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Steam reforming of gasification-derived tar for syngas production

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ABSTRACT

In this study, the steam reforming of tar was catalyzed by dolomite, Ni/dolomite, and Ni/CeO₂ for syngas production under different reaction temperature and weight hourly space velocity (WHSV, h⁻¹). The tar was the major side product from the biomass gasification.

Current results revealed that the nickel doped catalyst on dolomite with CO₂ in the feed stream yielded the highest H₂ and syngas production among all reaction conditions. Comparing to the use of dolomite, when Ni–dolomites was used as catalyst, the yield of H₂ increased by 33%, the yield of syngas increased by 7%, and the yield of CH₄ decreased by 59%. It was also found that the yield of syngas, H₂, or CO under the Ni/dolomite catalyst were significant higher ($p < 0.001$) than those under the dolomite catalyst using analysis of covariance.

Taguchi method was also applied to elucidate the sensitivity of experimental condition in this study. The impact of the factors on the entire performance had the following order: types of the catalyst > CO₂ concentration in the feed stream > reaction temperature > weight hourly space velocity.

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Introduction

The depletion of energy resources and the greenhouse emission for the environment are the major concerns for energy supply scheme. Seeking for the sustainable energy resources is the most important task at this moment. Modern sustainable energies include solar, wind, marine, and biomass energy. Biomass energy can be generated by direct combustion, thermal chemical conversion, and biological conversion. Thermal chemical biomass conversion can also be achieved by torrefaction, pyrolysis, or gasification to produce solid,

liquid, and gas phase energy carriers, respectively. Syngas is the most important product during the biomass gasification and it can be further converted into C-1 fuel or long-chain alkane fuels. So, biomass gasification can be considered as one of the most important energy production method in the future [1].

Tar is the major side product of the biomass gasification. It was formed by very complex chain reactions from cellulose, hemicellulose, and lignin in biomass during the gasification processes. The major components of tar are heterocyclic aromatics (such as: pyridine, phenol, cresols, quinoline, isoquinoline, and dibenzophenol), light aromatic (such as:

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toluene, ethylbenzene, xylenes, and styrene), light poly-aromatic compounds (such as: idene, naphthalene, methyl-naphthalene, biphenyl, acenaphthalene, fluorene, phenanthrene, and anthracene), and heavy poly-aromatic compounds (such as: fluoranthene, pyrene, chrysene, perylene, and coronene). The molecular weight of tar is too high to be easily analyzed by gas chromatograph [2–5]. Moreover, tar causes the reduction of syngas production, system blockage, and system corrosion. Syngas could be the resources of energy and tar could not be the resources easily without any treatment. It is very important to increase the yield of syngas and decrease yield of tar in the biomass gasification process.

Tar can be removed by the *in-situ* cleanup (the primary method) or the post treatment of the effluent flue gas (the secondary method) through the catalytic reactions or the physical processes. The physical tar removal process includes sand bed filter, wash tower, Venturi scrubber, rotational atomizer, wet electrostatic precipitator, fabric filter, or rotational particle separator [4]. They are operated at lower than 100 °C with less than 60% of tar reduction. Regeneration and cost of these equipments are the major concerns for the practical applications. However, the thermal chemical cleanup process, such as catalytic tar cracking, can convert tar into syngas at the biomass gasification temperature. Thus, tar can be converted to syngas and the overall yield can be enhanced.

In this study, tar (the side product of biomass gasification) steam reforming to generate syngas was proposed. The types of the catalyst, reforming temperature, CO₂ concentration in the sweeping gas, and weight hourly space velocity (WHSV, h⁻¹) were the major experimental parameters for the study. Experimental design by the Taguchi method was also explored to have the optimized condition. Part of the experiment results were also elucidated by the second order Response Surface Method (RSM).

Materials and methods

Catalyst preparation

Three different types of the catalyst, dolomite, nickel doped dolomite (Ni/D), and Ni–CeO₂ catalyst were prepared for this study. Dolomite was received from local stoneware supplier (Hualien County, Taiwan) and it was ground (particle size < 0.1 mm) prior further treatments.

To prepare the Ni/D catalyst (10 wt% of Ni), nickel (II) nitrate hexahydrate (Ni(NO₃)₂·6H₂O, 98%, SHOWA) was used as the nickel precursor and Ni was deposited at the surface of dolomite by the incipient wetness impregnation method. The Ni/CeO₂ (10 wt% of Ni) catalyst was prepared by coprecipitation method by using nickel nitrate and cerium (III) nitrate hexahydrate (Ce(NO₃)₃·6H₂O, 99.5%, ACROS) as the precursor and ammonium solution as the precipitation agent to form the catalyst. This method was revealed in our previous studies [7,8].

The fresh prepared catalyst powder (dolomite, Ni/D, and Ni/CeO₂) was calcined at 550 °C for 3 h. To make the catalyst pellet for the tar reforming reaction, the prepared catalyst

powder, carboxymethyl cellulose sodium salt (CMC, C₆H₇O₂(OH)₃, SHOWA), and Ludox TM-40 colloidal silica 40 in H₂O solution (40%, Sigma–Aldrich) were then mixed at the weight ratio of 5:1:15, molded into cylindrical shape (1/8" of outer diameter, 1/4" in length), air-dried for 48 h, and calcined at 900 °C for 4 h using air as the sweeping gas [9–11].

Materials characterization

The tar for this reaction was collected from our previous studies of the biomass gasification for hydrogen production [6]. The collected tar was analyzed by the proximate analysis (by the standard procedure of NIEA R205.01C, Republic of China) and ultimate analysis (Elementar, EA, Vario E1; for the composition of C, H, O, N, and S) to determine the water contents of tar and the elemental composition of the anhydrous tar. Then, the water content of the collected tar was adjusted to the designated ratios and then used as the substrate for reforming reaction.

The prepared catalyst was characterized by differential scanning calorimetry (DSC, TA Instruments, SDT2960) for thermal stabilities and properties. The surface area and the porosity were determined by BET (Micromeritics 2010C). The crystalline structure was characterized by the X-ray diffraction which is a continuous position sensitive detection technique (CPS) on a material analysis and characterization (MAC) MXP3 diffractometer using copper target (1.5405 Å Cu K α radiation) at room temperature. The surface morphology and composition were determined by Scanning Electron Microscopy with Energy Dispersive X-ray Spectroscopy (SEM/EDS, HITACHI S3000H). The bulk composition of the prepared catalyst was determined by inductively coupled plasma-optical emission spectrometer (ICPOES, Perkin Elmer OPTIMA 2000DV) for the composition analysis.

Reactor design and the reaction

During the reaction, tar was fed to the packed bed reactor by a syringe pump at the designated reaction conditions. The catalyst, reaction temperature, CO₂ concentration in the feed stream, and weight hourly space velocity (WHSV, h⁻¹) are the key kinetic parameters for this study. The prepared catalyst pellet was activated at 650 °C in H₂ prior the reaction. In a typical batch of the reaction, 3 g of the prepared catalyst was loaded into a packed bed reactor at the substrate feeding rate of 2.50 mL/min (WHSV = 2 h⁻¹) and the carrier gas (argon) at the flow rate of 90 mL/min at the designated reaction temperature. The reaction may operated at WHSV = 2, 3, or 4 h⁻¹ by changing the weight of the catalyst and substrate (carbon in tar) feeding rate. During the reaction, the effluent gas was cooled by an ice bath prior sampling. The collected sample was then injected to a gas chromatograph (G.C., CHINA Chromatograph, GC-1000, equipped with a Porapak Q packed column (1/8" ID, 3 m, 80/100 mesh), and thermo conductivity detector (TCD current 60 mA, Temperature 40 °C), oven temperature 40 °C, and argon as the carrier gas). The compositions and the yield of the gas can be then analyzed. The effluent gas flow rate was also monitored by a soap-film gas flow meter to determine the yield of the gas product.

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