

## Solar-driven biochar gasification in a particle-flow reactor

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### ABSTRACT

The steam-gasification of biochar with concentrated solar radiation is experimentally investigated using a 3 kW solar reactor prototype consisting of a cylindrical cavity-receiver containing an opaque tubular absorber. Particles of beech charcoal are used as the biomass feedstock in a continuous steam-particle flow through the tubular absorber. A reactor model that couples radiative, convective, and conductive heat transfer to the chemical kinetics is formulated and validated by comparing numerically computed and experimentally measured temperatures and carbon conversions. The simulation model is further applied to examine the thermal performance of 100 kW and 1 MW scaled-up solar reactor containing multiple tubular absorbers, yielding a theoretical maximum solar-to-chemical energy conversion efficiency of 39% and 50%, respectively. Major sources of irreversibility are associated with re-radiation losses through the cavity's aperture.

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### 1. Introduction

Solar steam-gasification of biomass makes use of concentrated solar energy to convert solid biomass feedstocks into high-quality synthesis gas (syngas) – mainly H<sub>2</sub> and CO – applicable for power generation in efficient combined cycles and fuel cells, or for Fischer-Tropsch processing of liquid biofuels [1,2]. Conventional autothermal gasification requires a significant portion of the introduced feedstock to be combusted with pure O<sub>2</sub> – usually produced by the energy-intensive air separation – to supply high-temperature process heat for the highly endothermic gasification reaction. For example, the energy required to gasify bituminous coal of LHV 34 MJ/kg is supplied by burning 35% of the injected coal mass [3]. In contrast, the solar-driven gasification eliminates the need for a pure stream of oxygen. Consequently, the gaseous products are not contaminated by the byproducts of feedstock combustion. Furthermore, the calorific value of the biomass feedstock is solar-upgraded by an amount equal to the enthalpy change of the reaction, resulting

in syngas with a negative CO<sub>2</sub> intensity. Ultimately, solar thermochemical gasification of carbonaceous feedstocks is a means of chemically storing intermittent solar energy in a dispatchable form. Solar pyrolysis and gasification of coke, coal, cellulose, and other carbonaceous materials was studied in directly irradiated fluidized-bed [4,5], vortex-flow [6,7], molten-salt pool [8], and packed-bed [3] solar reactors. Further, numerous studies have investigated the pyrolysis and gasification of biomass-derived char in non-solar reactors, e.g. in thermogravimeters [9–11], or fixed-bed microreactors [12,13].

Solar reactors for high-temperature thermochemical applications usually feature the use of cavity-type configurations to capture effectively the incident concentrated solar radiation entering through a small opening, the *aperture* [14]. Because of multiple internal reflections, the cavity approaches a blackbody and the optimum aperture size results from a compromise between maximizing radiation capture and minimizing re-radiation losses [15]. The direct irradiation of reactants was proven to provide efficient heat transfer to the reaction site [6]. However, a major drawback when working with controlled atmospheres is the requirement for a transparent window—a critical and troublesome component under high-pressure, severe gas environments, and scale-up designs. An alternative reactor design used in the present study that bypasses

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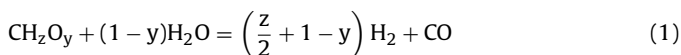
E-mail address: [aldo.steinfeld@eth.ch](mailto:aldo.steinfeld@eth.ch) (A. Steinfeld).

this problem is a cavity-receiver containing an opaque tubular absorber that encloses the reaction chamber [16]. The absorber is exposed to concentrated radiation entering through the cavity's aperture and to IR radiation emitted by the hot cavity walls, while heat is transferred by conduction to the reaction chamber. Such an indirectly irradiated reactor eliminates the need of a window at the expense of having a less efficient heat transfer through the absorber walls. Thus, the disadvantages are linked to the limitations imposed by the materials of construction of the absorber such as the maximum operating temperature, inertness to the chemical reaction, thermal conductivity, radiative absorptance, and resistance to thermal shocks.

In the present work, the steam-gasification of biochar is performed using the aforementioned reactor concept. Particles of beech charcoal are used as the biomass feedstock in a continuous steam-particle flow through the tubular absorber. The chemical thermodynamics and reaction kinetics are analyzed. A previously formulated heat transfer model [16] is applied to couple radiation/convection/conduction heat transfer to the reaction kinetics and validated by comparing numerically computed and experimentally measured temperatures and reaction rates. This model is further employed to examine the thermal performance of a scaled-up reactor for 100 kW and 1 MW solar radiative power input.

## 2. Thermodynamics and kinetics analyses

The overall process can be represented by the simplified net reaction:



Sulfur/nitrogen containing compounds and other impurities have been omitted from Eq. (1). The thermodynamic equilibrium composition of a system composed of a stoichiometric mixture of beech charcoal and water,  $\text{C}_1\text{H}_{0.418}\text{O}_{0.117}\text{S}_{0.0003}\text{N}_{0.006} + 0.883\text{H}_2\text{O}$ , at 1 bar and over a wide temperature range of interest is shown in Fig. 1. The HSC Outokumpu code [17] was used; species whose mole fraction is less than  $10^{-3}$  (e.g., HCN or  $\text{H}_2\text{S}$ ) have been omitted from the figure. At above 1400 K, the system at equilibrium consists of a gaseous mixture of 52%  $\text{H}_2$  and 48% CO. Also indicated in Fig. 1 is the enthalpy change of the reaction as a function of tem-

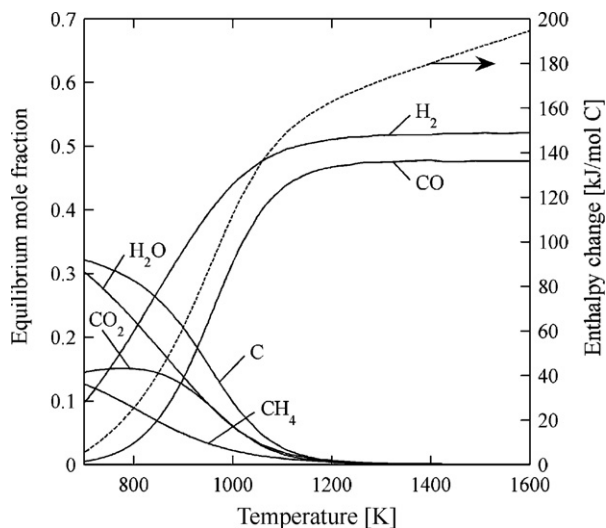


Fig. 1. Equilibrium composition of the system  $\text{C}_1\text{H}_{0.418}\text{O}_{0.117}\text{S}_{0.0003}\text{N}_{0.006} + 0.883\text{H}_2\text{O}$  as a function of temperature. Product species with mole fractions less than  $10^{-3}$  have been omitted. Also indicated is the enthalpy change of reaction (dashed line) when the reactants are fed at ambient temperature and the products are obtained in equilibrium at the reaction temperature.

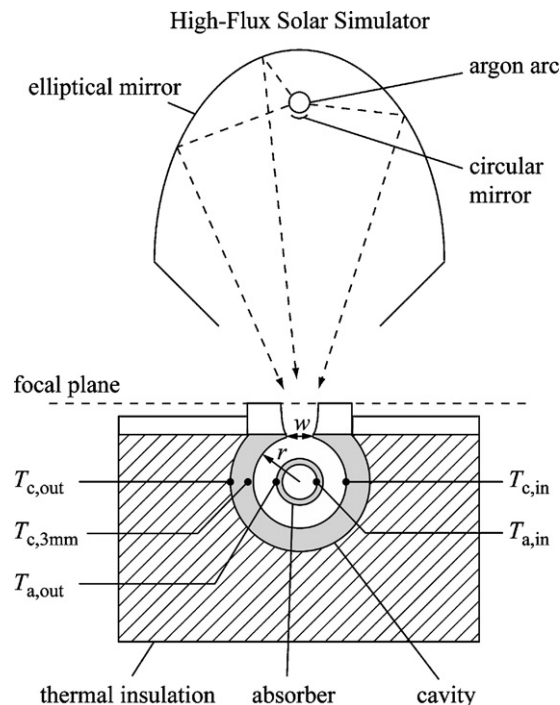
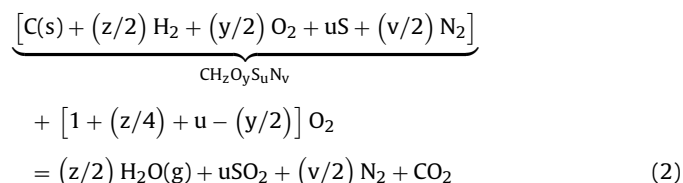


Fig. 2. Scheme of the solar chemical reactor configuration (cross-sectional view) at ETH's High-Flux Solar Simulator.

perature, when the reactants are fed at ambient temperature and the products are obtained in equilibrium at the reaction temperature. At 1400 K,  $\Delta H = 179.9\text{ kJ/mol C}$ . Note that since charcoal has no reference enthalpy, all enthalpy changes were calculated by assigning to charcoal the reference enthalpy of its elemental composition  $[\text{C}(s) + (z/2)\text{H}_2 + (y/2)\text{O}_2 + u\text{S} + (v/2)\text{N}_2]$  at 300 K, and further adjusting for the small offset between the heating value of charcoal (437.9 kJ/mol C, calculated from elemental composition [18]) and the enthalpy change of the combustion reaction at 300 K given by:



The reaction rate was determined by thermogravimetry [3]. Applying the shrinking spherical particle model for an unreacted spherical core [19], the rate law can be expressed as:

$$\frac{dX_C}{dt} = k'(1 - X_C)^{2/3} \quad (3)$$

where  $X_C = 1 - \dot{n}_C/\dot{n}_{0,C}$  denotes the carbon conversion and  $k'$  denotes the rate constant assumed to obey the Arrhenius law,  $k' = k_0 \exp(-E_a/R_u T)$ , with an apparent activation energy  $E_a = 43154\text{ J/mol}$  and a pre-exponential factor  $k_0 = 124.6\text{ s}^{-1}$ , the latter being adjusted for the experimental validation of the reactor model.

## 3. Solar reactor and experimental setup

A scheme of the reactor configuration is depicted in Fig. 2. The engineering design and construction has been previously described and applied to the thermal dissociation of ZnO [16]. Only the main features will be summarized here. It consists of a cylindrical cavity-receiver made of 10 wt%  $\text{YO}_2$ -stabilized  $\text{ZrO}_2$ , with an

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