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Development of an experimental test rig for the pyrolysis of plastic residues and waste tires

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> **Abstract.** The paper presents the design of the experimental apparatus developed in order to analyse the performance of a prototype of a pyrolysis system for the exploitation of the plastic residues of industrial processes and the end of life tires. The small scale prototype is specifically designed for carrying out an experimental campaign aimed at determining the influence of different plastic types on the syngas yield and on the quality of the liquid oil, gas and char obtained in the pyrolysis process. The study investigates the effect of different mixtures of various plastic products mainly made of polyethylene, styrene butadiene rubber, nylon and natural rubber. The prototype is equipped with a control system able to monitor the main operating parameters of the process, such as the syngas pressure and temperature as well as the temperature inside the reactor where the pyrolysis takes place. The monitored variables are employed for deriving correlations among the operating conditions and the yield of the pyrolysis process. Experimental measurements are carried out in order to estimate the main compounds that are contained in the syngas for the different plastic wastes analysed. Finally, the emissions of the small-scale prototype are evaluated.

1 Introduction

In the past fifty years, plastic has attracted considerable interest as a material for the enhancement of the properties of different products in many sectors, such as packaging, building and construction, automotive, electronics, biomedical and others.

In today's scenario, plastics production has reached about the 311 million tonnes in 2014 [1] determining a continuous growing in waste accumulation every year. Plastic solid waste is being produced on a massive scale worldwide and its production exceeds the 150 million tonnes per year globally, as presented in [2]. The European strategy for Plastics in a circular economy [3] reports that 25.8 million tonnes of plastic waste are generated in Europe every year. Due to its well established environmental impact, the plastic waste is becoming a key priority and different technologies for treating the plastic residues already exist.

Al Salem et al (2009) [4] presented a review on the different routes for recycling and recovery the plastic solid waste. They identified four different strategies for the PSW

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treatment such as re-extrusion, mechanical treatment, chemical treatment and energy recovery. According to statistics established in Europe [3], less than the 30 % of the plastic waste is collected for recycling, while the landfilling and incineration rates are 31% and 29 % respectively. Even though the percentage of the plastic waste disposed to landfill has slightly decreased over the last decades, it is still very high.

At Salem et al [1] presented the mechanical recycling as the most common technique for the recycling of plastic waste. In 2017, Ragaert K. et al [5] presented a description of the main steps of the mechanical recycling, which includes collection, sorting, washing and grinding of the material. Their study highlights that polymers will degrade during the mechanical recycling; this degradation occurs during lifetime and by reprocessing.

For this reason, many materials can be reused a limited number of times before they cannot be longer chemically active to produce products with certain chemical properties [6].

The amount of plastic disposed to landfill that is not treated via mechanical recycling can be used for energy recovery. In [3], it was estimated that the production and the incineration of the plastic give rise globally to approximately 400 million tonnes of CO_2 a year. As a consequence, the European Commission has introduced specific measures and procedure [7] in order to reduce the negative effect on the environment from the incineration of hazardous waste.

New sustainable processes of recycling have emerged, defined as chemical recycling [8], a process that converts plastic into liquids or gases which can be used as a fuel or as feedstock for the production of new plastic products [9, 10]. Most of the studies available in literature present the gasification and the pyrolysis processes as the advanced thermal treatment methods for the reduction of the plastic waste. The gasification is a process that operates at high temperatures, i.e. $600-800^{\circ}$ C, based on the thermo-chemical decomposition of the plastic into a synthesis gas that is suitable for the electricity production or for transportation fuel [11]. In 2013, A. Brems et al. [12] confirmed the importance of the introduction of the gasification process of the plastic solid waste into a valid recycling route of the PSW producing some syngas rich in H₂ and CO.

Pyrolysis is the thermal decomposition that occurs in the absence of oxygen and produces energy from waste in three different phases: solid bio-char, liquid oil and syngas [13].

In literature, many studies have been conducted regarding the influence of various types of plastics on the product yield. In [14], the set-up parameters that affects the pyrolysis process are identified and they include temperature, type of reactors, residence time, pressure.

The pyrolysis process can operate in large range of temperatures from 300 to 1000 $^{\circ}$ C [15-17].

In 2011, Kumar et al. [18] observed that the degradation of the polyethylene started at 400 °C and the maximum weight loss is at 427.7 °C.

In [19] the polystyrene pyrolysis was analyzed and the maximum yield of liquid oil was reached at 425 °C. Kan et al [20] investigated the pyrolysis process of natural and synthetic rubber. The study determined the syngas composition by chromatography technology and demonstrated that the syngas is mainly composed by hydrogen and carbon monoxide; the H_2 peaks are registered between 350 and 525 °C for the natural rubber, while in the range 300-500°C hydrogen is the dominant species for the synthetic rubber.

In literature there are many studies that investigates different technologies and layout of the systems for the pyrolysis. In particular, the pyrolysis systems can be divided in two different configurations. The first layout is composed by a reactor, a distillation column, a condenser and a reboiler; the latter configuration consists of a reactor and a combustion chamber [13]. In the second design there is no liquid production because there is no condenser.

The aim of this paper is the design of a test rig for investigating the pyrolysis of different mixture of plastic waste. The experimental campaign focuses on the pyrolysis of polyethylene, styrene butadiene rubber, nylon and natural rubber. The experimental apparatus reproduces the layout presented in [13]. The prototype is composed by a pyrolysis reactor, heat exchanger, a separator and a filter. This configuration enables to produce energy from different plastic waste obtaining syngas, oil and solid bio-char [13].

The test rig is equipped with an acquisition system that monitors the main variables of the process by means of temperature and pressure sensors placed in characteristic position of the prototype. As a consequence, it is possible to investigate the thermal behavior of the process and the influence of the operating temperature on the yield of the process.

Finally, the composition of the syngas and the main compounds generated by the syngas combustion are evaluated in order to asses the sustainability of the process.

2 MATERIALS AND METHODS

The present study focuses on the design of the experimental apparatus for the pyrolysis of different mixture of plastic waste. The aim of this paper is the development of a test rig for carrying out an experimental campaign in order to investigate the thermal behavior and the sustainability of the pyrolysis process of different plastic residues.

2.1 Pilot plant

2.1.1 Layout

Figure 1 depicts the experimental test rig developed for the experimental tests and the locations of the temperature and pressure sensors installed in the system.

Figure 1 depicts the structure of the pilot plant along with the main sensors used in the analysis. The experimental apparatus includes three main parts: the reactor, the condenser/separator and the filter for the treatment of the exhaust generated by the syngas combustion.

The main unit is the reactor where the pyrolysis process takes place and it is composed by two detached cavities. The burner is placed in the external chamber and the air is maintained in circulation in order to transfer the heat uniformly to the internal cavity. The chemical transformation of the plastic waste takes place in the internal cavity which is completely sealed in order to avoid syngas leakages. The sealing action is performed by two different gaskets: the first made by silicone and the second one composed by an insulation fabric.

The internal cavity is 1.5 mm thick and the internal volume is approximately about 1 m^3 and the maximum material load equals to 100 kg.

A heat exchanger and a separator are connected downstream of the reactor. The heat exchanger cools down the syngas in order to condensate the liquid part which is stored in the separator. The remaining syngas flows to the torch; in the real system the non-condensed syngas is used to power the reactor.

A scrubber system (not shown in Figure 1) is added before the exhaust chimney in order to filter the syngas gases generated by the syngas combustion.



Fig. 1. Schematic of the pilot plant and sensors location

2.1.2 Monitoring System

The sensors installed in the system are employed to monitor the main variable of the process. The temperature and pressure of the process are measured by eight thermocouples and four pressure transducers placed in characteristic parts of the system.

The temperature distribution inside the internal chamber is monitored by three different thermocouples (A); these sensors enable to investigate the suitable temperature for the pyrolysis reaction in order to maximize the yield of the process.

A temperature (D) and a pressure (E) sensor are installed to register syngas variables at the reactor outlet. The thermocouple (F) measures the temperature of the water inside the heat exchanger. Furthermore, two thermocouples (G, I) and two pressure transducers (H, L) are positioned upstream and downstream of the separator to monitor the temperature and pressure of the syngas respectively before and after its separation from the liquid phase.

Finally, a thermocouple (B) and a pressure transducer (C) are placed at the chimney in order to measure the exhaust temperature and pressure generated by the GPL burner.

Table 1 lists the operating range and the accuracy of the measuring instruments employed to characterize the system.

Table 1. Specifications of the sensors instance					
Sensor type	Operating range	Accuracy	Reference letter in Figure 1		
Thermocouples	-270÷1370 °C		A, B, D, F, G, I		
Pressure sensor	-1÷1 bar	<0.02% FS	Е		
Pressure sensors	0÷0.5 bar	<0.02% FS	C, H, L		

Table 1. Specifications of the sensors installed

The data acquisition is carried out using a National Instrument DAQ module integrated by a customized LabView interface. Through this interface the user is also able to control remotely all the working parameters of the system.



Fig. 2. LabVIEW monitoring interface

2.1.3 Industrial gas analyzer

The composition of the syngas at the outlet of the separator is analyzed by means of an industrial gas analyzer. The system acquires the composition of the syngas in real time, i.e.1 sample every 10 seconds.

The hardware technology is based on infrared sensors (NDIR) and electrochemical sensors (ECS) that enables the identification of the elements that compose the syngas.

The elements alanalyzed by the system are O_2 , CO, CO_2 , NO, NO_2 , CH_4 , H_2 and they are measured as a percentage of the total mass.

In Figure 3 the industrial gas analyzer is shown.



Fig. 3. Measuring instrument installed on the pilot plant.

2.3 Experimental tests

In this paper, an experimental campaign has been carried out in order to investigate the pyrolysis of different plastic types.

The collected plastic samples consist of waste tires, grocery bags, fabrics and vulcanized pipes that are mainly made of polyethylene, styrene butadiene rubber, natural rubber and nylon. The collected samples are used individually and in a mixture.

Table 2 lists the three experimental tests carried out in this study:

Tuble 2. Emperimental tests analysed						
Test	Typology	Main Compounds	Quantity [kg]			
#1	Waste tyres	SBR	34.5			
#2	Grocery bags	PE	11.5			
#3	Fabrics and rubberized pipes	Nylon, SBR, NR	21.7			

Table 2. Experimental tests analysed

The pyrolysis process operates approximately between 400-420 °C and a residence time in the reactor of 3-4 hours is used for all experiments depending on the composition of the materials of each batch cycle.

3 RESULTS

3.1 Temperature and pressure analysis

The experimental campaign explained in the previous paragraph is used for investigating the operating parameters in terms of temperature and pressure that optimize the pyrolysis process of the studied samples.

A total of 3 tests are carried out and in particular in test #1 the pyrolytic reactor is loaded with waste tires (4 SUV tires). The complete cycle lasted about 3h and the transients related to the temperatures and pressures recorded by the sensors are presented in the following.

In Figure 4(a) the temperatures of the eight thermocouples installed are depicted. The time evolution of the temperature measured at the chimney is directly relating to the heating process of the pyrolytic cavity. Indeed, in the first part, the temperature of the cavity is set approximately to 400 °C and remains constant for 15', afterwards a lower value is defined and maintained constant until the end of the experimental test. Consequently, the temperatures measured by the three thermocouples installed into the internal chamber follow a similar trend. The curve labelled "syngas input" shows the temperature of the syngas at the internal cavity exit. This temperature remains lower than 100 °C and then it increases when the syngas starts to flow out.

The experimental test #2 is conducted by introducing waste plastic bags into the reactor cavity. Unlike the previous test reported above, for this test the material was inserted into the pyrolytic reactor following a preheating phase of the cavity. In particular, the plastic bags are introduced inside the cavity once a temperature of 100 °C is reached.

The trend of the temperatures recorded during the test are shown in Figure 4(b).

Thanks to the temperature history represented in Figure 4(b), it is possible to reconstruct the entire development of the experimental test. Indeed, at 12h 00' a temperatures decrease at the chimney and in the internal cavity due to the opening of the front door can be seen in order to load the material following the preheating phase. For the initial period of time the pyrolytic reactor was kept at a constant temperature of about 350 °C and then raised by a hundred degrees in order to allow the complete development of the process. In the final phase the process was shut down and cooled and all temperatures consequently reduced.

The experimental test #3 is conducted by introducing vulcanized pipes and rubber and metallic fabric into the reactor cavity. The trend of the temperatures recorded during the test are represented in Figure 4(c).

After a first heating part of the pyrolytic reactor, the process is kept at a constant temperature in order to facilitate the transformation of all the different loaded materials. As can be seen from Figure 4(c), the increase in temperature measured at the syngas input coincides with the beginning of the formation phase of the syngas.

Figure 4(d) represents the trend of the pressures registered by the four pressure transducers during the 4h duration of the second experimental test and saved by the LabVIEW software.

The trend of the pressures resulted constant, following an initial settling phase. Indeed, the sensors, at the beginning of the test, registered the pressure due to the air inside the system, which is driven by the hot vapors exiting the pyrolytic reactor. Instead, in the final part of the process when the pressure produced by the syngas is very weak, the monitored pressures decrease significantly.



Fig. 4. (a) Waste tires pyrolysis temperatures graph, (b) Polyethylene bags pyrolysis temperatures graph, (c) Rubber fabric and vulcanized pipes waste pyrolysis temperatures graph, (d) Polyethylene bags pyrolysis pressures graph

3.2 Syngas yield

The effect of different plastic waste types on pyrolysis product yields is shown in Figure 5. All types of plastic waste are degradated at the pyrolysis temperature of 400-420° C.

At the end of the experimental test #1, as a result of the pyrolytic process, into the internal chamber of the reactor 16 kg (46% wt) of char and 5 kg (14% wt) of steel belts (used to reinforce SUV tires) remained. From the phase separator 12 kg (36% wt) of raw fuel oil are collected. Indeed, the pyrolysis of waste tires generally aims at maximizing the yield of the liquid-phase product, because of the valuable chemicals obtained from it. By subtracting the measured weight of the products, it is possible to calculate the amount of syngas generated by the pyrolytic process and burned by the flare: 1.5 kg (4% wt).

Thermal pyrolysis of grocery bags (PE) during the test #2 produced relatively less amounts of liquid oil (26%) and char (9%) along with high yield of gases (65%). Unlike the

waste tires, which produce a significant amount of fuel oil, the polyethylene bags are transformed mostly into syngas.

Results of the last experimental test, test #3, can be summarized as follows: 4.5 kg (21% wt) of char and 3 kg (14% wt) of steel fibers (used to reinforce pipes and fabrics). Within the phase separator 9 kg (42% wt) of raw fuel oil are collected. Finally, the amount of syngas generated by the pyrolytic process and burned by the torch can be estimated into 5.2 kg (23% wt).



Fig. 5. Pyrolysis yields for different plastic wastes

3.3 Composition

In the following the composition of the syngas in each experimental test is provided on the basis of the values obtained by the mean value from the measurements by the industrial analyzer.

In Table 2 the composition of the syngas related to the test #1 is presented. Usually the gas-phase products from the pyrolysis of waste tires are paraffins (that were clearly visible on the bottom surface of the pipes), olefins, hydrogen, carbon oxides and a small amount of other compounds. The H_2 and CH_4 encompassed the majority of the volumetric composition unlike CO_2 and O_2 that represent either the 20% of the total volume of the syngas. This particular volumetric composition of the syngas obtained from waste tires is characterized by a high-calorific power.

Table 2. Volumetric syngas composition						
O ₂	H ₂	CO	CO ₂	CH ₄	Total	
1	48	10	19	22	100	

The volumetric composition of the pyrogas related to the experimental test #2 is presented in Table 3. Unlike the previous case related to the pyrolysis of waste tires, in this case the volumetric composition varies slightly. The percentages of CH_4 and H_2 are reduced, conversely those related to CO and CO_2 are increased. Moreover, the syngas from the pyrolysis of polyethylene bags is also characterized by a small percentage of N_2 . Consequently, in this case the calorific value is reduced compared to the previous one due to the different concentrations found. The remaining composition is due to other components that the industrial analyzer was not able to detect.

 Table 3. Volumetric syngas composition

				10	1	
O ₂	N_2	H ₂	CO	CO ₂	CH ₄	Total
0.2	6	35.4	24	17.3	12.1	92

In Table 4, the volumetric composition of the pyrogas obtained in test #3 is presented. In the table below, the H_2 and CH_4 determined the main compounds in the of the volumetric composition and CO and O_2 represent the 10% of the total volume of the syngas.

Table 4. Volumetrie Syngus composition						
O ₂	H ₂	CO	CO ₂	CH ₄	Total	
1	25	9	28	32	95	

 Table 4. Volumetric syngas composition

4 CONCLUSIONS

In this paper a pyrolytic reactor prototype has been developed and implemented.

Real time acquisition and monitoring of temperatures and pressures in the main sections of the system was carried out by means of the LabVIEW software, and a control tool for the main parameters of the process was developed.

The temperature and pressure transients for each experimental test conducted were presented.

The percentages of the output products were monitored and compared with those already present in literature and a good agreement between was found.

The percentages of the main compound/element constituting the syngas were analyzed in real time and the values proved that the pyrolysis can be and efficient waste to energy technology to exploit plastic residues of different types.

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