

Integrated coal-pyrolysis tar reforming using steelmaking slag for carbon composite and hydrogen production

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HIGHLIGHTS

- Catalytic tar reforming over steelmaking slag to produce hydrogen and carbon material.
- Steelmaking slag increased hydrogen gas product and carbon content within the slag.
- High carbon deposition within slag was obtained at low temperature.
- Beside tar decomposition, methane reforming occurred at temperature above 600 °C.
- This system offered energy saving 104.3 MJ/ton steel in the steelmaking industry.

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ABSTRACT

Steelmaking slag contains high amounts of CaO, Fe₂O₃, SiO₂, and Al₂O₃, and has great potential as a catalyst for the tar-reforming reaction to produce a carbon composite and hydrogen. This paper describes chemical waste-heat recovery of tar and the effects of slag on the tar-reforming reaction. The results indicate that slag has a good activity for decomposing tar into the gas phase and for producing high carbon content within the slag. The introduction of coal-pyrolysis products to slag at a temperature of 500 °C caused an 18%vol increasing in the gas amount and a 6%mass enhancing in the carbon content within the slag compared to coal pyrolysis without slag. At higher temperatures, coal pyrolysis gave rise to lower carbon deposition within the slag but a higher amount of gas product. Moreover, the gas reforming reaction occurred simultaneously with tar decomposition at higher temperatures. This proposed system offers energy-saving benefits of 103 MJ/ton steel and a 6%mass reduction in coke-breeze usage in the steelmaking industry.

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1. Introduction

World steel consumption is rising continuously as a result of the growth in the world economy. The production of pig iron as the raw material of steel increased from 586 Mt in 2001 to 1025 Mt in 2010 [1]. However, steelmaking is one of the most energy-intensive industries, accounting for around 5% of the total world energy consumption, with a waste-heat recovery rate of only 17% [2]. Several actions have been taken to improve energy efficiency during the process, such as process development [3,4] and hot-slag energy recovery [5], but the results are still unsatisfactory. Coal is the primary energy source in the steelmaking industry, and is converted to coke, tar, and gas through pyrolysis in the coking plant. World coal resources are being depleted continuously

because of the high demand in the industrial sector. This has caused an increase in coal prices, in turn leading to high costs in the steelmaking industry. In order to achieve cost reduction and solve the problem of coal resources, the coal energy should be utilized effectively in the steelmaking industry. Besides being an energy source, coke is also used as an iron reducing agent and permeable support in blast-furnace operations [6]. Tar vapor and coke oven gas (COG) are by-products generated during the coking process, and contain high amounts of carbon and energy. In the pyrolysis process, tar material may cause operational problems such as pipe blocking, condensation, and tar aerosol formation [4].

In contrast, about 150–200 kg of steelmaking slag is produced as a by-product per ton of steel; this slag contains high amounts of CaO, Fe₂O₃, SiO₂, and Al₂O₃. Nowadays, steelmaking slag is utilized as a mixture material in cement production, soil conditioners, and fertilizers [7]. It has been reported that iron-oxide-based catalysts such as Fe₂O₃–Al₂O₃ could be used effectively for

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Table 1
Coal chemical properties.

Sample	Proximate (%mass)			Ultimate (%mass)				
	FC ^a	VM ^b	Ash	C	H	N	O ^c	S
Coal	66.9	24.4	8.7	80.2	5.3	1.8	12.8	0.5

^a FC = fixed carbon.

^b VM = volatile matters.

^c Calculated by difference.

Table 2
Chemical composition of steelmaking slag.

Comp. (%mass)	TFe ^a	MFe ^b	FeO	CaO	SiO ₂	Al ₂ O ₃	MgO	C	WC ^c
Slag	21.03	14.05	5.38	40.02	8.42	2.75	2.45	2.67	7.45

^a TFe = total iron.

^b MFe = metallic iron.

^c WC: water content (free water and hydroxides).

hydrogen production from biomass pyrolysis products [5,8]. Moreover, tar reforming/decomposition for hydrogen production is also quite effective over catalysts supported by CaO and MgO [7]. The CO₂ reforming of CH₄ over hot granulated slag that contains high amounts of CaO and SiO₂ was studied by Purwanto and Akiyama. The results showed that the slag behaved as a good catalyst for the reforming process [9]. Carbon deposition over the catalyst surface was the main problem during catalytic tar reforming [10,11]. Hosokai et al. suggested the possibility of direct tar reforming from pyrolysis over the char bed to improve the gas quality in terms of the amount and the heating value. This system gave some benefits such as increases in the amounts of CH₄ and H₂, and the elimination of tar to avoid pipe blocking and condensation [12]. The hydroxide molecules within a low-grade iron ore such as laterite created a highly porous material that is suitable for the tar-reforming reaction. During the reforming reaction, tar carbon could also be infiltrated within low-grade iron ore to produce a carbon composite

material [13]. Kashiwaya and Akiyama investigated the behavior and effect of deposited carbon on the iron-ore reduction reaction. There were two possible routes for the infiltration of deposited carbon: the tar itself diffusing into the pores, and gases decomposed from the tar diffusing into the pores. Both the deposited carbon and gas product promoted the iron reduction reaction [14].

On the basis of these two facts related to the tar problem and steelmaking slag, the utilization of steelmaking slag as a catalyst for tar reforming to produce hydrogen and carbon material is attractive to solve these problems. Hydrogen gas could be utilized as a fuel, whereas the slag–carbon material could be used as a supplementary fuel when the slag is recycled in the sintering machine, as it contains free CaO and FeO. An integrated system of pyrolysis and tar reforming will be applied in this proposed system to avoid tar condensation and to improve the gas quality. The objective of this paper is to study the chemical waste-heat recovery from steel-making slag and tar material. The effects of slag on the tar-reforming reaction and the amount of carbon deposition within the porous slag are also investigated.

2. Materials and experimental methods

2.1. Materials

Australian coal, crushed and sieved to approximately 250–500 µm, was used in these experiments. Table 1 shows the proximate and ultimate analysis results of this coal.

Table 2 shows the chemical composition of the steelmaking slag from the Japanese steel industry, which was used in these experiments. The steelmaking slag sample was also crushed and sieved to approximately 250–500 µm.

2.2. Experimental methods

The experimental apparatus is shown schematically in Fig. 1. The reactor was made from quartz with 30-mm inside diameter and 550-mm height, and was equipped with an SUS 304 mesh.

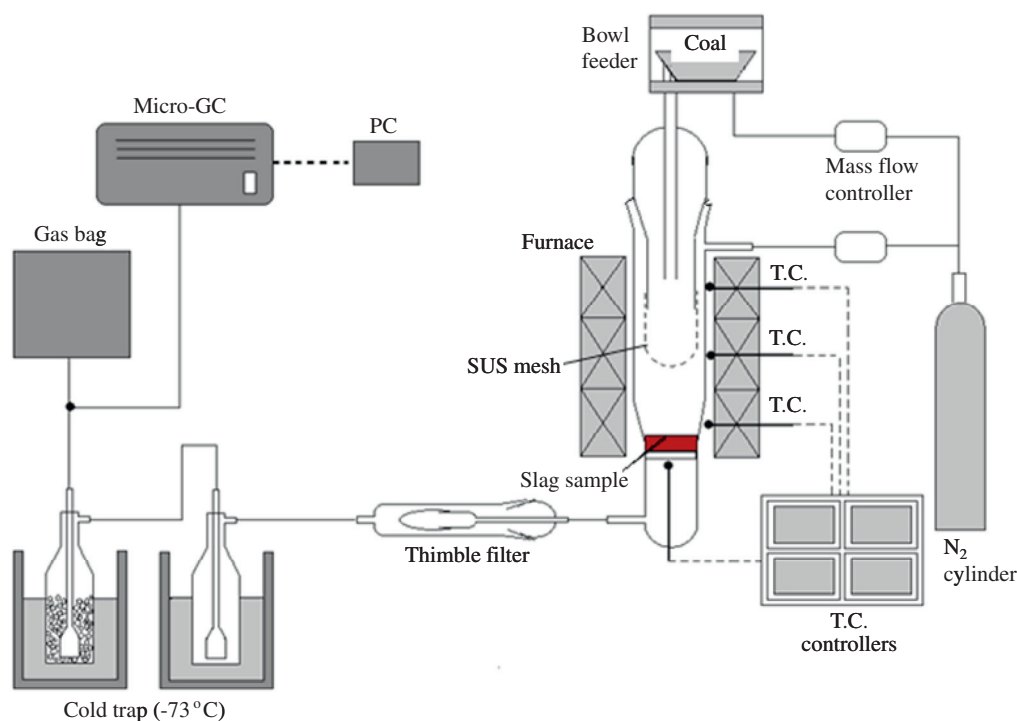


Fig. 1. Experimental apparatus for integrated coal-pyrolysis tar reforming using steelmaking slag.

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