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Separation of virgin plastic polymers and post-consumer mixed plastic waste by sinking-flotation technique

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Abstract

The main objective of this research is to separate virgin polymers (PA, PC, PP, HDPE; PS and ABS) and post-consumer plastic waste from municipal solid waste (MSW) using the sinking-flotation technique. The separation was carried out on a pilot scale in a container of 800 l of useful volume with agitation of 160 rpm for one hour. Tap water, ethanol solutions and sodium chloride at different concentrations were used as the densification medium. The virgin polymers were separated into two groups, that is, a group of low-density polymers (HDPE and PP) and a group of high-density polymers (PS, ABS, PA, and PC). Polymers whose density was less than that of the medium solution floated to the surface, while those whose density was greater than that of the medium solution sank to the bottom. The experimental results showed that the complete separation of HDPE from PP was achieved at 23% v/v of ethanol. For the separation of the high-density polymers, up to 40% w / v sodium chloride was used. The recoveries of the polymers ranged from 70 to 99.70%. In post-consumer recycled plastic waste, fractions of 29.6% polyolefins, 37.54% PS, 11% ABS, 8% PA and 12% PC, PET and PVC were obtained. Finally, cast plates were made of the post-consumer waste to improve the identification of the type of polymer present in the separated fractions.

1. Introduction

Plastics have become a crucial part of our lifestyles as they are highly functional, hygienic, lightweight, and inexpensive (Pol and Thiyagarajan, 2010). Therefore, its world production has increased exponentially during the last 50 years (Singh et al., 2017; Gu and Ozbakkaloglu, 2016; Bucknall, 2020). Mumbach et al. (2019) estimated that the world's cities generated 1.3 billion tons of solid waste in 2012 and forecast that it will increase to 2.2 billion tons by 2025. One of the reasons for the increased consumption of plastics is the various applications they have to replace traditional materials (Burat et al., 2009), especially ceramics and wood (Lackner, 2015). Plastics offer very advantageous characteristics such as light weight, resistance to heat, low density and low cost, making them poorly substitutable in different commercial and industrial uses (Pol and Thiyagarajan, 2010). According to Mancheno et al. (2016) the plastics that grow the most worldwide are those that are made up of polycarbonate (PC), polystyrene (PS), polyvinyl chloride (PVC), polyethylene terephthalate (PET), polymethylmethacrylate (PMMA) and acrylonitrile butadiene styrene (ABS) (Rahimi and García, 2017). All this has made the presence of plastics indispensable in the modern lifestyle due to versatility and low production costs (Al-Salem, 2019; Geyer et al., 2017).

However, the problem is that all the plastics generated end up as waste causing negative effects on the environment (Huysman et al., 2017). In addition, the management of plastic waste has not been very successful in recent years, which makes it a challenging task (Sharma et al., 2020; Ferronato and Torretta, 2019; Gupta et al., 2015; Vitorino de Souza et al., 2017; Sharma et al., 2020; Chand Malav et al., 2020; Law et al., 2020). Currently about 80% of plastic waste is sent to landfills (Ayeleru et al., 2020). However, most plastics take hundreds of years to disintegrate when they are dumped in a landfill. Thus, the increasing amount of plastic waste is exerting great pressure on the limited space of landfills

(Takoungsakdakun and Pongstabodee, 2007) causing improper management to negatively affect the environment (Aljerf, 2016). Furthermore, leachate from landfills can penetrate into surface waters posing a serious threat to the health of nearby residents (Du et al., 2020). In correspondence with the continuous growth of post-consumer plastic waste and the inadequate management of these, there is a special interest in continuing to search for efficient, economic and environmental alternatives to better manage plastic waste (Bing et al., 2016).

Currently there are various technologies to manage plastic waste. Thus, for example, incineration (Achilias et al., 2007; Guney et al., 2013; Huang et al., 2013) is a widely used alternative to eliminate bulky plastic waste; However, this technique generates a large amount of polluting gases such as CO, SO_2, NO_2, HCl and dioxins (Du et al., 2020; Tue et al., 2016). On the other hand, a promising alternative method that generates less pollution and could be very effective is the recycling of plastic waste (Chen et al., 2011). Recycling involves separating and identifying the plastic waste generated into individual categories. However, for the separation of plastic waste there are various techniques and methods (Ruj et al., 2015). A very economical separation technology that does not generate pollution to the environment is the sinking-floating separation. The application of this technique is simple; its use consists in varying the density of the aqueous media used in the dense process. Many authors have used this technique to separate polymers, such as polyolefins (HDPE, LDPE and PP) (Hu et al., 2013). In addition, the sinking-floating separation technique has been and continues to be widely used as a means of separating plastics, especially those that do not have similar density (Dodbiba et al., 2002; Shimoiizaka et al., 1976; Pongstabodee et al., 2008). Some of the means that have been used to recover plastics have been water, solutions saturated with water with sodium chloride, calcium chloride and ethanol solutions (Fu et al., 2017).

With the aim of improving the procedure and further enriching the literature on the applicability of the technique, the objective of this research is to present a work on the separation of different virgin polymers and plastic waste from post-consumer urban solid waste by means of the sinking method. -floatation. In addition, for the separation of polymers and plastics, three aqueous separation media are used: tap water, sodium chloride and ethanol. Finally, the characteristics of the separated fractions of post-consumer waste are evaluated by making melted plates at different melting temperatures.

2. Materials And Methods

2.1 Materials

Virgin plastics

Polymer samples from the Technological Institute of Plastic, AIMPLAS (Valencia-Spain) were used to separate the virgin plastics. Five different types of virgin polymers were used: high density polyethylene (HDPE), polypropylene (PP), acrylonitrile butadiene styrene (ABS), polyamide (PA), polystyrene (PS) and polycarbonate (PC). The virgin PP and HDPE granules had particle sizes between 3.36 and 2 mm in diameter. Although both polyolefins were white, they presented different tonalities in their coloration; thus, PP was much more transparent than HDPE. The shape of the ABS polymer was spherical and yellowish in colour. Likewise, the PS presented a spherical shape, although its colour presented a white-transparent hue. Instead, the PC featured a white cylindrical shape. For their part, the PA granules presented a rectangular shape and white coloration. **Table 1** shows some characteristics of the virgin polymers used.

Table 1. Standard main characteristics of virgin polymers

Polymer	Code	Characteristic	Standard	Units	Value
PP	BJ750	Density	D1505 ATMS	g/cm^3	0.910
		MFI (190 °C/21.,6 kg)	D1238 ATMS	g/10min	28
		Flexural modulus	D790 ATMS	Kg/cm^2	15.500
HDPE	Lotrène Q TR-571	Density	D792 ATMS	g/cm^3	0.953
		MFI (190 °C/21.6 kg)	D1238 ATMS	g/10min	0.020
		Flexural modulus	D790 ATMS	MPa	1300
PS	124N/L	Density	ISO 1183	g/cm^3	1,040
		MFI (190 °C/21.6 kg)	-	g/10min	-
		Flexural modulus	ISO 178	MPa	3400
ABS	ELIX ULTRA HH 4115 PG	Density	ISO 1183-1	g/cm^3	1.070
		MFI (230°C/3.8 kg)	ASTM D1238	g/10min	3
		Flexural modulus	ASTM D 790	MPa	2000
PA	PA 6 EXTRUDADA	Density	ISO 1183-1	g/cm^3	1.140
		MFI (190 °C/21.6 kg)	-	g/10min	-
		Flexural modulus	ISO 178	MPa	2800
PC	TECANAT PC	Density	ASTM 53479	g/cm^3	1.200
		MFI (190 °C/21.6 kg)	-	g/10min	-
		Flexural modulus	ASTM 53457	MPa	2200

Note: MFI= Melt Flow index

Post-consumer recycled plastics

As post-consumer recycled materials, plastic waste from solid urban waste (MSW) was used; the same ones that presented a certain degree of contamination and deterioration due to labels, inks, impurities and combinations. The waste analysed consisted of all types of plastic household packaging, such as sheets, bottles, cans, jars, margarine containers, yogurt jars, flowerpots, garden tables, etc. The samples used were mixed residues of various colours with a non-uniform size and shape.

2.2 Experimental procedure

Virgin plastics separation

2.0 kg samples of each of the different virgin polymers were thoroughly mixed. The mixture was carried out on a pilot scale in a tank, Ecomini ML-100, with a useful volume of 800 l. The waste was mixed with tap water with agitation of 160 rpm for one hour so that the mixtures were completely moistened and to avoid the formation of air bubbles. Polymers with a density lower than tap water (PP and HDPE) floated to the surface while plastic waste with a density higher than tap water (PA, PC, PS and ABS) sank to the surface. background. The polymers that sank and floated were centrifuged at 2950 rpm with an MC-250 centrifuge, to remove moisture from the polymers, and impurities and improprieties such as dust, dirt, etc. The polymers were then weighed to evaluate their recovery as a function of mass balance.

To further separate the polymer mixtures (PP and HDPE) and (PA, PET, PS and ABS) into individual polymers, the tap water was replaced by an ethanol solution and a sodium chloride solution. The sodium chloride solution was used with various densities (1.055, 1.100 and 1.175g/cm^3), while the ethanol solution was a solution of a single concentration (0.935 g/cm^3). **Figure 1** shows the separation process of the initial polymers, where the polymer that is denser than the dense medium sinks to the bottom and the polymer with a lower density than the dense medium floats on the surface. Finally, the agitation and centrifugation process in the separation of the polymers with ethanol and sodium chloride were carried out under the same conditions as in the separation with tap water.***Separation of post-consumer recyclable plastics.***

Figure 2 shows the process of separating automotive plastic waste with the different solutions used (tap water, ethanol and sodium chloride). Before starting the separation process, the plastics were crushed with a RC400 large cutting mill to reduce the size of the samples to diameters less than 50 mm; However, to homogenize the samples, their size was reduced to 3 mm diameters using a second C17.26s mill (**Figure 3**). In addition, the plastics were of different colours, which facilitated the analysis of their separation. Once all the samples had been crushed (12 kg in total), they were subjected to the separation process with the same conditions (concentration of solutions, tank volume, agitation and centrifugation) as those used in the separation of virgin polymers. Finally, the compositions and densities of the different mixtures were estimated. The typical densities in the mixtures were determined as an average value of five repetitions.

Recycled polymer plates

The composition of the separated plastic particle fractions should correspond to combinations of polymers with densities like those of the standards used to obtain the densities of the separation solutions of the first experiment.

To identify the type of plastic present in each separated fraction, plates or sheets were made from each of the fractions. The plates were made by compression using an industrial hydraulic press (FONTUNE PRESES model). The press consisted of two plates, one upper and one lower, driven by a hydraulic system that exerts high pressures. Previously the particles of each fraction were distributed in a mold of 20x20 cm and thickness of 0.2 mm (**Figure 4**). The whole (fractions of plastics, mold and metal plates) was placed in the hydraulic press for 10 min and a pressure of 100 kN with a progressive increase in temperature T_e, until reaching the melting temperature of the fraction. As a reference temperature (T_m) to melt the separated fractions the melting temperature of the virgin polymers was used. The range of temperatures used to melt the fractions ranged between 20 and -20% the T_f, because the fractions were not considered pure polymers since they were mixed with fillers, inorganic additives, and mixtures of impurities and improprieties. Subsequently, the temperature was lowered to 60 °C to cool the molten plate. The identification of the type of plastic was identified based on the percentage of molten area of the plate.

Table 2. Recycled polymer plates.

Separation solution	Concentration g/cm^3	Fraction used	Plate obtained	T_e ºC	Estimated polymer	T_f ºC
Ethanol	0,935	Mix 3	1	134-201	PP	168
		Mix 4	2	110-165	HDPE	137
Sodium chloride	1,055	Mix 5	3	176-264	PS	220
	1,100	Mix 7	4	180-270	ABS	225
	1,175	Mix 9	5	200-300	PA	250
		Mix 10	6	140-265	PC, PET, PVC	145-260

Note: T_f = melting temperature of virgin polymers, T_e = temperature range of the test

3. Results

3.1 Separation of the virgin polymer mixture with NaCl

To separate the mixture of polymers of HDPE, PP, PS, ABS, PA and PC into individual polymers, an aqueous solution of sodium chloride with various concentrations was used **Figure 5**. When only tap water was used, 97.5 was recovered % HDPE+PP. However, for an 11-12% w/v NaCl concentration, most of the PS (80.3%) floated to the surface of the solution, while the ABS, PA and PC sank to the bottom. With a further increase in the concentration of the solution (20% w/v), the complete separation of the ABS mixture (84.8%) from the PA and PC polymers was achieved. For a NaCl concentration of 40% w/v most of the PA (70%) floated to the surface of the solution, while 95.4% of the PC completely sunk.

3.2 Separation of the virgin polymer mixture with C_2H_5OH

The HDPE+PP fraction previously separated with water was separated with ethyl alcohol into individual polymers of PP and HDPE (**Figure 6**). The PP, which floated, began to separate from the HDPE when a concentration of 23% v/v of C_2H_5OH was used, obtaining a recovery of 95.60%. Complete separation of HDPE from PP was achieved when a concentration of 31% v/v was used. However, the recovery fraction of HDPE was much higher (99.70%). The experimental results demonstrated that the recoveries of HPDE and PP occurred for a density of the aqueous medium of 0.935 to 0.955 g/cm^3.

3.3 Separation of the post-consumer plastic waste mix with NaCl and C_2H_5OH

In **Figure 7**, 6 separate fractions of the MSW plastics are represented. The results showed that plastic waste was mainly composed of mixtures 5 (37.5%) and mixtures 3 and 4 with 15.4 and 14.1%, respectively. According to the estimated densities of each separation mixture, the above mixtures could be related to the polymers of PS, PP and HDPE. To a lesser extent the mixtures 7; 9 and 10 represented 11.9; 8.5 and 12.4% respectively that could be related to ABS, PA and PC polymers with some PET and PVC. However, the different fractions are mixed by inorganic fillers and additives, which causes the properties to vary, making the separated samples not completely pure and do not correspond to a single polymer.

3.4 Determination of plates of recycled polymers

One of the objectives of making the plates is to observe the diversity of colours present in them and determine if the material used melts completely at the melting temperature of the polymer. If the material melts completely, it means that the material has been separated with a high efficiency, that is, the material that makes up this fraction is sufficiently pure and in its composition, there is no presence of other polymers, nor the presence of impurities and improprieties such as dirt, wood, glass, metals, etc. On the contrary, if the plate has discontinuous castings or burned areas, it means that the material has not completely melted, causing there to be heterogeneity in the composition of the plate due to the low purity of the polymer.

Figure 8 shows the plates of post-consumer plastics obtained at different melting temperatures. Thus, for example, mixture 3 has a high percentage of cast iron, does not have cracks and does not have burned areas. However, the mixture has brown spots or spots. Stains caused by possible remains of wood and impurities that have prevented its complete melting (**Figure 8A**). Similarly, mixture 4 has few burned areas, denoting a more homogeneous casting (**Figure 8B**). However, spots of different colours are still noticeable, indicating the presence of other polymers of similar density.

In **Figure 8C**, a plate is represented that could mostly contain ABS due to its greater uniformity in its casting; the plate also shows some specific points where it is burned due to the presence of other polymers of lower density. On the other hand, the plate of **Figure 8D** contains less mixture of impurities due to the little presence of cracks. However, it has areas that have not completely melted, which means that there are higher density polymers. It also has some burned parts, which indicates the presence of polyolefins and ABS.

Finally, the last two plates of fractions 9 and 10 could correspond to PA, PC, PET and PVC due to their high densities. On the plate in **Figure 8E** you can see many burned areas and cracks, which means that the purity of the mixture is not very good because there is a mixture of different polymers. For its part, the last plate (**Figure 8F**) shows brown colours and many black spots due to the presence of impurities and improper elements (remains of wood, glass soil, etc.). In addition, this plate has areas that are burned, which indicates that it has polymer blends with a lower melting point.

4. Discussion

4.1 Separation of the virgin polymer mixture with NaCl

The results showed that polymers of polyolefins and PC are easier to recover by means of the float-sink technique due to their high recovery percentages. The use of this technique is presented as an interesting alternative to improve the recycling process. Likewise, in the ABS and PS groups, high recovery results were obtained, while in the PA samples the separation was not so interesting since their recovery percentages were quite low. The role played by the density of the medium, in the separation by sinking-floating, is a key parameter to separate the plastic particles (Fu et al., 2017). For example, PE and PP, as lightweight plastics, can be easily separated by density sorting in the medium of water, while PVC, PET, as heavy plastics, are considered unmanageable plastic blends due to their similar density (Wang et al., 2020). In this study, separation with tap water made the polyolefins separate easily, since these polymers have specific densities that are lighter than those of water (Ito et al., 2010). The recovery data for polyolefins was higher than 97%, results very similar to other studies in the literature. Thus, for example, Gundupalli et al. (2017) through laser-induced degradation spectroscopy obtained recoveries of 80-97% of polyolefins. Similarly, Wang et al. (2015) through foam flotation recovered percentages higher than 97% of and PE and PP. Similarly, Serranti et al. (2015) through the magnetic density separation technique managed to recover more than 94% of polyolefins. Bonifazi et al. (2013) using hyperspectral imaging technology (HSI) obtained recoveries higher than 96% of PE and PP.

Other fractions that had high separation percentages were the PS and ABS polymers. Obviously, these polymers possess an inherent buoyancy in higher density aqueous media, making them manageable polymers for recycling (Wang et al., 2016). Hence, there have been recoveries of over 80%. The high recovery of PS and ABS suggests the presence of an inherent hydrophobic surface of the tested polymers (Du et al., 2020). PA (70%) was the polymer that recovered the least. Their separation could be improved if a selective wetting of polymers was carried out, which allowed a reduction in surface tension (Wang et al., 2015; Alter, 2005; Fraunholcz, 2004). It should be considered that in this study a natural flotation of the polymers was carried out. However, the separation results were very consistent and much better than other previous studies. Qu et al. (2020), for example, through a natural flotation of ABS and PC, they achieved separation percentages of 68.41% and 59.4%, respectively.

4.2 Separation of the virgin polymer mixture with C_2H_5

As is known, all polymers both HDPE and PP are naturally hydrophobic. Thus, the set of HDPE+PP samples is easy to separate from the rest of polymers with higher density; however, separating HDPE from PP requires using a suitable solution due to their similarity in densities. Therefore, it is necessary to use a suitable agent to achieve the individual selective separation of polymers (Kangal and Üçerler, 2018).In this study, the use of ethanol as an aqueous solution to modify the separation medium was adequate since it allowed the two polymers to be separated with a high percentage of recovery. However, the small differences in density between HDPE and PP, when separated with ethanol, caused the sedimentation rates to be slow (Ferrara and Meloy, 1999).

In this work, the separation of the mixture between HDPE and PP began when the concentration used was below 23% v/v of C_2H_5OH. The ethanol concentration was much lower than that used by Pongstabodee et al. (2008), who used between 30-50% v/v of C2H5OH to separate HDPE from PP. However, in this study, 4.11% more PP was recovered than HDPE. This is because the density of the PP polymer was much closer than that of the aqueous medium. In general, the use of the sink-float separation technique can be very effective in separating polyolefins from each other (Bauer et al., 2018), especially since recoveries close to 100% can be obtained.

4.3 Separation of the post-consumer plastic waste mix with NaCl and C_2H_5OH

In this study the plastics were separated into 6 different groups based on their estimated density. The obtained fractions demonstrated that the analysed MSW plastic waste is mainly composed of ABS (mixture 7) and PS (mixture 5). These results are compatible with data from other studies that show that PS and ABS polymers added to other polymers are those that predominate in MSW, mainly when there are vehicle remains (Zhang et al., 2020). According to Dahlbo et al., (2018) and Burange et al., (2015) MSW plastic waste is usually composed of PP and HDPE and to a lesser extent by PS (Karmakar, 2020). However, variations in the percentage values of MSW plastic waste are usually associated with the consumption habits of the population (Vázquez et al., 2020).

In this work, the mixture 10 comprised a density of 1.20-1.40 g/cm^3, which led us to suppose that the mixture was composed of more than one polymer of the same or similar density. In accordance with the estimated density and with the percentage of separation obtained, this mixture can be composed of PVC, PET and some PC remains. In this case, the mixture represented between 8 and 12% of the total composition; Values very similar to the estimates of Buekens and Yang (2014), who consider that PET, PVC could represent 10-15% of the waste from a car. On the other hand, PVC and PET plastic waste do not have a density difference, which prevents them from being separated by sinking-floating. The density of PET changes from 1.33 to 1.37 g/cm^3 and the density of PVC is between 1.32 and 1.37 g/cm^3 (Burat et al., 2009). In other words, the separation of plastics of equal density is not possible by gravity methods (Hopewell et al., 2009). Hence, in mixture 10 a mixture of PET and PVC has been grouped together and they have not been separated into two different fractions. Furthermore, previous studies have found that a strong alkaline solution of NaOH could destroy the hydrophobicity of one of the polymers (Burat et al., 2009; Kangal, 2010), preventing the separation by sinking-floating.

4.4 Determination of plates of recycled polymers

In general, all the plates made had more than 90% cast area; moreover, they all melted above the melting temperature of virgin polymers due to the presence of inorganic additives, fillers and possible foaming agents. This was reflected in the presence of burned and cracked areas generated during combustion. The fact that the plates have undergone these reactions is due to the fact that the foaming material is usually composed of a large number of small foam holes that are often known as porous polymeric material (Jin et al., 2019). In addition, polymer foam is one of the most used polymeric materials and plays a very important role in the automotive industry (Zhang et al., 2018; Li et al., 2018), (Wang et al., 2017; Wang et al., 2018; Li et al., 2018). The results also revealed that the plates showed poor polymer homogeneity, indicating the presence of more than one polymer in each mix. This is because the foamers cause the density of the mixture to decrease, causing the plastic material to vary its density.

Conclusion

This study includes the separation by sinking-flotation of virgin polymers (HDPE, PP, PS, ABS, PA and PC) and mixed plastic waste from municipal solid waste. The sink-float method with water was very effective in separating the HDPE and PP polymers as they recovered up to 97.5%. The separation of the individual HDPE and PP fractions occurred when concentrations of 23% v/v of ethanol were used, obtaining recoveries of 96% of HDPE and 99.7% of PP. Likewise, the results of the separation of the higher density polymers (PA, PS, ABS and PC) were very promising, since recoveries of around 70-85% were obtained. The concentration of sodium chloride used to separate the polymers was 11-40% w/v. Finally, in the MSW plastics, separate fractions of 29.6% were obtained for polyolefins, 37.54% for PS, 11% for ABS, 8% for PA and 12% for PC, PET and PVC.

Declarations

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Compliance with ethical standards

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Figures

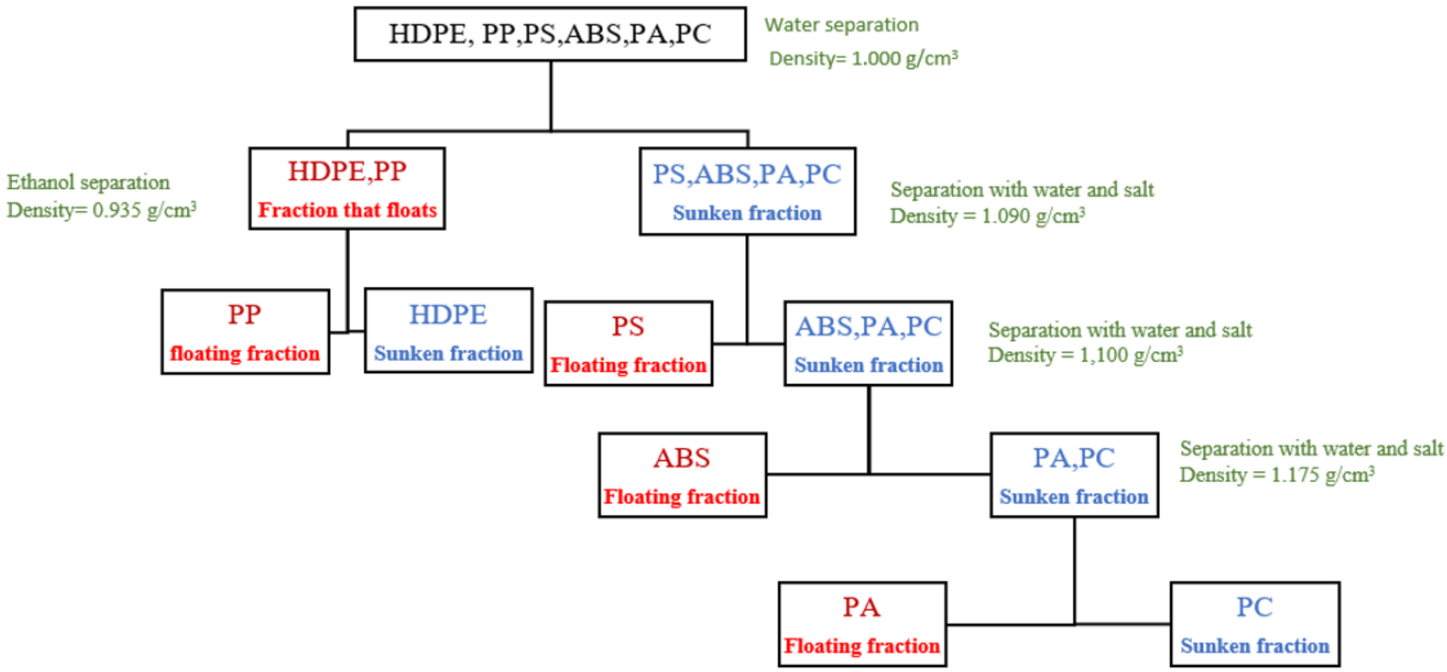

Figure 1

Separation of virgin polymers (HDPE, PP, PS, ABS, PA and PC) in water and NaCl. Note: The polymers written in red are those that float in the solution and the polymers written in blue sink in the solution used.

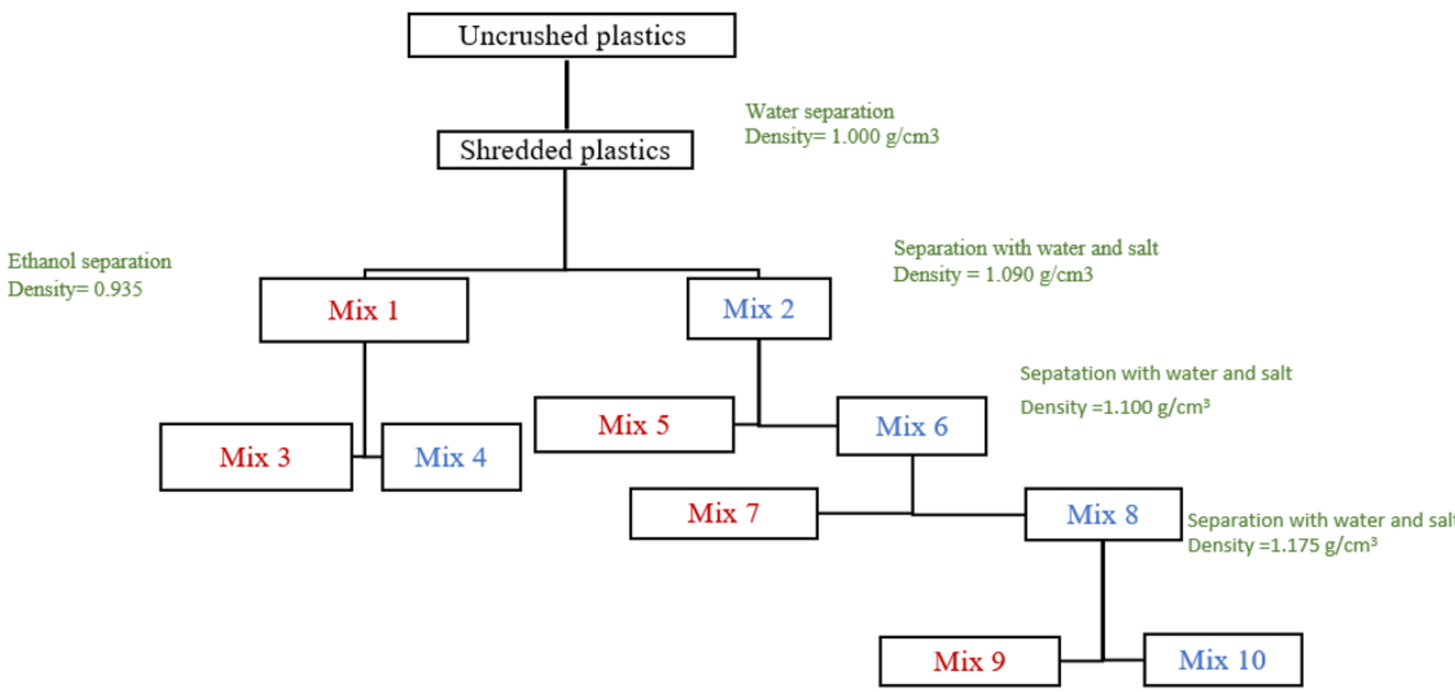

Figure 2

Separation of virgin polymers (HDPE, PP, PS, ABS, PA and PC) in water and NaCl. Note: The polymers written in red are those that float in the solution and the polymers written in blue sink in the solution used.

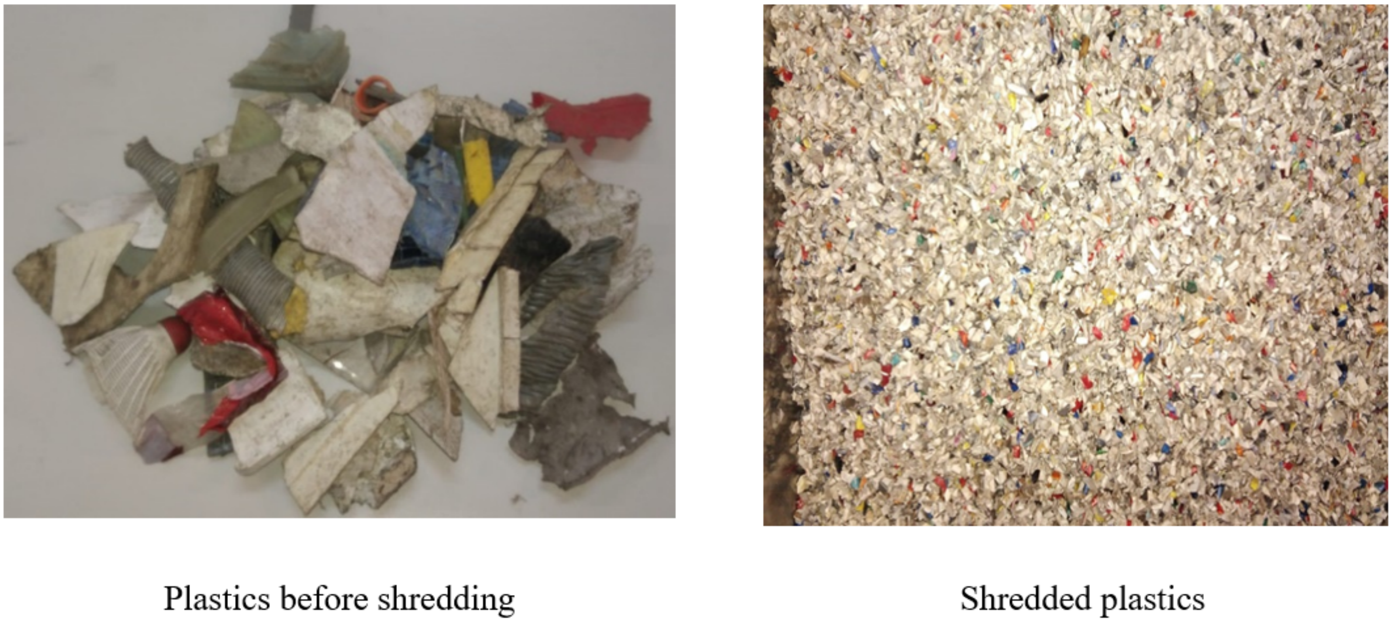

Figure 3

Post-consumer plastics from solid urban waste (MSW) waste before and after being shredded

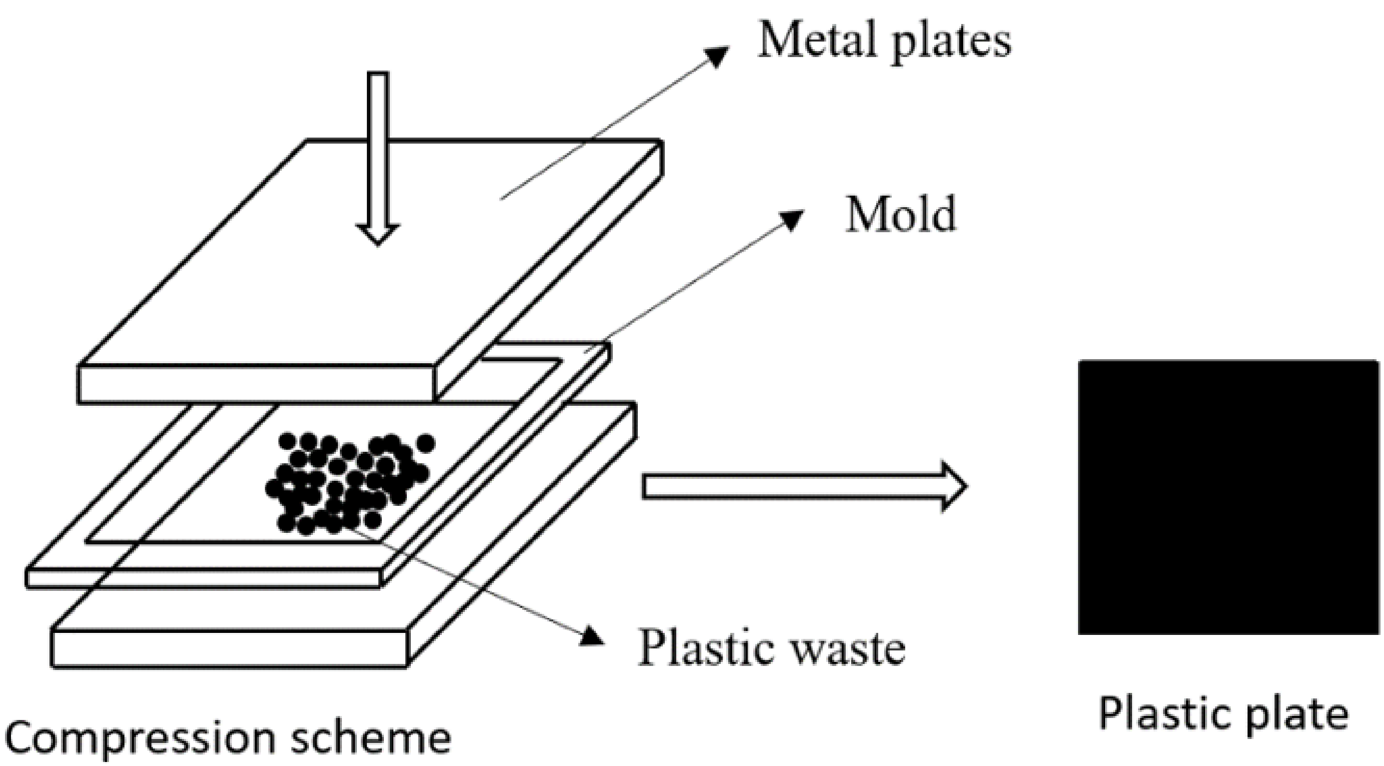

Figure 4

Process of obtaining the plates by means of the compression test.

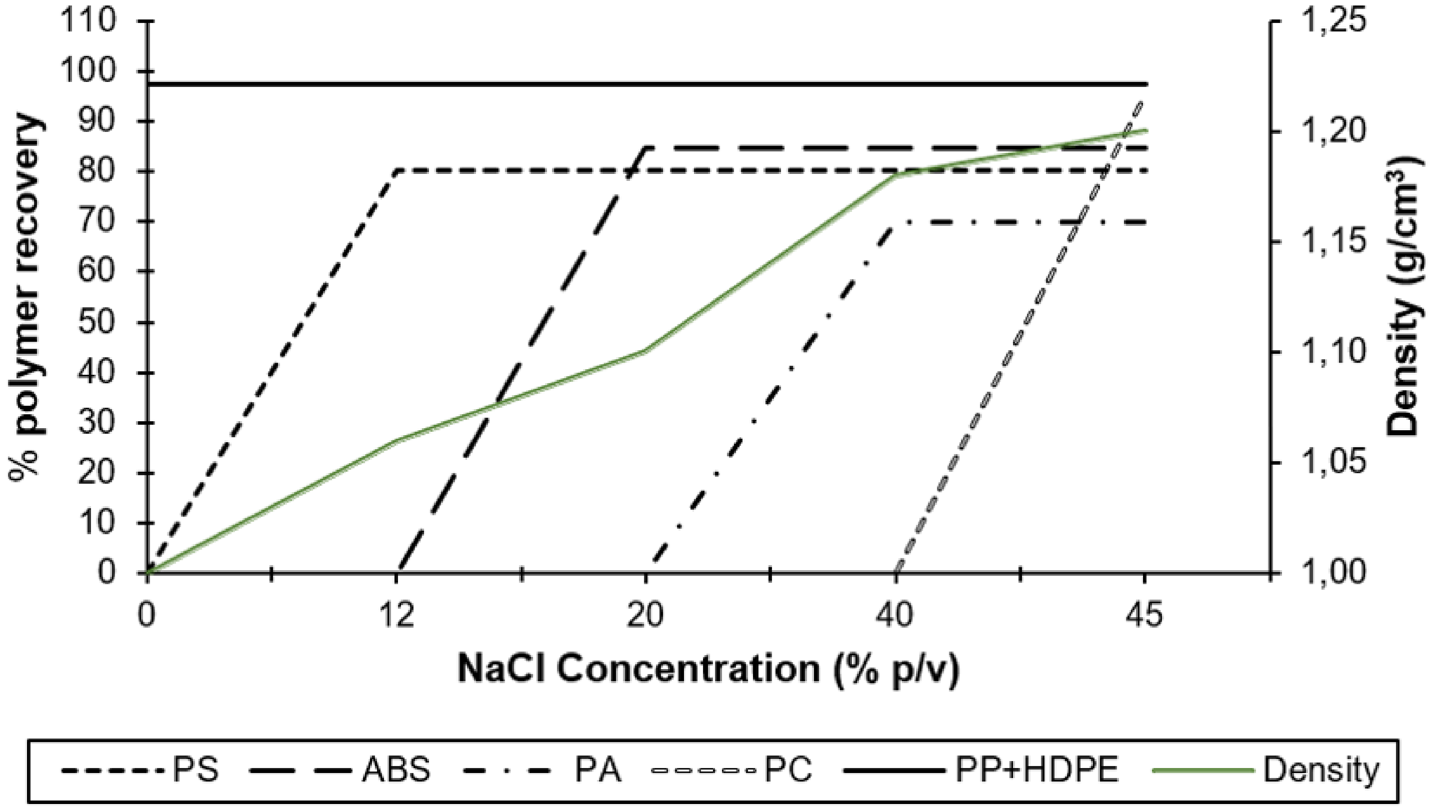

Figure 5

Separation of the PP+HDPE/PS/ABS/PA/PC mixture by the sinking-flotation method with sodium chloride: recovery of the polymers and density of the aqueous sodium chloride solution.

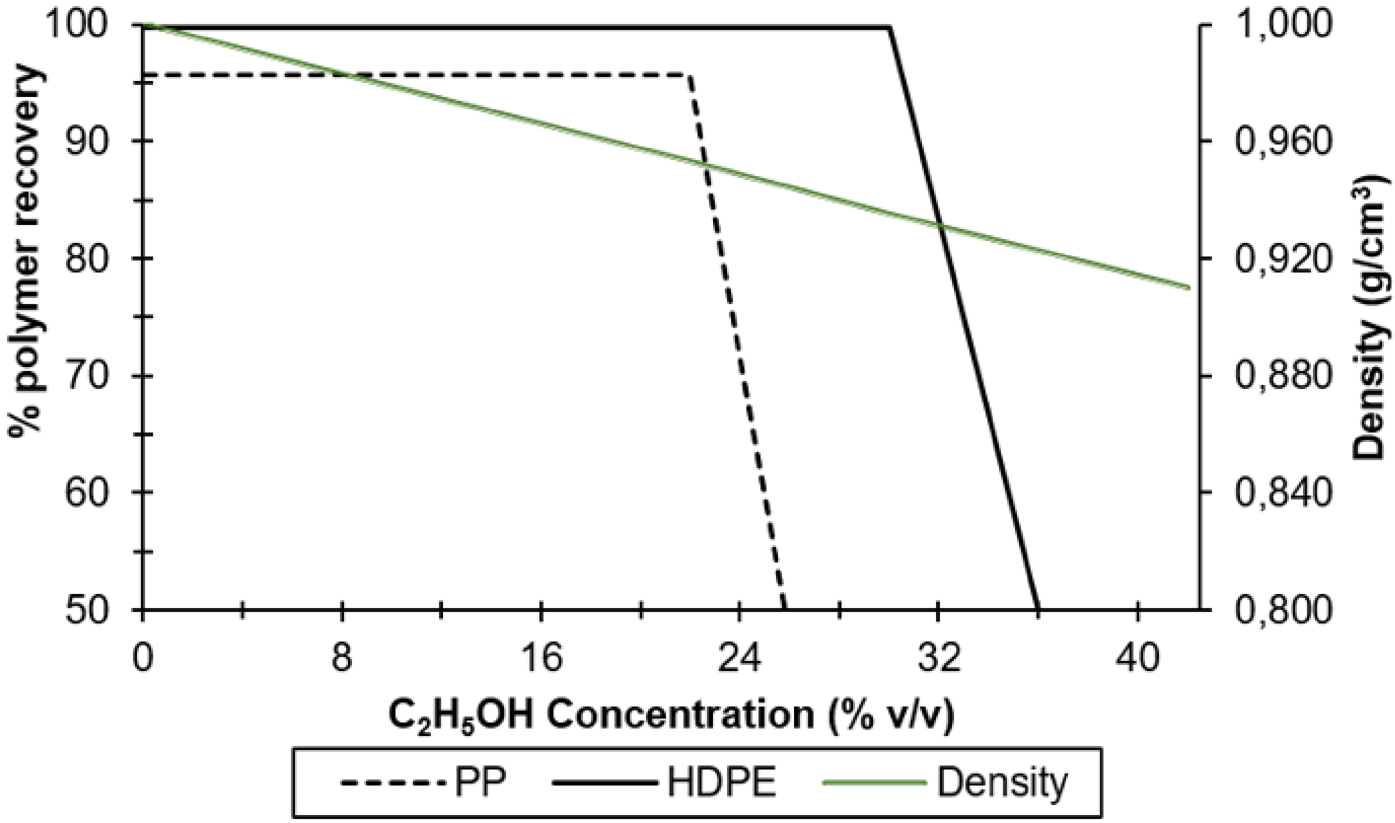

Figure 6

Separation of HDPE from PP by means of a sinking-flotation process with an ethyl alcohol solution: recovery of the polymers and density of the aqueous sodium chloride solution.

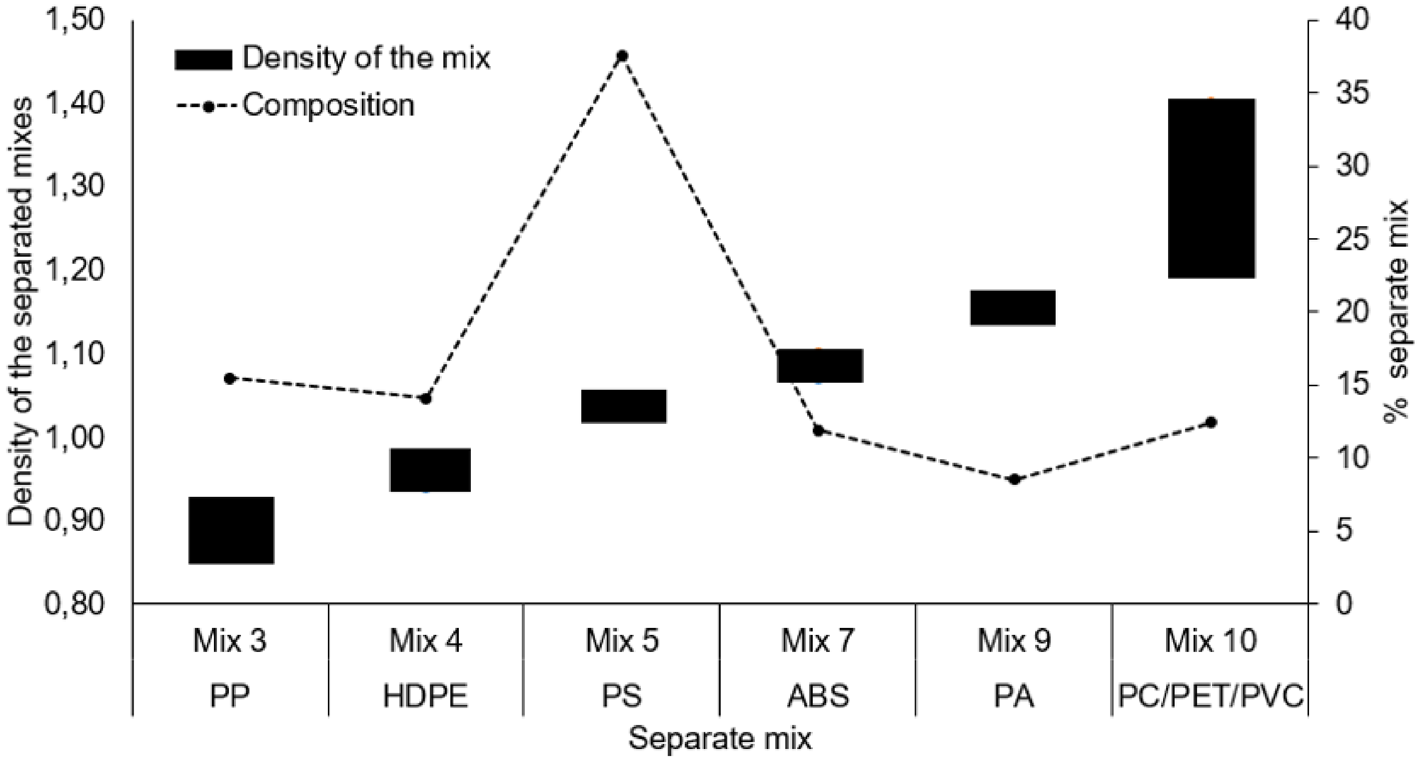

Figure 7

Range of densities and percentage of composition of types of plastics reported in post-consumer plastic waste from solid urban waste (MSW).

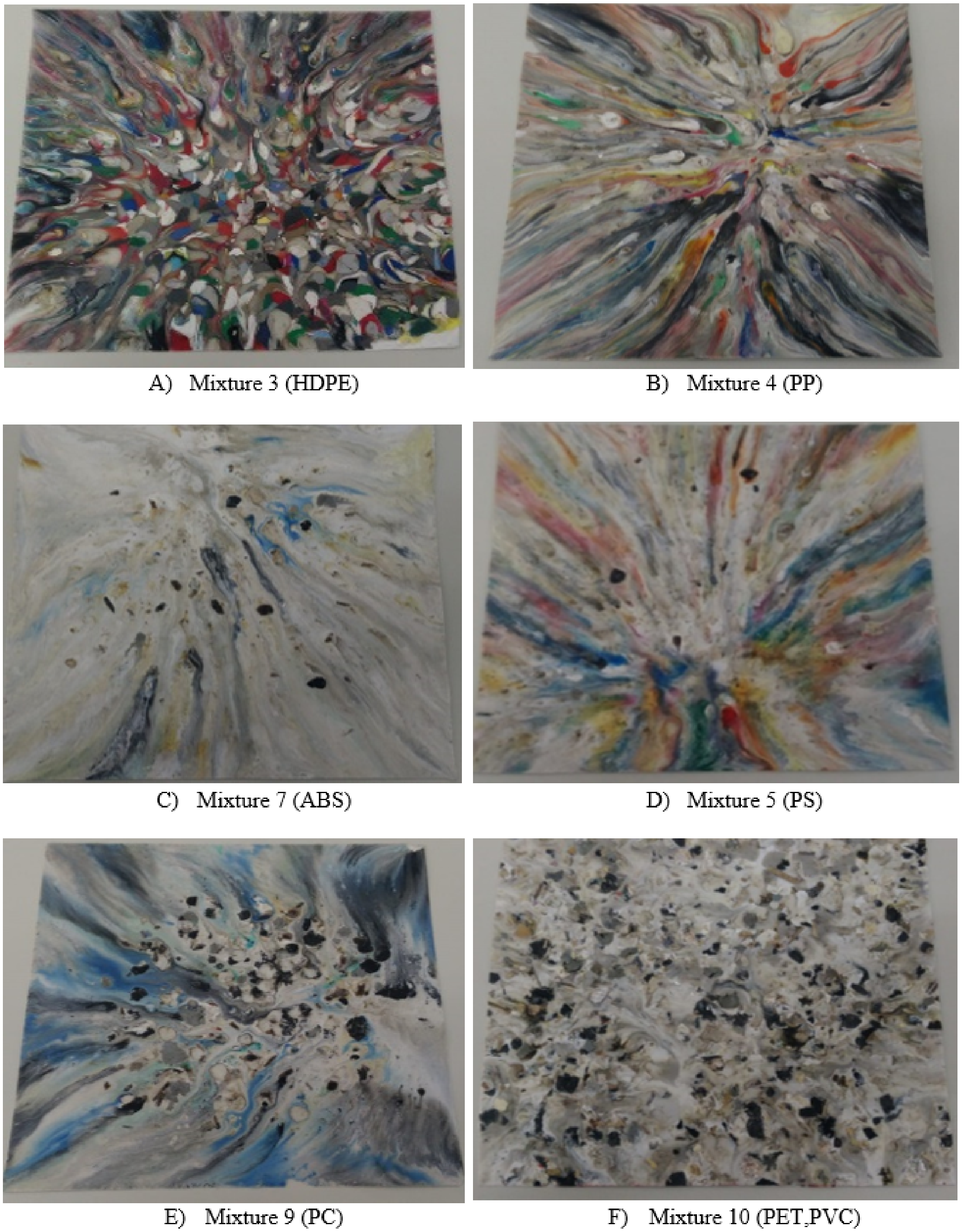

Figure 8

Plates of post-consumer plastic waste melted at different melting temperatures. Mixture 3 was melted at 175 °C, Mixture 4 at 195 °C, Mixture 5 at 220 °C, Mixture 7 at 235 °C, Mixture 9 at 240 °C, and Mixture 10 at 185 °C.