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DOI: https://doi.org/10.51573/Andes.PPS39.GS.PBA.1

December 2024

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Citation

Study of HDPE/PET/E-GMA Blends: Toughening, Rigidity, and Thermal and Morphology Behavior

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Abstract: The purpose of this study was to utilize E-GMA as a compatibilizer to enhance the mechanical properties of recycled blends of high-density polyethylene, polyethylene terephthalate, and ethylene-glycidyl methacrylate copolymer (HDPE/PET/E-GMA), with the aim of manufacturing plastic soda crates. The results showed that toughness increased without losing rigidity. Flexural strength improved by 11% and impact strength by 8% when the PET content was 20%, indicating that E-GMA acts as a compatibilizer. These results can be applied to improve the mechanical properties of recycled materials in an upcycling context.

Keywords: HDPE, PET, E-GMA, Blends, Recycled

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Introduction

Current approaches to resource efficiency focus on recycling and a circular economy. Recycling is considered one of the best options in waste management to reduce the impacts of post-consumer plastic packaging waste. Recycling offers opportunities for using polymers in sustainable manufacturing. However, each time a product reaches the end of its life cycle, the recycling process results in a loss of molecular weight and degradation of mechanical properties, primarily due to exposure to environmental factors and high temperatures [1].

As a result, the use of recycled material is limited or restricted to lower performance products. This limitation is particularly significant given that plastics constitute an increasingly large proportion of municipal solid waste worldwide. The bulk of this plastic waste comprises polyolefin-based plastics, such as high-density polyethylene (HDPE), low-density polyethylene (LDPE), polypropylene (PP), and polyethylene terephthalate (PET) [2].

PE and PP alone account for 50% of the 150 million metric tons of thermoplastics demanded globally, with PE making up two-thirds of all polyolefins used. Polyethylene terephthalate (PET) is an important polymer among plastics globally, contributing approximately 8% by weight and 12% by volume of global solid waste due to its desirable properties, applications, and industrial advancements, leading to a rapid increase in its generation [3].

This context highlights the critical need for improved recycling processes and innovative solutions capable of maintaining or even enhancing the quality and performance of recycled plastics. A significant area of research is the development of PET/HDPE blends, primarily focused on improving compatibility due to the substantial differences in their solubility parameters, which naturally render them incompatible. Key issues for high-performance polymer blend production include dispersion and embedding of the dispersed phase, strong interfacial interaction, and morphology design. Factors determining phase morphology include material characteristics and processing parameters. Some authors have shown that ternary blends of HDPE, PET, and E-GMA enhance matrix impact strength, including Lei et al. [2,4-6].

Experimental Section

Materials and Blends

Commercial grade HDPE (PADMEX 65080, MFI=8.0 g/10 min @ 190°C 2.16 kg, Petróleos Mexicanos), PET, and E-GMA copolymer (Lotader AX8840, MFI=5.0 g/10 min @ 190°C 2.16 kg, SK Functional Polymer) were used in this work to analyze their individual properties, prepare binary blends (HDPE/PET 95%/5% w/w, HDPE/E-GMA 95%/5% w/w, PET/E-GMA 95%/5% w/w), and prepare ternary blends (HDPE/PET/E-GMA 85%/10%/5% w/w, HDPE/PET/E-GMA 75%/20%/5%

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w/w, HDPE/PET/E-GMA 65%/30%/5% w/w). The decision to set the concentration of E-GMA at 5% w/w and use PET only up to 30% w/w was based on the previous work reported by [5], which showed that using more than 5% of E-GMA does not improve mechanical properties, and adding more than 30% of PET reduces the impact strength of the blend.

Blend Preparation

For the preparation of binary and ternary blends, the components were weighed at the designated mass ratio and blended in a Labtech Twin Screw extruder (LTE-2640, 26 mm diameter, L/D 40, OD/ID 1.3) at 280°C (flat profile) and 800 RPM. All the blends were produced at the same feed rate of 25 kg/h. The screw configuration was the same for all the blends and included distributive (30°, 60°) and dispersive (90°) mixing elements. A degassing vacuum zone at the end of the extruder was used to produce 30 cm Hg vac. In order to get rid of residual moisture, PET was dried prior to extrusion at 120°C for 4 hours in a Motan Bin S 15L dryer.

Test Specimen Preparation

All the test specimens for Izod impact strength and flexural tests were injection molded using a 35 A Boy Machine with an Axxicon AIM Quick Change mold and an ISO type B insert. All individual materials and blends were injected using the same parameters. The temperature profile was set to 250°C in the feeding zone and nozzle, and 280°C in the middle zone. Apart from adjusting the temperature due to the PET content, all injection molding parameters were defined according to the ISO 1872-2 standard. The mold temperature was 40°C. The theoretical flow front velocity was 100 mm/s in the cross-section of the test specimen. The specific hold pressure was set to 575 bar for 25 seconds and an additional 10 seconds for cooling. The tangential velocity during plastification was 0.2 m/s. The cycle time was approximately 40 seconds.

Impact Strength Test

The Izod impact tests were performed in a Tinius Olsen IT 504 pendulum according to ISO 180, with a type A notch. The capacity of the pendulum was 2.8 J.

Flexural Properties Test

The flexural properties were measured in a Shimadzu AGS-X 5 universal testing machine, according to ISO 178, at a test speed of 2 mm/min and a 64 mm span.

Scanning Electron Microscope Observation (SEM)

SEM Micrographs were obtained in a JEOL JXA8530F electron microscope. Fractured specimens from the test were polished. After this, the samples were gold coated in order to increase their electric conductivity for SEM observation. The operating voltage was 20 KV.

Differential Scanning Calorimetry Measurement (DSC)

DSC tests were conducted according to ASTM D3418-15 using a TA instrument DSC Q200. The samples were heated from 25°C to 280°C at a heating rate of 10°C/min, then cooled at the same rate to 40°C and reheated until reaching the melting temperature.

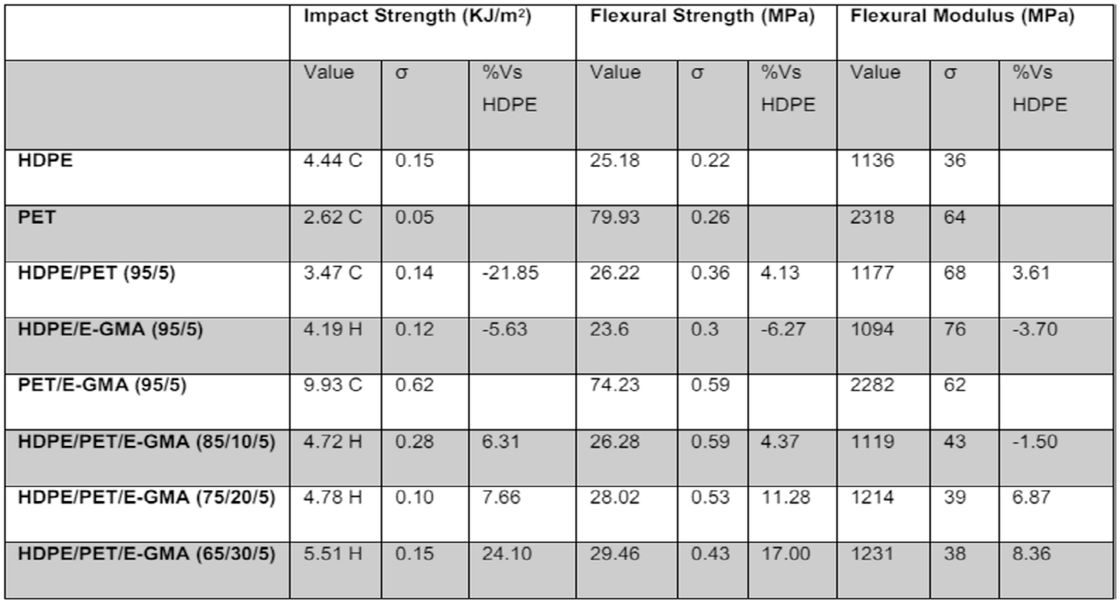
Dynamic Mechanical Analysis Measurement (DMA)

A TA Instrument DMA 850 was used in dual cantilever mode. The samples were heated to approximately 150°C at a heating rate of 5°C/min. The oscillatory frequency was 1 Hz, and the amplitude was 35 μm.

Results and Discussion

Mechanical properties. Table 1 shows the results of the mechanical properties.

Table 1. Mechanical properties of binary and ternary blends.



	Impact Strength (KJ/m^2)			Flexural Strength (MPa)			Flexural Modulus (MPa)		
	Value	σ	%Vs HDPE	Value	σ	%Vs HDPE	Value	σ	%Vs HDPE
HDPE	4.44 C	0.15		25.18	0.22		1136	36	
PET	2.62 C	0.05		79.93	0.26		2318	64	
HDPE/PET (95/5)	3.47 C	0.14	-21.85	26.22	0.36	4.13	1177	68	3.61
HDPE/E-GMA (95/5)	4.19 H	0.12	-5.63	23.6	0.3	-6.27	1094	76	-3.70
PET/E-GMA (95/5)	9.93 C	0.62		74.23	0.59		2282	62	
HDPE/PET/E-GMA (85/10/5)	4.72 H	0.28	6.31	26.28	0.59	4.37	1119	43	-1.50
HDPE/PET/E-GMA (75/20/5)	4.78 H	0.10	7.66	28.02	0.53	11.28	1214	39	6.87
HDPE/PET/E-GMA (65/30/5)	5.51 H	0.15	24.10	29.46	0.43	17.00	1231	38	8.36

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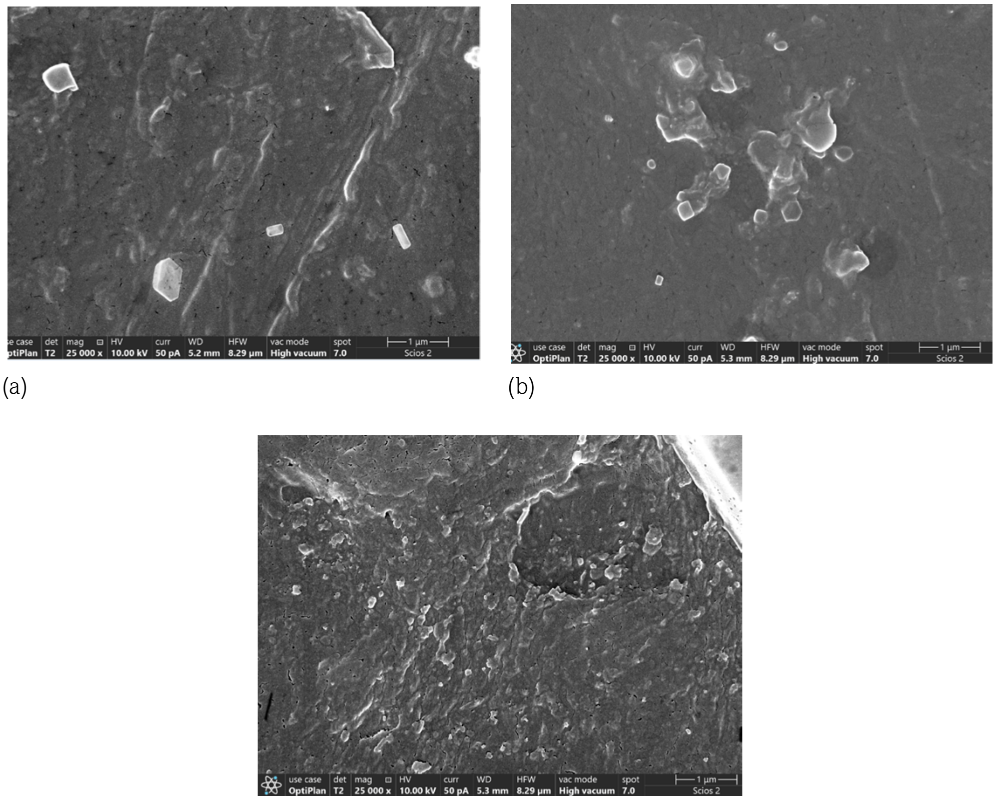
PET/HDPE blends are inherently incompatible due to the significant difference in their solubility parameters [2]. This incompatibility is evident in the impact strength of the binary HDPE/PET blend, which decreased by 21.85% compared to unmodified HDPE. On the other hand, ternary HDPE/PET/E-GMA blends exhibited higher impact strength than pristine HDPE, with a trend of improving in this property as PET concentration increased. Traditionally, adding elastomeric (rubbery) polymers to polyolefins could soften and toughen them, enhancing impact strength but at the cost of reduced rigidity, as expressed by the flexural modulus [7]. In our study, the ternary HDPE/PET/E-GMA blends demonstrated enhanced impact strength and flexural properties compared to pristine HDPE. A similar behavior has been reported by Chen et al. [8] for PP/EPR/HDPE blends, attributed to the reduction of the interparticle distance between the EPR domain, explained by the formation of a core-shell morphology where the PP content is located inside the EPR domain.

Scanning Electron Microscope Observation (SEM)

Figures 1a, 1b, and 1c show the morphology of HDPE/PET (95/5) binary blend, HDPE/PET/E-GMA (85/10/5), and ternary HDPE/PET/E-GMA (65/30/5) blend, respectively.



(a)

(b)

(c)

Figure 1. (a) HDPE/PET (95/5) binary blend, (b) HDPE/PET/E-GMA (85/10/5) ternary blend, (c) HDPE/PET/E-GMA (65/30/5) ternary blend.

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PET dispersed phase within an HDPE matrix can be observed in Figure 1(a). The addition of E-GMA in the HDPE/PET blends improves the dispersion of the PET dispersed phase within the HDPE matrix, as evidenced by a smaller particle size (Figure 1[b], Figure 1[c]).

Analyzing the mechanical properties alongside the SEM images it is possible to correlate the improved impact strength of HDPE/PET/E-GMA (65/30/5) compared to HDPE/PET/E-GMA (85/10/5), which can be attributed to the interparticle distance of the dispersed phase observed (Figure 1[b], Figure 1[c]). While pure PET exhibits low impact strength and poor miscibility in HDPE, PET itself has a toughening effect for HDPE, as can be deduced for the impact strength value of the HDPE/PET binary blend.

The addition of PET to the HDPE/E-GMA blend in the ternary blend demonstrates an excellent toughness, although E-GMA alone does not improve HDPE properties. This suggests a synergistic effect between PET and E-GMA in toughening HDPE. These results indicate that the increased toughness of the ternary HDPE/PET/E-GMA blends is due to E-GMA acting as a compatibilizer, improving the interfacial adhesion between HDPE and PET.

Differential Scanning Calorimetry Measurement (DSC)

The results of the DSC test are shown in Figure 2. As can be seen, the main melting transitions of PET and HDPE remain virtually unchanged compared with HDPE/PET/E-GMA (65/30/5). According to ASTM D3418-15, this minimal displacement in the transition temperature can be attributed to the method´s repeatability. The compatibilizer does not affect the melting temperature and crystallinity of PET and HDPE, which is advantageous because their mechanical properties are not affected.

The melting temperature of E-GMA was detected at 106.02°C, but it was not observed in the ternary blend. This is because the GMA part (8%) reacts with PET to form ester groups [6] Figure 3.

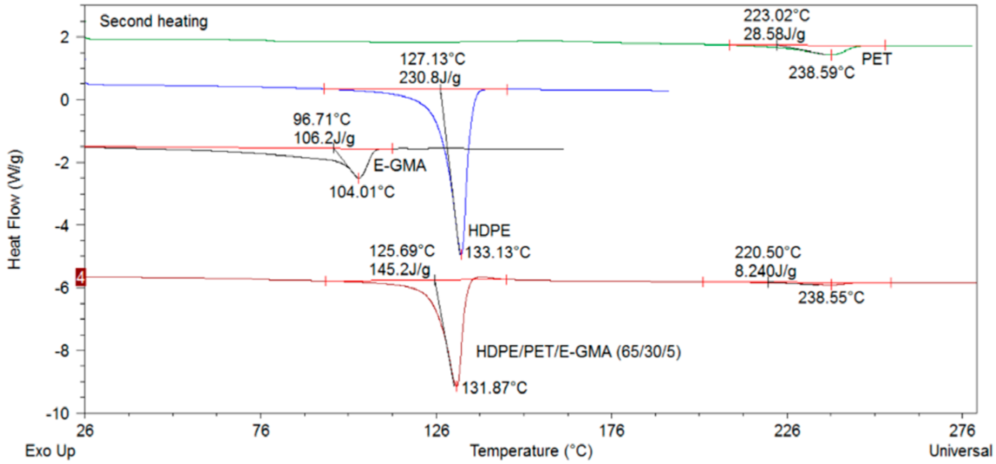

Figure 2. DSC thermogram of PET, HDPE, E-GMA, and HDPE/PET/E-GMA (65/30/5) samples.

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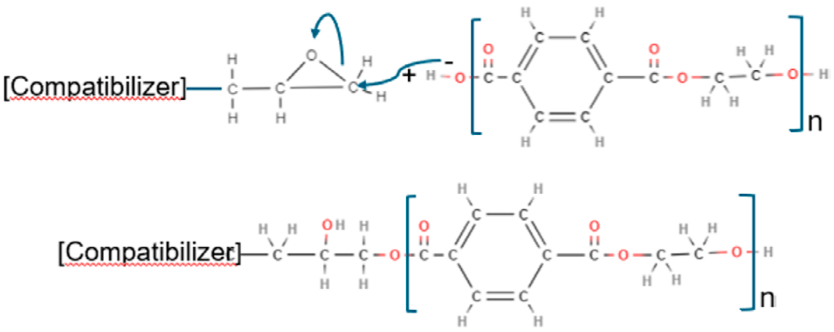

Figure 3. Reaction proposed by Kalfoglou et al. [6].

Dynamic Mechanical Analysis Measurement (DMA)

Figure 4 shows the DMA results for the pristine polymers and the ternary blend. The main α transition of PET is located at 83.2°C, HDPE has its broad α relaxation centered around 51.3°C, and E-GMA presents its main transition at -13.6°C. On the other hand, the ternary blend HDPE/PET/E-GMA (65/30/5) presents two transitions: one at 56.8°C, associated with the HDPE content, and another at 82.5°C, related to PET. Under these test conditions, in the HDPE/PET/E-GMA (65/30/5) ternary blend it is not possible to observe the transition associated with E-GMA. This fact, and the shift in the HDPE and PET transition temperatures towards each other, suggests that E-GMA functions solely as a compatibilizer, assisting in the dispersion of PET in the HDPE matrix.

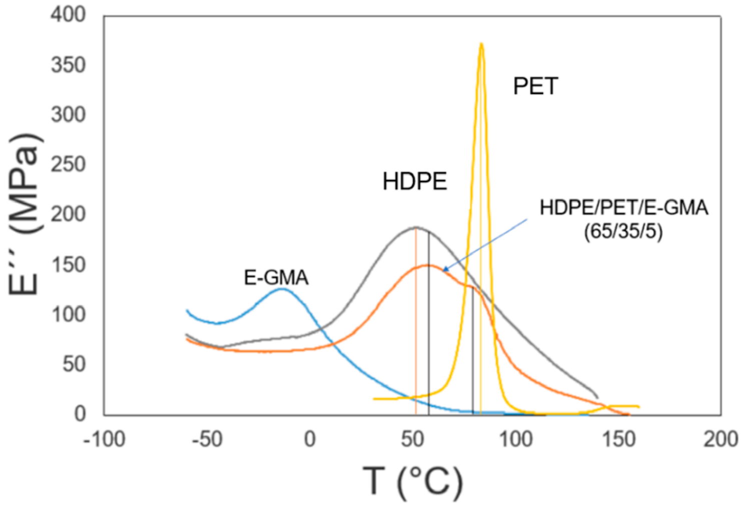

Figure 4. DMA test results showing loss modulus Vs temperature for HDPE, PET, E-GMA and HDPE/PET/E-GMA (65/30/5) (heating rate 5°C/min and oscillatory frequency 1 Hz).

Conclusion

To improve the mechanical properties of recycled materials, the morphology of HDPE/PET/E-GMA ternary blends demonstrates how the compatibilizer aids in the dispersion of PET in the HDPE matrix, improving the flexural modulus and impact strength of recycled materials in an upcycling context. E-GMA acts as a good compatibilizer.

The impact strength of the HDPE/PET/E-GMA ternary blends increases with higher PET content, which can be attributed to the decrease in the interparticle distance of the homogenous dispersed phase, as observed in SEM micrographs. Additionally, E-GMA helps reduce the size of the dispersed PET phase within the HDPE matrix, a fact also evident in the SEM micrographs. These results will be applied to recycled products, ultimately aiding in the design of materials from an upcycling perspective, which will enable the production of products with enhanced properties.

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