



Effects of Single and Binary Compatibilization on the Mechanical Properties and Thermal Behavior of Recycled PA66 and PPE Blends

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Abstract: Various compatibilizers, including PPO-g-MAH, SEBS-g-MAH, POE-g-MAH, SMA-1000, and E-MA-GMA, were incorporated into a 60/40 rPA66/PPE blend to investigate their effects on mechanical properties and thermal behaviors. The mechanical properties varied by compatibilizer type. At a 5 wt% compatibilizer content, the PPO-g-MAH-compatibilized blend exhibited higher tensile and flexural strengths, elongation at break, and modulus than the unmodified rPA66/PPE blend. The addition of POE-g-MAH yielded the highest impact strength of 8.5 kJ/m² owing to its elastomeric backbone structure. Both tensile and impact properties improved simultaneously through binary compatibilization using two different compatibilizers. The compatibilization restricted segmental mobility in the PPE-rich phase and enhanced interfacial interactions within the rPA66/PPE blends, thus increasing the glass transition temperature to 212°C for PPO/SEBS and PPO/POE. These findings demonstrate that the strength, ductility, impact resistance, and thermal transition of rPA66/PPE can be optimized by adjusting the molecular structure and reactive-compatibilizer combination.

Keywords: recycled polyamide 66, poly(phenylene ether), polymer blend, reactive compatibilizer, mechanical properties

Introduction

With the industrial transition toward carbon neutrality and a circular economy, coupled with increasingly stringent environmental regulations, upcycling technologies that convert post-use or process-generated polymer resources into high-value engineering materials, rather than low-grade applications, have become critical important. Owing to its intrinsic properties, such as high strength, wear resistance, heat resistance, and processability, utilizing recycled polyamide 66 (rPA66) to develop high-performance plastics is a compelling approach to advancing circular model and industrial applicability.^{1,2} However, rPA66 inherently undergoes molecular weight reduction, end-group alteration, viscosity variation, and severe fluctuation in mechanical performance due to cumulative thermal history, moisture exposure, oxidation, and repetitive processing. These degradative changes increase brittleness and compromise property reproducibility when rPA66 is utilized alone, particularly in components subjected to impact or cyclic

mechanical loading. Therefore, the practical deployment of rPA66 necessitates robust blend design and interfacial control strategies to compensate for property loss and achieve a synergetic balance between strength and ductility.^{3,4}

Poly(phenylene ether) (PPE), which features a high glass transition temperature, outstanding dimensional stability, low moisture absorption, and thermal resistance, holds great promise for mitigating the inherent brittleness of rPA66 and expanding its application spectrum. However, PA66 and PPE are intrinsically incompatible due to their distinct molecular structures and phase behaviors. Specifically, the polar and semicrystalline PA66 conflicts with the weakly polar and amorphous PPE. These structural and chemical disparities trigger macroscopic phase separation, high interfacial tension, non-uniform dispersion during melt compounding. The insufficient interfacial affinity limits stress transfer across phase domains, accelerating interfacial debonding and crack propagation, which ultimately deteriorates the tensile properties, elongation at break, and impact strength of the resulting blend.⁵

Therefore, simple melt blending is insufficient to improve

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rPA66/PPE blend performance, rendering robust compatibilization strategies essential to regulate interfacial interactions and stabilize the dispersed morphology. Such approaches are particularly vital for immiscible system containing recycled polymers, where enhancing interfacial adhesion and stress-transfer efficiency is indispensable for achieving reliable mechanical properties.^{6–10} In this regard, reactive compatibilizers play a critical role in lowering interfacial tension, refining dispersed domains, and strengthening interfacial adhesion. Compatibilizers functionalized with maleic anhydride (MAH) or epoxy groups are of particularly interest, as these functional groups can undergo in situ chemical reactions with the amine or carboxyl groups of PA66. These interfacial interactions induce grafting or anchoring effects at the phase boundaries, thereby altering the stress transfer mechanism and fracture behavior of the blend. These effects are primarily ascribed to localized interaction and enhanced phase compatibility.^{11–14}

In the present study, the effects of reactive compatibilizers on the interfacial compatibility, mechanical properties, and thermal transition behaviors of rPA66/PPE blends were systematically investigated. Notably, a synergistic improvement in mechanical performance was realized by employing a binary compatibilizer system. To isolate the effects of the compatibilizer type, the rPA66/ PPE weight ratio and compatibilizer content were maintained at 60/40 and 5 wt%, respectively. Macroscopic mechanical behaviors were assessed via tensile, flexural, and impact testing, while thermal degradation and phase transition profiles were monitored through thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC). Based on the distinct property variations induced by the different compatibilizers, the underlying structure-property relationships are induced, providing a robust interfacial control strategy for recycled PA66/PPE blends.

Experimental

1. Materials

Recycled PA66 (rPA66, ENPLON 2021BK grade) and virgin poly(phenylene ether) (PPE, ZM035 grade) were supplied by Yang Il Enpla Co., Ltd. (South Korea) and Dalian Zhongmu Chemical Co., Ltd. (China), respectively. The matrix components were melt-blended at a fixed 60/40 weight ratio. Five compatibilizers featuring distinct functional groups and backbone architectures were employed; maleic anhydride-functionalized poly(phenylene oxide) (PPO-g-MAH, SABIC); maleic anhydride-functionalized elastomeric compatibilizers, i.e., SEBS-g-MAH (SCONA TSKD 9103, BYK Chemie GmbH) and POE-g-MgMAH(SCONA TSPOE 1002 GBLL, BYK Chemie GmbH); a styrene-maleic anhydride copolymer with a high maleic anhydride functionality (SMA-1000); an ethylene-methyl acrylate-glycidyl methacrylate random terpolymer (E-MA-GMA, LOTADER AX8900, SK Functional Polymer). These additives were selected to achieve simultaneous impact modification and interfacial compatibilization of polyamide-based blends.^{15,16} All materials were used as received without further purification.

2. Melt Compounding

The compatibilizers were incorporated into the 60/40 rPA66/PPE blend at a fixed content of 5 wt%. Predetermined amounts of rPA66, PPE, and the respective compatibilizers were fed into a twin-screw extruder (JTE-32M, Jireh Tech, Korea) with a screw diameter of 31.6 mm, an L/D ratio of 44, and 11 barrel zones. The barrel temperature profile was set to 160-250-260-260-260-260-260-260-250-250°C from the feed zone to the die. All formulations were processed under identical thermal profiles and constant compounding parameters. The extruded strands were cooled and pelletized to yield composite pellets. To mitigate the adverse effects of moisture absorption inherent to PA66, the pellets were thoroughly dried at 90°C for 4 h prior to molding. Subsequently, the dried pellets were injection-molded into ISO standard specimens using a 150-ton injection molding machine (SELEX E150, Woojin, Korea) under the conditions listed in Table 1. The formulations and sample notations for

Table 1. Injection Molding Parameters for rPP/PPE/Compatibilizer Systems

Barrel Temperature (°C)	Mould Temperature (°C)	Injection Speed (mm/s)	Holding Pressure (MPa)	Cooling Time (s)
260/270/280 (Rear/middle/front)	80	60	60	25

Table 2. Composition and Sample Notations of rPA66/PPE/Compatibilizer Systems

Designation	Composition (wt%)						
	rPA66	PPE	PPO-g-MAH	SEBS-g-MAH	POE-g-MAH	SMA-1000	E-MA-GMA
rPA66/PPE	60	40					
PPO	57	38	5				
SEBS	57	38		5			
POE	57	38			5		
SMA	57	38				5	
GMA	57	38					5
PPO/SEBS	57	38	2	3			
PPO/POE	57	38	2		3		
POE/GMA	57	38			3		2
SEBS/GMA	57	38		3			2

the rPA66/PPE blends are summarized in Table 2.

3. Characterization

Tensile properties were evaluated according to ISO 527-2 using a universal testing machine (UTM, AGS-X, SHIMADZU, Japan) at a crosshead speed of 5 mm/min. Flexural tests were performed in accordance with ISO 178 at a test speed of 2 mm/min to determine the flexural strength and modulus, thereby assessing variations in mechanical stiffness induced by the compatibilizers. Izod impact properties were measured according to ISO 180/1A using a pendulum impact tester (CEAST 9050, Instron, USA) to compare the toughening efficiency and crack propagation resistance under the distinct compatibilizer systems. A machine-cut Type A V-notch was introduced to each specimen, featuring a notch angle of 45°, a depth of 2 mm, and a root radius of 0.25 mm, leaving a remaining specimen width of 8 mm. All specimens were conditioned at 23°C and 50% relative humidity for 24 h after molding, and the average values of at least five independent measurements were reported.

Thermal stability was evaluated using thermogravimetric analysis (TGA, Pyris 1 TGA, PerkinElmer, USA). The TGA measurements were conducted from ambient temperature to 600°C at a heating rate of 10°C/min under a nitrogen atmosphere. The 5% weight-loss temperature ($T_{5\%}$) and residual weight at 600°C ($R_{600^\circ\text{C}}$) were evaluated to compare the onset of decomposition and high temperature residue, respectively. Thermal transition behaviors were determined using differential scanning calorimetry (DSC, DSC 4000, PerkinElmer, USA) under a nitrogen flow of 20.0 mL/

min and a heating rate of 10°C/min. Specifically, the glass transition temperature (T_g) and the melting temperature (T_m) were monitored. Shifts in T_g were utilized as indirect indicators to assess the segmental mobility restrictions and altered interfacial interactions induced by the compatibilizers.

Results and Discussion

1. Mechanical Properties of rPA66/PPE with Single Compatibilizer System

The effect of compatibilizer type on the tensile strength of rPA66/PPE blend is illustrated in Figure 1(a). The neat rPA66/PPE blend exhibited a tensile strength of 59.7 MPa. Upon the addition of 5 wt% PPO-g-MAH (PPO), the tensile strength significantly increased to 73.3 MPa. Owing to the structural affinity for its PPO backbone with the PPE-rich phase and its reactive MAH functionality, PPO-g-MAH acts as a strength-oriented compatibilizer that enhances interfacial adhesion and stress transfer efficiency between the constituent phases.¹⁷ Conversely, the incorporation of other compatibilizers led to reduced tensile strength: 55.6 for SEBS, 57.2 for POE, 49.3 for SMA, and 51.3 MPa for GMA, respectively. These results indicate that while compatibilizers with flexible elastomeric backbones or low-molecular-weight functional groups may induce interfacial interactions, backbone rigidity and affinity with the PPE phase are more critical for sustaining mechanical loads under tensile stress.¹⁸ The elongation-at-break results revealed that the neat blend exhibited limited ductility at 3.8%, whereas the incorporation of PPO-g-MAH significantly increased this value to 17.3%,

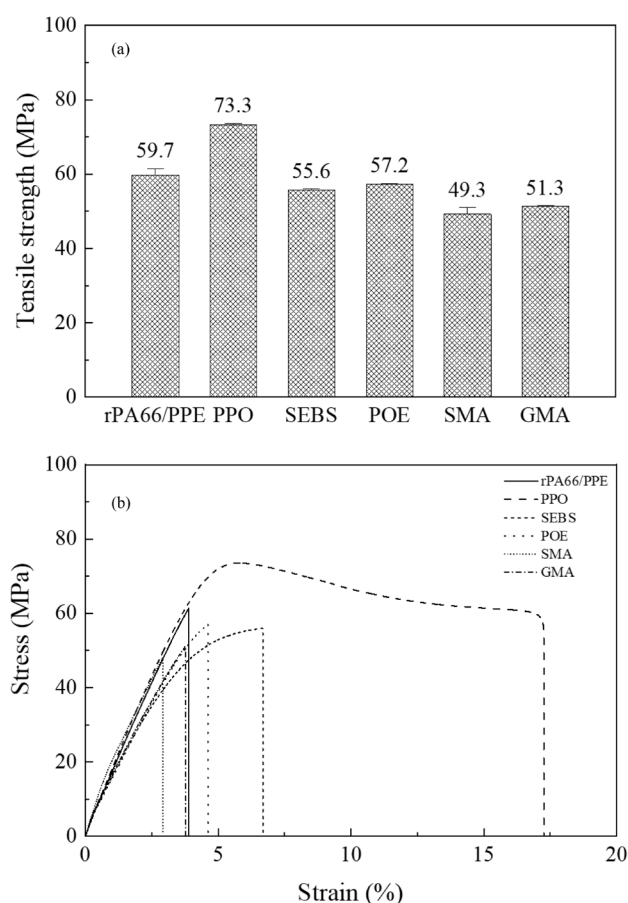


Figure 1. (a) Tensile strength and (b) representative tensile stress-strain curves of rPA66/PPE blends with single compatibilizer.

representing an approximately 4.5-fold enhancement. This confirms that PPO-g-MAH functions not merely as a strength modifier, but as a binary-functional compatibilizer capable of simultaneously improving both strength and ductility.^{19,20} The representative strain-stress curves are illustrated in Figure 1(b). The neat blend fractured at a low strain, demonstrating characteristic brittle behavior. In contrast, the compatibilized systems exhibited varied tensile responses governed by their backbone structure. Notably, the blend with PPO-g-MAH displayed both high maximum stress and extended fracture strain, proving that it simultaneously optimizes interfacial adhesion and energy dissipation during deformation.^{21,22}

Figure 2 illustrates the flexural strength and flexural modulus of rPA66/PPE blends as a function of compatibilizer type. The flexural strength of the neat blend was 101 MPa, which increased to 116 MPa upon the addition of PPO-g-MAH. Concurrently, the flexural modulus exhibited a significant enhancement, rising from 1,774 MPa to 2,841 MPa. These results indicate that PPO-g-MAH is highly effective

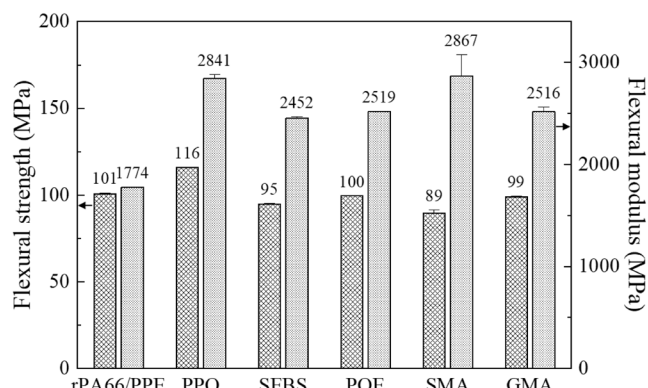


Figure 2. Flexural strength and flexural modulus of rPA66/PPE blends with single compatibilizer.

in increasing stiffness under flexural loading.²³ The blends modified with SEBS-g-MAH, POE-g-MAH, SMA-1000, and E-MA-GMA exhibited lower flexural strengths of 95 MPa, 100 MPa, 90 MPa, and 99 MPa, respectively. Although elastomeric modifiers, such as SEBS-g-MAH and POE-g-MAH, are advantageous for impact-energy absorption, they tend to compromise matrix stiffness. Similarly, while E-MA-GMA may provide auxiliary toughening benefits, its flexural-strength retention was inferior to that of the PPO-g-MAH system. The flexural modulus of all compatibilized blends exceeds 2,400 MPa irrespective of the compatibilizer type, remaining significantly higher than that of the neat blend. This universal improvement implies that the mechanical stiffness is critically governed by the underlying morphology stabilized by interfacial compatibilization.

Figure 3 shows the variations in notched Izod impact strength for the single compatibilizer systems. The neat blend exhibited a low impact strength of 4.1 kJ/m², whereas the

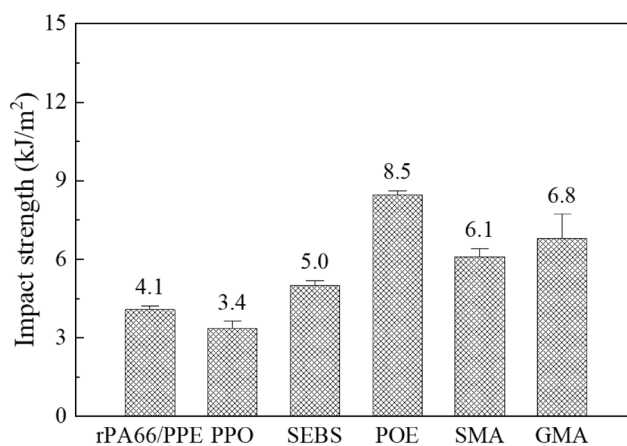


Figure 3. Izod impact strength of rPA66/PPE blends with single compatibilizer.

blend incorporating POE-g-MAH achieved the highest impact strength of 8.5 kJ/m², representing a two-fold improvement. This prominent toughening effect is primarily attributed to the flexible ethylene-octene elastomeric backbone of POE-g-MAH, which effectively dissipates impact energy and suppresses crack propagation.²⁴ The blend modified with styrene-maleic anhydride (SMA) and a random terpolymer of ethylene, methyl acrylate and glycidyl methacrylate (E-MA-GMA) also exhibited higher impact strengths of 6.1 and 6.8 kJ/m², respectively. For E-MA-GMA, the flexible backbone and the potential reaction of epoxy groups may contribute to this difference through energy dissipation and reactive compatibilization, respectively. Although SMA also facilitates interfacial anchoring via its maleic anhydride functionality, the elastomeric toughening efficiency of the POE-g-MAH remained far more pronounced. Meanwhile, the impact strength of the SEBS-g-MAH was 5.0 kJ/m² which is moderately higher than that of the neat blend. Although SEBS-g-MAH is widely recognized elastomeric impact modifier for polyamide-based systems, POE-g-MAH proved to be more effective within the rPA66/PPE matrix. The incorporation of PPO-g-MAH resulted in a reduced impact strength of 3.4 kJ/m², falling below that of the uncompatibilized blend.

2. Mechanical Properties of rPA66/PPE with Binary Compatibilizer System

Figure 4(a) presents the tensile strength of the binary compatibilization systems. The PPO/POE system exhibited the highest value of 67.9 MPa, followed closely by PPO/SEBS at 63.6 MPa, both of which surpassed that of the uncompatibilized neat blend. Meanwhile, the POE/GMA and SEBS/GMA systems showed values of 57.5 and 56.5 MPa, respectively. Regarding the elongation-at-break results, PPO/SEBS and PPO/POE combinations exhibited the values of 6.3 % and 6.8%, respectively. While these results were higher than that of the neat matrix, they remained substantially lower than that of the single PPO-g-MAH system, as illustrated in Figure 4(b). Therefore, whereas the individual PPO-g-MAH acts as a compatibilizer that simultaneously improves both strength and ductility, the binary PPO/POE system serves as an effective alternative to achieve an optimized balance of tensile strength and fracture elongation.

As illustrated in Figure 5, the flexural strength of the binary compatibilizer systems ranged from 81.6 to 87.9 MPa,

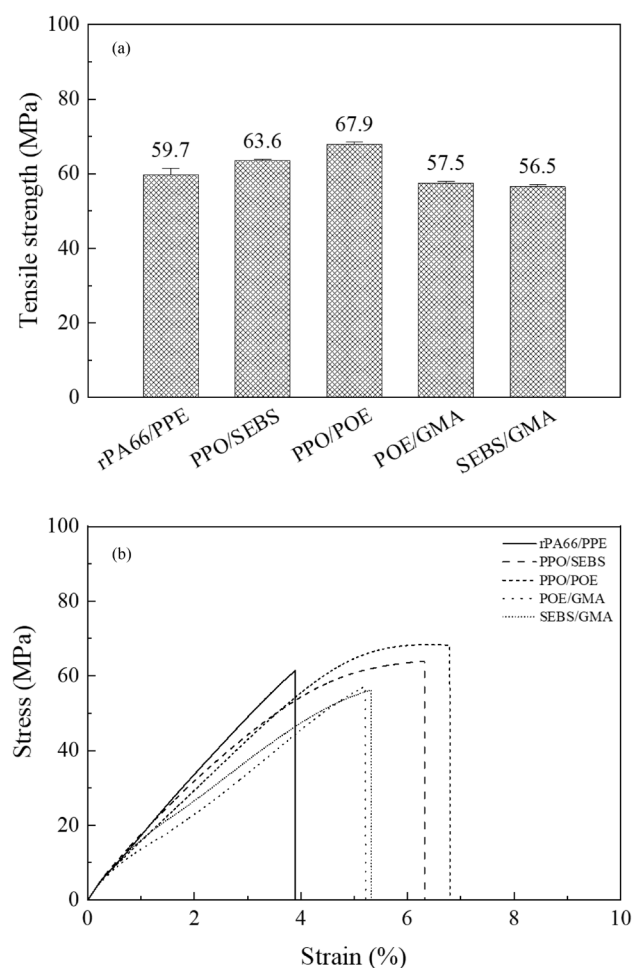


Figure 4. (a) Tensile strength and (b) representative tensile stress-strain curves of rPA66/PPE blends with binary compatibilizers.

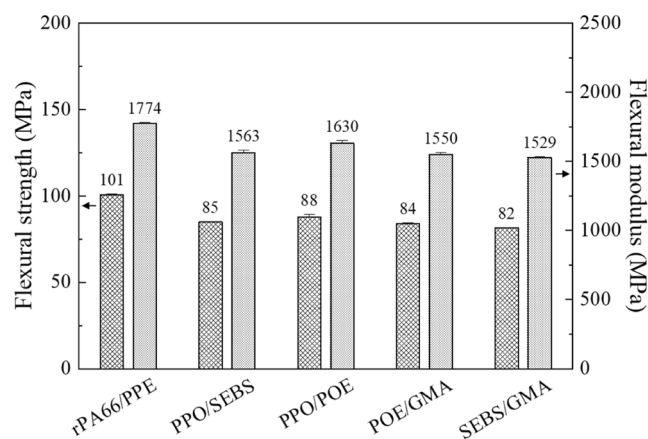


Figure 5. Flexural strength and flexural modulus of rPA66/PPE blends with binary compatibilizers.

and the flexural modulus from 1,529 to 1,630 MPa, both of which were markedly lower than those of the neat blend.

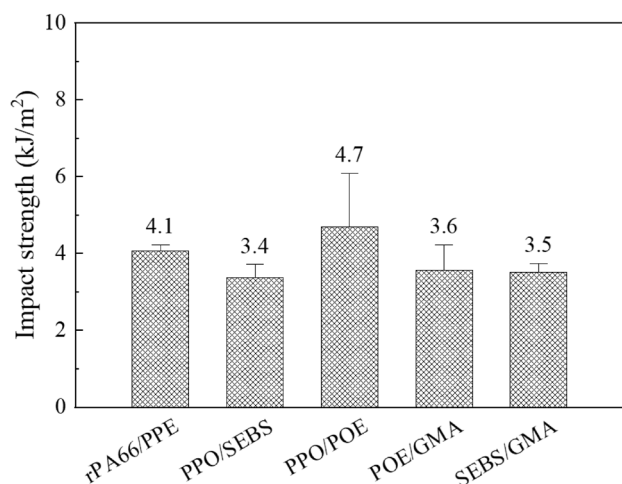


Figure 6. Izod impact strength of rPA66/PPE blends incorporated with binary compatibilizers.

It should be noted that the single compatibilizer system exhibited higher flexural modulus. This reduction is primarily attributed to the simultaneous incorporation of flexible elastomeric or epoxy-functionalized components, which collectively lowered the resistance to flexural deformation. Furthermore, the thermodynamic immiscibility between the two distinct compatibilizers can induce the formation of an isolated, phase-separated domain within the rPA66/PPE blends, acts as structural defects. Therefore, rather than evaluating binary systems merely for stiffness retention, their performance of binary system is further investigated in terms of potential interactions and segmental mobility alterations, as probed by DSC.^{25–27}

Figure 6 shows the Izod impact strength of the binary compatibilization systems. Similar to the flexural strength, the impact values were generally lower than those of the single component counterparts, which typically exceeded 5 kJ/m² with the exception of PPO-g-MAH. While the PPO/POE combination exhibited the highest impact strength of 4.7 kJ/m², the large error bars indicate a significant variation in strength between the test specimens. The PPO/SEBS, POE/GMA, and SEBS/GMA systems yielded values of 3.4, 3.6, and 3.5 kJ/m², respectively. These results indicate that binary compatibilization does not inherently guarantee improved impact properties. Instead, a moderately balanced toughening effect was observed only for the PPO-g-MAH/POE-g-MAH combination. Accordingly, the use of POE-g-MAH is confirmed as the optimum formulation for impact modification.²⁸

Table 3. Thermal Degradation and Transition Temperatures of rPA66/PPE Blends Incorporated with Single and Binary Compatibilizers

Designation	Thermal stability		Transition temperature	
	$T_{5\%}$ (°C)	$R_{600^\circ\text{C}}$ (%)	T_g (°C)	T_m (°C)
rPA66/PPE	416	5	202	254
PPO	416	7	203	254
SEBS	414	6	203	254
POE	416	6	204	253
SMA	405	7	204	252
GMA	415	6	203	253
PPO/SEBS	379	15	212	257
PPO/POE	410	9	212	257
POE/GMA	383	11	208	250
SEBS/GMA	418	9	209	257

3. Thermal Properties of rPA66/PPE with Single and Binary Compatibilization Systems

Thermal stability behaviors of the rPA66/PPE blend are summarized in Table 3. The initial decomposition onset and high-temperature char yield were monitored via the 5% weight-loss temperature ($T_{5\%}$) and the residual weight at 600°C ($R_{600^\circ\text{C}}$). The neat matrix exhibited robust thermal stability, with a $T_{5\%}$ of 416°C and an $R_{600^\circ\text{C}}$ of 5%. Incorporating individual compatibilizers yielded comparable $T_{5\%}$ values: 416°C for PPO-g-MAH, 414°C for SEBS-g-MAH, 416°C for POE-g-MAH, and 415°C for the E-MA-GMA. This indicates that the addition of 5 wt% compatibilizer has a negligible impact on the inherent thermal stability of the rPA66/PPE blend. The slight drop in $T_{5\%}$ value in the $T_{5\%}$ for the SMA-modified blend may stem from the lower molecular weight and higher maleic anhydride density of SMA-1000 copolymer. Nevertheless, it still preserved excellent thermal resistance, maintaining a decomposition onset temperature above 400°C. The residual weight at 600°C ($R_{600^\circ\text{C}}$) for all compatibilized blends ranged from 6% to 7%, which is comparable to that of the neat blend.

The thermal stability of the binary compatibilization systems differed slightly depending the types of compatibilizers and their combinations. The SEBS/GMA system exhibited the highest $T_{5\%}$ value of approximately 418°C, implying that the combination of SEBS-g-MAH and E-MA-GMA forms a relatively stable interfacial architecture. The PPO/POE system was also found to exhibit a relatively high $T_{5\%}$ value

of around 410°C. However, the PPO/SEBS and POE/GMA systems showed lower $T_{5\%}$ values of 379°C and 383°C, respectively, indicating that certain modifier combinations accelerate initial mass loss rather than improve thermal stability. The PPO/SEBS yielded the highest $R_{600^\circ\text{C}}$ value of 15%, while the PPO/POE, POE/GMA, and SEBS/GMA showed values of 8.6%, 10.8%, and 8.9%, respectively. These results demonstrated that high-temperature char residue formation does not necessarily follow the same trend as the initial degradation temperature. Therefore, overall thermal stability should be comprehensively interpreted by considering both the $T_{5\%}$ and $R_{600^\circ\text{C}}$ parameters.

The T_g corresponding to the PPE phase in the uncompatibilized rPA66/PPE blend was observed at 202°C. Upon the introduction of single compatibilizer, this value exhibited a slight increase of 1–2°C. The increase became more pronounced with binary compatibilizers. Specifically, PPO/SEBS and PPO/POE exhibited a T_g values of 212°C, representing an increase of over 10°C relative to the neat blend. These shifts indicate that incorporating PPO-g-MAH with an elastomeric modifier or a GMA-based compatibilizer enhances interfacial interactions, which restricts segmental motion in the PPE-rich phase. POE/GMA and SEBS/GMA also showed higher T_g values of 208°C and 209°C, respectively.

Formulation variations also influenced the melting transitions associated with the PA66 crystalline domains. Although the neat rPA66/PPE blend displayed dual melting peaks at around 246°C (β -crystal) and 254°C (α -crystal), only the major α -crystal peak is presented in Table 3. The T_m of the single-compatibilizer systems remained nearly unchanged at 252–254°C, closely matching the neat blend. However, the binary compatibilizer systems involving PPO/SEBS, PPO/POE, and SEBS/GMA shifted the T_m upward to approximately around 257°C. This behavior indicates that binary compatibilization systems affect the local interfacial environment of the PA66 crystalline domains, thereby altering their melting behavior.

Conclusions

This study systematically investigated the structure–property relationship of single and binary compatibilization systems to clarify the role of each compatibilizer in rPA66/PPE blend. Single-compatibilizer systems exhibited distinct trends in property enhancement. Specifically, PPO-g-MAH

increased the tensile and flexural strengths by up to approximately 23% and 15%, respectively, relative to the neat blend, with an increased flexural modulus observed in all single-compatibilized blends, contrary to the reduction in the binary systems. Meanwhile, POE-g-MAH effectively enhanced impact strength due to the energy absorption and crack-propagation resistance of its elastomeric backbone. For the binary compatibilizer systems, combining PPO-g-MAH and POE-g-MAH established an optimal balance of tensile strength, ductility, and toughness. The upward shift in T_g and T_m observed for the PPO/SEBS and PPO/POE confirm that interfacial interactions significantly constrain molecular chain mobility. These results prove that reactive functional groups, backbone structure, and phase affinity govern the interfacial structures and subsequent macroscopic properties. This finding offers practical insights for the high-value upcycling of recycled PA66-based engineering plastics and advanced interfacial-control compatibilizers.

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Conflict of Interest: The authors declare that there is no conflict of interest.

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