



Material balances of major and trace elements in hydrogen production process from coal with CO₂ recovery

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HIGHLIGHTS

- ▶ The material balances of major and trace elements in coal in the process were investigated.
- ▶ The behaviors of Na, Mg and Pb in the experiments were similar to those at equilibrium.
- ▶ The behaviors of As and Hg in the experiments were different from those at equilibrium.

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ABSTRACT

A new hydrogen production process “HyPr-RING” (hydrogen production by reaction integrated novel gasification), in which high-pressure steam (HPS) gasifies organic materials with a CO₂ sorbent (CaO) has been developed. In this study, the material balances of major elements such as sodium and magnesium, and trace elements such as arsenic, mercury and lead in coal in the HyPr-RING process at thermodynamic equilibrium were investigated using commercial simulation software package, and compared with those experimentally determined on the HPS gasification of coal with CaO at a temperature of 650 °C and calcination of the solid residue collected after gasification at 900 °C. The behaviors of sodium, magnesium and lead in the experiments were similar to those at equilibrium, whereas those of arsenic and mercury were different. Sodium and lead existed as solid-phase species after gasification, whereas they existed partly as gas-phase species after calcination. Magnesium existed as solid-phase species after gasification and calcination. After gasification, arsenic existed as gas-phase species at equilibrium, whereas solid-phase species were present in the experiments. After calcination, arsenic existed as solid-phase species at equilibrium and in the experiments. After gasification and calcination, mercury existed as gas-phase species at equilibrium, but part of the mercury remained in the solid residue collected in the experiments.

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1. Introduction

From the viewpoint of the depletion of fossil-fuel resources and global environmental issues, the clean use of unused carbonaceous resources such as biomass and organic wastes, as well as low-grade coal, should be promoted. In the production of energy from organic resources, the minimization of CO₂ emissions is required. To establish a new social system with clean and efficient energy sources such as hydrogen-based fuel-cells, a stable supply of hydrogen will be an important issue. The steam gasification of fossil fuels, biomass, or organic wastes is one of the key technologies for hydrogen production.

HyPr-RING (hydrogen production by reaction integrated novel gasification) is a new gasification method for producing hydrogen from carbonaceous resources using high-pressure steam (HPS) with simultaneous reduction of CO₂ using a calcium-based sorbent [1–15]. Fig. 1 shows the concept of the HyPr-RING method. Almost all organic materials, such as coal, can be reacted with steam and a sorbent to produce mainly hydrogen and CaCO₃. CaCO₃ can be regenerated to CaO by calcination, and a high-concentration stream of CO₂ is generated.

In our previous studies, attention was focused on the initial stage of the HyPr-RING reaction and the characteristics of the steam gasification of organic matter such as coal and sludge with Ca(OH)₂ were kinetically studied [16–18]. Most trace elements in coal and/or waste are expected to be in the solid and/or liquid phase under the HyPr-RING conditions because the reaction

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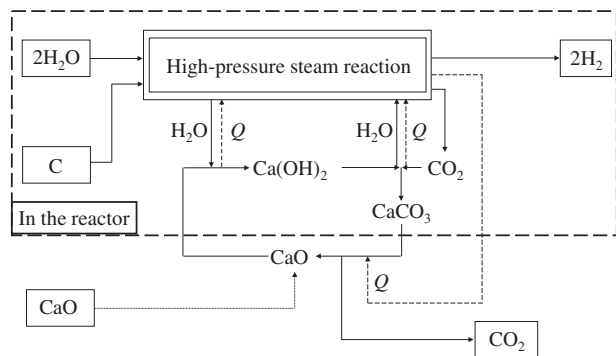


Fig. 1. Concept of the proposed hydrogen production method (HyPr-RING method).

temperature of HyPr-RING is lower than that of conventional steam gasification [19–22]. Up to the present time, however, there has been no report focusing on the behavior of trace elements under the HyPr-RING conditions [1–15]. It is necessary to study the behavior of trace elements in order to develop the HyPr-RING process, as well as conventional coal and/or waste combustion or gasification processes [23–28].

In this paper, the material balances of major elements such as sodium and magnesium, and trace elements such as arsenic, mercury and lead in coal in the HyPr-RING process at thermodynamic equilibrium were investigated using commercial chemical equilibrium simulation software and compared with those in experiments on HPS gasification of coal with CaO at a temperature of 650 °C and calcination of the solid residue collected after gasification at 900 °C.

2. Thermodynamic equilibrium simulation

The thermodynamic equilibrium simulations of carbon, hydrogen, nitrogen, oxygen, major elements such as sodium and magnesium, and trace elements such as arsenic, mercury, selenium, lead, cadmium, boron and chromium in Adaro coal for HPS gasification with Ca(OH)₂, cooling with decompression after the HPS gasification and the calcination of the solid residue collected after cooling were carried out using commercial software (FactSage 5.5) with the databases for slags (FACT-FeS and FACT-SlagA) to investigate the material balances of major and trace elements in the HyPr-RING process.

2.1. HPS gasification of coal with Ca(OH)₂

The properties of Adaro coal are shown in Table 1. In the thermodynamic equilibrium simulation of the HPS gasification with Ca(OH)₂, an Adaro coal feed of 1 g and molar ratios of steam and Ca(OH)₂ to carbon of 4 and 1.43 were used to determine the input values at a temperature and pressure of 25 °C and 0.1 MPa. Initially, the input values for arsenic, selenium, mercury, cadmium, lead, fluorine, boron and chromium were set in the actual contents in Adaro coal. As a result, the behaviors of the trace elements were unclear because compounds with the trace elements were not formed in the thermodynamic equilibrium simulation. By contrast, the set in the excess of the actual contents in Adaro coal clarified the behaviors of the trace elements. Therefore, finally, the input values for arsenic, selenium, mercury, cadmium, lead, fluorine, boron and chromium were set in excess of the actual contents in Adaro coal to clarify their behavior. The input values are shown in Table 2. The temperatures and pressure selected for the simulation system were 0–1000 °C and 3.0 MPa.

Table 1
Properties of Adaro coal used in high-pressure steam gasification.

<i>Proximate analysis (wt.%, as received basis)</i>	
Moisture	16.3
Ash	1.1
Volatile matter	57.0
Fixed carbon	25.6
<i>Ultimate analysis (wt.%, dry ash-free basis)</i>	
C	75.8
H	4.9
N	0.9
S	0.01
Cl	0.02
O (difference)	18.4
<i>Ash composition (wt.%)</i>	
CaO	37.1
SO ₃	17.3
MgO	15.1
Na ₂ O	12.9
Al ₂ O ₃	6.0
Fe ₂ O ₃	5.2
SiO ₂	4.6
K ₂ O	1.1
TiO ₂	0.3
Cr ₂ O ₃	0.02
<i>Trace elements (μg/kg-coal)</i>	
As	1.9
Hg	10.9
Pb	29.8

2.2. Cooling with decompression after HPS gasification

The thermodynamic equilibrium simulation for HPS gasification with Ca(OH)₂ was used to determine the input values at 650 °C and 3.0 MPa in the simulation of cooling with decompression after the HPS gasification. The temperature and pressure were 25 °C and 0.1 MPa.

2.3. Calcination of solid residue collected after cooling

The input values in the thermodynamic equilibrium simulation for the calcination of the solid residue collected after cooling were used at 25 °C and 0.1 MPa. The atmospheric gas composition was air, i.e., an N₂:O₂ ratio of 4:1. The temperatures and pressure were 0–900 °C and 0.1 MPa.

3. Experimental

3.1. HPS gasification of coal with CaO

The observed data from an updraft fluidized-bed gasifier were used for comparison with the simulation results for the HPS gasification of coal with CaO. A schematic diagram of the experimental system is shown in Fig. 2; it has been described in detail elsewhere [29]. Adaro coal from Indonesia was used as a typical sub-bituminous coal. The coal was pulverized to a sieve size ranging from 0.25 to 0.5 mm because coal with a smaller particle size was difficult to supply steadily. The properties of the coal have already been shown in Table 1. A commercial extra-pure-grade Ca(OH)₂ reagent (Kanto Chemical Co., Inc., Japan) was used to capture CO₂.

A solid mixture of coal and CaO in a hopper with a pressure vessel was supplied by a screw feeder. This feeder can steadily supply powder of size 0.25–0.5 mm to the fluidized-bed reactor. The reactor had an inner diameter of 70 mm and a length of 1500 mm. The height of the fluidized bed was approximately 800 mm [29]. The mixture was continuously supplied onto a perforated disk, which was set 1000 mm below the top of the reactor, with feed rates of coal and CaO of 1.25 and 3.14 kg/h. The mean residence time of

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