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Short Communication

The effect of preparation method on Fe/Al/Cu oxide-based catalyst performance for high temperature water gas shift reaction using simulated waste-derived synthesis gas

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ABSTRACT

The effect of preparation method on Fe/Al/Cu oxide-based catalysts was investigated for the high temperature water gas shift reaction using simulated waste-derived synthesis gas. Impregnated Fe/Al/Cu catalyst and sol–gel prepared Fe/Al/Cu catalyst were compared with a co-precipitated Fe/Al/Cu catalyst. The co-precipitated Fe/Al/Cu catalyst exhibited very high CO conversion ($X_{CO} > 85\%$ at 450 °C) with stable activity. This performance was attributed to the high surface area, the small crystallite size of the Fe_3O_4 , the ease of reducibility, and formation of reduced form of the Cu species.

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Introduction

Currently, there is growing interest in hydrogen as a clean, sustainable energy carrier because hydrogen utilization processes emit only water, without pollution or the production of greenhouse gases [1–3]. Hydrogen is produced commercially by the steam reforming of natural gas [4–7]. However, due to fossil fuel depletion and emission of greenhouse gases, it is necessary to produce hydrogen with different methods [8].

The generation of energy from waste has been attractive for many years and waste gasification is a viable technology for the production of clean hydrogen [9]. In addition, the application of waste gasification can reduce negative environmental impacts, such as air pollution, groundwater contamination, and other problems related to the various wastes caused by premature combustion [10].

The water gas shift (WGS) reaction is a key process in waste gasification for clean H_2 production using waste-derived synthesis gas [9]. Because the WGS reaction is slightly

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exothermic and favorable at low reaction temperatures thermodynamically, two reaction stages are employed in industry. One approach at 350–450 °C, high-temperature WGS (HT-WGS) reaction, is followed by low-temperature WGS (LT-WGS) reaction at 200–250 °C. Two types of catalysts are commonly used for these two stages of WGS: Fe–Cr catalyst for HT-WGS and Cu–Zn catalyst for LT-WGS [11–13]. In the waste gasification process, the outlet gas from the waste gasification process has a rather high temperature, making it reasonable to employ the HT-WGS approach for hydrogen production.

The typical composition of the commercial HT-WGS catalyst is Fe₂O₃ (88–92%) and Cr₂O₃ (8–12%) [14]. Magnetite (Fe₃O₄), which can be formed by partial reduction of hematite (Fe₂O₃), is a well-known active phase in the HT-WGS reaction [4,15,16]. The role of Cr in the Fe₂O₃–Cr₂O₃ catalyst is to prevent the thermal sintering of Fe₃O₄ and loss of catalyst surface area during the reaction [17]. Although there are some advantages to use Cr in the Fe-based catalyst, Cr⁶⁺ itself is a potent environmental toxin. To eliminate the use of Cr, Al is a potential substitute for Cr since it can effectively prevent thermal sintering and minimize surface area loss [18,19]. In addition, using a Cu promoter can enhance the HT-WGS activity of the Fe–Al catalyst [17]. Therefore, Fe/Al/Cu catalysts have attracted a great deal of interest due to their outstanding performance for HT-WGS [9,17,20].

Several studies have already been published concerning Fe/Al/Cu catalysts for HT-WGS [21,22]. Araújo and Rangel have investigated the HT-WGS over Fe/Al/Cu catalysts prepared by the precipitation-impregnation method and found that the Fe/Al/Cu catalyst exhibited similar catalytic performance compared to the commercial Fe–Cr–Cu catalyst [23]. Zhang et al. also reported that Fe/Al/Cu catalyst prepared by the sol–gel method showed higher catalytic activity than the commercial Fe–Cr–Cu catalyst. They also found that the catalytic activity of the Fe/Al/Cu catalyst can be affected by the catalyst preparation method [24].

In our earlier report, we prepared the Fe/Al/Cu catalysts by the impregnation method, and applied them to the HT-WGS reaction using waste-derived synthesis gas [9]. Our developed Fe/Al/Cu catalyst exhibited higher catalytic activity and stability than the commercial Fe–Cr catalyst. However, the high CO concentration of the waste-derived synthesis gas can deteriorate the catalytic performance of commercial Fe–Cr catalyst because it makes severe conditions [20]. We also reported that a Fe/Al/Cu catalyst prepared by the simple sol–gel method showed high catalytic performance without an activation procedure [9]. It has been reported that a Fe/Al/Cu catalyst synthesized by the sol–gel technique exhibited higher activity than Fe/Al/Cu catalysts prepared by precipitation-impregnation techniques [21]. Although many studies of Fe/Al/Cu catalysts have been reported [9,21–24], the effect of the preparation method on the HT-WGS activity has not been investigated over Fe/Al/Cu catalysts.

The objective of the present work is to study the effect of preparation method on physicochemical characteristics and to optimize the preparation method through catalytic performance of Fe/Al/Cu catalysts for the HT-WGS reaction using waste-derived synthesis gas. The Fe/Al/Cu catalysts in the present work were prepared using co-precipitation (CP),

sol–gel (SG), and impregnation (IM) methods, respectively. The results of the physicochemical characterization are discussed in relation to the catalytic performance of the Fe/Al/Cu catalysts.

Experimental

Catalyst preparation

The Fe/Al/Cu catalysts were synthesized by co-precipitation (FAC-CP), sol–gel (FAC-SG), and impregnation (FAC-IM) methods. All catalysts contained 83 wt.% Fe, 7 wt.% Al, and 10 wt.% Cu. The FAC-CP sample was prepared from well-mixed (stirrer speed of 1150 rpm) Fe(NO₃)₃·9H₂O (98%, Aldrich), Al(NO₃)₃·9H₂O (98%, Aldrich), and Cu(NO₃)₂·xH₂O (99%, Aldrich) water solutions by dropping the solution of K₂CO₃ (99%, Aldrich). The product was collected by filtration and washed with distilled water in order to remove the K⁺ ion impurity before drying in an oven at 100 °C for 12 h. The prepared catalyst was calcined at 400 °C for 3 h. The details of the impregnation and sol–gel methods can be found in our earlier paper [9,25].

Characterization

The BET surface areas of the catalysts were measured using a N₂ physisorption technique on a Micromeritics ASAP 2010 accelerated surface area and porosimetry instrument. Before measurement, samples were degassed for 12 h at 110 °C under a vacuum of greater than 0.5 mm Hg. XRD characterization was carried out using a Rigaku D/MAX–IIIC diffractometer operated at 40 kV and 100 mA with Ni filtered Cu–K α radiation. The sample was measured with a scan rate of 1°/min from 20° to 80°. The crystallite size was calculated from diffraction by the Debye–Scherrer equation. Hydrogen–temperature programmed reduction (H₂–TPR) experiments were carried out in an Autochem 2910 (Micromeritics). The detailed procedure for the TPR is described in a previous paper [26,27]. Transmission electron microscopy (TEM) images of catalysts were obtained in Tecnai G² T-20S (FEI) microscope with 200 kV operating voltage. All samples were suspended in ethanol by ultra-sonication. The suspension was deposited on a copper grid with carbon film for TEM measurements. XPS spectra were obtained using a K α spectrophotometer (Thermo-Scientific), with a high-resolution monochromator. The pressure of the analysis chamber was maintained at 6.8×10^{-9} mbar and with the detector in constant energy mode with a pass energy of 100 eV for the survey spectrum and 50 eV for the detailed scan. The binding energy was calibrated using the C1s transition, which appeared at 284.6 eV.

Catalytic reaction

The activity test was conducted using the same reactor used in our previous study [9]. Catalysts were first reduced at 400 °C for 1 h with a stream of 2% H₂/N₂. After that, catalysts were evaluated at different reaction temperatures, covering a range from 350 to 550 °C. A gas hourly space velocity (GHSV) of 40,057 h⁻¹ was employed to screen the catalysts in this study.

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