



Effect of Compatibilizer Type on Enhancement in Glass Fiber Reinforced Polyketone and Polyketone/Polyamide 6 Composites

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ABSTRACT

This study investigates the influence of compatibilizer type on the mechanical performance of 30 wt% glass fiber-reinforced polyketone (PK) and PK/Polyamide 6 (PA6) composites. Two compatibilizers, ethylene terpolymer (C1) and maleic anhydride grafted polyethylene (C2), were employed to improve interfacial adhesion and phase dispersion. Results show that the use of C2 significantly enhances impact resistance and elongation at break without severely compromising stiffness. Compared to unmodified composites, C2-modified samples demonstrated up to 113% improvement in impact strength and over 160% increase in strain at break. These findings highlight the critical role of compatibilizer chemistry in enhancing ductility and toughness in glass fiber-reinforced hybrid systems. In comparison with preceding studies, the present work introduces a systematic evaluation of compatibilizer chemistry in glass fibre-reinforced PK/PA6 hybrids. This evaluation reveals that the maleic anhydride group plays a dominant role in energy absorption. These findings demonstrate that selecting an appropriate compatibiliser is crucial for developing high-toughness and recyclable PK-based composites for engineering applications.



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1. INTRODUCTION

Polyketone (PK) is an environmentally sustainable engineering thermoplastic synthesized from carbon monoxide (CO), has attracted increasing interest for a variety of applications, including packaging, fibers and technical components. Given its exceptional equilibrium of mechanical properties and elevated chemical resistance, PK possesses the potential to emerge as a highly promising candidate for utilization in high-performance and multifunctional polymeric materials. The integration of additional polymers, reinforcing fibers, or micro-/nano-fillers within the PK matrix constitutes a robust methodology for the development of innovative PK-based materials, characterized by enhanced and customized properties [1].

Aliphatic polyketone (PK) is regarded as a sustainable and high-performance engineering plastic due to its distinctive molecular structure. Its notable chemical resistance, optimal wear and friction behavior, resistant mechanical strength, and advanced barrier properties make it a highly attractive material for industrial applications, including textile fibers, packaging films, and molded products.

Polymer blending is a widely adopted approach that involves the combination of different polymer types to adapt processing characteristics and unite the advantageous features of each constituent. In this context, studies on blends of polyketone terpolymers with polymers such as poly(styrene-co-acrylonitrile), poly (methyl methacrylate), and polyamide (PA) are critical to understanding the potential of such combinations.

It has been demonstrated by previous research that PK terpolymer exhibits partial compatibility with poly(styrene-co-acrylonitrile) [2], [3], and complete miscibility has been reported in blends involving poly(methyl methacrylate) and poly(styrene-co-acrylonitrile) [4], [5]. The results of this study suggest that homogeneous dispersion between blend components may be a viable method to develop materials with enhanced functional properties.

Asano *et al.* [6] reported that PK terpolymer/PA6 blends exhibit significantly higher impact resistance than expected. In particular, blends containing more than 30 wt% PA6 demonstrated significant enhancements in impact performance under humid conditions. This behavior underscores the remarkable resilience of these blends when confronted with particular environmental conditions.

The enhanced impact strength of the material has been attributed to its water-absorbing amorphous nature, which may facilitate better energy absorption. It is hypothesised that this effect contributes significantly to the overall enhancement in mechanical performance.

Morphological and thermal analyses conducted to investigate molecular-level interactions have revealed partial miscibility between the two polymers. These findings suggest that PK and PA6 are capable of integrating compatibly under certain conditions, with a concomitant positive influence on the blend's

impact properties.

The research conducted by Asano and colleagues provides significant insights into the design and performance of polymer blends under specific conditions, thus offering strategic directions for the development of more robust and effective materials for practical applications.

A more recent study by Zhou *et al.* [7] examined the morphology, miscibility, and mechanical properties of PK terpolymer/PA6 blends. Their observations revealed sub-micron phase domains and a highly stable morphology resistant to coalescence. The blends exhibited superior impact resistance in comparison to the neat components, particularly in formulations where PK formed a continuous phase. Moderate enhancements were also identified in PA6-rich blends. The increased compatibility was attributed to hydrogen bonding between PK and PA6, supported by the observed shift in the carbonyl absorption peak in the blends, indicating intermolecular interactions and favorable phase integration.

Similarly, Kim *et al.* [8] investigated the compatibility of a PK terpolymer with maleic anhydride-modified ethylene-octene rubber (mEOR). The findings of the study indicated a substantial enhancement in toughness, particularly in instances where PA6 was incorporated into the blend. The enhanced compatibility was largely attributed to chemical interactions between the maleic anhydride groups in mEOR and the amine end-groups in PA6, as well as the intrinsic compatibility between PK and PA6.

It is hypothesised that these chemical reactions form a more homogeneous structure within the blend, thus improving integration between mEOR and PA6 and contributing to superior mechanical performance. Furthermore, the inherent compatibility between PK and PA6 is hypothesised to further enhance the overall performance of the blend, thereby enabling synergistic interactions between the polymer components and resulting in enhanced strength and durability.

Despite these advances, a systematic comparison of different compatibilizer chemistries in glass fiber-reinforced PK/PA6 systems is still lacking. Conventional toughening strategies often increase processing costs or reduce stiffness. This study systematically compares two industrially relevant compatibilizers—an ethylene terpolymer (C1) and a maleic anhydride-grafted polyethylene (C2)—under identical processing conditions to elucidate the influence of compatibilizer functionality on interfacial adhesion, energy dissipation, and mechanical performance.

In this study, 30 wt% glass fiber-reinforced polyketone (PK) and polyketone/polyamide 6 (PK-PA6) blends were prepared using a twin-screw extruder to ensure homogeneous dispersion. Two types of 30 wt% glass fibers were used, depending on the PK-to-PA6 ratio. Type 534A glass fibre, surface-treated with a silane-based sizing compatible agent, was utilised for PK-rich compositions. Conversely, Type 568H glass fibre, which has been sized for compatibility with PA6 and PA66, was selected for PA6-rich formulations. The rationale behind this approach

is to enhance fibre-matrix interfacial adhesion and improve the mechanical performance of the resulting composites.

Furthermore, ethylene terpolymer (C1) and maleic anhydride-grafted polyethylene (C2) were utilised as compatibilizers with the objective of enhancing phase dispersion and interfacial interactions within the blends. The objective of this study is to evaluate the mechanical performance of glass fibre-reinforced PK and PK/PA6 composites with different compatibilizers.

This study builds upon our previously published work, which examined the mechanical and tribological behavior of glass fiber-reinforced PK/PA6 composites using a different commercial polyketone grade (M630A). Unlike the former study, which evaluated mechanical, thermal, and wear performance in detail, the present work focuses specifically on the influence of compatibilizer chemistry on mechanical reinforcement, using a distinct PK formulation (M330A) [9].

2. MATERIAL AND METHODS

The aliphatic PK terpolymer (Polyketone M330A) and PA-6 (Durethan B26) used in this study were supplied by Hyosung Corp. (Korea) and Lanxess GmbH (Germany), respectively. The basic physical properties of these polymeric materials are summarized in Table I.

Table I. Physical properties of aliphatic PK and PA-6.

Parameters	Test Method	PK	PA-6
Density (g/cm ³)	ISO 1183	1.24	1.14
Tensile Modulus (MPa)	ISO 527-1, -2	1500	3000
Elongation at Yield (%)	ISO 527-1, -2	22	4
Flexural Modulus (MPa)	ISO 178	1350	2700
Flexural Strength (MPa)	ISO 178	58	100
Izod Impact Strength (kJ/m ²)	ISO 180/A	7	<10
Melting Point (°C)	ISO 11357-1, -3	222	222
Vicat Softening Temperature (°C)	ISO 306	190	200

The PK and PK/PA-6 composites were prepared using two different types of E-glass fibers supplied by Jushi Group (Zhejiang, China). The glass fibers were selected based on their specific characteristics, with a focus on ensuring optimal processability and enhancing the final composite material's performance.

Zinc stearate was used as a processing aid during sample preparation to improve the ease of processing.

2.1. Extrusion Process

Glass fiber-reinforced PK and PK/PA6 composites were prepared using a COPERION ZSK 26 twin-screw extruder. The compounding process was conducted under a carefully controlled temperature profile ranging from 200 °C to 240 °C, with a constant screw rotation speed set at 350 rpm. All formulations and corresponding sample codes are presented in detail in Table II.

Table II. Formulation details of glass fiber-reinforced PK and PK/PA6 composites.

Sample	PK	PA 6	GF	Com	Lub	Tot
PK/GF30-C1	66.7	-	30	3	0.3	100
PK/GF30-C2	66.7	-	30	3	0.3	100
PK-15/PA6-50/GF30- C1	15	49.5	30	5	0.5	100
PK-25/PA6-40/GF30- C1	25	39.5	30	5	0.5	100
PK-15/PA6-50/GF30- C2	15	49.5	30	5	0.5	100
PK-25/PA6-40/GF30- C2	25	39.5	30	5	0.5	100

*GF: Glass Fiber, Compatibilizer (C1): ethylene terpolymer, Compatibilizer (C2): maleic anhydride grafted polyethylene, Lub: zinc stearate was used as a lubricant, Com: Compatibilizer, Tot: Total

The blends prepared using the twin-screw extrusion process were pelletized into granule form. The processing parameters used during extrusion are provided in Table III and Table IV.

Table III. Extrusion barrel temperature profile.

Barrel Zone	Temperature (°C)
Zone 1	200
Zone 2	240
Zone 3	235
Zone 4	235
Zone 5	235
Zone 6	235
Zone 7	235
Die Zone	235
Die Head	240

Table IV. Extrusion process parameters.

Side Feeder Speed (Rpm)	180
Production Rate (kg/h)	20
Material Melt Temperature (°C)	252

2.2. Injection Process

The compound mixtures obtained with the extrusion process were converted into granule form and subsequently molded into test specimens using a JONWAI brand injection molding machine, in accordance with ISO 179 and ISO 527 standards. The flow diagram of the experimental process is shown in Fig. 1.

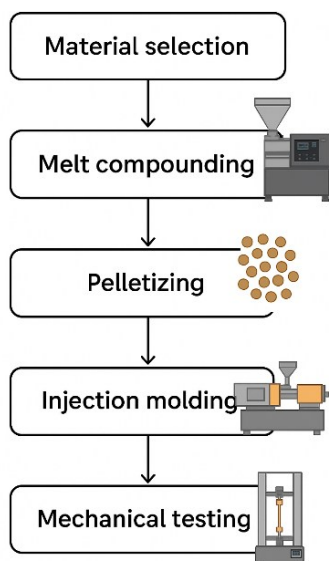


Fig. 1. Flow diagram of the experimental process.

3. RESULTS AND DISCUSSION

In this study, the effects of compatibilizers such as ethylene terpolymer (C1) and maleic anhydride grafted polyethylene (C2) on PK/GF30 and PK/PA/GF30 composite materials were thoroughly investigated.

The mechanical properties of the resulting composites were evaluated through tensile testing, flexural strength testing, and impact resistance testing.

Table V. Mechanical Properties of PK /GF30.

Sample	Tensile Modulus (GPa)	Tensile Stress at Break (MPa)	Tensile Strain at Break (%)	Flexural Modulus (GPa)	Flexural Strength (MPa)	Izod Impact (kJ/m)
PK/GF30	8.6	98	1.7	6.7	142	8.2
PK/GF30-C1	7.8	80	1.6	6.4	112	8.4
PK/GF30-C2	8.4	103	1.9	6.4	140	12.2

Table V provides that the mechanical performance and impact resistance of glass fiber-reinforced polyketone (PK/GF30) composites, formulated both with and without the incorporation of compatibilizing agents—namely, C1 (ethylene terpolymer) and C2 (maleic anhydride-grafted polyethylene).

Both compatibilizers reduced the tensile modulus compared to PK/GF30, indicating lower stiffness. This flexibilizing effect is typically attributed to the elastomeric characteristics of the compatibilizers, which may disrupt the continuity and rigidity of the PK-glass fiber matrix, thereby diminishing the load-bearing capacity of the composite.

Despite this decline in stiffness, the addition of C2 induced a significant enhancement in tensile strength at break, exhibiting an improvement of approximately 5% relative to the unmodified composite. Furthermore, the Izod impact strength increased by more than 50%, thus demonstrating the substantial toughening effect imparted by C2. C2 improves stress transfer by enhancing matrix–fiber bonding and increases energy

absorption during fracture. Conversely, the incorporation of C1 exerted a negligible influence on the overall mechanical performance. No significant alterations were detected in the tensile modulus, tensile strength at break, flexural modulus, or flexural strength. Although a marginal increase in impact strength was observed, it remained significantly below the enhancement achieved with the C2-modified composite.

An evaluation of the flexural properties of the three formulations (PK/GF30, PK/GF30-C1, and PK/GF30-C2) reveals a modest yet discernible improvement in both compatibilizers. However, the formulation containing C2 consistently demonstrated superior mechanical reinforcement, thereby reaffirming its effectiveness in strengthening the composite structure.

The improved performance of the C2-modified system is mainly due to its reactive maleic anhydride groups. It is well established that these functionalities form strong chemical interactions, including covalent bonds and hydrogen bonds, with the polar groups present on the polymer chains or glass fibre surfaces. This, in turn, promotes enhanced interfacial compatibility and mechanical integrity. Conversely, the ethylene terpolymer structure of C1 appears to exhibit a deficiency in interfacial activity, resulting in a comparatively muted impact on the mechanical behavior of the composite.

Table VI. Mechanical Properties of PK/PA6/GF30.

Sample	Tensile Modulus (GPa)	Tensile Stress at Break (MPa)	Tensile Strain at Break (%)	Flexural Modulus (GPa)	Flexural Strength (MPa)	Izod Impact (kJ/m)
PA6/GF30	10.6	168	3.00	8.3	266	11.3
PK-15/PA6-50/GF30-C1	10.6	100	2.54	6.7	144	10.3
PK-25/PA6-40/GF30-C1	7.6	81	2.76	6.3	117	8.3
PK-15/PA6-50/GF30-C2	8.6	122	5.86	6.4	176	15.4
PK-25/PA6-40/GF30-C2	8.0	114	7.84	5.9	163	24.1

Table VI provides a comparative overview of the mechanical and impact properties of standard 30 wt% glass fiber-reinforced polyamide 6 (PA6/GF30) and the polyketone–polyamide 6 hybrid composites with the same glass fiber content.

When benchmarked against the PA6/GF30 formulation, all hybrid composites containing PK exhibited a marked reduction in both tensile modulus and tensile strength at break. As a result, the incorporation of PK into the PA6 matrix introduced a softer character to the blend, weakening the hardness-oriented mechanical properties typical of unmodified PA6 composites.

Despite this reduction, the impact strength behavior revealed a different trend. For samples prepared with C1 as compatibilizer, no significant improvement in Izod impact strength was observed compared to the standard PA6/GF30 formulation. In contrast, the use of C2 as a compatibilizer resulted in substantial increases in impact resistance,

particularly in the PK-15/PA6-50/GF30-C2 and PK-25/PA6-40/GF30-C2 composites. Compared to PA6/GF30, these C2-modified blends demonstrated impact strength enhancements of approximately 36% and 113%, respectively.

This significant improvement is attributed to the maleic anhydride functional groups present in C2, which are believed to promote strong interfacial bonding between the PK/PA6 matrix and the glass fibers, as well as between the two polymer phases themselves. Moreover, the increased content of PK in these C2-based formulations appears to amplify the toughening effect, indicating a synergistic interaction between the flexible PK phase and the reactive compatibilizer.

In addition to impact performance, notable increases were also observed in tensile strain at break. The PK-15/PA6-50/GF30-C2 and PK-25/PA6-40/GF30-C2 composites exhibited increases of approximately 95% and 161%, respectively, when compared to the PA6/GF30 baseline. These results demonstrate that the presence of PK, combined with an effective compatibilizer, not only enhances impact resistance but also significantly improves ductility.

These strain improvements show that PK and C2 can significantly reduce the brittleness of PA6 composites, resulting in composites with a more balanced mechanical profile—combining moderate strength with high toughness and elongation capability. This tunability offers promising opportunities for applications requiring both structural integrity and energy absorption, such as in the automotive and electrical sectors.

4. CONCLUSION

This study provides a comparative evaluation of two compatibilizers on the mechanical behavior of glass fiber-reinforced PK and PK/PA6 composites. The results confirm that maleic anhydride-grafted polyethylene (C2) significantly enhances interfacial bonding and energy absorption, producing composites with up to 113% higher impact strength and 160% greater ductility than the unmodified system. These improvements stem from covalent and hydrogen bonding at the fiber-matrix and polymer-polymer interfaces.

Overall, the findings demonstrate that the selection of compatibilizer plays a pivotal role in tailoring the mechanical and impact performance of PK-based composites. Among the tested formulations, the C2-modified composite exhibited the most pronounced improvements in both strength and toughness, emphasizing the functional advantage of maleic anhydride in enhancing interfacial interactions. These results offer valuable insight into the design of high-performance, fiber-reinforced polyketone materials for demanding engineering applications.

In contrast to previous studies that primarily concentrated on unreinforced PK/PA6 blends, this research systematically evaluates the functionality of compatibilizers in fibre-reinforced hybrids, employing identical compositions and processing methodologies. This finding serves to elucidate the mechanistic role of compatibilizer chemistry in governing mechanical synergy.

Despite the absence of SEM analysis in the present study, earlier research [7], [9] has shown that maleic anhydride-based compatibilizers enhance interfacial bonding through covalent and hydrogen bonding mechanisms. The enhanced mechanical outcomes observed in this study are consistent with the morphological findings, thereby supporting the hypothesis of robust interaction.

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