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A new approach for high-yield hydrogen production: Atmospheric pressure vapour-phase catalytic reforming of biomass hydrolysate

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ABSTRACT

Aqueous-phase reforming (APR) and atmospheric pressure vapour-phase reforming (AVPR) of biomass hydrolysate were performed in a continuous flow fixed-bed reactor using Raney-Ni, supported Ru, Pd and Pt catalysts for high-yield hydrogen production. Sorghum biomass hydrolysate and glycerol were used as feeds. It was observed that AVPR process was considerably more effective than APR process for high-yield hydrogen production. The hydrogen yield was highest when Raney-Ni catalyst was used in AVPR process led to 3.4 fold increase in hydrogen yield. The results showed that AVPR is more attractive than APR process for production of high-yield hydrogen with low amount of CO₂ and CH₄. No pressure application in AVPR is also a big advantage over APR for implementation the process in industry.

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Introduction

Fuel cells have emerged as promising devices for future global energy needs. In particular, hydrogen fuel cells used in electrical power generation are environmentally clean and highly efficient devices compared to the conventional gasoline and diesel engines [1–3]. The full environmental benefit of generating power from hydrogen fuel cells is achieved when hydrogen is produced from renewable sources such as lignocellulosic biomass [1]. Fuels obtained from biomass are thought to be greenhouse gas neutral,

because CO₂ is produced in the process consumed by further biomass growth. Among the alternative biomass resources, sorghum is one of the most promising biomass sources for bioenergy because of its drought tolerance and fast growth rate. On the other hand, glycerol, which is byproduct of biodiesel production is a potential feed for generating hydrogen via aqueous-phase reforming (APR)¹ since it has no wide application in the other aspects. Several processes have been used to produce hydrogen from biomass. Two common conventional processes are pyrolysis [4,5] and gasification [6–8]. However; these processes require very high processing temperatures (670–1100 K) [4,6–9]. On the other hand, APR of biomass and related model compounds

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¹ APR: Aqueous-phase reforming.

takes place at relatively very low temperatures for hydrogen production [10–18]. The low reaction temperature (~230 °C) in APR makes the process potentially more attractive than pyrolysis and gasification. However, the pressure (to keep the water in liquid form) are the unfavorable aspect of APR process.

No research has been carried out on atmospheric pressure vapour–phase reforming (AVPR) process yet. The present study was designed to investigate AVPR of sorghum hydrolysate and glycerol using a pressureless continuous flow fixed–bed reactor system. Hydrogen yield and other gaseous products obtained in this process were compared with the products obtained in APR process at the same conditions (temperature, catalyst, feed, feed flow rate, etc.). The biomass hydrolysate and/or glycerol solutions were pumped into the continuous flow fixed–bed reactor and vaporization/gasification reactions took place directly in the reactor. The reforming process was performed at lower temperatures (230–270 °C) than traditional steam gasification process. An individual “steam generator” system that is used in traditional steam gasification process was not used in AVPR process. Therefore, we named the process *atmospheric pressure vapour–phase reforming* instead of steam reforming.

Experimental section

Materials

Sorghum biomass (*Green Go*) and glycerol (as a model compound) were used as feed materials in the present study. Sorghum biomass was grown at Cukurova University Agronomy Research Fields located in Adana, Turkey. The sorghum biomass was dried at room temperature and ground using a laboratory cutting mill. The sizes of biomass used in the experiments were between 120 and 140 mesh. The analytical data for the biomass are shown in Table 1. The cellulose, hemicellulose and lignin contents were determined as described elsewhere [19–21].

Pt 5% on activated carbon, Pd 5% on carbon, Ru 5% on carbon and Raney nickel 2400 catalysts were purchased from Sigma–Aldrich. All other chemicals used in the study were of either analytical or reagent grade.

Hydrolysis of sorghum

The hydrolysis of sorghum biomass was performed in a 500 ml PARR 4575 model HP/HT reactor equipped with a magne–drive mixing system. A pre–determined amount of ground sorghum (10 g, dry basis) and 350 ml water were placed in the reactor. The reactor was sealed and air was swept out by successive pressurizing and depressurizing with carbon dioxide. The detailed information about hydrolysis process was described elsewhere [11,13]. The percent hydrolysis of sorghum were found to be 72.7% in this procedure.

APR and AVPR of sorghum hydrolysates and glycerol

APR and AVPR of sorghum hydrolysate and glycerol were performed in a continuous flow fixed–bed reactor with

Table 1 – Analytical data for sorghum (*Green Go*) biomass.

C, wt % (daf) ^a	39.75
H, wt % (daf)	5.22
N, wt % (daf)	0.85
S, wt % (daf)	0.11
O, wt % (daf) ^b	54.07
Moisture, wt %	8.11
Ash, wt % (dry)	8.39
Cellulose, wt % (dry)	46.63
Hemicellulose, wt % (dry)	12.54
Lignin, wt % (dry)	18.95

^a daf: dry, ash-free.

^b Calculated from difference.

commercial Raney–nickel 2400, supported Pt, Pd and Ru catalysts (Sigma–Aldrich). The catalyst was loaded into a stainless steel tubular reactor (inner and outer diameter of 0.56 and 1.4 inches, respectively). The tubular reactor (40 cm in length) was placed in an oven with the programmable temperature controller [22]. The 20 ml of sorghum hydrolysate or glycerol was introduced into the tubular reactor at a feed flow rate of 1.0 ml/min (in some runs 0.5 ml/min) using a HPLC pump. The carbon content of biomass hydrolysate and/or glycerol were used in two different concentrations; 1000 mg/l and 2500 mg/l. The system pressure was controlled with backpressure regulator for all APR runs. The pressure of the system was adjusted to 5.86 MPa to keep the water in liquid form at the 250 °C. Inert gas was not used in the APR processes. The pressure was created with hydrolysate/water solution. In the AVPR process, the pressure was not created. The system pressure was at atmospheric pressure (0.10 MPa). Because of high hydrogen efficiency, more experiments were performed in AVPR using different reaction temperatures (230, 250 and 270 °C). The products formed in APR/AVPR runs were cooled to 20 °C by using PolyScience digital temperature controller cryostat. Liquid and gaseous products were separated in the gas–liquid separator unit after gasification. Gaseous products were analyzed by a gas chromatograph.

The carbon conversion to gas is defined as:

$$C \text{ conversion to gas (\%)} = \frac{[C \text{ in the gaseous products}]}{[C \text{ fed into reactor}]} \times 100 \quad (1)$$

The amount of gaseous products obtained from APR and AVPR runs were given as ml gas/g catalyst (metal + support).

Analysis of gas and liquid samples

Gaseous products were collected into gas buret that was filled with water and the volume of the gas mixture measured by water displacement. Gas products were characterized using an Thermo Finnigan Trace Gas Chromatography equipped with a TCD detector. GC column was Supelco Carboxen 1010 plot fused silica capillary (30 m × 0.32 mm i.d.). The detailed information about gas analysis was described in our previous studies [23,24].

The compositions of sorghum biomass hydrolysates before and after APR/AVPR processes were determined by a GC–MS (Thermo Finnigan Trace Gas Chromatograph and Mass Spectrometer) equipped with Thermo TR-5 MS capillary column

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