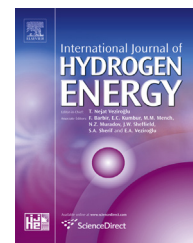


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Fluidized-bed gasification combined continuous sorption-enhanced steam reforming system to continuous hydrogen production from waste plastic

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ABSTRACT

This paper proposes a novel system for continuous hydrogen production from waste plastic (WP) by fluidized-bed gasification (FBG) combined sorption-enhanced steam reforming process (SERP). The system was operated in successive processes of several sections: (a) gasifying a WP into a raw gas comprising CO, H₂, CH₄, total hydrocarbons (THC), and small amount of HCl contaminant, etc.; (b) passing the raw gas to hydrogen production through two moving-bed reactors by continuous SERP process over Ni-based catalyst mixed CaO sorbent for in-situ CO₂ capture and HCl removal; (c) simultaneously regenerating CaCO₃ formed and catalyst with carbons deposited in other moving-bed reactor at the regeneration condition selected; and (d) carrying the particles of catalyst and sorbent to continuous steam reforming and their regeneration between two moving-bed reactors by riser. Gradually expanding chamber design of FBG reactor suitable for different particles flow to prolong the residence times of gas and solid phases makes high carbon conversion and the maximum value is up to 83.6% at 880 °C during FBG stage. The combination of FBG and SERP has produced a stream of high-purity hydrogen at some certain conditions, and about 88.4 vol % of hydrogen (H₂O- and N₂-free basis) was obtained at 818 °C of FBG temperature with 706–583 °C of SERP temperature. Reduced Ni-based catalyst efficiently converted raw gas from FBG and steam to H₂, and CaO sorbent in the moving-bed reactor are capable of reducing the HCl and CO₂ to low levels at all the temperatures tested.

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Introduction

Depletion of fossil fuels is creating opportunities in exploring alternative sources of energy, and at the same time, hydrogen is a clean energy carrier for the development of potential high efficiency power generation alternatives such as fuel cells. The suitable waste plastic (WP) management strategy for the manufacture of value added products is very important aspect of sustainable development in modern society. Economical hydrogen production from thermal-chemical conversion of WP as a viable renewable energy source in some ready manners remain a challenge [1,2]. It is very difficult for the design and operation of conversion systems due to various compositions of WP, and the differences in its thermal degradation behavior. Some limitations in fluidized-bed gasification (FBG) of WP still need to be tackled to increase the feeding conversion and decrease the contaminants in the gas products [3]. As we know, the continuous removal of CO₂ by CaO sorbent from the water-gas shift (WGS) reaction will incessantly drive the equilibrium-limited WGS reaction in the forward direction, which will ensure a high yield and purity of H₂ with near stoichiometric amount of steam needed for the reaction [4,5]. In addition, HCl gas contaminant from WP conversion can simultaneously be removed by solid sorbents including CaO at high temperatures [6].

A few papers specifically reported FBG to syngas and energy production [7–9], and the attention should be directed towards achieving high carbon conversion and obtaining the desire products. Seven mixtures of coals, plastics and wood have been fed into a pre-pilot scale fluidized-bed gasifier in order to investigate the main aspects of the co-gasification of these materials, and the raw gas consists of CO, H₂, CO₂, CH₄, C_nH_m, etc [8]. Ruoppolo et al. reported the results of gasification tests using a catalytic fluidized-bed gasifier to obtain a H₂-rich stream and only 32 vol% of hydrogen concentration was produced [9]. It has been indicated that the composition of the gas products from gasification depends on the inherent nature of the feeding and the process conditions employed, the concentration of raw gas components is very difficult to adjust and control. Tar and heavy hydrocarbons produced during FBG of WP are undesired and dangerous for typical post-process applications.

In the past few decades, the extensive studies on novel systems for low-cost hydrogen production have been carried out and the sorption-enhanced steam reforming process (SERP) with in-situ CO₂ capture has been considered to change the normal equilibrium limits of WGS reaction for producing high-purity hydrogen and reducing the number of processing steps required for subsequently separating CO₂ [10–13]. Recently, we reported a approach on continuous SERP [14,15] to high-purity hydrogen production and the process was characterized by a continuous flow concept of catalyst and sorbent for steam reforming and regeneration using two moving-bed reactors by the integration of continuous steam reforming, WGS, CO₂ capture, and hydrogen separation in one single reactor, which can result in improved process efficiency and reduced capital costs. The operating flexibility of continuous SERP by two moving-bed reactors makes it possible to utilize different types of gas and liquid feedstock such as raw

gas including CO, H₂, CH₄, total hydrocarbons (THC), etc. from FBG of WP. Our previous researches were directly related to high temperature pyrolysis, and gasification [16,17], and some reasonable amounts of gas products were produced during the processes.

To our knowledge, there is hardly any publication in the literature providing information on the ways of integrating FBG and continuous SERP for hydrogen production from a typical WP, which is not very needed to consider the composition of the raw gas produced from FBG. The integrated system indeed consolidates several unit operations including FBG of WP, steam reforming of raw gas, WGS, in-situ CO₂ capture and HCl removal. Fundamental studies and experimental works are also developed for high quality energy production from low-cost fuels as well as the reduction of environmental impact.

Experimental

Preparation of WP

Samples of WP from industrial plastic plant in China were prepared in order to obtain fine particles and sieved to 0.5–1.2 mm. The proximate analysis was carried out using the method described by ASTM D5142. The ultimate analysis was conducted using an element analyzer (EA1110, CE instrument). Thermogravimetric analysis (TGA) for WP pyrolysis was carried out using a Stanton–Redcroft thermogravimetric apparatus (STA 780). The calorific value of the sample (LHV) was calculated by the Dulong's equation:

$$\text{LHV} = \text{HHV} - 600(9H + W)$$

$$\text{where, } \text{HHV} = 8100C + 34000 \left(H - \frac{O}{8} \right) + 2500S$$

Preparation of catalyst and sorbent

The NiO/NiAl₂O₄ catalyst was prepared by the co-precipitation method with rising pH technique under controlled conditions and the calcination at high temperature of 900 °C in previous studies [14]. The 1 M NH₄OH was added to an aqueous solution (Ni(NO₃)₂·6H₂O and Al(NO₃)₃·9H₂O) with vigorous stirring. The precipitation was carried out at 50 °C until the final pH of 8.5 was kept. The precipitates were filtered and followed by drying at 110 °C over 15 h, and then calcined in air atmosphere at the temperature of 900 °C for over 3 h. The spinel composition of Ni–Al oxides was formed at the calcination temperature of 900 °C by the solid state reactions. The NiO content in the catalyst is about 29.6 wt% by elemental analysis. CaO sorbent was prepared from limestone decomposition, and it mainly consisted of higher than 96 wt% CaO based on the data provided by the industry manufacturer. Other compounds are CaCO₃ (<2.0 wt%) and MgO (<1.0 wt%). The particles of catalyst and sorbent in all the experiments were 0.10–0.25 mm.

Characterization of catalyst

The specific surface areas of fresh catalyst and sorbent were determined using a Micrometric Acusorb 2100E apparatus

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