

Article

## Fluidized-Bed Gasification of Plastic Waste, Wood, and Their Blends with Coal

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**Abstract:** The effect of fuel composition on gasification process performance was investigated by performing mass and energy balances on a pre-pilot scale bubbling fluidized bed reactor fed with mixtures of plastic waste, wood, and coal. The fuels containing plastic waste produced less H<sub>2</sub>, CO, and CO<sub>2</sub> and more light hydrocarbons than the fuels including biomass. The lower heating value (LHV) progressively increased from 5.1 to 7.9 MJ/Nm<sup>3</sup> when the plastic waste fraction was moved from 0% to 100%. Higher carbonaceous fines production was associated with the fuel containing a large fraction of coal (60%), producing 87.5 g/kg<sub>Fuel</sub> compared to only 1.0 g/kg<sub>Fuel</sub> obtained during the gasification test with just plastic waste. Conversely, plastic waste gasification produced the highest tar yield, 161.9 g/kg<sub>Fuel</sub>, while woody biomass generated only 13.4 g/kg<sub>Fuel</sub>. Wood gasification showed a carbon conversion efficiency (CCE) of 0.93, while the tests with two fuels containing coal showed lowest CCE values (0.78 and 0.70, respectively). Plastic waste and wood gasification presented similar cold gas efficiency (CGE) values (0.75 and 0.76, respectively), while that obtained during the co-gasification tests varied from 0.53 to 0.73.

**Keywords:** fluidized bed gasifier; co-gasification; plastic waste; wood; coal; mass and energy balances

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## 1. Introduction

There is a growing interest to apply thermo-chemical process to different kind of wastes, considering the environmental impact of these materials and economic aspects of power plants. Gasification is a viable technology for the thermo-chemical conversion of biomass and wastes due to its greater environmental sustainability and to the production of valuable products from different fuels. Among all gasification technologies, fluidization is often chosen as a reference for its great operating flexibility: the good mixing properties that ensure uniform process conditions, while also simultaneously feeding different fuels; the possibility to utilize various fluidizing agents [1–4]; to operate with or without a specific bed catalyst [5–10]; to add reagents along the reactor height [11–14], and to feed fuels in different positions of the reactor [15–17]. On the other hand, during the gasification process, the unavoidable formation of contaminants such as tar, carbonaceous particles, and inorganics leads to an increase in operating costs and efficiency loss. Since the formation of these by-products is strictly correlated to the fuel structure and composition, the possible synergy between the products and the intermediates produced during the gasification of different materials could lead to improving the process performance, reducing carbon losses, and increasing producer gas energy content. Gasification plants can be conducted by co-feeding different fuels in order to produce better results in term of producer gas quality and energy saving respect to those obtained by utilizing a single material.

Co-gasification is a relatively new process where the industrial know-how is far from exhaustive, even if the scientific literature is full of interesting research studies on the co-feeding effect of different fuels into fluidized bed gasifiers. Authors do not always agree about the effects of co-feeding on the gasification process performance [18–22]. These differences are probably also due to the difficult comparison of the results obtained from different gasification technologies, different fuels, and different operating conditions. Generally, synergistic effects are attributed to the interaction between the produced volatiles or between the volatiles and the ashes contained in the char of the gasifying fuels [23,24]. Some authors reported that the composition of the feedstock affects the producer gas quality by means of non-additive models with a synergistic interaction among different materials during co-gasification test. The changes in the gas composition were non-linear and, consequently, it could not be predicted on the basis of gasification of the individual materials [18–20]. On the contrary, several studies did not observe any interaction [21,22].

Wilk and Hofbauer [18] performed co-gasification tests in a pilot dual fluidized bed reactor by using different mixtures of wood and different types of plastic material as feedstock. The results indicated that the product gas composition was strongly influenced by fuel mixtures. During the gasification tests more CO and CO<sub>2</sub> were produced from co-gasification than would be expected from linear interpolation of mono-gasification of wood and plastic. On the other hand, light hydrocarbons and tar in the product gas were considerably lower than presumed. Saw and Pang [19] conducted co-gasification experiment of blended lignite and wood in a pilot-scale dual fluidized bed steam gasifier. The experimental results showed that the producer gas yield and gas compositions were non-linearly correlated to the lignite to wood ratio, which indicated a synergy effect of the blending. The authors attributed synergistic effect on the tar production to the catalytic elements (Ca and Fe) contained in the blended chars. Feroso *et al.* [20], utilizing a lab-scale high-pressure gasification reactor, studied the effect of several operating variables and that of blending bituminous coal with

petcoke and biomass on gasification process performances. The results showed a positive deviation from the linear additive rule in the case of H<sub>2</sub> and CO production for the blends of coal with petcoke.

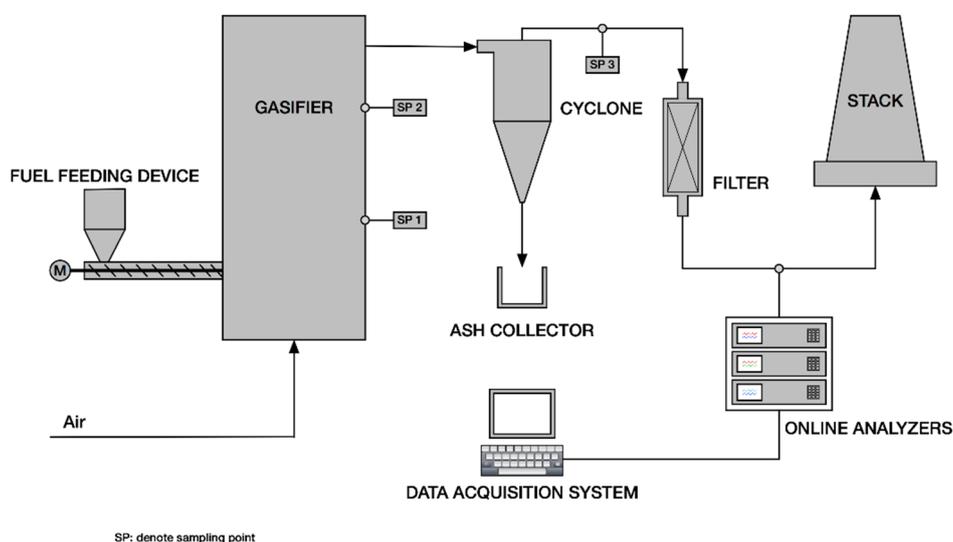
Conversely, Kumabe and co-workers [21] gasified woody biomass and coal with air and steam in a downdraft fixed bed reactor without observing an apparent synergy in terms of carbon distribution of products. The authors reported changes in the producer gas composition related to the increasing of biomass fraction in the mixture: a decrease in H<sub>2</sub> together with an increase in CO<sub>2</sub> content was observed. In addition, CO and hydrocarbons concentrations appeared independent by the fuel compositions. Aigner *et al.* [22] observed a linear relationship between the producer gas composition and wood ratio when a mixture of coal and wood was gasified. In particular, when H/O ratio in the fuel decreased, the H<sub>2</sub>/CO and H<sub>2</sub>/CO<sub>2</sub> ratios in producer gas decreased as well.

The aim of this study was to evaluate the effect of fuel composition on the gasification process by performing mass and energy balances on a pre-pilot bubbling fluidized bed reactor. The experimental runs were carried out by feeding five alternative fuels including mono-gasification tests of plastic waste and woody biomass, and co-gasification tests of plastic waste, wood, and coal mixtures.

## 2. Experimental Apparatus and Procedure

### 2.1. Experimental Apparatus

The experimental work was carried out utilizing a pre-pilot scale bubbling fluidized bed gasifier (BFBG) with a maximum feeding capacity of 5 kg/h, depending on the type of fuel (Figure 1).



**Figure 1.** Schematic illustration of the pre-pilot bubbling fluidized bed gasifier.

The BFBG is a 10 cm internal diameter cylindrical column, made of AISI 316L and electrically heated by five shell furnaces. Each furnace is controlled by a data acquisition system connected to five thermocouples, located in the reactor internal wall, which allow for independently setting the temperature of each reactor section (plenum, bed, and freeboard).

The air utilized as the fluidizing agent was injected at the bed bottom through a distributor plate composed of three nozzles. These have a truncate pyramidal shape and were specifically designed in order to ensure a homogeneous distribution of the fluidizing gas in the bed cross-section. The total

height is 2.5 m from the distributor plate to the product gas outlet. The feedstock was over-bed fed, continuously, by means of a screw-feeder device. A nitrogen flow of 0.32 kg/h was used to help the fuel feeding and to avoid the back flow of the hot gas. At the syngas outlet a high-efficiency cyclone allows the collection of elutriated fines. Downstream of this are two alternative symmetric gas conditioning lines, each one is composed by a bubbler filled with water and by a cartridge filter, which provide for tar, residual fly ash, and acid and basic gases removal. The gas coming from the producer gas treatment section is then sent to a stack.

## 2.2. Operating Conditions

The pre-pilot scale BFBG was fed with the five different fuels by keeping fixed the type and size range of the bed material (silica sand, 0.2–0.4 mm), the gasifying agent (air), the fluidized bed velocity (0.4 m/s) and the equivalence ratio (0.25), in order to obtain information about the role of fuel composition on the gasification process performance. Table 1 lists the values chosen for the complete set of experiments.

**Table 1.** Operating conditions of experimental runs.

Fuel	Bed Material	$U_g$ (m/s)	$W_{Air}$ (kg/h)	$W_{Fuel}$ (kg/h)	$A/F$ (kg <sub>Air</sub> /kg <sub>Fuel</sub> )	$ER$
RP	Silica sand	0.42	3.42	1.08	3.17	0.24
WRP	Silica sand	0.42	3.42	1.14	2.99	0.25
WRPC	Silica sand	0.42	3.43	1.56	2.20	0.25
WC	Silica sand	0.41	3.44	2.09	1.65	0.25
WD	Silica sand	0.42	3.42	2.46	1.39	0.25

With the purpose of obtain reliable data to perform accurate material and energy balances, sampling procedures of producer gas, elutriated fines, and tar were activated when the values of gas composition, temperature, and pressure were at steady state conditions and last for not less than 1 h.

## 2.3. Analytical Equipment

The main product gas compounds (CO<sub>2</sub>, CO, H<sub>2</sub>, CH<sub>4</sub>, C<sub>2</sub>H<sub>2</sub>, C<sub>2</sub>H<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>, C<sub>3</sub>H<sub>6</sub>, C<sub>3</sub>H<sub>8</sub>, and N<sub>2</sub>) were measured by using an Agilent 3000 micro gas chromatograph (micro-GC, Santa Clara, CA, USA) located downstream of the tar sampling line. In addition, an ABB AO2020 (for total hydrocarbons) (Zurich, Switzerland) and two HORIBA VA-3000 (for CO, CO<sub>2</sub>, and O<sub>2</sub>) (Kyoto, Japan) on-line analyzers were used as a check of accuracy of the micro-GC measurements. This double system allows a high reliability of measured gas composition.

The producer gas was further sampled by means of tedlar bags for off-line analyses in two other points along the reactor height (0.9 and 1.8 m). The flow rate of producer gas was determined by the tie component method applied to the value of nitrogen content in the dry gas, as obtained by on-line GC measurements.

Elutriated fines, consisting of char and fragmented bed particles, collected by the cyclone and particulate filter, were analyzed in a LECO TruSpec Elemental Analyzer (St. Joseph, MI, USA) in order to evaluate the content of carbon, hydrogen, nitrogen, and sulfur. For the sampling of

condensable species, a system consisted of four in-series cooling coils plugged into an ice bath; a suction pump and a flow meter were installed and operated with a gas flow rate of 2 NL/min for 1 h to obtain tar and water phase. The condensate is then washed from the coils using dichloromethane as solvent and collected in dedicated glass bottles. After a few minutes the condensate stratifies, forming two distinct solutions: water and hydrocarbons and dichloromethane. Water was separated from tar by means of a graduated syringe in order to evaluate its content in the producer gas. After water separation, the condensed hydrocarbons were analyzed off-line in a Perkin-Elmer Clarus 500 gas chromatograph (Waltham, MA, USA) coupled with a mass spectrometer (GC-MS). Elutriated fines and tar flow rate were obtained by dividing the total mass collected to the sampling time.

Hydrogen chloride, hydrogen sulfide, and ammonia were collected by bubbling the product gas through a pair of gas bubblers; each of them containing a solution of 50 mL of NaOH (0.5 M) and 50 mL of HCl (0.5 M). Subsequently, these solutions were analyzed by means of a Dionex DX-120 ion chromatograph (Sunnyvale, CA, USA).

Data obtained from on-line and off-line gas measurements and from chemical analyses of solid and condensed samples were processed to develop mass balance on atomic species and the related energy balance for each gasification tests.

#### 2.4. Feedstock

Gasification tests were carried out using five different materials including plastic waste, wood and coal. Two fuels were selected for the mono-gasification tests: recycled plastic (RP), a mixture of several plastic wastes obtained from the separate collection of post-consumer packaging materials and natural wood (WD), generally utilized to prepare fuel for domestic heating. Co-gasification tests were conducted utilizing three mixtures as fuels: the blend named WRP, composed of plastic waste and virgin wood; the fuel indicated as WRPC, obtained by blending recycled polyethylene, virgin wood, and brown coal and, finally, the fuel specified as WC, a mixture of virgin wood and brown coal. An overview of the employed fuels is given in Table 2.

**Table 2.** Composition and main physical properties of the tested fuels.

Items	RP	WRP	WRPC	WC	WD
Plastic waste, % <sub>wb</sub>	100	80	30	–	–
Wood, % <sub>wb</sub>	–	20	20	40	100
Coal, % <sub>wb</sub>	–	–	50	60	–
Size (diameter and length), mm	Irregular	6, 20	6, 20	6, 20	6, 20
Bulk density, kg/m <sup>3</sup>	590	580	615	620	570

The RP fuel has an irregular-spheroidal shape with a particle diameter of about 15–20 mm, while the other ones have a cylindrical shape, with a diameter of 6 mm and a length of about 15 mm. Plastic waste was selected as fuel because it saves the use of natural resources, for its large availability, and for its high calorific value. Woody biomass was chosen for the experimental tests since it offers credits resulting from the utilization of a renewable zero-emission energy resource. Lastly, coal utilization provides the benefits of wide fuel availability. Design and operation of thermochemical conversion systems need fuel composition as well as its chemical energy. In this context two types of

compositions are used: proximate and ultimate analyses. The proximate analysis provides moisture, volatile matter, fixed carbon, and ash content, while the ultimate analysis gives the fuel composition in terms of its basic elements such as carbon, hydrogen, nitrogen, sulfur, and oxygen. In this work the proximate analysis was performed as follows: the gross fuel sample was heated in air to 105 °C for 12 h to obtain moisture content, to 950 °C in inert ambient (nitrogen) for 5 h to obtain volatile matter, and to 750 °C in air for 2 h to obtain ash amount. Finally, the carbon-rich residue (fixed carbon) that remains after drying and devolatilization was calculated by subtracting the percentage of moisture, volatile matter, and ash from 100%. The ultimate analysis was carried out processing the fuel sample in the LECO TruSpec CHN/S Analyzer. Results of proximate and ultimate analyses of the tested fuels are listed in Table 3.

**Table 3.** Proximate and elemental analyses of the tested fuels.

Items	RP	WRP	WRPC	WC	WD
<b>Proximate analysis, %<sub>wb, ar</sub></b>					
Volatile matter	94.50	92.64	68.92	59.53	86.74
Fixed carbon	2.89	3.42	19.14	20.82	3.12
Moisture	0.67	2.11	4.11	9.42	9.93
Ash	1.94	1.83	7.83	10.23	0.21
<b>Ultimate analysis, %<sub>wb, ar</sub></b>					
C	79.54	73.20	62.28	51.93	45.31
H	13.06	11.15	8.11	5.44	5.59
N	0.18	0.30	0.19	0.22	0.26
S	0.08	0.10	0.13	0.16	<0.01
O (by difference)	4.53	11.31	17.35	22.60	38.70
<b>Heating value, MJ/kg<sub>Fuel</sub></b>					
HHV <sup>a</sup> <sub>db</sub>	42.69	37.45	29.35	22.00	18.40
LHV <sup>b</sup> <sub>ar</sub>	36.95	32.57	25.73	19.41	15.73

wb = weight basis; db = dry basis; ar = as received; <sup>a</sup> Evaluated by means of relationship proposed by Channiwala and Parikh [25]; <sup>b</sup> Evaluated by the HHV on dry basis by taking into account the latent heats of vaporization of the fuel moisture and the water obtained as product of hydrogen combustion.

### 3. Results and Discussion

#### 3.1. Producer Gas

The chemical characteristics of the fuels selected for the experimental tests differ considerably. Plastic waste contains a larger fraction of carbon and hydrogen than woody biomass that, conversely, contains more oxygen. The chemical energy of the fuels is greatly affected by the oxygen and water contents: the higher their values, the lower the chemical energy content. This agrees with the results reported in Table 3, which shows a decreasing of the lower heating value (*LHV*) when oxygen and moisture amounts in the feedstock increase. Plastic waste gives more volatile matter while the mixtures containing coal show the lower volatile content and a larger amount of fixed carbon. These marked differences in the fuels composition may present significant variations in the gasification products.

Gas measurements results from the micro-GC, that provides reading every 200 s, were used for all gasification tests calculations. The data acquisition system provides measurement data of temperature and pressure obtained from the gasifier and cleaning section in intervals of 1 s. These values were recorded and averaged over the entire sampling period. Figures 2 and 3 show gas composition, temperature, and pressure data averaged every ten minutes during the gasification test performed by feeding woody biomass (WD).

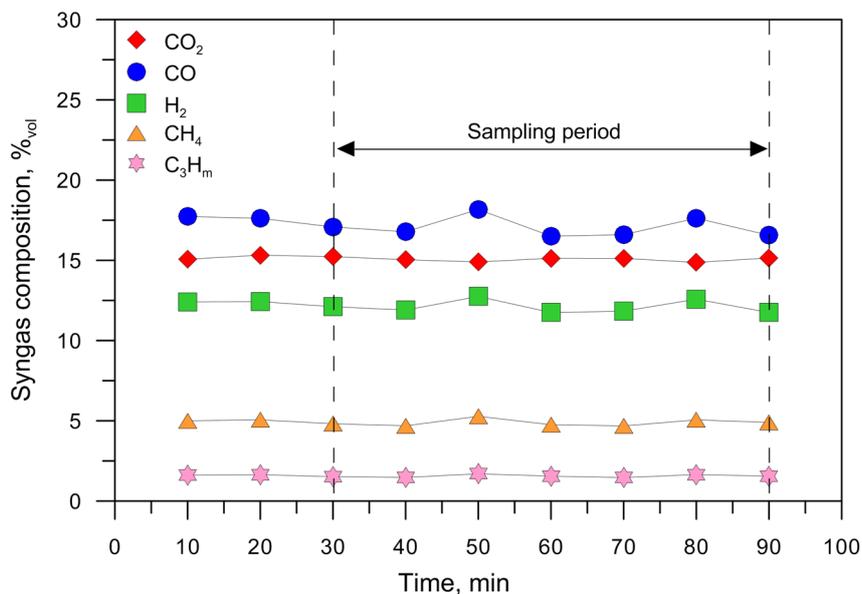


Figure 2. Producer gas composition profile obtained during wood gasification test.

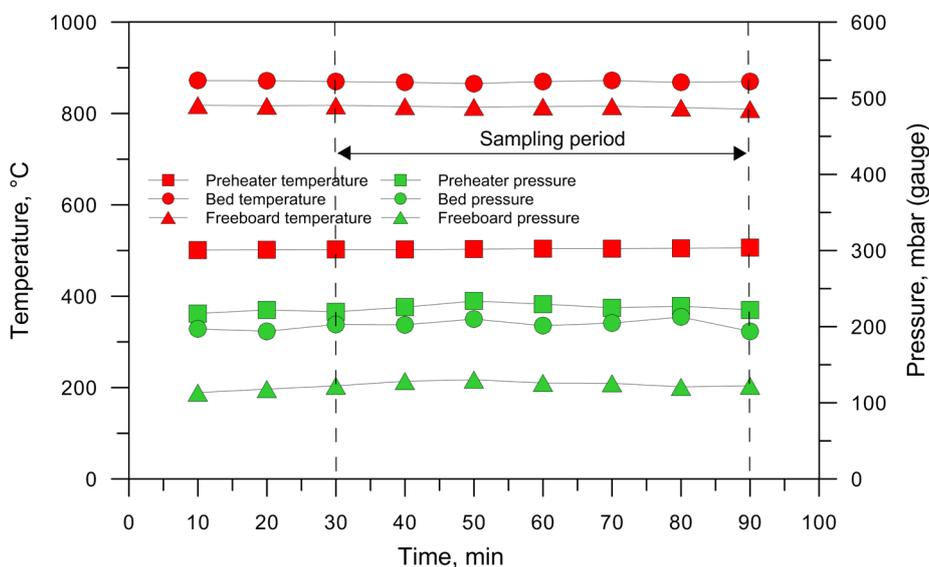
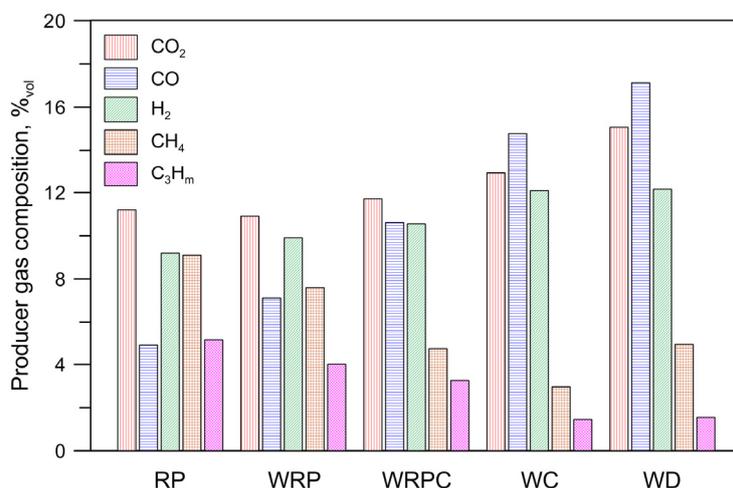


Figure 3. Temperature and pressure profiles obtained during wood gasification test.

Fuel composition strongly affects the characteristics of the producer gas obtained during the gasification tests. The results reported in Figure 4 show that the fuels containing plastic waste yield less hydrogen, carbon monoxide, and carbon dioxide and more methane and light hydrocarbons (sum of ethane, ethylene, acetylene, propane and propylene, C<sub>3</sub>H<sub>m</sub>) than the fuels including woody biomass. The considerable increase of carbon monoxide (from 4.9% to 17.1%) and carbon dioxide

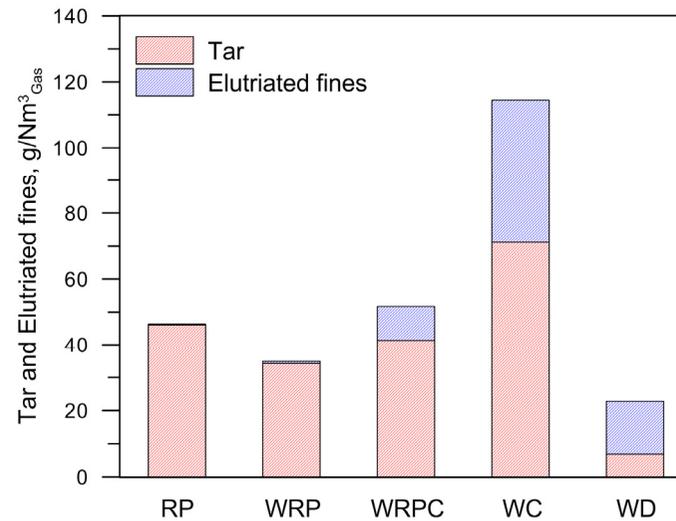
(from 11.2% to 15.1%) content in the product gas, as the wood fraction increases from 0% to 100%, can be attributed to the remarkable amount of oxygen and/or C–O groups in the biomass. On the other hand, the increase of plastic waste fraction, mainly made of polyethylene and polypropylene (consisting of long monomeric chains of  $C_2H_4$  and  $C_3H_6$ , respectively), promotes the production of methane and light hydrocarbons (that increase from 4.9% to 9.1% and from 1.6% to 5.1%, respectively), which lead to a reduction in hydrogen content (from 12.2% to 9.2%) in the producer gas.



**Figure 4.** Concentration of the main components of the dry product gas.

The above presented results are in agreement with the data obtained in other gasification studies using plastics and woody biomass as fuels. Pinto *et al.* [26] carried out experimental runs by means of a steam bubbling fluidized bed gasifier fed with mixtures of plastic and pine wood. They observed a decrease in the production of carbon monoxide as the fraction of plastic in the mixture increases. Wilk and Hofbauer [18] obtained similar results in a dual fluidized bed gasifier. In addition, the authors also reported an increase of methane and light hydrocarbon concentrations as the fuel plastic fraction increases from 0% to 100%.

The gasification tests carried out by utilizing mixtures containing coal further highlight the predominant role of plastic waste and woody biomass on the producer gas composition. They confirm that the production of hydrogen, carbon monoxide, and carbon dioxide can be attributed to wood degradation, whereas methane and light hydrocarbon generation can be assigned to plastic waste decomposition. Gasification tests carried out with WRPC, which contains 50% of coal, 30% of plastic, and 20% of wood, yields lower concentrations of hydrogen and carbon monoxide, and higher fractions of methane and light hydrocarbons than WC, which contains 60% of coal and a larger fraction of wood (40%). These results indicate that coal addition to the blends does not affect the trends shown by the main producer gas constituents obtained during the tests without coal. A possible explanation is that both lower coal reactivity and reduced volatile matter content, than that of plastic and wood, lead to a higher production of heavy tar and char [21,22]. As a consequence, coal addition slightly contributes to the change in the gas composition and to the increasing gas yield. This hypothesis is confirmed by the results displayed in Figure 5, which reports that the gasification tests with the blends containing coal (WRPC and WC) show a larger production of these by-products.



**Figure 5.** Elutriated fines and tar produced during the gasification tests.

**Table 4.** Main results obtained during the experimental runs.

Fuel	$T$ °C	$\text{CO}_2$ %vol	$\text{CO}$ %vol	$\text{H}_2$ %vol	$\text{CH}_4$ %vol	$\text{C}_3\text{H}_m$ %vol	$Q_{PG}$ Nm <sup>3</sup> /h	$C_{EF}$ g/Nm <sup>3</sup>	$C_{Tar}$ g/Nm <sup>3</sup>	$LHV_{PG}$ MJ/Nm <sup>3</sup>	$SE$ MJ/kg <sub>Fuel</sub>	$HRE$	$CCE$	$CGE$
RP	877	11.20	4.90	9.20	9.10	5.14	3.79	0.29	46.06	7.90	27.77	$0.91 \pm 0.03$	$0.84 \pm 0.11$	$0.75 \pm 0.04$
WRP	872	10.90	7.12	9.91	7.60	4.01	3.86	0.67	34.24	7.06	23.84	$0.90 \pm 0.19$	$0.83 \pm 0.22$	$0.73 \pm 0.15$
WRPC	868	11.71	10.62	10.55	4.73	3.25	4.20	10.31	41.38	6.09	16.40	$0.79 \pm 0.23$	$0.78 \pm 0.17$	$0.64 \pm 0.18$
WC	862	12.91	14.77	12.09	2.96	1.45	4.22	43.26	71.20	5.09	10.29	$0.70 \pm 0.22$	$0.70 \pm 0.14$	$0.53 \pm 0.16$
WD	870	15.07	17.13	12.16	4.93	1.55	4.80	15.94	6.86	6.15	12.03	$0.79 \pm 0.10$	$0.93 \pm 0.10$	$0.76 \pm 0.10$

PG = producer gas; EF = elutriated fines;  $\pm$  =  $\pm$ standard deviation.

As previously reported, the features of the feedstock affect the producer gas composition and, as a consequence, its *LHV*. The results reported in Table 4 show that the *LHV* trend is consistent to that obtained for methane and light hydrocarbons due to their high calorific value. Since plastic gasification generated a large amount of hydrocarbons, the *LHV* progressively increases up to 7.9 MJ/Nm<sup>3</sup> (gasification test with RP fuel) when more plastic waste is used in the fuel mixtures. The mono-gasification test with wood (WD) shows intermediate *LHV* value (6.2 MJ/Nm<sup>3</sup>) that is halfway between those obtained in the tests with the fuels containing plastic (RP and WRP) and those that include coal (WRPC and WC).

### 3.2. Mass Balance

As already mentioned, data obtained from on-line gas measurements and from chemical analyses of the elutriated fines and tar, produced during steady operation of the gasification plant, were processed to develop mass and energy balances for each gasification test. In particular, in this section, the acquired data were elaborated to perform overall mass and elemental balances. Mass inputs involve fuel, dry air, and nitrogen purge. The industrial area that houses the gasifier is equipped with an air dryer device; therefore, dry air was utilized as the gasifying agent during the experimental runs. Mass outputs include dry gas, reaction water, elutriated fines, and tar.

For accurate mass and energy balance calculations, it is also necessary to determine the composition of the elutriated fines and tar. The composition of elutriated fines sampled during the gasification tests was determined utilizing a LECO TruSpec Elemental Analyzer, while the tar elemental composition was evaluated from the molecular tar composition as obtained from the GC-MS analyzer (Table 5).

**Table 5.** Elutriated fines composition produced during the gasification tests.

Fuels	Ultimate Analysis, % <sub>wb</sub>									
	C		H		N		O		S	
	EF	Tar	EF	Tar	EF	Tar	EF	Tar	EF	Tar
RP	42.80	93.25	1.80	6.17	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
WRP	48.10	93.34	2.00	6.00	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
WRPC	39.70	94.13	0.90	5.85	0.50	<0.01	<0.01	0.01	0.16	<0.01
WC	49.70	93.49	0.80	6.08	0.71	<0.01	<0.01	0.02	0.59	<0.01
WD	65.45	93.82	1.10	6.15	0.39	<0.01	<0.01	0.02	<0.01	<0.01

Table 6 reports mass and elemental balances performed for the five gasification tests. The results indicate that the overall mass balances show values ranging between 96.1% and 99.6%. The non-closure of the mass balances is due to the sum of the systematic errors that affect the measurement instrumentations and those due to the effectiveness of the sampling procedures. Assuming that these errors were regularly repeated during all the experimental runs and that the overall maximum error is quite low (less than 4%), it is possible to carry out the following observations.

**Table 6.** Mass balance performed during the gasification tests.

Item	Input, kg/h					Item	Output, kg/h				
	Mass	C	H	O	N		Mass	C	H	O	N
Fuel, RP	1.08	0.86	0.14	0.05	<0.01	Dry gas	4.45	0.72	0.13	0.74	2.86
Air	3.42	–	–	0.80	2.62	Water	0.01	–	<0.01	0.01	–
Nitrogen purge	0.32	–	–	–	0.32	Elutriated fines	<0.01	<0.01	<0.01	–	<0.01
–	–	–	–	–	–	Tar	0.17	0.16	0.01	0.00	<0.01
<i>Total</i>	<i>4.82</i>	<i>0.86</i>	<i>0.14</i>	<i>0.85</i>	<i>2.95</i>	<i>Total</i>	<i>4.64</i>	<i>0.86</i>	<i>0.14</i>	<i>0.75</i>	<i>2.86</i>
<b>Mass balance, %</b>	<b>96.28</b>					–	–	–	–	–	–
Fuel, WRP	1.14	0.84	0.13	0.13	<0.01	Dry gas	4.52	0.70	0.12	0.80	2.92
Air	3.42	–	–	0.80	2.62	Water	0.03	–	<0.01	0.03	–
Nitrogen purge	0.32	–	–	–	0.32	Elutriated fines	<0.01	<0.01	<0.01	–	<0.01
–	–	–	–	–	–	Tar	0.13	0.12	0.01	0.00	0.00
<i>Total</i>	<i>4.88</i>	<i>0.84</i>	<i>0.13</i>	<i>0.93</i>	<i>2.95</i>	<i>Total</i>	<i>4.69</i>	<i>0.82</i>	<i>0.13</i>	<i>0.83</i>	<i>2.92</i>
<b>Mass balance, %</b>	<b>96.08</b>					–	–	–	–	–	–
Fuel, WRPC	1.56	0.97	0.13	0.27	<0.01	Dry gas	4.98	0.75	0.10	1.02	3.10
Air	3.43	–	–	0.80	2.63	Water	0.10	–	0.01	0.09	–
Nitrogen purge	0.32	–	–	–	0.32	Elutriated fines	0.04	0.02	<0.01	–	<0.01
–	–	–	–	–	–	Tar	0.17	0.16	0.01	<0.01	0.00
<i>Total</i>	<i>5.31</i>	<i>0.97</i>	<i>0.13</i>	<i>1.07</i>	<i>2.95</i>	<i>Total</i>	<i>5.30</i>	<i>0.94</i>	<i>0.12</i>	<i>1.11</i>	<i>3.10</i>
<b>Mass balance, %</b>	<b>99.75</b>					–	–	–	–	–	–
Fuel, WC	2.09	1.08	0.11	0.47	<0.01	Dry gas	5.00	0.76	0.08	1.22	2.94
Air	3.44	–	–	0.80	2.64	Water	0.13	–	0.01	0.11	–
Nitrogen purge	0.32	–	–	–	0.32	Elutriated fines	0.18	0.09	<0.01	–	<0.01
–	–	–	–	–	–	Tar	0.30	0.28	0.02	<0.01	<0.01
<i>Total</i>	<i>5.85</i>	<i>1.08</i>	<i>0.11</i>	<i>1.27</i>	<i>2.96</i>	<i>Total</i>	<i>5.62</i>	<i>1.13</i>	<i>0.11</i>	<i>1.33</i>	<i>2.94</i>
<b>Mass balance, %</b>	<b>96.06</b>					–	–	–	–	–	–
Fuel, WD	2.46	1.11	0.14	0.95	0.01	Dry gas	5.71	1.03	0.11	1.62	2.95
Air	3.42	–	–	0.80	2.62	Water	0.21	–	0.02	0.19	–
Nitrogen purge	0.32	–	–	–	0.32	Elutriated fines	0.08	0.05	<0.01	–	<0.01
–	–	–	–	–	–	Tar	0.03	0.03	<0.01	<0.01	<0.01
<i>Total</i>	<i>6.20</i>	<i>1.11</i>	<i>0.14</i>	<i>1.75</i>	<i>2.95</i>	<i>Total</i>	<i>6.03</i>	<i>1.11</i>	<i>0.13</i>	<i>1.81</i>	<i>2.95</i>
<b>Mass balance, %</b>	<b>97.33</b>					–	–	–	–	–	–

Plastic addition to the fuel mixture leads to an increase in gas specific yield (*GSY*). The results reported in Table 6 show that the *GSY* increases from about 2360 g/kg<sub>Fuel</sub>, with the fuels without plastic (WC and WD), to values of 3185.4, 3959.8, and 4126.8 g/kg<sub>Fuel</sub>, as the plastic content increases from 30% to 100% (WRPC, WRP and RP, respectively). Similar results were obtained in a previous study [27] and by other researchers. Ahmed *et al.* [28] conducted co-gasification experiments feeding polyethylene and woodchip mixtures in a semi-batch reactor, using steam as the gasifying agent. The authors observed a progressive increase in gas yield as the amount of plastic rises up to 80%.

The experimental runs conducted by feeding the fuels containing large amount of plastic (RP and WRP) yield small specific quantities of elutriated fines (1.0 and 2.3 g/kg<sub>Fuel</sub>, respectively), while the fuels containing coal (WRPC and WC) generate huge amounts of fines (27.8, and 87.5 g/kg<sub>Fuel</sub>, respectively). This could be due to the large quantity of fixed carbon contained in these fuels that

produce more char, which is then carried out from the BFBG in the form of carbonaceous fines. This phenomenon in the test with WRPC appears partially suppressed. This evidence could be explained considering the structure of the WRPC pellets: plastic, wood and coal were crumbled and intimately blended before being pelletized. When the WRPC pellets drop down in the reactor, the plastic contained in this fuel melts and binds the particles of coal and wood, protracting the permanence of these aggregates in the most reactive zone of the reactor (bed and splashing zone), favoring their conversion into gaseous products.

The mono-gasification test of plastic waste (RP) produces the highest specific amount of tar, 161.9 g/kg<sub>Fuel</sub>, while woody biomass (WD) gasification generates only 13.4 g/kg<sub>Fuel</sub>. The co-gasification tests yield similar (high) amounts of tar, *i.e.*, 115.7, 111.4 and 144.1 g/kg<sub>Fuel</sub> in the runs with WRP, WRPC, and WC, respectively.

Water content in the producer gas, as previously reported, was determined by applying the condensation method. In the runs with RP and WC fuels this procedure failed; thus, water content was estimated by applying the atomic balance method on the hydrogen element. Table 6 shows that woody biomass promotes the generation of reaction water. In particular, water content increases from 13.4 to 85.5 g/kg<sub>Fuel</sub> as the wood fraction in the fuel rises from 0% to 100%. Probably, water generation from the fuels containing a higher biomass fraction is due to both physical and chemical mechanisms: by evaporation of the larger fuel moisture content (Table 3) and by oxidation of hydroxyl and carboxyl groups widely present in lignocellulosic materials.

Data obtained from fuels characterization and from on-line GC measurements were utilized to evaluate the hydrogen recovery efficiency (*HRE*) and the carbon conversion efficiency (*CCE*). In this work *HRE* and *CCE* were calculated according to Equations (1) and (2):

$$HRE = \frac{m_{H,H_2} + m_{H,CH_4} + m_{H,C_3H_m}}{m_{H,Fuel}} \quad (1)$$

$$CCE = \frac{m_{C,CO_2} + m_{C,CO} + m_{C,CH_4} + m_{C,C_3H_m}}{m_{C,Fuel}} \quad (2)$$

where  $m_{H,i}$  and  $m_{C,i}$  are hydrogen and carbon mass flow rates of the constituent  $i$ , respectively.

The fuels containing high plastic fractions transfer more hydrogen in the producer gas than the blends without plastic. In particular, Table 4 shows that the *HRE* values obtained in the runs with RP, WRPC and WRP are 0.91, 0.90 and 0.79; while those found in the runs with WC and WD are 0.70 and 0.79, respectively. On the other hand, the gasification test conducted by feeding wood (WD) exhibits higher a *CCE* value (0.93). Furthermore, coal seems to have a negative effect on *CCE*. In fact, the gasification tests with the fuels WRPC and WC show the lowest *CCE* values (0.78 and 0.70, respectively). The low conversion efficiency in terms of *HRE* and *CCE* recorded in tests carried out with the blend WC is a direct consequence of carbon and hydrogen losses determined by the considerable production of elutriated fines and tar (Figure 5 and Table 6).

### 3.3. Energy Balance

Data obtained from material balances were utilized to perform energy balances for the gasification tests carried out by feeding all the five tested fuels. Energy input to the gasifier considered the

chemical enthalpy ( $CE$ ) of the fuel, while energy outputs took into account the  $CE$ s of the producer gas, elutriated fines, and tar. The  $CE$  was calculated as indicated in the Equation (3):

$$CE_i = m_i \times LHV_i \quad (3)$$

where  $m_i$  and  $LHV_i$  are volume or mass flow rate and lower heating value of the constituent  $i$ , respectively.

The  $LHV$ s of the fuels, elutriated fines, and tar were estimated from the  $HHV$  by taking into account the latent heats of vaporization of the fuel moisture and the water obtained as product of hydrogen combustion. The  $HHV$  was evaluated using the correlation, with a wide range of applicability, proposed by Channiwala and Parikh [25]:

$$HHV = 349.1C + 1178.3H + 100.5S - 103.4O - 15.1N - 21.1Ash \quad (4)$$

where  $C$ ,  $H$ ,  $S$ ,  $O$ ,  $N$ , and  $Ash$  are percentages of carbon, hydrogen, sulfur, oxygen, nitrogen, and ash as determined by ultimate analysis on a dry basis.

Table 7 reports the chemical enthalpy balances associated to input and output streams. As it shows, energy balance closure ranges from 78.4% to 88.5%.

**Table 7.** Energy balance performed during the gasification tests.

Item	Input, MJ/kg <sub>Fuel</sub>					Item	Output, MJ/kg <sub>Fuel</sub>				
	RP	WRP	WRPC	WC	WD		RP	WRP	WRPC	WC	WD
Fuel	36.95	32.57	25.73	19.41	15.73	Dry syngas	27.77	23.84	16.40	10.29	12.03
Air	–	–	–	–	–	Water	–	–	–	–	–
Nitrogen purge	–	–	–	–	–	Elutriated fines	0.02	0.04	0.38	1.49	0.72
–	–	–	–	–	–	Tar	4.92	3.52	3.39	4.38	0.41
<i>Total</i>	<i>36.95</i>	<i>32.57</i>	<i>25.73</i>	<i>19.41</i>	<i>15.73</i>	<i>Total</i>	<i>32.71</i>	<i>27.40</i>	<i>20.17</i>	<i>16.15</i>	<i>13.15</i>
<b>Energy balance, %</b>	<b>88.51</b>	<b>84.11</b>	<b>78.40</b>	<b>83.25</b>	<b>83.60</b>	–	–	–	–	–	–

During the gasification of plastic a large amount of light hydrocarbons are generated, resulting in a producer gas with high calorific value. As a consequence, the synthetic gas produced from the experimental runs carried out feeding the fuels containing plastic waste (RP, WRP and WRPC) exhibit higher specific energy ( $SE$ ) values (27.8, 23.8 and 16.4 MJ/kg<sub>Fuel</sub>, respectively), than those obtained with the fuels without plastic (WC and WD; 10.3 and 12.0 MJ/kg<sub>Fuel</sub>, respectively). These results are in agreement with those obtained during co-gasification studies utilizing fluidized bed reactors [18,26,29]. These authors observed that the addition of plastic waste to the fuel mixture generate a gas with higher hydrocarbon content, which results in a producer gas with high  $LHV$  and, thus, larger  $SE$  values.

The energy loss due to tar formation increases from 0.4 MJ/kg<sub>Fuel</sub>, for the test with woody biomass (WD), to 4.9 MJ/kg<sub>Fuel</sub>, for the test carried out by feeding plastic waste (RP). On the contrary, woody biomass gasification presents a greater energy loss, determined by a large production of carbonaceous fines (0.7 MJ/kg<sub>Fuel</sub>) than that observed during the gasification of plastic waste (<0.1 MJ/kg<sub>Fuel</sub>).

A remarkable amount of energy loss during the gasification tests is due to the energy needed to transform the fuel into gasification products; here, this energy is indicated as gasification loss ( $GL$ ) and evaluated according to Equation (5):

$$GL = 1 - \frac{CE_{PG} + CE_{EF} + CE_{Tar}}{CE_{Fuel}} \quad (5)$$

where  $CE_{PG}$  is the producer gas chemical enthalpy,  $CE_{EF}$  is the elutriated fines chemical enthalpy,  $CE_{Tar}$  is the tar chemical enthalpy, and  $CE_{Fuel}$  is the fuel chemical enthalpy.

Mono-gasification tests with RP and WD display  $GL$  values of 11.5% and 16.4%, respectively. These results indicate that plastic degradation requires lower amount of energy to be converted in the gasification products. This could be a consequence of the peculiar thermal behavior of plastic as a result of its different structure and composition. This experimental evidence is supported by the results obtained from the co-gasification tests with WRP and WRPC, which show increasing values of  $GL$  as the plastic fraction decreases from 80% to 30% (15.9% and 21.6%, respectively).

The chemical energy of the fuel transferred to the product gas, also known as cold gas efficiency ( $CGE$ ), was evaluated as indicated in Equation (6):

$$CGE = \frac{Q_{PG} \times LHV_{PG}}{W_{Fuel} \times LHV_{Fuel}} \quad (6)$$

where  $Q_{PG}$  is the volumetric flow rate of the producer gas,  $W_{Fuel}$  is the mass flow rate of the fuel,  $LHV_{PG}$  is the lower heating value of the producer gas and  $LHV_{Fuel}$  is the lower heating value of the fuel.

In accordance with the results presented above, the product gas generated during the mono-gasification tests of RP and WD shows higher values of  $CGE$  (75.2% and 76.5%, respectively), whereas the gas obtained from the co-gasification tests of WRP, WRPC, and WC, characterized by a greater production of by-products and by high  $GL$  values, yields lower  $CGEs$  (73.2%, 63.7% and 53.0%, respectively) as can be deduced from Table 7 (and reported in Table 4).

#### 4. Conclusions

The gasification process performance was evaluated by performing mass and energy balances on the BFB reactor fed with plastic waste, woody biomass, and their blends with coal. The results indicated that plastic waste induces the production of light hydrocarbons, which led to a reduction in hydrogen content in the producer gas. Additionally, woody biomass promotes the generation of  $CO_2$ ,  $CO$ , and  $H_2$  and depresses the production of light hydrocarbons. Coal addition to the blends does not seem to affect the trends shown by the main producer gas constituents obtained during the tests without coal. The experimental runs conducted by feeding the fuels containing large amount of plastic (RP and WRP) yield small specific quantities of elutriated fines and the highest amount of tar, similar to those generated with the fuels containing coal. On the contrary, the mono-gasification test of woody biomass (WD) generates the lowest amount of tar. The fuels containing high plastic fractions transfer more hydrogen in the producer gas than the blends without plastic, while the gasification test conducted by feeding wood (WD) exhibits higher  $CCE$  value.

The mono-gasification tests (RP and WD) show higher  $CGE$  values, whereas the co-gasification tests (WRP, WRPC and WC), which are characterized by a greater energy loss due to high  $GL$  values and by a large production of char and tar, inevitably transfer less fuel energy in the producer gas.

The results reported in this work indicate that the air gasification process of the tested fuels is technically feasible even if the co-gasification tests show the worst performance. These results suggest that the co-gasification process requires further research efforts with the purpose of reducing the production of char and tar in order to obtain higher conversion efficiencies in terms of  $HRE$ ,  $CCE$ , and  $CGE$ .

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## Author Contributions

Both authors contributed to publishing this paper. Lucio Zaccariello has contributed significantly to the conception, design, execution, analysis and interpretation of the data; participate in drafting, reviewing, and revising the manuscript for intellectual content; and approve the manuscript to be published. Maria Laura Mastellone, head of the project, has contributed to the analysis, interpretation and evaluation of the data; and approve the manuscript to be published.

## Conflicts of Interest

The authors declare no conflict of interest.

## Nomenclature

BFBG	bubbling fluidized bed gasifier
<i>CCE</i>	carbon conversion efficiency
<i>CE</i>	chemical enthalpy
<i>CGE</i>	cold gas efficiency
<i>EF</i>	chemical enthalpy
GC-MS	gas chromatograph-mass spectrometer
<i>GL</i>	gasification loss
<i>GSY</i>	gas specific yield
<i>HHV</i>	higher heating value
<i>HRE</i>	hydrogen recovery efficiency
$\text{kg}_{\text{Fuel}}$	kilogram of fuel on wet basis
<i>LHV</i>	lower heating value
$m_{C,i}$	carbon mass flow rate of the constituent <i>i</i>
$m_{H,i}$	hydrogen mass flow rate of the constituent <i>i</i>
$m_i$	volume or mass flow rate of the constituent <i>i</i>
micro-GC	micro gas chromatograph
$\text{Nm}^3$	cubic meters under normal conditions (0 °C and 1 atm)
PG	producer gas
$Q_i$	volumetric flow rate of the producer gas
RP	recycled plastic
SE	specific energy
$W_i$	mass flow rate of the fuel
WD	virgin wood
WC	mixture of virgin wood and brown coal
WRP	mixture of recycled plastic and virgin wood
WRPC	mixture of recycled polyethylene, virgin wood and brown coal

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