



Thermic pyrolysis of polypropylene waste as a source of fuel

Pirólisis térmica de polipropileno como fuente de combustibles

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Received: December 03, 2020; Accepted: March 30, 2021

Abstract

In this study, the pyrolysis of plastic wastes of polypropylene was investigated to study the quality of liquid products and to identify optimum process conditions for maximizing the yield of the liquid product. Experiments were performed in a stainless steel reactor under the nitrogen atmosphere. The effects of pyrolysis temperature with a constant heating rate on the yields of the products were investigated. The pyrolysis tests were performed to temperatures between 450 °C and 473 °C with a constant heating rate of 15 °C min⁻¹. The results showed that increasing the pyrolysis temperature from 400 °C resulted in a significant increase in the liquid and gas yield. Also, the presence of resins combined with polypropylene and additives modify the thermal degradation kinetics of polypropylene, although not in a significant way concerning the results reported by other researchers for virgin polypropylene. The physical-chemical characteristics of the liquid fraction of pyrolysis products have been compared with those of fossil fuel such as gasoline and diesel, the sulphur content of the liquid product fraction below 3 ppm was novel.

Keywords: Thermal pyrolysis, polypropylene, degradation temperature, alternative fuel.

Resumen

En este estudio, se investigó la calidad de los productos líquidos y la identificación de las condiciones óptimas del proceso de pirólisis de residuos plásticos de polipropileno para maximizar el rendimiento de la fracción líquida. Los experimentos se realizaron en un reactor de acero inoxidable bajo atmósfera de nitrógeno. Se investigaron los efectos de la temperatura de pirólisis con una velocidad de calentamiento constante sobre los rendimientos de los productos. Las pruebas de pirólisis se realizaron a temperaturas entre 450 °C y 473 °C con una velocidad de calentamiento constante de 15 °C min⁻¹. Los resultados mostraron que el aumento de la temperatura de pirólisis de 400 °C resultó en un aumento significativo en el rendimiento de líquido y gas. Así como, la presencia de resinas combinadas con polipropileno y aditivos modifican la cinética de degradación térmica del polipropileno, aunque no de manera significativa respecto a los resultados reportados por otros investigadores para polipropileno virgen. Se han comparado las características físico-químicas de la fracción líquida de los productos de pirólisis con las de los combustibles fósiles como la gasolina y el diesel, el contenido de azufre de la fracción de producto líquido por debajo de 3 ppm fue novedoso.

Palabras clave: Pirólisis térmica, polipropileno, temperatura de degradación, combustible alternativo.

1 Introduction

The global production of plastics, polymer resins, and fibers, in the year 2015 was estimated as 381 million metric tons (MT), which increased by 75% in the year 2020 (Lee *et al.*, 2020; Moldoveanu, 2019). It was also estimated that the global plastic waste generation in 2025 will increase to 9-13% of total Municipal Solid Waste (MSW) (Raman Sharma & Sharma, 2014). The

plastic demand in the European Union was 58 million tons, 29.7% was recycled, 39.5 was recovered in the form of energy, mainly incineration, and 30.8% was sent to landfill (Gu *et al.*, 2017).

Despite these benefits, its presence has caused serious environmental problems due to low degradability, for this reason, plastic waste occupies the landfill space. The presence of plastic waste in Municipal Solid Waste (MSW) has increased from less than 1% in 1960 to more than 10% in recent years.

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<https://doi.org/10.24275/rmiq/IA2321>
ISSN:1665-2738, issn-e: 2395-8472

Plastic waste generation leads to huge accumulation, instead of decay in the landfill and the various natural habitats like rivers and oceans (Thomas *et al.*, 2019). To reduce plastic waste in the landfill, a recycling method is considered as another alternative to manage this material. Recent studies indicate that in Europe alone, over 50% of plastic packaging could be recycled efficiently with today's available technologies (Kawai & Tasaki, 2016). But, in developing countries, there are limitations due to the lack of financial support and the necessary technical knowledge makes it difficult for the treatment of the waste, slowing down the achievements reached in this sector. There are two main categories of recycling: mechanical and chemical. Mechanical recycling is the most common method for the recycling of plastic waste. This process typically includes collection, sorting, washing and grinding of the material, depending on the origin and composition of the waste (Sardon & Dove, 2018). However, highly efficient separation processes, which are a pre-requisite for effective, mechanical recycling, are localized to only a few countries.

Recycling of polypropylene has been widely studied. One of the main applications of recycled polypropylene is its use as a polymeric matrix for the production of composite materials based on natural fibers; the mechanical properties of these materials are similar to those of virgin polypropylene, which facilitates the use of this recycled material (Balam-Cocom, *et al.*, 2006). In addition to recycling, it is important to develop environmentally friendly materials that have suitable fisicochemical properties for industrial use. This is the case of composite materials developed from monolayers of thermoplastic corn starch (TPS) combined with monolayers of polypropylene agafilated with maleic anhydride (PPMA), whose adequate proportion confers to this new material hydrophobic and biodegradable properties that could not be achieved independently (Diaz-Pedraza, *et al.*, 2020). The conversion of plastics to valuable energy is possible as they are derived from a petrochemical source, essentially having high calorific value (Awoyera & Adesina, 2020). Pyrolysis is the process of thermally degrading long-chain polymer molecules into smaller, less complex molecules through heat and pressure under inert condition. It is advantageous compared to mechanical recycling because it does not require sorted plastic to successfully be recycled (Anuar Sharuddin *et al.*, 2016; Dayana *et al.*, 2017). This process requires intense heat with shorter duration and absence of

oxygen. The three major products that are produced are oil, gas, and char. Pyrolysis has been chosen by many researchers since the process is able to produce a high amount of liquid oil up to 80 wt% at a moderate temperature around 500 °C (Ledesma Montaña, 2017). The liquid oil produced can be used in multiple applications such as furnaces, boilers, turbines, and diesel engines without the need of upgrading or treatment (Li *et al.*, 2020). Plastic selected for this investigation was polypropylene (PP), it is widely used in the industry due to lower density, respect other plastics such as high-density polyethylene (HDPE) which has higher hardness and rigidity. Also, PP contributes about 24.3% in the plastic wastes category which is the largest amount of plastics found in MSW (Matias *et al.*, 2020).

This study aimed to carry out the pyrolysis of PP obtained of a mixture of cut sheets of a chair, containers, and bottle cap through fast dynamic at moderate reaction conditions (heating rate 15 °C min⁻¹). Typical reaction conditions of steam cracking, such as very high temperatures, catalysts, and steam, were avoided. The liquid fraction obtained at various intervals (350- 450 °C) was identified and quantified by various analytical techniques, to determine its physical-chemical properties.

2 Materials and methods

2.1 Equipment

The total sulfur content of the liquid fraction of pyrolysis products was determined according to ASTM-D4294 using the Oxford Instruments Supreme-X8000 spectrometer. The elemental analysis was carried out into Vario Macro Cube Elementary Analyzer. The High Heat Values HHV was performed by Method PNE/DPEC/P/MI02 Ref. ASTM D-240, without correction of sulfur content and humidity. The Thermogravimetric Analysis (TGA) was performed using a thermogravimetric analyzer 1 STAR System METTLER TOLEDO. The main components were identified by FT-IR spectroscopy (Perkin-Elmer Frontier spectrometer) in the wavenumber ranging between 4000 cm⁻¹ to 600 cm⁻¹ with a resolution of 4 cm⁻¹.

2.2 Characterization of plastic waste (PP)

The samples of plastic waste were collected from a local recycling plant. Three PP samples were selected: S1 (cut pieces of chairs, containers, and bottle caps), S2 (orange and blue pieces that come from roof decks), and S3 (imported pellets for recycling), to choose PP's best characteristics. PP wastes were conditioned to a particle diameter of approximately 1 mm using an SM 300 cutter mill of the Restch brand for analysis.

The main components were identified by FT-IR spectroscopy (Perkin-Elmer Frontier spectrometer) in the wavenumber ranging between 4000 and 600 cm^{-1} with a resolution of 4 cm^{-1} . Thermogravimetric Analysis (TGA) was performed using a thermogravimetric analyzer 1 STAR System METTLER TOLEDO) was carried out to check the thermal degradation behavior of the feed material and determine the temperature ranges in the reactor. For this, PP samples were thermally degraded at a constant heating rate of 30 $^{\circ}\text{C min}^{-1}$ in a nitrogen atmosphere, 543.1 mmHg of pressure, and 19.5 $^{\circ}\text{C}$. The loss of mass concerning the time of reaction was studied through thermogravimetric curves (TGA), however, it was also possible to study the loss of mass concerning

temperature through differential thermogravimetric curves (DTG).

2.3 Pyrolysis of plastic

The pyrolysis of PP was carried out into a batch type reactor with a capacity of 12 L, with heating by electrical resistances. The sample of 100 g was exposed in a nitrogen atmosphere to displace air in the reactor. Based on the thermogravimetric analysis, the range of temperature was fixed at 350 $^{\circ}\text{C}$ to 450 $^{\circ}\text{C}$, the residence time was 15 minutes (Aboulkas *et al.*, 2010). The temperature and pressure data were taken every 30 seconds for the verification of the heating curve and gas formation. In the reactor the pyrolysis processes were carried out at a rate of heating of 15 $^{\circ}\text{C min}^{-1}$, the vapors produced were condensed at 10 $^{\circ}\text{C}$, the internal pressure of the reactor did not exceed the 100 psi. The liquid products were analyzed by gas chromatography and the gaseous products were collected in a Tedlar bag for chemical analysis. At the end of each experiment, the residue deposited at the bottom of the reactor corresponding to the solid fraction was collected and weighed. The schematic representation of the process is shown in figure 1.

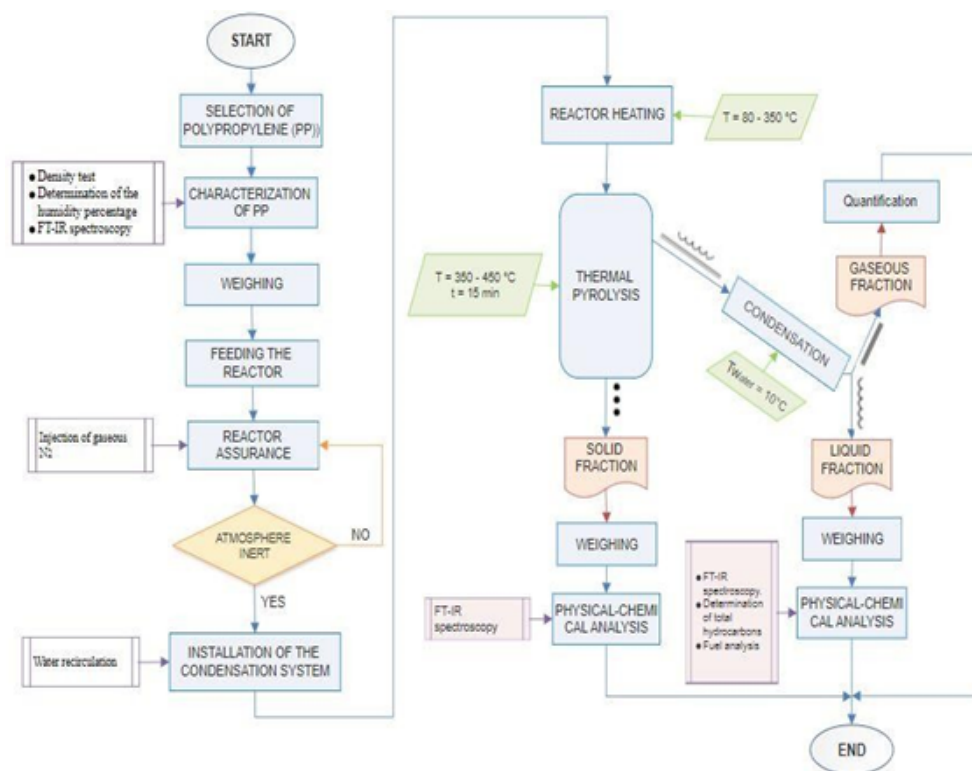


Fig. 1. Schematic representation of the pyrolysis process.

2.4 Characterization of liquid products

2.4.1 Chemical characterization of liquid products

The five samples of liquid products obtained at temperatures between 350 °C to 450 °C were characterized by gas chromatography (GC) Thermo Scientific™ TRACE™ TR-5, with a column of the following characteristics: L 30m/ ID 0.53/ DF 1 μ m, a run time of 21.50 min and flow of helium of 3.0 mL min⁻¹.

3 Results and discussion

3.1 Characterization of plastic waste (PP)

3.1.1 Elementary analysis and High Heat Values of PP

The elemental analysis of the three samples of PP (S1, S2, and S3) evidenced the potential of the use of these residues as raw material for obtaining fuels due to its high carbon content and high calorific value as shown in table 1.

The HHV of PP (44.144 MJ Kg⁻¹) is similar to that of virgin polypropylene (44 MJ kg⁻¹), showing that the effect of additives or other impurities is not significant in the composition of PP samples; besides, they represent an alternative source of energy

comparable to that of conventional fuel such as gasoline or diesel whose average higher calorific power is 45.536 MJ kg⁻¹ (Kalargaris *et al.*, 2017, 2018) as show in table 2.

3.1.2 FT-IR analysis of PP

The infrared spectrum of PP has strong bands close to 2900 cm⁻¹ corresponding to tension movements of bonds C-H and tension movements of C-C close to 1350-1450 cm⁻¹ and flexion movements of -CH₃ between 1200-1000 cm⁻¹, as are shown in figure 2.

Table 1. Elementary analysis and High Heat Values of PP.

% w/w	S1	S2	S3
Nitrogen	0.120	0.100	1.500
Carbon	84.460	85.680	81.440
Hydrogen	15.100	14.037	16.905
Sulfur	0.320	0.183	0.155
HHV (MJ kg ⁻¹)	44.144	45.265	44.627

Table 2. High Heat Value of PP (HHV).

Sample	HHV experimental MJ kg ⁻¹
S 1	44.1440
S 2	45.2654
S 3	44.6271

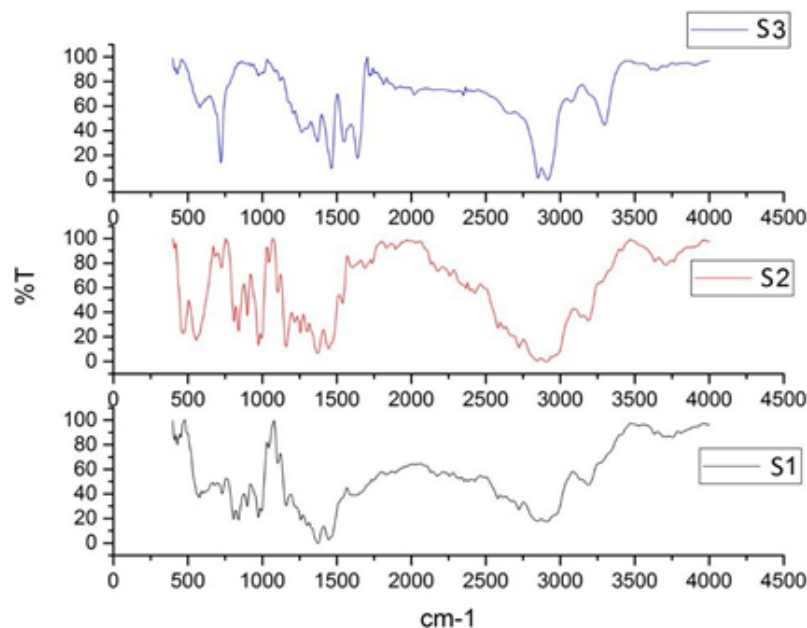


Fig. 2. The infrared spectrum of PP.

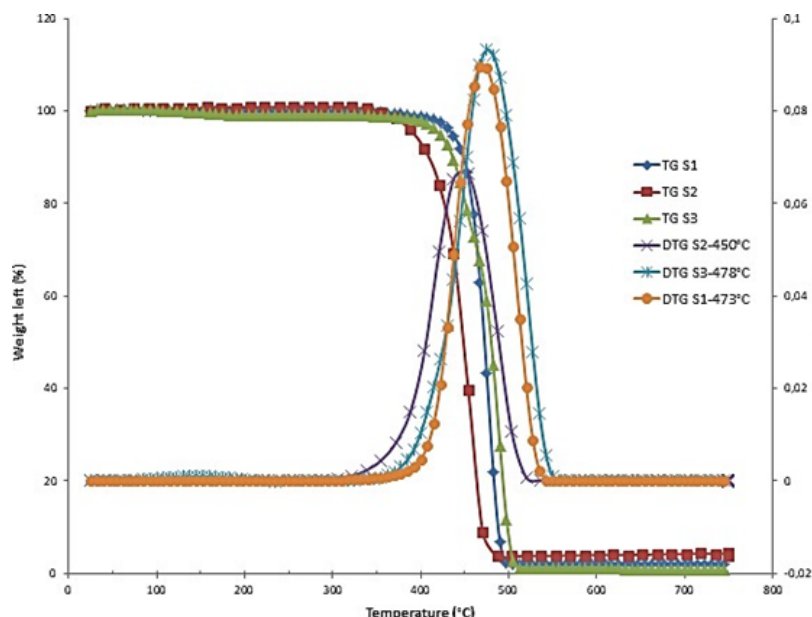


Fig. 3. TGA and DTG curves of PP.

For isotactic PP the intensities of the bands are shown in 998 cm^{-1} . In the case of the S1 sample, the presence of isonitriles and amines is related to the type of polymerization. In the S2 sample, the chloride groups are shown in 1686.57 cm^{-1} , possibly attributed to the UV-327. In the case of the S3 sample, the bands show additives such as alcohols, phenols, and chlorinated acids (Kassargy *et al.*, 2017). Considering that the samples will be subjected to thermal pyrolysis, it is emphasized that in the case of S1 there is no generation of hydrochloric acid; this represents an advantage since the products of pyrolysis are non-halogenated hydrocarbons that can be used as fuels. Based on these preliminary results, it was decided to work with sample S1.

3.1.3 Thermogravimetric analysis of PP

The mass loss curves obtained from TGA experiments with a constant rate of 30 °C min^{-1} are shown in figure 3; these show degradation temperatures between $350\text{--}450\text{ °C}$. This result is consistent with other investigations about the pyrolysis of plastic, in which degradation temperatures are between $447\text{ and }490\text{ °C}$ (Lovás *et al.*, 2017; Matias *et al.*, 2020). The weight losses show that degradation of 98% of the sample occurs in one step, as shown in figure 3; in which DTG curve show only one peak, moreover, no prominent peaks are observed at temperatures lower than 100 °C that indicate the presence of free humidity in this type of material, which is consistent with the other results.

3.2 The yield of pyrolysis products of PP

The influence of the thermal degradation on the performance of pyrolysis products is showed in figure 4, the formation of solid residues is favored at close temperatures of 350 °C and the formation of gaseous products is favored at temperatures close to 450 °C , and the liquid products without wax are favored close to 400 °C .

3.3 Characterization of liquid products

The Figure 5 shows the results of chemical characterization obtained by gas chromatography (GC) for liquid products of pyrolysis taken at different temperature. The range of characterization was between C6 and C40 as a function of temperature. The highest percentage of hydrocarbons obtained in the range of C12 to C20 was at a temperature of 400 °C , while the highest generation of hydrocarbons between C20 to C28 occurs at a temperature of 350 °C . The highest percentage of hydrocarbons obtained in the range of C12 to C20 was at a temperature of 400 °C , while the highest generation of hydrocarbons between C20 to C28 occurs at a temperature of 350 °C . Analysis of the variance showed a significant difference in the liquid fraction at temperatures of 350 °C , 375 °C , and 400 °C , without variability at temperatures above 400 °C .

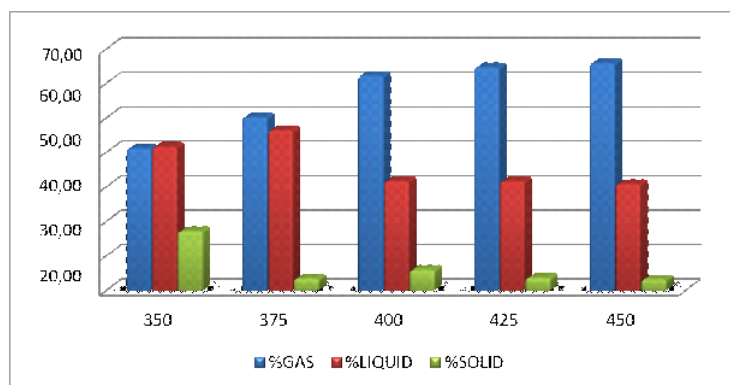


Fig. 4. The yield of pyrolysis products of PP at different temperatures.

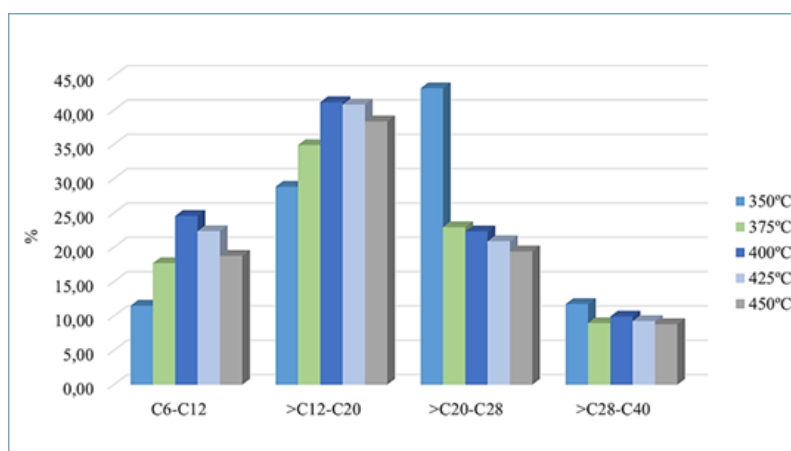


Fig. 5. Chemical composition of liquid fraction.

At low temperatures, there is a 40% conversion of liquids. However, hydrocarbons between C20 to C40 are obtained, which causes them to be partially (375 °C) or completely (350 °C) solidified at the time of cooling. This is why the maximum conversion temperature for the liquid fraction is set at 400 °C with a yield of 32% of hydrocarbons between C6 and C28, which is very close to the results of some investigations (Miandad *et al.*, 2016, 2017)

Taking into account that gasoline, kerosene, and diesel cover a range of C6 to C20, the liquid fraction obtained in this research can be considered as a fuel. However, this hydrocarbon mixture needed to be fractionated for use, the light fraction should be collected for gasoline engines, while the heavy fraction for diesel engines; the most suitable hydrocarbons are in the range of C5-C8 (Rehan *et al.*, 2017).

Table 3 shows some properties of the liquid products obtained respect other investigations show that the isoalkanic structure of the PP is maintained in the thermal decomposition products, so the cetane index is (27.60) the measure of the ignition quality of diesel fuel based on ignition delay in an engine, the higher the cetane number, the shorter the ignition delay and the better the ignition quality. Important for ease of ignition, better starting in cold temperature, reduced engine noise, and to control legislated emissions. Low flash point (19 °C) is the lowest temperature at which vapours will ignite under a low flame; important for safe handling and storage, this value limits the use of this fuel as a pure substance, so blends with fuels such as diesel or bunker must be prepared for engine testing. The very low sulfur content is within international regulations for use as fuel in internal combustion engines ASTM D975, which is novel.

Table 3. Properties of liquid products PP.

Test	Liquid Products	Diesel No. 2 On Road	Unit	Reference
HHV	47103	43000	kJ kg^{-1}	(Moro, 2016, p. 43)
TBP				
10%	106	221.1		
50%	149	265.6	$^{\circ}\text{C}$	ASTM D975
90% 357	329.4			
Cetane Index	27.60	min 40	-	ASTM D975
Flash Point	19	min 52	$^{\circ}\text{C}$	ASTM D975
API a 60°F	38.1	34 - 38	$^{\circ}\text{API}$	ASTM D975
Specific gravity (15.6°C/15.6°C)	0.8343	-	-	-
Density a 15°C	833.9	820 - 845	Kg m^{-3}	(González, 2015, pp. 323, 434)
Sulfur	3	max 500	ppm	ASTM D975
Kinematic viscosity 40°C	1.6	1.9 - 4.1	cSt	ASTM D975
BSW	0.60	max 0.05	%v/v	(NTE INEN 1489, 2012, p. 5)

Conclusions

The pyrolysis of polypropylene was performed in a steel reactor and the effects of the pyrolysis temperature on product yields were investigated. The maximum yields of liquid fraction were obtained at temperatures between 350 to 375 $^{\circ}\text{C}$ and the yield decreases at temperatures close to 400 $^{\circ}\text{C}$ at a heating rate of 15 $^{\circ}\text{C min}^{-1}$. The results showed that the liquid fraction obtained as a result of the pyrolysis of polypropylene at the described conditions has similar physical-chemical characteristics to that of fossil gasoline, even being of better quality because of its ultra-low sulfur content.

Acknowledgements

The authors deeply thank the ESPOCH CESTTA laboratory for the opening to carry out research analysis at its facilities.

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