

Production of Bio-oil from Cigarette Butts and a Circular Economy System

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Abstract

Global population is constantly growing and therefore greater pressure has been placed on natural resources, environment, and sustainability. Circular economy is a concept which when applied in business strategy enables the use of end-of-life materials to produce new solutions, contributing to a more sustainable and efficient economy. Worldwide, cigarette butts are among the most common type of litter. This type of waste represents a global threat to the environment and there are few solutions for its reuse and management. A complete literature review on the circular economy, energy recovery, current recycling solutions of cigarette butts, reverse logistics and municipal solid waste management is carried out to provide a theoretical background for a full understanding of the discussed matters and drive future work. This research aims at filling the literature gap on the development of a solution for cigarette butts valorisation at an industrial scale within a circular economy perspective. The investigation adopted allowed the development of a practical solution for the valorisation of cigarette butts: the production of bio-oil and a logistics solution to integrate the collection of this type of waste in the municipal waste system, thus avoiding environmental pollution and facilitating its reuse.

Keywords: Sustainability, Circular Economy, Valorisation, Methodology, Bio-oil

1. Introduction

Given the recent trends in population growth and the predictions about its evolution over the next decades (UN, 2019), understanding the extent to which demographic changes may affect the prospects of sustainable development is a priority. It is important to understand the main habits of the population and their consequences for the environment.

Environmental pollution is not a new phenomenon, yet it remains the world's greatest problem facing humanity, and the leading environmental causes of morbidity and mortality (Ukaogo, 2020). One form of waste that has been brought to the public's attention in recent years is plastic waste.

Since the 1950s, billions of tons of plastic have been produced with only a small fraction of this being recycled. Because of this, huge quantities of plastics end up in oceans, leading to ecological disasters (Tiseo, 2020). The world's most littered plastic item is cigarette butts. Cigarette filters are made of plastic called cellulose acetate. This type of waste has a severe impact on the environment, not only because of the plastic but also the nicotine, heavy metals, and many other chemicals. Cigarette butts accumulate due to the poor biodegradability of the cellulose acetate filter, and all the toxicity threatens human life, marine ecosystems and the environment (Kadir, 2015). With the increasing concern arising about landfills toxic incinerator emissions,

there is a critical need for an alternative method for cigarette butts waste disposal which is sustainable and resilient. Based on the circular economy's principles it is possible to create value-added alternatives for the reuse of cigarette butts, while reducing resources used, and the waste and leakage created and helps to reduce environmental pollution. The energy sector is undergoing major changes and developments to find new sustainable ways of producing energy. Energy production from waste (particularly plastics) is being explored and some processes such as anaerobic digestion, pyrolysis, hydrothermal liquefaction, and gasification are proving to be viable alternatives (Foster et al., 2021). This Dissertation aims at filling the literature gaps on the development of an integrated process of cigarette butt's valorisation at an industrial scale by adopting a circular economy strategy. The goal is to develop a form of valorisation and logistics scenarios that enable the creation of a structured supply chain for this waste.

2.Literature Review

During the past two decades, tobacco use has dropped from 1.397 billion in 2000 to 1.337 billion in 2018 (World Health Organization, 2019), even so tobacco is still considered an epidemic and one of the greatest public health threats the world ever faced, killing around 8 million people a year (World Health Organization, 2020). Even though there is a lot of information these days about the consequences of smoking, according to Euromonitor estimation in 2016, still 5.5 trillion cigarettes were consumed worldwide (Elflein ,2019). Figure 1 represents the number of tobacco smokers among those 15 years and older worldwide from 2000 to 2025 by region. First of all, it is noticeable a constant decrease during the period between 2000 and 2020, as well as it is expected to maintain the pattern until 2025.

According to two separate reports, the ratio of littered butts to the cigarette consumption was about 76% in 2013 (Patel et al., 2013) and 84% in 2015 (Lee and Lee, 2015), indicating the effect of smoker's behaviour

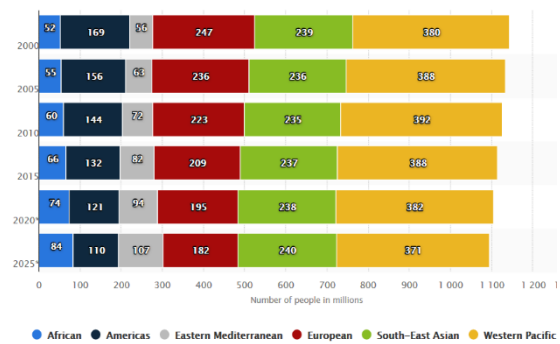


Figure 1- Number of tobacco smokers among those 15 years and older worldwide from 2000 to 2025-Statista October 2020

on the amount of these litters. Cigarette butts are commonly found in coasts, roads (Haseler et al., 2018), aquatic and sea environments (Becherucci et al., 2017), urban and public areas (Chevalier et al., 2018). It is recognized as one of the most common and epidemical wastes (Pon and Becherucci, 2012) and contributes to the largest number of wastes in the world (Micevska et al., 2006). Many studies have been carried out with the purpose of finding effective management solutions for this waste. Conventional methods such as landfilling or incineration are neither universally sustainable nor economically feasible for this purpose (Bandi et al., 2018; Mohajerani et al., 2016).

Recycling cigarette butts is difficult because there are no easy mechanisms or procedures to ensure efficient and economical separation of the butts and appropriate treatment of the entrapped chemicals (Mohajerani et al., 2016). According to Puls et al., 2011, cellulose is degradable thanks to organisms that utilize cellulose enzymes. At the other hand, Haske-Cornelius et al., 2017, state that cellulose acetate-based cigarette filters do not biodegrade under most circumstances because of their compressed make up and the presence of acetyl molecules. Conclusions demonstrate that nicotine poses an important risk to aquatic organisms. Cigarette butts littered on streets commonly get washed away into stormwater drains and end up on beaches, or in rivers and harbours, leaching toxic chemicals (Araújo and Costa, 2019).

Ultimately, circular economy refers to the idea of extending the useful life of products, materials, or resources (Gregson et al., 2015). A central topic in the concept of circular economy is the use of resources within closed-loop systems, reducing pollution or avoiding resource leakage while sustaining economic growth (Winans et al., 2017). A literature review of possible butt recovery solutions was developed and can be concluded that there is some research in this field. Some papers were selected as good approaches to the problem: **Fired clay bricks** (1) Mohajerani et al., 2016; **Sound porous absorber** (2) Maderuelo-Sanz et al., 2018; **Blocks for paving** (3) Wadalkar et al., 2018; **Superhydrophobic adsorbent** (4) Xiong et al., 2018b.

Biochemical and thermochemical waste-to-energy technologies can exploit the energy content of municipal solid waste, thus replacing fossil fuels and diverting waste from landfills (Maria et al., 2018). This solution could be explored using cigarette butts since cellulose acetate (main filter component) is a very good source of organic carbon and can be converted into high valued liquid products by suitable thermochemical conversion methods. Pyrolysis and hydrothermal liquefaction are two possible solutions for energy recovery through waste. Both processes are used approaches to convert biomass or organic wastes into liquid bio-oils or other fuels and value-added chemicals (Araujo, 2018; Baloch, 2018; Li, 2015; Lian, 2017).

To achieve a process that allows energy recovery using cigarette butts it is necessary to have a logistics system that enables the creation of a closed loop supply chain. In fact, proper reverse logistics management is related to many different measures which are implemented in the supply chain (Alshamsi and Diabat, 2015). This concept plays a key role in promoting an environmentally friendly operation, since with the collection and reuse of disposed products, the generation of new waste is avoided, as well as its incorrect disposal in the environment (Guarnieri et al., 2020). Uncertainty could be found in market

demand, processing capacity of each facility, return quantity of the recycled materials, quality variation of the return flow and relevant cost parameters (Yadollahinia et al., 2018), and for this reason it is important to have a resilient model to support decisions to be made in supply chain management.

3. Methodology

Figure 2 provides an overview of the research. The methodology of this work will be divided into two parts: laboratory research to develop a practical solution for cigarette butt's valorisation and an analysis of a logistic process and development of an optimization model to minimize the costs of a company that will recycle cigarette butts according to the solution found in the laboratory.

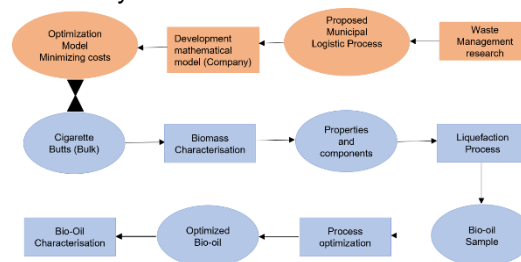


Figure 2- Research Overview

Laboratory Research:

It was followed the laboratory research overview presented in figure 3. Initially cigarette butts were characterized through analyses such as Fourier transform infrared (FTIR) spectroscopy, Thermogravimetry (TGA) and Gel permeation chromatography (GPC). After this initial characterisation stage, the liquefaction reactions were carried out. The liquid obtained from the reaction was subsequently filtered to produce bio-oil and later characterized, using the techniques mentioned above and also elemental analysis to obtain higher heating values for each bio-oil.

Materials and Reagents

The liquefaction was performed in a 2 L reactor plugged with an electric stirring motor, a temperature controller and a Dean-Stark mounted on a heating blanket. A sample of virgin filters was used for the first

experiment and used filters for the second and third experiments. As for the solvent and catalyst, was used 2-ethylhexanol (2EH) and *p*-toluenesulfonic acid (PTSA) respectively.

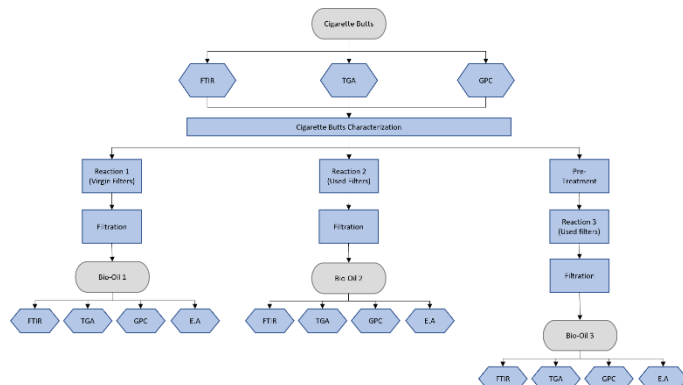


Figure 3-Laboratory research overview

Cigarette butts' characterization

A virgin and used filter were characterized using three different analyses. The **FTIR-ATR** analysis was performed using a *Spectrum Two-PerkinElmer*. The wavenumber was ranged between 600 and 4000 cm⁻¹. The results were treated through the *Perkin Elmer Spectrum IR* software. Then a **Thermogravimetric (TGA)** analysis was performed to study mass degradation with temperature variation. The heating programme consisted of an initial holding for 3 min at 30°C, then a ramp at 5 °C per min from 30°C to 240 °C, followed by an isothermal at 240°C for 5 min and finally a ramp at 5 °C per min from 240 °C to 900 °C. It was used a *Hitachi-STA7200* apparatus. Finally, the **Gel Permeation Chromatography (GPC)** allowed to obtain the molecular weight distribution of the bio oils. It was used a set which include a *JASCO* pump, a column, a UV and refraction detector.

Bio-oil production- Liquefaction process

The operating conditions for the liquefaction was set at 160°C for a reaction time of 4 hours, a solvent/biomass ratio of 5:1 and a mass of catalyst equivalent to 3% of the sum of feedstock and solvent. In the first experiment virgin filters were used and a simple pre-treatment of splitting filters into small pieces was performed. In the second experiment, the same pre-treatment was

used, however used filters were fed as biomass. In the third experiment, the pre-treatment involved the removal of the wrapping paper of the filter and the splitting of the filter into small pieces. The liquefaction process was followed by a filtration process to separate the liquid and solid products. After the cleaning step with acetone, the solids were stored in an oven at 100°C for over 24 hours. The reaction yield was calculated according to the Equation 1.

$$(1) \text{ Liquefaction Yield (\%)} = \left(1 - \frac{Mr}{Mi}\right) \times 100$$

Where *Mr* is the mass of the residue and *Mi* is the initial mass of the biomass.

Bio-oil Characterisation

After Bio-Oil Production, characterisation was carried out through **FTIR**, **TGA** and **GPC**, using the same method and technology described above in the characterisation of virgin and used cigarette butts. After the solvent extraction, an analysis was held to obtain values of higher heating values (MJ/KG).

Logistic Process

The solution found for the valorisation of cigarette butts will always depend on the availability of biomass for the process. For this reason, it is essential to analyse the management of cigarette butts at the municipal and district level and to optimize the interactions between municipalities, districts, and a (fictitious) recycling company.

Municipal cigarette butts waste management proposal

Based on the Law no. 88/2019 (Diário da República n.º 168/2019, 2019), conditions must be created so that it can be effectively enforced, foreseeing where cigarette butts must be disposed and who is responsible for doing so. Therefore, it is suggested for the management of cigarette butts the incorporation of (removable) cigarette butts' containers in the waste containers available in the streets of the municipalities. A 3D model was created in SketchUp program, figure 3, to make the proposal clearer.

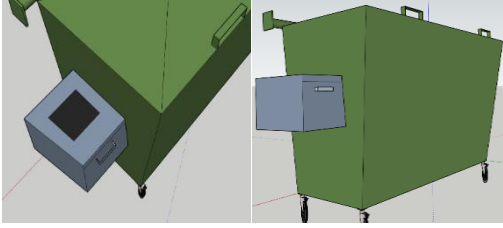


Figure 4- 3D model of cigarette butt's container developed in Sketchup

Therefore, this proposal makes available to all population a specific place for the disposal of this waste, making its collection and processing more efficient. This proposal also suggests the existence of a specific place inside the municipal waste trucks for waste workers to deposit butts and prevent it from mixing with normal municipal waste.

The implementation of this proposal for the collection of cigarette butts at the municipal level allows for the existence of a company dedicated to its valorisation. Therefore, a company will be able to access the biomass to produce bio-oil by purchasing cigarette butts from different districts that integrate the waste from the respective municipalities. An implementation scheme of the proposal is presented in figure 5.

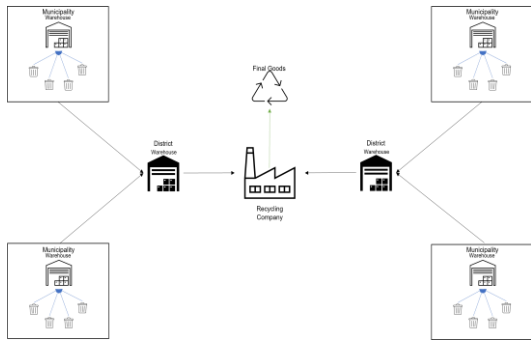


Figure 5- Implementation scheme of the proposal

Recycling company logistics optimization

The recycling company will have to perform an analysis to decide how much biomass and from which district it should buy the cigarette butts, considering the demand for the final product, the price of access to the biomass, and the transportation costs. A optimization model was developed to solve this case at the strategic and tactical level, considering: Fixed locations of the district warehouses, Fixed locations of the recycling

plants, Implementation costs of each plant, Demand by the different markets, Distance associated with moving the Bio-oil from the plant to the markets, Distance associated with moving cigarette butts from the warehouse to the plants, Maximum capacity of each warehouse, Maximum capacity of the recycling plants, Yield of recycling process, Production cost of Bio-oil and Cost of biomass from the different districts. The model intends to support the optimal values for the Flow from each warehouse to each plant and the Flow from each plant to each market, the decision of opening the recycling plant or not and also the location of the plants (Lisbon vs Porto). An objective function was defined to optimize the problem:

$$(2) \text{Min} \sum_s \text{probs} \sum_k \sum_i P_k \times XW_{kis} + \sum_s \text{probs} \times CT \left(\sum_k \sum_i Distw_{ki} \times XW_{kis} \right) + \sum_s \text{probs} \times CT \left(\sum_i \sum_m Distw_{im} \times XP_{ims} \right) + \sum_s \text{probs} \sum_m CP \times Demand_{ms} + \sum_i CF_i \times Y_i$$

Subjected to the constraints:

$$(3) \sum_i XW_{kis} = \sum_s \frac{Demand_{ms}}{\eta} \times Y_{i,s} \quad \forall k, m, i$$

$$(4) \sum_k XW_{kis} \leq CPW_k \quad \forall k, s$$

$$(5) \sum_i XP_{ims} \leq CPP_i \times Y_{i,s} \quad \forall i, s, m$$

$$(6) \sum_i XP_{ims} = Demand_{ms} \quad \forall m, s$$

$$(7) \sum_s Y_{i,s} \geq 1 \quad \forall i$$

$$(8) \sum_i XW_{kis} \times \eta = \sum_i XP_{ims} \quad \forall k, s, m$$

Where, Sets: K: set of k warehouses (each per district) where the cigarette butts are stored $k \in K, k = 1, \dots, |K|$, I: set of i Recycling plants $i \in I, i = 1, \dots, |I|$, S: set of s scenarios, $s \in S, s = 1, \dots, |S|$, M: set of markets $m \in M, m = 1, \dots, |M|$. Parameters : probs - probability of each scenario, CPW_k - Warehouse capacity, CPP_i - Plant capacity, $Dists_{im}$ - distance from plant i to market m, in

km, Distwki - distance from warehouses k to plants i , in km, CT- transportation cost, P_k - Price/ton of cigarette butts from district warehouse k , Demand_{ms}- Demand of market m in scenario s , in ton, η - Bio-oil Process Yield and CP - Bio-oil Production cost. Decision variables: XW_{kis} flow from warehouse k to plant i in scenario s , in ton, XP_{ims} flow from plant i to market m in scenario s , in ton and $Y_{i,s}$ - variable that assumes value 1 if plant i is open and 0 otherwise, in each scenario s . Some data was collected, and some estimations were made to define values for: plants capacity 204 ton/year, (Biofuels Canada, 2010), Fixed costs of plants- Plant 1- Porto- 161 M€ and Plant 2- Lisbon-180 M€, (INE, 2021), Distances, Price of the biomass- 239 €/ton (Maalouf et. al., 2019), Warehouse capacities (Qamar, 2020, World Population review, 2021, Forey et.al., 2015), production costs- 4620 €/Ton, Labour costs-2600€/Ton (Rogers and Brammer, 2012) and Electricity and Operating costs- 8664 €/Ton.

4. Results

Laboratory Research

In terms of Process yield, **Bio-oil 1**- 88,3%, **Bio-oil 2**- 56.1% and **Bio-oil 3**- 98,15%. With these results it is possible to say that the pre-treatment of the cigarette butts is crucial since, using the same type of biomass and the same experimental conditions, completely different yields were obtained: 98.15% for bio-oil 3, which underwent a more aggressive pre-treatment (removal of the paper around the filter) and 56.1% for bio-oil 2, which had a pre-treatment that only involved cutting the filter into smaller pieces.

Cigarette Butts Characterization

In figure 6, the cellulose acetate (CA) composition of the cigarette filters can be identified by the acetyl groups characteristic bands at 1240, 1370, and 1750 cm^{-1} wavenumbers. Hence, it can be observed that the FTIR spectra of the virgin filters and used filters are practically identical. It is important to note that the FTIR results indicate that the contaminants in the cigarette butts are in trace amounts and

therefore there are no major changes in the chemical structure of the used and virgin filters.

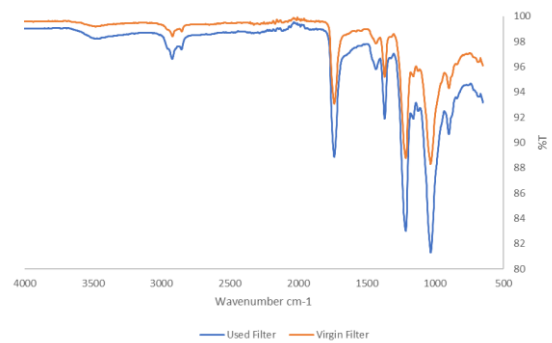


Figure 6-Transmittance vs wavenumber in Used and Virgin filters

As figure 7 shows, the initial mass loss up to 100 °C was mainly due to water and solvent evaporation (Banyasz, 1999). The second mass loss region, between 100 °C and the 240 °C holding temperature, resulted in a further mass loss of ca.15%. This region may be accounted for by the evaporation of glycerol, nicotine, and some volatile chemicals from the tobacco. The initial thermal decomposition of some biopolymers such as sugars and pectin might also contribute to mass loss in this temperature range (Wang et al., 2009). Isothermal holding at 240 °C continued to reduce the mass to a small extent. Up to approximately 350 °C. In the temperature region above 240 °C, there was a slightly faster rate of mass loss. The three main components of common biomass e hemicellulose, cellulose, and lignin- each display a well-defined thermal decomposition region: hemicellulose starts to decompose between 220 and 315 °C, followed by cellulose at temperatures close to 400 °C, and lignin at temperatures above 400 °C.

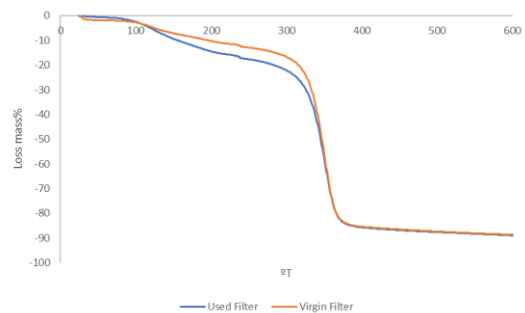


Figure 7- TGA Curves for virgin and used filters

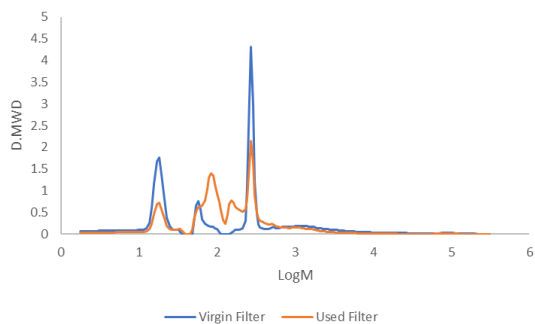


Figure 8- Distribution of the average molecular weight of the filter samples

From figure 8 it is possible to obtain the average molecular weight distribution of each filter sample. It was also possible to extract from the software the data concerning the average numerical and mass molecular weights (Mn and Mw respectively) as well as their polydispersity index, which can be seen in the following table.

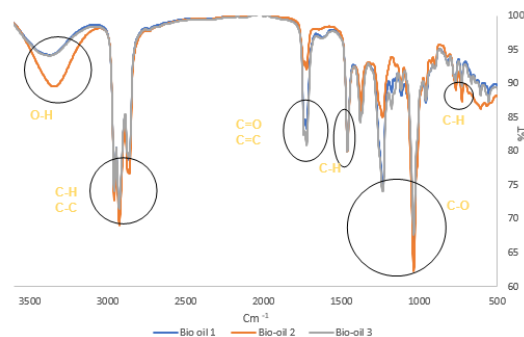
Table 1- GPC results for filter samples

Sample	Mn (g/mol)	Mw(g/mol)	PDI
Virgin Filter	1263.3	49505.8	39.2
Used Filter	538.7	29370.8	54.5

Liquefaction products characterisation

It is interesting to note in figure 9 a band with transmittance at 3400 cm^{-1} corresponding to O-H bonds, this due not only to the presence of water in the sample but also phenolic groups and alcohols. The bio-oils 1 and 3 present higher conversion rates (88.5% and 98.15%) and therefore the transmittances are identical at this wave number (3400 cm^{-1}), in contrast with the bio-oil 2 that presents a more intense band, indicating a greater presence of water, phenols and alcohols, something that is in accordance with the low conversion that had, about 56.1%. Considering then the wavenumbers of the order of 3000 cm^{-1} , a broad band of considerable transmittance was detected corresponding to alkane molecules derived from the fragmentation of polysaccharide constituents. Between 1600 cm^{-1} and 1730 cm^{-1} two peaks equivalent to the stretching of C=O and C=C bonds were noted, with bio-oil 3 showing the longest bands, resulting from higher concentrations of ketones, aldehydes, carboxylic acids, and olefins.

The C-H bond transmittance bands, located approximately at 1460 cm^{-1} , prove that a more efficient liquefaction results in a better fragmentation of the lignin polymer chain and consequently in a higher concentration



of aromatic compounds in the liquid phase.

Figure 9-FTIR transmittance spectrum Of Bio-oils

Finally, it is important to highlight the peaks referring to the minimum values of transmittance at about 1030 cm^{-1} , equivalent to C-O bonds indicating a large influx of aromatic compounds and alcohols in the composition of the bio-oils. It is worth mentioning that the highest band at this wave number and adjacent ($660\text{--}800\text{ cm}^{-1}$) does not belong to the bio-oil corresponding to the liquefaction with the highest conversion, but rather relative to bio-oil 2, which recorded low conversion value (56.1%).

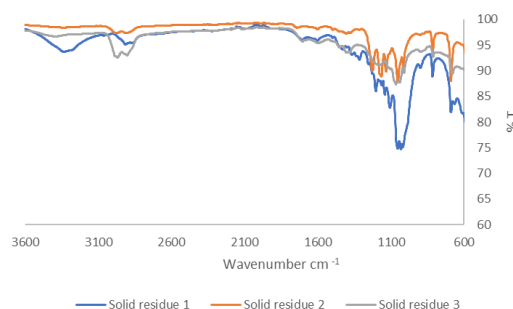


Figure 10- FTIR transmittance spectrum of the solid residues

Comparing the spectrum for solid residues (Figure 10), the bands are in approximately the same wavenumber ranges as for the bio-oils, indicating, as would be expected, much lower transmittance values. Again, higher conversions provide in general higher transmittance peaks. Some C=O are found (ketones, carboxylic groups, and aldehydes) but the great predominance resides, as

before in the C-O bonds, indicative of the presence of aromatic groups resulting from the depolymerization of lignin, with the maximum and minimum representations for the products of Bio-oil 1 (88% conversion), and 2 (56%), respectively. It will be interesting to point out the fact that the solid residues that have the most similar bands are not the ones with highest yields. It should be noted that solid residues 2 and 3 show similar bands and both are the result of a reaction with used filters despite the large difference between yields (56% and 98%) respectively.

Table 2- GPC Results for bio-oil samples

Sample	Mn (g/mol)	Mw(g/mol)	PDI	Yield
Bio-oil 1	2574.7	13632.0	5.3	88.3%
Bio-oil 2	805.1	4566.5	5.7	56.1%
Bio-oil 3	2790.8	17543.6	6.3	98.15%

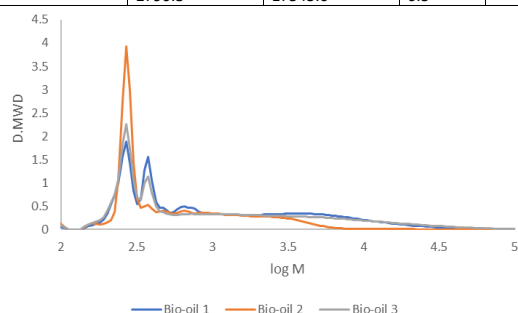


Figure 11-Distribution of the average molecular weight of the bio-oil samples

Based on these results it is possible to state that the value of Mw and Mn for bio-oil 2 is negligible, having been obtained because of some experimental error. In the production of bio-oil 2 the same biomass was used as in bio-oil 3 and for this reason the order of magnitude of Mw should be the same as of bio-oils 1 and 3. Moreover, given its low yield the Mw value should be the highest, indicating that there were not as many bond breaks as in the other bio-oils, and therefore the presence of compounds with higher values of Mw. For the bio-oil 1 there is a substantial reduction of the Mw (13632.0 g/mol) when compared with the biomass used for its production (49505.8 g/mol) about 1/4 of reduction of the value, something expected, due to the breaking of bonds of larger compounds during the process of liquefaction, thus giving rise to compounds of smaller dimensions and therefore with lower Mw. The same happens and for the same reason with the bio-oil 3, there is a reduction of about half of the Mw of the

biomass used (29370.8 g/mol) when compared with the Mw of the bio-oil (17543.6 g/mol), something also expectable due to the explanation given above. The greater reduction of Mw observed in bio-oil 3 (about half) when compared with the reduction in bio-oil 1 (about a quarter), is explained by the difference in yield, being this in bio-oil 3 of 98.15% and 88.3% in bio-oil 1. This means that the greater the yield, the greater the number of breaks in the bonds of the

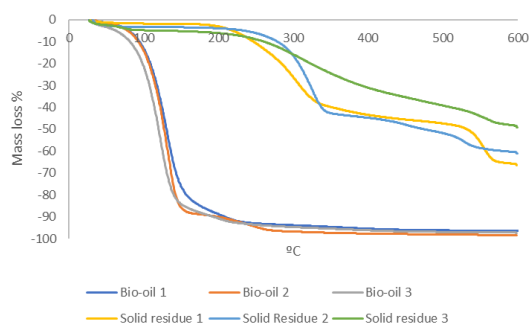


Figure 12- TGA results for the bio-oils and solid residues

compounds and therefore greater presence of compounds with lower Mw. The TG (Figure 12) and DTG (Figure 13) results of the pyrolysis (in N₂ atmosphere) of bio-oils using a heating rate of 5°C/min are shown in figure 28 and 29, respectively. The bio-oil undergoes thermal decomposition due to the absence of oxygen and a notable mass loss is observed between 70 °C and 140 °C. This region is characteristic of the elimination of water - which forms a major portion of the bio-oil - and removal of acids, alcohols, and other aromatic oxygenate such as mono-phenols and furans. DTG peaks of bio-oils 1,2 and 3 are observed in this region at temperatures of 134 °C, 136°C and 123 °C, respectively. At temperatures between 200 °C and 350 °C, the decomposition of mono-sugars such as levoglucosan and poly-sugars such as cellobiose takes place as reported (Perez et. al., 2017).

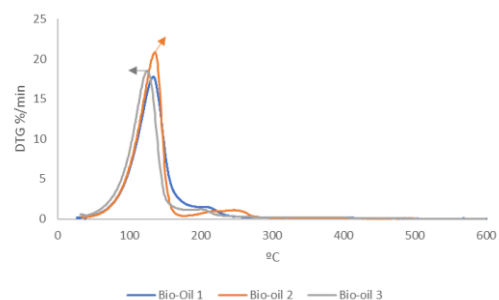


Figure 13 - DTG Results for Bio-oil samples

warehouses and plants and between plants and markets

Table 5- Variable Costs

	Scenario 1		Scenario 2	
	€	%	€	%
Transportation Costs W-P	11542	0%	6055	0%
Transportation Costs P-M	333657	9%	156460	8%
Biomass Costs	100165	3%	54245	3%
Production Costs	3231672	88%	1750128	89%
Total Costs	3677036		1966887	

5. Conclusion

Cigarette consumption is not expected to decrease, and it is also not expected that consumer behaviour will change in the coming years, cigarette butts will remain as one of the waste products with the greatest impact on the environment. Cigarette Butts waste management is almost non-existent worldwide, and this represents a serious problem for the environment, since plastic, the main constituent of the cigarette filter, takes many years to degrade and the chemicals are highly toxic. The development of this research shows, even if in an embryonic stage, it is possible to have a viable solution for value creation through cigarette butts. The laboratory results of the bio-oils produced are very encouraging, insofar as the yield of the most optimised process was around 98.15% and the higher heating value of 33.287 MJ/KG is, within the literature, well above the average of the bio-oils produced worldwide. However, it is necessary to stress once more that the valorisation solution will only have an impact if a different management of the cigarette butts is done, and a valid proposal was also presented during the development of this work. This research may represent great advances in the resolution of the problem characterised and contributes fully to sustainability and circular economy.

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