

Article

Hydrogen-Rich Syngas Production from Gasification of Sewage Sludge: Catalonia Case

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Abstract: The continuous tightening of legislation regulating the agricultural usage of sewage sludge in the province of Catalonia (Spain) leads us to propose its gasification to produce hydrogen-rich syngas. A thermodynamic equilibrium model was developed using Aspen Plus[®] to simulate the air and steam gasification of sewage sludge from a wastewater treatment plant in Catalonia. The syngas generated is analyzed in terms of composition and lower heating value (LHV), as a function of equivalence ratio (ER), gasification temperature (T_{gas}), steam-to-biomass ratio (SBR), and moisture content (MC). Results show that air-blown gasification finds the highest LHV of 7.48 MJ/m³ at 1200 °C, ER of 0.2, and MC of 5%. Using steam as the gasifying agent, an LHV of 10.30 MJ/m³ is obtained at SBR of 0.2, MC of 5%, and 1200 °C. A maximum of 69.7% hydrogen molar fraction is obtained at 600 °C, MC of 25%, and SBR of 1.2. This study suggests using steam as a gasifying agent instead of air since it provides a higher LHV of the syngas as well as a hydrogen-richer syngas for the implementation of gasification as an alternative method to sewage sludge treatment in the region of Catalonia. Since the economic aspect should also be considered, in this regard, our sensitivity analysis provided important data demonstrating that it is possible to reduce the gasification temperature without significantly decreasing the LHV.

Keywords: gasification; syngas; hydrogen; sewage sludge; Aspen Plus



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1. Introduction

The development of today's society is closely linked to two very important aspects, energy, and environmental sustainability. Conventional energy conversion systems use fossil fuels and are associated with greenhouse gas emissions and other atmospheric pollutants. These negative environmental impacts are increasing due to the growing demand for energy. The progressive depletion of fossil fuels and the growing concern about global warming and climate change have driven the search for renewable energy generation techniques. In addition to the growing energy demand, another consequence of the pace of development of today's society is the increase in waste generation, there is a great concern to manage these in a sustainable and economically viable way. For these reasons, it is reasonable to think that one of the ways to make the development of society energetically and environmentally sustainable is the maximum use of waste for energy production, since the benefit would be twofold: waste would be reduced and at the same time the consumption of natural resources would be reduced by producing and using alternative energy.

Sewage sludge (SS) is one of the wastes whose production has significantly grown. Due to urbanization, industrialization, population expansion, an increase in the proportion of people using the sewer system, and better wastewater treatment facilities, the rate of sewage sludge (SS) creation is rising globally. Sewage sludge is hazardous waste, which is

produced by wastewater treatment plants (WWTPs) [1,2]. The management and disposal of the large amount of sewage sludge generated are increasingly complicated due to the strict conditions imposed by current legislation regarding its disposal in landfills and its use as an agricultural fertilizer [3]. Currently, 92.5% of Catalonia's sewage sludge, of the total 120,000 tons of dry matter produced annually, is destined for soil application and 2.5% for landfill [4]. For this reason, the implementation of new ways of valorization of sludge that cannot be destined for agriculture and/or landfills are relevant. One of these ways is gasification in order to produce syngas which could be used to produce chemicals, alternative fuels, hydrogen or combined heat and power (CHP). The sewage sludge gasification process involves multiple reactions and transformations and is therefore considered a complex process. Thus, it is useful to use simulation models that help study the system behavior and allow predicting the process's efficiency under different operating conditions with high reliability and low cost [5]. There are numerous process simulation software tools, among which Aspen Plus stands out for its flexibility and intuitive handling. It has been used by several authors.

Given that supercritical water gasification has emerged as a preferred means of converting wet biomass to hydrogen-rich gases [6], sewage sludge has been studied almost exclusively using supercritical water gasification. For example, Qian et al. [7] experimentally investigated the effects of moisture and pressure on mole fraction, yield, gasification efficiency of gaseous products from supercritical water gasification of sewage sludge. Ruya et al. [8] simulated the supercritical water gasification of various sewage sludge for power generation in Aspen Plus. Chen et al. [9] studied the sewage sludge gasification in supercritical water with a high heating rate batch reactor. Hantoko et al. [10] evaluated through experimental and thermodynamic analysis the sewage sludge supercritical water gasification for hydrogen-rich syngas production. In this work, the sewage sludge will be subjected to autothermal gasification, which, as discussed by Ramos et al. [11] requires the feedstock to be exposed to a drying process.

The thermochemical techniques for hydrogen production from biomass were reviewed by Pandey et al. [12]. They found that hydrogen produced from biomass has satisfactory energy efficiency and the potential to minimize greenhouse gas emissions. They also found that higher temperatures, an appropriate steam-to-biomass ratio, and catalyst type increase hydrogen output. However, production costs remain expensive. The hydrogen production costs issue based on three different gasification processes of high-moisture forest residues was studied by Martins et al. [13]. They found that supercritical water gasification is the most appropriate process for hydrogen production. Conventional gasification is viable for steam-to-biomass ratios below 3. Process intensification procedures used in supercritical water gasification make this process feasible for feed concentrations ranging from 15% and 25%. Alves et al. [14] conduct a techno-economic analysis of a co-gasification plant processing solid recovered fuels and sewage sludge, assuming an 883 kg/h capacity and two alternative scenarios: power production or hydrogen production. The results demonstrated that both possibilities were feasible for implementation. Although the generation of power scenario was more appealing in the short term due to the shorter payback period and higher internal rate of return, the other alternative was more advantageous at the end of the plant's life as the net present value increased. The exploration of conventional gasification of sewage sludge is a recent topic of research, mostly in experimental studies. Kang et al. [15] explored the catalytic gasification of sewage sludge using activated carbon and sawdust biochar catalysts, bed temperatures, and gasifying agents. The porosity of activated carbon was studied for its impact on tar adsorption and cracking, as well as biochar containing alkali and alkaline earth metals species. A temperature of 800 °C and particle sizes of 0.5–1.7 mm exhibited the best conditions for enhancing hydrogen and carbon monoxide contents and decreasing carbon dioxide content. The injection of steam had a positive effect on the H₂ amount, owing to the enhancement of water gas shift and hydrocarbon steam reforming reactions. Tezer et al. [16] experimentally studied sewage sludge gasification

on updraft and downdraft reactors. The effect of temperature and gasifying agents (air and oxygen) on gasification efficiency were investigated. The highest hydrogen molar fraction in syngas from updraft and downdraft gasifiers was 42% and 46%, respectively, when air was used as the gasifying agent. Carotenuto et al. [1] developed an Aspen Plus model of sewage sludge gasification based on a restricted chemical equilibrium. The novelty of this work consists of considering different sludge samples. Through sensitivity analysis, the established Aspen Plus model is utilized to determine the gasification temperature (900 °C) and equivalency ratio (0.2) that results in a higher calorific value for the syngas. The model is then used to evaluate the possibility for sewage sludge to generate both heat and electricity. They found that the proposed technique may provide about 50% of the electricity required to run wastewater treatment plants, as well as 60–75% of the thermal energy required for the drying of sewage sludge for gasification. Andrés et al. [3] performed sewage sludge gasification tests in a fluidized bed gasifier using air and air-steam blends as the gasifying agents. The objective of this work was to determine the influence of dolomite, olivine and alumina catalysts in the syngas distribution and tar production. They discovered that dolomite had the highest activity in tar removal, followed by alumina and olivine. In addition to boosting tar removal, the combination of steam and catalysts boosted the concentration of hydrogen by about 60%.

This brief overview of the current state-of-the-art shows that the sewage sludge conversion to hydrogen-rich gas is mostly approached using supercritical water gasification. The economic studies on high-moisture biomass also show that conventional gasification is more suitable to the economic viability of a gasification plant producing hydrogen. Therefore, in this work, a thermodynamic equilibrium model has been developed using Aspen Plus® V12 to simulate the air and steam gasification process of sewage sludge in a wastewater treatment plant in Catalonia (Spain). The main contribution of this work is to determine the potential of sewage sludge for hydrogen-rich gas production and provide fundamental data for economic studies of the implementation of this waste-to-energy technology.

2. Materials and Methods

2.1. Aspen Plus Model Description

The model developed in this study is based on the thermodynamic equilibrium model. This model is independent of the gasifier design and is based on minimizing the system's Gibbs free energy. The gasifier is represented by multiple blocks, as seen in Figure 1, which correspond to the gasification phases: feed drying, decomposition or pyrolysis and gasification. The configuration starts by defining the components concerned with the process. For the conventional components, the RKS-BM property method, abbreviation of Redlich–Kwong–Soave equation of state with Boston Mathias modifications, is defined [17]. For nonconventional components, biomass and ash, the only physical properties calculated are enthalpy and density as they do not participate in chemical or phase equilibrium. In this simulation, HCOALGEN and DCOALIGT models are used to calculate the enthalpy and density of biomass and ash based on the proximate, ultimate, and sulfur analyses [18].

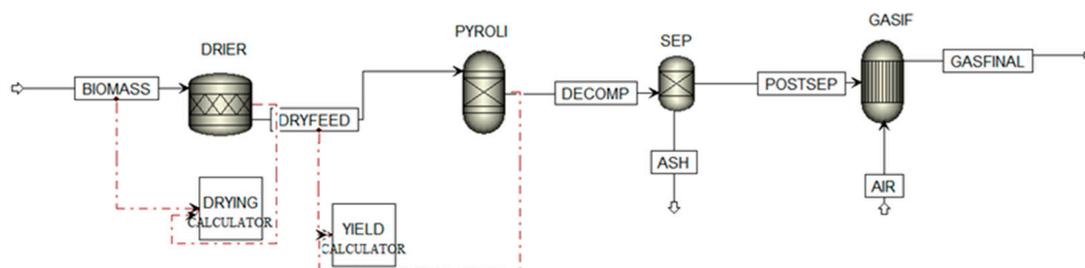


Figure 1. Sewage sludge gasification model using air as the gasifying agent.

The first stage of the process occurs in the DRIER where the evaporation of moisture is simulated. It is necessary to define the water coefficient as 1/18 in the stoichiometric reactor (RStoic) unit, since the molecular weight of biomass and water are 1 g/mol [19] and 18 g/mol, respectively. In addition, a Fortran subroutine calculator is implemented to control this drying process. The second stage occurs in the PYROLI where biomass is converted into its main components C, H, O, N, S, and ash, with the yield distribution determined by the biomass ultimate composition. A Fortran subroutine calculator is implemented to control the decomposition. The resulting stream is directed to SEP where C, H₂, H₂O, O₂ and N₂ are separated from the ash content which is discarded. The mixture is redirected to GASIF, where the reactions take place with the oxidizing agent, giving rise to the synthesis gas (GASFINAL).

2.2. Aspen Plus Model Validation

To validate the results of the developed Aspen Plus model, the composition of the syngas is compared with the experimental results of Ong et al. [20] and with a kinetic model developed by Rabea et al. [21]. Both studies are based on the same biomass and conditions as the present study. The biomass used is wood chips, and the gasifying agent for the analysis is air. The ultimate and proximate compositions are given in Table 1, where MC, VM, and FC stand for moisture content, volatile matter, and fixed carbon, respectively.

Table 1. Proximate and ultimate analysis of wood chips [21].

Proximate Analysis (wt.%)				Ultimate Analysis (wt.%, db.)			
MC	VM	FC	Ash	C	H	N	O
8.35	74.8	18.4	6.8	43.75	5.75	1.65	42.05

Figure 2 compares the molar fractions of the syngas produced by the proposed model and the kinetic model of Rabea et al. [21] to the experimental data of Ong et al. [20]. The findings were achieved using air flow rate of 7 dm³/s, a biomass flow rate of 16.2 kg/h, and a gasifier temperature of 995 °C.

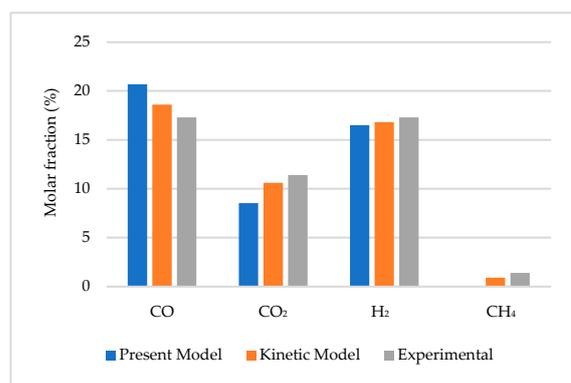


Figure 2. Comparison of produced syngas composition.

Relative error is used to determine the deviations between the results and verify if the model is overestimating (positive relative errors) or underestimating (negative relative errors) the results. The relative error is calculated as expressed in Equation (1) and is present in Table 2.

$$\text{Relative error (\%)} = \frac{\text{Aspen Plus value} - \text{Literature value}}{\text{Literature value}} \times 100 (\%) \quad (1)$$

Table 2. Syngas composition and relative error between the present model, Rabea et al. [21] kinetic model and the experimental data of Ong et al. [20].

Species	Gas Composition (Vol.%)		Relative Error (%)
	Present Model	Experimental [20]	
CO	20.67	17.30	19.50
CO ₂	8.53	11.40	−25.18
H ₂	16.50	17.30	−4.62
	Present Model	Kinetic Model [21]	
CO	20.67	18.60	11.13
CO ₂	8.53	10.60	−19.53
H ₂	16.50	16.80	−1.79
	Experimental [20]	Kinetic Model [21]	
CO	17.30	18.60	−7.51
CO ₂	11.40	10.60	7.02
H ₂	17.30	16.80	2.89

The main syngas species have relative errors of less than 25%. Methane (CH₄) has been excluded in the assessment due to its near-zero molar fractions. The greatest relative errors are obtained for CO₂, which occurs frequently in thermodynamic equilibrium models [22].

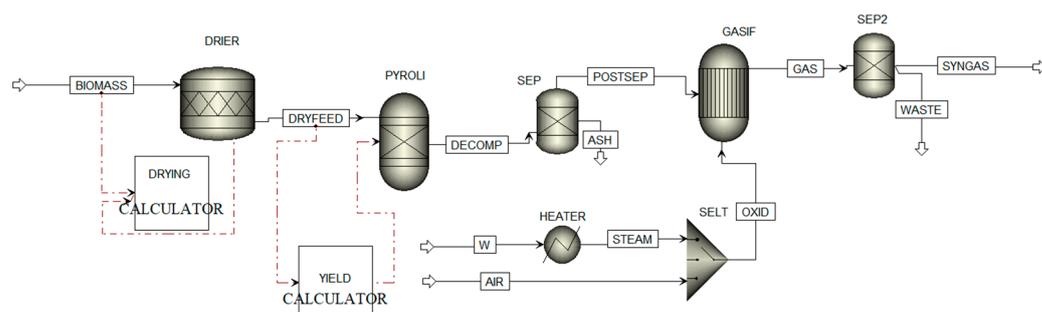
Furthermore, Ong et al. [20]’s experimental data lack an error analysis that might lower the relative errors in the comparison. Table 2 also includes the relative error between literature data (experimental versus kinetic model). The agreement of the kinetic model with experimental data is better than the Aspen Plus model developed in this study. This is not surprising since kinetic models are recognized to be more accurate than thermodynamic equilibrium models for describing the behavior of a gasifier [23].

The rationale for this behavior is that kinetic models are based on kinetic mechanisms, which explain the chemical processes that occur throughout the biomass gasification process. Residence time, gasifier design, biomass feeding rate and reactor hydrodynamic, for example, may all be predicted using kinetic models [24], but not with thermodynamic equilibrium models.

Nevertheless, the results obtained with the Aspen Plus-developed model accurately reproduce the thermodynamic behavior of the biomass gasification conversion process, especially for the hydrogen molar fraction (<5%), making it an effective tool for estimating syngas composition and process design prior to experimental investigations.

2.3. Aspen Plus Model Improvement

Since it is intended to perform the sensitivity analysis for both air and steam, a selector (SELT) is added to the model to easily switch from one gasifying agent to another. In addition, a separator (SEP1) is added at the gasifier outlet, which basically separates the components of interest from those that are not of interest. The final model used to perform both sensitivity analyses is shown in Figure 3.

**Figure 3.** Sewage sludge gasification model improvement to use air or steam as the gasifying agent.

2.4. Sewage Sludge Characterization

Dried sewage sludge was selected as waste biomass material for this study. Their origin was an urban wastewater treatment plant in Catalonia (Spain), whose proximate and ultimate analyses are presented in Table 3.

Table 3. Proximate and ultimate analysis of sewage sludge from a wastewater treatment plant in Catalonia (Spain) [3].

Proximate Analysis (wt.%)				Ultimate Analysis (wt.%, Dry Basis)				
MC	VM	FC	Ash	C	H	N	O	S
7.0	46.0	10	44	27.3	4.8	4.1	18.9	0.9

The main chemical reactions that are performed in the gasification process and enthalpy of formation are depicted in Table 4.

Table 4. Main biomass gasification reactions [25,26].

Reaction	Chemical Reaction	Enthalpy of Formation
R1—Carbon combustion	$C + O_2 \rightarrow CO_2$	$\Delta H = -394 \text{ J/mol}$
R2—Partial oxidation	$C + \frac{1}{2} O_2 \rightarrow CO$	$\Delta H = -111 \text{ J/mol}$
R3—Hydrogen combustion	$H_2 + \frac{1}{2} O_2 \rightarrow H_2O$	$\Delta H = -242 \text{ J/mol}$
R4—Boudouard reaction	$C + CO_2 \leftrightarrow 2CO$	$\Delta H = 172 \text{ J/mol}$
R5—Reforming of the char	$C + H_2O \leftrightarrow CO + H_2$	$\Delta H = 131 \text{ J/mol}$
R6—Water-gas shift	$CO + H_2O \leftrightarrow CO_2 + H_2$	$\Delta H = -41 \text{ J/mol}$
R7—Methanation	$C + 2H_2 \leftrightarrow CH_4$	$\Delta H = -75 \text{ J/mol}$
R8—Steam methane reforming	$CH_4 + H_2O \rightarrow CO + 3H_2$	$\Delta H = 206 \text{ J/mol}$

3. Results and Discussion

3.1. Evaluation Using Air as the Gasifying Agent

The composition and LHV of the syngas will be analyzed as a function of different process parameters: equivalence ratio, gasification temperature, and moisture content. The same conditions used for model validation (6.2 kg/h of sewage sludge and a gasifier temperature of 995 °C) were used in this assessment, except for the gasification temperature.

3.1.1. Effect of Equivalence Ratio

The ER is defined as “the ratio of the air-to-biomass ratio to the stoichiometric air-to-biomass ratio” [26]. The ER was adjusted by varying the air flow rate into the Gibbs reactor. Figure 4 shows the effect of the ER on the syngas composition and LHV.

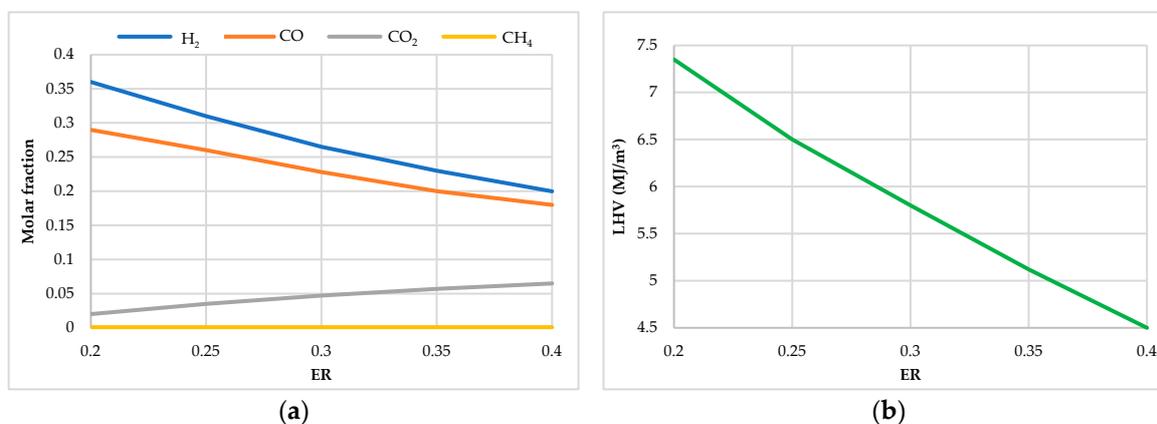


Figure 4. (a) Effect of ER on syngas composition when using air as the gasifying agent; (b) Effect of ER on the LHV of syngas when using air as the gasifying agent.

Figure 4a shows that an increase in the ER leads to a decrease in H₂ production due to a lower magnitude of the reforming reaction (R5) and the water-gas shift reaction (R6) versus the combustion reactions (R1, R2, and R3). The molar fraction of CO decreases because of the higher presence of oxygen, which promotes the oxidation of the carbon in the sewage sludge and part of the combustible gases to produce CO₂, which explains the increase of this compound in the syngas [3,27].

Figure 4b shows the LHV of the syngas obtained according to Equation (2) [28]:

$$\text{LHV}_{\text{syngas}} = 35.84 Y_{\text{CH}_4} + 12.62 Y_{\text{CO}} + 10.79 Y_{\text{H}_2} \quad (2)$$

Therefore, the reduction of H₂ and CO content leads to lower heating values in the produced syngas. Although CH₄ has a high calorific value (Equation (2)), its molar fractions are near-zero, as seen in Figure 4a. The rationale for such a low molar fraction of CH₄ in our model predictions is the assumption that thermodynamic equilibrium is attained, which deviates from a real gasification scenario [29]. This behavior is consistent with thermodynamic equilibrium models, which are supported by the fact that methane produced in the pyrolysis phase cannot avoid the reduction phase [30]. In a real gasification scenario, CH₄ is able to avoid reduction, which is considered a non-equilibrium factor characteristic of the gasifier [30]. Similar trends for the different syngas species with ER can be found in the stated literature [31–33].

3.1.2. Effect of Gasification Temperature

According to Le Chatelier's principle, high temperatures favor the products of endothermic reactions and reactants in exothermic reactions [34]. Figure 5 shows the effect of the gasification temperature on the syngas composition and LHV.

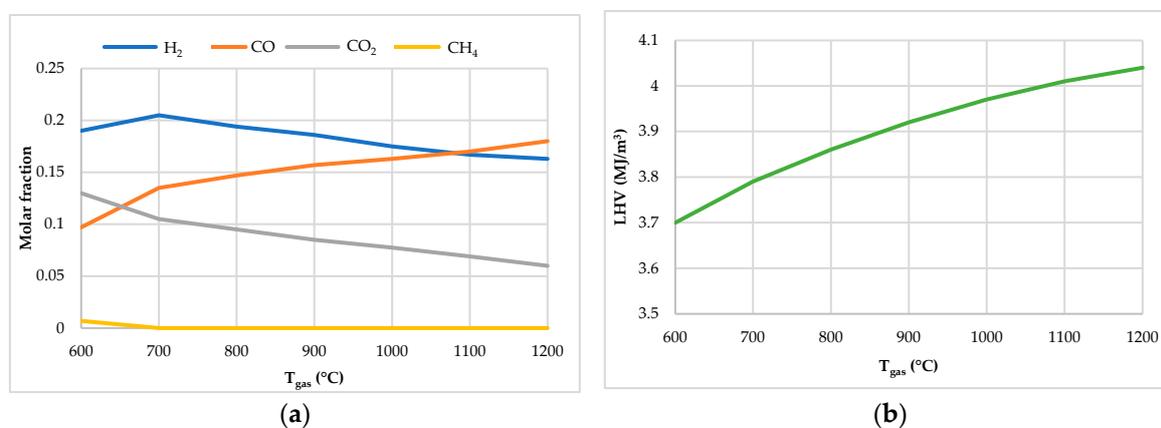


Figure 5. (a) Effect of the gasification temperature on syngas composition when using air as the gasifying agent; (b) Effect of the gasification temperature on syngas LHV when using air as the gasifying agent.

Figure 5a shows that the molar fractions of CO increase while CO₂ decreases, mainly due to the Boudouard reaction (R4) and the reforming of the char reaction (R5), which are enhanced with increasing temperature. Figure 5a also shows that the H₂ content increases between 600–700 °C decreasing its molar fractions at higher temperatures. This change in H₂ trend may be due to the mutual effect of the reactions taking place in the reactor. The R6 (water-gas shift) reaction is successful at lower temperatures, creating syngas that is rich in H₂, but it is hampered at temperatures above 720 °C because the reverse reaction is encouraged, which reduces the amount of H₂ that is produced. CH₄ has near-zero molar fractions due to the reasons listed in Section 3.1.1.

Regarding Figure 5b, the LHV of the syngas shows a continuous increase with temperature. Between 600–700 °C the molar fractions of H₂ and CO increase and the CH₄ molar fraction decreases. Since the CH₄ molar fractions are very low, this results in an increase in

the LHV of the syngas, according to Equation (2). Above 700 °C, the molar fraction of H₂ decreases, the CO molar fraction increases, and the CH₄ molar fraction is negligible. Since the CO molar fraction increases more than that of H₂, the LHV continues to increase with temperature increases. Similar trends for the different syngas species with temperature can be found in [35,36].

3.1.3. Correlation between Variables

The following assessment examines the correlation between the process parameters temperature, ER, and moisture content (Figure 6) to determine the optimal conditions that maximize the LHV of the produced syngas.

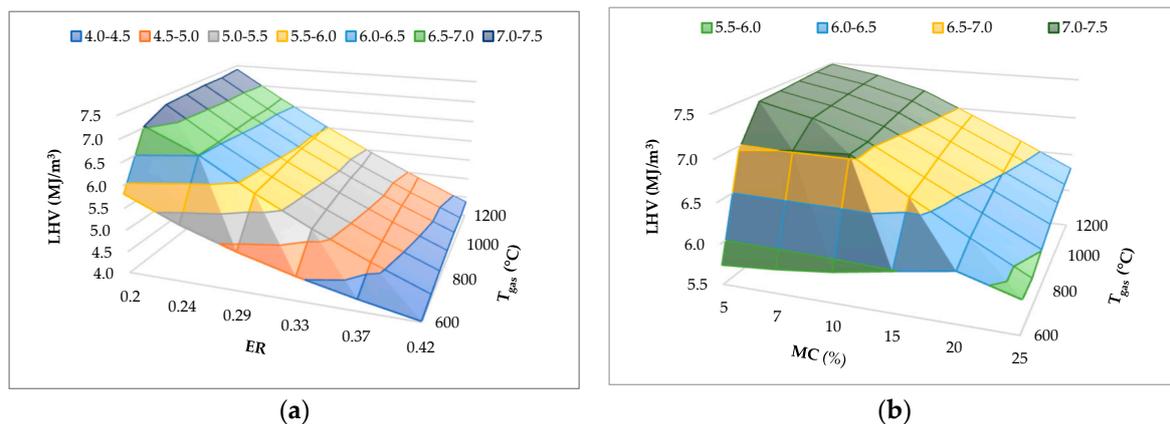


Figure 6. (a) Combined effect of temperature and ER on the LHV of syngas; (b) Combined effect of moisture content (MC) and temperature on the LHV of syngas.

Figure 6a shows that the lower the ER, the lower the temperature required by the gasifier to produce syngas with nearly the same LHV. This result is of great interest since the lower the temperature, the lower the energy, and the lower the operational expenditures. For example, an ER of 0.29 at $T_{\text{gas}} = 1000$ °C produces syngas with the same LHV as an ER of 0.24 at $T_{\text{gas}} = 600\text{--}700$ °C. Figure 6b shows that the lower the MC, the lower the temperature required by the gasifier to produce syngas with virtually the same LHV. For example, sewage sludge gasification with an MC of 15% at $T_{\text{gas}} = 1200$ °C produces syngas with almost the same LHV as sewage sludge with an MC of 10% at $T_{\text{gas}} = 600\text{--}700$ °C. It is known that moisture content considerably affects the gasification process because the water is vaporized in the reactor, absorbing heat, and reducing the temperature, while the generated steam can react with other compounds [29]. The obtained trends for the effect of moisture content on the LHV of the syngas are confirmed by the literature [29,37].

3.2. Evaluation Using Steam as the Gasifying Agent

The composition and LHV of the syngas will be analyzed as a function of SBR, gasification temperature, and moisture content. The same conditions used for model validation (16.2 kg/h of sewage sludge and a gasifier temperature of 995 °C) were used in this assessment, except for the gasification temperature.

3.2.1. Effect of Steam-to-Biomass Ratio

Figure 7 shows the effect of SBR on the molar fraction of the syngas main species and LHV. Since more steam is added with increasing SBR, it enhances the water-gas shift reaction (R6) and steam reforming reaction (R5) which result in an increase in H₂ and CO molar fractions [32]. However, the CO concentration will decrease due to the water-gas shift reaction (R6), which reduces the CO molar fraction concentration by reacting with steam and increasing H₂ and CO₂ concentrations as seen in Figure 7a.

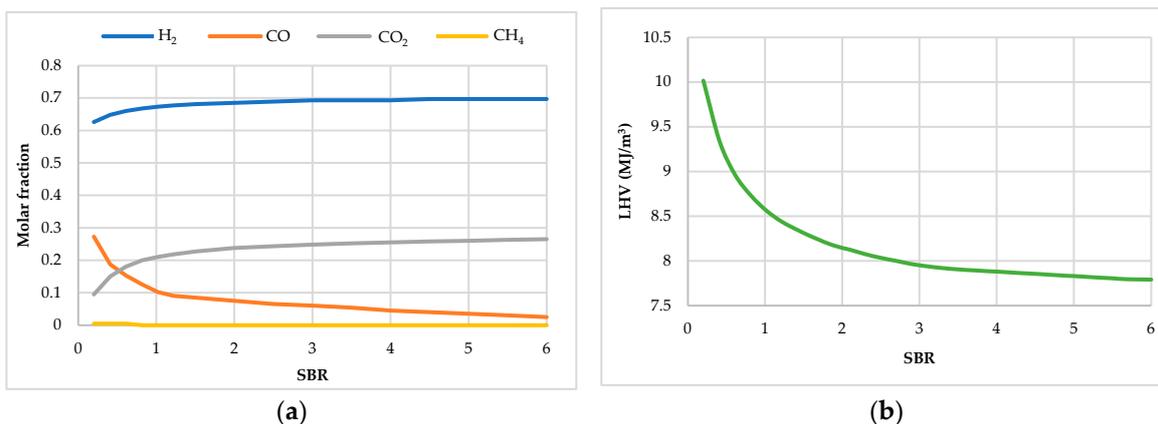


Figure 7. (a) Effect of SBR on syngas composition when using steam as the gasifying agent; (b) Effect of SBR on syngas LHV when using steam as the gasifying agent.

Regarding the CH₄ molar fraction, it presents a near-zero value for the entire SBR range. The reasons for this behavior are pointed out in Section 3.1.1, which are also valid for steam as a gasifying agent. Figure 7b shows that the syngas LHV decreases with SBR. According to Equation (2), the rationale for this behavior is that the CO molar fraction decreases more prominently than that of H₂. Similar results for the different syngas species and LHV as a function of SBR can be found in [38].

3.2.2. Effect of Gasification Temperature

One of the most important parameters influencing syngas composition is gasification temperature. Because the main reactions of gasification are endothermic, raising temperature strengthens them [32]. Figure 8 shows the effect of temperature on the molar fraction of the main syngas species and LHV. Figure 8 shows that the mole fractions of CO and H₂ increase up to 600 °C because the reforming of the char reaction (R5) and the Boudouard reaction (R4) are endothermic and, therefore, the temperature increase causes an increase in CO and H₂. The water-gas shift reaction (R6) is the dominant reaction above 750 °C, so CO increases while CO₂ and H₂ decrease. Reactions (R5) and (R4) contribute to the increase in CO above 800 °C. CH₄ reduction is dictated by the methanation reaction (R7) and the steam methane reforming reaction (R8).

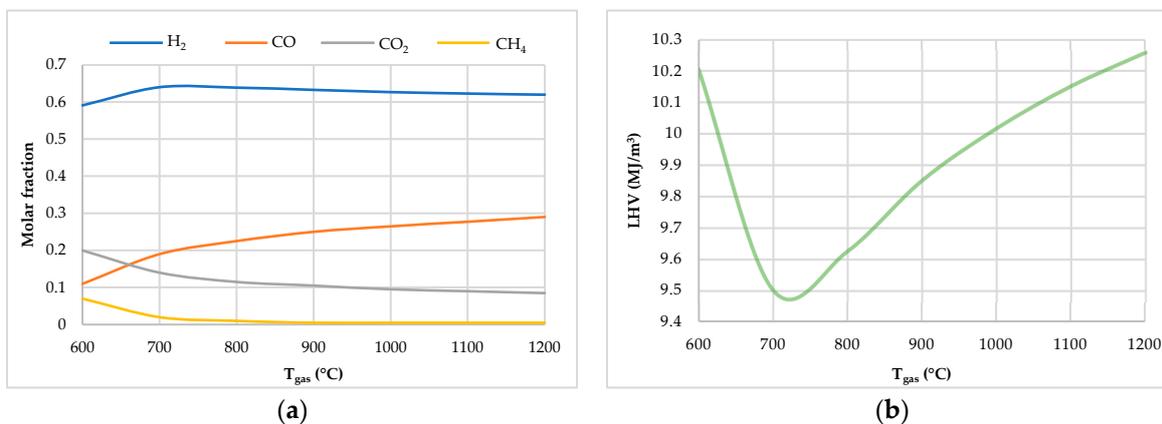


Figure 8. (a) Effect of temperature on syngas composition when using steam as the gasifying agent; (b) Effect of temperature on syngas composition and LHV when using steam as the gasifying agent.

The LHV decreases until reaching the temperature of 700 °C because CH₄ decreases more than H₂ increases, but from then on, the LHV follows a clear increasing trend as H₂, and CO continue to increase while CH₄ values remain low. It can be said that the higher

the temperature, the higher the LHV, but this does not imply higher H_2 . Similar trends for the different syngas species and LHV as a function of temperature can be found in [39,40].

3.2.3. Correlation between Variables

The following assessment examines the correlation between the process parameters SBR, gasification temperature, and moisture content. The objective is to find the optimal conditions that maximize the syngas quality in terms of heating value and H_2 content. Figure 9 shows the combined effect of SBR and gasification temperature on the LHV and H_2 content of the syngas.

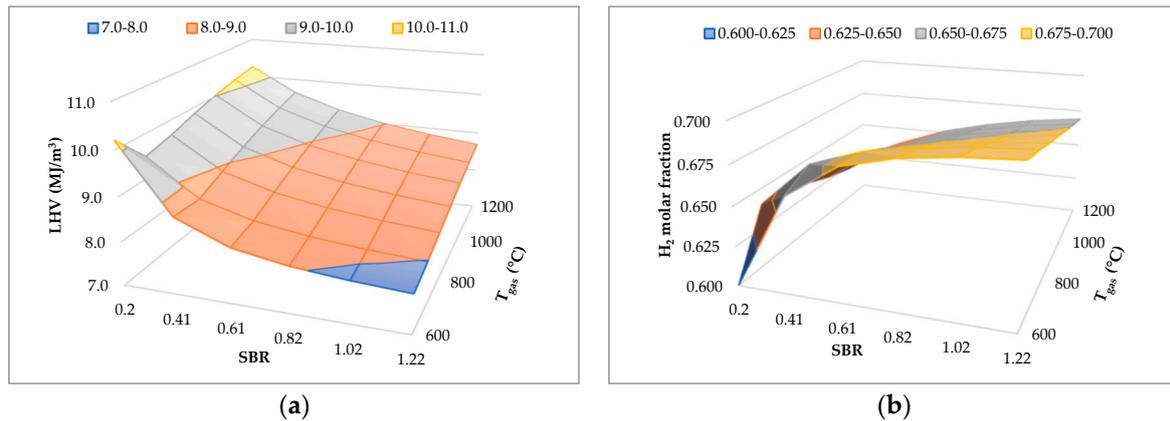


Figure 9. (a) Combined effect of SBR and temperature on the LHV of syngas; (b) Combined effect of SBR and temperature on the H_2 molar fraction of syngas.

The results of Figure 9a show that the lower the SBR, the lower the temperature required by the gasifier to produce the same LHV. As mentioned before, lower temperatures are beneficial for the reduction of operational expenditures. On the other hand, in Figure 9b, it can be seen that the higher the SBR, the lower the temperature required by the gasifier to produce similar H_2 molar fractions. Figure 10 shows the combined effect of the sewage sludge moisture and temperature on the LHV and H_2 content of the syngas.

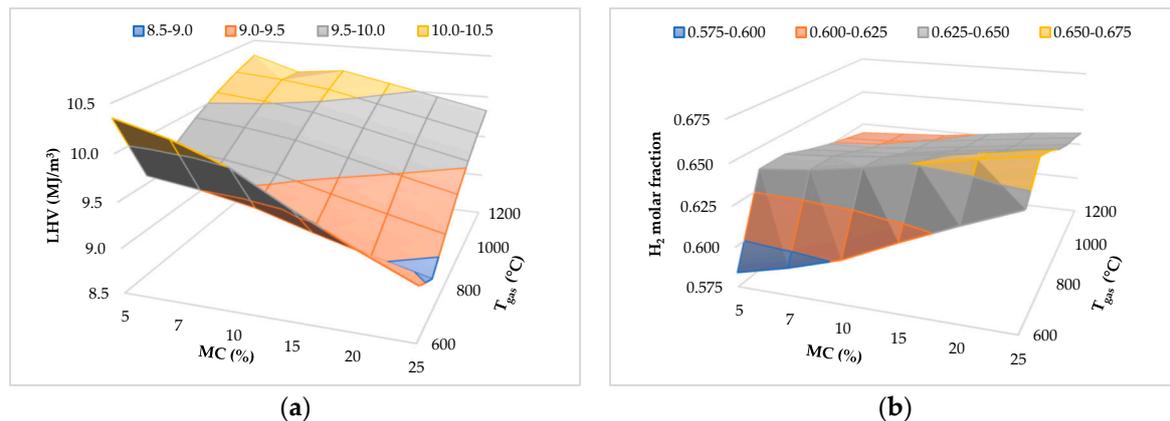


Figure 10. (a) Combined effect of moisture content and gasification temperature on the LHV of syngas; (b) Combined effect of moisture content and temperature on the H_2 molar fraction of syngas.

It can be observed that the lower the moisture content, the lower the temperature required to achieve similar LHV values. On the other hand, to obtain higher H_2 molar fractions, the higher the MC, the higher the H_2 molar fraction. The condition in which maximum H_2 production is reached is the one with high moisture content (20%) at a temperature of 700 °C. As has been reported elsewhere [29], higher moisture contents promote a slight increase in the H_2 molar fraction. This behavior can be explained based on

the water-gas shift reaction (R6). When there is greater moisture, CO levels decrease due to consumption, subsequently raising the H₂ content [41]. Regarding the effect of moisture content in the LHV, it can be seen that higher moisture content leads to the decrease of LHV. Similar trends for LHV as a function of moisture can be found in the literature [29,37]. Figure 11 shows the combined effect of SBR and sewage sludge moisture content on the LHV and H₂ of the syngas.

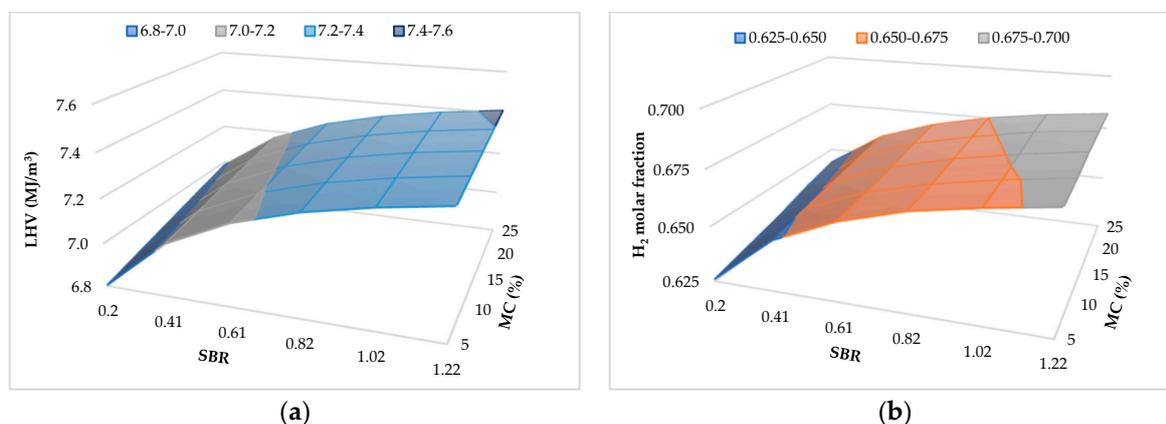


Figure 11. (a) Combined effect of SBR and moisture content on the LHV of syngas; (b) Combined effect of SBR and moisture content on the H₂ of syngas.

From Figure 11, it can be deduced that the higher the moisture content and the higher the SBR, the higher the LHV and H₂ values. However, the SBR has a more pronounced effect on the LHV and H₂ content of the syngas than that of the moisture content. This behavior is explained by the fact that the moisture content of sewage sludge evaporates in the reactor lowering the gasification temperature, which does not occur when steam is delivered to the reactor as a gasifying agent. In both cases, the steam generated or provided will be used in steam reforming reactions during the gasification process. Lower temperatures will favor exothermic reforming reactions like the water-gas shift (R6), leading to the consumption of CO and the production of H₂ and CO₂. The increase in H₂ molar fraction with moisture content can be seen in Figure 11b.

4. Conclusions

In this work, a thermodynamic equilibrium model was developed using Aspen Plus[®] to simulate the air and steam gasification processes of sewage sludge from a wastewater treatment plant in Catalonia (Spain). Sensitivity analyses to various process parameters were performed to determine the optimal conditions for higher calorific value of the syngas and higher hydrogen molar fractions.

Air-blown sewage sludge gasification finds the highest LHV conditions under the minimum sewage sludge moisture content and equivalence ratio (and high temperature). Specifically, under conditions of ER = 0.2, MC = 5%, and T_{gas} = 1200 °C, it generates a syngas with a calorific value of 7.48 MJ/m³. On the other hand, using steam as the gasifying agent, the lower the SBR and the lower the MC, the higher the LHV (at high temperature). Working at SBR = 0.2, MC = 5%, and T_{gas} = 1200 °C generates syngas with a calorific value of 10.30 MJ/m³. Under optimal operating conditions that maximize the LHV of the syngas and considering a moisture content of 7% in the dried sewage sludge, the LHV of the syngas is 7.37 MJ/m³ and 10.26 MJ/m³ when using air and steam as the gasifying agents, respectively. The hydrogen molar fraction is maximized when steam is used as the gasifying agent in combination with high moisture content sewage sludge, high steam-to-biomass ratios, and low gasification temperatures. Particularly, a moisture content of 25%, an SBR of 1.2, and a temperature of 600 °C allow for a maximum H₂ molar fraction of 69.7%.

This study suggests the use of steam as a gasifying agent instead of air for the implementation of gasification as an alternative method to sewage sludge treatment in the region of Catalonia (Spain). When suggesting such a paradigm shift in sewage sludge treatment, the economic component must be considered. In this regard, our sensitivity analysis provides valuable information on several gasification parameters that can be lowered without compromising syngas quality while lowering operational expenditures. Cases involving energy-intensive parameters such as moisture content and SBR are highly relevant from an economic point of view. Therefore, it is imperative to reduce the sewage sludge pre-drying needs or inject less steam into the gasifier. Further studies should be carried out, such as cost-benefit studies, which can be performed using the tool developed. The main contribution of this work is to determine the potential of sewage sludge for hydrogen-rich gas production and provide fundamental data for economic studies of the implementation of this waste-to-energy technology.

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References

1. Carotenuto, A.; Fraia, S.; Massarotti, N.; Sobek, S.; Uddin, M.R.; Vanoli, L.; Werle, S. Predictive modeling for energy recovery from sewage sludge gasification. *Energy* **2023**, *263*, 1258. [CrossRef]
2. Quan, L.M.; Yabar, H.; Idaa, W.; Yuzir, A. Simulation of sewage sludge air gasification and application of electricity and steam generation: Case study of Pantai 1 sewage treatment plant. *Energy Convers. Manag.* **2022**, *270*, 116196. [CrossRef]
3. Andrés, J.M.; Narros, A.; Rodríguez, M.E. Behaviour of dolomite, olivine and alumina as primary catalysts in air–steam gasification of sewage sludge. *Fuel* **2011**, *90*, 521–527. [CrossRef]
4. Agència Catalana de l'Aigua. La Gestió dels Biosòlids a Catalunya. Generalitat de Catalunya, 2019. Available online: https://aca.gencat.cat/web/.content/10_ACA/J_Publicacions/07-estudis-informes/10_gestio_biosolids_a_catalunya.pdf (accessed on 9 January 2023).
5. Ahmed, T.Y.; Ahmad, M.M.; Yusup, S.; Inayat, A.; Khan, Z. Mathematical and computational approaches for design of biomass gasification for hydrogen production: A review. *Renew. Sustain. Energy Rev.* **2012**, *16*, 2304–2315. [CrossRef]
6. Hu, Y.; Gong, M.; Xing, X.; Wang, H.; Zeng, Y.; Xu, C.C. Supercritical water gasification of biomass model compounds: A review. *Renew. Sustain. Energy Rev.* **2020**, *118*, 09529. [CrossRef]
7. Qian, L.; Wang, S.; Wang, S.; Zhao, S.; Zhang, B. Supercritical water gasification and partial oxidation of municipal sewage sludge: An experimental and thermodynamic study. *Int. J. Hydrogen Energy* **2021**, *46*, 89–99. [CrossRef]
8. Ruya, P.M.; Purwadi, R.; Lim, S.S. Supercritical water gasification of sewage sludge for power generation—thermodynamic study on auto-thermal operation using Aspen Plus. *Energy Convers. Manag.* **2020**, *206*, 112458. [CrossRef]
9. Chen, Y.; Yi, L.; Wei, W.; Jin, H.; Guo, L. Hydrogen production by sewage sludge gasification in supercritical water with high heating rate batch reactor. *Energy* **2022**, *238*, 121740. [CrossRef]
10. Hantoko, D.; Antoni; Kanchanatip, E.; Yan, M.; Weng, Z.; Gao, Z.; Zhong, Y. Assessment of sewage sludge gasification in supercritical water for H₂-rich syngas production. *Process Saf. Environ. Prot.* **2019**, *131*, 63–72. [CrossRef]
11. Kumar, J.A.; Sathish, S.; Prabu, D.; Annam Renita, A.; Saravanan, A.; Deivayanai, V.C.; Anish, M.; Jayaprabakar, J.; Baigenzhenov, O.; Hosseini-Bandegharai, A. Agricultural waste biomass for sustainable bioenergy production: Feedstock, characterization and pre-treatment methodologies. *Chemosphere* **2023**, *331*, 138680. [CrossRef]
12. Pandey, B.; Prajapati, Y.K.; Sheth, P.N. Recent progress in thermochemical techniques to produce hydrogen gas from biomass: A state of the art review. *Int. J. Hydrogen Energy* **2019**, *44*, 25384–25415. [CrossRef]
13. Martins, A.H.; Rouboa, A.; Monteiro, E. On the green hydrogen production through gasification processes: A techno-economic approach. *J. Clean. Prod.* **2023**, *383*, 135476. [CrossRef]
14. Alves, O.; Calado, L.; Panizio, R.M.; Gonçalves, M.; Monteiro, E.; Brito, P. Techno-economic study for a gasification plant processing residues of sewage sludge and solid recovered fuels. *Waste Manag.* **2021**, *131*, 148–162. [CrossRef]

15. Kang, B.S.; Farooq, A.; Valizadeh, B.; Lee, D.; Seo, M.W.; Jung, S.-C.; Hussain, M.; Kim, Y.M.; Khan, M.A.; Jeon, B.-H.; et al. Valorization of sewage sludge via air/steam gasification using activated carbon and biochar as catalysts. *Int. J. Hydrogen Energy* **2024**, *54*, 284–293. [[CrossRef](#)]
16. Tezer, Ö.; Karabağ, N.; Öngen, A.; Ayol, A. Syngas production from municipal sewage sludge by gasification Process: Effects of fixed bed reactor types and gasification agents on syngas quality. *Sustain. Energy Technol. Assess.* **2023**, *56*, 103042. [[CrossRef](#)]
17. Lan, W.; Chen, G.; Zhu, X.; Wang, X.; Liu, C.; Xu, B. Biomass gasification-gas turbine combustion for power generation system model based on ASPEN PLUS. *Sci. Total Environ.* **2018**, *628–629*, 1278–1286. [[CrossRef](#)] [[PubMed](#)]
18. Aspen Technology Inc. *Aspen Plus User Guide, Version 10.2*; Aspen Technology Inc.: Cambridge, MA, USA, 2000.
19. Aspen Technology Inc. *Aspen Plus 12.1 Getting Started Modeling Processes with Solids*; Aspen Technology Inc.: Cambridge, MA, USA, 2003.
20. Ong, Z.; Cheng, Y.; Maneerung, T.; Yao, Z.; Tong, Y.W.; Wang, C.-H.; Dai, Y. Co-gasification of woody biomass and sewage sludge in a fixed-bed downdraft gasifier. *AIChE J.* **2015**, *61*, 2508–2521. [[CrossRef](#)]
21. Rabea, K.; Michailos, S.; Akram, M.; Hughes, K.J.; Ingham, D.; Pourkashanian, M. An improved kinetic modelling of woody biomass gasification in a downdraft reactor based on the pyrolysis gas evolution. *Energy Convers. Manag.* **2022**, *258*, 115495. [[CrossRef](#)]
22. Puig-Arnavat, M.; Bruno, J.C.; Coronas, A. Review and analysis of biomass gasification models. *Renew. Sustain. Energy Rev.* **2010**, *14*, 2841–2851. [[CrossRef](#)]
23. Sharma, A.K. Equilibrium and kinetic modelling of char reduction reactions in a downdraft biomass gasifier: A comparison. *Sol. Energy* **2008**, *52*, 918–928. [[CrossRef](#)]
24. Hajinajaf, N.; Mehrabadi, A.; Tavakoli, O. Practical strategies to improve harvestable biomass energy yield in microalgal culture: A review. *Biomass Bioenergy* **2021**, *145*, 105941. [[CrossRef](#)]
25. Molino, A.; Chianese, S.; Musmarra, D. Biomass gasification technology: The state of the art overview. *J. Energy Chem.* **2016**, *25*, 10–25. [[CrossRef](#)]
26. Basu, P. *Biomass Gasification, Pyrolysis and Torrefaction*, 2nd ed.; Elsevier Inc.: London, UK, 2013.
27. Ismail, T.; Ramos, A.; Monteiro, E.; El-Salam, M.; Rouboa, A. Parametric studies in the gasification agent and fluidization velocity during oxygen-enriched gasification of biomass in a pilot-scale fluidized bed: Experimental and numerical assessment. *Renew. Energy* **2020**, *147*, 2429–2439. [[CrossRef](#)]
28. Kaewluan, S.; Pipatmanomai, S. Potential of synthesis gas production from rubber wood chip gasification in a bubbling fluidised bed gasifier. *Energy Convers. Manag.* **2011**, *52*, 75–84. [[CrossRef](#)]
29. Altafini, C.R.; Wander, P.R.; Barreto, R.M. Prediction of the working parameters of a wood waste gasifier through an equilibrium model. *Energy Convers. Manag.* **2003**, *44*, 2763–2777. [[CrossRef](#)]
30. Aydin, E.S.; Yucel, O.; Sadikoglu, H. Development of a semi-empirical equilibrium model for downdraft gasification systems. *Energy* **2017**, *130*, 86–98. [[CrossRef](#)]
31. Couto, N.; Silva, V.; Monteiro, E.; Rouboa, A.; Brito, P. An experimental and numerical study on the Miscanthus gasification by using a pilot scale gasifier. *Renew. Energy* **2017**, *109*, 248–261. [[CrossRef](#)]
32. Couto, N.; Monteiro, E.; Silva, V.; Rouboa, A. Hydrogen-rich gas from gasification of Portuguese municipal solid wastes. *Int. J. Hydrogen Energy* **2016**, *41*, 10619–10630. [[CrossRef](#)]
33. Kombe, E.Y.; Lang'at, N.; Njogu, P.; Malessa, R.; Weber, C.-T.; Njoka, F.; Krause, U. Process modeling and evaluation of optimal operating conditions for production of hydrogen-rich syngas from air gasification of rice husks using aspen plus and response surface methodology. *Bioresour. Technol.* **2022**, *361*, 127734. [[CrossRef](#)]
34. Heer, J. The principle of Le Châtelier and Braun. *J. Chem. Educ.* **1957**, *34*, 375–380. [[CrossRef](#)]
35. Ferreira, S.; Monteiro, E.; Calado, L.; Silva, V.; Brito, P.; Vilarinho, C. Experimental and Modeling Analysis of Brewers' Spent Grains Gasification in a Downdraft Reactor. *Energies* **2019**, *12*, 4413. [[CrossRef](#)]
36. Zhang, W.; Gu, Y.; Fang, H.; Chen, J.; Chen, H.; Zhu, Y.; Mu, L. Thermodynamic modeling and performance analysis on co-gasification of *Chlorella vulgaris* and petrochemical industrial sludge via Aspen plus combining with response surface methodology. *Int. J. Hydrogen Energy* **2024**, *55*, 1037–1049. [[CrossRef](#)]
37. Jarungthammachote, S.; Dutta, A. Thermodynamic equilibrium model and second law analysis of a downdraft waste gasifier. *Energy* **2007**, *32*, 1660–1669. [[CrossRef](#)]
38. Ferreira, S.; Monteiro, E.; Brito, P.; Vilarinho, C. Experimental Analysis of Brewers' Spent Grains Steam Gasification in an Allothermal Batch Reactor. *Energies* **2019**, *12*, 912. [[CrossRef](#)]
39. Ramzan, N.; Ashraf, A.; Naveed, S.; Malik, A. Simulation of hybrid biomass gasification using Aspen plus: A comparative performance analysis for food, municipal solid and poultry waste. *Biomass Bioenergy* **2011**, *35*, 3962–3969. [[CrossRef](#)]
40. Baratieri, M.; Baggio, P.; Fiori, L.; Grigiante, M. Biomass as an energy source: Thermodynamic constraints on the performance of the conversion process. *Bioresour. Technol.* **2008**, *99*, 7063–7073. [[CrossRef](#)] [[PubMed](#)]
41. McKendry, P. Energy production from biomass (part 3): Gasification technologies. *Bioresour. Technol.* **2002**, *83*, 55–63. [[CrossRef](#)] [[PubMed](#)]

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