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Dissolution and recovery of waste expanded polystyrene using alternative essential oils

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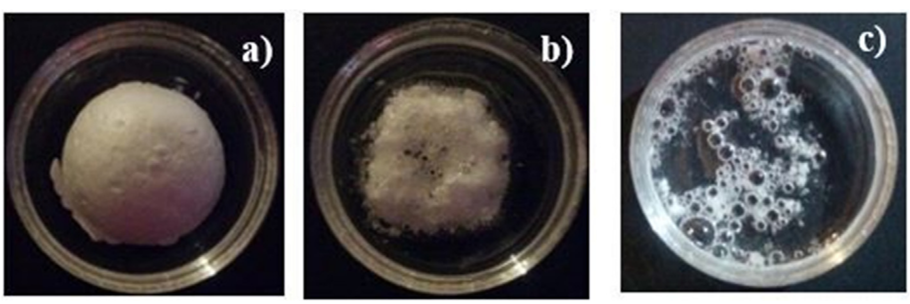
GRAPHICAL ABSTRACT

An efficient method to dissolve and recover waste expanded polystyrene with alternative natural oils.



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ABSTRACT

The disposal of expanded polystyrene contributes to the current pollution problem. To deal with such issues, there are different recycling methods. However, most of these options are expensive and environmentally unfriendly.

Herein, we report an alternative, environmentally friendly and cheaper recycling process for recovering expanded polystyrene at room temperature based on commercial essential oils used without further purification. Full dissolution of the polystyrene was achieved with a maximum ratio of 1:1 wt%. This dissolution time is at least four times faster than with limonene. Pure waste expanded polystyrene (WEP) was recovered with methanol and finally dried. All the essential oils were recovered and used again. The raw materials: WEP, the essential oils, and the recovered WEP were characterised using infrared spectroscopy, X-ray powder diffraction and thermal gravimetric analysis.

1. Introduction

The use of existing and widely commercialised polymers has become a disposal problem due to the time they take to decompose naturally and the impact they have in the environment during the degradation process. Several methodologies and actions have been

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proposed to tackle this problem, ranging from the development of biodegradable materials to more sustainable recycling methods [1].

Some of the methods used to manage plastic waste include: land-filling, incineration, chemical recycling and mechanical processes [2,28–30]. The disadvantage of land-filling is the non-degradability or slow degradation rate of the plastics. With incineration, toxic emissions as dioxins, furans, mercury and polychlorinated biphenyl are produced during the process [20]. Mechanical recycling allows reuse of the plastic solid waste to form a new product with the same inherent characteristics [1,3,4]. On the other hand, methods that include dissolution processes are among those proposed as the most environmentally friendly and profitable [5].

Expanded polystyrene (EPS) is made of 98% air and 2% polystyrene. According to the Society of the Plastics Industry (SPI) is a Type 6 polymer. Because of its mechanical properties, chemical stability, thermal insulating ability, simple manufacturing and low cost, it is one of the most widespread polymers around the world. It is mainly produced from styrene obtained from petroleum. Polystyrene is used in many applications such as disposable containers and packaging materials, and causes severe environmental problems due to its high consumption and low recyclability [6,23]. In 2015, 6200 kt of expanded polystyrene was produced, and just around 40% was reused [7,25]. One methodology for polystyrene recycling is its dissolution in organic solvents, thereby solving the low density problem by achieving a volume reduction of more than 100 times. However this is still an expensive and not environmentally friendly process. The solvents most commonly used are toxic (toluene, xylene, benzene, chloroform, acetone, cyclohexane, butyl acetate, ethyl acetate and methyl ethyl ketone) [8–11,19]. The natural oils are composed mainly of terpenes, sesquiterpenes and aromatic compounds. Terpenes play roles in the defence mechanisms of plants and to attract insects for pollination. Terpenes possess isoprene subunits; of which about 30 000 different ones have been identified in roots, rhizomes, stems, leaves, blossoms, fruits and seeds for a variety of plants. These include conifer wood, balm trees, citrus fruits, coriander, eucalyptus, lemon grass, carnation, caraway, pepper-mint species, rosemary, sage and thyme, among others. Terpenes are the cause of the well-known pleasant smells, spicy tastes, and pharmacological properties that the aforementioned plants exhibit [12,13,18,26]. Limonene, the principal component of the essential oils found in citric fruits, is allegedly the most investigated terpene [27]. Limonene dissolves the same amounts of waste expanded polystyrene (WEP) as some organic solvents, but has the problems of low yield and high cost of extraction. The use of alternative natural oils is a promising and environmentally friendly alternative for recycling WEP [24]. It can be considered a green process because CO_2 emissions are reduced compared to other methods, and offers the opportunity to re-use the material; although this is still a developing area [21,22].

Herein, we report a simple and inexpensive method for recycling polystyrene, using commercial natural oils (used without further purification) to dissolve WEP. We also discuss the maximum dissolution concentration and time for each of the different oils studied. Both WEP and natural oils were recovered and characterised using a variety of techniques. The sources of the natural oils studied were: star anise, chamomile, thyme and eucalyptus. These oils contain: anethole and estragole (present in star anise); thymol, myrcene, ocimene and linalool (present in thyme); α-pinene, 1,8-cineol, p-cymene, β-pinene, a-terpineol and linalool (present in eucalyptus) and bisabolol, farnesol, caryophyllene, myrcene, linalool, geraniol, carvone and cineol (present in chamomile). This information was provided in the technical data sheet provided by the oil supplier.

2. Materials and methods

2.1. Materials

The WEP collected was waste packaging. The essential oils (from star anise, eucalyptus, thyme and chamomile) were purchased from a local drugstore (Botica la Moderna ®) and used without further purification. The selection of the oils was according to their price and availability. The goals were to find an accessible method for WEP recovery and recycling that would be cheaper and faster than with limonene [6], and would avoid the use of organic solvents. The properties of the natural oils were provided by the supplier.

Star Anise Oil: Density 0.9866 g/cm^3, flashpoint 90 °C. Trans-Anethol 72.1%, Cis-Anethol 11.6%, Estragole 16.3%. Eucalyptus Oil: Density 0.9114 g/cm^3, flashpoint 53 °C. 1.8 Cineol 79.9%, Limonene 7.4%, α-Phellandrene 1.0%, α-Pinene 1.4%, β-Pinene 0.4%. Thyme Oil: Density 0.9236 g/cm^3, flashpoint 63 °C. Thymol Carvacrol Chamomile Oil: Density 0.9872 g/cm^3, flashpoint 252 °C.

Methanol and isopropanol were purchased from Sigma-Aldrich Co., and used without further purification.

2.2. Dissolution of expanded polystyrene

For each experiment, 1 g of essential oil was placed in a test tube, then different amounts of expanded polystyrene (10–100 wt%) were added and the time to complete dissolution noted. All experiments were carried out at room temperature.

2.3. Recovery of polystyrene

To recover dissolved polystyrene, 1 mL of methanol was added to the mixture and stirred for 2 min. After this time, a sticky white solid precipitated, was filtered and then was placed in 1 mL of fresh methanol. Next, it was washed three times with 1 mL of isopropanol. Finally, the white solid was squeezed between layers of filter paper and let dry at room temperature.

2.4. Recovery of essential oil

The filtrate was placed in a rotary evaporator at 65 °C to remove the alcohols. The remaining residue was the essential oil which was recovered for further use.

2.5. Characterisation

Infrared (IR) data about the solution was collected utilising an IR Shimadzu® spectrometer, with attenuated total reflection (ATR) accessory. Thermal gravimetric analysis (TGA, model Netzsch STA 449 F3 Jupiter) was carried out in the range 25–300 °C with 10 °C/min ramp.

The recovered polystyrene was characterised using a Rigaku Ultima IV X-ray Powder Diffractometer equipped with CuKα radiation. Measurements were taken using a step size of 0.5° and a scan rate of 2° per minute. Scanning electron microscopy (SEM, JEOL JSM-6510lV) was used to analyse the surface morphology.

3. Results and discussion

As mentioned before, the sources of the natural oils used were: star anise, chamomile, thyme and eucalyptus. The latter had been used in previous reports and herein, was used just as a reference oil.

Dissolution assays were carried out in test tubes, in which small blocks of WEP were immersed in each of the different natural oils (Fig. 1).

All the natural oils successfully achieved full dissolution of WEP at room temperature. The maximum dissolution concentration was 1 g WEP per gram of natural oil, which is a higher concentration than reported for organic solvents (Table 1) [6]. The times for complete dissolution of WEP at different concentrations (10 to 100 wt%) are presented in Fig. 2. The results show that star anise oil is the best solvent, being the fastest at dissolving WEP at all different concentrations tested. Even though chamomile was faster than eucalyptus and thyme at low

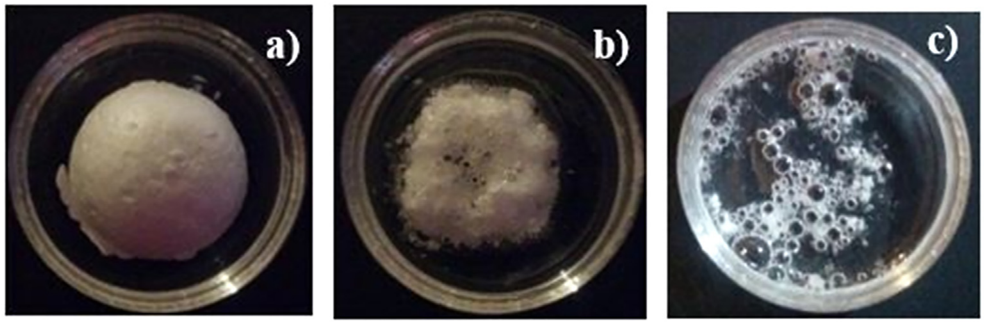

Fig. 1. Dissolution of WEP at room temperature by essential oil: a) Before essential oil, b) After 1 min and, c) After 2 min.

concentrations, above 45 wt%, eucalyptus became faster. However, all these times are faster than d-limonene (800 s) at 10 wt% [6].

The WEP was recovered from the solution in the beginning as a white sticky precipitated by adding methanol. Then it was rinsed with isopropanol to take out the remaining oil and dried at room temperature to finally get a white solid mass. The WEP recovery was fastest from the eucalyptus solution, followed by thyme. In the cases of chamomile and star anise, it took more time or required more methanol.

All the natural oils, WEP, natural oil with dissolved WEP, and recovered WEP were analysed using IR spectroscopy (Fig. 3 and Table 2). The IR spectrum shows that all the natural oils can be recovered in pure form at the end of the process, and that they can be reused again for WEP dissolution (Fig. 4 and SI). The IR spectrum of the recovered WEP indicates that only WEP is present; thus, all the oil and alcohol was removed during purification.

The recovered WEP was analysed using scanning electron microscopy (SEM); the micrographs show an irregular and porous surface (Fig. 5).

The X-ray powder diffraction (XRD) patterns indicate amorphous phases for all the recovered WEP, which are identical to those in the untreated polystyrene samples, confirming the successful recovery of WEP (Fig. 6).

In Fig. 7, thermogravimetric analysis shows that regular WEP undergoes thermal decomposition in the range 390–500 °C with a weight loss of 99.29%. All the WEP recovered from each of the natural oils present a first loss at 120 °C, and decomposition at 400 °C. The first loss in the recovered WEP samples is attributed to remaining methanol, propanol, or even natural oil. The WEP recovered from chamomile has a weight loss of approximately 24.08%, while for the WEP from star anise, it is 15.86%. The TGA curve of the WEP recovered from eucalyptus and thyme oil shows a single loss of 12.07% and 10.45%, respectively, suggesting the loss of humidity.

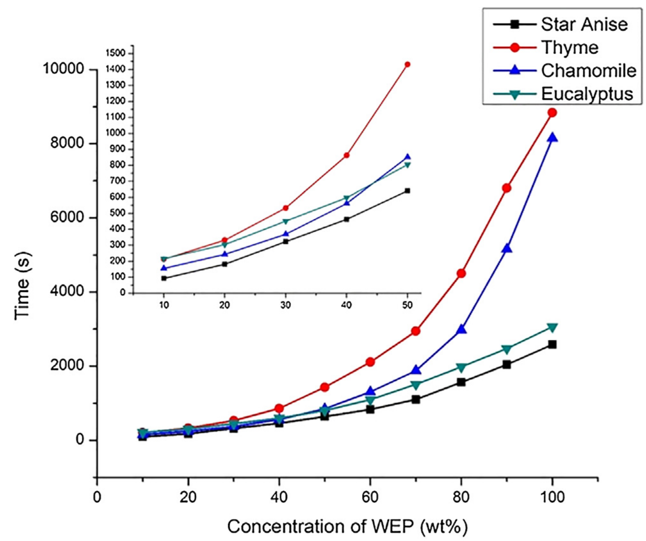

Fig. 2. Dissolution time with the different essential oils.

The TGA analysis of the WEP dissolved in oils (10 wt%; Fig. 8) shows that for star anise and chamomile oil dissolutions, the maximum weight loss is 81.29% and 78.01% at 175 °C and 140 °C, respectively. For thyme and eucalyptus it is 54.33% and 20.61% at 100 °C and 125 °C, respectively. This difference could explain why it is more difficult to recover WEP from star anise and chamomile dissolutions than from eucalyptus and thyme dissolutions. Eucalyptus and thyme evaporation occurred at room temperature even before the TGA

Table 1
Common solvents used for WEP dissolution [5].

Solvent	Non solvent	Temp. (°C)	PR	Experimental conditions	References
DCM	Methanol	100	98%	5 g of PS in 100 mL	[8]
Toluene			97%		
Toluene	n-hexane	100	94.%	1 g PS was dissolved in 20 mL with heating for 30 min.	[8]
Xylene	Methanol	100	97.%		
d-Limonene	————	25	100%	Solution of 10% of EPS in d-limonene was dissolved in 800 s.*	[6]
Cyclic monoterpenes	Water	————	————	0.2 g PS film was dissolved in 2 cm^3 cyclic monoterpenes	[14]
Benzene	Water	————	95%	PS was dissolved in Toluene or benzene, 3 times of water was added, stirring (800 rpm) and dried at	[15]
Toluene			96%	70 °C for 24 h.	
Methyl Ethyl Ketone	Methanol	————	≈100%	The PS solution (0.25–0.3 kg/L) was filtered (200–150 mm) under pressure (0.2–0.3 MPa, 20 °C), and	[16]
	n-hexane			a non-solvent (4–10 times volume of solvent) was added with stirring for 30 min, followed by	
p-xylene	n-hexane			centrifugation.	
Star Anise Oil	Methanol	25	> 95%	Solution of 60% of WEP in essential oil was dissolved in 833 s.	This work
Eucalyptus Oil					
Thyme Oil					
Chamomile Oil					

PR: Polystyrene recovered.
* This data was taken from Fig. 2 in reference [7].

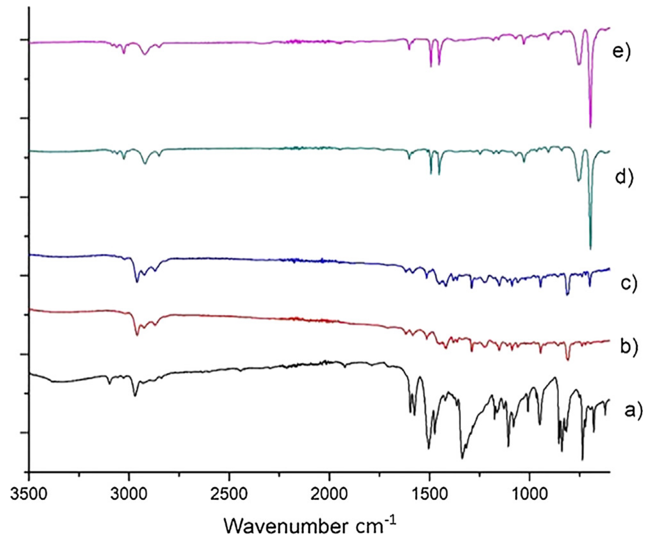

Fig. 3. IR Shimadzu® Spectrometer, with ATR accessory spectra: a) Essential thyme oil, b) Solution of polystyrene in thyme oil, c) Recovered essential thyme oil, d) Polystyrene, e) Recovered polystyrene.

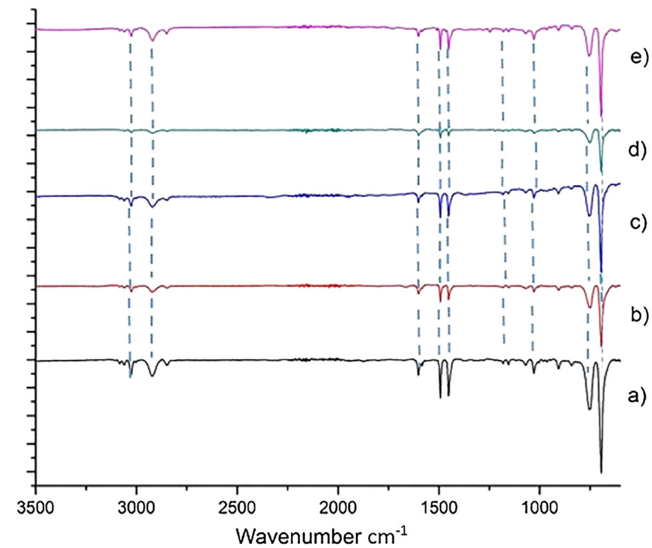

Fig. 4. IR Shimadzu® Spectrometer, with ATR accessory spectra: a) Polystyrene recovered from star anise oil, b) Polystyrene recovered from eucalyptus oil, c) Polystyrene recovered from chamomile oil, d) Polystyrene recovered from thyme oil, e) Waste Expanded Polystyrene.

measurements started due their low flash point, as can be confirmed by their weight loss. On the other hand, chamomile and star anise oils both have higher flash points closer to the WEP decomposition temperature, making separation of WEP from the oil harder, which is in agreement with the observed recovery process.

The glass temperature (T_g) was obtained by Differential Scanning Calorimetry analysis and was used to calculate the activation energy (Eq. (1)) and the molecular weight (Eq. (6)) of the WEP recovered (See Fig. 9). These results show that the molecular weight of the WEP recovered from eucalyptus is the closest to the original expanded polystyrene [17] (Table 3).

The activation energy was calculated using the Arrhenius Eq. (1)

$$k(T) = Ae^{-\frac{E_a}{RT}} \tag{1}$$

where k is a constant, A is a frequency factor, E_a is activation energy, R is the gas constant ($R = 8.314$ J/K mol) and T is temperature.

To use the Arrhenius equation as a linear regression model between the variables k and T^{-1}, the equation is rewritten as (Eq. (2)),

$$\ln(k) = \ln(A) - \frac{E_a}{R}\left(\frac{1}{T}\right) \tag{2}$$

Which follows the equation for a line (Equation (3))

$$y = mx + b \tag{3}$$

With $m = \dfrac{E_a}{R}$ (4)

and

$$b = \ln(A) \tag{5}$$

Eq. (6), for T_g (glass temperature), is used to calculate molecular weight.

$$T_g\,[K] = \frac{1.9 \times 10^5}{M_n} \tag{6}$$

The mechanical properties of the recovered WEP should be investigated to determine for sure if the quality of the recovered product can substitute virgin polymer. In addition, it is necessary to study the fuel properties of such materials. Therefore, there is a wide range of topics for future studies.

4. Conclusions

In this study, we successfully proved that WEP can be dissolved by

Table 2
Comparison of the characteristic peaks of polystyrene with the polystyrene recovered from the different oils.

WEP		PR.SA		PR.TH		PR.EU		PR.CH	
Peak	Intensity	Peak	Intensity	Peak	Intensity	Peak	Intensity	Peak	Intensity
695.35	s	695.35	s	696.31	s	694.38	s	695.35	s
756.1	m	754.17	m	754.17	m	747.42	m	749.35	m
1028.07	w	1028.07	w	1029.04	w	1028.07	w	1028.07	w
1155.38	w	1154.41	w	1154.41	w	1154.41	w	1155.38	w
1452.42	m	1452.42	m	1452.42	m	1452.42	m	1452.42	m
1492.92	m	1492.92	m	1492.92	m	1491.96	m	1492.92	m
1600.94	w	1602.87	w	1601.9	w	1599.98	w	1601.9	w
2849.87	w	2848.91	w	2850.83	w	2849.87	w	2848.9	w
2921.24	w	2921.24	w	2921.24	w	2921.24	w	2922.2	w

WEP: Waste Expanded Polystyrene.
PR.SA: Polystyrene recovered after dissolution in star anise oil.
PR.EU: Polystyrene recovered after dissolution in eucalyptus oil.
PR.TH: Polystyrene recovered after dissolution in thyme oil.
PR.CH: Polystyrene recovered after dissolution in chamomile oil

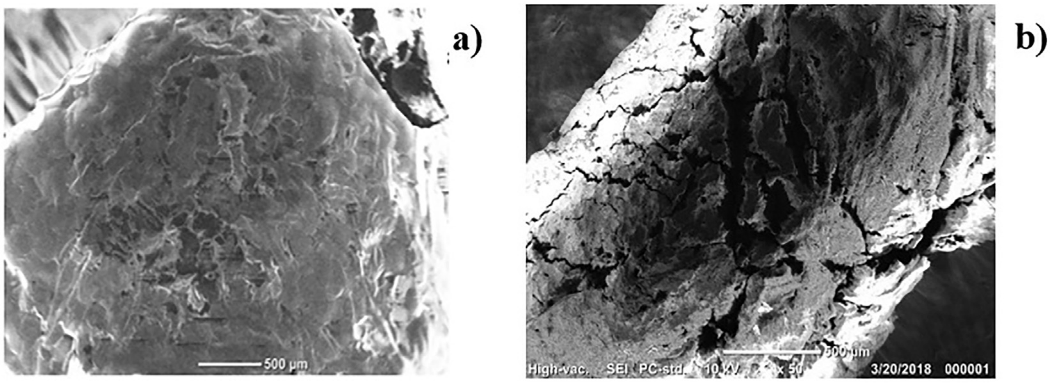

Fig. 5. SEM images of recovered polystyrene: a) WEP, b) Polystyrene recovered from thyme oil.

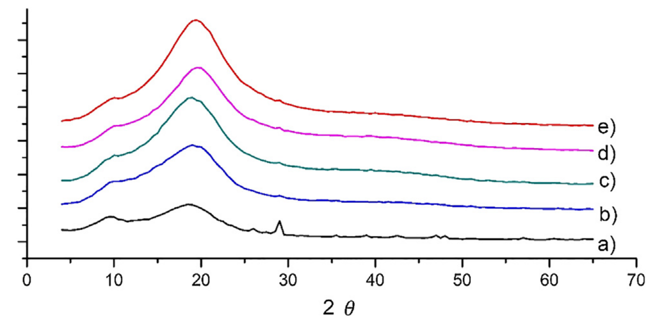

Fig. 6. XRD of recovered polystyrene; a) WEP, b) Polystyrene recovered from star anise oil, c) Polystyrene recovered from eucalyptus oil, d) Polystyrene recovered from chamomile oil, d) Polystyrene recovered from thyme oil.

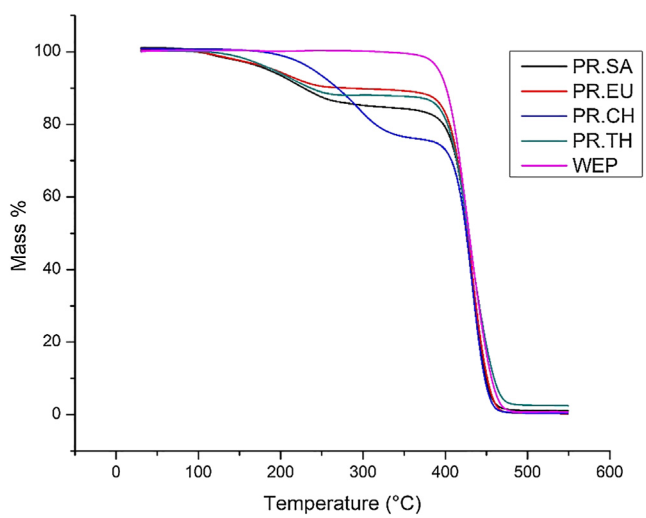

Fig. 7. TGA curve of WEP and polystyrene recovered from different oils.

commercial essential oils, which is a cheaper process than the commonly used d-limonene, without further purification or expensive and complex extractions. Polystyrene was dissolved fastest by star anise oil, followed by eucalyptus oil. The WEP was successfully recovered from all the natural oils tested. The WEP recovered from eucalyptus was the purest and easiest to recover. Finally, we report an efficient recovery process with the opportunity of recycling WEP to form sticks, paints and adhesives. This method also facilitates the handling and transportation of WEP. This study presents a more sustainable and environmentally friendly procedure for recycling WEP, providing an alternative for handling of waste plastic pollution.

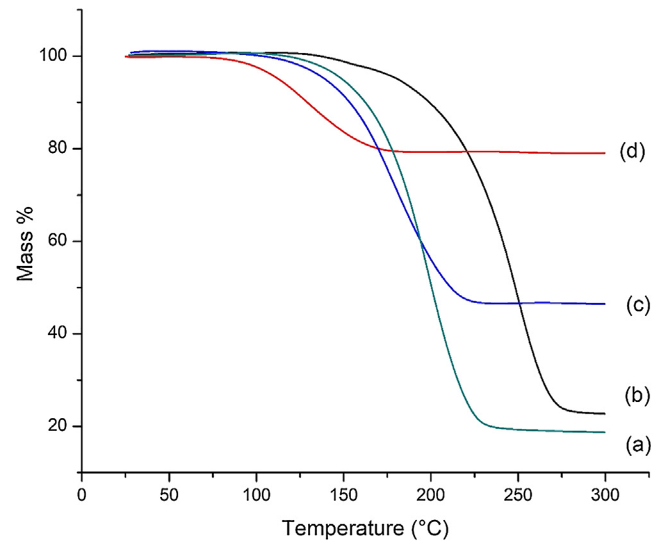

Fig. 8. TGA curves of the solutions with polystyrene: a) Star anise (mass change 81.29%), b) Chamomile (mass change 78.01%), c) Thyme (mass change 54.33%), d) Eucalyptus (mass change 20.61%).

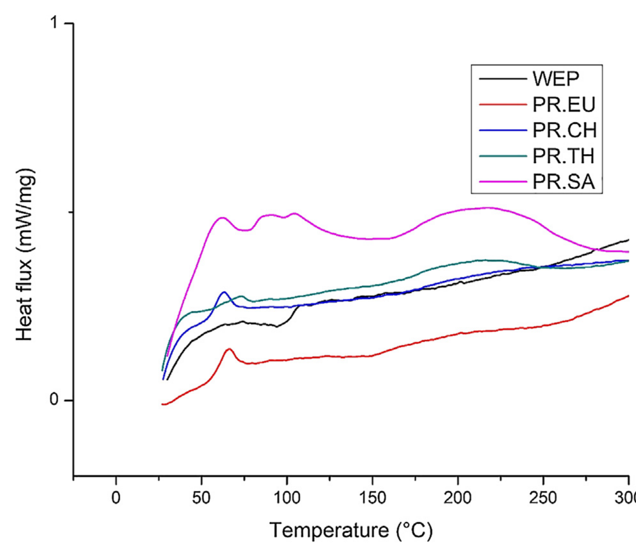

Fig. 9. DSC curve of recovered polystyrene and glass temperature. PR.EU (Polystyrene recovered from solution in essential oil of eucalyptus) T_g, 58.1 °C. PR.TH (Polystyrene recovered from solution in essential oil of thyme) T_g, 75.3 °C. PR.CH (Polystyrene recovered from solution in essential oil of chamomile) T_g, 63.2 °C. PR.SA (Polystyrene recovered from solution in essential oil of star anise) T_g, 56.21 °C. WEP; T_g, 60.5 °C).

Table 3
Activation energy and molecular weight.

	E_a (J/mol)	Mw
WEP	4133.37347	4828.46
WEP recovered from star anise	4146.51033	4353.80
WEP recovered from eucalyptus	4148.75524	4552.2
WEP recovered from chamomile	4139.69246	5196.36
WEP recovered from thyme	4133.78919	7757.31

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.fuel.2018.11.055.

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