

Process simulation and energetic analysis of different supercritical water gasification systems for the valorisation of biomass

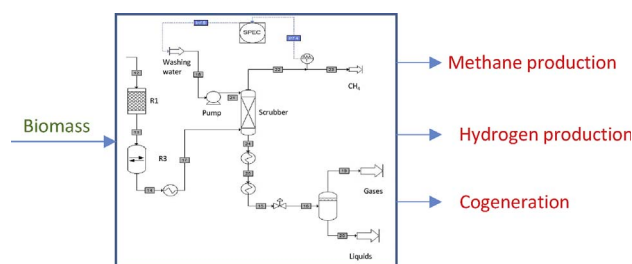


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GRAPHICAL ABSTRACT



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ABSTRACT

The energetic efficiency for the supercritical gasification of biomass is studied for three cases: hydrogen and methane production, heat and electricity cogeneration. Experimental results from the gasification of glucose are used to represent the gasifier. The other unit operations are simulated using the software ProsimPlus through thermodynamic equilibrium calculations. Simulations are conducted at different pressure, temperature and initial biomass concentration. The energetic and exergetic yields are calculated, as well as the minimum heat requirement estimated from a pinch analysis. All the results are then exploited to determine optimal conditions for two systems: adiabatic and isothermal. The optimal temperature ranges from 584 to 626 °C, whereas the optimal initial concentration ranges from 51 to 87 g L⁻¹. The system giving the best result in term of global energetic optimisation is methane production in isothermal conditions, followed by methane production in adiabatic conditions. The optimal energy efficiencies are 94% and 91% respectively.

1. Introduction

Among different processes of biomass conversion, an attractive way is gasification: the “dry” gasification, which operates at high temperature (up to 950 °C) and a pressure close to atmospheric pressure, and supercritical water gasification (SCWG) with temperature ranging from 450 to 600 °C and pressure up to 40 MPa. This last one is more specifically dedicated to wet biomass as no pre-drying step is needed.

Gasification in supercritical water can be used for the production of hydrogen [1–3]. However a mixture of carbon monoxide, hydrogen and methane are generally obtained depending on the operating conditions [4–6]. Gas compositions from experimental results for various biomass or chemical compounds are available and specific calculations have been performed to predict gas composition at thermodynamic equilibrium [7–9]. Equilibrium predictions are in accordance with experimental data when light gas molecules are considered but are not of use

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when bigger molecules are presents in complex mixtures.

Regarding conventional lignocellulosic biomass, it is generally accepted that glucose is a model compound for cellulose (also sometimes for hemicellulose), aromatic compounds a model for lignin and glycine a model for protein [10]. A study of many different biomass of agricultural origin shows the wide range of results and the difficulty of predicting yields regarding the composition of the initial biomass [11]. For residues, SCWG has been tested for industrial effluent: black liquor from paper mills, food industry or municipal in the case of sewage sludge with massic water content higher than 90%. Yakaboylu et al. [12] and Casademont et al. [13] provide a complete overview on biomass gasification, from experimental approaches to modelling.

Contrary to the important quantity of experimental results for SCWG of biomass, less studies have paid attention to the use of experimental results for a global energetic analysis of the systems, from the raw biomass to the final useful product. In literature, first approaches are based on the estimation of overall energy balances. For instance, a classical and complete approach on energetic evaluation has been proposed by Marias et al. [14]. The same kind of work have been conducted by Guan et al. [15] for partial oxidation of biomass. However, these works are dedicated only to the gasification reactor and not to the complete system up to the production of the final useful gas. A more complete study including other unit operations, up to the production of a syngas, is proposed by Wan [16]. Different operating parameters have been tested, the different stages being modelled through thermodynamic equilibrium. However, in all these studies, a systemic study of the operating conditions as well as an optimisation for the gas production are not developed.

This work presents the evaluation of three scenarios (production of hydrogen, production of methane and cogeneration) simulated using a commercial software, based on experimental data for the gasification reactor and mainly thermodynamic equilibrium for other operation units. These simulations give energy and exergy yields, and along with a Pinch analysis, the minimum energy required for each case studied. Eventually, using a numerical design approach, the influence of operating conditions on these three parameters is studied.

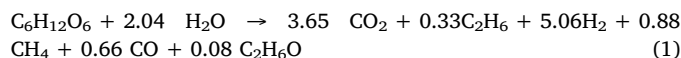
2. Process simulations

Process simulations have been carried out with the numerical simulation software ProSimPlus (version 3.5.18, France). This software allows the calculation of mass and energy balances for each unit operation. It is also a data bank for the physical properties of the different compounds considered. Except for supercritical gasification, process units are mainly calculated with thermodynamic equilibrium using Predictive Soave Redlich Kwong model. For few cases, another thermodynamic model can be used; this model being indicted in the text. Experimental data from the supercritical water gasification of glucose are used at the entering and the exit of the gasifier [17]. For the three scenarios, the different unit operation are calculated either as an isothermal system or an adiabatic system. Detailed conversion systems are given bellow. A brief description of the simulation tool is given in the Supplementary materiel, Appendix A.

2.1. Gasifier

The simulation of the generation of gases from the gasification reactor is common to the three scenarios. The data correspond to the experimental results obtained from the gasification of glucose [17]. The gasifier is fed by a glucose solution with a flowrate of 1 kg h⁻¹ (Flow 1 in Fig. 1). The initial concentration of Total Organic Carbon (TOC) varies from 20 to 80 g L⁻¹. The incoming flow is pressurized by a high pressure pump (HP pump on Fig. 1) having an isentropic efficiency of 90%. The stream (2) is then preheated between 450 and 600 °C. The gasification reactor (GR) is simulated by a subroutine considering experimental conversion rates: a balance equation is written for each

experimental condition tested on glucose. Indeed, as a matter of fact, equilibrium composition cannot be used due to the remaining carbon in the experimental results. All balance equations used in the gasifier module are presented in the supporting Information part (Supplementary material, Appendix B). For example, for the simulation of the production of gas at 600 °C with an initial glucose solution of 20 g L⁻¹, the following reaction is considered (Eq. (1)).



The last term “C₂H₆O” (ethanol) is added to the equation in order to complete the atom balance and to fulfil with experimental results. Indeed, for each conversion, the mass balance and the atom balance on C H and O is calculated. In order to fulfil all atom balance another compounds has to be used. The use of ethanol, in this case, allows to make atom balance correct and are consistent with the remaining TOC of liquids after conversion. Compounds in the outlet are not quantified except by TOC measurement. The heat of reaction accompanying gasification is calculated on the basis of the balance equation using the enthalpy of formation of the various compounds. The stream leaving the reactor (4) is fed to a two-phase separator downstream with an overflow valve and a cold heat exchanger.

2.2. Hydrogen production

For hydrogen production, the different stages of enrichment and purification are based on the data from literature [18–20]. Fig. 1 shows the system for hydrogen production from biomass, including the gasifier reactor.

The gasification reactor outlet stream (6) is cooled down and is separated by a liquid flash stream (LF1) at given temperature and pressure. The temperature specification (noted “SPEC” on Fig. 1) is performed on the ratio of the flow of water necessary for the enrichment step over the flow of gaseous carbon (flux 10). It reduces both the heat needed for flux 11 and the CO₂ content in the gas mixture at the outlet of the separator. Remaining gases consist of H₂, H₂O, CO₂, CO, CH₄ and C₂–C₄ at a pressure of 3 MPa and in varying proportions depending on the temperature of the gasification reactor. At this step, the hydrogen flow ranges from 1.5 to 8 g h⁻¹ and represents from 5 to 30 mol% of the producted gas, for a temperature range between 450 and 600 °C in the gasification reactor.

To increase hydrogen concentration in the mixture an enrichment step consists of two hydrocarbons reforming reactors in series:



- i- A reforming reactor for light hydrocarbons (R1). For instance, the reforming of methane to hydrogen is simulated by steam reforming reactions through Eq. ((2) and (3)).



Under the conditions of temperature and pressure of 3 MPa and 700 °C, the formation of coal that may occur during the process of reforming [18] is avoided.

- ii- A reforming reactor for CO (R2), following Eq. (4) (water gas shift reaction):



For other hydrocarbons, stoichiometric equations are written and the excess of initial water is used for reactions. The gaseous mixture is then cooled to separate residual water from the product gas (LF2) prior to final purification of the hydrogen stream. After this step, the gaseous

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