

Preparation and Characterization of Recycled PET/PP Blends from Automotive Textile Waste for Use in the Furniture Edge Banding Sector

Merve Ozer, Tolga Gokkurt, Yasemen Gokkurt, Ezgi Bozbey

Abstract—In this study, research has been conducted on the recovery of automotive textile waste, which has heavy use in the automotive sector and consists of PET/PP content, through the upcycling technique of post-product and post-consumer usage. The aim is to investigate the formulation and production methods that will enable the substitution of original PP raw materials, used in the production of plastic edge bands, with PP/PET alloys. The lamination structure of the mentioned waste makes it impossible to separate the incompatible PP and PET phases, thereby hindering the production of high-quality raw materials or products through recycling. In this study, a comprehensive process was examined through a two-step production process using different types of block and maleic-grafted copolymers to achieve compatibility between these two incompatible phases. The obtained plastic raw materials, referred to as PP/PET blends, were examined in detail, with a focus on their mechanical, thermal, and morphological properties, to discuss their substitutability for the original raw materials.

Keywords—Twin screw extruders, mechanical recycling, melt blending, plastic blends, polyethylene, polypropylene, recycling of plastics.

I. INTRODUCTION

THE use of plastic materials worldwide has rapidly increased over the past half century, reaching incredible levels. The growth in plastic material usage has been surpassing global growth rates. Global plastic production was only around 1.7 million tons in 1950, but it rose to 47 million metric tons in 1976, and in 2002 and 2015, it further increased to 204 million metric tons and 322 million metric tons respectively [1]. In 2018, plastic production and consumption almost reached 360 million metric tons [2]. Since 2004, the world has produced as much plastic as it did in the previous half-century, and as of 2015, the total amount of plastic ever produced had reached 8.3 billion metric tons [3].

Approximately 50% of the 360 million tons of plastic raw materials used globally consist of Polyolefins [1], [2]. Considering their extensive use in the packaging sector and their disposable nature, these plastics contribute to an escalating environmental pollution problem [1]-[3]. Numerous studies have highlighted the increasing magnitude of this environmental pollution issue. For instance, between 1950 and 2015, it has been reported that only 6.3% of the total primary

and secondary (recycled) plastic waste was recycled, 12% was incinerated, and the remaining 79% was either landfilled or released into the environment as waste [3]. Similarly, it is estimated that plastic waste production worldwide has reached approximately 6.3 billion metric tons (MT) with the most optimistic estimate [4], [5].

It is evident that recycling is the most effective method to prevent environmental pollution caused by these wastes. These wastes, also known as above-ground mines, actually constitute valuable raw materials that still retain commercial value and should be reintroduced into the economy. However, unfortunately, the recycling rates/figures have not reached the desired levels, resulting in the inability to utilize this value in the economy and leading to significant environmental pollution. For example, even within the European Union, where the most effective efforts to promote recycling have been made, only 32.5% of the 29 million tons of waste collected annually have been recycled for reuse. The remaining plastic-based waste, 42.6% of it, is incinerated for energy recovery, while 24.9% is disposed of in landfill sites [2].

When considering the industries and sectoral products that use plastic raw materials, it can be observed that a large portion of the plastic waste causing environmental pollution consists of polyolefins used in packaging production. These polyolefins include LDPE (Low-Density Polyethylene), HDPE (High-Density Polyethylene), and PP (Polypropylene) derived raw materials and their mixtures [6], [7]. The inadequate collection and separation processes for these wastes, their tendency to contain multiple different types of plastics, their predominantly laminated or coextruded layered structures comprising different polymers, and the difficulties associated with separating these plastics due to their similar physical properties collectively severely limit recycling rates and uses [8]-[10].

However, the hurdles mentioned above can be overcome by implementing appropriately designed and intelligent recycling processes, which offer the potential to generate recycled plastic raw materials while ushering in new opportunities. One such avenue gaining prominence is the exploration of polymer/plastic blends, which have been extensively researched. These blends present a novel approach to boosting recycling rates while transforming the above-mentioned challenges into advantageous prospects [11], [12].

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Polymer-based blends fundamentally serve as a highly efficient and cost-effective approach for integrating the distinct characteristics of various polymers. This enables the attainment of desired technical properties across a wide spectrum of applications without necessitating the synthesis of novel polymers. Referred to as the melt blending method, this technique enhances the properties of polymers by physically mixing them with additional polymers introduced into the structure. By adopting this approach, a significant opportunity emerges to recycle plastics. Particularly, those comprising mixed compositions and exhibiting non-degradable attributes can be rendered reusable with the application of polymer blending techniques. However, these techniques encounter challenges due to inherent incompatibilities and morphological immiscibility between the constituent polymers, primarily arising from variations in viscosity and thermodynamic properties. Consequently, achieving the desired mechanical and optical properties becomes challenging. To address these limitations, blends or mixtures must undergo morphological compatibility through the incorporation of compatibilizers, which serve to reinforce the interface between the components. Various additives are employed in this process to enhance blend miscibility by reducing interfacial tension and minimizing stress at the interface. As a result, the miscible polymer blends exhibit notable improvements in their properties [13], [16]. For successful compatibilization, specific requirements need to be met [14], [15]:

1. Improvement of interfacial adhesion and dispersion of mixture/blend components/phases
2. Increasing adhesion between different phases/components in a solid state facilitates stress transfer
3. Stabilization of the dispersed phases/droplets against coalescence in such a way that a stable (balanced/stabilized) morphological structure can be achieved against the effects of temperature and shear stresses during the process
4. Strengthening the molecular structure of the blend composition by promoting chemical reactions during melt processing.

Several academic studies have focused on investigating the effects of different additives on the morphological properties of polymer blends. For instance, Vervoortden et al. [17] examined the impact of PP-based olefin block copolymers on the morphology of PP/PE blends. They utilized three types of PP-based olefin block copolymers exhibiting variations in viscosity, molecular weight, and octene comonomer content. Through scanning electron microscope (SEM) images and rheological analyses, the researchers observed that adding these block copolymers led to the forming a finer and smaller-grained PP/PE blend structure, indicating improved compatibility between the constituent plastics. In a similar study, Akbari et al. [18] investigated the effects of PP.g.MA (Maleic Anhydride Grafted Polypropylene) copolymers on PP/PET blends, serving as the foundation of their scientific inquiry. They explored the influence of the PP.g.MA copolymer at three different weight ratios (5%, 10%, and 15%) on the morphology of the PP/PET blends. Elemental Analysis (EDAX) results unveiled that PET

consistently constituted the dispersed phase, while PP formed the matrix phase across all cases of the blend compositions. These studies shed light on the potential of specific additives to reshape the morphological characteristics of polymer blends, thereby offering valuable insights into enhancing their properties.

A study conducted by Tao & Mai [19] focused on addressing the environmental pollution caused by the disposable nature of PET-based products, particularly packaging bottles and films. Their research aimed to explore the utilization of these waste materials by investigating PP/PET blends. Specifically, the study examined the compatibility effects of PP copolymers grafted with functional groups, such as glycidyl methacrylate (PP.g.GMA) and maleic anhydride (PP.g.MA), on the blend composition of PP and recycled PET (r.PET). Additionally, the researchers investigated the non-isothermal crystallization and melting behavior of the resulting PP/PET blend structure. Based on their findings, SEM images demonstrated that incorporating PP.g.MA copolymer facilitated the formation of a finer and smaller-grained morphological structure, indicating improved compatibility among the blend phases. Notably, the study emphasized that the best outcomes were attained with a 10% incorporation of PP.g.MA copolymer. The results obtained from the study demonstrate the establishment of a chemical bonding between the maleic anhydride (MA) and glycidyl methacrylate (GMA) functional units present in the PP.g.MA and PP.g.GMA copolymers, respectively. These copolymers were utilized as compatibilizers to harmonize with the terminal groups of r.PET. The formation of this chemical bonding indicates the achievement of a compatible blend structure. Moreover, differential scanning calorimetry (DSC) and optical polarizing microscope (POM) analyses revealed that the r.PET phase within the blend structure exhibits nucleating characteristics. This behavior leads to an inhomogeneous nucleation effect, facilitating faster processing of the final blends during the production of finished products. In a similar context, Oyman & Tinçer [20] investigated the influence of silane binders on PP and recycled r.PET blends using the melt blending method. Silane binders, commonly employed as active ingredients, serve to modify the interface between an organic polymer and an inorganic substrate [21].

Including silane binding agents in the blend system was considered as a potential method to react with the hydroxyl end groups of PET and achieve a modified morphology that promotes compatibility in PP/PET blends, leading to enhanced mechanical properties. To investigate this, the PET surface was treated using silane binding agents, and subsequent examination of the mechanical and morphological properties was conducted by preparing PP/PET blends with varying compositions. The findings indicated that the incorporation of silane connectors generated a unique morphological structure that was not extensively documented in literature. This structure exhibited compatibility, lacked any discernible gaps, and displayed tubular-type fibril extensions. Moreover, it was observed that the inclusion of silane connectors facilitated the development of mechanical properties crucial for structural applications, such as improved tensile and impact strength.

Consequently, this research highlights the potential of silane binding agents in modifying the blend morphology and enhancing the mechanical performance of PP/PET blends. Extensive research efforts have been undertaken to enhance polymer blends' compatibility and mechanical properties by utilizing various approaches [18]. A comprehensive literature review shows that many of these studies primarily focus on utilizing original raw plastic materials. However, a notable gap exists regarding investigations specifically targeting the development of plastic blends derived from recycling processes, which heavily rely on plastic waste. This gap is significant because the majority of plastic waste available for recycling consists of complex and non-separable mixtures containing different polymers. The limitations associated with the recycling rates for such mixed plastic waste present a unique and unparalleled opportunity for the development of plastic blends. Consequently, there is a pressing need for further research endeavors to explore and harness the potential of polymer blends derived from recycled plastic waste. In this direction, this study investigates formulation and production methods for the recovery of automotive textile waste, specifically PET/PP-based production (Post-Product) and post-consumer usage (Post-Consumer) waste, which are extensively used in the automotive sector. The aim is to achieve their recovery through the upward recycling technique and explore the substitution of original PP raw materials with PP/PET alloys. The lamination structure of the waste mentioned makes it impossible to separate the incompatible PP and PET phases in the content and produce qualified final raw materials or products resulting from recycling. To ensure the compatibility of these two incompatible phases with the study, a comprehensive process was examined using block and maleic grafted copolymers of different characteristics within the scope of a two-stage production process. The resulting plastic materials, referred to as PP/PET blends, underwent a thorough investigation of their mechanical, thermal, and morphological properties. Comparisons were drawn between these blends and the original raw materials, focusing on exploring their potential as substitutes through meticulous analysis.

II. MATERIAL AND METHOD

A. The Materials

Within the scope of the study; PET/PP laminated production (post-product) and post-consumer plastic waste with intensive use in the automotive sector were used. These wastes have a layered structure expressed as co-ex or laminated as shown in Fig 1 and cannot be separated by the known techniques of today.



Fig. 1 Schematic representation of automotive textile waste and/or fire structures

Within the scope of the recycling processes, three different compatibilizers were used to eliminate the phase separations arising from the viscosity and thermodynamic differences between the PP and PET main phases that constitute the structure, to obtain a final structure with finer and shell-cell morphology, compatible and technical characteristics that can be used in structural applications. Dow's ELVALOY™ PTW Terpolymer, Addivant's ROYALTUF™ 498 grafted Terpolymer and Lushan's LUSHAN™ AR493 grafted copolymer were used as compatibilizers.

ELVALOY PTW Terpolymer has the "Ethylene/N-butyl acrylate/Glycidyl methacrylate" structure. It is called ethylene terpolymer. Melt Flow Index value (MFI/MFR) is 12 g/10 min under 190 °C and 2.16 kg load in accordance with ISO 1133 (ASTM D1238) standard, density value is 0.94 g/cm³ in compliance with ISO 1183 (ASTM D792) standards. The melting temperature is determined around 720 °C following ISO 3146 (ASTM D3418) standard. The content of the terpolymer methyl acrylate/acrylate comonomers (methyl acrylate comonomer) is about 28%.

ROYALTUF™ 498 grafted Terpolymer has an Ethylene-Propylene-Diene (EPDM.g.MA) structure grafted with MA. By ASTM D-6047 standard, MA content can vary between 0.8% and 1.2%. Following ISO 11357 standard, the glass transition temperature is -460 °C and by ASTM D-1646 standard, the Mooney Viscosity (Mooney Viscosity 1+4, 125 °C) value describing the fluidity is 30.

LUSHAN™ AR493 suspended copolymer has a maleic anhydride-grafted ethylene-octane copolymer (POE.g.MA) structure. Density value was measured at 0.87 g/cm³ following ISO 1183 (ASTM D792) standards, melt Flow Index value (MFI/MFR) was measured at 190 °C by ISO 1133 (ASTM D1238) standard and 0.2 g/10 min under 2.16 kg load. According to ASTM D-6047, MA content can range from 1.0% to 1.2%.

B. Recycling and Melting Process

1) Single-Screw Extruder

Classic recycling lines have a classic structure that includes single-screw extruders with different structural characteristics. The schematic appearance of single-screw extruder line is given in Fig. 2. This process is also known as mechanical recycling.

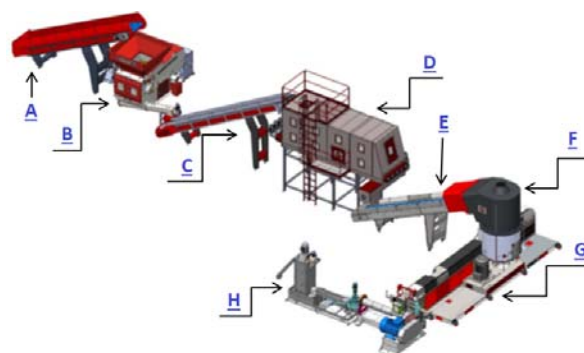


Fig. 2 Single-screw extruder line suitable for mechanical recycling technique

The single screw recycling line, which is given to schematic representation in Fig. 2 and used within the scope of the studies, has a structure composed of 8 different sub-equipment summarized below.

- a) Crusher (Shredder) Feed Conveyor: This equipment serves the purpose of loading waste plastics into the crusher
- b) Single Rotor Crusher (Shredder): This equipment is designed to break down PET/PP-containing polymers commonly found in the automotive industry, including various derivatives such as composite waste. It is positioned before the single-screw recycling extruder. The rotor of this crusher has a length of 1000 mm and a diameter of 500 mm. During initial operation, the rotor operates in the opposite direction for a specific period of time to prevent material blockage.
- c) Crusher Output Auger and Transport Converter: This equipment is responsible for conveying the crushed plastics from the crusher to the Balancing silo.
- d) Balancing Silo: This equipment serves the purpose of achieving a balanced mixture by homogeneously blending the broken plastics. It automatically feeds the waste into the single screw recycling extruder line. The Balancing Silo incorporates an auger transport system, ensuring the recycling extruder's stable and consistent feeding through the feed conveyor. There are five augers in the silo, each with a minimum diameter of Ø250 mm. A 2.2 kW motor powers the augers, while the output auger with a diameter of Ø400 mm is driven by a 5.5 kW motor. If the material reaches the upper limit, as detected by level sensors in the silo, the shredder feed conveyor is automatically stopped.
- e) Extruder Feed Conveyor: This component of the production process facilitates the transfer of materials from the silo where broken plastics are balanced to the recycling extruder.
- f) Condenser: This one represents a single-screw recycling extruder equipped with a condenser. This extruder further reduces the dimensions of the extruded, balanced, and homogenized waste through a series of sequential blades, heating the waste particles and eliminating moisture from the content. With inline and fixed blades, the motor-driven disc generates friction heat to heat the plastic components to the desired temperature. The moisture remaining in the plastics, heated by the friction heat, evaporates, ensuring complete drying. The evaporative discharge system absorbs the moisture generated during this process and is discharged from the condenser boiler. Feeding the plastics to the main screw in a heated state enhances the extruder's efficiency, resulting in energy savings. The condenser drive system is powered by a 200 kW motor.
- g) Single Screw Recycling Extractor: This crucial recycling equipment is responsible for melting and recycling plastic waste from the condenser. The single-screw extruder used in the study possesses a screw diameter of 130 mm and an aspect ratio (L/D) of 38. In a particular region, the utilization of twin gassing technology enables the release of gases generated during the plastic melting process in the

extruder, thereby enhancing the quality of the final granules. The extracted gas passes through a tank equipped with a stainless filter, allowing solid particles to be filtered prior to reaching the vacuum pump. A hydraulic filter with Ø300 mm sieve eyes, featuring a single plate with 2 sieve eyes, is utilized at the outlet of the extruder. This filter effectively eliminates the plastic's non-organic impurities and non-melted agglomerates, resulting in higher-quality granules.

- h) Granulator: This equipment serves the purpose of shaping the melting plastics from the outlet of the extruder, transforming them into granules with a round-scale shape. The granules are subsequently cooled. The granulator can produce plastic granules with diameters ranging from Ø1 to Ø3 mm.

In the mechanical recycling processes, single-screw extruders with lower slip stresses are preferred over twin-screw extruders. However, due to these lower shear stresses, it becomes challenging to conduct studies aimed at enhancing the final properties of the structure or waste through the incorporation of functional additives. Therefore, the primary focus in mechanical recycling processes is to eliminate organic or inorganic components/contaminants, expressed as impurities, from the plastic waste contents and achieve the utmost homogeneity in the resulting plastic granules.

However, as mentioned in the preceding section, the majority of plastic waste in today's scenario comprises mixed content containing more than one polymer. The polymers within this content typically lack compatibility due to differences in thermal and viscosity properties. Consequently, morphological phase separations occur, reducing mechanical and technical characteristics and hindering their utilization. Therefore, these waste materials must be compatibilized in production processes by incorporating various additives such as di-block copolymers, thermopolymers, or copolymers grafted with functional units. This necessitates using twin screw extruders, widely employed in the melt blending technique, to enable the addition of functional contributions during the process. This study integrates classical mechanical recycling processes utilizing single screw extruders with melt blending processes involving twin screw extruders, thereby showcasing a comprehensive approach that can be regarded as an upward transformation technique.

A twin screw expander, illustrated in Fig 3, with a screw diameter of 60 mm and an aspect ratio (L/D) of 57/1, is employed as the extruder line in this study. The twin screw recycling line utilized for melt blending investigations consists of nine distinct sub-equipment components, as summarized below.

2) Twin-Screw Extruder

- a) Automatic Filler (Refill): This equipment is utilized to facilitate the automatic feeding of recycling-based granules and the incorporation of compatibilizer components needed to ensure a seamless production process in the extruder. It serves as the initial stage of gravimetric dosing.

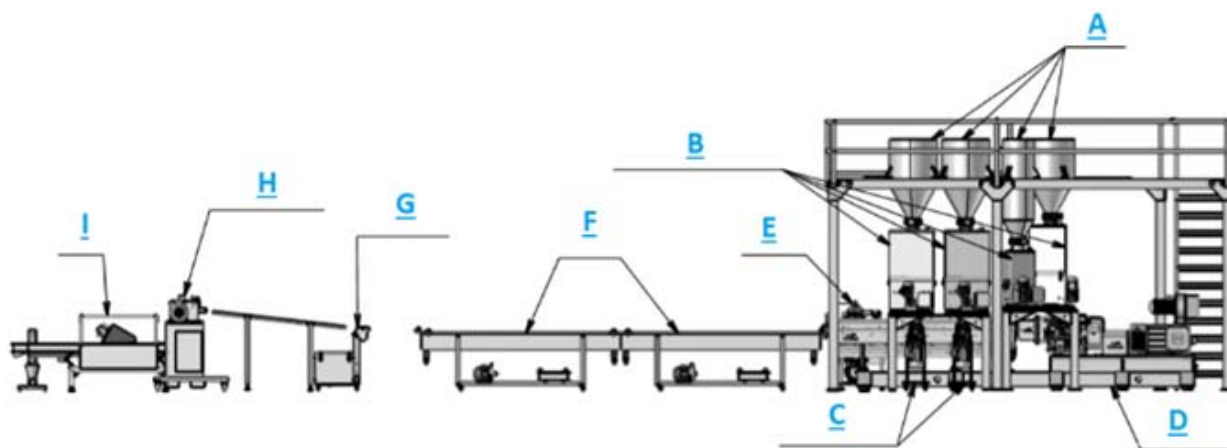


Fig. 3 Twin-screw extruder line for melt blending technique

- b) Gravimetric Dosing: The components constituting the structure in extruder production processes are responsible for accurate gravimetric feeding at desired rates. The extruder line employed in this study consists of five main feed zones and two side feed zones. These feeding sites are integrated with gravimetric dosing, enabling the incorporation of seven different components into the structure on a gravimetric basis.
- c) Side Feed: In the production of plastic blends or composites, it is essential to provide support and maintain/improve the properties of the structure through various contributions. For instance, introducing functional components with low thermal stability, such as triazine-based UV additives, directly into the extruder content via the primary feeds may lead to a loss of effectiveness of these additives. Therefore, the presence of side feeds positioned in different regions becomes necessary depending on the specific additive derivatives to be introduced. The extruder line also incorporates two side feeds situated in different regions. Within the scope of the study, side feed zones were employed to incorporate into the structure compatibilizer components with functional properties, such as thermopolymers and maleic grafted polymers.
- d) Twin Screw Co-Rotating Extruder: It represents the primary recycling equipment where plastic waste from the condenser is recycled by melting. The extruder has a screw diameter of 60 mm, with an aspect ratio of 57/1. The extruder is equipped with five main feeding zones, allowing the inclusion of five distinct components into the structure. Additionally, two side feeding zones are located in different regions to enable the incorporation of specific materials.
- e) Vacuum: The vacuum system is employed to extract and eliminate gas-form degradation products that are created as a result of thermal-induced interactions within the extruder. Its purpose is to maintain the desired environment within the extruder and effectively remove any undesirable by-products.
- f) Cooling Pool: The 3-meter long water bath is used to

achieve the hardness necessary for easily cutting and transforming the molten thread-like plastic structure, ranging from $\text{Ø}1$ to $\text{Ø}3$ mm in diameter, at the extruder output into granules. This process, also referred to as pasta cutting, marks the beginning of the cutting process. The water inside the bath is consistently maintained at a temperature of $23\text{ }^{\circ}\text{C}$.

- g) Dryer: It is used to dispose of excess water on the surface of plastic thread/pasta that has reached a temperature with the hardness that can be cut from the water bath. With air at a temperature of $23\text{ }^{\circ}\text{C}$, the water on the surface is removed from the structure.
- h) Granulator: This is the equipment used to transform plastics in the form of pasta or thread into round-scale grains. Plastic granules with diameters ranging from $\text{Ø}1$ to $\text{Ø}3$ mm can be obtained.
- i) Sieve: Vibrating sieves, which are utilized for the sieving and separation of fines and agglomerates other than plastic granules ranging from $\text{Ø}1$ to $\text{Ø}3$ mm, obtained after the granulator, are equipped with vibration capabilities. They are used for obtaining granules in a homogeneous size.

The content information of the recycling-based polymer blend samples prepared within the scope of this process is given in Table I.

C. Characterization

- 1) Thermal Analysis with Differential Scanning Calorimeter (DSC)

Thermal property characterization and determination of structural compositions related to these properties were conducted using DSC analyses by the ISO 11357-3:2018 standard [22]. All measurements were performed on 50 mL samples to prevent thermal degradation. The DSC analysis was conducted under a nitrogen atmosphere using a dak-1 apparatus.

The analysis involved placing 9-10 mg samples in an aluminum crucible. The samples were heated to $300\text{ }^{\circ}\text{C}$ at a rate of $10\text{ }^{\circ}\text{C}/\text{min}$ to ensure the erasure of their thermal history, followed by a holding period of 5 minutes at this temperature. Subsequently, the samples were cooled to $23\text{ }^{\circ}\text{C}$ at a $5\text{ }^{\circ}\text{C}/\text{min}$

rate to evaluate their crystallization behavior. The crystallization temperature (T_c) and the corresponding

crystallization enthalpy (ΔH_c) of the samples were determined throughout the cooling process.

TABLE I
PREPARED RECYCLING BASED PP/PET BLENDING COMPOUNDS

Code	Textile waste (PET/PP LAMINATED)	Terpolymer (ELVALOY™ PTW)	EPDM.g.MA (ROYALTUF™ 498)	POE.G.MA (LUSHAN™ AR493)
P1	100	-	-	-
P2	95	5	-	-
P3	90	10	-	-
P4	95	-	5	-
P5	90	-	10	-
P6	95	-	-	5
P7	90	-	-	10

After this phase, known as melt crystallization behavior, the samples were hold at 23 °C for 5 minutes, providing sufficient time for equilibration. The samples were then heated again to 300 °C at a 5 °C/min rate. During this heating process, the melting temperature (TM) of the samples and their corresponding melting enthalpy (ΔH_m) were determined.

2) Morphological Analyses with SEM

The morphological properties of the prepared samples were examined with the FEI Quanta FEG 450 brand SEM. Before the examination, the samples were broken with liquid nitrogen, the broken surfaces were covered with gold spray, and images were taken directly on the electron microscope.

3) Tensile and Impact Tests and Mechanical Specification Analyses

The mechanical properties of the specimens, which are essential for structural applications such as tensile strength, elastic modulus, and elongation at break, were made at room temperature according to ISO 527-2:2012, using the Zwick/Roell Z020 universal tensile device [23]. A testing speed of 10 mm/minute was applied to each sample, and a minimum of five different test specimens were used. Another important mechanical property for structural applications, impact resistance, was evaluated using the Izod notched impact strength test in accordance with ISO 180:2019 standard [24].

4) Measurement of Melt Flow Index, Density and Hardness and Physical Property Analysis

Under the physical property analysis of the samples, melt flow index (MFI) values were performed according to ISO 1133-1:2012, density measurements according to ISO 1133-1:2019, and hardness measurements according to ISO 868:2003 [25]-[27].

III. FINDINGS AND DISCUSSION

A. Thermal Analyses with Differential Scan Calorimeter

The determination and documentation of the structural composition of the PP/PET-layered wastes used in this study, both before and after recycling, are significant. Specifically, the ratios of PP and PET components in the waste material prior to recycling processes play a crucial role in determining the types and usage rates of functional additives to be employed in compatibilization processes. Consequently, granular samples

obtained after mechanical recycling with pre-recycled wastes were subjected to DSC analyses for characterization purposes.

Fig. 4 illustrates the DSC diagram of PP/PET laminated automotive textile waste intended for inclusion in the recycling procedures. The diagram exhibits two distinct melting temperatures around 164 °C and 251 °C, as well as two different crystallization temperatures approximately at 122 °C and 210 °C. The DSC analyses clearly reveal the presence of a two-phase structure within the samples. Based on these temperature observations, it can be deduced that the structure primarily comprises PP and PET components.

It is possible to reach many different studies carried out in the past to determine the proportions by weight of the polymer components that form structures that contain such different polymers [28]-[31]. In the study carried out, the use of the method represented by (1) was preferred to determine the rate expressed.

$$\%PP = \frac{\Delta H_m(PP)}{\Delta H_m(PP) + \Delta H_m(PET)} \times 100 \quad (1)$$

% PP represents the weight ratio of the PP polymer in the content, $\Delta H_m(PP)$: The melting enthalpy of the PP phase, $\Delta H_m(PET)$: The melting enthalpy of the PET phase. In short, it is possible to obtain an approximate composition value about the structure's content by dividing the polymer's melting enthalpy value, which is desired to be found in the ratio by weight, by dividing the total melting enthalpy ratio.

Calculations based on (1) show that the waste content has an average composition of 75% PP and 25% PET by weight. The thermal properties of the samples obtained as a result of the recycling processes, supported by different compatibilizers, and the contents calculated according to (1) are presented in Tables II and III.

Based on the data presented in Table II, it is evident that the adapters introduced into the composition to enhance the structural integrity during recycling processes have no impact on the melting temperature of the PET and PP phases. The calculated PP/PET ratio, obtained using (1) prior to recycling, remains at comparable levels even after the recycling processes. Moreover, following the utilization of compatibilizers to reinforce the structure, the DSC diagrams continue to exhibit distinct melting temperatures, indicating the persistence of the two-phase structure.

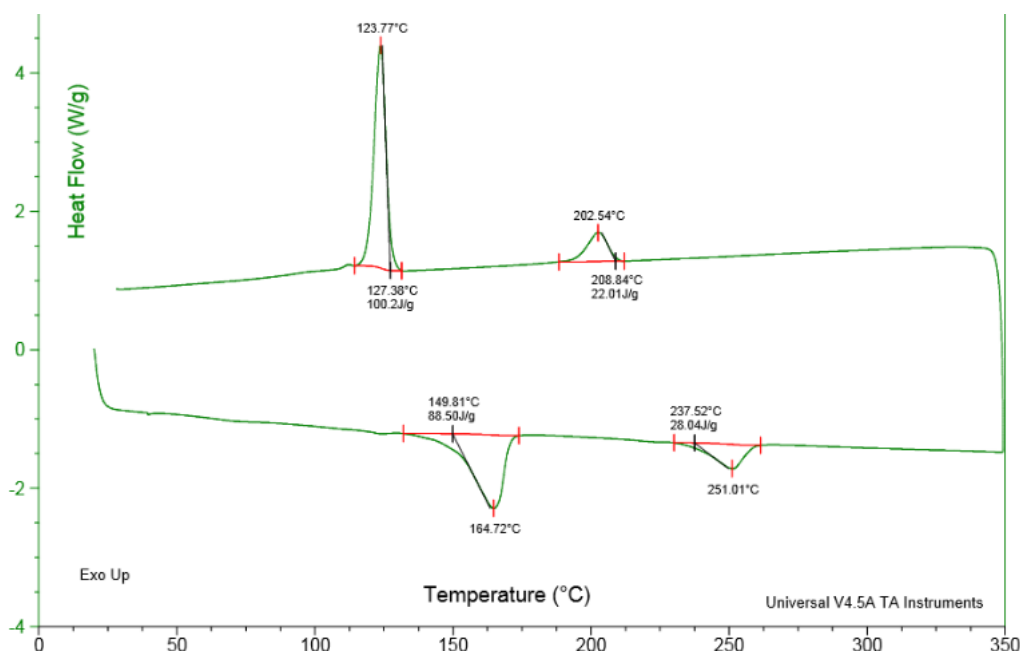


Fig. 4 DSC diagram of automotive textile waste before recycling processes

TABLE II
MELTING TEMPERATURE AND ENTHALPY OF RECYCLING-BASED PP/PET BLENDS

Samples	TM (PP) (°C)	TM (PET) (°C)	ΔH_m (PP) (°C)	ΔH_m (PP) (°C)	PP/PET ratio
p1	166.1	252.7	39.4	11.5	77% PP + 23% PET
p2	166.3	252.1	42.9	12.9	77% PP + 23% PET
p3	165.1	252.3	38.8	11.2	78% PP + 22% PET
p4	165.4	250.9	46.9	14.6	76% PP + 24% PET
P5	165.2	250.1	38.7	12.1	76% PP + 24% PET
P6	166.1	252.1	45.4	13.8	77% PP + 23% PET
P7	165.6	252.6	38.1	12.2	76% PP + 24% PET

TABLE III
CRYSTALLIZATION TEMPERATURES AND ENTHALPY OF RECYCLING-BASED PP/PET BLENDS

Samples	Tm (PP) (°C)	Tm (PET) (°C)	ΔH_m (PP) (°C)	ΔH_m (PP) (°C)
p1	120.8	210.1	46.6	10.1
p2	120.7	204.4	52.8	11.2
p3	121.5	201.9	44.9	9.6
p4	121.4	212.4	53.1	13.7
P5	119.6	209.8	46.9	10.9
P6	120.2	207.1	54.4	11.7
P7	119.8	206.3	45.5	8.7

The analysis of crystallization temperatures, as presented in Table III, reveals that adding a compatibilizer can lead to alterations in the crystallization temperatures, particularly in the PET phase of the blend. With the utilization of Therpolymer and POE.g.MA compatibilizers, in contrast to the EPDM.g.MA type compatibilizer, a proportional decrease in the crystallization temperature of the PET phase is observed as the usage rate increases. Conversely, no significant change in the crystallization temperature of the PP phase is observed.

B. Morphological Analyses with SEM

Polymer-based blending studies frequently encounter challenges related to the viscosity and thermodynamic properties of studied plastics, leading to an incompatible and

inconsistent morphological structure [11]-[13], [32]-[34]. Consequently, achieving desired levels of various technical characteristics, particularly in terms of mechanical properties, can be quite difficult. To address this issue, supplemental components known as compatibilizers are required to enhance the morphological compatibility of the blends.

To assess the current state in this regard, morphological and microstructural analyses were conducted using SEM on the blends produced within the scope of this study. The SEM analyses provided insights into the interaction, compatibility, and phases of the PP and PET components in the sample compositions. The results of the SEM analyses highlight notable differences between the blend samples and the pure components.

Fig 5 depicts SEM images taken at 5000x and 10000x magnification of a recycling-based PP/PET blend sample (P1) that does not incorporate a compatibilizing additive. The images clearly illustrate the two-phase structure identified through DSC analyses. Additionally, the images reveal interfaces between the phases, indicating poor adhesion at the interface and phase separation. Furthermore, the migration of some PET phases from the PP matrix to the surface results in a distinct morphological structure within the blend. This morphological structure is anticipated to hinder the attainment

of satisfactory mechanical properties for practical applications, a hypothesis supported by the data obtained from mechanical

property analyses.

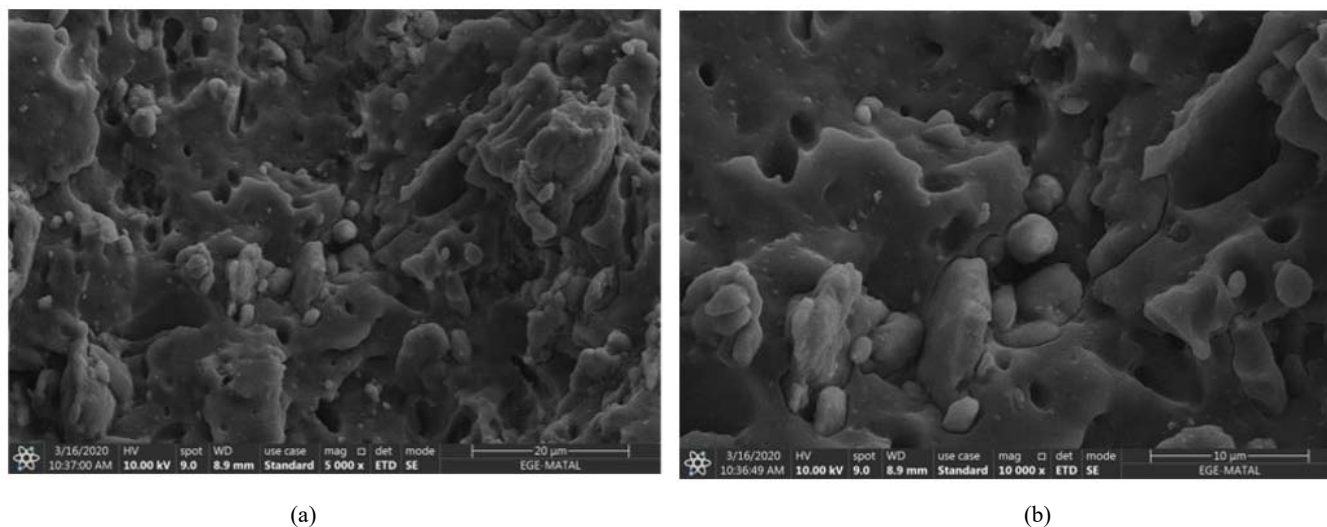


Fig. 5 5000x (a) and 10000x (B) magnification SEM images of a recycling-based PP/PET blend sample without a compatibilizer additive

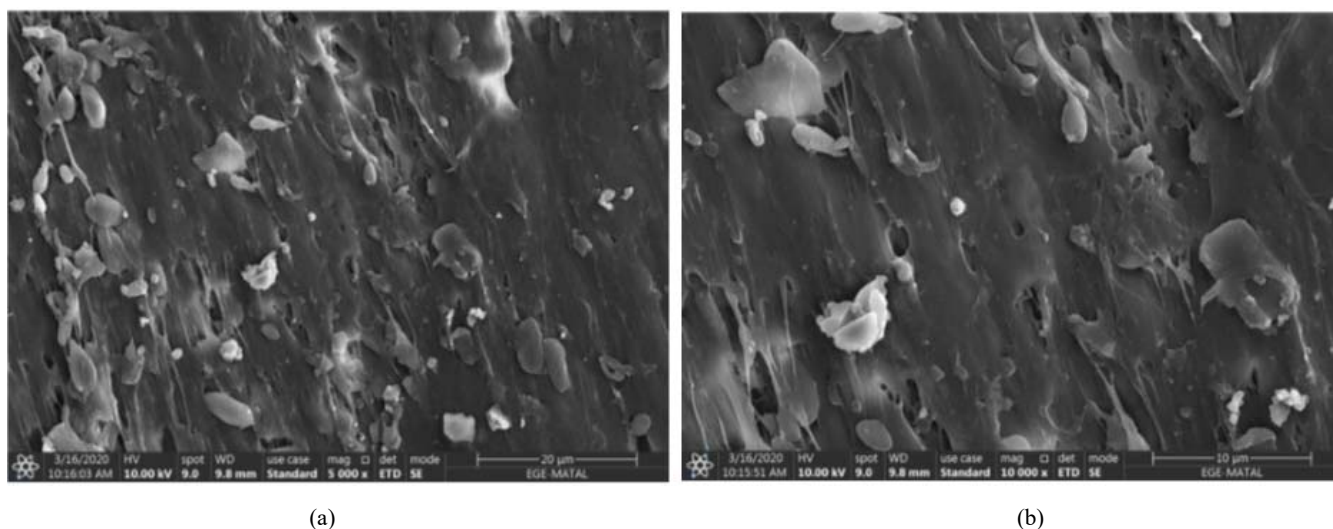


Fig. 6 5000x (a) and 10000x (B) magnification SEM images of a recycling-based PP/PET blend sample compatibilized with 5% Terpolymer

In contrast, the PP/PET blend samples illustrated in Fig. 6, supported by 5% weight of the terpolymer, reveal a notable adjustment of the PET phase within the PP matrix. However, it is worth noting that a stratified morphology is observed in the flow direction, rather than the core-shell morphology commonly associated with the compatibilizer described in the literature [35]. This observed morphological structure, along with the subsequent analysis data, demonstrates a significant improvement in the mechanical properties of the blend.

In the recycling-based PP/PET blend samples depicted in Fig. 7 and compatibilized with 5% weight of EPDM.g.MA, the presence of spherical PET phases dispersed within the PP matrix is visually apparent. The observed morphological characteristics display some resemblance to the blend samples without compatibilizer additives (P1). However, it is noteworthy that the size of the PET phases diminishes, and

there is an absence of migration or separation that would result in micro gaps within the PP matrix. These observations suggest that the EPDM.g.MA component, employed as a compatibilizer, exhibits a partial effect in achieving morphological compatibility. The mechanical analysis data have further validated such partial effectiveness. It is observed that, although PP/PET blends with acceptable mechanical properties can be attained using EPDM.g.MA compatibilizers suitable for various plastic-based products, the mechanical properties remain lower compared to other compatibilizers.

The SEM images shown in Fig. 8 depict the recycling-based PP/PET blend samples compatibilized with 5% weight of POE.g.MA, providing informative insights. These images demonstrate the homogeneous distribution of PET phases within the PP matrix, exhibiting a core-shell morphology, which suggests the potential for enhanced mechanical

properties. This prediction is supported by the subsequent mechanical analysis data presented in the following section.

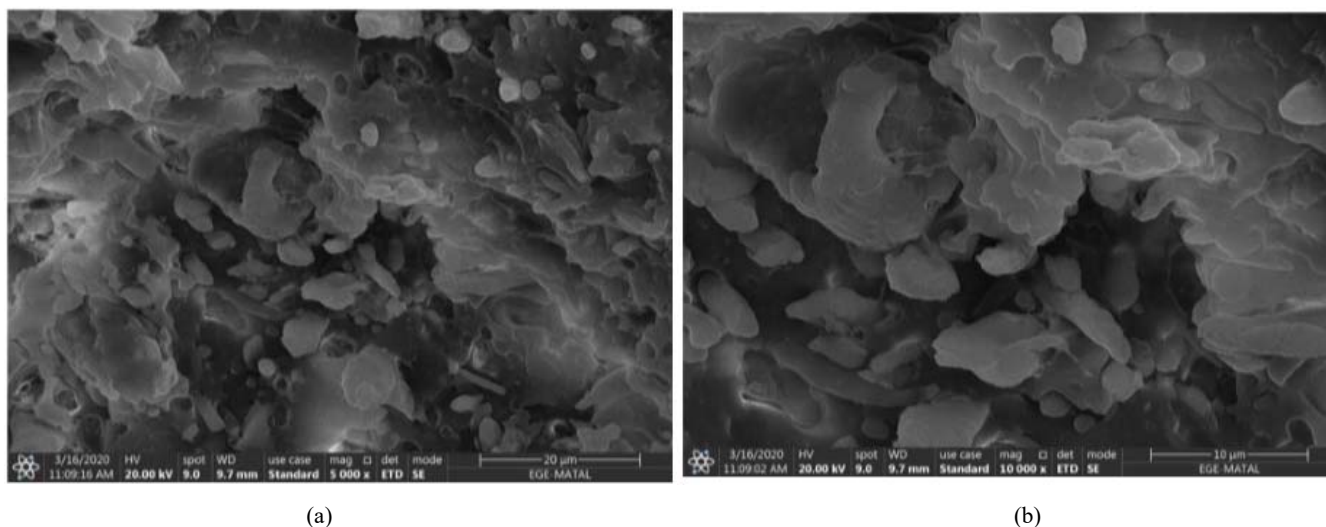


Fig. 7 5000x (a) and 10000x (B) magnification SEM images of a recycling-based PP/PET blend sample compatibilized with 5% EPDM.g.MA

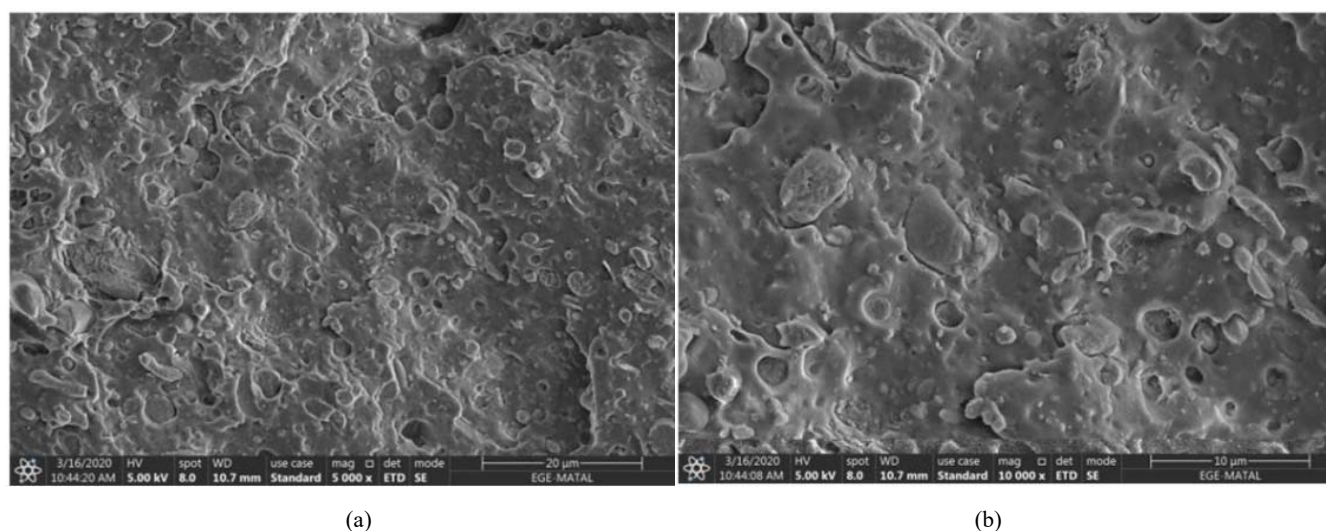


Fig. 8 5000x (a) and 10000x (b) magnification SEM images of a recycling based PP/PET blend sample, compatibilized with 5% POE.g.MA

C. Tensile and Impact Tests and Mechanical Specification Analyses

Table II presents the mechanical properties of the samples obtained through a comprehensive recycling process, where automotive textile furnishings consisting of PET and PP polymers in a laminated or coex structure are supported by various harmonizers.

The results indicate that the samples without harmonizers (P1) exhibit low break elongation and impact strength values, rendering them unsuitable for structural applications. Even simple plastic-based products such as pots or garbage buckets cannot be produced with impact strength values of 2 kJ/m² and elongation at break values of 8%. In contrast, when the structure is supported with the compatibilizers employed in the study, it is evident that the limitations above can be effectively addressed, enabling the attainment of mechanical properties

applicable in various practical scenarios. For instance, samples (P2) incorporating 5% by weight of the terpolymer with the "Ethylene/N-butyl acrylate/Glycidyl methacrylate" structure demonstrate elongation at break of 172% and impact strength values of 128%. These improved mechanical properties make them suitable for the manufacturing of diverse products. Moreover, increasing the compatibilization ratio to 10% by weight allows for achieving even higher values than initially observed.

The observed properties align well with the morphological characteristics derived from SEM analyses, confirming that the PET phase within the PP matrix can be effectively harmonized. The impact strength value of 4.8 kJ/m² achieved with a compatibilizer utilization rate of 6.3% demonstrates that favorable impact strength values can be obtained. Similarly, by increasing the compatibilizer rate to 10%, impact strength

values of 5 kJ/m² can be achieved. These values closely resemble the impact strength values exhibited by many

pristine/original PP-based raw materials.

TABLE IV
MECHANICAL PROPERTIES OF PREPARED RECYCLING BASED PP/PET BLENDS

Samples	Tensile Strength (MPa)	Yield Strength (MPa)	Yield elongation (%)	Elongation at Break (%)	Elastic Module (MPa)	Impact strength (KJ/m ²)
P1	17.7	17.5	3.15	8.5	1523	2.1
P2	20.3	18.1	3.5	23.2	1192	4.8
P3	18.6	17.8	3.8	52.3	1020	6.3
P4	17.8	17.6	3.6	21.9	1078	4.1
P5	15.8	14.9	3.7	42.4	950	5.2
P6	19.8	18.3	3.8	31.3	1035	5.4
P7	18.5	17.7	3.9	58.9	980	6.9

When the compatibilizer with 5% and 10% weight of maleic anhydride-grafted Ethylene-Propylene-Diene (EPDM.g.MA) structure (P4 and P5) was employed, improvements in mechanical properties were observed, albeit not as significant as those achieved with the thermopolymer. The morphological analysis of these samples (P4 and P5) using SEM revealed that the effectiveness of EPDM.g.MA-based compatibilizers is limited.

The presence of a core-shell morphology, which is considered an indicator of component compatibility in blend studies, was only observed in samples obtained with maleic anhydride-grafted ethylene-octane copolymer (P6 and P7). As a result, it was anticipated that the highest mechanical values would be obtained in these blend samples. Corresponding to this expectation, the results of the mechanical analysis demonstrate that the use of POE.g.MA adapters yields the highest levels of elongation at break and impact strength. With a 5% dosing rate, impact strength values of 6.9 kJ/m² were achieved, while a dosing rate of 10% resulted in an impact strength value of 5.4 kJ/m². These values meet the criteria for many structural applications. Thus, it is evident that the most successful blend samples within the study were those prepared using the POE.g.MA type compatibilizer.

D.Measurement of MFI, Density and Hardness and Physical Property Analysis

Table V presents the technical characteristics of PP/PET blend samples obtained through an integrated recycling process, with the added support of various compatibilizers. These characteristics are expressed in terms of physical properties, including MFI, density and hardness.

The PP/PET blend samples, obtained as a result of recycling, are intended to substitute original PP raw materials. Consequently, the MFI values, which play a crucial role in plastic forming processes, have been measured by the ISO 1133-1:2012 standard for PP materials. Analyzing these values under a temperature of 230 °C and load of 2.16 kg reveals that the MFI values decrease by incorporating compatibilizers into the blend structure. Interestingly, the obtained values are relatively close to each other, ranging from 1.0 to 1.3 g/10 min. Although these values may not be suitable for injection molding or plastic product manufacturing processes, they seem to be suitable for plastic forming operations like sheet formation or thermoforming techniques.

TABLE V
MFI, DENSITY AND HARDNESS VALUES FOR PREPARED RECYCLING-BASED PP/PET BLENDS

Samples	MFI (g/10 min)	Density (g/cm ³)	Hardness (Shore D)
P1	2.3	1.03	75
P2	1.3	1.03	71
P3	1.1	1.02	69
P4	1.7	1.03	70
P5	1.5	1.03	66
P6	1.4	1.03	70
P7	1.0	1.01	68

According to Table V, all the samples display similar density values ranging from 1.01 g/cm³ to 1.03 g/cm³. When comparing these values with the density of PP-derived raw materials (0.89 g/cm³) and PET-derived raw materials (1.38 g/cm³), as well as considering the determined blend content of 75% PP and 25% PET based on DSC analyses, the observed density values align with the expected blend composition.

The hardness values vary depending on the type and proportion of the compatibilizer used. Nonetheless, the hardness generally falls within the range of 68 Shore D to 71 Shore D, which is consistent with commercially available PP copolymers. Notably, the samples incorporating POE.G.MA compatibilizers, which yield the most favorable results according to SEM and Mechanical Analysis, closely resemble many commercial copolymer PP raw materials commonly employed in the industry.

IV. CONCLUSION

This study focuses on the formulation and production methods for recovering automotive textile waste containing PET/PP (Post-product) and post-use (Post-Consumer) materials commonly found in the automotive sector. The goal is to employ the upward recycling technique by substituting original PP raw materials with PP/PET alloys. However, the lamination structure of the waste hinders the separation of incompatible PP and PET phases, which limits the production of high-quality raw materials or products during recovery. To address this challenge, a comprehensive process has been developed by combining mechanical recycling using single-screw extruders and melt blending involving twin-screw extruders. The compatibility of the incompatible PP and PET phases in the waste structure is achieved using three different block and

hanging copolymers, namely Terpolymer, EPDM.g.MA, and POE.g.MA. These copolymers, equipped with functional groups like GMA and MA, require high shear stresses for effective operation. Therefore, the addition of compatibilizers is carried out during the melt blending process using twin-screw extruders.

SEM analysis and mechanical testing, particularly the use of POE.g.MA compatibilizers, demonstrate that inadequate interface adhesion resulting from differences in viscosity and thermal properties can be significantly improved. SEM images confirm that the core-shell morphology, which is crucial for successful compatibilization, can be achieved. The obtained tensile strength of approximately 18 MPa from tensile and impact tests, notched impact strength exceeding 5 kJ/m², and elongation values exceeding 20% indicate that the technical requirements for numerous applications can be met. Consequently, it is evident that, under standard circumstances, automotive textile waste, which would otherwise be disposed of in landfills or incinerated in waste collection plants, can be reintroduced into the economy as a valuable resource through an integrated production process and suitable compatibilizers.

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