EVALUATION OF THE TECHNICAL FEASIBILITY OF COMMERCIAL COMPATIBILIZERS FOR MULTILAYER FILM RECYCLING

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Abstract

This study aims to evaluate major commercial compatibilizers for the recycling of multilayer films. Samples were sourced from post-consumer and post-industrial materials and processed using standard industrial equipment, including agglomerators, granulators (knife mills), and an extruder for compatibilizer incorporation. The materials were subsequently tested according to the criteria established by ASTM D648 (Standard Test Method for Deflection Temperature of Plastics Under Flexural Load in the Edgewise Position) and ASTM D6110 (Standard Test Method for Determining the Charpy Impact Resistance of Notched Specimens of Plastics). Compatibilizing agents were added at the same concentration and using a consistent incorporation methodology to produce pellets, which were then injection-molded into test specimens for evaluation. The raw material was initially characterized using Differential Scanning Calorimetry (DSC) and Thermogravimetric Analysis (TGA). These techniques were used to assess the basic composition of the material under study, as well as its thermal decomposition behavior and inorganic residue content. Overall, the resulting specimens tested using HDT and Charpy impact tests showed significant changes in certain mechanical and thermal properties, while others remained virtually unchanged. The results indicate that specific compatibilizers demonstrated great potential for the recycling of multilayer films.

Keywords: *plastics recycling, multilayer films, post-consumer packaging.*

Introduction

Plastics recycling is essential for the circular economy, which is based on the continuous reuse and recycling of materials. This practice reduces the reliance on virgin raw materials and promotes efficient resource utilization.

Mechanical recycling is the most widely used method for processing plastics. In this process, the plastic material is agglomerated, washed, dried, compatibilized, and extruded, resulting in pellets that can be used to manufacture new plastic products.

This process comprises the following stages:

- Collection and Sorting: Waste is collected and separated by material type.
- Reclamation: The sorted material is reprocessed to become a secondary raw material.
- Conversion: The raw material is transformed into a new product.

Plastic has revolutionized the way we live, enabling significant advancements in diverse fields, from packaging and sanitary protection to cutting-edge technology. However, improper waste management and incorrect disposal have led to serious environmental consequences [1].

Packaging, which accounts for approximately 30% of household waste, has been identified as a major challenge to sustainability. The pressure for green products and packaging has intensified in recent years, with Europe and Japan leading the search for more eco-efficient solutions [2].

The accumulation of plastic waste poses a severe global environmental problem. When material quality permits, mechanical recycling is the most suitable option for managing these wastes. However, the recycling of plastic films, particularly post-consumer films, presents significant challenges due to their inherent complexity and contamination.

In summary, while the recycling of plastic films involves complex challenges, it also offers significant opportunities for building a more sustainable future. Through investment in research and development, the adoption of new technologies, and an integrated approach, it is possible to overcome these difficulties and promote the circularity of plastics [3].

Packaging design requires a comprehensive understanding of various factors, such as protection, barrier properties, product-packaging interaction, and manufacturing processes. However, a lack of focus on sustainability during the planning stage often results in complex packaging structures that are difficult to recycle. To minimize environmental impact, it is essential to integrate a life cycle perspective, covering the packaging from its production through to its end-of-life.

Throughout their history, plastic materials have widely replaced traditional materials such as glass, metals, and paper. This trend is justified by their versatility, lower energy consumption, barrier properties, chemical resistance, and ease of decoration, all of which contribute to their market success.

The plastic transformation and recycling industries have found national representation and support for more than five decades in the Brazilian Plastic Industry Association (ABIPLAST), since the segment began to develop in the country. The work started in 1967, currently accounts for a total of more than 12,000 jobs and almost 360,000 professionals[4].

As every year, ABIPLAST continues to work on the development of the plastic transformation and recycling sectors. It is with satisfaction, and with hope, that we see that the good debate has gained momentum and that there are new efforts to enhance the role of industry with concrete measures in national development, guaranteeing a sustainable future for everyone and not just for niches in the productive sector[4].

All over the world, industrial policies have considered the green economy a national strategy to guarantee a more sustainable future for humanity. The same is happening in Brazil, with the debate on ecological transition and reindustrialization. Support for growth includes the main points to be encouraged: the carbon credit market and the use of renewable energy sources, such as the production of solar panels and the increased participation in exports of products from properly managed forests. Recycling and reverse logistics are no longer an option [4].

For years, the concrete implementation of the Circular Economy in the production chain has been at the top of ABIPLAST's priorities. The entity works in this direction, developing together with its associates, actions that prepare the sector for the current reality, advancing towards effective results. The circular economy, conscious production and consumption, demand new applicability of plastic material, which are adding greater value to processed products, leading to innovations both in raw materials and products as well as in processes and business models. Innovation, by the way, has been the beacon that guides the course of ABIPLAST's actions, with the objective of maintaining plastic products, including those that use recycled content, as the best solution for many human needs, integrating plastic material to new demands and market trends[4].

This study aims to identify the compatibilizers that exhibit superior thermal and mechanical properties. This will enable expanded testing on the most promising candidates, in pursuit of optimizing their thermal and mechanical performance. These properties are of high value to the recycling market, and achieving them could establish the technical and economic feasibility of reusing this material, which would otherwise be sent to landfills.

Experimental

The present work is focused on evaluating the compatibilization of blends formed from multilayer materials, analyzing their behavior relative to a non-compatibilized control sample. For this purpose, the methodology involved using ten different commercial compatibilizers, all at a consistent loading level.

Materials

The samples were collected from household waste, consisting of metallized monolayer and multilayer materials from both post-consumer and post-industrial sources.

An effort was made to include a wide variety of materials, sourcing from both post-industrial and post-consumer streams. This approach yielded a broad range of base materials and, consequently, a diverse set of resulting blends.

The rationale for this selection is to simulate the actual waste streams of materials that are not currently viable for industrial recycling due to their inherent complexity.

Samples Preparation for Extrusion and Compatibilization

The collected material was agglomerated and, after cooling, subsequently ground and dried. This step prepared the material for its characterization, including polymeric composition and inorganic content analysis, and for the subsequent compatibilization process.

Compatibilizers

- Masterfil Coupling Agent/Flexibilizer – M9593 is a (compatibilizing) agent designed to improve the interaction between polyolefins (PP and PE), as well as with other thermoplastic resins.
- OREVAC® CA100 is a coupling agent functionalized with maleic anhydride, used as a compatibilizer.
- POE NT015 B MA POLYOCTENE, grafted with maleic anhydride, is a chemically functionalized ethylene-polyoctene with a high maleic anhydride content.
- SCONA TSPE 2102 GAHD is a coupling agent for polymers and polymer blends, functionalized with maleic anhydride.
- SCONA TSPOE 1002 GBLL is a coupling agent and impact modifier recommended for PET and polyamides; it is functionalized with maleic anhydride.
- SCONA TSEB 2113 GB is an ethylene butyl acrylate-based coupling agent, functionalized with maleic anhydride.
- Engage 8100 is a coupling agent based on an ethylene-octene copolymer.
- RETAIN 3000 is a compatibilizer that promotes adhesion between polar and non polar materials.
- TK-COMP® PR-3D is a compatibilizer and impact modifier used in the recycling industry.
- TK-COMP® AR493 is a functionalized TPE-based compatibilizing agent for polymers such as polyamides, among others.

Extrusion and Injection

All samples were prepared with a 5% addition of each additive and then extruded to yield pellets. Specimens for Charpy impact and HDT tests were molded on a manual mini-injection molder.

Methods

Differential Scanning Calorimetry (DSC) is a technique used to measure the variation in heat flow into or out of a sample as it is subjected to a controlled temperature program. This variation is analyzed as a function of temperature or time. In this case you can identify many polimers and use this information, Additionally, Thermogravimetric Analysis (TGA), as per the Standard Test Method for Compositional Analysis by Thermogravimetry, was employed to determine the sample's thermal decomposition and inorganic residue contente.

The Charpy Impact test determines a material's toughness by measuring the energy absorbed during a sudden impact without fracture. Results are reported in Joules (J) or normalized by the sample's cross-sectional area (J/m or kJ/m²).

The HDT analysis was performed according to the ASTM D-648 standard and evaluates a material's capacity to resist deformation under load at elevated temperatures. A higher HDT value signifies greater thermal resistance and better dimensional stability under heat

Results and Discussion

Based on the experimental findings, while the compatibilizer TK-COMP AR493 exhibits the highest impact resistance, its thermal performance is comparatively limited. In contrast, TK COMP PR 3-D emerges as the most promising candidate, offering a well-balanced combination of properties, with an impact resistance of 3.16 kJ/m² and a Heat Deflection Temperature (HDT) of 114 °C. Following closely are RETAIM 3000 and SCONA TSPE 2102 GAHD, both demonstrating satisfactory thermal stability (HDT of 102 °C) along with acceptable impact resistance values (2.76 kJ/m² and 2.66 kJ/m², respectively). These results highlight the need for further investigation into these promising compatibilizers to optimize their performance in thermoplastic blends.

Figure 1 shows the TGA/DTG curves obtained under inert atmosphere, with heating from 25 °C to 800 °C. The material remained thermally stable up to ~350 °C. Between 350 °C and 500 °C, a major mass loss occurred, indicating polymer matrix degradation. Beyond 500 °C, stability was observed

with ~20% residual mass, attributed to inorganic fillers or carbonaceous residue from pyrolysis. The DTG curve revealed a single degradation peak at ~460 °C, suggesting a main decomposition stage, typical of thermoplastics without volatile additives. These results indicate thermal stability up to 350 °C, maximum degradation rate at 460 °C, and the presence of mineral fillers, corroborated by literature [5,6].
Figure 1 also presents the DSC curves from two consecutive heating cycles (-100 °C to 400 °C) under inert atmosphere. In the first heating, a baseline shift between 80–100 °C marked the glass transition (T_g), followed by an endothermic peak from 170–200 °C related to melting of the semicrystalline phase. The second heating confirmed the T_g and showed a sharper, lower-intensity peak, indicating the elimination of thermal history and internal stress, leading to a stabilized molecular structure. The material thus exhibits T_g around 80–100 °C, melting behavior near 170–200 °C, and a semicrystalline PA6 structure with thermal stabilization upon reheating.

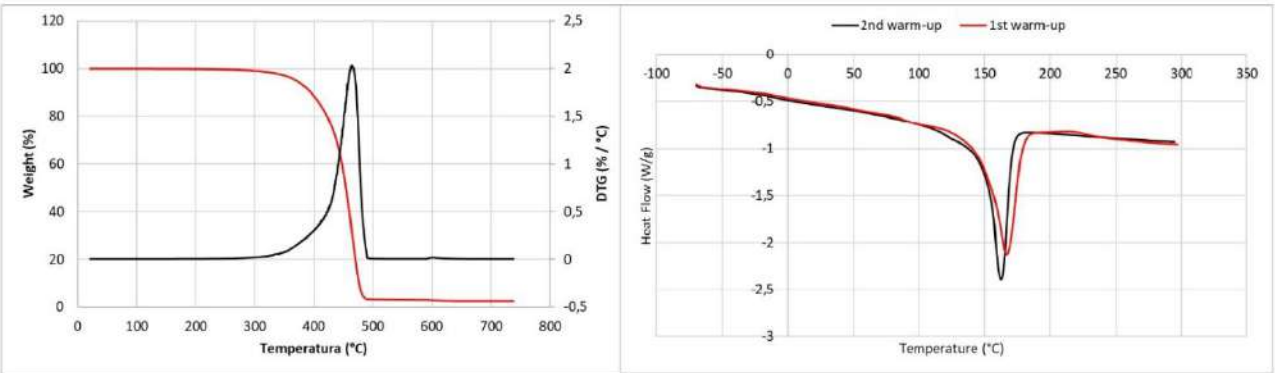

Figure 1: DSC curves and TGA curves

Results of Charpy Impact test are reported in Joules (J) or normalized by the sample's cross-sectional area (J/m or kJ/m²) (Fig 2).
According to the ASTM D6110 the TK-Comp AR compatibilizer delivered the best performance regarding impact resistance, while the other materials performed similarly to the reference material. The main exceptions were Orevac CA100, which showed a significant decrease, followed by SCONA POE 1002 GBLL. While this test alone cannot confirm incompatibility, a significant drop in performance is observed.
The HDT analysis was performed according to the ASTM D-648 standard and evaluates a material's capacity to resist deformation under load at elevated temperatures. A higher HDT value signifies greater thermal resistance and better dimensional stability under heat (Fig. 2).

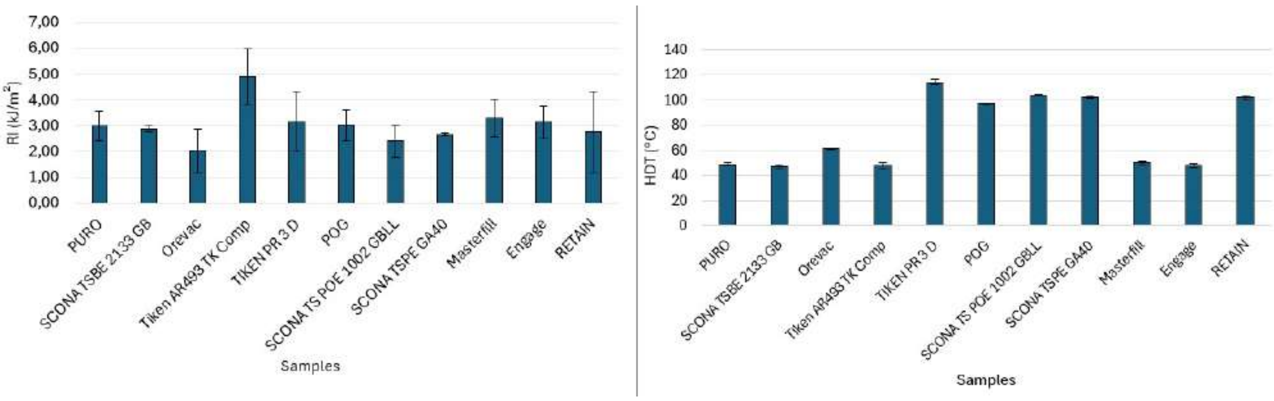

Figure 2: Results of heat deflection temperature analysis

According to the results and when compared to the reference, a significant improvement is noted for the compatibilizers TK-COMP PR 3D, POE NT015B MA, SCONA TS POE 1002 GBLL, SCONA TSPE GAHD, and RETAIM 3000. The other materials showed no improvement.

Conclusion

Based on this analysis, it is recommended that additional studies be carried out with these three compatibilizers for the recycling of multilayer materials, ensuring that the studies simulate the recovery methods currently practiced by Brazilian industries.

The compatibilizer functionalized the polymer, leading to significant enhancements in its physicochemical and mechanical properties. Its mechanism of action involved introducing reactive functional groups that formed chemical bonds or intermolecular interactions between the phases of the system, thereby improving the compatibility and cohesion of the polymer matrix.

TK-COMP® PR-3D offers the best balance between mechanical performance (impact resistance) and thermal resistance (HDT), making it the ideal compatibilizer, its efficiency as a compatibilizer is related to its chemical structure, which contains reactive functional groups grafted onto the main polymer chain. These functional groups act as 'chemical bridges' between the different phases of the polymer blend (e.g., between polar and non-polar polymers), promoting effective interfacial adhesion."

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