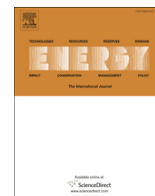




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## Thermodynamic analysis of biomass gasification for biomethane production

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### ABSTRACT

A thermodynamic analysis has been performed to predict the biomass gasification performance on the basis of the minimization of Gibbs free energy. The theoretical methane yield was further predicted based on the equilibrium in the downstream processes. A C–H–O ternary diagram is proposed to predict the boundary for the formation of carbon deposit. Three zones, namely carbon deposit, carbon-free and intermediate zone, are defined, with possible carbon formation or elimination mechanisms being postulated. The biomass gasification performance using different gasifying agents, namely H<sub>2</sub>O, CO<sub>2</sub>, O<sub>2</sub> or air, was then analyzed. Results showed that the steam addition is conducive to enhance carbon conversion and shrink the carbon deposit zone, but the use of air, O<sub>2</sub> and CO<sub>2</sub> will impose a negative impact on the methane yield and H<sub>2</sub>/CO ratio. The maximum methane yield can be achieved at the temperature at which carbon is completely converted. The effects of pressure are dependent on whether or not solid carbon is absent. Based on the analysis on thermodynamic and exergetic efficiencies, it is concluded that steam gasification is the preferred conversion scheme for biomethane production.

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### 1. Introduction

Compared to coal, natural gas is a cleaner energy source, but it is non-renewable and will one day be depleted. Another general characteristic of fossil energy sources is non-uniform geographical distribution, both globally and domestically. Thus, the disparity between supply and demand on the natural resources will continue for a long time, and becomes more intense with the development of economy and enhanced awareness of the environmental concerns about global warming. In China, there is a relatively abundant coal resource but very scarce natural gas resource. In 2007, the share of natural gas in China's energy mix is only 3.5%, while coal's share accounts for 69.5% [1]. Globally, natural gas supplied approximately 24% of the primary energy consumed around the world in 2011. To solve the serious air pollution problem encountered in major cities in China, the use of natural gas is being promoted as a substitute for coal and oil. With the increasing natural gas demand, the

supply–demand gap on the natural gas is predicted to be 186–210 billion cubic meters [2,3], presenting a huge challenge to China's gas supply.

To alleviate the rising demand on natural gas, synthetic natural gas can be generated from other domestic resources such as coal and biomass. Synthetic natural gas from coal has a large carbon and environmental footprints. In addition, the hydrogen sulfide and NO<sub>x</sub> byproducts are potentially harmful to human health, if not properly scrubbed or treated [4]. The production of coal-derived SNG is also water intensive. Due to the opposite distribution of water and coal sources in China, there is a potential water shortage in the region with coal-to-SNG plants.

Another way to generate the substitute natural gas, bio-methane, is sourced from biomass. Bio-methane can potentially avoid the environmental issues and water shortage referred above. Biomass is renewable and carbon-neutral. So recently there has been a growing interest in biomethane production. There are two production pathways for bio-methane namely thermochemical gasification and anaerobic digestion. Two pathways are not competing each other due to the different requirements of feedstocks. For the latter, the capacity of Biogas plant is relatively small due to the local availability of the (wet) feedstock [5]. But the feedstock for the biomass gasification are diverse and abundant. So

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Bio-methane from biomass gasification is more promising to meet the demand on the natural gas. Additionally, there is an abundant biomass energy resource in China. But its primitive utilization, such as direct combustion, causes serious environmental issues and energy waste [6,7]. So it will be a clean and efficient pathway if the biomass is converted to biofuels via thermochemical gasification.

Recently, many activities on biomass gasification for the production of Bio-methane have been or being performed. Bench-scale, demonstration and commercial scale projects were proposed or built by Energy Research Center of the Netherlands (ECN), center for Solar Energy and Hydrogen Research (ZSW) and Paul-Scherrer Institute (PSI) etc. [8,9]. Among these programs, the main difference is the design of the gasifier and operating conditions [9]. The process is in general similar to the thermochemical routine to produce SNG from coal. As the largest exergy loss step, the gasifier performance strongly depends on the operating conditions. As a result, the thermodynamic efficiency (energy and exergy) and methane yield vary [9]. It is thus necessary to optimize the operating conditions of biomass gasification based on thermodynamics in order to achieve high biomethane yield and efficiency.

Thermodynamic analysis of biomass gasification involved thermodynamic equilibrium calculation [10–12] and thermodynamic efficiency (first-law and second-law) analysis [9,13–17]. Thermodynamic equilibrium calculation provided the compositions of target species under specific conditions, which is especially suitable for systems with unknown reaction mechanisms and precise chemical compositions like biomass and coal. For such multi-element system, it is prerequisite to achieve the higher thermodynamic efficiency and avoid the system being operated in the carbon deposit region. Though carbon deposit boundaries were given on such a C–H–O Ternary System [18,19] as well as a C–H–O–N–S system [11,20], possible mechanisms on carbon formation needed to be analyzed in combination with the specific operating condition. Furthermore, analysis of thermodynamic efficiency and optimization of operating parameters are also essential to achieve a high bio-methane yield in consideration of the subsequent downstream units.

It is the aim of this study to reduce the exergy loss in the bio-methane production process using biomass as the feedstock. Thermodynamic equilibrium calculations were performed for biomass gasification. The operating condition was optimized, under which carbon formation can be avoided and meanwhile a high methane yield and exergy efficiency can be achieved.

## 2. Methodology

The thermodynamic equilibrium model and the source code originally developed by Li [21] were adopted in this study. The model based on the non-stoichiometric approach was coded in Matlab software. The input biomass composition is treated as its equivalent elemental abundance. An iterative RAND algorithm was used to solve the equations involving elements in abundance to minimize the Gibbs free energy of the reactive system. The calculation accuracy strongly depends on thermodynamic input data like heat Capacity, enthalpy and entropy. So the database on correlation coefficients was re-established in the present study, with correlation factors being obtained from McBride et al. [22] in the polynomial form as given, respectively, below:

$$\text{Heat capacity : } \frac{Cp(T)}{R} = a_1 + a_2T + a_3T^2 + a_4T^3 + a_5T^4 \quad (1)$$

$$\text{Enthalpy : } \frac{H(T)}{RT} = a_1 + a_2\frac{T}{2} + a_3\frac{T^2}{3} + a_4\frac{T^3}{4} + a_5\frac{T^4}{5} + \frac{b_1}{T} \quad (2)$$

$$\text{Entropy : } \frac{S(T)}{R} = a_1 \ln(T) + a_2T + a_3\frac{T^2}{2} + a_4\frac{T^3}{3} + a_5\frac{T^4}{4} + b_2 \quad (3)$$

$$\text{Gibbs free energy : } G(p, T) = H(T) - T*S(T) \quad (4)$$

The calculation accuracy is improved. Furthermore other sub-processes were coupled into the numerical model such as water–gas shift, clean-up and methanation. On this basis, the bio-methane yield was predicted. The boundary for the formation of carbon deposit was predicted, and the optimal gasifying parameters were obtained to achieve the maximum biomethane yield and energy efficiency. Meanwhile possible reactions occurred under various operating conditions were proposed in combination with the shift on the compositions. In addition, exergy calculation methods were added into the model, and the post-processing program was introduced to calculate the gasification efficiency.

In the present study, the biomass fuel is represented by a general formula of  $\text{CH}_{1.4}\text{O}_{0.59}\text{N}_{0.0017}$ , which is also used in the equilibrium calculation [24]. In addition, the abundances of other elements are negligibly small [25]. The system was simplified to 32 gaseous species (C (g), CH, CH<sub>2</sub>, CH<sub>3</sub>, CH<sub>4</sub>, C<sub>2</sub>H<sub>2</sub>, C<sub>2</sub>H<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>, C<sub>3</sub>H<sub>8</sub>, H, H<sub>2</sub>, O, O<sub>2</sub>, CO, CO<sub>2</sub>, OH, H<sub>2</sub>O, H<sub>2</sub>O<sub>2</sub>, HCO, HO, N, N<sub>2</sub>, NCO, NH, NH<sub>2</sub>, NH<sub>3</sub>, N<sub>2</sub>O, NO, NO<sub>2</sub>, CN, HCN, HCNO) and one solid specie (C(s)).

## 3. Energy and exergy calculations

The energy balance for a system can be expressed as

$$Q - W + \left( \sum_i \frac{m_i}{M_i} H_i \right)_{in} - \left( \sum_i \frac{m_i}{M_i} H_i \right)_{out} = 0 \quad (5)$$

in which the heat flux  $Q$  is positive when the system absorbed heat from the environment. Meanwhile, work is positive when it is produced by the system.

The exergy destruction is as follows,

$$E_D = \left( \sum_i \frac{m_i}{M_i} e_i \right)_{in} - \left( \sum_i \frac{m_i}{M_i} e_i \right)_{out} + \left( 1 - \frac{T_0}{T} \right) Q - W \quad (6)$$

For biomass gasification, the heat loss and work generated or consumed were neglected in the present study.

Statistical correlations are used to calculate the heating value and chemical exergies of biomass. The higher heating value was predicted by the following correlation developed by Channiwala and Parikh (in MJ/kg) [26]:

$$\text{HHV}_{fuel} = 0.3491z_C + 1.1783z_H - 0.1034z_O - 0.0151z_N - 0.0211z_A \quad (7)$$

For the specific chemical exergy of dry biomass, the statistical correlation of Song and Shen [27] (in kJ Kg<sup>-1</sup>) was used as follows:

$$e_{fuel}^{ch} = 1812.5 + 295.606z_C + 587.354z_H + 17.506z_O + 17.735z_N - 31.8z_A \quad (8)$$

In Equations (7) and (8), only ultimate analysis data plus ash content were used in weight% on a dry basis, and the sulfur content is neglected.

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