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Gasification of Solid Recovered Fuels with Variable Fractions of Polymeric Materials [†]

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Abstract: Gasification is a promising thermochemical technology used to convert waste materials into energy with the introduction of low amounts of an oxidant agent, therefore producing an environmental impact that is lower when compared to incineration and landfilling. Moreover, gasification allows a sustainable management of wastes and reduces the use of fossil fuels responsible for the increment of greenhouse gases. This work aimed to perform gasification tests with solid recovered fuels (SRF) containing organic fractions mainly retrieved from construction and demolition wastes to assess the potential for energy conversion. Tests were conducted in a pilot-scale downdraft gasifier (maximum feedstock input of 22 kg/h) at c.a. 800 °C, using SRF samples containing different proportions of polymeric wastes ranging between 0 and 20 wt %. Gas and chars obtained as by-products were analysed to evaluate their properties and to establish valid pathways for their valorisation. The addition of polymeric wastes reduced char production but rose both tar and HCl concentrations in the gas. The SRF with 10 wt % of polymeric wastes generated the best results, producing the highest calorific value for the gas (3.5 MJ/Nm^3) and the highest cold-gas efficiency (45%). Possible char applications include their use as catalysts for tar decomposition, or as an additive in construction materials. Gasification can therefore be considered a valid solution for the energetic valorisation of these SRFs.

Keywords: gasification; solid recovered fuel; construction and demolition waste; polymeric waste

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1. Introduction

The production and management of construction and demolition wastes (C&DW) have established challenges in the last years due to the high amounts generated and to the need of defining convenient solutions for their treatment and valorisation. During the period of 2008–2018, the generation of C&DW inside the European Union (EU) increased 7% and achieved a final value of 924 × 10^6 t. The situation in Portugal also showed an increasing trend but with a higher growth rate (26%) that contributed to reach a final amount of 1.71 × 10^6 t [1].

On the other hand, overall, society has evolved with increasing technology developments, therefore having higher energy demands [2]. The production of electricity in Portugal increased 29% between 2008 and 2017 to a final value of 57.7 TWh [1]. Around

59% of this production was obtained from fossil fuels (59%), but these present several drawbacks such as the geographic and politic dependencies, exhaustion of feedstocks in a near future, instability of prices, and aggravation of greenhouse gas (GHG) emissions to the atmosphere [3]. These problems raised the pressure to find out other sources and technologies for energy production [2]. As a result, recent European policies were created specifically to promote a low carbon economy, which contributed to reduce GHG emissions by 16% between 2008 and 2018 across the EU [1].

Many typologies of wastes present in their composition a relevant amount of organic compounds, enabling them to have calorific properties that can be exploited for energy purposes. C&DW incorporates a considerable fraction of inert ceramic materials such as bricks, concrete and asphalt (65–72 wt %), incoherent fine materials (20–26 wt %), metals (4–5 wt %), and a small parcel composed of organic wastes (3–5 wt %) that include mainly wood, paper, plastics, and rubber [4]. Despite the low percentage, this organic parcel still represents a significant amount of waste in the total values of C&DW reported previously for the EU and Portugal (37×10^6 t and 68×10^3 t in average, respectively). If this parcel is separated and processed to obtain an adequate solid recovered fuel (SRF), the calorific value may achieve 18 MJ/kg wb, presenting a good potential for energy recovery [5].

The main final treatments attributed to C&DW in Portugal were the recovery/recycling of materials (44.1 wt %), landfilling (4.5 wt %) and, in much smaller extensions, the valorisation through energy production (0.03 wt %) and incineration (0.02 wt %) [6]. Landfilling of C&DW still occurred at some extent possibly because the material fractions composing them were not adequately separated at the source of production, or no sufficient action plans were put in practice to perform this primary separation aiming to facilitate posterior recycling treatments. Considering these data, the production of energy from C&DW represents a small parcel when compared with landfilling, which is a treatment alternative that poses environment issues such as the contamination of natural resources, the need for large areas to be implemented, loss of material resources that could be recovered, and high GHG emissions [7]. In fact, recent European legislation has discouraged landfilling practices and has promoted other valorisation strategies to convert wastes into valuable products (directive 2008/98/EC). Thus, new ways to valorise wastes into useful energy have to be established in order to contribute for a more sustainable environment and to fulfil the requirements of legislation in force.

Gasification may be a promising option for the treatment of organic fractions present in C&DW. Here, feedstocks are thermally converted into a combustible gas rich in H_2 and CO under a partially reducing atmosphere, typically with the injection of low amounts of air (<30% of the amount required for complete combustion). Temperatures inside the reactor may reach 700–900 °C and, instead of air, the oxidant agent may be replaced by pure oxygen or steam that promotes the increase in the lower heating value (LHV) of the gas; this property is commonly in the range of 4–16 MJ/Nm^3 [7,8]. The gas contains a series of contaminants that incorporate small particles, condensable tars, sulphur and nitrogen gases (H_2S, SO_x, NH_3, NO_x, …), and also acid gases (HCl, HF) that must be removed to achieve acceptable levels before being used in final applications [9,10]. Then, the gas can be burnt in an internal combustion engine (ICE) or in a boiler–turbine system to produce heat and electric energy, or it even can be converted into other liquid biofuels through catalytic processes [2,8].

Gasification also originates chars and tars as by-products that can be used in different applications for valorisation. Depending on the respective composition and physical properties, chars can be used as adsorbents of contaminants in effluents, fertilisers for agriculture practices, and as catalysts in the gasification process itself; by its turn, tars can be reformed to obtain other valuable chemical products such as naphthalene [11].

Several feedstocks have been used in gasification such as coal, biomass (wood, forest, and agricultural residues), organic wastes (sewage sludge and animal wastes), and municipal wastes [7,8,12–14]. However, the presence of plastic fractions may cause problems during the process, as is the case of tar production in excess and chlorine gases that are responsible for obstructions and corrosion of equipment [15,16]. Since C&DW may contain plastic compounds, similar gasification problems may occur, and therefore, optimal conditions and appropriate cleaning systems must be established to improve the overall efficiency and lifetime of equipment.

Another thermochemical approach that has been investigated with organic fractions from C&DW is pyrolysis, which consists in a thermal conversion of materials in the absence of oxygen. Different materials such as waste wood, plastics, textiles and rubber (eventually mixed with other municipal solid waste (MSW) fractions) were processed at temperatures and residence times in the ranges of 400–900 °C and 30–60 min, respectively, and using different pyrolysis reactor configurations (e.g., rotary kiln, fixed-bed, fluidised-bed, and batch). Depending on the feedstock considered and on the operational conditions, interesting results have been obtained after the tests: biochars with high carbon contents (70–80%), gases with high concentrations of H_2 (30–40 vol %), and high oil yields (about 50–55% at 500 °C). In addition, the use of HZSM-5 catalysts during pyrolysis favours the formation of phenolic and aromatic compounds in the oil fraction, which helps the upgrading step aiming to improve oil quality [17–19]. Compared to pyrolysis, gasification offers the advantage of producing high amounts of a valuable gas that presents a greater flexibility in terms of final applications (e.g., production of energy and gas/liquid fuels); moreover, gasification can achieve a thermal self-sustainability status, therefore disregarding the use of external heat sources (only required at the start-up stage).

Some works were found in the scientific literature focused on the gasification of C&DW or its individual fractions. Nuss et al. [20] developed a life cycle analysis for a plasma gasification facility aiming to process mixtures of wood from C&DW, forest biomass, and coal. They concluded that the environmental burden was similar to a conventional combustion plant due to the use of coal in gasification, and that energy conversion was lower as a consequence of high heat losses in the reactor and use of a steam turbine; however, the landfill area occupied by other by-products generated was lower. Hwang et al. [21] performed a steam gasification of wood from C&DW and other residues, and they concluded that the process is viable for a temperature of 900 °C, with the disadvantage of incrementing the generation of polycyclic aromatic hydrocarbons. In addition, a clean-up system for tar removal (e.g., catalysers) was required, and the increment of the plastic fraction in feedstocks promoted a greater production of tars. The work of Willeboer [22] was based on a project for a fluidised-bed gasifier processing wood from C&DW, aiming to replace a part of coal that was originally admitted. For a feedstock input of 150,000 t/year, the project reduces CO_2 emissions in 170,000 t/year and electricity can be generated with an efficiency of 35%. Littlejohns et al. [23] performed pilot gasification tests in a downdraft reactor using wood fractions from C&DW and other lignocellulosic feedstocks, and found that materials with low densities and high angles of repose caused process instabilities (in particular, bridge formation inside the reactor); in addition, tar concentrations in the gas ranged between 1 and 9 mg/m^3, and it rose with system instability and moisture content in the feedstock. Lotfi et al. [24] developed a downdraft gasifier model which predicted that gas LHV as well as CO and H_2 concentrations assume the highest values when using wood from C&DW containing 5% of moisture (5.4 MJ/m^3, 20% and 30%, respectively); in parallel, the ratio between the lengths of drying + pyrolysis zone and combustion + reduction zone must be greater than two to achieve optimal results for gas yield and gas quality. Peres et al. [25] tested different wood-based feedstocks from C&DW in a bench-scale gasifier, and they obtained a syngas with a LHV between 8 and 11 MJ/m^3 at temperatures of 700–900 °C. When the gas was injected in an engine-generator for electricity production, the maximum amount of energy produced was equivalent to 13 GJ/t, which demonstrated the suitability of the gas for electricity production. Beyond the fact of being scarce, the existing literature does not explore conveniently the effect of varying the composition of mixtures of different C&DW organic fractions in a downdraft gasification process for evaluating the quality of the gas produced and the performance of the overall process.

The present work aimed at performing gasification tests in a downdraft reactor using SRFs containing different proportions of lignocellulosic and polymeric wastes, which were derived mainly from C&DW. The influence of varying these proportions was evaluated in the properties of the produced gas and in the global efficiency of the process as a way to determine the viability of SRFs for energy conversion. In addition, chars generated as by-products were analysed to establish feasible valorisation pathways.

2. Materials and Methods

2.1. Raw Materials

Nine different residues provided by a Portuguese waste management company were used and mixed in various proportions to perform the gasification tests. These residues were provided separately and included organic components obtained mostly from C&DW and also some from MSW but in smaller proportions. Only organic components were selected due to their potential energetic content (soils and other inert materials were therefore excluded). These waste components were classified in two categories, according to the type of material: lignocellulosic and polymeric. Some of the waste components included both categories; for these cases, it was admitted that they belong to the polymeric category for simplification once this was the most predominant material type. Table 1 lists all the individual residues to prepare the SRFs for gasification and their classification along with the annual mass fractions received during the year 2017.

Table 1. Residues used in the gasification experiments.

Code	Description	ELW * Code	Category	Annual Mass Fraction (wt % wb)
R1	Wood from construction and demolition wastes (C&DW)	17 02 01	Lignocellulosic	65.9 (R1 + R2)
R2	Wood from municipal solid wastes (MSW)	20 01 38	Lignocellulosic	
R3	Paper/card (MSW)	20 01 01	Lignocellulosic	9.2
R4	Plastic packages	15 01 02	Polymeric	0.7
R5	Composite packages	15 01 05	Polymeric	4.9
R6	Mixture of packages	15 01 06	Polymeric	4.2
R7	Plastics (C&DW)	17 02 03	Polymeric	9.9
R8	Plastics (MSW)	20 01 39	Polymeric	2.8
R9	Polymeric insulations (C&DW)	17 06 04	Polymeric	2.4

* European List of Waste, as defined by the European Commission's Decision 2014/955/UE.

To comply with the gasification requirements, all residues were milled, cut and sieved to obtain fractions with dimensions between 10 and 40 mm; in addition, inert materials that occasionally were found (stones, metals, and glass) were manually separated. Three waste mixtures were then prepared to obtain solid recovered fuels (SRF) with distinct proportions of lignocellulosic (L) and polymeric (P) residues, identified by SRF1 (100 wt % L), SRF2 (90 wt % L + 10 wt % P) and SRF3 (80 wt % L + 20 wt % P). A final homogenisation step was performed to all SRF samples before the tests. These SRF samples were mixed based on the proportions defined for each category (L between 80 and 100 wt %, and P between 0 and 20 wt %), and simultaneously on the annual amount of individual residues R1–R9 received by the Portuguese waste management company during 2017. The choice of these criteria for preparing the SRFs was due to practical reasons related with a possible construction of a gasification plant in the company's area. Moreover, the variation assumed for the polymeric fraction (0–20 wt %) may eventually simulate seasonal oscillations on total waste composition received by the company. Considering these observations, the mass fraction of each individual residue R_i (i = 1 to 9) inside each waste category (lignocellulosic or polymeric) that composes the mixture SRF1, SRF2 or SRF3 (X_{Ri}, in wt % wb) was calculated through Equation (1).

$$X_{Ri} = X_{Ri\ annual} \times \frac{X_{wc}}{100} \tag{1}$$

where $X_{Ri\,annual}$ is the mass fraction of residue R_i (i = 1 to 9) in the total annual waste amount received by the company during the year 2017 (in wt % wb), and X_{wc} is the mass fraction of the waste category to which R_i belongs (lignocellulosic (80, 90 or 100 wt % wb) and polymeric (20, 10 or 0 wt % wb)). Values for $X_{Ri\,annual}$ are presented in Table 1.

The composition and illustrations of the three SRFs are presented in Table 2 and Figure 1, respectively.

Table 2. Composition of the elaborated solid recovered fuels (SRFs) for gasification tests.

Mixture	General Composition	Fraction of Residues (wt % wb *)
SRF1	100 wt % lignocellulosic	R1 + R2 **: 87.7 R3: 12.3
SRF2	90 wt % lignocellulosic + 10 wt % polymeric	R1 + R2: 78.8 R3: 11.1 R4: 0.3 R5: 2.0 R6: 1.7 R7: 4.0 R8: 1.1 R9: 1.0
SRF3	80 wt % lignocellulosic + 20 wt % polymeric	R1 + R2: 70.1 R3: 9.8 R4: 0.6 R5: 3.9 R6: 3.4 R7: 8.0 R8: 2.3 R9: 1.9

* Wet basis. ** The residue R1 + R2 resulted from a combination of both R1 and R2, and it was originally provided by the company in this form.

Figure 1. Illustrations of the prepared mixtures: (**a**) SRF1, (**b**) SRF2 and (**c**) SRF3.

2.2. Gasification Experiments

The gasification tests were conducted in a small downdraft gasifier unit (All Power Labs, model PP20) with a maximum throughput capacity of 22 kg/h. The unit is composed by seven modules: (i) a feedstock feeder, (ii) a drier, (iii) an Imbert downdraft reactor, (iv) a char collector system, (v) a gas cleaning system, (vi) a stack and (vii) a control board. Figure 2 illustrates the equipment and the block diagram of the gasification unit as well as the interconnection among the various modules.

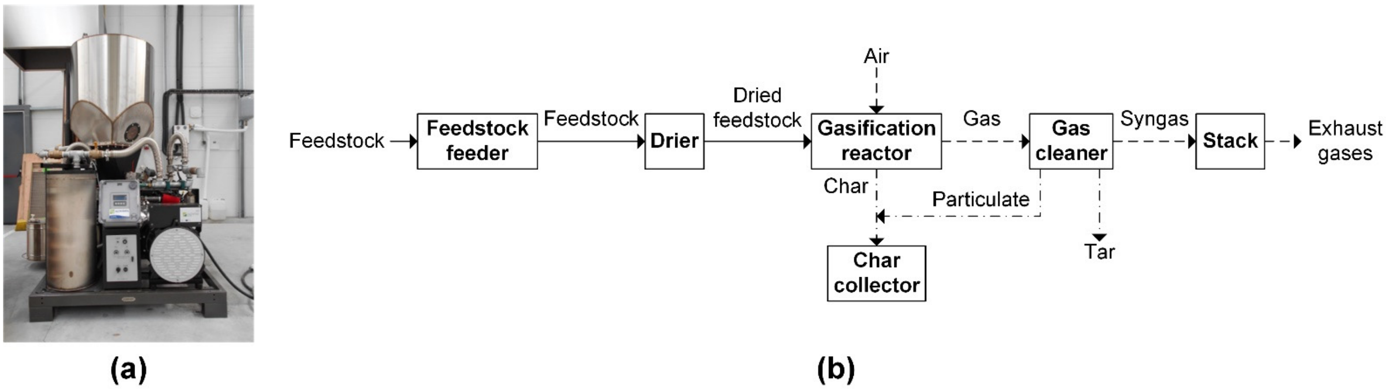

Figure 2. Downdraft gasifier used in the experiments: (**a**) image of the equipment; (**b**) block diagram.

The feedstock is initially introduced in a hopper and then it falls to the feeder, which consists in an auger that displaces the material to the drier and later to the reactor. The auger moves depending on the amount of material present in the reactor, and is automatically commanded by the control board.

Meanwhile, a flux of air is pre-heated through a recirculation around the walls of the downdraft reactor, and then, it is injected inside it. The air flux was measured with an anemometer (Amprobe TMA10A), and it can be controlled manually in the control board. This control allows one to adjust both the equivalence ratio (ER) and the temperature of the reactor.

A sequence of gasification reactions takes place inside the reactor to convert the feedstock in product gas and chars with the temperature in the oxidation zone maintained at c.a. 800 °C. Chars produced at the bottom of the reactor are collected automatically by the char collection system, moving them to an auxiliary deposit. The gas generated at the base of the reactor is conducted to the gas cleaning system, composed of a cyclone to separate particulates, and also by a filter filled with wood biomass to retain tars from the gas. To minimise the decomposition of this biomass filter, the hot gas exiting the reactor passes first through the drier to transfer some of its heat to the feedstock, and the latter is conducted to the filter at a colder temperature.

After the cleaning stage, the gas is forwarded to the stack to be burnt. The movement of gases along all the gasification unit is performed by two vacuum pumps located before the stack.

When the reactor temperature (oxidation zone) was stabilised at 800 °C and a flame was produced in the stack, product gas samples were collected at the reactor's exit using a laboratory vacuum pump (KNF model N 022 AN.18) and were stored in Tedlar bags of 3–5 L for further analysis. The experiments lasted until the flame in the burner was extinguished (no more feedstock to be gasified) or when the temperature at the oxidation zone started to increase significantly; this situation indicates that the feedstock was completely converted to ashes inside the reactor, and therefore, there was no more feedstock to be gasified. Long-run experiments in industrial downdraft reactors processing the SRFs created may lead to possible corrosion problems, especially when the fraction of polymeric materials was increased. This is explained by the presence of chlorine in some polymeric fractions, in particular R7 and R9. Moreover, obstruction problems and slag formation may occur inside the reactor after a while, depending on the mineral composition of feedstocks that tend to vary in highly heterogeneous wastes, such as those presented in C&DW and MSW.

The quantification of HCl present in the product gas was performed by capturing it in an assembly composed by two impinger bottles containing a solution of NaOH 1 M, which is in accordance with the study of Gai and Dong [26]. The measurement was conducted for c.a. 30 min and the volumetric flow of the gas passing through the assembly was periodically recorded.

After the reactor was cooled, chars and tars were collected for quantification and complementary analysis. The total number of gasification experiments was three (one for each SRF, without repetitions).

2.3. Calculation of Gasification Performance Indicators

To assess the efficiency of gasification experiments, seven performance indicators were determined: gas yield (Y_{gas}, m^3/kg feedstock), feedstock flow ($\dot{m}_{waste}$, kg/s), char and tar yield (both in kg/kg feedstock), ER, cold-gas efficiency (*CGE*, %), and carbon conversion efficiency (*CCE*, %).

The gas yield (Y_{gas}) was estimated by using Equation (2), which was derived from the volume conservation of N_2 that enters and exits the gasifier (i.e., the volumetric flow of N_2 present in the air and in the gas, respectively). Nitrogen contents present in the feedstocks were not considered in the balance due to their negligible concentrations, as can be confirmed latter through the ultimate analysis.

$$Y_{gas} = \frac{0.781 \times \dot{V}_{air}}{x_{N2} \times \dot{m}_{waste}} \tag{2}$$

$\dot{V}_{air}$ is the volumetric flow of the intake air (m^3/s), x_{N2} is the volumetric proportion of N_2 in the gas, and $\dot{m}_{waste}$ is the mass flow of waste (kg/s). The coefficient 0.781 represents the volumetric fraction of N_2 present in the air.

The ER was defined as the ratio between the real amount of air introduced in the gasifier and the stoichiometric amount needed for the complete combustion of the feedstock. The stoichiometric amount of air was estimated based on the quantity of O_2 required to oxidise completely the organic elements N, C, H and S present in the feedstock (for reference, please see Table 4 in Section 3.1).

The *CGE* was calculated using Equation (3) [27].

$$CGE = \frac{LHV_{gas} \times \dot{m}_{gas}}{LHV_{waste} \times \dot{m}_{waste}} \tag{3}$$

LHV_{gas} and LHV_{waste} are the LHVs of the gas and the feedstock mixture (both in MJ/kg), and $\dot{m}_{gas}$ and $\dot{m}_{waste}$ are the mass flows of gas produced and waste admitted, respectively (kg/s).

The *CCE* was determined according to Equation (4) [7].

$$CCE = \frac{x_{C\ gas} \times \dot{m}_{gas}}{x_{C\ waste} \times \dot{m}_{waste}} \tag{4}$$

$x_{C\ gas}$ and $x_{C\ waste}$ are the mass fractions of carbon contained in the gas and in the feedstock, respectively, while $\dot{m}_{gas}$ and $\dot{m}_{waste}$ are the mass flows of gas and of feedstock (kg/s).

2.4. Analysis Procedures for Feedstocks and Gasification Products

The nine individual residues R1–R9 were examined separately to assess their chemical and fuel properties. All residues were milled and sieved to obtain particles with dimensions of 12 mm and 425 µm, and some of these treated fractions were subsequently dried at 105 °C. The following is a list of all analysis that were performed:

- Proximate analysis (with moisture, volatile matter and ash contents determined by ASTM E949-88, ASTM E897-88 and ASTM E830-87, respectively; fixed carbon was calculated by difference). The final results for each parameter were determined through the average of two experimental replicates;
- Ultimate analysis (the contents of N, C, H and S were quantified in a ThermoFisher Scientific Flash 2000 CHNS-O analyser; the concentrations of O were calculated by difference);
- Higher heating value—HHV (using a calorimetric bomb IKA C200);

- Chlorine concentration (using an X-ray fluorescent analyser Thermo Scientific Niton XL 3T GOLDD+). Two readings of chlorine concentrations were performed to obtain the final results through the average of experimental values;
- Mineral composition of ashes (using the same procedure as for chlorine concentration). Again, this composition was obtained through the average of two readings, using the analyser;
- Apparent density (by following EN 15103). Final results were determined through the average of two replicates.

In addition, the SRF mixtures that were prepared (SRF1, SRF2 and SRF3) were analysed through the previous methods, with the exception of ultimate analysis and chlorine concentration. Instead, these two latter analyses were determined by calculation of the weighted average of the elements present in the individual residues (R1 + R2 to R9). The reason for this choice was due to the high heterogeneity of materials and to the difficulty of obtaining small particle sizes required for equipment analysis.

The composition of product gas samples was determined in a gas chromatography (GC) equipment (Varian 450-GC), which was equipped with two thermal conductivity detectors (TCD) and two capillary columns (Ultimetal 1.5 × 0.002 m and 1 × 0.002 m). The major gaseous components N_2, H_2, CO, CO_2 and CH_4, as well as other minor hydrocarbons (C_2H_x) were all quantified. GC calibration was performed with two standard gases (Linde) containing known compositions of H_2 (5 vol % and 10 vol %), CO (20 vol % and 15 vol %), CO_2 (10 vol % and 20 vol %), CH_4 (15 vol % and 3 vol %), C_2H_2 (2 vol % and 1 vol %), C_2H_4 (2 vol % and 1 vol %), C_2H_6 (2 vol % and 1 vol %), H_2S (2 vol % and 1 vol %), and N_2 (42 vol % and 48 vol %). The product gas samples from Tedlar bags were injected into the equipment through a peristaltic pump, while the carrier gases used were N_2 (to detect H_2) and He (to detect the remaining gases), both with a volumetric flow of 20 mL/min. The TCD detector was set to 175 °C. The temperature program for the oven that was used to detect component gases was the following: (i) constant temperature of 50 °C during 7.4 min; (ii) increase in temperature at a constant rate (+7.4 °C/min), until a final value of 90 °C was achieved; (iii) constant temperature of 90 °C for 3.5 min. By their turn, the lower heating value (LHV) and gas density were calculated based on the proportions of the different gaseous components, which were present in the product gas samples. Tar concentrations were estimated through the ratio of the total tar amount collected after the experiments and the total volume of gas produced.

To determine the concentration of HCl present in the gas, chloride ions captured by the NaOH solution were measured by a potentiometric method similar to the one described in USEPA 9212, using an ion selective electrode Hanna HI 4007 connected to a potentiometer Hanna HI 98185. Using these chloride concentrations, capture durations, and the volumetric flows of gas in the impinger bottles, an estimation of HCl concentrations in gas samples were therefore possible to be obtained. Equation (5) was applied to predict these concentrations (“$C_{HCl\ gas}$”, in mg/Nm^3).

$$C_{HCl\ syngas} = 1.028 \times \frac{V_{solution} \times C_{Cl^-}}{\dot{V}_{gas} \times t} \tag{5}$$

$V_{solution}$ is the total volume of solution contained in the impinger bottles (m^3), $C_{Cl}{}^-$ is the mass concentration of chloride ions detected in solution (mg/L), $\dot{V}_{gas}$ is the average volumetric flow of product gas that passed through the HCl capturing assembly (Nm^3/s), and t is the duration of the capture test (s). Final results were calculated considering the average of two determinations of chloride concentrations present in solution.

Chars produced during the tests were characterised to quantify the following properties: moisture and ash contents, HHV, chlorine concentration, and mineral composition of ashes. All these parameters were determined by following the same procedures as for the raw residues, which were previously described.

3. Results and Discussion

3.1. Characterisation of Raw Residues and Solid Recovered Fuels

Table 3 presents the characterisation of the individual residues R1 through R9, which were used to prepare the SRF mixtures.

Table 3. Properties of raw individual residues used to prepare SRFs.

Property		**Residue**							
		R1 + R2	**R3**	**R4**	**R5**	**R6**	**R7**	**R8**	**R9**
Proximate analysis (wt %) *	Moisture	12.5 ± 0.1	6.2 ± 0.2	0.6 ± 0.0	6.1 ± 0.0	4.3 ± 0.1	0.4 ± 0.1	0.7 ± 0.2	1.0 ± 0.1
	Volatile matter	85.7 ± 0.1	82.0 ± 1.5	98.0 ± 0.1	94.0 ± 0.6	85.7 ± 0.4	81.8 ± 0.5	98.1 ± 0.1	93.6 ± 0.2
	Fixed carbon	6.3 ± 0.7	0.0 ± 1.5	0.0 ± 0.2	0.0 ± 0.8	5.2 ± 0.5	7.3 ± 0.9	0.4 ± 0.2	4.5 ± 0.3
	Ash	8.0 ± 0.6	18.0 ± 0.0	2.0 ± 0.1	6.0 ± 0.2	9.1 ± 0.1	10.9 ± 0.4	1.5 ± 0.1	1.9 ± 0.1
Ultimate analysis (wt % daf **)	C	52.3 ± 0.0	47.2 ± 0.0	85.6 ± 0.0	42.8 ± 0.0	46.0 ± 0.0	57.1 ± 0.0	73.6 ± 0.0	72.3 ± 0.0
	H	6.3 ± 0.0	6.4 ± 0.0	14.3 ± 0.0	5.8 ± 0.0	6.5 ± 0.0	7.4 ± 0.0	13.1 ± 0.0	7.5 ± 0.0
	N	1.7 ± 0.0	0.4 ± 0.0	0.0 ± 0.0	0.2 ± 0.0	0.0 ± 0.0	0.3 ± 0.0	0.8 ± 0.0	3.4 ± 0.0
	S	0.1 ± 0.0	0.1 ± 0.0	0.0 ± 0.0	0.1 ± 0.0	0.0 ± 0.0	0.1 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
	O ***	39.6 ± 0.0	45.9 ± 0.0	0.1 ± 0.0	51.1 ± 0.0	47.5 ± 0.0	35.1 ± 0.0	12.5 ± 0.0	16.8 ± 0.0
Ash composition (wt % db)	CaO	17.26 ± 1.30	42.19 ± 0.06	23.69 ± 1.39	54.66 ± 0.22	36.72 ± 0.00	22.66 ± 0.00	36.22 ± 2.07	17.61 ± 0.14
	SiO_2	14.58 ± 1.69	16.32 ± 0.02	17.97 ± 0.49	18.03 ± 0.36	16.21 ± 0.06	6.72 ± 0.24	18.48 ± 0.30	57.67 ± 0.80
	Al_2O_3	4.78 ± 0.45	8.24 ± 0.26	4.65 ± 1.38	8.37 ± 1.02	8.22 ± 0.25	0.00 ± 0.00	3.59 ± 0.60	2.49 ± 0.13
	Fe_2O_3	5.43 ± 1.46	1.37 ± 0.01	4.59 ± 0.23	2.69 ± 0.07	1.47 ± 0.06	1.39 ± 0.03	4.68 ± 0.27	6.11 ± 0.33
	K_2O	1.60 ± 0.27	0.11 ± 0.01	1.35 ± 0.07	0.64 ± 0.00	0.33 ± 0.00	0.08 ± 0.00	0.58 ± 0.04	0.52 ± 0.01
H/C mass ratio (daf)		0.12	0.13	0.17	0.13	0.14	0.13	0.18	0.10
O/C mass ratio (daf)		0.76	0.97	0.00	1.19	1.03	0.61	0.17	0.23
HHV (MJ/kg db)		18.5 ± 0.0	14.9 ± 0.1	44.9 ± 0.1	17.0 ± 0.1	26.5 ± 2.3	25.0 ± 0.5	38.2 ± 3.3	30.3 ± 0.1
Chlorine (wt % db)		0.2 ± 0.0	0.3 ± 0.0	0.1 ± 0.0	0.2 ± 0.0	0.1 ± 0.0	7.3 ± 0.0	0.1 ± 0.0	44.1 ± 0.3
Apparent density (kg/m^3 wb)		262 ± 0	37 ± 1	26 ± 0	25 ± 2	43 ± 3	543 ± 11	69 ± 17	19 ± 2

* Moisture content in wt % wet basis (wb); volatile matter, fixed carbon and ash contents in wt % dry basis (db); ** dry and ash free basis; *** calculated by difference.

HHVs were essentially higher for residues with plastic fractions (namely R4, R6, R7, R8 and R9), with results greater than 25 MJ/kg db. Therefore, these residues present a higher potential to be converted to energy in a gasification process, a fact that is essentially explained by the high amounts of volatile matter (>81 wt % db), lower oxygen contents (<48 wt % daf), and lower H/C and O/C mass ratios (average of 0.14 and 0.41, respectively). Generally, lower mass ratios of H/C and O/C are typically found in feedstocks with high energy densities, as is the case of fossil coals [28].

Moisture levels were adequate for a gasification process since they assumed values lower than 15 wt %, with a maximum result of 12.5 wt % wb for wood residues R1 + R2 [27]. A drying pre-treatment is therefore unnecessary and the associated energy requirements for this step can be saved.

CaO and SiO_2 were the most prevalent oxides in the ashes of all residues, with average concentrations of 31 wt % db and 21 wt % db, respectively. R5 had a significant proportion of CaO (55 wt % db), and R9 was especially rich in SiO_2 (58 wt % db); these results were probably justified by the aggregation of cement and sand contained in the inert fraction of C&DW. Moreover, the presence of paper and cardboard in R3, R5 and R6 increased the amounts of Ca and Al. K_2O and Fe_2O_3 also appeared with lower concentrations (<6 wt % db), the former with the highest result for the wood residue R1 + R2.

Taking into account that agglomerations and slagging issues may occur when using feedstocks with high concentrations of ash, K, Ca and Fe, residues R3 and R5 may be more problematic for a gasification process [29–31]. Thus, the proportions of these two residues must be reduced in the prepared SRFs. The lower fractions of residues R3 and R5 included in SRF1, SRF2 and SRF3 (up to 13.7 wt % wb) may help to mitigate this problem.

With the exception of R1 + R2 and R9, all residues contained lower amounts of N and S (both <1 wt % daf) that would promote lower emissions of pollutants such as SO_2, H_2S and NO_x, during gasification. When using R1 + R2 and R9, the ER must be adjusted to adequate levels to reduce the generation of oxide pollutants (e.g., SO_2 and NO_x). An alternative is to dilute those residues with others possessing lower concentrations of N and S.

Chlorine contents were generally lower for all samples ($\leq$0.3 wt % db), except for R7 and R9 (composed of plastics) that exhibited significant concentrations (>7 wt % db), which was possibly due to the existence of chlorinated compounds such as polyvinyl chloride. These residues may promote the accumulation of deposits and formation of HCl, which would degrade downstream equipment [32]. To minimise problems induced by chlorine-rich wastes in a gasification process, two possible solutions may be considered: (i) to dilute these wastes in a mixture containing materials with low chlorine concentrations, e.g., lignocellulosic wastes; (ii) to perform a pre-treatment of carbonisation with a continuous flux of an inert gas (nitrogen), which releases part of chlorine to the gas phase that can be further treated through decontamination techniques, e.g., lime slurry atomisation.

The majority of plastics and paper/card residues had lower apparent densities (<70 kg/m^3) which would increase their volume during transportation routes and storage operations, hence rising the corresponding costs. On the other hand, the wood residue R1 + R2 and the C&D plastic R7 may reduce such operational costs due to their greater apparent densities (>260 kg/m^3).

Another problem reported by the use of plastic materials in gasification is the formation of larger amounts of tars that are responsible for obstruction problems in equipment, such as engines and turbines for energy generation [15,33]. Although these materials presented good calorific properties, solutions must be implemented to minimise tar production.

A gasification process may be viable for all residues R1 + R2 to R9 if an adequate combination of them is performed instead of processing the individual residues. These waste combinations would compensate the disadvantages presented by each individual residue: on one hand, the plastic fractions would favour the calorific value of the global mixture, but on the other hand, the lignocellulosic fractions would dilute the chlorine present, produce low tar amounts, and increase in the apparent density of the global mixtures.

To evaluate the effect of lignocellulosic and polymeric proportions in the properties of final SRFs, Table 4 presents the characterisation of all SRFs prepared (SRF1, SRF2 and SRF3) and used in gasification experiments.

Table 4. Properties of SRFs used in gasification experiments.

Property		**Mixture**		
		SRF1	**SRF2**	**SRF3**
Proximate analysis (wt %) *	Moisture	11.9 ± 0.3	9.4 ± 1.1	6.0 ± 0.3
	Volatile matter	92.4 ± 0.7	93.9 ± 0.4	85.5 ± 0.8
	Fixed carbon	1.6 ± 0.8	0.0 ± 0.9	9.5 ± 1.2
	Ash	6.0 ± 0.1	6.1 ± 0.5	5.0 ± 0.4
Ultimate analysis (wt % daf)	C	51.7 ± 0.0	52.2 ± 0.0	52.7 ± 0.0
	H	6.3 ± 0.0	6.5 ± 0.0	6.6 ± 0.0
	N	1.5 ± 0.0	1.4 ± 0.0	1.3 ± 0.0
	S	0.1 ± 0.0	0.1 ± 0.0	0.1 ± 0.0
	O **	40.4 ± 0.0	39.8 ± 0.0	39.3 ± 0.0
Ash composition (wt % db)	CaO	34.7 ± 1.09	41.5 ± 0.13	45.0 ± 0.88
	SiO_2	25.0 ± 0.51	24.7 ± 0.66	23.1 ± 0.00
	Al_2O_3	13.9 ± 0.32	18.7 ± 0.96	21.3 ± 3.00
	Fe_2O_3	6.6 ± 0.63	4.2 ± 0.17	4.9 ± 0.00
	K_2O	2.5 ± 0.11	1.2 ± 0.00	1.1 ± 0.02
H/C mass ratio		0.12	0.12	0.13
O/C mass ratio		0.78	0.76	0.75
Higher heating value (HHV, MJ/kg db)		18.5 ± 0.0	21.0 ± 0.0	24.7 ± 0.0
Chlorine (wt % db)		0.2 ± 0.0	0.9 ± 0.1	1.6 ± 0.1
Apparent density (kg/m^3 wb)		152 ± 5	117 ± 2	93 ± 8

* Moisture content in wt % wb; volatile matter, fixed carbon and ash contents in wt % db; ** calculated by difference.

As expected, the increase in the polymeric fraction in SRFs contributed to rise carbon and hydrogen contents (+1.0 wt % db and +0.6 wt % db, respectively), and to decrease both the oxygen concentration (−1.1 wt % db) and apparent density (−38.8%). These observations were in line with the properties of the individual polymeric residues R4 to R9, as shown in Table 3. In addition, HHV increased from 18.5 to 24.7 MJ/kg db (+33.5%), which is a fact that can be explained by two main reasons: (i) the slight decrease in O/C mass ratio (−0.03); (ii) the high amount of fixed carbon and low ash content detected particularly in SRF3 (9.5 wt % db and 5.0 wt % db, respectively) [28,34]. Similar results for HHV were also obtained in other literature [35,36].

On the other hand, the chlorine content increased +1.4 wt % db due to the large fraction of residues R7 and R9 present in SRF2 and SRF3, which are rich in that element. The presence of high amounts of chlorine would contribute to accentuate the corrosiveness of such mixtures. Moreover, the concentration of CaO in ashes rose +10.3 wt % db, possibly encouraging ash fusibility at low temperatures and the formation of agglomerations during thermochemical conversion processes [30]. In light of these results, the introduction of polymeric materials in SRFs had benefits in terms of increased energy density, but the tendency for corrosiveness of equipment and agglomerate formation may be appreciable. Therefore, the addition of polymeric wastes must be carefully controlled to obtain an adequate balance between process efficiency and maintenance costs in gasification plants.

3.2. Assessment of Gasification Performance

Table 5 shows the results obtained for gasification performance parameters that were reached in each experiment, while Figure 3 presents the temperature profile over each experiment period.

Table 5. Performance indicators of gasification tests.

Indicator		**Mixture**		
		SRF1	**SRF2**	**SRF3**
Average temperature (°C) *		780	783	805
Test duration (h)		1.5	2.3	2.8
SRF feed rate (kg/h)		6.6	4.1	3.1
Equivalence ratio (ER)		0.39	0.33	0.45
Gas yield (m^3/kg waste)		2.62	2.56	3.51
Char yield (g/kg waste)		61	46	28
Tar yield	g/kg waste	14	29	26
	g/m^3 gas	5.3	11.3	7.4
Cold-gas efficiency (*CGE*, %)		41.7	44.8	43.3
Carbon conversion efficiency (*CCE*, %)		67.1	64.2	84.4

* Measured at the oxidation zone of the downdraft reactor.

Despite the implementation of the best efforts to stabilise the gasification temperature at 800 °C, some difficulties were encountered once the average values oscillated between 780 and 805 °C. The high heterogeneity of materials present in SRFs in terms of composition and particle dimension may justify the occurrence of these stabilisation difficulties.

The addition of polymeric materials to the SRFs reduced their mass densities and, consequently, the waste flows also diminished from 6.6 to 3.1 kg/h. The gas yield diminished slightly from 2.62 m^3/kg in SRF1 to 2.56 m^3/kg in SRF2, and it rose significantly to 3.51 m^3/kg in the case of SRF3. The increase in gas yield can be explained by the greater volatilisation of the polymeric fractions found in SRF3 and by the reduction in char yield. The relationship between the increment of gas yield and polymeric concentration in the SRFs is corroborated by other literature [15,36].

The results for ERs when using SRF1 and SRF2 were acceptable, once they were in the range 0.2–0.4 that are typically obtained in traditional gasification operations reported in the literature. This range contributes for a better control of tar and char production [37]. However, the experiment with SRF3 generated a high ER (0.45) as a consequence of an excessive admission of air in the reactor. SRF2 exhibited the lowest ER among all experiments (0.33), therefore demonstrating that lower amounts of oxygen were needed to keep stable the gasification temperature at 800 °C. This may represent a reduction in energy for pumping and heating the air entering the reactor. Greater thermal savings can therefore be achieved when using SRF2.

*CGE*s assumed similar values in all experiments, with results in the range 41–45%. This parameter possessed the highest result for SRF2 (44.8%), which can be justified by the highest LHV obtained for the gas (please see ahead Section 3.3.1 regarding gas properties). This highest result was close to the values obtained by Park et al., who used fluff-type solid refuse fuels in their experiments [38]. The other SRFs were less efficient in terms of energy transfer from the feedstock to the gas once lower *CGE*s were obtained.

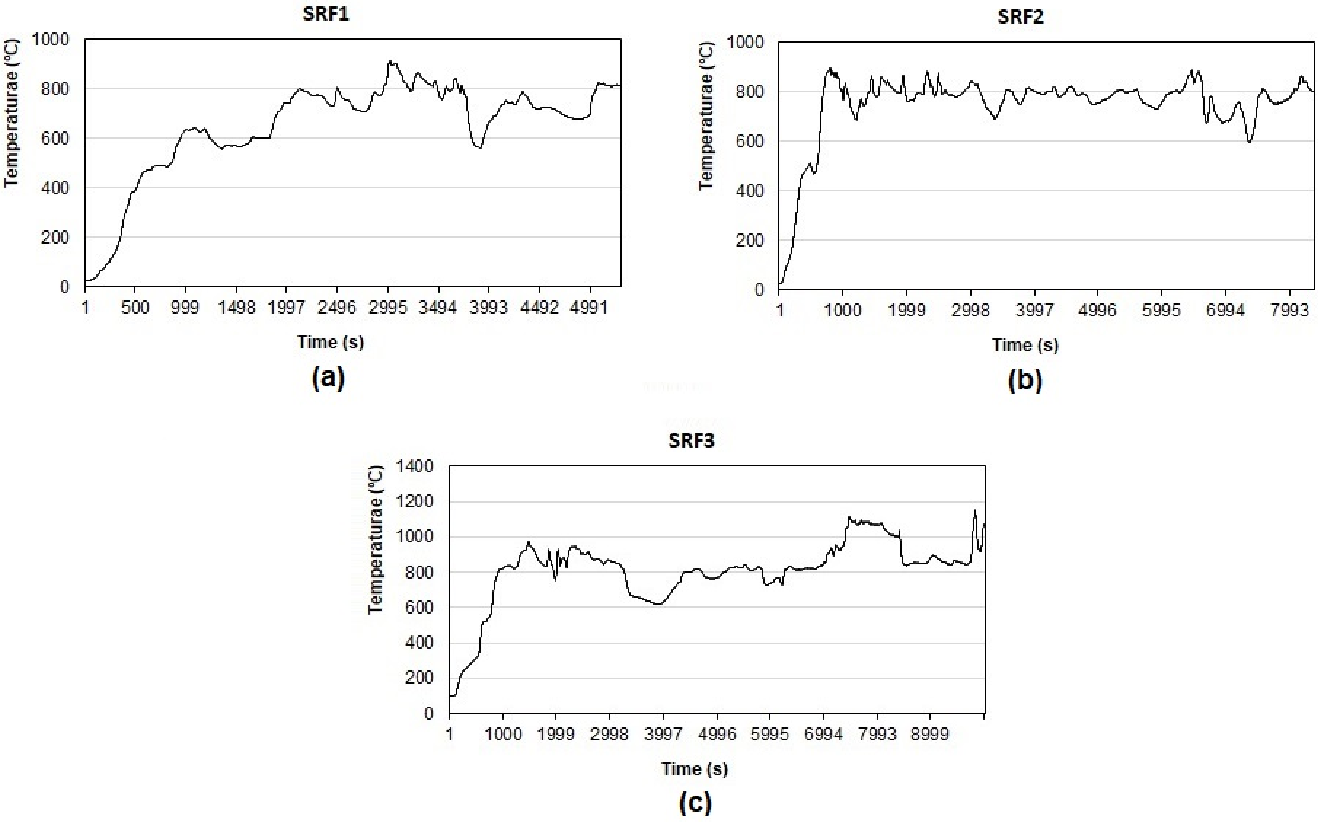

Figure 3. Temperature profiles over time for each gasification test using (**a**) SRF1, (**b**) SRF2 and (**c**) SRF3.

*CCE*s varied between 67 and 84%, which are typical values found in other literature [15,36]. This parameter assumed the highest result in the test with SRF3 (84%), and it is an indication that a significant portion of carbon present in the feedstock was converted into gases, and that other by-products (chars and tars) possibly exhibited a less carbonaceous nature. Conversely, the tests using SRF1 and SRF2 produced lower results (64–67%), showing that the migration of the carbon contained in the feedstock to the gas was not highly effective. All of these behaviours may be explained by the high ratio of air: feedstock mass flow observed for SRF3, which increased the mass of carbon transferred from the feedstock to the product gas. No linear correlation between *CGE* and *CCE* was detected.

All SRFs exhibited tar levels in the gas that were well above the limit defined for ICE's (0.1 g/m^3), thus requiring the implementation of mechanisms for their attenuation [9]. Although SRF2 generated the highest *CGE*, tar production was also the highest (11.3 g/m^3). The increase in polymeric fractions is known to increase the formation of tars; therefore, SRF1 presented the lowest amount (5.3 g/m^3), while SRF2 and SRF3 produced more tars (>7.4 g/m^3) [15]. Hence, the gasification of wastes containing more polymeric materials needs a fine control of process parameters and the use of gas cleaning methods to retain tars, aiming to minimise their production that induces issues such as obstruction and abrasion in the downstream equipment, in particular turbines and engines [33]. For example, the operation of the gasifier at high temperatures (>800 °C), high ERs (>0.2), and with the introduction of catalyst materials (dolomite, olivine, or Ni-based materials) and gas cleaning systems (catalytic filters and scrubbers) may be employed to attenuate the generation of tars in excess [33].

3.3. Analysis of Gasification Products

3.3.1. Gas Properties

The results for the composition and other relevant properties of gas samples obtained from the gasification experiments are summarised in Figure 4 and Table 6.

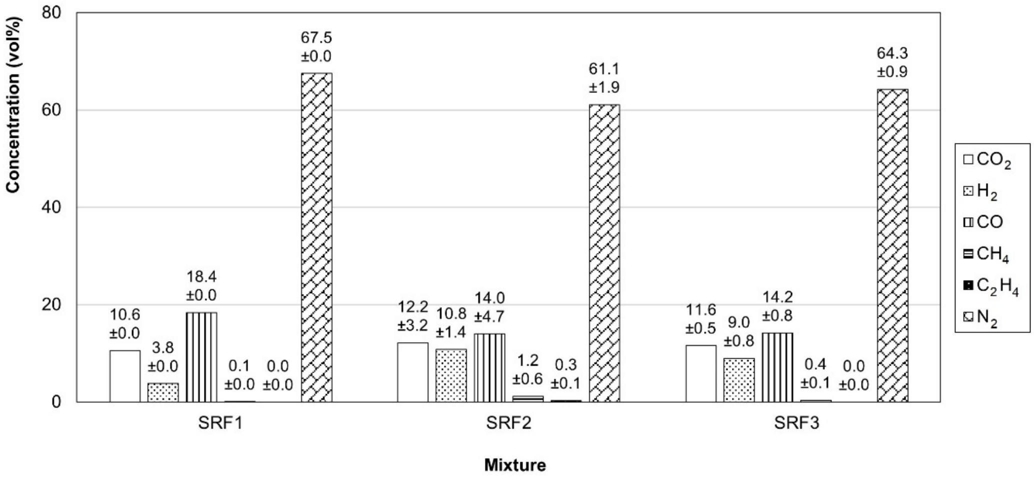

Figure 4. Composition of gas samples for each gasification test.

Table 6. Properties of gas samples.

Mixture	Lower Heating Value (LHV, MJ/Nm^3)	Density (kg/Nm^3)	Tar Amount (g/m^3)	HCl Concentration (g/Nm^3)
SRF1	2.8 ± 0.0	1.28 ± 0.00	5.3	1.1 ± 0.1
SRF2	3.5 ± 0.5	1.20 ± 0.03	11.3	1.9 ± 0.0
SRF3	2.9 ± 0.2	1.22 ± 0.01	7.4	1.7 ± 0.3

Regarding combustible components, the proportions of H_2 and CO found in gas samples oscillated among 3–11 vol % and 14–18 vol %, respectively, while those of CH_4 and C_2H_4 were almost irrelevant (<1.2 vol %). These results were near to those achieved in previous literature [36,39].

The addition of polymeric fractions to SRFs increased the formation of H_2, CH_4 and C_2H_4, particularly in the case of SRF2 where those values achieved 10.8 vol %, 1.2 vol % and 0.3 vol %, respectively. The formation of light gaseous hydrocarbons (namely CH_4 and C_2H_4) may be the result of the decomposition of long polymeric chains contained in plastics into simple monomers [15]. These light hydrocarbons may also be derived from the breakdown of tar compounds occurring at the oxidation zone of the reactor. The presence of high amounts of H_2, CH_4 and C_2H_4 contributed to the highest result of LHV measured in the gas from SRF2 (3.5 MJ/Nm^3). This means that the inclusion of plastic materials in the feedstocks improves the LHV of the gas. In addition, CO levels lowered from 18 vol % to 14 vol % when the plastic fraction was incremented, which was possibly due to the lower level of oxygen present in plastics and to the lower lignocellulosic fraction in the mixture; these aspects contributed to the diminishment of oxygen detected in mixtures SRF2 and SRF3 [15]. The gas from SRF1 had a high CO amount (18.4 vol %), which was probably due to the high admission of oxygen in the reactor, as deducted from the relatively high

ER obtained for this experiment (0.39). This significant admission of oxygen promoted the reaction that describes the partial oxidation of carbon, as represented by Equation (6) [40].

$$C + 0.5\ O_2 \leftrightarrow CO \qquad (6)$$

The slight reduction in gas LHV between SRF2 and SRF3 (from 3.5 to 2.9 MJ/Nm^3) was explained by the decrease in H_2, CH_4 and C_2H_4 contents, although the concentration of CO increased slightly. The presence of a higher amount of N_2 in the gas from SRF3 may also explain the attenuation of gas LHV. Based on this result, one may declare that there is no linear relationship among the polymeric fraction and the LHV of the gas produced; in fact, Ahmed et al. [41] argued that this occurrence can be explained by the synergetic effect with the lignocellulosic fraction that is present. Therefore, the LHV of the gas achieved a maximum result for SRF2, and it started to decrease for greater polymeric fractions as observed in SRF3. The results for gas LHV reported in other literature that considered the downdraft gasification of refuse-derived fuels (similar to SRFs studied in this work) revealed a disparity of results ranging from 0.5 to 5.9 MJ/m^3. This fact occurred due to the characteristics of materials and conditions of operations, but the present work generated results that fitted in the same range [39,42].

The presence of the toxic compound H_2S was negligible (<0.05 vol %) mainly due to the inexistence of sulphur in all SRFs; therefore, the gas does not present a relevant toxicity level regarding this specific compound.

Gas density assumed the highest result for SRF1 (1.28 kg/Nm^3) due to the significant amounts of dense gases (CO and N_2) when compared to the remaining experiments.

As reported by Zaccariello and Mastellone [15], the incorporation of plastics in SRFs increased the tar amount detected in the gas, which in the current study rose from 5.4 to 9.1 g/m^3 between SRF1 and SRF2. Such results are corroborated by the literature [43]. However, the direct use of product gases generated from all experiments in gas engines and turbines is not allowed, because tar concentrations are above the recommended limit of 50 mg/Nm^3 [43]. In this way, a convenient cleaning treatment is required to remove the excess of tars by using filters, water scrubbers, or catalytic beds before the final gas is converted into energy, as was mentioned previously [10,33].

The concentration of HCl in the gas ranged between 1 and 2 g/Nm^3, which is in accordance with the literature [9,36]. This range of values is above the limits recommended for practical gas applications, such as methanol synthesis (<0.1 mg/Nm^3) or electric energy generation through ICEs (<50 mg/Nm^3). A cleaning stage to remove HCl in excess is necessary, which can be performed through sand bed filters and rotational atomisers with removal efficiencies of 90% [9,44].

The gas produced from SRF2 exhibited the largest HCl concentration (1.9 g/Nm^3), showing a greater potential to corrode engines and turbines when compared with other SRFs. The content of this contaminant increased for greater plastic fractions present in the mixtures due to the high chlorine amounts found in some polymeric chains, especially in residues R7 (plastics from C&DW) and R9 (polymeric insulations from C&DW).

In spite of having the highest calorific value (3.5 MJ/Nm^3), the gas originated by SRF2 exhibited the largest tar and HCl amounts that may increase the costs for their removal during the cleaning treatment. Because these amounts are similar to those obtained from SRF3 and due to the dilution effect caused by mixing chlorine-rich polymeric fractions with lignocellulosic residues, the use of SRF2 seems to be the best choice for a gasification process in terms of energy recovery from the gas as well as for exploiting a wider variety of waste materials.

3.3.2. Char Properties

The results for the properties of the different chars originated from the gasification experiments are reported in Table 7.

Table 7. Composition and properties of gasification chars.

Property		Mixture SRF1	SRF2	SRF3
HHV (MJ/kg db)		0.0 ± 0.0 [1]	5.8 ± 0.1	4.6 ± 0.0
Ash (wt % db)		90.7 ± 0.1	79.9 ± 0.4	81.7 ± 0.0
Chlorine (wt % db)		3.4 ± 0.1	2.9 ± 0.0	4.2 ± 0.2
Mineral oxides in ash (wt % db)	CaO	40.3 ± 0.1	32.4 ± 0.3	29.5 ± 1.5
	SiO_2	12.3 ± 0.2	15.8 ± 0.6	15.9 ± 0.4
	Al_2O_3	3.9 ± 0.2	4.5 ± 0.4	4.9 ± 0.2
	Fe_2O_3	5.1 ± 0.4	7.5 ± 1.9	10.4 ± 2.3
	K_2O	2.1 ± 0.1	3.4 ± 0.1	2.7 ± 0.0
Heavy metals in ash (ppmw db)	Cd	0 ± 0	0 ± 0	0 ± 0
	Cr	799 ± 56	804 ± 68	1564 ± 584
	Cu	892 ± 41	3757 ± 769	2988 ± 391
	Hg	0 ± 0	0 ± 0	0 ± 0
	Ni	599 ± 31	421 ± 25	713 ± 53
	Pb	494 ± 18	514 ± 82	410 ± 33
	Zn	888 ± 14	1382 ± 3	1568 ± 36

[1] HHV for SRF1 was assumed to be null, because char samples did not ignite during the determination in the calorimetric bomb.

The high ash concentrations found in all chars (>80 wt % db) invalidate their use for energy production, since their HHVs are relatively low (<6 MJ/kg db). The char from SRF1 did not present any potential for heat production due to the highest ash amount (91 wt % db), therefore constituting the worst case among all char samples. The high ash concentrations found in chars were also observed in other works [45].

The chlorine amounts did not vary significantly with the polymeric fraction added to the mixtures. The average result was 3.5 wt % db, with a slight decrease in the case of SRF2 (2.9 wt % db), which was possibly due a stronger migration of chlorine to the gas phase. This fact may explain the higher HCl concentration found in the gas.

Regarding the mineral analysis, higher concentrations of CaO and SiO_2 were detected (>12 wt % db), which followed the same trends already described for raw mixtures SRF1–SRF3 before gasification. Other oxides were identified as well but with lower quantities, namely Fe_2O_3, Al_2O_3 and K_2O, with average values of 7.7 wt % db, 4.4 wt % db and 2.7 wt % db, respectively. The increment of the polymeric fraction fomented the rise of Si, Al and Fe contents, as well as a decline of the quantity of Ca. The presence of Ca and Fe in chars makes them adequate for the catalytic conversion of hydrocarbons during the process, thereby reducing tar formation and obstruction problems in downstream equipment [46,47].

All chars presented considerable amounts of heavy metals such as Cr, Cu and Zn, with concentrations above 750 ppmw db. The incorporation of polymeric wastes in SRFs contributed to a notable increase in Cu (+321% in SRF2) and for a less pronounced rise of Cr and Zn (+96% and +77% in SRF3, respectively), all increasing the toxicity of chars. According to the Portuguese legislation regarding the application of fertilisers for agriculture purposes (decree-law 103/2015, of June 15th), the amounts of Pb, Cu, Cr, Ni and Zn were all above the imposed limits (>100 ppmw db), which made unfeasible the use of chars for fertilisation.

Based on these results, chars may be valorised as catalysts for tar reforming and as an additive in construction materials to improve their properties (e.g., concrete, roofs, and roads) [45]. The first application is justified by the significant amounts of Fe and Ca that were observed, and which present catalytic activity during tar decomposition; the second can be explained by the reduction in material's bulk density and thermal conductivity [46–48]. These uses avoid their conduction to landfills that would represent an environmental problem and a waste of material resources.

3.4. Summary of the Influence of SRF Composition in Gasification Performance

To summarise the effect that the composition of SRFs had in the performance of gasification tests, Figure 5 illustrates the influence of varying the SRF polymeric fraction in gasification product yields (gas, chars and tars), LHV of product gas, and *CGE*.

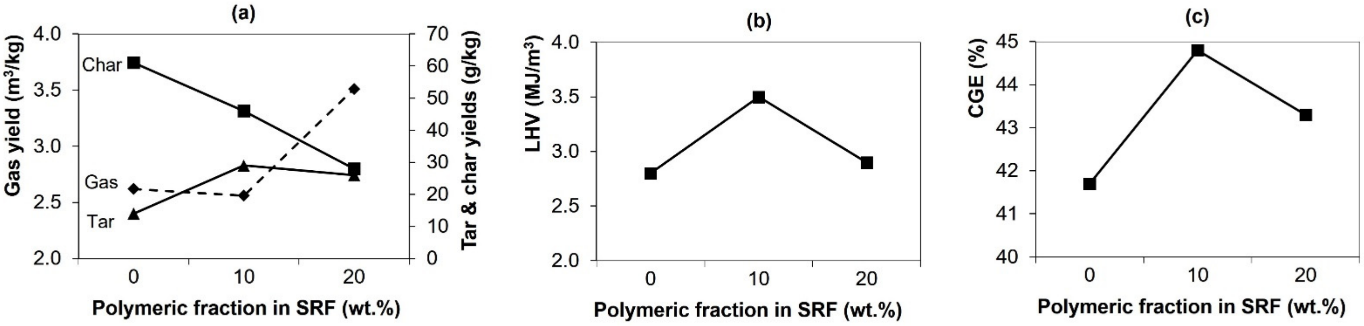

Figure 5. Summary of the effect of SRF polymeric fraction in some gasification parameters: (**a**) product yields, (**b**) lower heating value (LHV) of gas and (**c**) cold-gas efficiency (*CGE*).

The increase in the polymeric fraction contributed to a reduction in char yield through an almost linear relationship. Tar yield, gas LHV and *CGE* achieved their maximum values for the intermediate mixture SRF2 (with a polymeric fraction of 10 wt %), hence demonstrating that such parameters did not present a linear correlation with the amount of plastics in SRFs. According to the graphical results, the best mixture suited for gasification was SFR2 due to the highest LHV obtained for the product gas (3.5 MJ/Nm^3) and to the highest *CGE* (45%); however, SRF2 presented also some drawbacks, namely in terms of tar yield that was the highest (29 g/kg waste), and gas yield assumed the minimum result between all SRFs (2.6 m^3/kg waste). High tar production provokes obstruction, abrasion and corrosion problems in downstream equipment, but these can be minimised by implementing adequate gas cleaning processes (e.g., thermal or catalytic cracking, wet scrubbers, and absorption through oil scrubbing) [10]. The decomposition of tars into light gaseous compounds through thermal or catalytic cracking may eventually help to increase gas yield.

By comparing the results obtained for some relevant gasification parameters with those from other works, both gas yields (2.6–3.5 m^3/kg waste) and gas LHVs (2.8–3.5 MJ/Nm^3) were similar than those reported by Zaccariello and Mastellone [15] (2.0–3.5 m^3/kg waste and 6.2–7.9 MJ/Nm^3, respectively), who carried out air fluidised-bed gasification experiments in a pilot-scale reactor (feed of 5 kg/h) using mixtures composed of wood and plastics (0 wt %, 80 wt %, and 100 wt %). However, *CGE*s obtained in this work were substantially higher (73–76%), probably highlighting the beneficial effect in energy efficiency by increasing the contact time between feedstock particles, bed material, and gases produced. Khosasaeng and Suntivarakorn [39] conducted air downdraft gasification pilot tests with refuse-derived fuel (initial mass of 10 kg), and achieved gas LHVs and *CGE*s in the ranges of 4–6 MJ/Nm^3 and 40–70 %, respectively; these results are close to those obtained in the present work. By its turn, the use of oxygen as gasification agent and a slightly higher process temperature (900 °C) in a lab-scale downdraft gasifier (feed rate of 1 g/min) generated a gas with high LHVs (12.5–12.8 MJ/Nm^3) and similar *CGE*s (47.8–51.4 %) in comparison to the results from the current work [38]. This demonstrates the benefits of such operating conditions in the improvement of energy quality of the final gas.

4. Conclusions

In the present work, a series of gasification experiments were conducted in a pilot downdraft gasifier using SRF mixtures prepared with different waste components, which were collected mostly from C&DW. The polymeric fraction contained in these mixtures was varied between 0 and 20 wt %, with the purpose of studying the influence in gasification performance, as well as in the properties and yields of the products obtained.

SRFs containing polymeric wastes (SRF2 and SRF3) reduced the formation of chars inside the gasifier. At the same time, the product gas composition obtained from both mixtures contained greater amounts of H_2, CH_4 and C_2H_4, therefore contributing to the rise in gas LHV; however, tar and HCl production also increased, meaning that further cleaning treatments must be employed in order to make the gas suitable for practical applications, such as electric energy production or liquid fuels synthesis. In addition, both SRF2 and SRF3 produced gases with similar compositions. For all SRFs, no linear relationship was identified between the polymeric fraction and gas LHV, or *CGE*. Chars obtained from the experiments revealed a composition that may enable their valorisation as gasification catalysts for tar decomposition. According to the results that were achieved, SRF2 generated the best ones in terms of energetic performance during gasification and gas calorific value for energy purposes. These benefits may eventually balance the costs associated with HCl and tar removal from the gas, both assuming the highest results for SRF2.

This study presented a possibility to valorise organic fractions from C&DW through gasification for energy purposes. If economically feasible solutions can be applied to remove the excess of tars and HCl present in the product gas, then the gasification technology may potentially improve the sustainability in waste management and reduce the environmental impact caused by the use of conventional fossil fuels for energy applications. However, future techno-economic studies are required to evaluate accurately the feasibility of implementing the solution proposed in this work.

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