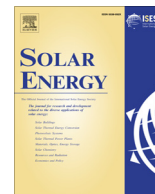




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A review of the state-of-the-art in solar-driven gasification processes with carbonaceous materials

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ABSTRACT

Gasification of carbonaceous feedstock with process heat derived from concentrated solar irradiation has been shown as a promising renewable pathway towards producing synthesis gas (mixtures of H₂/CO and some CO₂). The carbonaceous feedstock is upgraded in calorific content equal to the enthalpy change of the endothermic reaction, which results in the net storage of solar energy in a chemical form. The process is carbon-neutral when biomass is used and the feedstock is transformed into a fuel with applications to more efficient processes (e.g., Air-Brayton cycle), and the resulting syngas can be converted to liquid hydrocarbon fuels via known catalytic routes. A comprehensive summary is provided of the state-of-the-art in solar gasification, including thermodynamic and kinetic analyses and thermochemical reactor modeling, fabrication and testing for a range of carbonaceous feedstocks.

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1. Introduction

Each hour, approximately $4.3 \cdot 10^{20}$ J of energy from the sun impinge directly upon the earth's surface (Lewis and Nocera, 2006), providing the most abundant and exploitable renewable energy resource available. However, the sunlight arrives in a relatively dilute form, and it is intermittent and unequally distributed. These barriers can be overcome by concentrating and storing sunlight in a chemical form [i.e., solar fuels (Perkins and Weimer, 2009; Piatkowski et al., 2011; Steinfeld and Meier, 2004)]. Solar fuels such as H₂ and CO can be used to greatly alleviate worldwide dependency on fossil fuels. The combination of CO, CO₂, and H₂ constitutes synthesis gas (syngas): The precursor to liquid hydrocarbon fuels via Fischer-Tropsch synthesis (Corporan et al., 2007; Dry, 2002; Swanson et al., 2010) or other known catalytic routes required to drive transportation and power sectors. Syngas can also be directly combusted in a combined cycle or converted completely to H₂ via a water-gas shift reaction. Solar fuels can be produced in uninhabited desert regions with optimal solar irradiation and transported to population centers. Solar facilities like power towers with secondary concentrators coupled to arrays of heliostat fields and parabolic dishes are capable of concentrating solar irradiation in excess of 1000 suns (where 1 sun = 1 kW·m⁻²), optimal for driving solar thermochemical processes and cycles that produce syngas. Several thermochemical pathways have been pro-

posed and investigated to produce syngas by harnessing the power of the sun (Steinfeld, 2005).

Of special interest are solar technologies aimed at the gasification of carbonaceous feedstock (Perkins and Weimer, 2009; Piatkowski et al., 2011) with steam and/or CO₂, where the process heat needed to drive the reaction is derived from concentrated solar irradiation (i.e., solar-driven allothermal gasification). The carbonaceous feedstock is upgraded in calorific content equal to the enthalpy change of the endothermic reaction, which results in the net storage of solar energy in a chemical form. The carbonaceous feedstock is transformed into a fuel with a broader range of more efficient applications. This is an attractive alternative to conventional autothermal gasification processes where pure streams of O₂ are introduced into the system to combust a portion of the feedstock (i.e., oxy-combustion) for process heat to drive the reactions, thereby, lowering the overall energy content of the products. Solar-driven gasification processes are also relatively free of combustion products and tars that are associated with autothermal gasification.

The overall process is schematically depicted in Fig. 1, which shows the inputs of concentrated solar irradiation coupled to an array of carbonaceous feedstocks and steam and/or CO₂ to produce syngas. The resulting syngas can be directly combusted, shifted to H₂, or transformed into a liquid hydrocarbon for powering the transportation sector.

Solar-driven gasification for an array of carbonaceous feedstocks, both biomass and solid fossil fuels, has been performed to analyze theoretical thermodynamic performance of different

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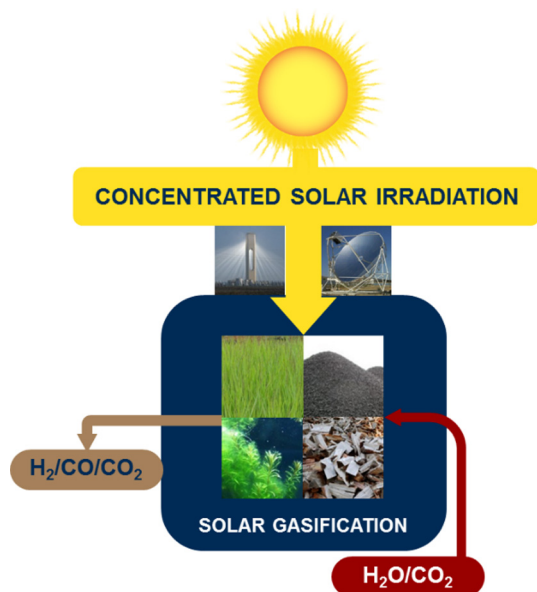


Fig. 1. Schematic of solar gasification processes with an array of carbonaceous feedstocks.

processes, to determine reaction mechanisms and associated kinetic parameters, and to develop solar thermochemical reactor technologies (*i.e.*, solar gasifiers) adapted to specific carbonaceous feedstocks for optimal performance over a range of scales. These results are contained in numerous publications that report work performed over many years. Our aim is to consolidate the principal findings and provide a comprehensive summary of the work to date in the area of solar-driven gasification.

2. Economic and thermodynamic analyses

The framework for comparing solar gasification to conventional gasification both with biomass was proposed in an economic assessment where four different solar routes compared to the conventional gasification route (Nickerson et al., 2015). Solar gasification was competitive with conventional gasification for certain scenarios. Due to the relatively low cost of natural gas in the United States in 2012, it was difficult for solar gasification to be cost effective without subsidies. However, natural gas is more expensive in other countries, making solar gasification a viable option.

Thermodynamic analyses have been employed to determine the compositions at chemical equilibrium for reactions of carbonaceous feedstock with H_2O over a range of temperatures and pressures and to impose first and second law thermodynamic constraints to assess the theoretical performance for a range of feedstocks. Equilibrium compositions were determined using Gibb's free energy minimization for a reaction of steam and lignite coal (LHV = 28,500 kJ/kg): $CH_{0.81}O_{0.23} + 0.77H_2O$ as a function of temperature for isobaric reactions at 1, 10, and 100 bars (Muroyama et al., 2014). The results are given in Fig. 2 for (a) C-containing species and (b) H-containing species. Only H_2 and CO are forecast at 1 bar for $T^{eq} > 1200$ K, and this temperature was shifted to an even higher temperature for increased pressures according to Le Chatelier's principle. At these temperatures, the reaction is highly endothermic, necessitating a large heat input that may be provided by concentrated solar irradiation.

Similar analyses were performed to forecast equilibrium compositions for a range of carbonaceous feedstocks, including anthracite coal (Zedtwitz and Steinfeld, 2003); anthracite, bituminous,

lignite coal, and peat (Ng and Lipiński, 2012); Flexicoke and PD coke (Trommer et al., 2005); beech charcoal (Piatkowski and Steinfeld, 2011); and Brazilian sugarcane bagasse (Kruesi et al., 2013) with similar equilibrium compositions forecast at 1 bar for $T^{eq} > 1200$ K.

First and second law thermodynamic analyses (which in combination represent an exergy analysis) were used to assess the potential for the cycle for a range feedstocks and operating conditions. Exergy analyses were performed to investigate the viability of two routes for extracting power from the products of solar-driven steam gasification: (1) a combined cycle where the resulting syngas is combusted with air and (2) water-gas shift reaction where the syngas is shifted to H_2 for use in a fuel cell. The results were compared to the direct combustion of the feedstock in a Rankine cycle assumed to be 0.35. The overall cycle efficiency (also described as the exergy efficiency) for the various routes was determined as:

$$\eta_{\text{cycle}} = \frac{\dot{W}_{\text{cycle}}}{\dot{Q}_{\text{solar}} + LHV} \quad (1)$$

where \dot{W}_{cycle} is the net power produced by the cycle; \dot{Q}_{solar} is the concentrated solar irradiation input to the cycle; and LHV is the lower heating value for the carbonaceous feedstock. Table 1 contains the cycle efficiencies for the various cycles of the analyses of different feedstock.

3. Kinetic analyses

Accurate determination of chemical kinetics is vital for designing and optimizing solar gasifiers that are customized for different feedstocks with a range of size distributions. The identification of the rate limiting mechanism(s) and determination of kinetic parameters is essential for designing and modeling efficient solar gasifiers. Pyrolysis represents an important reaction mechanism to consider in gasification processes as volatiles diffuse through pores and release tars. For a range of biomass feedstocks, the onset of pyrolysis occurs at 473–533 K where CO_2 and CO are released using thermogravimetry and varies based on biomass. At higher temperatures, CO, CO_2 , hydrocarbon vapors, tars and H_2 are produced (Raveendran et al., 1996) leaving behind fixed carbon. Due to the relatively high temperatures required for gasification, numerous studies have looked at determining the reaction mechanisms and kinetics to varying degrees of complexity for the fixed carbon that remains after pyrolysis. The coupling of the analyses with thermogravimetry or laboratory-scale reactors allows for temporal monitoring of conversion as a function of partial pressure of reactant and product gases and temperature. The temporal conversion of the feedstock can be determined from temporal mass loss as:

$$\alpha(t) = \frac{m_{\text{initial}} - m(t)}{m_{\text{initial}} - m_{\text{final}}} \quad (2)$$

where t is time; $m(t)$ is the temporal mass; m_{initial} is the sample mass at the beginning of the experiment; and m_{final} is the final mass after the experiment. The reaction rates for particles can expressed as a first-order model in differential or integrated forms in terms of α (or a related form) for changing particle surface area with specific knowledge of the initial effective surface areas, given as:

$$r_C = \frac{1}{1 - \alpha} \frac{d\alpha}{dt} \quad (3)$$

The rate has also been determined in other works in terms of mass of the carbonaceous feedstock and measured molar amount of products as:

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