CHEMICAL ENGINEERING TRANSACTIONS
VOL. 101, 2023
A publication of AIDIC
The Italian Association of Chemical Engineering
Online at www.cetjournal.it
Guest Editors: Paolo Ciambelli, Luca Di Palma

ISBN 979-12-81206-00-7; **ISSN** 2283-9216
DOI: 10.3303/CET23101043

Gasification of Recycled Plastic Waste in a Fluidized Bed Reactor: Effect of Operating Conditions

Lucio Zaccariello*, Daniele Battaglia, Maria L. Mastellone

Università degli studi della Campania "L. Vanvitelli" - Dipartimento di Scienze e Tecnologie Ambientali, Biologiche e Farmaceutiche - Via Vivaldi, 43 – 81100 Caserta - Italy
lucio.zaccariello@unicampania.it

In this work, a lab-scale bubbling fluidized bed reactor was used to investigate the effect of operating conditions on the plastic waste gasification process performance. The effect of the equivalence ratio (0.25 and 0.30) and gasifying agent (air and oxygen-enriched air – 35% of oxygen) was investigated by keeping fixed the fluidization velocity (0.4m/s) and the type of the bed material (silica sand, 0.2-0.4mm). The process performance was assessed based on different parameters, such as producer gas heating value, yield of undesired by-products (tar and elutriated fines), carbon conversion efficiency, and cold gas efficiency. To further investigate the effect of the operating parameters, a Material Flow Analysis method was applied. The results indicated that the test with oxygen-enriched air generated a producer gas with the highest calorific value, while the test with air and equivalence ratio of 0.25 produced a gas with the most elevated concentration of tar.

1. Introduction

In the last decades, plastics have become an indispensable part of the human living style due to their low production cost, low density, and high durability (Lopez et al., 2018). Consequently, the global production of plastics has steadily increased, reaching 391 million tons in 2021. China reached almost one-third of the world's plastics production (32%), followed by North America (18%), and Europe (15%). The largest end-use markets are packaging (44%), building & construction (18%), and the automotive industry (8%). In Europe, polyolefins account for about half of the total plastics produced (40.6%), followed by polyvinyl chloride (11.4%), polystyrene (6.1%), polyurethane (5.5%), polyethylene terephthalate (5.3%). Although the amount of plastic waste sent to recycling has significantly increased, 6.9 million tons (23.4%) were still sent to landfill (Plastics Europe, 2022). Due to their low degradability, plastic waste can cause significant environmental and human health concerns (Pandey et al., 2023). More than 99% of plastics are produced from fossil fuels leading to a remarkable increase in greenhouse gas emissions and, as a consequence, elevated global warming levels (Sharma et al. 2022; Zaccariello and Montagnaro, 2023). Moreover, ingestion of food containing microplastics can cause severe complications to human and animal health (Prata et al., 2020).In the last years, numerous efforts have been made to mitigate the environmental impact of plastic waste by increasing the recovery of such products in an economical and ecologically-friendly manner. A possible solution for recovering valuable products from plastic waste is the gasification process (Huang et al. 2022; Zaccariello and Mastellone, 2023). On the other hand, during gasification, the formation of by-products such as char and tar can cause efficiency losses and operational troubles. Thus, the operating conditions of this process still need to be investigated to a deeper extent to reduce or prevent the formation of these by-products. The objective of this study is to investigate the effect of two crucial operating parameters, such as the equivalence ratio and the type of gasifying agent, on the plastic waste gasification process performance. The experimental runs were carried out at two different values of equivalence ratio (i.e., 0.25 and 0.30), utilizing air and oxygen-enriched air as gasifying agents, and with silica

Paper Received: 12 April 2023; Revised: 28 May 2023; Accepted: 15 July 2023
Please cite this article as: Zaccariello L., Battaglia D., Mastellone M.L., 2023, Gasification of Recycled Plastic Waste in a Fluidized Bed Reactor: Effect of Operating Conditions, Chemical Engineering Transactions, 101, 253-258 DOI:10.3303/CET23101043

sand as bed material. Moreover, the gasification process performance was assessed by applying the Material Flow Analysis (MFA) method.

2. Materials and Method

2.1 Experimental apparatus

The gasification tests were carried out using a lab-scale scale bubbling fluidized bed gasifier (BFBG) having a fuel feeding rate up to 4 kg/h, depending on the type of the fuel and operating conditions. The BFBG is made of AISI 316L, has an internal diameter of 0.10 m, and has an overall height of 2.5 m. The reactor is electrically heated by five shell furnaces, which allow for independently setting the temperature of each reactor section (plenum, bed, and freeboard). The gasifying agent is injected at the bed bottom through a distributor plate composed of three nozzles to ensure a homogeneous distribution of the gas in the bed cross-section. The feedstock was over-bed fed using a screw-feeder device. More detailed informations on the experimental apparatus and procedure are reported by Mastellone and Zaccariello (2013).

2.2 Analytical Equipment

The producer gas was analyzed by using a four channels Agilent 3000 micro gas chromatograph (μGC), which provides the gas composition in terms of CO_2, CO, H_2, CH_4, N_2, and light hydrocarbons, C_8H_m, i.e., hydrocarbons containing 2–8 atoms of carbon. The volumetric flow rate of the producer gas was determined using the tie component method applied to the nitrogen content in the dry producer gas.
The carbonaceous solid particles (elutriated fines, EF) collected by the cyclone and particulate filter, were analyzed in a LECO TruSpec Elemental Analyzer to determine the content of carbon, hydrogen, nitrogen, and sulphur. For the sampling of tar, a system composed of a heated probe and a series of five impinger bottles containing 50 mL of dichloromethane (DCM) was used. The first two bottles were placed in a water bath at 20°C, while the last three bottles were placed in a salt and ice bath at about -15°C. After the tar sampling, the content of the bottles containing tar dissolved in DCM was mixed, filtered, and stored in dark bottles at 4°C. Next, 5 mL of the tar sample was microfiltered and analyzed in a Perkin-Elmer Clarus 500 gas chromatograph coupled with a mass spectrometer (GC-MS).

2.3 Feedstock and bed material

The gasification tests were carried out using lenticular-shaped granules of recycled polyethylene with a diameter of 5 mm and a thickness of about 2 mm. Results of proximate and ultimate analyses of the feedstock utilized for the experimental runs are listed in Table 1.
Silica sand with a size range of 200-400 μm and a Sauter mean diameter of 210 μm as bed materials was used.

Table 1. Proximate and ultimate analyses, and heating value of the plastic waste

Feedstock	Moisture	Ash	C	H	N	O	LHV
-	wt.%		wt.%				MJ/kg_{Fuel}
PW	0.2	1.00	85.00	13.80	0.00	0.00	39.88

3. Results and Discussion

The effects of ER and gasifying agent on the gasification of plastic waste were studied keeping fixed the fluidization velocity (Ug = 0.4 m/s) and conducting the experimental runs in autothermal conditions, i.e., without an external heat energy supply in addition to the sensible heat of the gasifying agent (heated at 500°C in all the gasification tests). The results display that the reaction temperature (T_{Bed}) increased from 885°C to 892°C as the ER was moved from 0.25 to 0.30, and further increased to 927°C in the test with ER=0.30 and oxygen-enriched air (Table 2).

Table 2. Main results obtained from the gasification tests

ID	T_{Bed}	CO_2	CO	H_2	CH_4	C_8H_m	Q_{Gas}	LHV_{Gas}	C_{EF}	C_{Tar}	CCE	CGE
-	°C	%	%	%	%	%	Nm^3/h	kJ/Nm^3	g/Nm^3	g/Nm^3	%	%
PW-25-A	885	8.04	11.25	13.1	4.92	3.45	3.90	7327	0.37	17.62	81.48	68.42
PW-30-A	892	9.97	8.51	8.19	3.84	3.87	3.51	5455	0.72	10.61	82.76	60.44
PW-30-AE	927	11.18	16.1	17.54	6.53	2.24	4.32	8143	23.06	6.08	84.57	65.24

The increase in reaction temperature in the tests PW-30-A and PW-30-EA was due to the larger amount of oxygen available in the gasifier that promoted exothermic reactions. This hypothesis was supported by the increase in the concentration of CO_2 in the producer gas in the test PW-30-A and PW-30-EA with respect to the test conducted at the lower ER value (PW-25-A). The test PW-25-A showed higher concentrations of CO (+32.2%), H_2 (+59.9%), CH_4 (+28.1%), and C_8H_m (+21.9%) than that obtained in the test PW-30-A, this was due to the lower ER value, which determined more enhanced reducing conditions. The effect of oxygen-enriched air on the producer gas composition can be evaluated by comparing the tests PW-30-A and PW-30-EA. Table 2 displays that oxygen-enriched air produced an increase of CO_2 (+12.1%), CO (+89.2%), H_2 (114.2%), CH_4 (+70.1%), and a reduction of C_8H_m (-20.9%). The composition of the producer gas obtained in the test PW-30-EA was affected by the reduced amount of nitrogen in the gasifying agent, which produced a limited diluting effect compared to the PW-30-A test. To a large extent, the impressive increase of CO, H_2, and CH_4, and the reduction of C_8H_m can be attributed to the greater development of the Boudouard reaction and thermal dehydrogenation/cracking of heavy hydrocarbons due to the higher reaction temperature. This led to the production of a producer gas with the highest lower heating value (LHV), which resulted equal to 8143 kJ/Nm^3. On the other hand, the test PW-30-A (with the higher concentration of N_2) showed an LHV of only 5455 kJ/Nm^3.

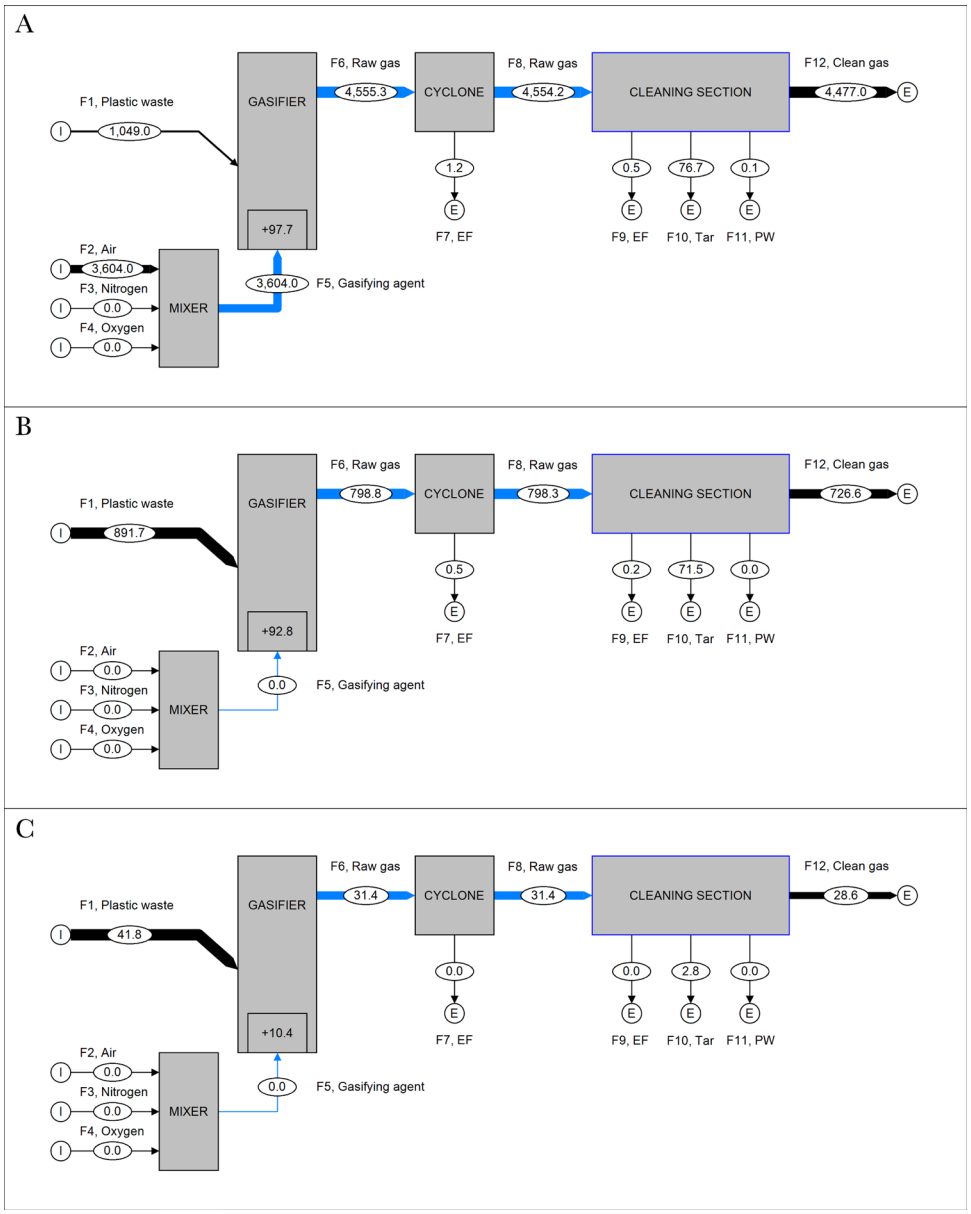

Figure 1: Material and energy flow analyses applied to the test PW-25-A. (A) Total mass layer (g/h); (B) Carbon layer (g/h); (C) Energy layer (MJ/h).

The main advantage of using oxygen-enriched air as a gasifying agent is the reduced concentration of tar in the producer gas. It resulted equal to 6.08 g/Nm^3 in the test PW-30-EA while it was 17.62 and 10.61 g/Nm^3 in the tests PW-25-A and PW-30-A, respectively. On the other hand, the test with oxygen-enriched air showed an impressive production of EF (23.06 g/Nm^3), much higher than that produced during tests with air at different ERs (0.37 and 0.72 g/Nm^3). The remarkable production of EF observed during the test PW-30-EA can be explained by considering that the greater reaction temperature, due to the greater concentration of oxygen in the gasifying agent, supports the thermal carbonization of heavy hydrocarbons (tar) through dehydrogenation and polymerization reactions, which increase the production of hydrogen and solid condensed aromatic molecules (soot).

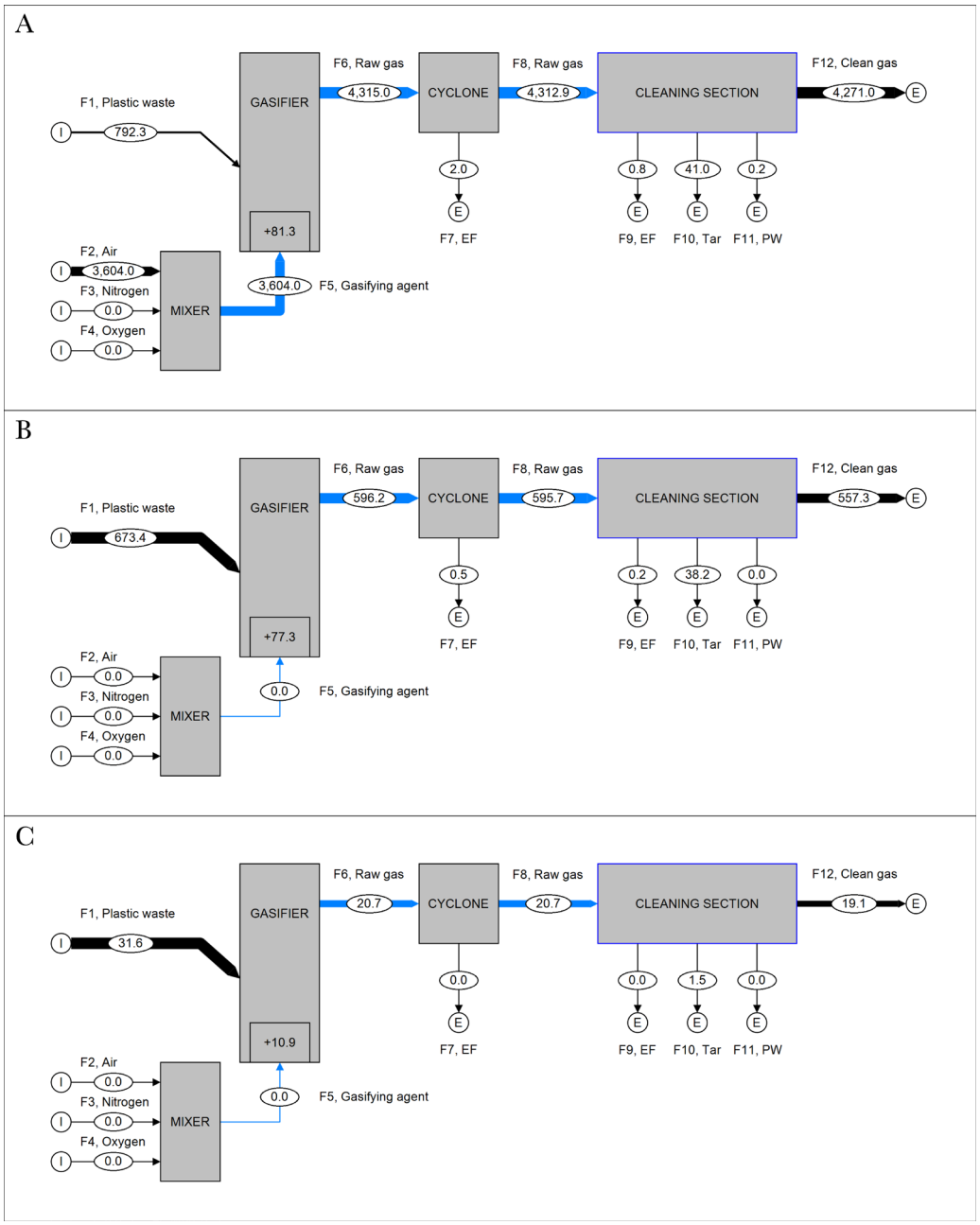

Figure 2: Material and energy flow analyses applied to the test PW-30-A. (A) Total mass layer (g/h); (B) Carbon layer (g/h); (C) Energy layer (MJ/h).

Figure 1 displays the result of MFA applied to the gasification apparatus under the operating conditions of the run PW-25-A. Figure 1A represents total mass flow rates. It shows that 1049 g/h of plastic waste (F1) and 3604

g/h of preheated air (F5) were fed to the gasifier and transformed into 4555 g/h of raw gas (F6). Then, the raw gas was sent to the cyclone which, removed 1.2 g/h of EF (F7). Successively, the raw gas enters the cleaning unit composed of a hot quartz wool filter, a scrubber, and a demister, where the remaining EF (F9), tar (F10), and process water (PW, F11) were removed. Finally, a stream of 4477 g/h of conditioned gas (F12) was sent to a gas flare. The chemical characterization of plastic waste, producer gas, EF, and tar streams allowed to perform the mass balance applied to the carbon element, i.e., the carbon layer in Figure 1B and the energy balance, i.e., the energy layer in Figure 1C.

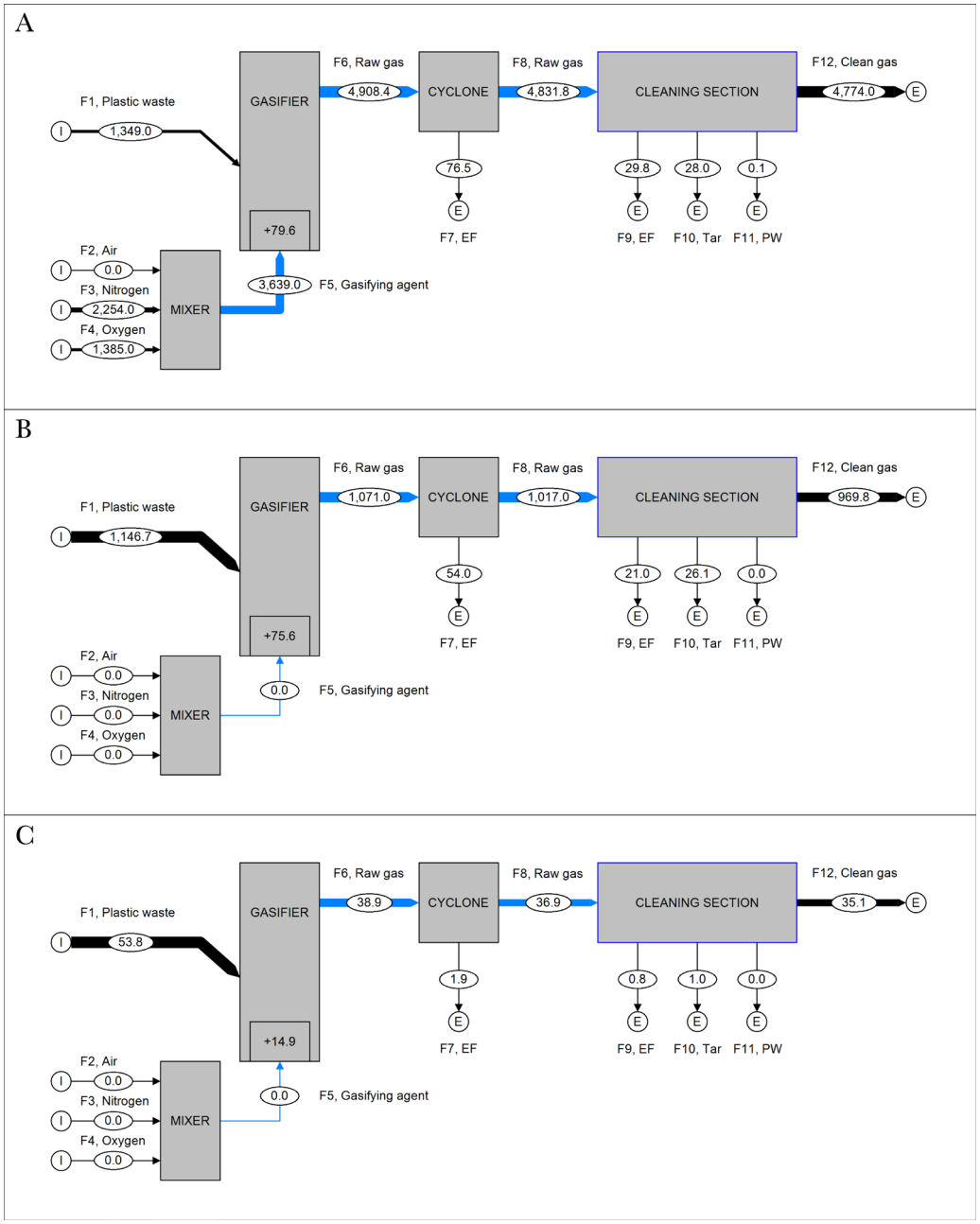

Figure 3: Material and energy flow analyses applied to the test PW-30-EA. (A) Total mass layer (g/h); (B) Carbon layer (g/h); (C) Energy layer (MJ/h).

Results of mass balance on the carbon element allowed to calculate the carbon conversion efficiency (CCE), defined as the ratio between the mass flow rate of carbon contained in the producer gas (F12-Figure 1B) and the mass flow rate of carbon fed to the reactor through the plastic waste (F1-Figure 1B). The results of mass balance on the carbon element for the test PW-25-A showed that the CCE was equal to 81.48% (Table 2).

Figure 1B displays that the CCE was mainly affected by the carbon losses related to the tar collected by the cleaning unit (71.5 g/h, F10-Figure1B). Also in the test PW-30-A the main carbon loss was associated with the tar stream at the exit of the cleaning unit (38.2 g/h, F10-Figure 2B) and CCE resulted equal to 82.76%. Otherwise, in the test PW-30-EA the larger carbon loss was due to the EF discharged by the cyclone (54.0 g/h, F7-Figure 3B) and the CCE resulted equal to 84.49%.Data from the energy layer (Figure 1C) allowed to calculate the cold gas efficiency (CGE), defined as the ratio between the chemical energy of the producer gas generated during the gasification process (F12-Figure 1C) and that entering to the gasification reactor through the plastic waste (F1-Figure 1C). CGE for this test was 68.42% and resulted mainly affected by the energy used by the system to convert the plastic waste into producer gas, which amounted to 10.4 MJ/h. Also for the tests PW-30-A (Figure 2C) and PW-30-EA (Figure 3C) the CGE was mainly influenced by the energy the system used to convert plastic waste into gasification products, and it resulted 60.44 and 65.24% (Table 2), respectively.

4. Conclusions

The lab-scale bubbling fluidized bed gasifier was operated by varying the ER (0.25-0.30) and the gasifying agent (air or oxygen-enriched air) to optimize the process performance and obtain a producer gas with low concentrations of by-products. The results indicated that the test conducted with air and an ER of 0.25 generated a producer gas with a high concentration of tar (17.62 g/Nm^3) and a low concentration of elutriated fines (0.37 g/Nm^3). Instead, the test with oxygen-enriched air and an ER of 0.30 produced a lower concentration of tar (6.08 g/Nm^3) and an impressive amount of elutriate fine (23.06 g/Nm^3). Furthermore, in this test, the best lower heating value of producer gas was obtained (8143 kJ/Nm^3). As regards the test with air and an ER of 0.30, intermediate values of EF, tar, and lower heating value of producer gas were obtained. Material flow analysis showed that the carbon conversion efficiency was mainly affected by the carbon loss due to the tar collected by the cleaning unit in the tests with air at different ERs. Instead, in the test with oxygen-enriched air, the carbon conversion efficiency was mainly influenced by the elutriated fines collected by the cyclone. The energy flow analysis revealed that the cold gas efficiency was greatly impacted, in all the gasification tests, by the energy the system used to convert the plastic waste into producer gas, with the higher value (14.9 MJ/h) detected in the test with oxygen-enriched air. Despite this, the test PW-30-EA showed an intermediate CGE value (65.24%), due to the high energy content of the producer gas and the lower tar concentration.

Acknowledgments

Authors are indebted to Mr. Giuseppe Barbato and Mr. Donato Santoro that performed the off-line analyses of the reported experimental runs.

References

Huang J., Veksha A., Chan W.P, Giannis A., Lisak G., 2022, Chemical recycling of plastic waste for sustainable material management: A prospective review on catalysts and processes, Renewable and Sustainable Energy Reviews, 154, 111866.

Lopez G., Artetxe M., Amutio M., Alvarez J., Bilbao J., Olazar M., 2018, Recent advances in the gasification of waste plastics. A critical overview, Renewable and Sustainable Energy Reviews, 82, 576–596.

Mastellone M.L., Zaccariello L., 2013, Gasification of polyethylene in a bubbling fluidized bed operated with the air staging, Fuel, 106, 226–233.

Pandey K.P., Jha U.R., Kushwaha J., Priyadarsini M., Meshram S.U., Dhoble A.S., 2023, Practical ways to recycle plastic: current status and future aspects, Journal of Material Cycles and Waste Management, 25, 1249–1266.

Plastics Europe, 2022, Plastics – the Facts 2022. An analysis of European plastics production, demand and waste data <plasticseurope.org/wp-content/uploads/2022/12/PE-PLASTICS-THE-FACTS_FINAL_DIGITAL.pdf> accessed 30.03.2023.

Prata JC., da Costa J.P., Lopes I., Duarte A.C., Rocha-Santos T., 2020, Environmental exposure to microplastics: an overview on possible human health effects, Science of Total Environment, 702, 134455.

Sharma V., Hossain A.K., Griffiths G., Duraisamy G., Krishnasamy A., Ravikrishnan V., Sodré J.R., 2022, Plastic waste to liquid fuel: A review of technologies, applications, and challenges, Sustainable Energy Technologies and Assessments, 53, 102651.

Zaccariello L., Mastellone M.L., 2023, Fuel Gas Production from the Co-Gasification of Coal, Plastic Waste, and Wood in a Fluidized Bed Reactor: Effect of Gasifying Agent and Bed Material, Sustainability, 15, 7547.

Zaccariello L., Montagnaro F., 2023, Fluidised bed gasification of biomasses and wastes to produce hydrogen-rich syn-gas – a review, Journal of Chemical Technology and Biotechnology, 98, 1878–1887.