Clean Hydrogen Production via Novel Steam-Air Gasification of Biomass

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ABSTRACT:
Gasification converts biomass into $H_2$ and CO rich product gas by using air, oxygen and/or steam as reaction agent. An experimental and modelling study of hydrogen production was conducted using a laboratory scale counter-current fixed bed gasifier with preheated air and steam as the gasifying agents. Commercial charcoal was used as the fuel and the reactor bed was maintained at temperatures of 750-960$^\circ$C by external heaters. $H_2$ (17-28vol\%), CO (5-11vol\%) and CO\textsubscript{2} (18-21vol\%) forms a large portion of the synthesis gas produced depending on the ratio of the gasifying agent. The experimental work was coupled with both equilibrium and kinetic modelling to predict the equilibrium composition, conversion rates and temperature profile.

KEYWORDS : gasification, hydrogen, synthesis gas, kinetic modelling, equilibrium modelling.

1. INTRODUCTION

The demand for hydrogen is increasing due to the amount of hydrogen needed in refineries and the urgent need for a sustainable and renewable green energy. The Kyoto Protocol imposes emission targets under which the UK has agreed to the reduction of carbon emission by 12.5% on the 1990 level, by 2010 [1]. The shift towards a low CO\textsubscript{2} emission would mitigate the method of hydrogen and synthesis gas production from the common catalytic reforming of natural gas towards the production of hydrogen from water electrolysis or thermal conversion processes. The use of gasification to provide fuel for motor vehicles is an old art developed during the World War 2 where the shortage of petroleum products in Europe led to the use of a fixed bed gasifier unit attached to the front of a vehicle [2]. Similarly, the current energy and environmental situation led to many major companies such as Toyota, Daimler-Benz and General Motors investing in hydrogen-powered vehicle.

There are many types of gasification processes with a broad range of reactor types and operating conditions. The end-use of the gasification product and the economic factor are the primary factors that govern the selection of a particular type of gasification process. The gas turbine application is very sensitive to the quality of the gas and only extreme low levels of contaminants, principally tar, can be tolerated. This has traditionally rule out the application of counter-current gasification although the overall efficiency of the process is higher compared to the co-current gasification.

The three main parameters in the operation of a gasifier are the pressure, reactant and temperature. Gasification at high pressure increases the overall reaction rates at the expense of added complexity and lower formation of carbon monoxide and hydrogen. The gasification process comprises of both endothermic and exothermic reactions. The changes in temperature were found to influence the product gas composition [3]. Operation at high temperature increases the reaction rate especially for chemical reaction rate controlled processes. The presence of methane, steam and carbon dioxide in the product gas is favoured by low temperatures and high pressures whereas the presence of hydrogen and carbon monoxide is favoured by high temperatures and low pressures. Equilibrium consideration points to a high temperature, low pressure operating regime unless methane is the desired product [4].

The positive effect of an increase in reaction temperature on the tar content have been reported by several authors [5-8]. It is found that homogeneous secondary tar reactions become important at temperatures
higher than 650°C, indicated by the increasing concentrations of pyrolysis gases. In the investigation carried out by Morf [5], the gravimetric tar yield was found to decrease with increasing reactor temperatures during homogeneous tar conversion. Therefore, in view of the positive effect of higher bed temperature on thermal tar cracking, work on high temperature gasification is underway. High bed temperatures in an air/steam gasifier can be achieved by either raising the feed temperature or by means of external heating. The latter is applied in this research to maintain the bed temperature at about 750-960°C.

The design of this gasifier is part of a two stage integrated unit that aims ultimately at utilizing biomass fuel such as wood chips or fuel pellets derived from agricultural waste. The gasifier differs from conventional system as it produces practically tar-free gases. The high operating temperature acts as a catalyst for the pyrolysis of tar to carbon and gas. Proposal is underway to combine this unit with a first stage pyrolysis chamber. The high exit gas temperature opens up opportunity to combine thermal gasification with Stirling engines for use in small combined heat and power plant due to increase in electricity generation efficiency (>15%). Another important feature of this gasifier is the rotating grate at the bottom of the system that allows ash particles to be removed from below the bed for a more efficient operation.

Mathematical modelling has always been a useful tool to provide detailed information on the temperature profile, gas compositions, bed height, pollutant formation and combustion stoichiometry. The three main types of models typically used for gasification system are the equilibrium, kinetic and Computational Fluid Dynamic (CFD). The type of model chosen depends on the objective and experimental information available. The gasification experiment was modelled using two different mathematical approaches in this study. The chemical equilibrium calculations were carried out using version 2 of NASA Glenn’s computer program Chemical Equilibrium with Application (CEA) [2002]. The Fluid Dynamic of Incineration Combustion Code (FLIC) developed at the Sheffield University Waste Incineration Centre (SUWIC) to model packed bed combustion of solid fuel was adopted to model the gasification process [9-11].

2. EXPERIMENTAL SETUP

2.1 Gasification System

The experimental work was carried out using a batch type counter-current and updraft fixed-bed reactor at atmospheric pressure as presented in Fig 1. The reactor is a vertical cylindrical chamber suspended from a weighing scale. The height of the reactor is 1.5m with an inner diameter of 200mm and an interior tube of 4mm thick constructed with 316-stainless steel. The gasifier was heated electrically by ultra-high temperature, helically wound ceramic ribbon heaters. The grate at the bottom of the furnace was a perforated cone made with Inconel, with the centre slightly higher than the sides. A gear was fastened to the grate so that it can be rotated manually from the outside of the furnace to discharge ash if required.

The reactants (preheated steam and air) were fed into the gasifier through an L-shaped stainless steel tube located 100mm below the grate. Several outlet holes were located at the side of the tube to allow a uniform air flow across the cross-sectional area. The air was preheated electrically to 150°C. Saturated steam at temperature of 100°C was supplied from a steam generator. The temperature profiles were measured by a set of thermocouples located in the centre of the gasifier. The first thermocouple was placed just above the grate with the other 5 thermocouples placed 20mm apart from each other. This was followed by another 2 thermocouples placed 50mm apart.

The flue gas exits from the top of the furnace into two shell and tube condenser units to cool the gases down before gas measurements were taken. The condensed liquid which captures solid particles entrained by the gas flows down the tubes into collection bottles. The outlet gases are then flowed through a demister consisting of a glass cylinder filled with glass wool. This ensures that the outlet gases are sufficiently clean for online gas-chromatographic analysis (PCG2000) equipped with thermal conductivity detector (TCD) with a span time of 570s. The CO, CO₂ and O₂ were also measured using an MGA 3000 gas analyser. The GC and the gas analyser were connected to a data logger together with the thermocouples.
2.2 Test Conditions

The fuel used for the gasification tests was commercial lumpwood charcoal, sized manually to about 3mm in diameter. The furnace was initially filled with 4kg of lumpwood charcoal giving a bed height of about 348mm and preheated to about 700°C under the influence of nitrogen to prevent any oxidation reaction. The mass loss during the heating up of the charcoal is about 20% wt as the initial material contains 2.5% wt moisture and 20% wt volatile. Table 1 lists the results of the standard fuel analyses for the materials before and after preheating.

<table>
<thead>
<tr>
<th>Samples</th>
<th>Before preheating</th>
<th>After preheating</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Proximate analysis</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Moisture</td>
<td>2.59</td>
<td>0.86</td>
</tr>
<tr>
<td>Volatile matter</td>
<td>20.46</td>
<td>3.86</td>
</tr>
<tr>
<td>Fixed Carbon</td>
<td>74.15</td>
<td>91.81</td>
</tr>
<tr>
<td>Ash</td>
<td>2.81</td>
<td>3.47</td>
</tr>
<tr>
<td><strong>Ultimate analysis</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>75.32</td>
<td>90.66</td>
</tr>
<tr>
<td>H</td>
<td>3.39</td>
<td>0.71</td>
</tr>
<tr>
<td>O*</td>
<td>15.91</td>
<td>4.30</td>
</tr>
<tr>
<td><strong>Gross Calorific Value (MJ/kg)</strong></td>
<td>28.45</td>
<td>30.32</td>
</tr>
</tbody>
</table>

Table 1: Lumpwood charcoal analysis.

The supply of gasifying agents started when the bed temperature of the gasifier reached the target temperature. The operating conditions are as listed in Table 2. The effect of air and steam were investigated for the air flow rate ranging from 220kg/m²hr to 294kg/m²hr and steam flows of 95-196kg/m²hr. The test runs are named according to the volume of each reactant supplied. For example, the “A90-S30” run refers to the
gasification using 90l/min of air (220kg/m²hr) and with steam making up 30%wt of the total input reactant (preheated air and steam).

<table>
<thead>
<tr>
<th>Air flow rate (lit/min)</th>
<th>Steam flow rate (kg/m²hr)</th>
<th>Mass percentage of steam (%)</th>
<th>Case Name</th>
</tr>
</thead>
<tbody>
<tr>
<td>90</td>
<td>220.60</td>
<td>30</td>
<td>A90-S30</td>
</tr>
<tr>
<td>90</td>
<td>220.60</td>
<td>35</td>
<td>A90-S35</td>
</tr>
<tr>
<td>90</td>
<td>220.60</td>
<td>40</td>
<td>A90-S40</td>
</tr>
<tr>
<td>100</td>
<td>245.04</td>
<td>25</td>
<td>A100-S25</td>
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<td>245.04</td>
<td>30</td>
<td>A100-S30</td>
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<td>245.04</td>
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<td>100</td>
<td>245.04</td>
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<td>A100-S40</td>
</tr>
<tr>
<td>110</td>
<td>269.55</td>
<td>30</td>
<td>A110-S30</td>
</tr>
<tr>
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<td>269.55</td>
<td>35</td>
<td>A110-S35</td>
</tr>
<tr>
<td>110</td>
<td>269.55</td>
<td>40</td>
<td>A110-S40</td>
</tr>
<tr>
<td>120</td>
<td>293.94</td>
<td>40</td>
<td>A120-S40</td>
</tr>
</tbody>
</table>

Table 2: Process conditions for the test cases.

### 3.0 MATHEMATICAL MODELLING

#### 3.1 Chemical Equilibrium

The chemical equilibrium calculations were carried out using version 2 of NASA Glenn’s computer program Chemical Equilibrium with Application (CEA) [2002]. CEA is based on a non-stoichiometric formulation to calculate reaction equilibrium based on the minimisation of Gibbs free energy [12]. The program obtains chemical equilibrium composition for the assigned thermodynamic state such as temperature and pressure. The CEA programme was used to calculate the chemical equilibrium for three cases (A110-S30, A110-S35 and A110-S40) from the gasification test. The gas phase composition was assigned to take into account the main species in the outlet gas such as CH₄, CO, CO₂, H₂, H₂O and also minor species that might be present, C₂H₆, C₃H₈, HCN, O₂ and NH₃. The elemental composition of the solid fuel (commercial charcoal) was set according to the ultimate analysis and the enthalpy of formation of the commercial charcoal was calculated to be -254kJ/mol based on the higher heating value.

The main objective of this calculation is to give an insight into the chemical equilibrium based on the carbon conversion rate of the gasification experiments.

#### 3.2 Comprehensive Mathematical Modelling

The FLIC code is a one-dimensional, unsteady mathematical model which includes heat and mass transfer for gas, solid and reaction rate kinetics for the combustion and gasification of char and gaseous species. In this calculation, major bed properties, i.e. temperatures of gas and solid phases inside the bed, gas compositions and solid compositions is described on dimensionally as a function of bed height. The surface-volume averaged diameter used is the shape of a particle assuming it to be spherical. The individual bed processes is viewed to take place layer by layer, from the bed top to the bottom. The FLIC code was previously designed and used to simulate packed bed combustion of solid fuels [9-11].

In the simulation, the whole bed is divided into 60 sections along the bed height and time-dependent solutions were sought. The relaxation factor was set to 0.5 and the convergence criteria at 0.005. The calculated space height was set to 500mm. The model was used to simulate a laboratory-scale plant, which is cylindrical in shape with a 0.1m internal diameter and 1.5m length. The bed properties for the charcoal samples were those of experiments, as listed in Table 1. The particle size was taken as 30mm and the bed porosity Φ was taken as 0.69. The wall temperature was set and maintained at 725°C assuming no heat losses to the surrounding as the equipment was well-insulated in the laboratory experiment. The steam and air flow rates were selected in accordance with the experimental conditions and at temperature of 150°C. The calculations were made based on the reaction kinetics given by Hobbs [13] for char/CO₂, char/steam , char/O₂ and char/H₂. The kinetics for the water-gas shift reaction was based on the values given by Biba [14].
4.0 RESULTS AND DISCUSSION

4.1 Dynamic behaviour

Figure 2 shows the temperatures measured at several positions along the centre of the gasifier for Case A110-S30. The temperature profile in the bed was fairly steady and reproducible as this case was repeated several times. The temperature profile for the rest of the test cases also showed similar behaviour with the bottom of the bed the hottest due to the different chemical processes stratified along the reactor height. The temperature measurement at the bottom of the bed reflects the removal of ashes. The temperature at y=46mm is the highest at the start of the experiment. The lower temperature at the bottom of the bed (y=6mm) could be due to the contact with steam and air cooling the bed. The bottom temperature starts to drop 20°C during t=442-449min. The temperature decrease corresponded to the time when the ashes were removed from the bottom of the bed. The temperature at y=6mm started escalating after the discharge of the ashes while the temperature at the upper section starts declining (y=46mm, y=86, y=106mm) due to the bed movement. It can also be seen from the dynamic temperature oscillations at the bottom section of the bed that the conversion processes were very active in this region. The maximum bed temperature observed for this case was about 980°C.

4.2 The Effect of Reactant Supply on the Production of Hydrogen Gas: Composition of Dry Product Gas and HHV

The product gas contains the combustible and non-combustible gases, moisture and soot. Figure 3 shows the dry product gas composition and higher heating values (HHV) of five representative gasification conditions operated with 40%wt of steam and air flow rates of 50l/min, 90l/min, 100l/min, 110l/min and 120l/min. The showed the effect of higher input reactant on the gasification reactions under a constant proportion of steam to air ratio. The synthesis of H₂ (28%) and CO (10%) was the highest for Case A90-S40 and declines as the total flow rate of reactant increases. The concentration of CO₂ was about 21% for all the cases except Case A90-S40. The evolution of H₂ and CO was considerably low for Case A50-S40 due to the limited amount of reactant supplied. CH₄ concentration was negligible with the highest concentration of only 0.32% shown in Case A100-S40. The average bed temperature for all the five cases increased with a higher input of reactant. Therefore, it can be said that the decrease in H₂ and CO arise from more air entering the reactor resulting in the exothermic burning of these two gases in the oxidation zone and thus, contributing to the increase in bed temperature. Therefore, it can also be postulated that the air flow rate have an exceeding influence over the production of H₂ compared to the steam flow rate as the higher mass flow rate in the input steam did not result in a higher production of H₂ in the test cases. The result obtained in this study agrees
with the observations made by Midili [15] in his experiment with an air-blown downdraft gasifier where the flow rate of hydrogen gas and temperatures at oxidation zone decreased arising from more air entering the reactor bed. Thus, combustion was said to occur in the reactor bed rather than the gasification. The dry product gas has around 19-38% of combustible gases with higher heating values of 2-4.4MJ/kg. The heating value of the flue gas decreases with increasing reactant feed because of the low amount of CO and H₂. The char conversion rates for the four cases (cases A90-S40, A100-S40, A110-S40 and A120-S40) with higher input reactants varied in the range of 40-50kg/m³/hr.

Table 3 presents the conversion ratios of hydrogen gas from char gasification via steam-air blown downdraft gasification technique. The flow rate of hydrogen and combustible gas from char was calculated using the mass balance of the input reactants, conversion rate and flue gas concentration. The production ratio of the gases was calculated based on the mass of gases produced per mass of char converted.

<table>
<thead>
<tr>
<th>Case</th>
<th>Hydrogen gas from char (kg/h)</th>
<th>Combustible gas from char (kg/h)</th>
<th>Hydrogen gas production ratio (%)</th>
<th>Combustible gas production ratio (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A50-S40</td>
<td>0.057</td>
<td>0.179</td>
<td>10.83</td>
<td>34</td>
</tr>
<tr>
<td>A90-S40</td>
<td>0.232</td>
<td>1.394</td>
<td>14.56</td>
<td>87.5</td>
</tr>
<tr>
<td>A100-S40</td>
<td>0.22</td>
<td>1.176</td>
<td>13.90</td>
<td>74.3</td>
</tr>
<tr>
<td>A110-S40</td>
<td>0.142</td>
<td>0.692</td>
<td>10.90</td>
<td>54</td>
</tr>
<tr>
<td>A120-S40</td>
<td>0.139</td>
<td>0.705</td>
<td>10.30</td>
<td>52</td>
</tr>
</tbody>
</table>

Table 3: The conversion ratios of char to hydrogen gas.
4.3 The Effect of Increasing Steam to Air Ratio on Hydrogen Production: Composition of Dry Flue Gas and HHV

The effect of steam flow rate on the gas composition was investigated in order to establish the role of the different reactions in the gasification process. The air flow rate is kept constant at 110 l/min while the steam composes of 30, 35 and 40%wt of the total reactant flow rate. A higher formation of H₂ was attained as the feed steam was increased from 30% in Case A110-S30 to 35% in Case A110-S35. The molar fraction of CO decreases at the expense of the production of CO₂. The bottom bed temperature was found to decrease from 900 to 880°C as the steam flow rate was increased from 30% to 40%. The char conversion rate was also found to be slower at a higher flow rate of steam feed. Two gasification reactions that have significant effects on the formation of H₂ are the water gas reaction \((\text{H}_2\text{O})_g + \text{C}_g \rightarrow \text{CO}_g + \text{H}_2(g)\) and the water-gas shift reaction \((\text{CO})_g + \text{H}_2\text{O})(g) \leftrightarrow \text{CO}(g) + \text{H}_2(g)\). The increase of H₂ is favoured by the increase of the exothermal character of the water-gas shift reaction which begins and predominates between 500 to 600°C, and the water gas reaction which becomes significant at temperatures from 1000°C upward [15]. Assuming that the water-gas shift reaction and the water gas reaction are the main gasification reactions, the ratio of CO₂/CO would provide an indication of the main reaction governing the production of H₂, which in the case of this work is the water-gas shift reaction.

![Dry flue gas composition graph](image)

**Figure 4:** Dry flue gas composition and its HHV for all cases for 110 l/min of air (preheated) and with steam at 30, 35 and 40% of the total reactant input.

The increase in H₂ evolution in Case A110-S35 may be due to the addition of steam in the reacting. This enhances the water-gas shift reaction and as consequence, result in a decrease of CO and an increase of CO₂. The higher formation of H₂ with increasing steam was also reported in work carried out by Lucas [16] where the steam-biomass reaction rates was enhanced relative to the competing reactions by increasing the reactive agent steam in the feed gas. A parallel increase in H₂ yield was seen with increasing steam/biomass was found in the gasification experimental investigation by Turn [17].

From Figure 4, the H₂ concentration declined to about 18% when the steam feed rate was further increased to 40%. The drop in H₂ evolution could be due to the influence of the lower bed temperature. Both the reactor temperature and feed temperature was proven to have an effect on the H₂ yield [3, 17, 18]. The water-gas and Boudouard reactions are endothermic and thermodynamically favourable at high temperature. The lowering of bed temperature due to the addition of steam as in Case A110-S40 could have possibly reduce the influence of these two reactions resulting in both a slower char conversion rate, H₂ and CO₂ evolution. Similar trend was reported by Franco [18] whereby the H₂ formation reaches a maximum with increasing
steam/biomass ratio and then declines as the ratio was further increased. In his work, the CO was found to decrease significantly while no significant changes was observed for CO2.

The lack of existence of higher hydrocarbon, CxHy and negligible amount of CH4 (0.1-0.2%) promises a tar free synthesis gas by utilizing a high temperature two-stage gasification process in which a large part of the higher hydrocarbons and contaminant were removed during the pyrolysis step. The result of which is a clean synthesis gas, without the necessity of excessive gas cleaning. Work is currently underway to combine this high temperature gasifier with a first stage pyrolysis chamber using char for catalytic cracking of tar.

Figure 5 shows the effect of additional steam on the H2/CO ratio at varying air flow rates. The H2/CO molar ratio was quite high for steam and air gasification of the commercial char, ranging up to 3.6. The results show that as the steam feed increased, the ratio of H2/CO increased sharply. The increase in H2/CO ratio in the product gas was mainly due to the decrease in concentration of CO with increasing steam feed.

Therefore, the H2/CO in the synthesis gas can be adjusted to suit different applications by manipulating the steam input.

4.4 Chemical Equilibrium Modelling

Figure 6 shows the results from the equilibrium calculation compared to the experimental measurements. Comparison was made based on the measured average temperature in the gasifier. The composition of the wet flue gas was calculated through mass balances as experimental measurements were made based on the dry flue gas composition.

On the overall, the calculation prediction of the influence of steam on the composition flue gas showed good agreement with the trend obtained from the experiment measurements. An increase of steam input causes a drop in the CO and H2 concentration and a step up in H2O concentration for cases carried out with 110l/min (A110).
4.5 Detailed Mathematical Modelling (FLIC)

Figure 6: Comparison between the experimental and calculations showing the influence of steam cases with air feed of 110l/min (preheated).
Figure 7: Calculated dry flue gas composition in the bed vs. reaction time. (a) CO; (b) H₂; (c) CO₂

The calculation demonstrates that CO is the dominant component at 100mm from the bottom of the bed compared to the other two components (H₂ and CO₂) due to the partial combustion of the carbonaceous char material (Figure 7). H₂ is formed further along the bed due to the gasification reactions with water leading to a release of CO and CO₂. The simulations showed layer of inactive material at the bottom of the bed due to the accumulation of ash. The accumulation of ashes at the bottom of the bed resulted in the loss of sensible heat in heating up the ashes in the calculation. Figure 8 below shows the calculated solid bed temperature for Case A110-S30. The temperature at the bottom of the bed (y=4mm, y=21mm) decreases with time due to the loss in sensible heat in ashes. The temperature profile obtained from the calculation was about 150°C lower compared to the experimental measurements (Figure 2). However, the calculation was able give a close representation of the dynamic change in temperature along the stratified gasifier.

Figure 9 compares the calculated and measured wet flue gas composition for various steam feed. Both the calculation and measurements showed similar trend in the flue gas composition as flow rate of the feed steam was increased. The calculations showed a slight over-prediction of CO and a much lower release of CO₂ and H₂. This could be due to the under estimation of the CO combustion or water-gas shift reaction. The slow prediction of these exothermic reactions also explains the lower bed temperature in the calculations compared to the experiment. The calculated mass burning rate at 40kg/m²hr which is slower compared to the experimental mass burning rate of about 49kg/m²hr would in consequence result in a lower bed temperature.
CONCLUSION

The following conclusion from the results of the high temperature air and steam gasification experiments and modelling can be summarised as follows:

1. The percentage of hydrogen gas was the highest among all the combustible gases.
2. The production of hydrogen gas was influenced by the air to steam ratio and the total feed flow rate. By entering more air in the oxidation zone, the production of hydrogen and carbon monoxide decreased as combustion occurred in the reactor bed. The addition of steam could lower the bed
temperature resulting in a decrease of synthesis gas production. The ratio of H₂/CO could be manipulated by adjusting the steam feed.

(3) The conversion rate obtained in the experimental work ranged in the region of 40-50kg/m²hr depending on the reactant input.

(4) The hydrogen yield obtained in the counter-current reactor was about 10-14kg/hr of hydrogen from 100kg of biomass char.

(5) The rotating grate applied in the counter-current reactor successfully reduced the sensible heat losses in the ashes and helped maintain a high temperature and active region at the bottom of the bed.

(6) The production of methane was negligible and no higher hydrocarbons were detected. Therefore, proposed combination of this gasification chamber with a first stage tar-cracking pyrolyser would allow a low tar content in the synthesis gas.

(7) The calculated results agreed qualitatively with experiments and showed similar trends in the performance of the gasifier with varying flow rates of the reactant feed.

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