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Parametric Modelling Study to Determine the Feasibility of the Co-Gasification of Macroalgae and Plastics for the Production of Hydrogen-Rich Syngas

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Abstract: Seaweed is of importance to the environment due to properties such as its CO_2 sequestration capability. On the other hand, plastic is a versatile material important to society. Unfortunately, the abundance of both plastics and seaweed represents a challenge due to the need of adequate waste management. At first, algae and plastics might not appear suitable for energy recovery; however, with co-gasification, the results are promising. This work presents a model to simulate the co-gasification of *Chlorella vulgaris*, *Sargassum fluitans*, and *Sargassum natans* with plastics. The effect of the gasification temperature (650–850 °C), equivalence ratio (0.25–0.45), and plastics/biomass ratio (0.0–1.0) on the produced gas's lower heating value, tar concentration, and composition is assessed. Moreover, the environmental performance of using plastic to enhance the syngas produced from the co-gasification with sargassum is assessed using the life cycle assessment methodology. The results indicate that the lower heating value increases with the temperature and plastics/biomass ratio. Moreover, tar increases with the quantity of plastics, varying between 20 and 50 g/Nm^3. Finally, adding plastics to algae during gasification reduces the variability in the syngas composition, simplifying the post-processing stage. With respect to environmental performance, gasification has an impact similar to those of pyrolysis and recycling, but the performance could improve with sufficient research and development.

Keywords: gasification; co-gasification; thermodynamic equilibrium model; Gibbs free energy minimization; plastics; sargassum

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1. Introduction

Adequate waste management is of worldwide concern because materials such as plastics have a long degradation time and can decompose into smaller debris, such as microplastics, which present hazards to the environment and human health [1–3]. Plastic waste (including microplastic waste) is usually found as a mixture of plastics, where the three most prevalent are polypropylene (PP), polyethylene (PE), and polyethylene terephthalate (PET) [4,5]. However, plastics are not the only kind of solid waste that represents a challenge to society. For example, macroalgae, such as sargassum, cause problems in different coastal environments due to its sheer abundance; it has been reported that around 1 million tons wash ashore the Gulf of Mexico every year [6,7]. Highly abundant macroalgae cause environmental, health, and economic damage [2,6,7]. To combat the issues, researchers have proposed different alternatives to use sargassum; for example, sargassum has been used to develop food supplements [8,9]. Unfortunately, the opportunities for food production using sargassum can become compromised in some circumstances, such as when microplastics are present in the algae [10–12]. When algae are contaminated with microplastics, these cannot be used for food production, and an alternative methodology must be employed for their usage. Technologies, such as pyrolysis and gasification, are popular to convert biomass and waste to energy and have received attention due to the versatility of the products; however, challenges exist in different forms [13–15]. For example, sargassum usually has a low hydrogen content; hence, its use as a fuel precursor can be limited [16]. Li et al. [17] gasified *Sargassum horneri* biochar and found that hydrogen-rich syngas was produced, attributed to its high ash content; it was noted, however, that the high ash content might be related to the algae-growing environment. Hence, it might be different for different cases. He et al. [18] gasified macroalgae in a fixed-bed reactor using sand and ash as the bed materials and found that the large amount of ashes in the macroalgae caused agglomerates in the bed. On the other hand, the thermal treatment of plastics can be complicated due to the materials melting and clogging the reactors [19,20]. Co-feeding biomass with other substances (coal, peat, or plastics) during either pyrolysis or gasification is an alternative to enhance the product properties, as, for example, the addition of coal can increase the overall carbon conversion [21–27]. Kositkanawuth et al. [22] experimented with the co-pyrolysis of sargassum and polystyrene and found that the addition of plastics to the pyrolysis of sargassum increased the product yield and hydrogen contents in the formed bio-oils. Cheng et al. [23] co-pyrolyzed sargassum and poplar and found that the co-pyrolysis enhanced the process since lower temperatures were required to achieve the material's pyrolysis, when compared to the pyrolysis of a single feedstock. Understanding the characteristics of the products of the co-gasification of sargassum with plastics represents an opportunity to address two important types of solid waste while producing a chemical of interest [28,29]. When reactors have a long residence time (sufficient to achieve thermodynamic equilibrium), thermodynamic equilibrium models can be used [15]. The thermodynamic equilibrium assumption is adequate for downdraft and some fluidized bed gasifiers [15].

This work consists of a modelling study to determine the characteristics of the syngas (a mixture of H_2, CO, CO_2, CH_4, and other compounds) produced from the air co-gasification of different biomasses (namely, sargassum) with a mixture of plastics at different operation conditions, with the aim of understanding the potential of gasification as a waste-management option, in terms of energetic and environmental performance. Particularly, the algae that grows in abundance and are washed ashore beaches in Mexico (*Chlorella vulgaris* (CV), *Sargassum natans* (SN), and *Sargassum fluitans* (SF)) were used as biomass feedstock [6], while a mixture of PP/PE/PET (3.6% PP, 51.9% PE, and 29.3% PET) was used to represent plastics; the plastic mixture composition was based on the common plastic mixture composition found in waste [30,31]. In the study, the effect of the gasification temperature, feedstock composition, and equivalence ratio on the syngas' lower heating value (LHV), cold gas efficiency (CGE) process, syngas tar content, and gas composition was analyzed. The gas LHV is an indicator of the amount of energy released during the combustion of a fuel, while the process CGE is an indication of how much energy is released from oxidizing the produced syngas in comparison with the feedstock. The tar content is an important parameter for gasification because tar must be reduced significantly (or completely) for most downstream syngas applications [32]. The gas composition is important since it defines potential upgrade routes for usage (for example, the H_2/CO ratio defines the capabilities of the syngas for Fischer–Tropsch synthesis) [33]. Finally, plastic has several well-studied and well-documented waste management methodologies; hence, these must be compared with the proposed co-gasification process. For this, an environmental performance analysis is presented in the form of a life cycle assessment comparing different waste disposal alternatives for plastics: landfill, recycling, and gasification [34,35].

The structure of the work is as follows: Firstly, the thermodynamic model is described, together with the methodology followed for the validation of the developed model. Afterwards, details on the LCA methodology are presented. Subsequently, Section 3 shows the results from the parametric analysis, where the input parameters (fuel to air, gasification temperature, and mixture composition) are varied to determine their effect on the gas quality and composition. Afterwards, a life cycle assessment (LCA), where the different waste management scenarios are analyzed, is presented. Finally, a summary and a series of conclusions are presented.

2. Methodology

2.1. Model Description

A thermodynamic equilibrium stoichiometric model, based on an in-house-developed Python code, was used to simulate the gasification. The model calculates the equilibrium composition (based on the feedstock ultimate analysis) of the gasification products (syngas) following the Gibbs free energy minimization method [36]. The model calculates the Gibbs free energy at different compositions and temperatures fulfilling a material and energy balance; the composition of the syngas is then fixed at the point where the Gibbs free energy is the lowest and the balance is fulfilled. For the material balance, it is assumed that the potential products are common products reported in experiments [37,38].

To represent the decomposition of biomass and plastics, the following pseudo-reactions were used:

$$CH_{\alpha}O_{\beta} + \gamma_1(O_2 + 3.76N_2) \leftrightarrow x_1H_2 + x_2CO + x_3CO_2 + x_4CH_4 + x_5C_6H_6 + x_6H_2O + x_7C + 3.76\gamma_1N_2 \quad (1)$$

$$C_{10}H_8O_4 + \gamma_2(O_2 + 3.76N_2) \leftrightarrow y_1H_2 + y_2CO + y_3CO_2 + y_4CH_4 + y_5C_6H_6 + y_6H_2O + y_7C + 3.76\gamma_2N_2 \quad (2)$$

$$C_3H_6 + \gamma_3(O_2 + 3.76N_2) \leftrightarrow z_1H_2 + z_2CO + z_3CO_2 + z_4CH_4 + z_5C_6H_6 + z_6H_2O + z_7C + 3.76\gamma_3N_2 \quad (3)$$

$$C_2H_4 + \gamma_4(O_2 + 3.76N_2) \leftrightarrow w_1H_2 + w_2CO + w_3CO_2 + w_4CH_4 + w_5C_6H_6 + w_6H_2O + w_7C + 3.76\gamma_4N_2 \quad (4)$$

where α and β are the mass factors corresponding to the ultimate analysis of the biomass; γ_i is a mass factor associated to the air used for gasification; and w_i, x_i, y_i, and z_i are the molar fraction of species i, products from the decomposition of the feedstock [37]. Benzene (C_6H_6) is used to represent tar, since it is a well-known tar precursor refractory in nature [37]. Moreover, it is assumed that benzene can react and form other tar, toluene (C_7H_8) and naphthalene ($C_{10}H_8$), through the following reactions [38,39]:

$$C_6H_6 + CH_2 \leftrightarrow C_7H_8 \quad (5)$$

$$C_6H_6 \leftrightarrow C_6H_5^* + H^* \quad (6)$$

$$C_6H_5^* + C_2H_2 \leftrightarrow C_8H_7^* + H^* \quad (7)$$

$$C_8H_7^* + C_2H_2 \leftrightarrow C_{10}H_9^* \quad (8)$$

$$C_{10}H_9^* \leftrightarrow C_{10}H_8 + H^* \quad (9)$$

The mass of air added to the gasifier is calculated with the equivalence ratio, ER [28]:

$$ER = (F/A)/(F/A)_{Stoic} \quad (10)$$

where F/A is the fuel/air mass ratio, and the subindex “stoic” corresponds to stoichiometric quantities. The fuel mass is calculated using the sum of the ultimate analysis percentages of carbon, hydrogen, oxygen, and nitrogen in the feedstock, multiplying by the corresponding molecular weight.

To complete the thermodynamic equilibrium calculations, the following equations were used [37]:

$$CO + H_2O \leftrightarrow CO_2 + H_2 \tag{11}$$

$$C + 2H_2 \leftrightarrow CH_4 \tag{12}$$

$$k_6 = (P_{CO2}P_{H2})/(P_{CO}P_{H2O}) \tag{13}$$

$$k_7 = P_{CH4}/P_{H2} \tag{14}$$

where k_i and P_j correspond to the equilibrium constant for equation I and the partial pressure of gas j, respectively [6,16]. The ultimate analysis of the gasified biomasses was extracted from references [6,22], whose details can be found in Table 1, while the plastic mixture composition was obtained from [31].

Table 1. Ultimate analysis (in % weight) of the biomass feedstock used in the simulations.

Element	**Algae Species**		
	Chlorella vulgaris	***Sargassum natans***	***Sargassum fluitans***
C	52.1	39.78	38
H	6.5	8.91	5.5
O	30.9	43.89	55
N	9.95	5.48	1.5
S	0.55	1.92	0

The energy balance for the reactions is calculated as follows:

$$\sum_j \Delta H_j^0 = \sum_j \sum_i x_{ji}\ \Delta H_i \tag{15}$$

where x_{ji} is the molar fraction of the ith product from the decomposition of reactant j, ΔH_j^0 is the standard enthalpy of the formation of species j, and ΔH_j are the changes in enthalpy of species j. The heat of formation for the biomass and the plastics is calculated following the relationship found in [40]:

$$\Delta H^0_{Biomass} = \Delta H^0_{CO2} + \Delta H^0_{H2O} + (12 + \alpha + 16\beta)\ LHV_{Biomass} \tag{16}$$

The following mathematical expressions were used to calculate the LHVs [41]:

$$LHV_{Feedstock} = LHV_{Biomass} + LHV_{PP} + LHV_{PE} + LHV_{PET} \tag{17}$$

$$LHV_{Biomass} = 34.835w_C + 93.87w_H - 10.80w_O + 6.28w_N + 10.465w_S \tag{18}$$

$$LHV_{Syngas} = 4.180(2.57\Phi H_2 + 3.0\Phi CO + 8.54\Phi CO_2 + 15.13\Phi CH_4) + 0.0422\Phi C_6H_6 \tag{19}$$

where Φ_i is the dry mole fraction of the species i. The LHV values of the different plastics can be found in reference [42].

Finally, the gas yield is calculated using the following relationship [41]:

$$v_{Syngas}/m_{feedstock} = 22.4(\Phi H_2 + \Phi CO + \Phi CO_2 + \Phi CH_4 + \Phi N_2 + \beta \sum \gamma_i)/m_{feedstock} \tag{20}$$

where $m_{feedstock}$ is the feedstock mass.

The produced syngas was analyzed with respect to the gas lower heating value (LHV), gas volume composition, H_2-to-CO ratio, and CGE.

The CGE is a relationship between the LHV of the produced gas (LHV_{Syngas}) and the LHV of the feedstock ($LHV_{Feedstock}$), and can be used to determine the efficiency of the process with respect to the employed feedstock [28]:

$$\mathrm{CGE} = \mathrm{LHV_{Syngas}}/\mathrm{LHV_{Feedstock}} \tag{21}$$

2.2. Model Validation

To validate the model, the experiments from the literature [26,43–48] were replicated and the experimentally produced syngas composition was compared with the composition estimated by the model. The mean absolute error (MAE) was used as an indicator of the accuracy of the model. The validation was conducted in two stages: (1) by experiments (i.e., the MAE of the whole experiment was calculated) and (2) by species (that is, for all of the experiments, the MAE of each species was calculated to determine whether the model is less accurate on the calculation for a particular species). Information on the validation by experiment can be found in Table 2, while the validation by species can be seen in Table 3. In Table 3, it can be seen that the model is the least accurate when the tar concentration is calculated; this occurs because tar has a diverse composition (while the present model only considers benzene, toluene, and naphthalene) and formation/decomposition rates [32].

Table 2. Details on the feedstock from the experiments used to validate the model and validation by experiment.

Reference	**Gasifier Type**	**Feedstock Composition**				**MAE**
		C	**H**	**O**	**N**	
[47]	FB	53.2	5.5	37.1	0.3	2.407
[44]	DD	43.83	5.95	45.01	0.97	1.556
[46]	FB	51.0	6.0	42.8	0.1	4.017
[45]	DD	50.6	6.5	0.2	42.0	3.601
[48]	FB	86.42	12.28	0	0.72	2.045
[43]	FB	80.8	12.8	5.1	0.2	1.038
[26]	DD	42.38	5.24	35.41	1.78	0.019
[26]	DD	85.32	14.68	0	0	0.019

Table 3. Validation of the model by species.

Species	**MAE**
H_2 [a]	1.872
CO [a]	1.165
CO_2 [a]	1.142
CH_4 [a]	1.015
N_2 [a]	0.044
Tar [b]	2.5

[a] In volume percent [b] In g/Nm^3.

The MAE was calculated using the equation:

$$\mathrm{MAE} = \sum |\mathrm{Y_i} - \mathrm{X_i}|/\mathrm{n} \tag{22}$$

where Yi are the volumes of the species i (H_2, CO, CO_2, CH_4, and tar) calculated using the developed model, Xi are the volumes of species i reported in the literature, and n is the number of values.

2.3. Life Cycle Assessment

A life cycle assessment (LCA) is a methodology where the impact of the different stages in the life cycle of a product are evaluated [49]. A basic principle of a LCA is to define the goal and scope of the study; in this work, since the aim is to assess co-gasification (COG) of plastics with biomass as a waste-handling alternative, the goal is to determine the environmental impact of using plastics as feedstock for gasification and comparing it with pyrolysis (PY), recycling (REC), and landfilling (LF) as end-of-life scenarios. The system boundary considers the processes involved in the different end-of-life scenarios (for example, plastic separation and grinding). The functional unit is the management of 1 ton of plastics. ReCiPe 2016 endpoint was used as an impact assessment method due to its robustness and ease of interpretation [50]. The inventory data for the gasification and pyrolysis of different plastic wastes were collected from the literature [51–53], and the energy consumption was calculated using the energy balance, assuming 80% efficiency (that is, only 80% of the energy consumed during the process is actually used for the gasification); the information was complemented with the Ecoinvent 3 database [54].

Importantly, since gasification leads to the production of syngas, it was assumed that the produced gas energy (in MJ, calculated from the gas yield and the gas LHV) prevented the environmental burden of the same amount of energy from fossil sources. Potential uses of the syngas, such as the synthesis of additives or other liquid fuels, is not considered in the LCA.

3. Results and Discussion

The simulations were conducted under different conditions to determine the effect of the parameters on the syngas LHV, CGE, tar concentration, and H_2/CO ratio. The following parameters where varied: gasification temperatures (650, 750, and 850 °C), ERs (0.25, 0.35, and 0.45), and plastics/biomass mass ratios (called MR from here onwards) (0, 0.25, 0.50, 0.75, and 1.0); all the simulations were conducted using the three biomasses (CV, SN, and SF). The MR is defined as follows:

$$\mathrm{MR} = \mathrm{m}_{\mathrm{Plastics}}/\mathrm{m}_{\mathrm{Biomass}} \tag{23}$$

where $\mathrm{m}_{\mathrm{Plastics}}$ is the mass of the plastic mixture, while $\mathrm{m}_{\mathrm{Biomass}}$ is the mass of sargassum added to the gasifier. The LCA results are presented considering the gasification of plastics only, since, of the different feedstocks, plastics comprise a significant environmental burden.

3.1. Effect of the Gas LHV

Figure 1 shows the LHV of the syngas at different gasification temperatures and ERs using pure (a) CV, (b) SN, and (c) SF as biomass feedstocks. It can be seen that, regardless of the biomass, the LHV increases with the temperature and decreases with the ER. Higher temperatures promote the formation of H_2 (from the decomposition of the feedstock) and CO (from the water–gas shift reaction, WGS), which increase the gas LHV. On the other hand, the LHV decreases with the ER because the syngas becomes diluted with N_2 from the gasifying agent. The trend followed by the different biomasses (in terms of LHV) changes with the operation conditions. At ER = 0.25, SN is the biomass that produces the best gas (in terms of LHV), followed by CV and SF. At ER = 0.45, the best biomass is still SN, but CV leads to the gas with the lowest LHV. Interestingly, the LHVs of the three algae are 2.160 MJ/kg for the CV, 1.803 MJ/kg for the SN, and 1.255 for the SN, calculated using Equation (18).

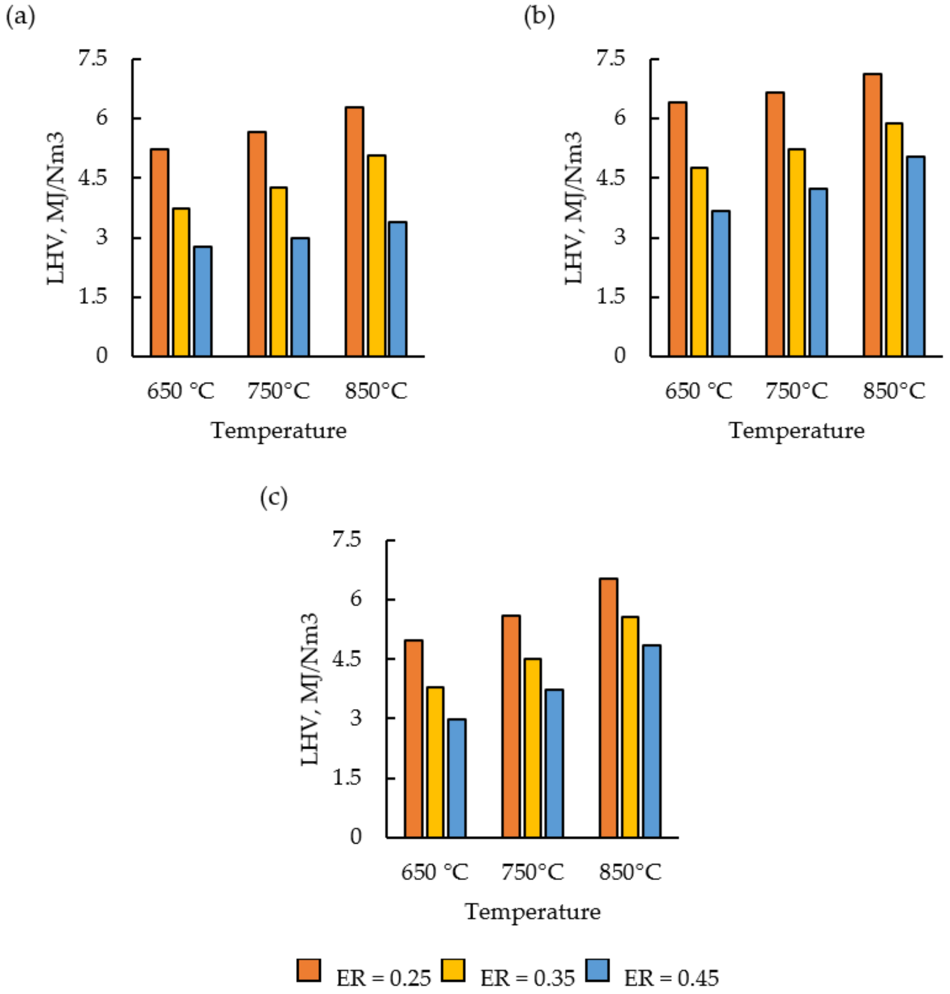

Figure 1. Lower heating values of the syngas produced from the gasification of (**a**) CV, (**b**) SN, and (**c**) SF as a function of the temperature and equivalence ratio.

When the plastic mixture was added to the feedstock, the composition of the produced gas changed; the results obtained from mixing the algae with plastics, in terms of LHV, are shown in Figure 2, where (a) shows CV, (b) shows the gasification of SN, and (c) shows the gasification of SF. It can be seen that, for CV and SF, the LHV increased with the increase in MR, while SN has the opposite behavior. The behavior for the CV and SF cases is associated to the abundance of H atoms in plastics, which favors the formation of H_2; on the other hand, since SN has a higher H content than the other biomasses, the plastics contribute to the formation of tar instead of hydrogen (as is explained in following subsections). The effect of the presence of plastics is particularly noticeable when ER = 0.25, where there is little N_2 diluting the gas. Moreover, it can be seen that the LHV approaches a maximum value at MR = 1.0 and T = 850 °C (with the exception of the SN case); this can be associated to the produced tar being completely decomposed, as is discussed in Section 3.2. In the case of SN, the LHV decreases with the MR because of the relatively high hydrogen content in SN; the addition of carbon atoms to plastics results in the formation of CO and CO_2 instead of H_2, which does not contribute as much to the LHV [41]. With respect to the biomasses, the gas with the highest LHV is produced from SN, followed by SF and CV.

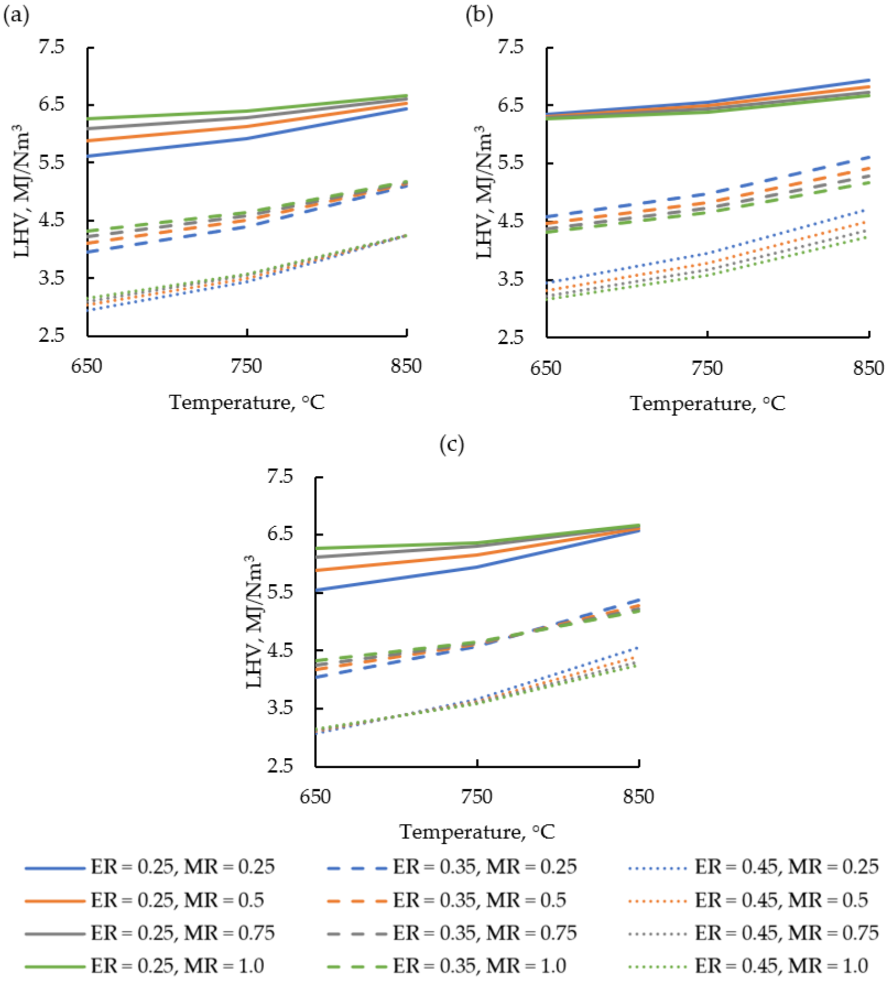

Figure 2. Lower heating values of the syngas produced from the co-gasification of a mixture of plastics with (**a**) CV, (**b**) SN, and (**c**) SF as a function of the temperature and equivalence ratio.

3.2. Effect of the Gas Tar Concentration

Figure 3 shows how the tar concentration varies with the operation conditions. For every case, the tar concentration decreases when the temperature or the ER increase; this occurs because higher ERs and temperatures promote the decomposition of tar (hydrocarbons) into CO and CO_2 due to oxidation reactions (that is, the oxygen in the air oxidizes the heaviest hydrocarbons to produce CO and CO_2), which also results in an increase in LHVs (although, in the end, the ER also results in an increase in N_2 content, with the dilution of the gas and the decrease in the LHV). At the same time, it can be seen that the tar concentration increases with the MR; this occurs because the C atoms (present in PP, PE, and PET) and the O atoms (present in the PET) thermodynamically favor the formation of tar. However, since tar is practically completely removed at high temperatures and ERs, these operation parameters can be tweaked to achieve the required syngas LHVs. At high ERs and temperatures, tar is practically completely decomposed for each of the different biomasses and regardless of the mixing ratio. Therefore, in order to produce gas with a low amount of tar, either a large amount of air (or oxygen) or high gasification temperatures should be used.

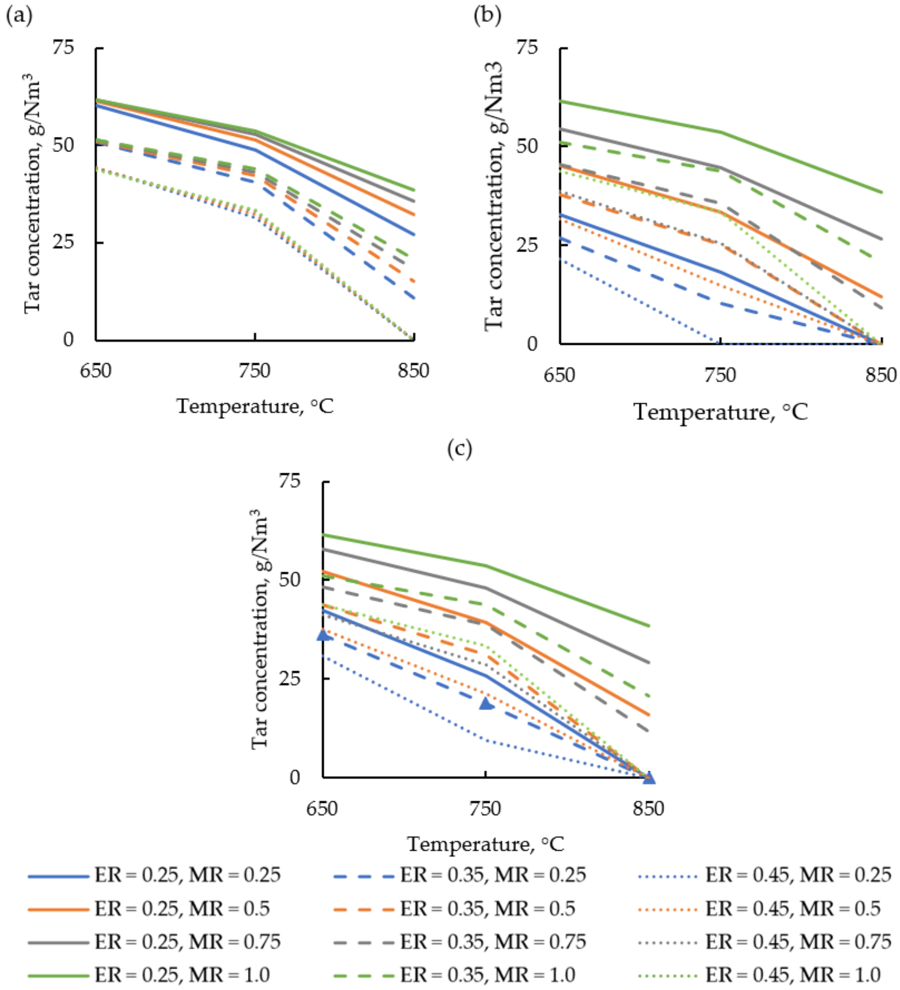

Figure 3. Tar (benzene) concentration in the syngas produced from the co-gasification of a mixture of plastics with (**a**) CV, (**b**) SN, and (**c**) SF as a function of the temperature and equivalence ratio.

3.3. *Effect of Cold Gas Efficiency*

Figure 4 shows the CGE ratio of the gasification process for the different operation conditions. For all the biomasses, MRs, and ERs, the CGE increases with the temperature, since high temperatures lead to the decomposition of hydrocarbons into H_2 and CO, both of which have important heating values. The CGE decreases with the ER due to the addition of the practically inert N_2 to the syngas. Moreover, although not particularly noticeable for the CV case, increasing the MR decreases the CGE; this occurs due to increasing the feedstock LHVs (due to the addition of plastics with a large percentage of hydrogen). For example, at MR = 0, the feedstock LHV is 17.83 MJ/kg, whereas at MR = 0.5, the feedstock LHV is 30.11 MJ/kg. On the other hand, the gas LHV is constrained by the amount of H_2 formed from the decomposition of the feedstock. Moreover, since the WGS reaction is exothermic (that is, favored at low temperatures), the reverse WGS reaction leads to the formation of CO and H_2O (gases with a low LHV), instead of H_2 (a gas with a high LHV) and CO_2. This indicates that, even without accounting for the energy required to achieve high temperatures, exceedingly high temperatures might be prejudicial to the formation of syngas with high H_2, and to the whole gasification process.

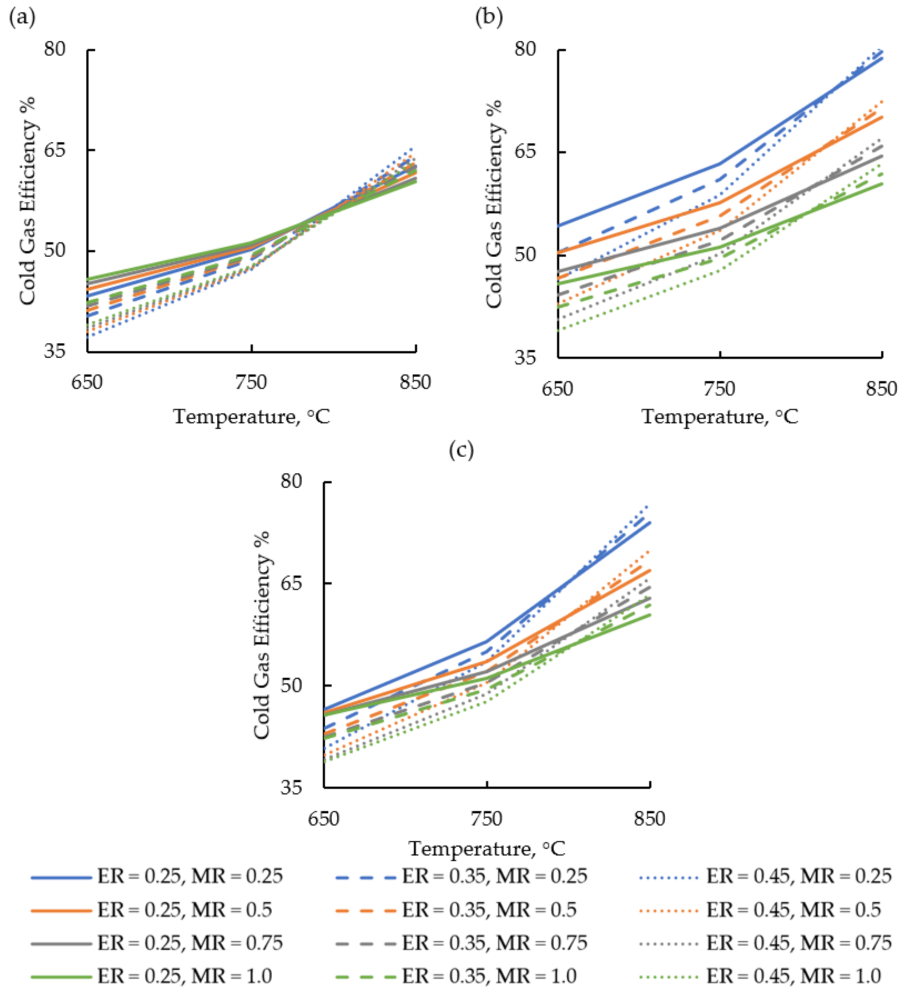

Figure 4. Cold gas efficiency estimated from the co-gasification of a mixture of plastics with (**a**) CV, (**b**) SN, and (**c**) SF as a function of the temperature and equivalence ratio.

3.4. Effect of the Gas Composition

The effect of the co-gasification conditions in the gas composition were explored by calculating the dry gas composition of the syngas under different conditions (in volume percentages). The dry composition expresses the percentage of H_2, CO, CO_2, CH_4, and N_2 in the syngas and can be found in Figure 5, while Figure 6 shows the normalized syngas composition (without considering the N_2 in the gas composition); in all cases, the data correspond to simulations using CV with the plastic mixture. It can be seen that the N_2 increases with the ER, which is expected, since a higher ER implies more air (hence, N_2) is added to the syngas. The N_2 decreases with the temperature, not because less N_2 is present, but because more of the feedstock decomposes to the other products (H_2, CO, CO_2, and CH_4). On the other hand, the H_2 and CO contents increase with the temperature, while the CO_2 and CH_4 decrease, and the later decreases marginally. Ideally, since CO_2 is a byproduct, its contents in the produced syngas should be as low as possible from the production stage or otherwise removed in an additional process stage [55]. With respect to CH_4, although the amount of methane produced is not particularly large, it can be reformed to increase the H_2 contents in the syngas or recovered for the production of other chemicals of interest [56,57]. Additionally, it can be noted that the N_2 increases slightly with the MR; this effect is not associated to an actual increase in the amount of N_2 in the syngas, but to a decrease in the percent of CO, CO_2, and H_2 due to tar formation (which is expressed in g/Nm^3 and not volume percent), in agreement with Figure 3.

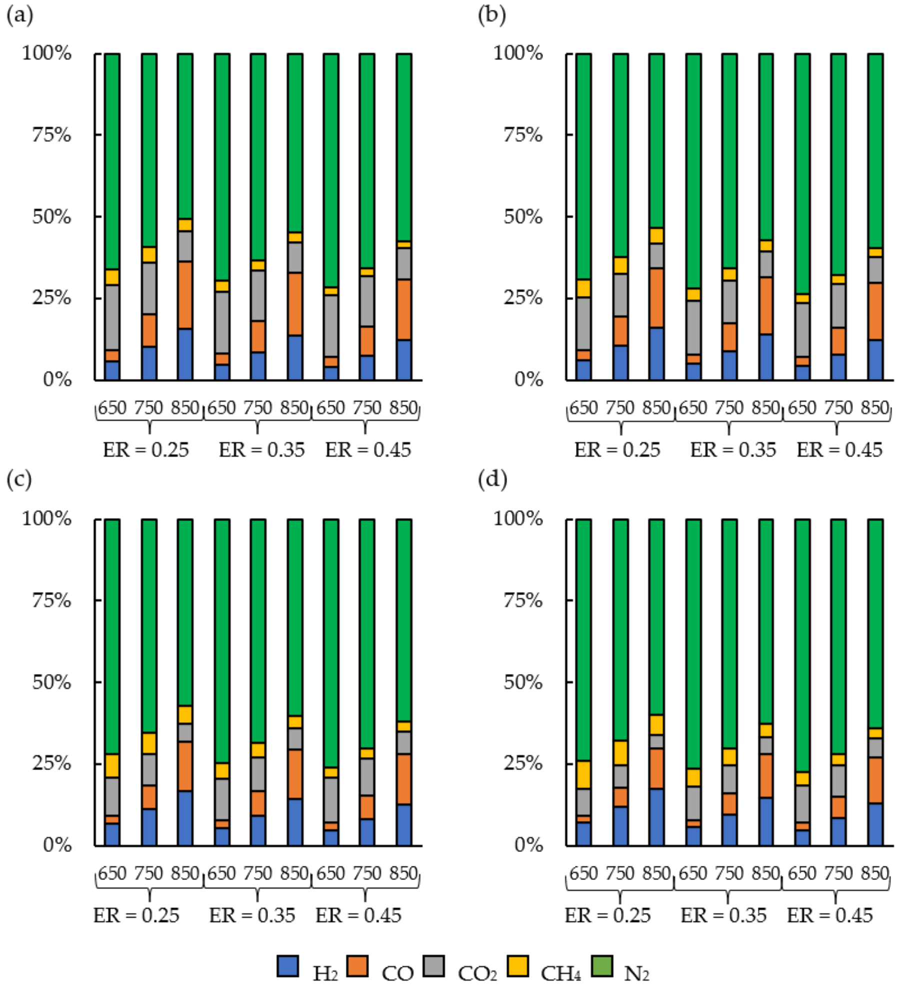

Figure 5. Syngas percent composition, on a dry basis, from the co-gasification of CV at (**a**) MR = 0, (**b**) MR = 0.25, (**c**) MR = 0.75, and (**d**) MR = 1.5.

When observing the normalized (without N_2) syngas composition, it can be seen that the gas that has the largest increase with the temperature is CO. The normalized syngas composition allows for a better analysis of the effect of the MR in the syngas composition; when the MR increases, the amount of H_2 and CH_4 increases, while the CO and CO_2 decreases. The highest increase is exhibited by the H_2 (from 17% at 650 °C, an ER of 0.25 and MR of 0, to 27% at the same temperature and ER, but an MR of 1.5), while the highest decrease occurs for the CO_2 (from 58% at 650 °C, an ER of 0.25 and MR of 0, to 32% at the same temperature and ER, but an MR of 1.5). This indicates that the addition of plastics results in a decrease in the produced CO_2, which could result in a decreased environmental burden when considering syngas post-treatment (which is outside of the scope of the present work). Arguably, depending on the end-use of the syngas, the addition of plastics is beneficial to increase the H_2 and CO in the syngas (with the exception of SN) and diminish the production of CO_2, resulting in less syngas post-treatment [58].

An important parameter for syngas is the H_2/CO ratio, since it dictates what kind of usage and/or upgrading the syngas can receive [33,59]. Figure 7 shows the H_2/CO ratio of the syngas using the different operation conditions. The results indicate that the H_2/CO ratio decreases with the temperature and ER and increases with the increase in MR (as seen when comparing Figure 7a–c). The results indicate that, without the addition of plastics, the biomass that yields the syngas with the highest LHV is SN, but with the addition of plastics, the three algae result in similar quality syngas.

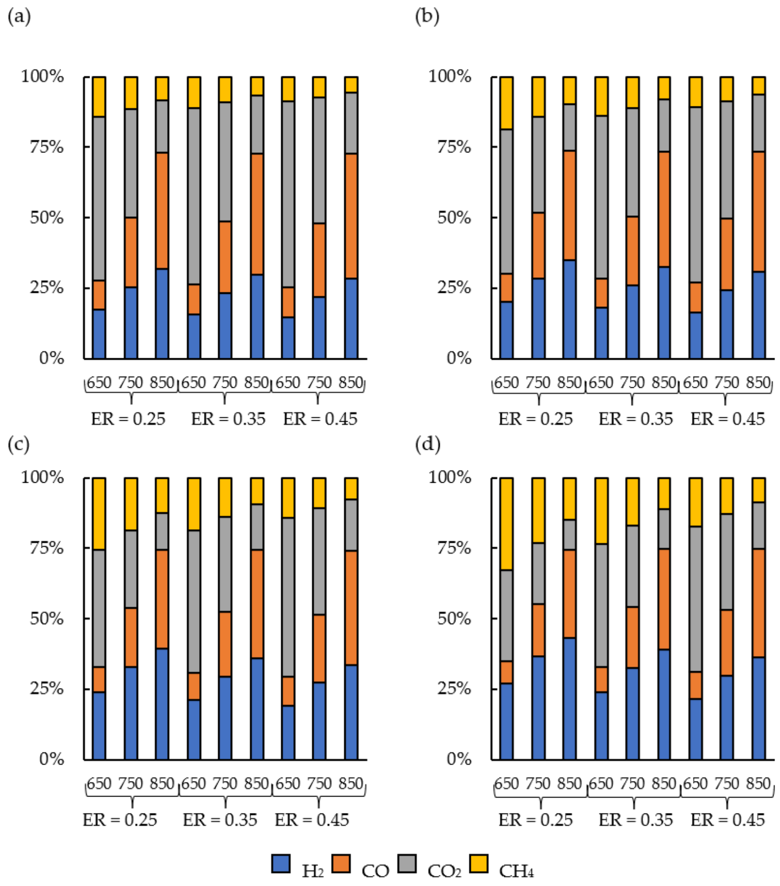

Figure 6. Normalized syngas percent composition, on a dry basis, from the co-gasification of CV at (**a**) MR = 0, (**b**) MR = 0.25, (**c**) MR = 0.75, and (**d**) MR = 1.5.

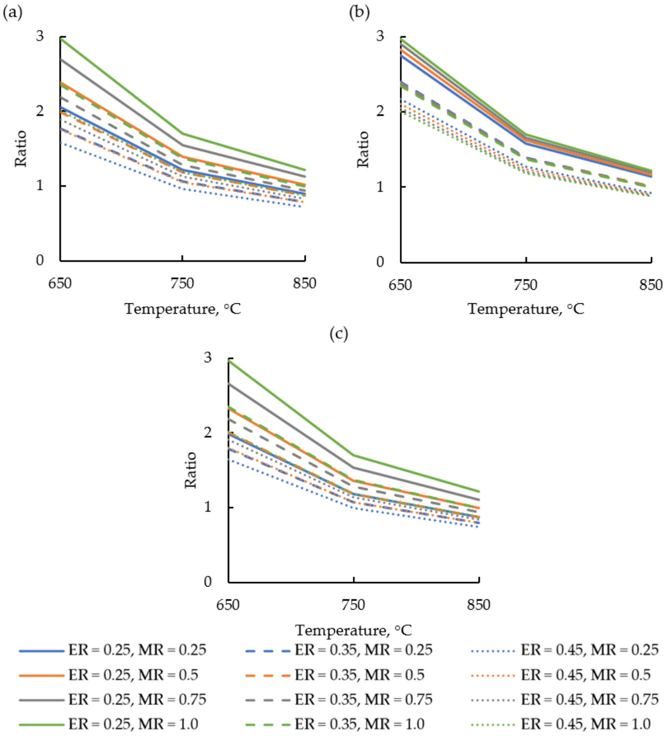

Figure 7. H_2/CO ratio in the syngas produced from the co-gasification of a mixture of plastics with (**a**) CV, (**b**) SN, and (**c**) SF as a function of the temperature and equivalence ratio.

All in all, it can be concluded that the addition of plastics to algae during gasification has advantages in terms of syngas quality and provides an alternative for waste handling. Moreover, since the H_2/CO ratio decreases with the temperature, for applications where the H_2/CO must be more than 2 (for example, the synthesis of fuel additive), gasification at low temperatures is preferred, even if the syngas has a lower LHV.

3.5. Effect of the Used Biomass

The studied algae have different characteristics; hence, the products from gasification are expected to vary. Figure 8 shows (a) the gas LHV, (b) the gas H_2/CO ratio, (c) the gas tar contents, and (d) the CGE from gasifying the biomass at T = 650 °C, ER = 0.25, and a different MR. As discussed in Section 3.1, the syngas LHV increases with the MR (with the amount of plastics in the mixture). However, Figure 8a shows that, when SN is gasified, albeit slightly, the syngas LHV decreases with the increase in the plastic content; this occurs because of the relatively high hydrogen content in SN (the H content in SN, in weight percent, is 8.91%; as a reference, Vassilev et al. [60] analyzed 86 varieties of biomass and found that the average hydrogen content in biomass is 6.3). The same behavior occurs with the CGE (Figure 8b), since the biomass has a higher LHV than the biomass with the plastic mixture. With respect to the tar concentration, although it increases with the MR for all the biomasses, the effect is almost negligible in the CV case due to the higher C and intermediate H contents. Since tar are aromatic hydrocarbons, they require C for their formation, and the tar formation is limited by the presence of C in the biomass sample; this limitation does not occur in the CV case.

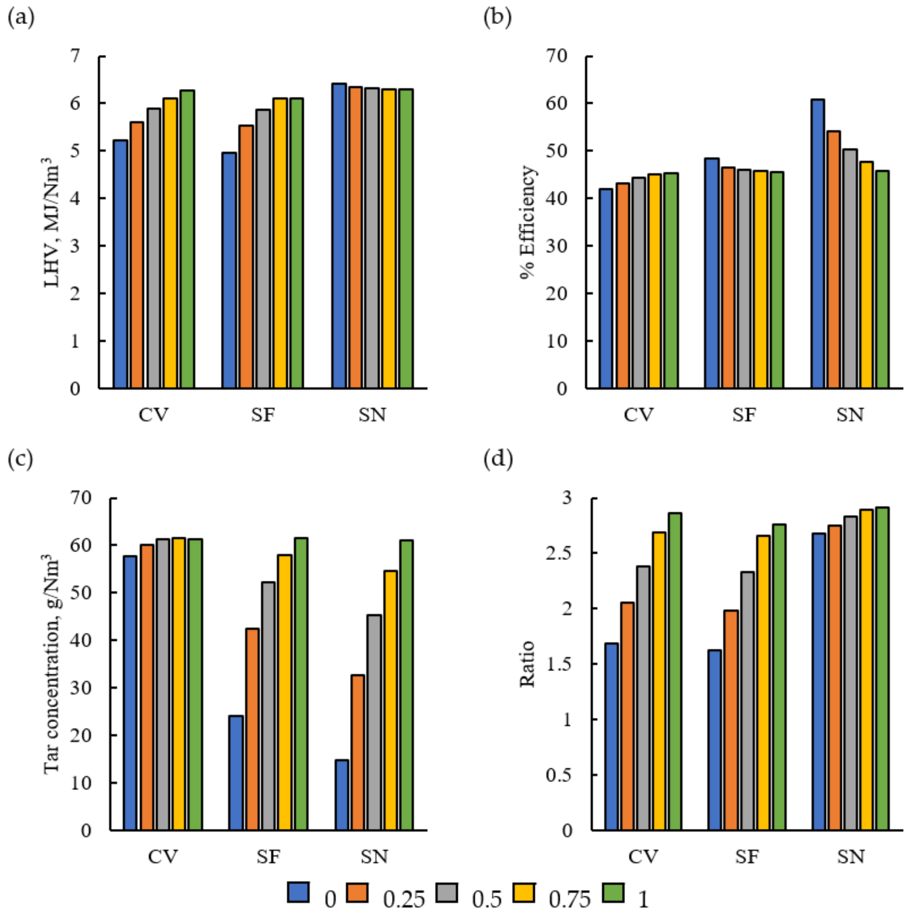

Figure 8. (**a**) Lower heating value, (**b**) cold gas efficiency, (**c**) tar concentration, and (**d**) H_2/CO ratio, calculated in the simulations from the co-gasification of a mixture of plastics with CV, SN, and SF at 650 °C and an equivalence ratio of 0.25 as a function of the MR.

Finally, Figure 8d shows that the H_2/CO ratio increases with the MR, and that the SN sees an almost negligible increase attributed to its composition.

Importantly, it can be seen that, although for low MRs (almost pure biomass gasification) the gas H_2/CO ratio is different for the three algae, when the MR approaches 1, the difference becomes smaller; hence, mixing plastics with biomass in co-gasification can be useful to regulate the produced syngas H_2/CO ratio. This might be particularly useful, since some applications require a specific H_2/CO ratio (for example, Fischer–Tropsch synthesis).

3.6. Life Cycle Assessment

A life cycle assessment of the different end-of-life scenarios for the plastic mixture was conducted. For the environmental burden prevention, it was assumed that the produced syngas has a LHV of 6.2 MJ/Nm^3 and that the gas yield was 2.2 $Nm^3{}_{syngas}/kg_{feedstock}$, which are the mean values from the different co-gasification simulations from this work. Mean values were used to provide mean LCA results, since the operation conditions of an actual process using sargassum and plastics might be different from the particular cases reported in this work but might be closer to the average values. Figure 9 shows the impact scores of the different processes; Figure 9a portrays the impact to ecosystems (ES), Figure 9b shows the impact to resources (RE), Figure 9c shows the impact to human health (HH), and Figure 9d shows the normalized damage assessment considering the three above categories. Importantly, impact categories whose impact was lower than 1% of the maximum impact are not shown in the graphics. In the impact to ecosystems graphic (Figure 9a), the categories shown are the ones with the largest effect to the species.year indicator, and are water consumption in terrestrial ecosystems (WC), land use (LU), freshwater eutrophication (FWE), terrestrial acidification (TA), ozone formation in terrestrial ecosystems (OF), and global warming in terrestrial ecosystems (GWT). The fossil resource scarcity is the only indicator that contributes significantly to the resources category, while the human health category has contributions from the human non-carcinogenic toxicity (NCT), human carcinogenic toxicity (HCT), fine particulate matter formation (PM), and global warming and human health (GWG) indicators. In Figure 9a, it can be seen that most of the damage occurs in the GWT category; the performance, from worst to best, follows the sequence LF, REC, COG, and PY. With regards to RE, LF and PY are the best performing scenarios, whereas RE exhibits the worst performance (due to the energy consumption in mechanical separation and waste treatment); this burden can be alleviated with the implementation of technologies such as renewable energy [52]. On the other hand, when inspected together, Figure 9c,d show that most of the damage to human health occurs due to particulate matter (since PM in Figure 9c follows the same profile as HH in Figure 9d). Disregarding the LF scenario, PM and GWG are the two most important areas of opportunity. Particularly, fully developed and matured co-gasification or systems could reduce the problems related to particles, since, for example, instruments such as filters could diminish the PM environmental burden. Moreover, the processes could benefit from process intensification, co-generation, and similar technologies [61]. Arguably, REC, COG, and PY have similar environmental performances and are superior to the LF option. Without considering the level of maturity of the different technologies, the use of REC, COG, or PY in a particular location will depend on the end application of the recovered materials, either from mechanical or chemical recovery. For example, if the desired application is the production of polymers, REC or PY should prove superior than COG; on the other hand, for FT synthesis, COG could be the better option when compared to PY [33]. Finally, with mature enough technology, COG can have a particular advantage over REC in developing countries (developing countries have a low recycling rate due to a lack of incentives [62]), where energy and commercial chemical productions can prove attractive.

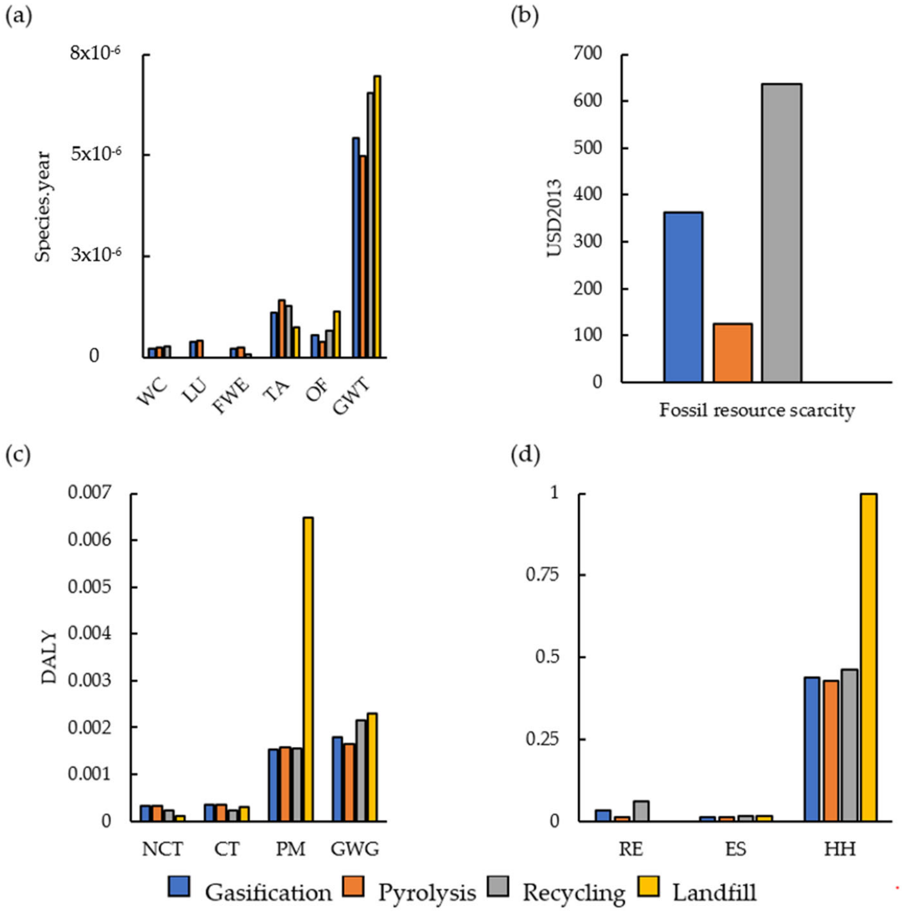

Figure 9. Results from the LCA using the ReCiPe Endpoint method. (**a**) Impact to the ecosystem, (**b**) impact to resources, (**c**) impact to human health, and (**d**) normalized impacts by category.

4. Summary and Conclusions

The co-gasification of three macroalgae species (*Chlorella vulgaris*, *Sargassum natans*, and *Sargassum flutans*) with a mixture of plastics (polypropylene, polyethylene, and polyethylene terephthalate) under various operation conditions was simulated and studied parametrically using an in-house-developed thermodynamic equilibrium model. To ensure the accuracy of the results presented in this work, the developed model was validated by reproducing results from the literature. The gasification temperature, equivalence ratio, and plastics/biomass ratio were varied, and the effects of said variation in the produced gas LHV, tar concentration, gas composition, H_2/CO ratio, and process cold gas efficiency were analyzed. In order to measure the environmental performance of the co-gasification of sargassum with the mixed plastics, a life cycle assessment was conducted to compare gasification as an end-of-life option for plastic management.

From the study, the following was concluded:

- Adding plastics to biomasses for co-gasification can enhance the gas LHV; however, this does not occur when the biomass has a high elemental hydrogen content, as in the case of *Sargassum natans*. Similarly, the cold gas efficiency of the process increases with the increase in plastic content, except in cases where the biomass has a high hydrogen content.
- The presence of plastic in the biomass during co-gasification promotes the formation of tar; hence, although the co-gasification of plastics and sargassum can be a useful waste-handling alternative, and increasing the plastic content increases the gas LHV, a high plastic content (high mixing ratios) might not be operational in cases where

the application of the produced gas requires a low tar concentration (for example, gas turbines).

- The addition of plastics for biomass co-gasification can be useful to regulate the produced syngas H_2/CO ratio, since increasing the amounts of plastics tends to homogenize the ratio.
- Increasing the MR increases the amount of H_2 and CH_4 produced during gasification, while the CO and CO_2 contents decrease.
- The damage assessment information obtained from the life cycle analysis indicate that the gasification of plastics has an impact somewhat similar to that of pyrolysis and recycling. Using the ReCiPe Endpoint methodology, it was found that the categories with the largest impact are human health followed by ecosystem damage, due to the formation of particulate matter and greenhouse gas releases, respectively.

All in all, the results indicate that, although the gasification of macroalgae might lead to otherwise low-quality syngas, co-gasifying them with plastics can be a waste management alternative. The gasification of plastic competes evenly with pyrolysis and recycling as waste management options, but with adequate process integration and technology maturity, it can become feasible. This can be particularly useful, since algae contaminated with microplastics can be limited in terms of management alternatives (for example, algae with microplastics cannot be use for food production).

Future work will include a model of a complete Integrated Gasification Combined Cycle Plant, considering the first and second law of thermodynamics, as well as the environmental performance of the system.

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