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Performance Enhancement of COC-Modified LLDPE Films: Optimization of Stiffness, Tear Resistance, and Barrier through Plasma Surface Modification

Hetal Shah[1,*], Purvi Dave[2], Sudhir Kumar Nema[3]

Abstract

This study explores the development of high-performance packaging materials by blending commodity Linear Low-Density Polyethylene (LLDPE) with engineering cyclic olefin copolymer (COC) as a performance modifier. To leverage the processability of LLDPE alongside the structural rigidity of COC, blends were prepared at various weight ratios (95/05, 90/10, 85/15, and 80/20) using twin-screw melt extrusion, followed by the production of films and sheets via blown film extrusion and compression molding. The addition of COC significantly improved the mechanical properties of the LLDPE matrix; specifically, the tensile modulus increased three-fold with 10 wt% COC and five-fold with 20 wt% loading. While the inherent gas barrier properties of the films—measured through the Oxygen Transmission Rate (OTR) and Water Vapor Transmission Rate (WVTR)—improved with the addition of high-density COC domains, further optimization was achieved through surface modification. Thin organosilicon coatings were deposited onto 5 and 10 wt% COC blend films using plasma-enhanced chemical vapor deposition (PECVD) with hexamethyldisiloxane (HMDSO) and oxygen precursors. These PECVD coatings led to a substantial reduction in both the OTR and WVTR. As compared to uncoated $L_{90}C_{10}$ film, about 34% reduction in OTR and 43% reduction in WVTR is observed in SiOx coated $L_{90}C_{10}$ film. Scanning electron microscopy (SEM) confirmed the partial compatibility and interfacial adhesion between the COC phase and LLDPE matrix, providing a structural explanation for the enhanced mechanical and barrier properties. The findings indicate that LLDPE/COC blends, especially when enhanced by plasma surface treatments, represent a high-performance solution for the food, pharmaceutical, and industrial packaging applications.

Keywords: LLDPE, cyclic olefin copolymer (COC), polymer blend, mechanical, SiO_x coating

INTRODUCTION

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Polymer blending is a conventional approach for altering polymer characteristics. Polymer blends comprise approximately 30 percent of plastic products and are increasingly utilized in the development of innovative materials. There are several advantages of polymer blends because of which they are used in widespread applications. It also offers a cost-effective solution by improving the efficacy of more expensive polymers, ultimately leading to high performance at reasonable cost. Furthermore, polymer blending enables rapid property modification and reduces the time required to successfully develop a new material. The process is highly practical because it can be performed using existing manufacturing equipment [1, 2].

LLDPE is a popular choice for packaging owing to its remarkable lightweight nature, flexibility, strength, toughness, sealability, transparency, and superior processability. However, there are several limitations such as poor thermal properties and increased permeability [3]. Improvement in mechanical, thermal, and barrier properties of LLDPE are advantageous for numerous applications. Over 60% of LLDPE has been combined with other polyolefins, such as polypropylene (PP), polyethylene (PE), and copolymers, such as ethylene-vinyl acetate (EVA). The enhancement of several physical qualities and the processability of polyolefins through blending have been extensively established [4]. The characteristics of LLDPE blends often rely on interfacial adhesion, phase structure, and the compatibilizer employed owing to the limited compatibility of the polymers [5].

COC is a copolymer composed of ethylene and norbornene, which is synthesized using metallocene catalysts. COC is anticipated to be compatible with polyolefins because of its significant proportion of ethylene units, which negates the need for specialized compatibilizers [6, 7]. COC is an amorphous polymer characterized by transparency, low density, high rigidity, moisture resistance, elevated strength, and chemical resistance to alcohols, as well as significant resistance to acids and bases. They possess exceptional biocompatibility and electrical insulation properties. It possesses superior barrier qualities compared to LLDPE and can be utilized as a blend component to adjust gas transmission rates for certain applications [8, 9].

Incorporating COC into LLDPE is expected to improve the stiffness or modulus of the material. A higher modulus is advantageous because it allows for a reduction in film thickness while also providing better performance for packaging hot products and for materials that undergo thermal processes, such as metallization, printing, or sterilization.

In addition, COC is composed entirely of carbon and hydrogen. This structure allows for clean combustion, producing only carbon dioxide and water. These characteristics are beneficial for recycling and waste management [9, 10].

Research has consistently shown that the addition of COC to polyolefin blends significantly alters their mechanical and morphological characteristics. For instance, studies on polypropylene (PP)/COC blends [11] demonstrated that the incorporation of COC led to an increase in the tensile and flexural strengths and modulus. However, this enhancement in stiffness was accompanied by a decrease in both the impact strength and elongation at break. Similarly, investigations into COC/ethylene-vinyl acetate (EVA) blend films [12] reported a decrease in the modulus and an increase in the strain at break, along with the characteristic "matrix-droplet" morphology indicative of immiscibility. This immiscible "matrix-droplet" morphology was also observed in COC/ethylene-octene copolymer (EOC) blends [13], indicating phase separation. In the rheological, morphological, and heat seal properties of LLDPE/COC blends, partial phase compatibility was systematically confirmed, first suggested by dynamic rheological analysis (including Cole-Cole and equivalent plots) and then validated by subsequent morphological characterization via SEM [14].

The thermal and rheological properties of polymer blends with COC have also been extensively studied. The immiscibility of COC with other polymers is often confirmed through thermal analysis. For example, in blends of COC with polyolefin elastomer (POE), dynamic mechanical analysis (DMA) revealed two distinct peaks in the loss modulus versus temperature graph, corresponding to the glass transition temperatures (Tg) of the individual components [15]. Similarly, incorporating ethylene-vinyl acetate (EVA) into high-density polyethylene (HDPE) and low-density polyethylene (LDPE) resulted in a decrease in all transition temperatures, with the tan δ peak widening as the EVA concentration increased [16].

Rheological investigations, such as those conducted on PP/COC blends [17], found that the viscosity ratio of COC to PP increased with the shear rate but decreased with an increase in temperature. In

HDPE/COC mixtures, the resistance of the blend to creep was dependent on the COC content, with both pure HDPE and HDPE-rich blends exhibiting highly nonlinear creep behaviors [18].

Beyond the mechanical properties, the integration of COC is known to enhance the barrier performance of polymer blends. Even with signs of poor compatibility indicated by studies such as SEM, the addition of COC can significantly improve gas permeability. For example, cast films of a PE matrix blended with 5–20 wt% COC exhibited a noticeable reduction in both oxygen (O_2) and carbon dioxide (CO_2) permeability. This study further highlighted a noteworthy mechanical improvement, with a blend containing only 5 wt% COC showing a fivefold increase in elasticity compared to that of the pure PE matrix [19].

The thermal behavior of the blends was significantly influenced by the amorphous nature of COC. It was observed that the increase in the loading of amorphous COC increased the molecular disorder of the LLDPE chains, which hindered the crystallization kinetics and reduced the overall degree of crystallinity[20]. In our previous investigations employing melt rheology, Fourier-transform infrared (FTIR) spectroscopy, X-ray diffraction (XRD), and differential scanning calorimetry (DSC) on LLDPE/COC blends, we revealed that the melting and crystallization peak temperatures of the LLDPE phase remained largely unaffected by the addition of COC. This indicates that while the presence of COC domains restricts crystal formation by obstructing chain folding, it does not interfere with the thermodynamic stability or lamellar thickness of the LLDPE crystals that form. Furthermore, rheological analysis showed that the Melt Flow Index (MFI) values of the LLDPE/COC blends remained within the optimal range (0.9-2.0 g/10 min) required for stable processing [21, 22]. These favorable flow characteristics directly facilitated the subsequent blown film extrusion process, ensuring stable bubble formation and the successful production of the film samples. To assess the suitability of such polymer blends for packaging applications, it is essential to evaluate their dynamic mechanical properties, mechanical performance (e.g., tensile strength, modulus, and tear strength), and barrier properties. Furthermore, there is a notable gap in the existing literature regarding the enhancement of barrier properties in LLDPE/COC blended films via the application of a silicon oxide (SiO_x) coating using plasma-enhanced chemical vapor deposition (PECVD), an area of significant scientific and industrial interest.

This study explores the creation and characterization of LLDPE and COC blends. The blends were prepared using a Haake twin-screw extruder with COC concentrations of 5, 10, 15 wt%, and 20 wt%. Samples for analysis were subsequently produced via film extrusion and compression molding. A comprehensive analysis of the LLDPE/COC blends was conducted, focusing on their dynamic mechanical, tensile (strength, modulus, and percent elongation), and tear properties, in addition to their gas barrier and morphological characteristics. Furthermore, this study examines the effect of thin SiO_x films deposited by PECVD on the barrier performance of blended materials. This study aims to create improved packaging materials by combining the excellent mechanical, thermal, and barrier properties of COC with the cost-effectiveness and processability of LLDPE.

EXPERIMENTAL

Materials

LLDPE, F19010 (density = 0.918 g/cc, MFI at 190°C and 2.16 kg = 0.90 g/10 min), was procured from Reliance Industries Limited, whereas COC, TOPAS 8007 (density = 1.02 g/cc, MFI at 190°C and 2.16 kg = 1.8 g/10 min), was provided by TOPAS Advanced Polymers. The norbornene concentration in the chosen COC grade is approximately 35%, and the glass transition temperature (Tg) of COC is 78°C.

Preparation of Blends

Blends of LLDPE and COC were created through a melt-blending process utilizing a co-rotating intermeshing twin-screw extruder (Haake Rheomix TW100). The compositions of the investigated samples are denoted as LXCY, where X and Y indicate the weight percentages of LLDPE and COC in the blend, respectively. The parameters for processing included a screw speed of 50 rpm, cylinder

temperature profile of 210, 220, and 230 °C, and die exit temperature of 230 °C. The extrudate was quenched in a cold-water bath and subsequently granulated following the melt blending process, as shown in Figure 1.

SAMPLE PREPARATION

Compression Molding

Granules of LLDPE, COC, and LLDPE/COC blends underwent pre-drying before being subjected to compression molding to produce sheets (Figure 2) at a temperature of 250ºC and a pressure of 200 psi for 10 minutes. The resulting sheets exhibited a thickness between 3±0.2 mm. The prepared samples were then used to analyze dynamic mechanical properties.

Blown Film

Blends of LLDPE and COC were produced utilizing a Haake tubular blown film extruder as shown in Figure 3(a). The measurement of film thickness was conducted utilizing a digital thickness gauge meter, revealing an approximate thickness of 40±3 μm. The processing temperature across various zones was maintained at 210–240ºC. Blown film samples, as shown in Figure 3(b), were prepared and utilized to assess mechanical and barrier properties and conduct a morphological study.

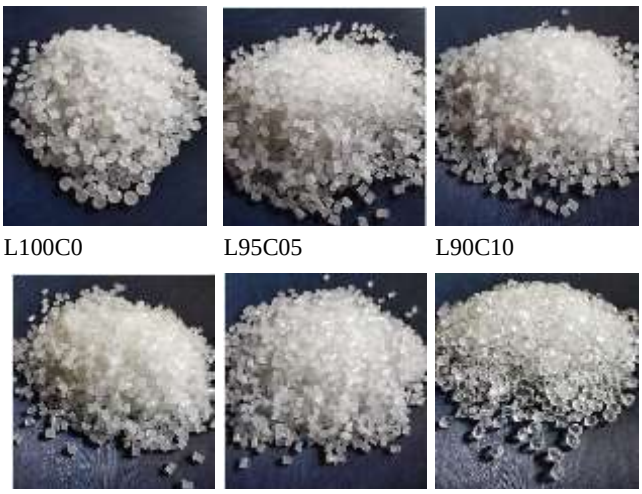


Figure 1. Granules of $L_{100}C_{0,}$ $L_{95}C_{05,}$ $L_{90}C_{10,}$ $L_{85}C_{15,}$ $L_{80}C_{20}$ and L_0C_{100}.

Figure 2. Compression-molded samples of $L_{100}C_{0,}$ $L_0C_{100,}$ $L_{95}C_{05,}$ $L_{90}C_{10,}$ $L_{85}C_{15,}$ and $L_{80}C_{20}$..

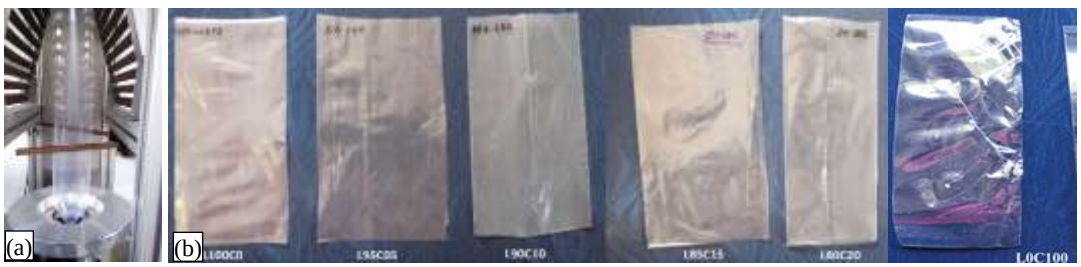
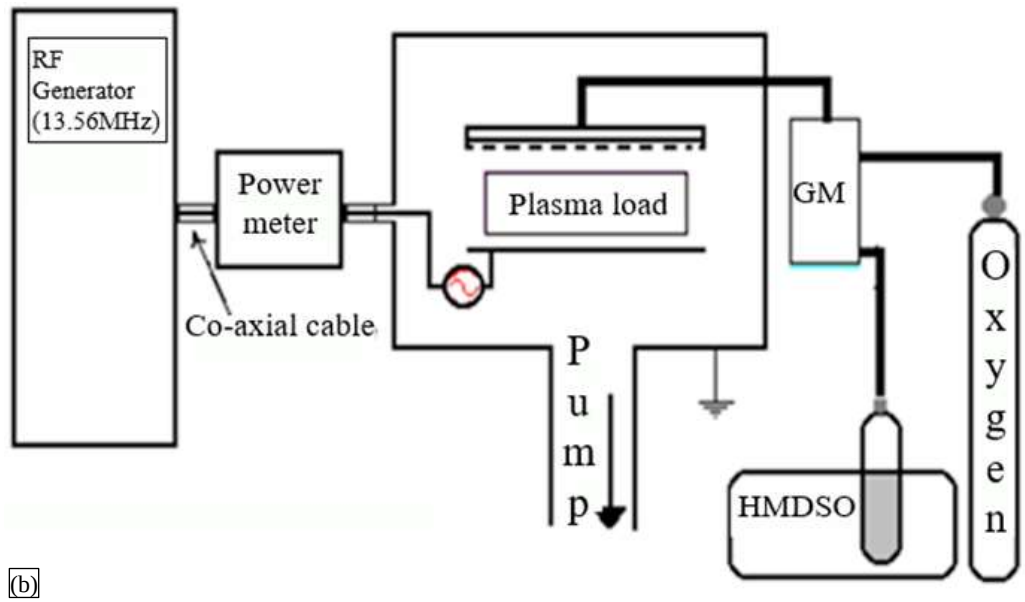


Figure 3. (a) Blown Film Preparation (b) Blown film samples of $L_{100}C_{0,}$ $L_{95}C_{05,}$ $L_{90}C_{10,}$ $L_{85}C_{15,}$ $L_{80}C_{20}$ and L_0C_{100}.

SiO_x Coating

This investigation involved the deposition of thin SiO_x films through the PECVD method, as shown in Figure 4(a). Hexamethyldisiloxane (HMDSO) and oxygen (O_2) were utilized as plasma-forming gases, introduced into the process zone through a multipoint gas-feeding showerhead. Plasma was produced at a power level of 50 watts between two parallel plate electrodes with a diameter of 35 cm, utilizing a radio frequency source operating at 13.56 MHz with an O_2/HMDSO mixture, as illustrated in Figure 4(b). The process for SiO_x involved a cleaning duration of 10 minutes, followed by a coating period of 20 minutes, conducted at a pressure of 0.07 Mbar in an O_2/HMDSO gas mixture, and concluded with a cooling phase lasting 30 minutes. The thickness of the deposited films was assessed utilizing a NanoMap-500ES contact mode stylus profilometer. In our experiments, we focused on inhibiting the ingress of oxygen and water molecules from the film surface through the deposition of thin SiO_x films, ranging from 100 to 500 nm, using the PECVD method. SiO_x coating was applied to $L_{95}C_{05}$ and $L_{90}C_{10}$ films, which were subsequently assessed for OTR and WVTR.

(a)



(b)

*GM= Gas mixing chamber

Figure 4. (a) PECVD Equipment and (b) Process of plasma-enhanced chemical vapor deposition (PECVD) using hexamethyl disiloxane.

CHARACTERIZATION

Dynamic Mechanical Properties

DMA is widely utilized to characterize the viscoelastic properties, transition, and relaxation behavior of polymers. The DMA was performed using the Inkarp Instruments EXSTAR DMS 6100 dynamic mechanical analyzer, employing a three-point bending method at a constant frequency of 1 Hz. The test was conducted at a heating rate of 2°C/min, within a temperature range of -130 to +130°C. The dimensions of the test specimen were approximately 54 mm × 13 mm × 3 mm. The samples underwent scanning to assess their viscoelastic responses, which included the measurement of storage moduli (E'), loss moduli (E"), and loss factor (Tan δ).

Mechanical Properties

Tensile strength, percentage elongation, and tensile modulus in the machine direction (MD) and transverse direction (TD) were measured using a universal testing machine (Deepak Polyplast) in accordance with ASTM D882 at 23°C and 50% relative humidity for the prepared film samples. The tensile testing apparatus operates at a crosshead speed of 500 mm/min. The dimensions of the test specimen were approximately 100 × 25 mm. Trouser tear strength in the MD and TD was determined using a universal testing machine (Deepak Polyplast). The prepared film samples were tested in accordance with ASTM D1938 at 23°C and 50% relative humidity. The dimensions of the test specimen measured approximately 90 × 25 mm. A standard tensile tester operates at a crosshead speed of 250 mm/min. All reported values represent the averages derived from five specimens for each sample.

Barrier Properties

The evaluation of the oxygen transmission rate (OTR) was conducted using a gas permeation analyzer (Model: N500) in accordance with ASTM D1434 at a temperature of 23°C and relative humidity of 50%. The water vapor transmission rate (WVTR) was assessed using Extra solution (Permeh$_2$O) following ASTM F1249 at a temperature of 38°C and relative humidity of 90%. The OTR and WVTR values for both uncoated and SiO_x -coated film samples were measured. All reported values are derived as averages from three specimens for each sample.

Morphological Properties

The morphology of the film samples was analyzed using SEM with a JOEL FESEM (JSM-7600F). Prior to the SEM analysis, the surfaces of the samples were coated with gold using a sputtering coating unit, model JOEL JFC-1600.

RESULTS AND DISCUSSION

Dynamic Mechanical Properties

Figure 5 illustrates the storage modulus (E') for LLDPE, COC, and their blend samples as a function of temperature. COC has a rigid structure that gives it a higher modulus at high temperature; the high crystallinity of LLDPE results in a higher modulus at lower temperature. Blends with higher COC content exhibited a higher E' value at temperatures above 0°C than $L_{100}C_0$. A particularly notable improvement was observed at approximately 25°C, where the E′ of the $L_{90}C_{10}$ blend reached 2179 MPa, a substantial increase over the 794 MPa measured for pure LLDPE. For $L_{90}C_{10,}$ $L_{85}C_{15}$ and $L_{80}C_{20}$ samples, they have almost the same E' at temperatures above 0°C, higher than $L_{100}C_0$ but lower than L_0C_{100}. Even at 90°C, E' of $L_{90}C_{10}$ is 362 MPa as compared to 20 MPa of L_0C_{100}. Below -100°C (except for $L_{95}C_{05}$) and above 80°C, E' of all blends is higher than that of L_0C_{100}. One of the possible reasons for $L_{95}C_{05}$ exhibiting a lower E' compared to $L_{100}C_0$ at temperatures below 0°C is that this phenomenon represents a negative deviation from the linear rule of mixtures, a behaviour often observed in immiscible or partially compatible polymer blends at low secondary-phase concentrations. Instead of reinforcing the matrix, they act as stress concentrators that allow the surrounding LLDPE to deform more easily under stress at low temperatures. Once the COC concentration reaches 10%, the reinforcing effect is finally achieved over the disruption, and the material becomes significantly stiffer.

Several factors, including improved crystallinity, may lead to an increase in the storage modulus. Our prior research [22] using DSC on LLDPE/COC blends indicates that the addition of COC does not significantly alter the percentage crystallinity of these blends.

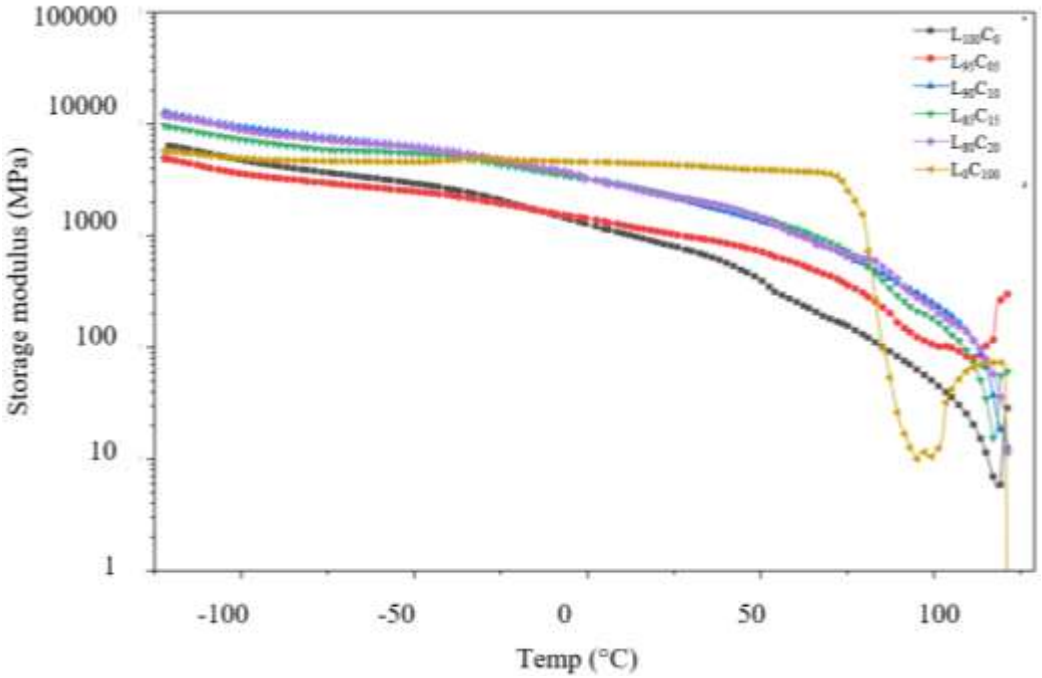


Figure 5. Storage modulus (*E′*) of $L_{100}C_0$, $L_{95}C_{05}$, $L_{90}C_{10}$, $L_{85}C_{15}$, $L_{80}C_{20}$ and L_0C_{100}.

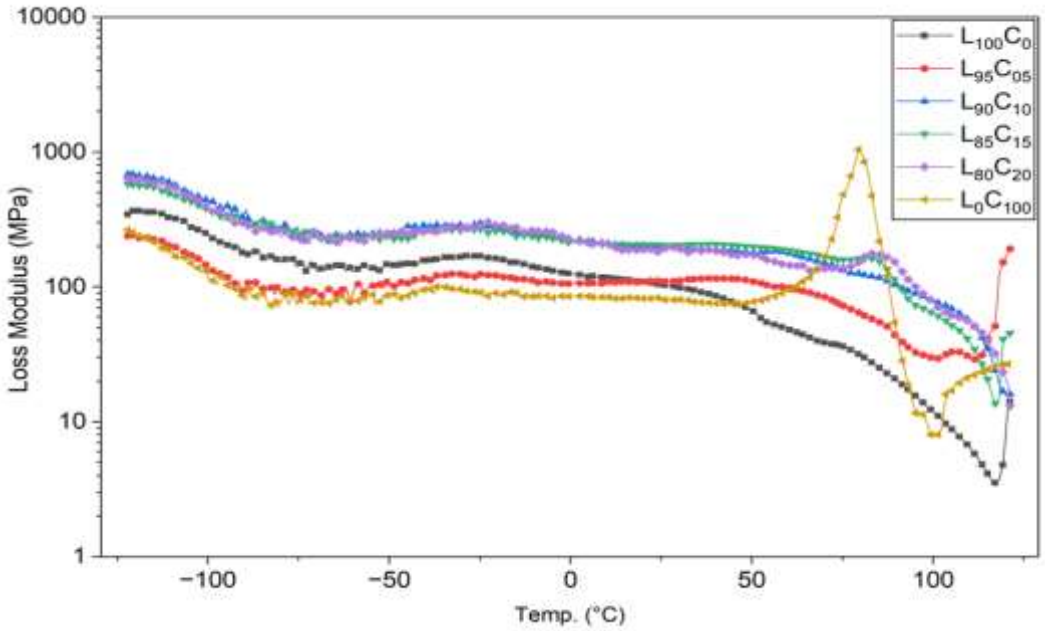


Figure 6. Loss modulus (E*″*) of $L_{100}C_0$, $L_{95}C_{05}$, $L_{90}C_{10}$, $L_{85}C_{15}$, $L_{80}C_{20}$ and L_0C_{100}.

The loss modulus peak observed at approximately -120, -25, and 40°C is indicative of the γ-transition, β-transition, and α-transition for pure LLDPE, respectively. A peak at about −120°C is thought to be caused by the movement of molecules in the amorphous areas. The peak at −25°C is the glass transition temperature (Tg) of the LLDPE. A peak around 40°C is linked to molecular movements in the crystalline and interfacial areas. The glass transition of L_0C_{100} is observed by a peak at 87°C, also visible in the blends, and corresponds to a significant drop observed in the storage modulus. The data shows that the blends have two separate glass transitions, which is evidence of immiscibility and the presence of two distinct phases.

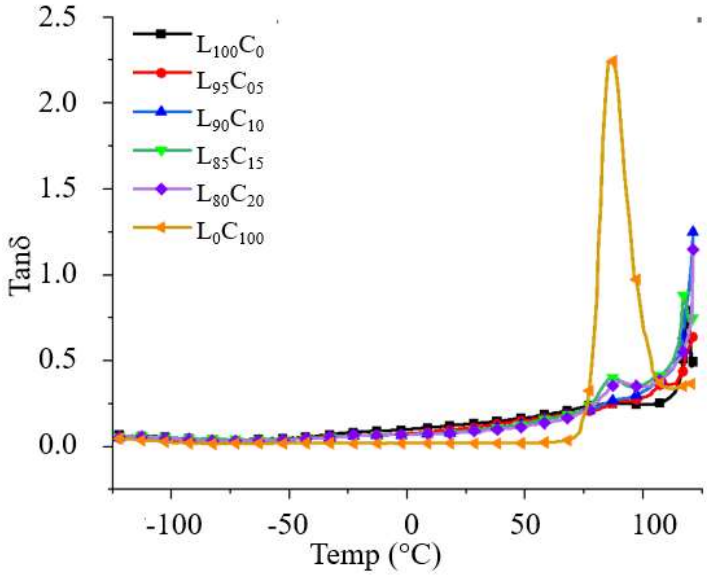


Figure 7. Tan δ of $L_{100}C_0$, $L_{95}C_{05}$, $L_{90}C_{10}$, $L_{85}C_{15}$, $L_{80}C_{20}$ and L_0C_{100}.

As the amount of COC increases, the E″ curve undergoes a systematic change. Blends with $L_{90}C_{10}$ or more show higher loss modulus, which states that the elastic recovery is low. This effect is because the COC makes the material more rigid. This trend aligns with similar observations reported for other blends, like COC/POE systems [15].

Figure 7 illustrates the tan δ versus temperature diagram; it indicates the relative energy dissipation or damping of the material. The tan δ (~0.25) for $L_{100}C_0$ shows a relatively low and wide peak, because $L_{100}C_0$ has a semicrystalline structure and limited mobility of its amorphous chains, which means that it didn't lose much energy. It's usual for semicrystalline polyolefins not to have a visible Tg peak. L_0C_{100} shows a clear Tg at 87.93°C with a crisp and prominent tan δ peak (~2.34), because of its stiff amorphous structure [23, 24].

The intensity of the tan δ peak for COC varies with the weight fractions within the blends. At $L_{95}C_{05}$, the tan δ went up to 0.31, and a Tg showed up at about 80.19°C. Adding more COC raised the damping factor and increased the Tg values to higher temperatures. It shows that even little amounts of COC control the relaxation dynamics, which makes it harder for the chains to move around in the blend.

The increase in E′ and tan δ shows that COC not only makes the LLDPE matrix stronger, but it also modifies how the segments relax by reducing chain movement and moving the thermal response to higher temperatures. In short, DMA results demonstrate that adding COC to LLDPE generates stiff blends. The synergistic improvement in modulus, damping properties, and relaxation transitions show that LLDPE/COC systems could be useful for applications that need better thermal and mechanical performance. The blends are partially compatible due to the fine dispersion, which aligns with the microscopic data. The presence of separate Tg peaks confirms immiscibility, while the overlapping nature of the peaks suggests some degree of interaction or fine dispersion.

Mechanical Properties

The mechanical properties of film are influenced by various processing variables, which affect molecular orientation and crystallinity within the film to varying degrees. The orientation in the film results in directional tear and tensile properties, with values specified for both the MD and the TD. Figures 8, 9, and 10 illustrate the tensile strength, percentage elongation, and tensile modulus for LLDPE and its blends with COC, respectively. The tensile strength of the LLDPE films is greater in the MD than in the TD, which can be attributed to the presence of the long fibril structure [23, 24]. The

data indicates that as the ratio of COC in the blend increases, there is a corresponding increase in tensile strength for both MD and TD. The tensile strength for $L_{90}C_{10}$ exhibits an increase of 60% in the MD and 58% in the TD when compared to $L_{100}C_0$. Similarly, we observe an increase in tensile strength across other blend compositions.

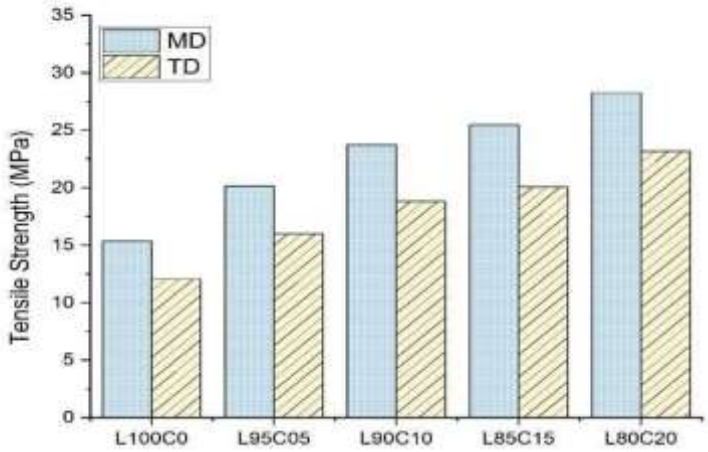


Figure 8. Tensile strength of LLDPE/COC blends.

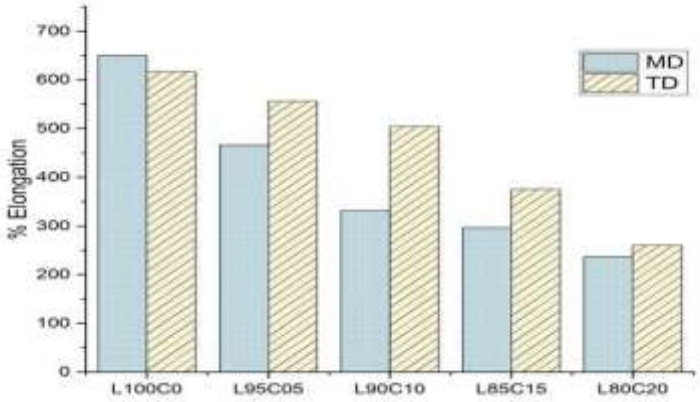


Figure 9. Percent Elongation of LLDPE/COC blends.

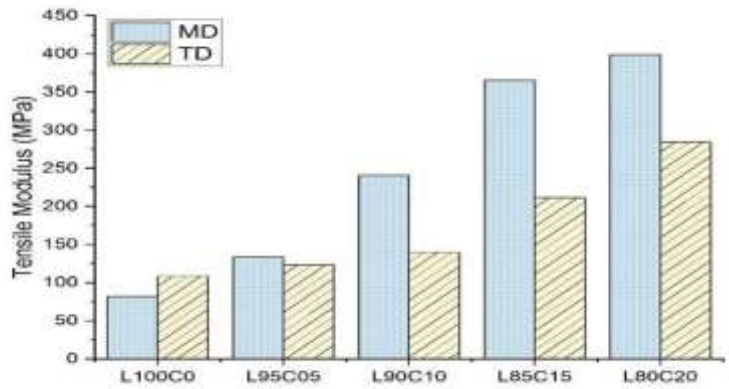


Figure 10. Tensile modulus of LLDPE/COC blends.

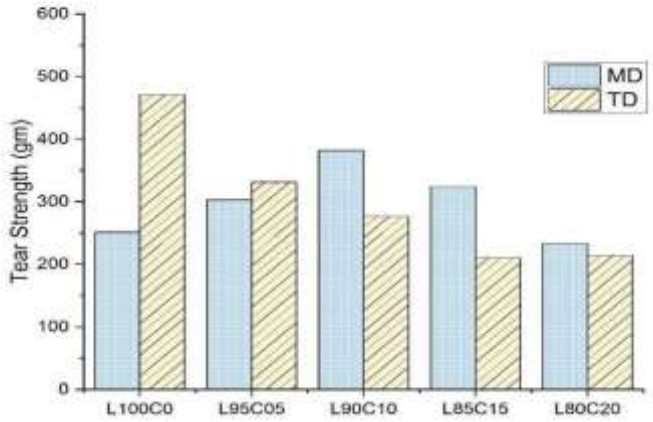


Figure 11. Tear strength of LLDPE/COC blends.

The tensile modulus of the films exhibits a notable increase as the COC content rises. The tensile modulus for $L_{90}C_{10}$ exhibits an increase of 194% in the MD and 30% in the TD compared to $L_{100}C_0$. The increase in tensile modulus aligns with the findings for storage modulus, which indicates a 174% increase in storage modulus (at 25°C) following the addition of COC for $L_{90}C_{10}$ in comparison to $L_{100}C_0$. The introduction of a stiffer component correlates with an increase in the modulus of the blends, consistent with the existing literature [26, 27]. This gauge reduction can be achieved without compromising properties. The storage modulus and tensile modulus exhibited an increase in relation to the COC weight fraction. Gopanna et al. observed a similar trend in their research on PP/COC blends [11].

The incorporation of stiffened COC contributed to the decrease in percent elongation observed in the LLDPE/COC blend film. The elongation in the MD exhibits a significant decrease when contrasted with the elongation in the TD. The $L_{90}C_{10}$ blend film exhibits a reduction of approximately 49% in percent elongation in the MD and 18% in the TD. The elongation of the LLDPE/COC blends is situated within the range established by pure LLDPE and COC, with the value varying according to the COC fraction present in the blend [19]. The inclusion of a small quantity of COC enhances the mechanical properties of the pure polymer, as demonstrated by the results.

The data obtained from tensile properties were compared with findings from similar studies to validate the observed trends. It was found that the addition of COC leads to an increase in tensile strength and modulus, which is consistent with the results reported by other researchers [5, 28]. This enhancement is attributed to the high stiffness of the cyclic olefin domains, which reinforce the softer LLDPE matrix. Conversely, a progressive decrease in elongation at break was observed as the COC content increased. This trend is a well-documented consequence of incorporating a brittle, glassy phase into a ductile polyolefin matrix, as the dispersed COC domains act as points of restricted molecular mobility, thereby reducing the overall ductility of the blend.

Figure 11 presents the tear strength of LLDPE and the LLDPE/COC blend film in both MD and TD. $L_{100}C_0$ exhibits the anisotropy for blown films, where the polymer chains undergo molecular orientation primarily along the MD during processing. Tear propagation along the TD requires higher energy to cut across these highly aligned molecular chains. Hence, the TD tear strength is significantly higher than the MD tear strength. Addition of COC disrupts this anisotropy of LLDPE. As the COC content increases, these rigid domains act as stress concentration points and interfere with the molecular alignment of the continuous LLDPE matrix, causing the MD tear strength to initially increase (as seen in $L_{90}C_{10}$) while the TD tear strength generally decreases. This relationship between film orientation and anisotropic tear strength in LLDPE is consistent with established literature concerning processing

effects on polyolefin film properties [29]. The higher tear strength of $L_{100}C_0$ in the TD compared to the MD presents challenges for its application in directional tear scenarios oriented in the transverse direction. The incorporation of COC into LLDPE results in a reduction of TD tear, which may enhance the ease of tear performance [25].

The results from the tear strength analysis indicate that the $L_{90}C_{10}$ film exhibits the highest increase in MD tear strength, showing a 52% improvement compared to the $L_{100}C_0$ and all other blends. The selection of plastic film is determined by the specific properties required for the intended end-use application.

Morphological study

Figures 12 (a) to (f) present the SEM micrographs of the LLDPE/COC blend film and SiO_x -coated samples. The micrographs usually show that the LLDPE makes up the continuous matrix phase, and the rigid COC is spread out throughout this matrix as the discontinuous phase in the form of small, spherical domains [15, 19]. This fine dispersion suggests excellent kinetic mixing during the melt-blending process and is consistent with the DMA results that indicated a degree of partial compatibility and strong interfacial interaction. The SEM images show that the blends are immiscible, but some level of partial compatibility is observed due to this fine dispersion.

The fine, rigid COC domains act as reinforcing sites within the softer LLDPE matrix. This morphology directly explains the observed significant increase in the E′ and tensile modulus of the blends. The integrity of the dispersed phase signifies strong interfacial adhesion, facilitating effective stress transfer between the phases.

The SEM micrographs of the SiO_x-coated films as shown in figures 12(e) and 12(f) are essential for evaluating the effectiveness of the barrier enhancement procedure. The micrographs were inspected for topographic defects such as pinholes, microcracks, or large particulate inclusions, which can impact the barrier properties of the film.

The presence of a smooth, featureless surface confirms that the SiO_x-coated LLDPE/COC blend film provides a suitable, low-roughness substrate for the uniform nucleation and growth of the inorganic barrier layer, thereby optimizing the diffusion path length for gas molecules. The absence of significant flaws in the SiO_x layer is essential for maximizing the barrier efficiency of the final multilayer structure.

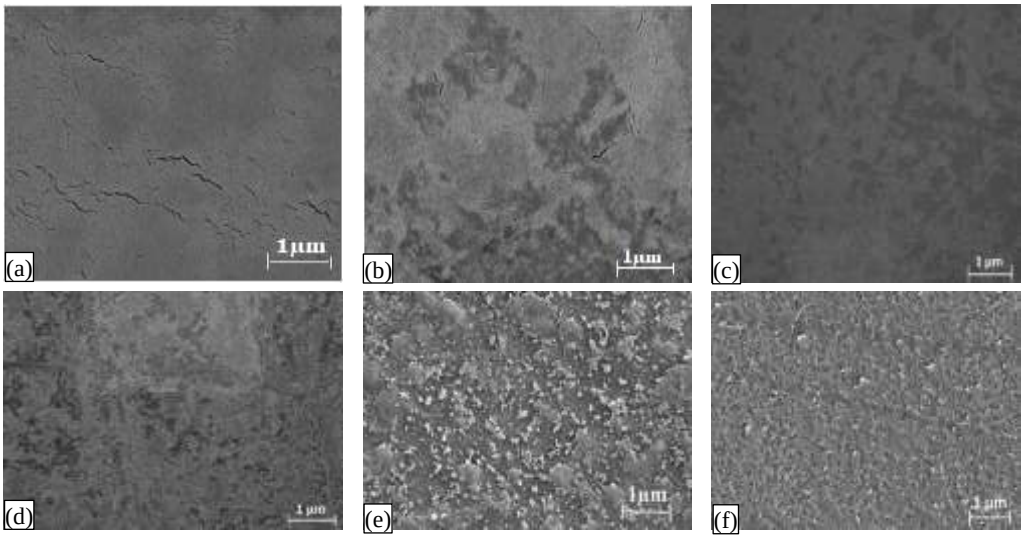


Figure 12. SEM micrographs of (a) $L_{95}C_{05}$ (b) $L_{90}C_{10}$ (c) $L_{85}C_{15}$ (d) $L_{80}C_{20}$ films at 1 µm; SEM micrographs of SiO_x coated films of (e) $L_{95}C_{05}$ (f) $L_{90}C_{10}$ at 1 µm.

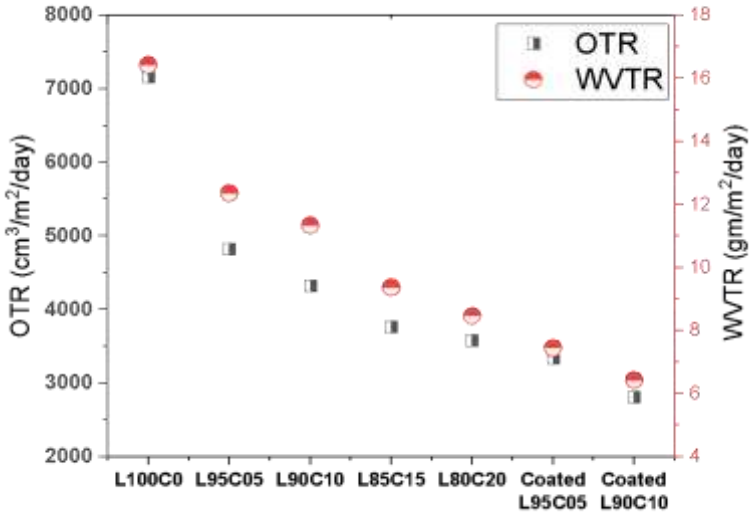


Figure 13. Oxygen and water vapour transmission rate for LLDPE/COC blend film samples.

Barrier Properties

The barrier to the permeation of gas and water vapor is a critical consideration from a packaging perspective. The transmission rates of oxygen and water vapour for LLDPE/COC blend film samples were measured, and the results are presented in Figure 13. The results indicate a reduction in OTR and WVTR for all blend compositions following the addition of COC. The OTR decreases to approximately 33% for $L_{95}C_{05}$ film and 41% for $L_{90}C_{10}$ film. In a similar manner, the WVTR decreases to approximately 25% for the $L_{95}C_{05}$ film and 31% for the $L_{90}C_{10}$ film. The results obtained are comparable to the data reported by the other researcher [19,30].

According to Ou et al. [31], the decrease in transmission rate of various gas molecules is attributed to the incorporation of high-density COC, which compels the gas molecules to traverse a longer pathway. COC is not a perfect barrier to moisture and oxygen. However, the incorporation of melt-blended COC results in some improvement in barrier properties by altering the diffusion path of gas molecules within the bulk material. Chain stiffness hinders molecular movement, thereby decreasing the formation of temporary voids that allow permeant molecules to enter the system [25]. The results obtained are significant, as they indicate the potential for improved barrier properties in monolayer polyolefin films while also enhancing the feasibility of recycling.

The improved barrier performance is strongly linked to the morphology observed via SEM. Despite COC's dispersed phase reduces free volume and enhances tortuosity. The rigid COC domains act as physical barriers, forcing gas molecules to navigate a longer, less direct path through the continuous LLDPE matrix. COC itself has a high Tg (~87°C) and a rigid structure, giving it an inherently lower free volume and higher density compared to LLDPE, thus making it a superior intrinsic barrier material. The dispersion of these high-barrier domains effectively reduces the overall permeability of the blend film.

Methods for achieving high barrier properties of monolayer polymers include deposition of thin metal coatings and deposition of oxide films such as SiO_x. This work adopts the PECVD process to deposit SiO_x coating, which improves barrier properties. To deposit the SiO_x coating, we have primarily focused on $L_{95}C_{05}$ and $L_{90}C_{10}$ blend films, aiming to keep the COC content smaller. After SiO_x coating on $L_{95}C_{05}$ film, there was about a 31% and 39% decrease in OTR and WVTR, respectively, as compared to uncoated $L_{95}C_{05}$ film. Whereas for the SiO_x coated $L_{90}C_{10}$ film, it shows about 34% and 43% reduction

in OTR and WVTR, respectively, as compared to uncoated $L_{90}C_{10}$ film. There is a remarkable reduction in OTR and WVTR for SiO_x coated $L_{95}C_{05}$ and $L_{90}C_{10}$ film as shown in Figure 13.

Figure 12 (e and f) illustrates that the SiO_x coating applied to the film surface has effectively filled the cracks, resulting in a significant enhancement [32]. This improvement in OTR and WVTR suggests the possibility of enhanced barrier properties in a monolayer polyolefinic film. Such improvements can be achieved by adjusting the COC content within the range of 5–10 wt%, utilizing existing processing equipment and conditions. This approach moderately influences product costs and supports end-use recycling efforts.

CONCLUSIONS

This study successfully demonstrated that melt-blending LLDPE with COC (5–20 wt%) is a robust strategy for engineering high-performance packaging films. By combining the cost-effectiveness of commodity polyolefins with the structural rigidity of engineering copolymers, this research established a clear pathway for tailoring film properties through both internal blending and surface modification.

DMA showed a substantial increase in E′ across a wide temperature range. At room temperature (25°C), the E′ of the $L_{90}C_{10}$ blend reached 2179 MPa, nearly tripling the 794 MPa of $L_{100}C_0$. The blends retained significant structural integrity at elevated temperatures. At 90°C, the $L_{90}C_{10}$ blend maintained a storage modulus of 362 MPa, providing a critical advantage for hot-fill packaging where pure LLDPE would typically lose its dimensional stability.

Mechanical properties revealed that the incorporation of COC significantly alters the anisotropic nature of LLDPE blown films, leading to a substantial increase in structural rigidity and more balanced tear properties. The $L_{90}C_{10}$ blend emerged as an optimal composition, achieving a 60% increase in MD tensile strength and a remarkable 194% improvement in tensile modulus, a trend that closely aligns with the result observed in storage modulus. COC played a role in modifying the film's directional tear resistance; specifically, the $L_{90}C_{10}$ sample showed a 52% enhancement in MD tear strength compared to $L_{100}C_0$. This modification disrupts the extreme molecular orientation of the LLDPE matrix, reducing the high TD tear resistance and facilitating a more controlled, 'easy-tear' performance for packaging applications.

Additionally, the investigation verified that the diffusion barrier capabilities against oxygen and water vapor are significantly improved by the addition of COC. One of the main reasons for the observed decrease in gas and vapor transmission rates is the good dispersion and remarkable degree of adherence of the COC domains inside the LLDPE matrix, as revealed by the SEM investigation. Thin organosilicon films were applied to the films using PECVD in order to further enhance these characteristics. The OTR and WVTR were both shown to be greatly decreased by this coating, indicating a practical method for building high-performance multilayer structures.

The practical implications of these findings are substantial for the packaging industry. Such films are well-suited for demanding applications requiring improved stiffness, moisture/gas protection as well as thermal performance, such as food, pharmaceutical, and industrial packaging, where extended shelf life and product integrity are critical. Future research should focus on optimizing the PECVD coating parameters to achieve even greater barrier performance and investigate the long-term stability and recyclability of these modified multilayer films. Further investigation into the specific interfacial interactions between the COC and LLDPE phases at a molecular level may yield enhanced understanding of the observed property improvements.

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Conflict Of Interest

The authors declare that they have no conflict of interest.

Abbreviations

COC	:	Cyclic Olefin Copolymer
DSC	:	Differential scanning calorimetry
DMA	:	Dynamic mechanical analysis
EOC	:	Ethylene-octene copolymer
EVA	:	Ethylene-vinyl acetate
FTIR	:	Fourier-transform infrared spectroscopy
Tg	:	Glass transition temperatures
HMDSO	:	Hexamethyldisiloxane
HDPE	:	High-density polyethylene
LLDPE	:	Linear Low-Density Polyethylene
Tan δ	:	Loss factor
E"	:	Loss moduli
LDPE	:	Low-density polyethylene
MD	:	Machine direction
MFI	:	Melt Flow Index
OTR	:	Oxygen Transmission Rate
PECVD	:	Plasma-enhanced chemical vapor deposition
PE	:	Polyethylene
POE	:	Polyolefin elastomer
PP	:	Polypropylene
SEM	:	Scanning Electron Microscopy
SiO_x	:	Silicon oxide
E'	:	Storage moduli
TD	:	Transverse direction
WVTR	:	Water Vapor Transmission Rate
XRD	:	X-ray diffraction

REFERENCES

1. Utracki LA. Polymer blends handbook. Kluwer Academic Publishers; 2002. 1442 p.
2. Shonaike GO, Simon GP. Polymer Blends and Alloys. Marcel Dakker Inc.; 1999.
3. Nwabunma D, Kyu T. POLYOLEFIN BLENDS. John Wiley & Sons, Inc., Hoboken, New Jersey; 2008.
4. Hay JN, Zhou XQ. The effect of mixing on the properties of polyethylene blends. Polymer (Guildf). 1993;34(11):2282–8. doi:10.1016/0032-3861(93)90809-O
5. Dorigato A, Pegoretti A, Fambri L, Lonardi C, Šlouf M, Kolařik J. Linear low density polyethylene/cycloolefin copolymer blends. Express Polym Lett. 2011;5(1):23–37. doi:10.3144/expresspolymlett.2011.4
6. Kolařík J, Pegoretti A, Fambri L, Penati A. Non-linear long-term tensile creep of poly(propylene)/cycloolefin copolymer blends with fibrous structure. Macromol Mater Eng. 2003;288(8):629–41. doi:10.1002/mame.200300005
7. Kolar J. Phase Morphology of PP / COC Blends. J Appl Polym Sci. 2004;91:253–259.
8. Kaufman, J J Falcetta HS. Introduction to polymer science and technology: An SPE text-book. Wiley-Interscience, New York; 1978.
9. JESTER R. Cyclic Olefin Copolymer Enhances Polyolefin Blends for Film Packaging. Plastics Technology. 2011 Apr.
10. Starck P. Dynamic mechanical thermal analysis on Ziegler-Natta and metallocene type ethylene copolymers. Eur Polym J. 1997;33:339--348.

11. Gopanna A, Thomas SP, Rajan KP, Rajan R, Rainosalo E, Zavašnik J, et al. Investigation of mechanical, dynamic mechanical, rheological and morphological properties of blends based on polypropylene (PP) and cyclic olefin copolymer (COC). Eur Polym J. 2018;108:439–51. doi:10.1016/j.eurpolymj.2018.09.030
12. Durmus A, Alanalp MB, Aydin S. Investigation of morphological, rheological, and mechanical properties of cyclic olefin copolymer/poly(ethylene-co-vinyl acetate) blend films. Journal of Plastic Film & Sheeting. 2018;34(2):140–59.
13. Doshev, D. Tomova, A. Wutzler, P. Morphology and Mechanical Properties of Reactive and Non-Reactive COC/EOC Blends. Journal of Polymer Engineering. 2011;25(5):375–392.
14. Lamnawar Khalid and Vion-Loisel F and Maazouz Abderrahim. Rheological, Morphological, and Heat Seal Properties of Linear Low Density Polyethylene and Cyclo Olefine Copolymer (LLDPE/COC) Blends. J Appl Polym Sci. 2010;116:2015–22. doi:10.1002/app.31804
15. Khonakdar HA, Jafari SH, Hesabi MN. Miscibility analysis, viscoelastic properties and morphology of cyclic olefin copolymer/polyolefin elastomer (COC/POE) blends. Compos B Eng. 2015;69:111–9. doi:10.1016/j.compositesb.2014.09.034
16. Khonakdar HA, Wagenknecht U, Jafari SH, Hässler R, Eslami H. Dynamic mechanical properties and morphology of polyethylene/ethylene vinyl acetate copolymer blends. Advances in Polymer Technology. 2004;23(4):307–15. doi:10.1002/adv.20019
17. Fambri L, Kolarik J, Pasqualini E, Penati A, Pegoretti A. Rheological study on polypropylene/cydoolefin copolymer blends. Macromol Symp. 2008;263(1):114–20. doi:10.1002/masy.200850314
18. Jan Kolarˇı´k, Alessandro Pegoretti, Luca Fambri, Amabile Penati. High-Density Polyethylene/Cycloolefin Copolymer Blends, Part 2: Nonlinear Tensile Creep. Polym Eng Sci. 2006;46:1363–73. doi:10.1002/pen.20580
19. S. Taglialatela Scafati, L. Boragno, S. Losio, S. Limbo, M. Castellano, M. C. Sacchi, et al. Modulation of Barrier Properties of Monolayer Films from Blends of Polyethylene with Ethylene-co-Norbornene. Journal ofAppliedPolymer Science. 2011;121:3020–3027. doi:10.1002/app.33901
20. Alwaan IM. Effect of cyclo-olefin copolymer loading on kinetics, thermodynamics and model of linear low density polyethylene crystallinity. Progress in Rubber, Plastics and Recycling Technology. 2017;34(1):55–74.
21. Shah HC, Nema SK. The Effect Of Cyclic Olefin Copolymer Loading On Linear Low Density Polyethylene Blends : Characterization By Fourier-Transform Infrared Spectroscopy And X-Ray Diffraction. INTERNATIONAL JOURNAL OF SCIENTIFIC & TECHNOLOGY RESEARCH. 2019;8(08):1019–25.
22. Shah HC, Nema SK. Investigation of Thermal and Melt rheological properties of Linear Low Density Polyethylene / Cyclic olefin copolymer blends. International Journal for Research in Engineering Application & Management. 2019;(11):259–63.
23. Sabu Thomas, Yves Grohens, P Jyotishkumar. Characterization of Polymer Blends: Miscibility, Morphology and Interfaces. Wiley VCH Verlag GmbH & Co. KGaA; 2014. doi:10.1002/9783527645602
24. Moly KA, Bhagawan SS, Groeninckx G, Thomas S. Correlation between the morphology and dynamic mechanical properties of ethylene vinyl acetate/linear low-density polyethylene blends: Effects of the blend ratio and compatibilization. J Appl Polym Sci. 2006;100(6):4526–38. doi:10.1002/app.22466
25. Morris BA. THE SCIENCE AND TECHNOLOGY OF FLEXIBLE PACKAGING Multilayer Films from Resin and Process to End Use. Elsevier Inc.; 2017.
26. Pegoretti A., Kola!ík J., Fambri L. PA. Polypropylene/cycloolefin copolymer blends: effects of fibrous phase structure on tensile mechanical properties. Polymer (Guildf). 2003;44(11):3381–7.
27. Kolařík J, Kruliš Z, Šlouf M, Fambri L. High-density polyethylene/cycloolefin copolymer blends. Part 1: Phase structure, dynamic mechanical, tensile, and impact properties. Polym Eng Sci. 2005;45(6):817–26. doi:10.1002/pen.20337

28. Yingao Zhang CJHLAXDLJHMJ. Melt-reprocessable linear low-density polyethylene/cyclic olefin copolymer blends with low dielectric constant and low thermal expansion. J Appl Polym Sci. 2024;141(13).
29. Ren Y, Shi Y, Yao X, Tang Y, Liu LZ. Different dependence of tear strength on film orientation of LLDPE made with different co-monomer. Polymers (Basel). 2019;11(3). doi:10.3390/polym11030434
30. S. Sanchez-Valdes et al. Physical and mechanical properties of linear low density polyethylene/cycloolefin copolymer/layered silicate nanocomposite. Polym Compos. 2015. doi:10.1002/pc.23514
31. Ou CF, Hsu MC. Preparation and characterization of cyclo olefin copolymer (COC)/silica nanoparticle composites by solution blending. Journal of Polymer Research. 2007;14(5):373–8. doi:10.1007/s10965-007-9119-5
32. Dave P, Chandwani N, Nema SK, Mukherji S. Enhancement in gas diffusion barrier properties of polyethylene by plasma deposited SiOx films for food packaging applications. In: Nayak SK, Mohanty S, editors. Trends and Applications in Advanced Polymeric Materials. Scrivener Publishing LLC; 2018. p. 255–74.

