



# Combined effect of interfacial strength and fiber orientation on mechanical performance of short Kevlar fiber reinforced olefin block copolymer



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## ABSTRACT

As a novel thermoplastic elastomer, olefin block copolymers (OBCs) show a tremendous application potential in many areas. However, the practical use of OBCs is mainly restricted to the low load-bearing environments because of its poor low-strain mechanical strength and stiffness. In this work, short Kevlar fiber was introduced into OBC matrix to improve its mechanical properties. The results show that introduction of pristine Kevlar fiber only gives rise to a slight increase in tensile strength of OBC owing to the poor interfacial strength, regardless of the presence of compatibilizer maleic anhydride-grafted polypropylene (MA-g-PP). However, a largely enhanced tensile strength can be easily obtained when hydrolyzed or polydopamine-coated Kevlar fiber was incorporated into the OBC matrix using the MA-g-PP as the reactive compatibilizer. More importantly, the tensile strength can be further enhanced with the orientation of Kevlar fiber in the composites. As compared with compression molded composites with randomly orientated fibers, injection molded composites with highly oriented fibers exhibit a significantly higher tensile strength at the same composition. Moreover, it is interesting to observe that Kevlar fiber can be easily exfoliated into several microfibrils during melt-mixing with OBC, but the exfoliation has no evident effect on the reinforcement effect possibly because of the poor interfacial strength between the newly-formed microfibrils and the OBC matrix. Our work demonstrates that mechanical performance of short fiber reinforced elastomer composites could be significantly improved by enhancing interfacial strength and fiber orientation using a facile surface modification strategy and conventional melt-processing technology.

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## 1. Introduction

In the past two decades, thermoplastic elastomers (TPEs) combining good melt processability of thermoplastics with inherent elasticity of conventional elastomeric materials (e.g., chemically cross-linked ethylene-propylene-diene monomer (EPDM) and nature rubber (NR)) have been widely used in various fields such as automotive interiors, household appliances, sports equipment, packaging and hygiene products. Olefin block copolymers (OBCs), consisting of crystallizable ethylene-octene blocks (hard segments) with very low 1-octene concentration and amorphous blocks (soft segments) with high 1-octene concentration, is a novel TPE synthesized by Dow Chemical Company via a chain-shutting polymerization technology using two distinct catalysts with different monomer selectivities and one chain-shutting agent [1–3]. The

chain architecture of OBCs including lengths and distributions of both segments can be readily regulated by the chain-shutting agent capable of switching the growing chains between the two catalysts at random intervals [1,3]. Because the tunable chain architecture can impart much better crystallizability and mechanical properties while maintaining good elasticity as compared to other polyolefin elastomers, such as corresponding random ethylene-octene copolymers, OBCs exhibit a tremendous application potential in many areas and have been becoming the focus of intense academic and industrial research recently [4–9]. However, similar to other TPEs, the poor mechanical properties such as low-strain strength and stiffness make OBCs unable to be used in many application environments where high load-bearing capacity is required to avoid deformation during their service life, such as automotive components.

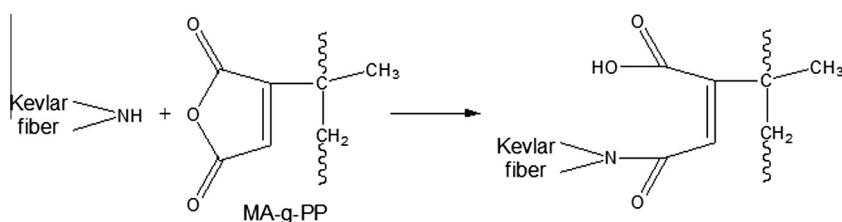
To meet the demands of various application environments, several strategies including polymer blending [10] and addition of nanoparticles [10–18] or fibers [13,19–24] into elastomer matrix

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have been developed to realize effective reinforcement. Among them, short fiber reinforced elastomer composites have attracted growing interest in recent years due to the superior low-strain reinforcement effect at low fiber loading and ease of preparation [19–22]. Para-aramid (Kevlar) fiber is one of the most promising candidates ever developed as a reinforcing agent for high-performance polymer composites because of its high specific strength, high modulus, and high thermal resistance when compared to metallic and polyester fibers [21,23]. Unfortunately, the use of Kevlar fiber to reinforce elastomers including OBCs faces many challenging obstacles mainly related to the interfacial incompatibility between Kevlar fiber and elastomer matrices because the lack of active functional groups on the smooth fiber surface to interact strongly with elastomer matrices via hydrogen bond or covalent bond. The ineffective interfaces make the stress applied to the elastomer/fiber composites unable to be efficiently transferred from the elastomer matrix to the fiber and thus the potential reinforcement effect of the high-performance fiber cannot be adequately exploited [25,26]. Various chemical treatments are frequently utilized to introduce or activate functional groups on the fiber surface for direct interacting with elastomer matrices or reacting with a reactive compatibilizer to form grafted polymer chains at the interface of the composites [27–32], finally greatly improving the interfacial strength. However, most of these treatments could dramatically destroy the fiber structure and involve toxic solvents, in addition to being time-consuming and costly.

In this work, to significantly improve the interfacial strength and subsequent low-strain mechanical strength, hydrolyzing and polydopamine-coating were selected as the two green, economical and feasible modification technologies to treat the short Kevlar fiber before being incorporated into OBC matrix and maleic anhydride-grafted polypropylene (MA-g-PP) was used as the reactive compatibilizer. Hydrolyzing treatment is a facile chemical treatment technique to activate amine group on Kevlar fiber surface without evidently changing its internal structure and bulk properties [30,32]. Polydopamine with superior adhesion properties is a newly-developed surface modification agent [33–37]. It has been reported that polydopamine can be facilely coated on various nanofillers via strong physical adhesion and provide abundant amine group to react with the reactive compatibilizer [33,34]. It was expected that the anhydride group of MA-g-PP can help the PP chains be anchored onto the fiber surface via readily reacting with the activated amine group of the fiber or abundant amine group of the polydopamine coating (as illustrated in Scheme 1) while the PP chains could entangle strongly with the matrix, finally forming effective stress transfer links at the interface. Moreover, the role of fiber orientation in improving the tensile strength was also highlighted by comparing the mechanical performance of compression molded composites with randomly orientated fibers and that of injection molded composites with highly oriented fibers. The results clearly demonstrate that a fairly large reinforcement effect can be obtained via enhancing interfacial strength and fiber orientation.



**Scheme 1.** Schematic illustration of the interfacial compatibilization reaction between anhydride group of MA-g-PP and the amine group of Kevlar fiber or polydopamine coating covered on the fiber surface.

**Table 1**

Nomenclature used for Kevlar fibers without and with various surface treatments.

Code	Surface treatment methods
KF	Untreated Kevlar fiber
H-KF	Hydrolyzed Kevlar fiber
D-KF	Polydopamine-coated Kevlar fiber

## 2. Experimental section

### 2.1. Materials

OBC (trade name D9530) was obtained from Dow Chemical Company, U.S.A. The molecular characteristics including density, octene content, weight-average molecular weight ( $M_w$ ), contents of the soft segment and the hard segment, melt flow index (MFI), and melting temperature ( $T_m$ ) measured by DSC are summarized in Table S1 (See in supporting information). Kevlar short-cut fibers with average length of 3 mm, diameter of 12  $\mu\text{m}$ , and aspect ratio of 250 were supplied by BlueStar Chengrand Research & Design Institute of Chemical Industry, China. Maleic anhydride-grafted polypropylene (MA-g-PP, trade name Epolene-E43), used as a reactive compatibilizer for OBC matrix and Kevlar fibers, was purchased from EASTMAN Chemicals Ltd., UK. It contains 3.8 wt.% maleic anhydride. Dopamine hydrochloride (99%) was provided by J&K scientific Ltd. Tris buffer solution and acetone were purchased from Chengdu Best Reagent Co., Ltd. and Tianjin Bodi Chemical Reagent Co., Ltd., respectively, and used as received.

### 2.2. Fiber surface treatment

Two different treatment methods including hydrolysis and polydopamine coating were used to modify the fiber surface. Hydrolysis treatment of Kevlar fibers with 10 wt.% sodium hydroxide aqueous solution was carried out at room temperature, according to the method reported by Chatzi et al. [38]. For the coating of polydopamine on the fiber surface, pre-dried Kevlar fibers were firstly soaked in dopamine aqueous solution at room temperature for 2 h to allow the absorption of dopamine molecules on the surface and then the dopamine-absorbed fibers were transferred into the Tris buffer solution with a pH value of around 8.5 for 24 h to encourage the polymerization of dopamine at the fiber surface [33]. Nomenclature for the Kevlar fibers treated with different methods is listed in Table 1.

In both the cases, Kevlar fibers were firstly immersed in boiling acetone for 4 h using Soxhlet extraction method before the surface treatments to remove the sizing agents and impurities attached on the fiber surface. After the treatments, all the obtained fibers were copiously washed with large amounts of distilled water for at least 3 times to ensure the complete remove of the remaining chemicals, followed by drying in a vacuum oven at 60  $^{\circ}\text{C}$  for 48 h.

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