% in 20 min at 900 °C in the presence of benzene. Significant increases of hydrogen and CO/CO<sub>2</sub> ratio are observed in the gas. It is indicated that iron reduction is accompanied by the reforming and decomposition of benzene. The degree of metallization and reduction increases with the increasing benzene concentration. Iron oxide can nearly completely be converted into cementite with benzene present in the gas under the experimental conditions. No sintering is found in the reduced sample with benzene in the gas.

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## 1. Introduction

Iron and steel production is in continuous high demand as a result of the growth in the world economy. A large amount of coke is required as both reducing agent and fuel for iron production using the conventional blast furnace process. Metallurgy coke is produced from high grade bituminous coal. However, reserves of the high grade bituminous coal are limited, which account for only 6.2% of coal reserves in China [1]. The limited reserves and the high demand cause a high price of coking coal, in turn leading to high costs of iron production. In addition, the environmental issues associated with the coke production impose serious health and ecological concerns. Thus, there is an increasing interest in exploring new iron making approaches [2–4]. An alternative iron-making route is direct iron reduction through either natural gas or coal-based technology [3,4]. The coal-based technology has attracted considerable attention in China due to the lack of natural gas. In the coal-based technology, syngas (CO and H<sub>2</sub>) derived from coal gasification is used as a reducing agent for iron production.

Coal is the primary energy resource in China, which provides 67.5% of the total energy consumption in 2013 [5]. Low rank coals such as lignite and sub-bituminous coal are abundant, comprising up to 55% of

coal reserves [6]. These low rank coals contain high volatile matters. These volatiles can be released through pyrolysis and further converted into liquid fuels and gaseous products. In order to bridge the gaps between the demands and the supplies of oil and gas, various processes for the production of liquid fuel and gas from coal based on pyrolysis are being developed by many research institutions and coal companies in China [7,8]. However, over 50% of heavy components in the obtained tar not only increase the difficulty in further processing the tar, but also cause serious operation problems in downstream equipment such as pipe blocking. One option to overcome this problem is *in-situ* catalytic upgrading of tar to increase its selectivity to light liquids. Catalytic upgrading of tar has been studied by a number of researchers [9–11]. Many researches have shown that iron ore exhibits a high activity toward tar cracking/reforming [12–14]. Compared with the conventional nickel-based catalysts, iron ore is cheaper and abundant. Moreover, the reactivation procedure could be avoided since the deactivated and reduced iron ore can be used as the feedstock of steel making industry.

In view of the abovementioned facts, an integrated coal pyrolysis with iron ore reduction was proposed in order to obtain high yields of light liquids and gas [15]. In the proposed scheme, heavy components of tar in the gas are decomposed/reformed into light species and gas over the iron ore. Simultaneously, the tar-containing gas from pyrolysis of coal is used as a reductant in the production of iron. In the proposed scheme, the iron reduction behaviors could be different from those using natural gas or syngas (CO and H<sub>2</sub>) as a reductant [3] since the reductant employed in the proposed scheme contains a large amount of

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tar. It is known that low rank coal contains high volatile matters, which are released in the form of incondensable gases and tar during pyrolysis. The tar content is around 30 wt.% or even higher in the pyrolytic gas from low rank coal [8,16]. However, research on the effects of tar on iron reduction especially for the case in the presence of a large amount of tar is rare.

As the first step, the objective of this work is to understand the reduction behavior of iron ore in the presence of a large amount of tar when using coal pyrolysis gas as a reduction agent. The reduction behavior of iron oxide is studied in a lab-scale fixed bed reactor using a simulated coal pyrolysis gas with benzene as a model compound of coal tar. Effects of reduction temperature, reduction time and benzene concentration on iron oxide reduction are discussed. The experiments using the simulated gas without the presence of benzene is also performed for comparison. Various characterization techniques such as X-ray diffraction (XRD) and scanning electron microscopy (SEM) are employed.

## 2. Experimental

### 2.1. Apparatus and operation conditions

The experiments were performed in a lab-scale fixed bed reactor as shown in Fig. 1. The reactor is quartz glass tube with an inner diameter of 25 mm and a length of 800 mm, which is placed in an electric-heated oven consisting of four silica carbon rods. The reactor temperature is measured by a chromel–alumel thermocouple. Benzene is fed into the reactor through a syringe pump. The simulated coal pyrolysis gas is introduced into the reactor at the top where it is mixed with benzene and its flowrate is regulated by a mass flow controller. The exit of the reactor is connected to two cold traps immersed in an ice–water bath to recover the benzene residues. The exhausted gas is collected with a gas sample collection bag, which is analyzed with a micro-gas chromatograph (CP-4900, Varian).

The composition of coal pyrolysis gas is related to coal type, operation conditions and reactor configuration. The coal pyrolysis gas obtained at low temperatures mainly consists of hydrogen, carbon monoxide, carbon dioxide and methane, which generally accounts for 85 vol%–95 vol% [17,18], with the rest is C2 and C3 with a very low amount. The composition of the simulated coal pyrolysis gas prepared in this work is shown in Table 1, which is made by referring to the gas composition from sub-bituminous coal pyrolysis at low temperatures [19]. The

**Table 1**  
Composition of the simulated gas

Gas	H <sub>2</sub>	CO	CH <sub>4</sub>	CO <sub>2</sub>	C <sub>2</sub> H <sub>6</sub>	C <sub>2</sub> H <sub>4</sub>	Total
Composition (by volume)/%	15	18	30	25	7	5	100

iron oxide sample used in the experiments is pure, purchased from Xilong Chemical Cooperation, Ltd. About 2.9 g sample of iron oxide are placed in the reactor for each test. The reduction temperature is set at 700, 800 and 900 °C. The simulated pyrolysis gas is fixed at a rate of 77 ml·min<sup>-1</sup>. The feeding rate of benzene is fixed at 0.05 ml·min<sup>-1</sup> (corresponding to 573 g·m<sup>-3</sup>) if not specified. The reduction time is controlled at 10, 20, 40 and 60 min, respectively. Nitrogen is used as purge gas during heat-up and cool-down to protect the sample from oxidation by air. The tests without benzene addition were carried out as a reference base for investigating benzene effect.

### 2.2. Analyses of iron oxide reduction

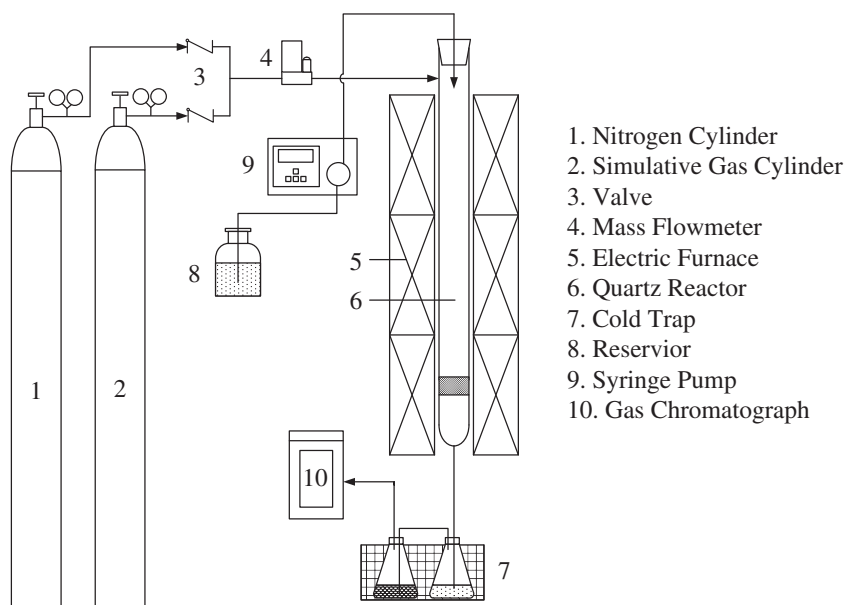
The quality of sponge iron is primarily ascertained by the degree of metallization, which is defined as follows:

$$\text{Degree of metallization} = \frac{\text{mass of metallic iron (free iron and iron combined with carbon)}}{\text{mass of total iron}} \times 100. \quad (1)$$

Both the metallic iron and the total iron are determined through the titrimetric method based on National Standard GB 223.7–2002 of China. Another index normally used to measure the extent of iron oxide reduction is the degree of reduction, expressed as follows:

$$\text{Degree of reduction} = \frac{\text{oxygen removed by reduction}}{\text{oxygen content before reduction}} \times 100. \quad (2)$$

The amount of oxygen removed by reduction can be calculated through the weight loss of the sample after reduction. Due to the presence of benzene in the gas, a large amount of carbon may deposit on the sample during reduction, which partially offsets the weight loss caused by oxygen removal. Thus, the amount of deposited carbon should be taken into account in calculating the weight loss.



**Fig. 1.** Schematic diagram of the experimental set-up.

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