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Disciplines
Biomechanical Engineering | Bioresource and Agricultural Engineering | Other Mechanical Engineering | Thermodynamics

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Thermochemical Generation of Hydrogen from Switchgrass

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Thermochemical production of hydrogen is anticipated to be one of the most cost-effective means of producing hydrogen fuel. Switchgrass, a warm-season perennial grass that is native to many areas of the United States, is an attractive feedstock for this purpose. The goal of this study is to convert switchgrass into hydrogen by the sequential processes of thermal gasification in a fluidized bed reactor, catalytic steam reforming of tars, and the use of water-gas shift catalysts to enhance the concentration of hydrogen. Air-blown gasification of switchgrass produced relatively low concentrations of hydrogen (about 8.5 vol-%). Steam reforming of tars and light hydrocarbons and reacting steam with carbon monoxide via the water-gas shift reaction increased the hydrogen content in the producer gas to 27.1 vol-%. The catalysts used in the steam reformer and water-gas shift reactors were examined at the end of the trials using X-ray photoelectron spectroscopy and BET analysis. These analyses showed changes in pore size and pore size distribution. Although not evident during the tests, eventual degradation of the catalysts can be expected as the result of deposition of coke, sulfur, and chlorine on the catalysts.

1. Introduction

Thermochemical production of hydrogen is anticipated to be one of the most cost-effective means of producing hydrogen fuel.1 Switchgrass, a warm-season perennial grass that is native to many areas of the United States, is an attractive feedstock for this purpose. It is prolific, drought resistant, and can be grown and harvested using conventional haymaking agricultural implements.2

Gasification of switchgrass produces a tar-laden producer gas of relatively low hydrogen content, typically containing no more than about 8.5 vol-% hydrogen. Downstream processing of producer gas must eliminate this tar and increase the concentration of hydrogen. Although the tar can be removed by scrubbing the gas stream with water, a more environmentally sound approach is steam reforming of tar, which cracks the large molecular weight organic compounds associated with tar into hydrogen (H₂) and carbon monoxide (CO).3 This approach also has the advantage of increasing the cold-gas efficiency of the gasification process since tar is converted into combustible gases instead of being removed from the producer gas.

Since biomass gasification yields relatively high CO/H₂ ratios, higher H₂ contents can be achieved by using commercial CO-shift catalysts in two fixed bed reactors operated in series: a high-temperature shift reactor for rapid reaction and a low-temperature shift reactor to shift thermodynamic equilibrium to very low levels of CO.4,5 The high-temperature shift reaction takes advantage of faster kinetics at elevated temperatures to convert about 75% of the CO into H₂. Since conversion is limited by thermodynamic equilibrium, which favors hydrogen formation at low temperatures, the gas is cooled before entering the second shift reactor.

The goal of this study is to convert switchgrass into hydrogen fuel by the sequential processes of thermal gasification, steam reforming of tars, and reacting carbon monoxide in the producer gas with steam to enhance the concentration of hydrogen. In an effort to better understand the long-term performance of the catalysts, this study includes an examination of changes in pore size and pore size distribution in the catalysts and the deposition of coke, sulfur, and chlorine on the catalysts.

2. Experimental Apparatus and Methodology

Tests were conducted at the Biomass Energy Conversion Facility (BECON) in Nevada, Iowa, which is operated by the Iowa Energy Center. A pilot-scale fluidized bed gasification system was used to perform the experiments. The system is rated at 800 kW (2.8 MMBtu h⁻¹) thermal input, which corresponds to an average throughput of 180 kg h⁻¹ (400 lb h⁻¹) of solid biomass fuel at a heating value of 16 000 kJ kg⁻¹ (7000 Btu lb⁻¹). The major components of the system include

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the fluidized bed reactor, fluidization gas system, fuel delivery system, data acquisition system, and gas sampling system. Details on the operation of the biomass gasifier can be found in Smeenk and Brown (1998).

The gasifier was fluidized with air at an equivalence ratio between 0.25 and 0.35, which maintained the reactor in the temperature range of 700 to 760 °C. The feed rate of switchgrass during these tests was in the range of 160–200 kg h⁻¹.

The composition of the switchgrass, grown in southern Iowa, is given in Table 1.

A 5 L min⁻¹ slipstream from the gasification stream was used to evaluate gas cleaning and hydrogen enhancement. The gasifier was fluidized with air at an equivalence ratio between 0.25 and 0.35, which maintained the reactor in the temperature range of 700 to 760 °C. The feed rate of switchgrass during these tests was in the range of 160–200 kg h⁻¹. The composition of the switchgrass, grown in southern Iowa, is given in Table 1.

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Table 2. Composition of Catalysts

<table>
<thead>
<tr>
<th>catalyst</th>
<th>active components</th>
<th>promoter/carrier</th>
<th>binder</th>
<th>trace contaminants</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni-based ICI-46-1 high-temperature shift, LB</td>
<td>NiO</td>
<td>CaO, K_2O/SiO_2, Al_2O_3</td>
<td>Black Carbon 0.5%</td>
<td>S &lt; 80 ppm; Cl &lt; 100 ppm</td>
</tr>
<tr>
<td>high-temperature shift</td>
<td>FeO_2 78 ± 2%</td>
<td>Cr_2O_3 9 ± 2% CuO 2.0%; rare earth 1.5%</td>
<td>Black Carbon 0.5%</td>
<td>S &lt; 1000 ppm; Cl &lt; 100 ppm</td>
</tr>
<tr>
<td>low-temperature shift</td>
<td>CuO &gt; 29%</td>
<td>ZnO 41–47% Al_2O_3 8.1–10%</td>
<td>Black Carbon 0.5%</td>
<td>S &lt; 80 ppm; Cl &lt; 100 ppm</td>
</tr>
</tbody>
</table>

Table 3. Operating Conditions of Catalytic Reactors

<table>
<thead>
<tr>
<th>reactor</th>
<th>guard bed</th>
<th>steam reforming</th>
<th>high-temperature shift</th>
<th>low-temperature shift</th>
</tr>
</thead>
<tbody>
<tr>
<td>set-point temperature of reactor (°C)</td>
<td>650</td>
<td>800</td>
<td>400</td>
<td>200</td>
</tr>
<tr>
<td>temperature range of reactor (°C)</td>
<td>600–670</td>
<td>750–850</td>
<td>350–420</td>
<td>180–240</td>
</tr>
<tr>
<td>SV (h⁻¹)</td>
<td>900</td>
<td>3000</td>
<td>1500</td>
<td>1200</td>
</tr>
<tr>
<td>catalyst</td>
<td>calcined dolomite</td>
<td>ICI 46-1</td>
<td>Fe–Cr-based LB</td>
<td>Cu–Zn–Al-based B202</td>
</tr>
<tr>
<td>catalyst volume (mL)</td>
<td>200</td>
<td>60</td>
<td>120</td>
<td>150</td>
</tr>
<tr>
<td>inert material*(mL/mL)</td>
<td>20/20</td>
<td>20/20</td>
<td>20/50</td>
<td>25/50</td>
</tr>
</tbody>
</table>

* Volume of inert material above/below catalyst layer.

normal conditions. Since the focus of this work was the water-gas shift reactors, direct measurements of tar content in the producer gas was not performed although previous work indicated that heavy tar in the raw producer gas was on the order of 20 g m⁻³ and steam reforming was able to reduce heavy tar to undetectable levels.

Analysis of the extent of conversion of CO by the water-gas shift reaction requires careful consideration of changes in mole fractions of the reacting gases. Although the water-gas shift reaction is equimolar, the total number of moles at the change in moles of dry gas through the reactor is equal to the change in moles of CO through the reactor, it can be shown that the molar conversion of CO expressed in terms of X_CO is

\[ \chi = \frac{n_{CO_{inlet}} - n_{CO_{outlet}}}{n_{CO_{inlet}}} \]  

where \( n_{CO} \) designates the number of moles of CO at the inlet or outlet of the reactor as appropriate. Since the water-gas shift reaction is equimolar, the total number of moles at the inlet and outlet of the reactor are equal and it follows that

\[ \chi = \frac{X_{CO_{inlet}} - X_{CO_{outlet}}}{X_{CO_{inlet}}} \]  

where X designates mole fractions based on water vapor being one of the constituents of the gas mixture. In practice, gas analysis is performed on dry gas, the water vapor having been removed from the gas mixture before analysis; thus, it is convenient to define mole fractions that are based on moles of dry gas:

\[ X'_{CO} = \frac{n_{CO}}{n_{DG}} \]  

where \( n_{DG} \) is the total moles of dry gas. Solving eq 3 for \( n_{CO} \), substituting this expression into eq 1, and recognizing that the change in moles of dry gas through the reactor is equal to the change in moles of CO through the reactor, it can be shown that the molar conversion of CO expressed in terms of \( X'_{CO} \) is

\[ \chi = \frac{X'_{CO_{inlet}} - X'_{CO_{outlet}}}{X'_{CO_{inlet}}(1 + X'_{CO_{outlet}})} \]  

This formula was used to calculate CO conversions presented in the results.

The guard bed was designed to capture fine particulate, absorb hydrogen sulfide, and steam-reform some of the heavy tars in the producer gas. It plays an important role in protecting the nickel catalyst in the steam-reforming reactor, which is susceptible to coking by heavy tars and poisoning by hydrogen sulfide. Calcined dolomite was used in the guard bed reactor. No attempt was made to regenerate the dolomite, which is an inexpensive sorbent.

The tar reactor converts light tars into carbon monoxide and hydrogen. ICI 46-1, a Ni-based catalyst produced by Imperial Chemical Industry, was used in the steam reforming (tar cracking) reactor. A Fe–Cr-based LB catalyst, employed commercially in fertilizer factories in China, was manufactured according to China National Patent No. ZL 96102477.1 for use in the high-temperature water-gas shift reactor. A Cu–Zn-based catalyst B202 was used in the low-temperature shift reactor. The chemical compositions of the shift catalysts are given in Table 2.

All three catalysts were reduced prior to evaluation of catalytic activity. This was done by treating them with producer gas in the amount of 1 L/min along with steam injected at a steam/gas ratio of 0.8. The Ni-based ICI-46 catalyst in the tar-cracking reactor was reduced at a temperature of 700 °C, while the Fe–Cr-based catalyst in the high-temperature shift reactor was reduced at 250 °C and the Cu–Zn-based catalyst in the low-temperature shift reactor was reduced at 180 °C. Judging from the hydrogen content exiting the reactors, the Fe–Cr-based catalyst was substantially reduced after 20 min while the Cu–Zn-based catalyst required 45 min. Reduction was assumed complete when the hydrogen concentration exiting the reactor stopped increasing and reached a steady-state concentration. The catalysts were readily reduced by producer gas without requiring the addition of hydrogen as a reducing agent.

Table 3 details operating conditions of the reactor, including the amount of inert material added above and below the catalyst in the reactor. During trials with the steam reformer and water-gas shift reactors, producer gas was passed through the gas conditioning system at a flow rate of 3.0 L/min with steam added to achieve steam/gas volumetric ratio of 1.2. The guard bed was operated at a temperature T_GB equal to about 650 °C and space velocity of 900 h⁻¹ while the steam reformer (tar reactor) was operated at a temperature T_TR equal to about

Gas composition was measured after each of the three catalytic reactors and CO conversion calculated for each of the water-gas shift reactors. Upon completion of the trials, both fresh and spent catalysts were characterized by BET specific surface and porosity and by X-ray photoelectron spectroscopy (XPS). Analysis by XPS was performed to check for coking and poisoning of the catalysts by chlorine and sulfur. Specific surface area and porosity were tested using ASAP 2010 with analysis adsorptive N2 at 77.35 K. Analysis by XPS was performed using a Physical Electronics 5500 Multitechnique system with monochromatic Al and standard Mg/Al sources using sample sizes of less than 2 cm² cm. Although analysis of reduced catalyst would have been useful in understanding the development of pore structure, it was not possible with the present experimental facilities to remove a reduced catalyst sample from the reactors without it being exposed to the oxidizing environment of the atmosphere. Thus, only fresh and spent catalysts were analyzed.

Table 4. Gas Composition at Various Locations in the Gas Conditioning System

<table>
<thead>
<tr>
<th>gas composition²</th>
<th>raw gas</th>
<th>outlet steam reformer</th>
<th>outlet high-temperature shift</th>
<th>outlet low-temperature shift</th>
</tr>
</thead>
<tbody>
<tr>
<td>H₂</td>
<td>8.6 ± 0.2</td>
<td>19.4 ± 0.2</td>
<td>23.7 ± 0.1</td>
<td>26.7 ± 1.9</td>
</tr>
<tr>
<td>CO</td>
<td>14.3 ± 0.5</td>
<td>9.0 ± 0.2</td>
<td>1.4 ± 0.5</td>
<td>0.11 ± 0.04</td>
</tr>
<tr>
<td>CO₂</td>
<td>18.0 ± 1.0</td>
<td>20.5 ± 0.2</td>
<td>26.8 ± 0.1</td>
<td>27.4 ± 0.03</td>
</tr>
<tr>
<td>CH₄</td>
<td>4.5 ± 0.1</td>
<td>3.36 ± 0.02</td>
<td>3.1 ± 0.1</td>
<td>1.9 ± 0.9</td>
</tr>
<tr>
<td>C₂H₆</td>
<td>1.5 ± 0.05</td>
<td>0.28 ± 0.01</td>
<td>0.08 ± 0.05</td>
<td>0.13 ± 0.08</td>
</tr>
<tr>
<td>CO shift conversion</td>
<td></td>
<td></td>
<td>83%</td>
<td>98.7%</td>
</tr>
<tr>
<td>tar content (g/Nm³)</td>
<td>19.5</td>
<td></td>
<td>(b)</td>
<td>(b)</td>
</tr>
</tbody>
</table>

² Gas composition is dry basis (vol-%) measured by gas chromatography. ³ No heavy tar by observation.

Table 5. Atomic Concentrations of Catalysts As Determined by X-ray Photoelectron Spectroscopy (XPS)

<table>
<thead>
<tr>
<th>Ni-based catalyst</th>
<th>Fe–Cr-based catalyst</th>
<th>Cu–Zn-based catalyst</th>
</tr>
</thead>
<tbody>
<tr>
<td>fresh</td>
<td>used</td>
<td>fresh</td>
</tr>
<tr>
<td>Carbon C 1s</td>
<td>9.93</td>
<td>14.18</td>
</tr>
<tr>
<td>Oxygen O 1s</td>
<td>60.61</td>
<td>52.44</td>
</tr>
<tr>
<td>Sulfur S 2p</td>
<td>0.00</td>
<td>0.15</td>
</tr>
<tr>
<td>Chlorine Cl 1p</td>
<td>0.23</td>
<td>0.34</td>
</tr>
<tr>
<td>Nickel Ni 2p</td>
<td>8.55</td>
<td>9.70</td>
</tr>
<tr>
<td>Aluminum Al 2p</td>
<td>3.43</td>
<td>6.33</td>
</tr>
</tbody>
</table>

Figure 2. X-ray photoelectron spectra (XPS) of Ni-based catalysts using Physical Electronics 5500 equipped with Al Kα source (a) carbon 1s peak, (b) chlorine 2p peak, (c) sulfur 2p peak.

Figure 3. X-ray photoelectron spectra (XPS) of Fe–Cr-based catalysts using Physical Electronics 5500 equipped with Al Kα source (a) carbon 1s peak, (b) chlorine 2p peak, (c) sulfur 2p peak.

Upon completion of the trials, both fresh and spent catalysts were characterized by BET specific surface and porosity and by X-ray photoelectron spectroscopy (XPS). Analysis by XPS was performed to check for coking and poisoning of the catalysts by chlorine and sulfur. Specific surface area and porosity were tested using ASAP 2010 with analysis adsorptive N₂ at 77.35 K. Analysis by XPS was performed using a Physical Electronics 5500 Multitechnique system with monochromatic Al and standard Mg/Al sources using sample sizes of less than 2 cm × 2 cm. Although analysis of reduced catalyst would have been useful in understanding the development of pore structure, it was not possible with the present experimental facilities to remove a reduced catalyst sample from the reactors without it being exposed to the oxidizing environment of the atmosphere. Thus, only fresh and spent catalysts were analyzed.
3. Results and Discussion

Table 4 presents gas composition at different points in the gas conditioning system. The raw producer gas entering the system contained 8.6 vol-% H₂, 14.3 vol-% CO, 18.0 vol-% CO₂, 4.5 vol-% CH₄, and 1.5 vol-% C₂H₄. The producer gas contained about 19.5 g/Nm³ heavy tar. Exiting the steam reformer, the average gas composition was 19.4 vol-% H₂, 9.0 vol-% CO, 20.5 vol-% CO₂, 3.36 vol-% CH₄, and 0.28 vol-% C₂H₄. No condensable (heavy) tar was detectable at the exit of the steam reformer. As might be expected, the reaction between steam and tar increased the hydrogen content of the producer gas. Although steam reforming might also be expected to produce CO, this gas actually decreased 5.3 vol-%, indicating that the water-gas shift reaction is occurring even at the elevated temperature of the steam reformer. The steam reformer substantially reduced the concentration of C₂H₄, but only moderately reduced CH₄. Although CH₄ is expected to be more resistant to catalytic cracking than C₂H₄, the most probable reason for its persistence arises from thermodynamic equilibrium of the steam reforming reaction for methane:

\[
\text{CH}_4 + 2\text{H}_2\text{O} \rightarrow \text{CO}_2 + 4\text{H}_2 \quad (5)
\]

This reaction is strongly endothermic and highly reversible. Thus, complete reaction favors high temperatures and low partial pressures of CO₂. At the operating conditions of the tar reactor in this study (750–850 °C, 20.5 vol-% CO₂) a few percent of CH₄ in the product gas is not surprising. Temperatures approaching 1000 °C would be required to substantially reduce methane.

Exiting the high-temperature shift reactor, the average gas composition was 23.7 vol-% H₂, 1.4 vol-% CO, 26.8 vol-% CO₂, 3.1 vol-% CH₄, and 0.08 vol-% C₂H₄. The high-temperature shift reactor reduced CO content by 7.6 vol-%, representing 83% conversion. The concentration of CH₄ is essentially unchanged through the
high-temperature shift reactor, while the concentration of C2H4 decreases substantially. The fact that CH4 is essentially unchanged gives some confidence that the high-temperature shift reactor was operated with adequate steam. Otherwise, the Fe-Cr-based catalyst has a tendency to be over-reduced by H2 and CO to form metallic iron, which catalyzes the methanation reaction:

$$\text{CO} + 3\text{H}_2 = \text{CH}_4 + \text{H}_2\text{O} \quad (6)$$

Exiting the low-temperature shift reactor, the average gas composition was: 26.7 vol-% H2, 0.11 vol-% CO, 27.4 vol-% CO2, 1.9 vol-% CH4, and 0.13 vol-% C2H4. The total decrease in CO content through the shift-reactor system of 8.9 vol-% represents an overall CO conversion of 98.7%. The overall increase in H2 due to the combined action of the steam reforming and shift reactors is 18.1 vol-%. Within the uncertainty of the measurements, the concentrations of CH4 and C2H4 were not significantly affected through the low-temperature shift reactor.

Table 5 summarized the atomic concentrations (mol-%) of carbon, oxygen, sulfur, chlorine, and metals found on fresh and spent catalysts as determined by X-ray photoelectron spectroscopy (XPS). Figures 2–4 illustrate the X-ray photoelectron spectra of carbon, sulfur, and chlorine for the Ni catalyst, Fe-Cr catalyst, and Cu-Zn catalyst, respectively. Comparison of fresh and spent catalysts indicates that carbon accumulated on all three kinds of catalysts to some extent. Coking was most serious on the Cu-Zn catalyst where the relative amount of carbon almost doubled during the gasification test. Coking on the Ni catalyst was also serious, with carbon content increasing by nearly 40%. Accumulation of sulfur of atomic concentration on the catalysts was less than 0.2 mol-% while chlorine accumulation represented about 1.0 to 2.5 mol-%. Clearly, some hydrogen chloride and hydrogen sulfide in the raw producer gas broke through the guard bed and deposited on the catalysts. Although the reactors did not show any evidence of catalyst deactivation, the design of the guard bed needs to be improved to protect the metal catalysts from these trace contaminants.

Table 6 summarizes the specific surface and average pore diameters of catalysts as determined by BET analysis. In all cases, the specific surface area of spent catalyst greatly decreased compared to fresh catalyst. The average pore diameter of Ni-based catalyst decreased while it increased for the other two catalysts.

4. Conclusions

A gas conditioning system consisting of a steam reformer and two stages of water-gas shift reaction operating in conjunction with a switchgrass-fueled, air-blown gasifier was able to upgrade the hydrogen content of raw producer gas from 8.6 vol-% to 26.7 vol-%. Carbon monoxide concentration of 14.3 vol-% in the raw producer gas was reduced to less than 0.2 vol-%. Carbon monoxide conversion in the high-temperature shift reactor reached 83% while the overall conversion in the two-stage shift reaction system reached 98.7%. Although the steam reformer destroyed essentially all condensable tars and significantly reduced C2H4, it reformed very little of the CH4, which may reflect thermodynamic limitations as a result of inadequate temperature and too-high partial pressure of CO2.

Characterization of the catalysts by X-ray photoelectron spectroscopy showed that coke and small quantities of sulfur and chlorine deposited on the catalysts. BET analysis revealed losses in micropores and mesopores. Although no sign of catalytic deactivation was evident during the tests, these changes indicate the need for improvements in the design of the guard bed.

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