



Co-processing methane in high temperature steam gasification of biomass

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HIGHLIGHTS

- ▶ CH₄ and biomass were reacted simultaneously in an entrained flow steam-gasifier.
- ▶ Equilibrium accurately predicts the syngas composition at 1500 °C; not at 1200 °C.
- ▶ ANOVA reveals temperature dominates syngas composition and conversion.
- ▶ Product syngas composition can be tuned for downstream processing.

ARTICLE INFO

Article history:

Received 14 August 2012
 Received in revised form 9 October 2012
 Accepted 10 October 2012
 Available online 23 October 2012

Keywords:

Gasification
 Entrained flow
 Methane reforming
 High temperature
 Concentrated solar

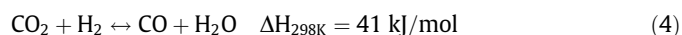
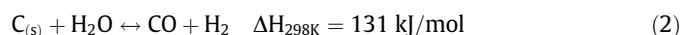
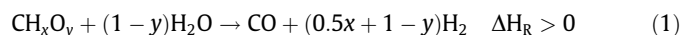
ABSTRACT

High temperature steam gasification/reforming of biomass-methane mixtures was carried out in an indirectly heated entrained flow reactor to analyze the feasibility of controlling the output composition of the major synthesis gas products: H₂, CO, CO₂, CH₄. A 2³ factorial experimental design was carried out and compared to thermodynamic equilibrium predictions. Experiments demonstrated the product gas composition is mostly dependent on temperature and that excess steam contributes to CO₂ formation. Results showed that with two carbon-containing reactants it is possible to control the gas composition of the major products. At 1500 °C, the equilibrium results accurately predicted the syngas composition and can be used to guide optimization of the syngas for downstream liquid fuel synthesis technologies.

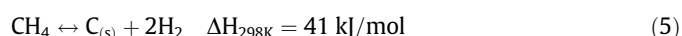
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1. Introduction

High temperature, steam-only thermochemical processing of biomass initially involves rapid pyrolysis into char, permanent gases, and condensable vapors. Condensable vapors continue to undergo secondary homogeneous reactions such as cracking and reforming to form more permanent gases. The resulting char particles undergo conventional heterogeneous gasification per Eqs. (1)–(3). The reverse water–gas shift reaction, Eq. (4), is also a significant gas phase reaction whose products are thermodynamically favored above approximately 850 °C. If the gasifier temperature is higher than about 1100 °C, little to no condensable species are present in the synthesis gas (syngas) product (Milne et al., 1998).

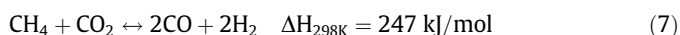
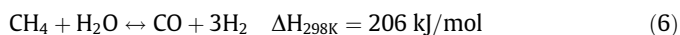


Several researchers (Adams and Barton, 2011; Dufour et al., 2009; Fiaschi and Carta, 2007; Perkins et al., 2010; Rodrigues et al., 2003; Song and Guo, 2007; Steinberg, 1997; Sudiro and Bertucco, 2007, 2009; Valin et al., 2009) have considered using methane, or natural gas, as an additive in thermochemical processes to increase the calorific value of the syngas or to improve combustion characteristics. Methane can be co-processed simultaneously with a carbonaceous solid (Adams and Barton, 2011; Perkins et al., 2010; Song and Guo, 2007; Sudiro and Bertucco, 2007, 2009), reacted in a parallel step to gasification (Adams and Barton, 2011; Borgwardt, 1997; Steinberg, 1997; Sudiro and Bertucco, 2009), added to the products of gasification and reacted further in a serial step (Adams and Barton, 2011; Dufour et al., 2009; Valin et al., 2009), or simply added to the products of gasification without further modification (Fiaschi and Carta, 2007; Rodrigues et al., 2003). The primary reactions methane undergoes in steam-only processing are outlined in Eqs. (5)–(7).

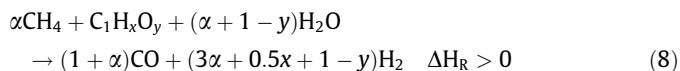


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Combining methane and biomass in single-step gasifier-reformer results in the overall reaction described by Eq. (8) where α is the methane-to-biomass ratio. Compared to Eqs. (1) and (8) reveals that the product gas composition is not only dependent on the biomass composition, but also on α .



While reactor configuration (fluidized bed, circulating fluidized bed, fixed bed, entrained flow, etc.) (Alvarez et al., 2011; Gomez-Barea and Leckner, 2010; Lee et al., 1996; Mandl et al., 2010; Umeki et al., 2010; Xie et al., 2012) and operating conditions (equivalence ratio, residence time, etc.) (Alvarez et al., 2011; Antal, 1981; Smoot and Brown, 1987; Yoon et al., 2011; Zhang et al., 2010) are known to affect the product gas composition, the combination of methane, biomass, and steam at high temperatures has been left unexplored.

In autothermal gasification, combustion of biomass with air or pure oxygen generates large amounts of CO_2 from combustion in order to provide the heat necessary for the endothermic gasification reactions (Smoot and Smith, 1985; Yoon et al., 2011). This CO_2 must be separated from the desired product gases downstream, typically through pressure swing adsorption, an energy-intensive process (Adams and Barton, 2011; Anex et al., 2010). If pure oxygen is used, air separation units may require 50–72% of the total onsite energy use and a significant initial capital investment (Adams and Barton, 2011; Liu et al., 2011; Swanson et al., 2010).

For processes that utilize syngas in downstream liquid fuel synthesis, the syngas is often modified in a water–gas shift reactor to obtain an H_2/CO ratio of approximately 2.0–2.1; this in turn increases the overall quantity of CO_2 to generate more H_2 (Adams and Barton, 2011; Liu et al., 2011). In a conventional gasification-to-liquid fuel process, the biomass-carbon utilization from start to finish is around 20–40% (Adams and Barton, 2011; Liu et al., 2011; Smoot and Smith, 1985; Swanson et al., 2010), where the balance is mostly CO_2 and is vented to the atmosphere.

Concentrated solar power (CSP) processes are well-known for converting solar thermal energy into more useful forms of energy and are poised for significant contributions in electricity generation (Coelho et al., 2010), gaseous and liquid fuel production (Steinfeld and Weimer, 2010), and even coffee bean roasting (Rosenblum, 2011). Using CSP for the thermochemical treatment of carbonaceous feedstock has also been considered by many researchers, and a review of many of these processes may be found in (Lede, 1999; Piatkowski et al., 2011; Steinfeld and Weimer, 2010). CSP has been demonstrated to provide sufficient process heat to drive the endothermic gasification reactions achieving temperatures in excess of 1200 °C, excluding the need for combustion of the feedstock, eliminating tar production, and increasing char conversion (Haussener et al., 2009; Lichty et al., 2010). A high temperature, adiabatic solar gasifier with added methane has a theoretical carbon-to-liquid fuel utilization greater than 95% and an operational CO_2 output of nil per kilogram of biomass.

In this paper, the technical feasibility of high-temperature, steam-only gasification of biomass with added methane is demonstrated. Thermodynamic equilibrium results are presented as a theoretical basis for the process and are used to guide the design of an experimental matrix. An electrically heated entrained flow reactor is used to simulate the conditions of an indirectly heated solar reactor to experimentally explore the effects of some operating conditions on yield and two performance indicators for the

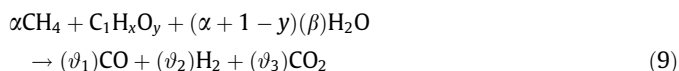
purposes of future work on reactor and plant design. The goal is to provide theoretical and experimental evidence for an H_2 -enriched syngas product with low CO_2 content and high carbon conversion, which cannot be obtained from any other known biomass gasification technology.

2. Methods

2.1. Thermodynamic equilibrium analysis

The thermodynamic computational software package FactSage was used for calculating equilibrium results. The calculations are based on a Gibbs free energy minimization technique and use an extensive database of physical and thermochemical properties of thousands of potential gaseous, liquid, and solid compounds to calculate the most stable products over large ranges of temperatures and pressures. Only major gas constituents, CO , CO_2 , H_2 , H_2O , and CH_4 , were included in the analysis.

The purpose of this analysis was to demonstrate the relationship between product composition and temperature using various hypothetical scenarios of reactant concentrations, specifically α and β as shown in Eq. (9) where $\beta \geq 1.0$, representing steam in excess of the stoichiometric amount required for a given quantity of methane and biomass. Minor products and impurities such as C_2+ hydrocarbons, halides, and alkali vapors were not considered in this study. The stoichiometric coefficients of the major products, ϑ_i , in Eq. (9) are unknown and depend on the equilibrium constants for all of the major reactions involved. Experimental studies at temperatures below 1200 °C showed that equilibrium predictions often overestimate the quantity of CO and H_2 in the product stream while underestimating the quantities of CO_2 , CH_4 , other hydrocarbons, char, and tar (Gomez-Barea and Leckner, 2010; Yoon et al., 2011). Above 1300 °C, equilibrium predictions were shown to be adequate and often used in simulations (Adams and Barton, 2011; Anex et al., 2010; Sudiro and Bertuccio, 2007)



2.2. Materials

Rice hulls (Sundrop Fuels Inc., Broomfield, CO) were used as the biomass source for experiments with ultimate and proximate analyses performed by Huffman Labs in Golden, Colorado (C, 37.6%; H, 5.5%; O, 37.2%; N, 0.98%; S, 0.10%; moisture, 4.5%; ash, 18.6%; HHV, 15156 kJ/kg). They were ground in a laboratory grinder and sieved to <150 μm . The overall moisture- and ash-free molecular composition was found to be $\text{CH}_{1.583}\text{O}_{0.6629}\text{N}_{0.0223}\text{S}_{0.0010}$ with a molecular weight of 24.56 g/gmol. For simplicity, the molecular formula $\text{CH}_{1.583}\text{O}_{0.6629}$ was used.

2.3. Experimental apparatus and procedure

A tubular, entrained flow reactor set-up was determined to be appropriate for pulverized fuel, high-temperature steam-gasification. An alumina reaction tube had an I.D. of 0.092 m and the hot zone length was 0.46 m. The reactor tube was indirectly heated by a graphite element, simulating the conditions that would be seen by an opaque tubular absorber similar to the configuration found in Lichty et al. (2010). Biomass and diluted, non-flammable methane entered the reactor via a 20–25 °C water-cooled lance and product gases exited the reactor through a quench zone with a water jacket at 15 °C. Unconverted solids and ash were separated using a gravity collection vessel and a 0.22 μm HEPA filter. Due to the difficulty of sampling char and the fact that it deposited

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