Fuel-Flexible Hybrid Solar Coal Gasification Reactor

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Abstract:

Current power generation systems in the United States are based on combustion of fossil fuel technologies and emit carbon dioxide, which is costly to mitigate and has a negative impact on the environment. Coal gasification is a conversion process that utilizes vast, domestic coal resources to generate cleaner syngas for power generation and has gained significant attention due to its reduced carbon emissions. Carbon emissions reduction has motivated research into power generation from renewable resources, such as solar and wind. However, these technologies are more expensive than current power generation technologies, and are a long term goal to securing energy independence. Hybridizing solar energy from concentrated solar power towers, a renewable resource technology, to provide process heat for coal gasification, a fossil fuel based technology, will create an intermediate solution that reduces carbon emissions while maintaining power generation capacity.[1] Preliminary energy balance calculations indicate that producing the stoichiometric amount of steam required for the gasification reaction from solar energy, such as concentrated solar power towers, can reduce the energy required for gasification by 29%. Furthermore, through an innovative fuel-flexible gasification reactor, the remaining energy can be provided by low-carbon emitting fuels, such as natural gas, thereby reducing the carbon emissions of the gasification process by 60%. The fuel-flexibility of the reactor is supported by an allothermal gasification reactor wherein the combustion and gasification section are thermally linked by high temperature heat pipe. A prototype annular heat pipe with an integrated fluidized bed gasification reactor was fabricated to verify the feasibility of the design. Through gas chromatography analysis, this prototype demonstrated conversion of 88% of the input steam to hydrogen at 900 °C, with an effluent composition comprised of 1.3:1 H_2:CO ratio. Therefore, through modeling and experimentation, the hybridization of solar energy and coal gasification demonstrated an intermediate solution that supports the transition between fossil fuel based power generation to systems based on more sustainable renewable resources.

Introduction

Gasification is an endothermic thermo-chemical process that converts steam and carbon, into syngas, a flammable mixture of hydrogen and carbon monoxide. This process occurs at high temperature (>700 °C) and a multitude of carbonaceous materials, such as coal and biomass, can be gasified. In the gasification of carbonaceous materials, the carbonaceous material is pyrolyzed to char which is subsequently gasified to produce syngas. The produced syngas can be used in a combined cycle to produce electricity or converted to higher value added products, such as transportation fuels, through the Fischer-Tropsch process.[2] Thus, utilizing coal in the gasification process can generate energy and valuable products from domestic resources.

Two classes of gasification reactors that are commonly used are autothermal and allothermal reactors. In autothermal gasification reactor technologies, a mixture of steam and air or steam and oxygen are introduced into the reaction chamber where the carbonaceous material is combusted under oxygen deficient conditions providing heat to pyrolyze the carbonaceous material to char which is subsequently gasified producing syngas. However, the effluent stream not only contains syngas but also contains combustion and pyrolysis by-products that need to be removed in order to purify the syngas for use in further power generating or chemical processes. Alternatively, the combustion or heat source can be applied externally in allothermal gasification reactor technologies, thereby separating combustion by-products from the effluent stream. In either reactor technology, approximately 37% of the energy required for the gasification process is dedicated to producing steam, 8% is required to heat the coal to gasification reaction temperatures, and the remaining 55% is required for the endothermic gasification reaction. Providing this energy through combustion can produce significant carbon emission.

In a climate requiring reduction in carbon emissions from power generation processes, hybridization of clean alternative energy sources and gasification technologies can help meet these carbon reduction goals while matching energy demands.[3] Key to this hybridization approach is new gasification reactor technologies. Therefore, an innovative allothermal gasification reactor was designed and demonstrated that utilizes high temperature heat pipe technology to thermally connect the combustion and gasification zones, as depicted in the conceptual drawing in Figure 1.

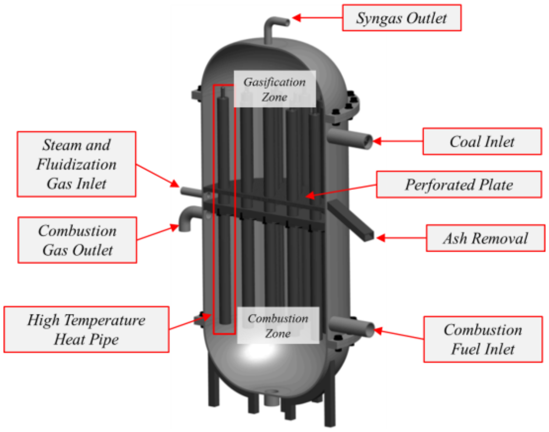

Figure 1. Model of the conceptual allothermal gasification reactor thermally linking the gasification and combustion zones by high temperature heat pipes thereby enabling the required process heat to be supplied by low-carbon emitting fuels.

This design enables the energy required for the gasification reaction to be provided by the combustion of low-carbon emitting fuels, such as natural gas, thereby imparting fuel flexibility to the gasification process. Furthermore, the steam required for the gasification reaction is provided through solar energy, reducing the overall energy requirements of the gasification process, and the associated quantity of fuel required to provide the energy. Solar technologies, such as concentrated solar power towers, can produce steam at temperatures of 560 °C.[4]

A preliminary energy balance of this reactor configuration indicates that producing the stoichiometric required amount of steam at a temperature of 560 °C with solar energy sources would reduce the energy required for the gasification process by 29%. In return, this reduces the amount of fuel required for combustion to provide the energy for the remaining processes, such as coal pyrolysis and gasification. Thus, utilizing the combustion of low-carbon emitting fuels, such as natural gas, to provide the remaining heat required for the remaining processes can reduce the carbon emissions of the gasification process by 60%.

Essential to enabling this new reactor design are high temperature heat pipes that thermally connect the combustion zone and gasification zone while keeping the two zones hydraulically separated. A heat pipe is a passive two-phase heat transfer device wherein an evacuated metal envelope material lined with a wick is filled with a working fluid. Heat is applied at the evaporator of the heat pipe, thereby evaporating the working fluid which is transported along the axial direction of the heat pipe where it is subsequently condensed liberating heat. The condensed liquid working fluid is then pumped back to the evaporator by the capillary pressure of the wick structure.[5] This evaporation/condensation cycle of heat pipes is depicted in Figure 2.

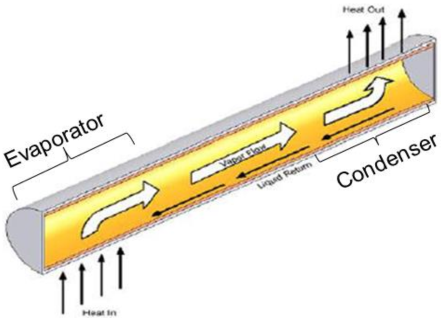

Figure 2. Illustration of a heat pipe demonstrating heat input at the evaporator, vapor flow of the working fluid along the axial direction of the heat pipe, heat removal and condensation of the working fluid at the condenser, and return of the liquid working fluid by capillary pressure of the wick.

The passive, two-phase heat pipe has an effective thermal conductivity of >10,000 W/mK, thereby enabling effective heat transport and isothermal axial temperatures over the length of the heat pipe without active control requirements.[*] Thus, heat pipes operating in the range of 700-1100 °C integrated into an allothermal gasification reactor will efficiently transfer heat between the combustion and gasification sections of the gasification reactor.

Experimental Setup

To evaluate the capability of using high temperature heat pipes in coal gasification processes, a subscale prototype annular heat pipe with an integrated fluidized bed reactor was fabricated and tested at temperatures between 800 and 900 °C and the conversion of the steam and carbon into hydrogen and carbon monoxide were evaluated by gas chromatography. A computer aided drawing (CAD) model and picture of the fabricated subscale gasification reactor are presented in Figure 3 and the design parameters of the annular heat pipe are presented in Table 1.

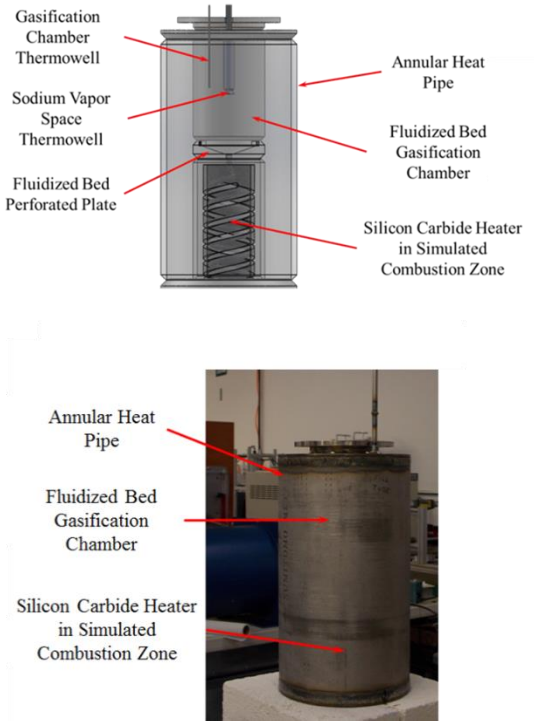

Figure 3. ***(Top)*** *CAD model and* ***(Bottom)*** *picture of the fabricated subscale prototype annular heat pipe with an integrated fluidized bed reactor.*

Table 1. Subscale Annular Heat Pipe Design Parameters

Parameter	**Value**
Envelope Material	Inconel 600
Gasification Reactor Diameter	3.07"
Fluidized Bed Height	6.00"
Outer Tube OD	6.63"
Working Fluid	Sodium
Operating Temperature	500-900 °C
Wick Material	100x100 Stainless Steel Mesh Screen

As shown in Figure 3, the vapor space of the heat pipe was formed by welding together two concentric Inconel 600 tubes, thereby creating an annual vapor space for the sodium working fluid. The top and bottom half of the interior of the annular heat pipe were separated by a perforated plate, wherein the top half of the annular heat pipe (*i.e.* the condenser of the heat pipe) became the fluidized bed gasification reactor and the bottom half (*i.e.* the evaporator of the heat pipe) was a simulated combustion zone heated by a silicon carbide heater.

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[*] More details on heat pipes and their capabilities at www.1-act.com.

Prior to conducting gasification experiments, the proper operation of the annular heat pipe was evaluated by measuring the axial temperature distribution at five locations along the outer tube of the heat pipe, which is presented in Figure 4, with the inert argon fluidization media flowing at a superficial velocity such that bubbling fluidization occurred in the charcoal bed.

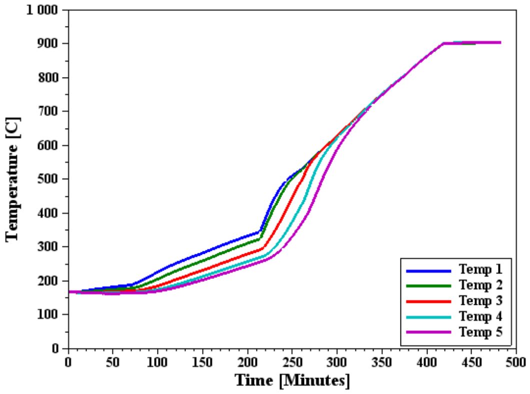

Figure 4. Axial temperature distribution at five locations along the length of the annular heat pipe with integrated fluidized bed reactor demonstrating an isothermal temperature distribution and proper heat pipe operation.

As shown in Figure 4, the axial temperature distribution converges at 700 °C and maintained a maximum axial temperature difference of 2 °C up to the operating temperature of 900 °C, which demonstrated the proper operation of the annular heat pipe.

After demonstrating proper heat pipe operation, the annular heat pipe gasification reactor was assembled into the test apparatus shown in Figure 5, where inert argon fluidizing gas and steam were injected through the perforated plate and through the carbon bed. The effluent exited through the top of the reactor, where it was collected for analysis by gas chromatography (GC).

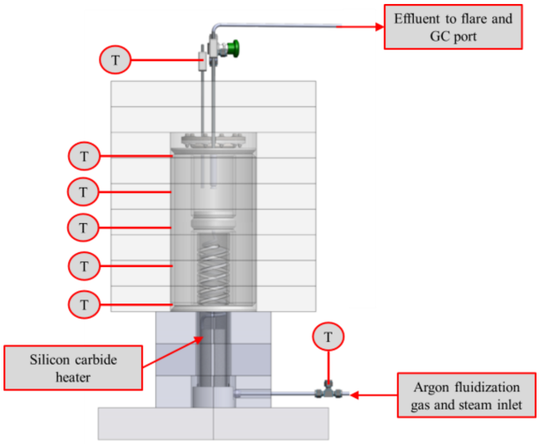

Figure 5. Illustration of experimental gasification test apparatus.

To eliminate inconsistencies in testing from impurities in coal samples, charcoal (Sigma Aldrich product number C3014) was used as a coal substitute. The steam required for the gasification reaction was generated by pumping de-aerated water through a syringe pump onto a section of heated tube, where water was boiled and the argon/steam mixture was controlled to a temperature of 130 °C. This apparatus allowed precise control over the entering steam mass flow rate by controlling the mass flow rate of water injected onto the heated tube. The argon/steam mixture was then pumped through a coil tube within the simulated combustion zone, where it was pre-heated to operating temperature before being distributed across the perforated plate for fluidization through the charcoal bed.

Results

While the objective of the experiment was to create syngas through gasification, carbon dioxide and hydrogen could have been formed from reaction of carbon monoxide and steam through the water gas shift reaction. The gasification and water gas shift reactions are presented in Equation 1 and Equation 2, respectively. Summing these two reactions, the overall conversion of the charcoal bed and steam can be represented by the net reaction in Equation 3. Thus, evaluating the ratios of CO/H_2, $2CO_2/H_2$, and H_2/O yield information about the dominant conversion process within the fluidized bed gasification reactor.[6]

$$C + H_2O \rightarrow CO + H_2 \qquad \textit{Equation 1}$$

$$CO + H_2O \rightarrow CO_2 + H_2 \qquad \textit{Equation 2}$$

$$C + \beta H_2O \rightarrow (2-\beta)CO + (\beta - 1)CO_2 + \beta H_2 \qquad \textit{Equation 3}$$

Gasification reactions were conducted at temperatures of 800, 850, and 900 °C. Prior to testing, the gasification reactor was brought to steady state temperature while the charcoal bed was being fluidized by the inert argon fluidization gas. Once steady state conditions were achieved, a blank injection of the effluent inert fluidizing gas was analyzed by GC to ensure that no side reaction were occurring between the fluidizing media and the charcoal bed. Steam was then injected into the fluidization stream and the composition of the effluent was analyzed via GC at five minute intervals for six samples at steady state operation. A sample chromatogram of the blank injection and gasification effluent are presented in Figure 6, which demonstrates the fluidization gas was inert to the charcoal bed and that gasification occurred during injection of steam thereby producing hydrogen and carbon monoxide.

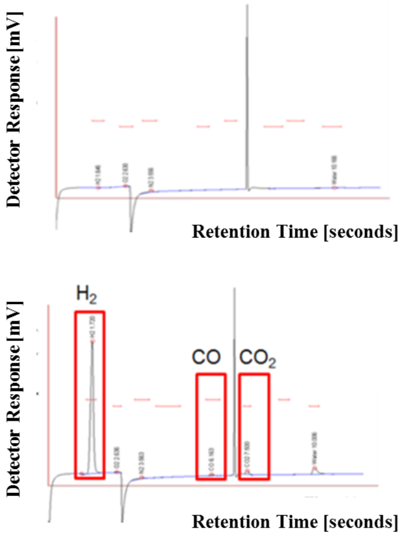

Figure 6. ***(Top)*** *Gas chromatogram of blank injection exhibiting no response for the formation of hydrogen, carbon monoxide, and carbon dioxide.* ***(Bottom)*** *Gas chromatogram collected during gasification reaction indicating the formation of hydrogen, carbon monoxide, and carbon dioxide.*

During all reaction scenarios, the steam was the limiting reactant and therefore was used to determine the percent yield of hydrogen and carbon monoxide based on their respective concentrations determined from GC analysis. The percent yield of hydrogen and carbon monoxide for gasification reactions conducted at 800, 850, and 900 °C with a steam mean residence time (also known as space time) of 2 seconds, are presented in Figure 7.

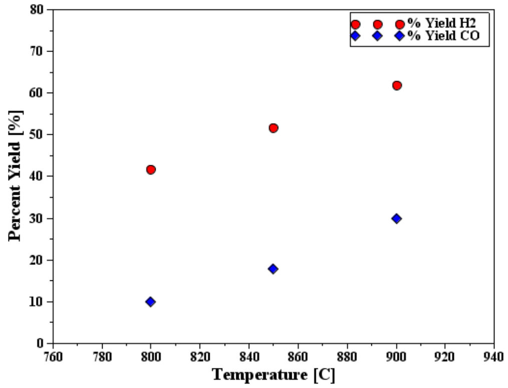

Figure 7. Percent yield of hydrogen and carbon monoxide at gasification temperatures of 800, 850, and 900 °C with a steam mean residence time of 2 seconds, demonstrating a 60% conversion of steam and charcoal to hydrogen and 30% conversion to carbon monoxide at 900 °C.

The conversion of steam to hydrogen increased with temperature at a constant steam mean residence time of 2 seconds reaching a conversion of 60% at 900 °C. This trend demonstrated the effect of the reaction kinetics on the gasification process.

The ratios of the molecular species in the net chemical reaction in Equation 3 are presented in Figure 8, and provide information on the dominant conversion process (*i.e.* gasification or water-gas shift) in the annular heat pipe gasification reactor.

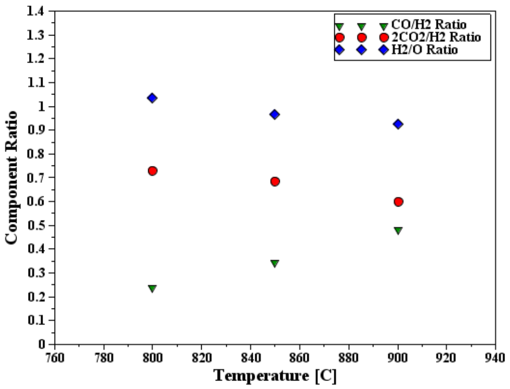

Figure 8. The ratio of molecular components in the effluent stream which demonstrates the conversion at all temperatures was dominated by the water-gas shift reaction. However, the proportion of the conversion by the steam gasification increases with temperature.

The evaluations of the molecular component ratios in Figure 8 demonstrated that the conversion to hydrogen was dominated by the water-gas shift reaction, indicated by larger ratio $2CO_2/H_2$ than the CO/H_2 ratio. At 800 °C approximately 20% of the conversion was conducted through the steam gasification reaction while the remaining 80% of the conversion of hydrogen was conducted through the water-gas shift process. At 900 °C, approximately 43% of the conversion to hydrogen occurred through the steam gasification reaction while 57% of the conversion occurred through the water-gas shift reaction. Thus, while the water gas shift reaction was still the dominant conversion of steam and charcoal to hydrogen, the steam gasification reaction became a larger portion of the conversion as the temperature of the reaction increased. However, the steam gasification reaction is required to initiate the water-gas shift reaction, thereby demonstrating that the steam gasification reaction can be conducted by transferring process heat to the gasification reaction via high temperature heat pipes. Furthermore, the H_2/O ratio of approximately unity for all temperatures indicates that steam was the only oxidant in the conversion process demonstrating that impurities, such as oxygen did not contribute to the formation of carbon dioxide and confirms the presence of the water gas shift conversion process.

To maximize the conversion of charcoal and steam to syngas, the conversion at 900 °C was evaluated at mean steam residence times ranging from 1 to 5 seconds, as demonstrated in Figure 9.

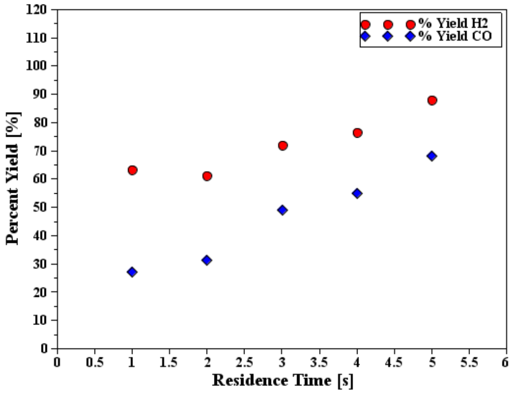

Figure 9. Percent yield of hydrogen and carbon monoxide at 900 °C with steam mean residence times from 1 to 5 seconds demonstrating an 88% conversion of hydrogen with a steam mean residence time of 5 seconds.

The conversion of steam and charcoal to hydrogen and carbon monoxide increased with reaction temperature reaching 88% and 62%, respectively, at 900 °C. Evaluating the molecular component ratios of the effluent stream, presented in Figure 10, provides insight into the dominant conversion processes.

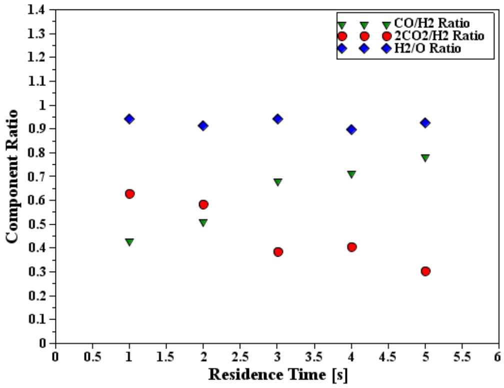

Figure 10. Molecular component ratios of the effluent stream at 900 °C at mean steam residence time of 1 to 5 seconds demonstrating the steam gasification process dominates at mean residence times greater than 2 seconds.

As demonstrated in Figure 10, the water gas shift was still the dominant conversion process at low residence time; however, at residence time greater than approximately 2.5 seconds, the steam gasification conversion process dominated the formation of hydrogen. The proportion of hydrogen formed through the steam gasification reaction increased with the steam mean residence time as the steam and carbon required in the gasification reaction had longer time to react thereby increasing the conversion to syngas.

Thus, the conversion of charcoal and steam to syngas through the steam gasification and water-gas shift reaction indicates that high temperature heat pipes can transfer heat from the combustion zone to the gasification zone in an allothermal gasification process. Coupling these developments with low-carbon emitting fuels will aide in meeting the goal to reduce carbon emissions in gasification processes.

Conclusion and Future Research

The conversion of 88% of the input steam into hydrogen through the gasification and water-gas shift reactions at temperatures of 900 °C in an annular heat pipe fluidized bed reactor demonstrates that gasification can successfully be conducted in an allothermal reactor with heat pipes thermally linking the gasification and combustion zones. Furthermore, this demonstration provides supporting evidence that heat pipes can be used to generate fuel-flexible gasification reactors that can utilize low-carbon emitting fuels to reduce carbon emissions from the gasification process. Lastly, utilizing alternative, renewable resources, such as solar energy, to produce the steam required for the gasification reaction can decrease the energy requirement for gasification thereby further reducing carbon emissions.

With the successful demonstration of a heat pipe based gasification reactor, further research should be focused on scaling up the reactor to match energy demands while maximizing the conversion efficiency of the reactor. This scale up research should contain efforts to optimize the conversion efficiency through arranging the heat pipes to maximize heat transfer from the combustion zone to the gasification zone and designing modular reactors that can be scaled to match energy demands. Furthermore, these research efforts should focus on integrating the scaled up gasification reactor with steam generated from renewable resources, such as in concentrated solar power towers, and utilizing waste heat as process heat for the endothermic gasification process.

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