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Analyzing Impact Strength Improvement in Polymer Laminates: A Comparative Study of Machine Direction Oriented (MDO)-PE/LLDPE and Polyamide (PA)/LLDPE Structures

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Abstract

The mechanical strength of 2 film structures, machine-direction-oriented polyethylene (MDO-PE) laminated with linear low-density polyethylene (LLDPE) and polyamide (PA) laminated with LLDPE was evaluated. PA/LLDPE films demonstrated superior impact strength (0.834 J/mm²) compared to MDO-PE/LLDPE films (0.445 J/mm²). Tensile strength for modified LLDPE (C_6) was 40 N/mm², higher than standard LLDPE (C_8) at 33 N/mm². The elongation at break for LLDPE reference C_8 was 689 %, compared to 682 % for LLDPE sample C_6. Film stiffness was measured at 162 MPa for the LLDPE reference C_8 and 122 MPa for the LLDPE sample C_6. PA films exhibited increased mechanical strength due to their intrinsic polyamide structure, while MDO-PE films benefited from molecular orientation during production. The modified LLDPE or C_6 displayed improved impact resistance and tensile strength with reduced stiffness. MDO-PE/LLDPE films are promising for sustainable and robust food packaging.

Keywords: Flexible multilayer films, MDO-PE/LLDPE, PA/LLDPE, Mechanical properties, Laminated structures, Polyamide

Introduction

Flexible multilayer films have significantly transformed the food industry by extending the shelf life of products while preserving their texture, flavor, and moisture content, among other essential attributes. Achieving these desirable properties, including resistance to water vapor, gases, flavors, light, and flexibility or rigidity, often necessitates using a combination of polymers. While one type of polymer may offer advantages in terms of mechanical strength, it may fall short in aspects like transparency and permeability. Consequently, the food packaging sector must create multilayer films with distinct chemical, physical, and mechanical characteristics, employing various polymers through lamination or co-extrusion methods [1,2].

Typically, plastics with high impact resistance are employed in the packaging industry, featuring polyamides as the outer layer in packages that utilize Polyamide/Linear low-density polyethylene (PA/LLDPE). This combination is commonly used for packaging rice, cooking oil, and products where high impact resistance is essential to prevent leakage during the filling and distribution processes to end-users [3-5]. Polyamides arise from condensation polymerization, formed by merging diamines with diacids. Their naming convention hinges on the count of carbon atoms in the foundational materials. As an illustration, Nylon 6,6 comes from the union of hexamethylene diamine ($H_2N–(CH_2)_6–NH_2$) and 1,6-hexanediol acid ($HOOC–(CH_2)_4–COOH$), which is frequently referred to as adipic acid. In another approach, when originating from amino acids, they carry a singular numeric label indicating the carbon atom count of the precursor.

Machine Direction Orientation (MDO) technology is widely utilized to achieve the unidirectional elongation of polymer sheets. During the MDO procedure, the material is stretched in a semi-fluid condition, ensuring that the temperature during processing remains above the glass transition point but below the melting point. The extension of the material is facilitated by the tension created by a pair of rollers that turn in opposite directions and feature incrementally increasing rotation speeds [6]. Crucially, the differential linear velocities of the roller surfaces induce film elongation, not the direction of their rotation. The MDO apparatus can be seamlessly integrated into the extrusion process or operated independently, offering versatility with both blown and cast film techniques. The orientation procedure bestows enhanced smoothness and rigidity upon the film. Consequently, films processed through MDO exhibit improved performance at elevated speeds during subsequent processing stages, such as printing and lamination [7,8].

Typically, MDO-PE applications are employed in the MDO-PE/LLDPE laminated structure, which is envisioned as a more environmentally friendly alternative to the PA/LLDPE structure because MDO-PE and LLDPE are both polyethylene (PE) based, which often makes them more compatible with existing recycling streams. PA, on the other hand, can be more challenging to recycle due to its different chemical structure and melting points, which may not align with standard recycling processes for PE [3-5]. This study evaluates and determines the optimal LLDPE structure for a durable MDO-PE/LLDPE structure. The evaluation is based on various impact and mechanical tests, which will be compared head-to-head with PA/LLDPE, a material known for its significantly higher impact strength. This study contributes to the ongoing efforts in developing advanced polymer materials, expanding their utility across industry sectors.

Materials and methods

Figure 1 is a flowchart depicting the critical steps in the fabrication process of flexible multilayer film samples using the dry lamination process. Following fabrication, the samples are tested for characterization.

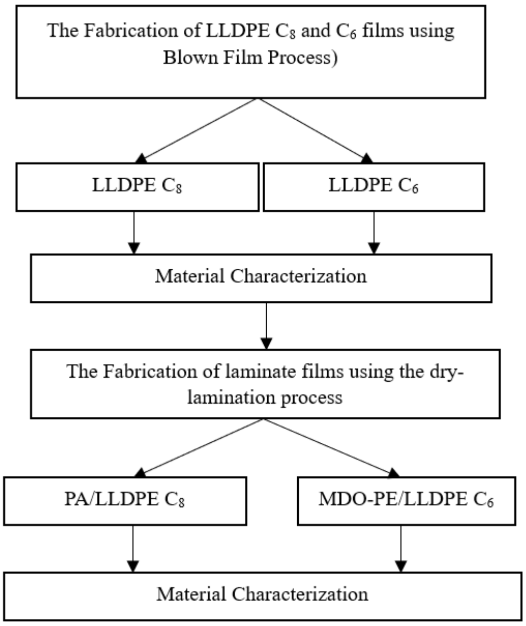


Figure 1 The process flow diagram for producing LLDPE films and constructing material structures using PA/LLDPE and MDO-PE/LLDPE lamination.

Materials fabrication

LLDPE films were produced using the blown film process that uses the BL-05 blown film machine, which operates with 3 extruders A/B/C. Each extruder has a diameter of 70, 105, and 70 mm, respectively. The die for film fabrication has a width of 630 mm and a maximum winder width of 2,600 mm. The feedstock for film formation was LLDPE C_8 and LLDPE C_6, respectively. Both materials were processed from 160 to 190 °C with a blow-up ratio (BUR) of 2.0. These results are film densities of 0.918 and 0.914 g/cm^3 and a consistent thickness of 80 µm for both films. The parameters set during film fabrication and the feedstock are summarized in **Table 1**.

Producing laminate films involves using dry lamination techniques to integrate LLDPE with PA and MDO-PE films. The DL-03 machine, which operates at an optimal speed of 120 m/min, was used in this process. The temperature of the lamination chamber is precisely controlled within the 70 - 80 °C range to facilitate optimal adhesion. The coating weight is standardized at 3.0 g/m^2. A specially formulated polyester-type adhesive is applied during the adhesive phase. The viscosity of the adhesive was evaluated using Zahn Cup DIN #3. The viscous material lasts for 16 - 17 s. To ensure uniform lamination, the temperature of the laminating rolls is kept constant at 45 to 50 °C. The MDO-PE films intended for lamination undergo a surface treatment to achieve a wettability of 38 dyne/cm². These films are defined by a thickness of 25 mm and a density of 0.930 g/cm³.

Table 1 Distribution of LLDPE C_8 & LLDPE C_6 resin and corresponding film thickness across extruders in the blown film fabrication process.

Extruder	Feedstock		Distribution ratio (%)	Setting film thickness per layer (µm)	Total film thickness (µm)
A	LLDPE C_8 n-octane	LLDPE C_6 n-hexane	25	20	80
B	LLDPE C_8 n-octane	LLDPE C_6 n-hexane	50	40	
C	LLDPE C_8 n-octane	LLDPE C_6 n-hexane	25	20	

Materials characterization

Material characterization assessed the impact and peel strength, tensile strength, elongation, modulus, and thermal properties. Impact strength for the LLDPE C_8 and C_6 films was determined using a Labthink Co Ltd FIT-01 equipped with a 19 mm pendulum. The apparatus was calibrated to assess impact energies ranging from 1 to 3 J/mm², applied to test specimens each 100 mm wide. ASTM D882 and ASTM F-904 standards [12,13] were used to evaluate tensile and peel strength, elongation, and young modulus. Thermal characterization of the LLDPE films was performed using a Mettler Toledo differential scanning calorimetry (DSC) in a nitrogen atmosphere. The heating rate was 10 deg./min, running from 0 to 200 °C. LLDPE samples were also analyzed using a Keyence VHX high-resolution microscope with a magnification of 1,000x to reveal their surface morphology.

In comparison, PA films undergo the same surface treatment level but vary with a thinner thickness of 15 µm, a higher molecular weight of 115.10, and a greater density of 1.16 g/cm³.

They are also evaluated for their dimensional stability under thermal stress, precisely their shrinkage rate in hot water. The mechanical strength of the laminated films is thoroughly examined through a set of standardized tests. These include impact strength and peel strength tests to evaluate the quality of the adhesive bond. Furthermore, tensile strength and elongation tests were also performed to measure the film's resistance to stretching forces and its ductility [14,15]. The young modulus is measured to determine the stiffness, providing a comprehensive understanding of the film's mechanical properties [3]. These evaluations are necessary to ensure the film performs in its intended applications and meets industrial standards.

Results and discussion

Comparative analysis MDO-PE vs PA film

Figure 2 compares the impact strength of MDO-PE and PA films. The impact strength values differ depending on the applied force's orientation. MD is the direction of the machine, while TD is the transversal direction, which is perpendicular to the direction of film processing. The impact strength of the PA film is recorded at 0.834 J/mm² along the MD, with a slightly lower value of 0.820 J/mm² along the TD. This minor difference indicates a quasi-isotropic impact resistance characteristic in PA, with minimal distinction in resistance between the MD and TD.

In contrast, MDO-PE films exhibit significant anisotropy in their mechanical response. The recorded impact strength in the MD is 0.445 J/mm², markedly inferior to that of PA films, and further diminishes to 0.321 J/mm² in the TD. This notable difference between the MD and TD readings of MDO-PE reflects its dependence on the directionality of mechanical stress, contrasting with the more uniform behavior of PA films.

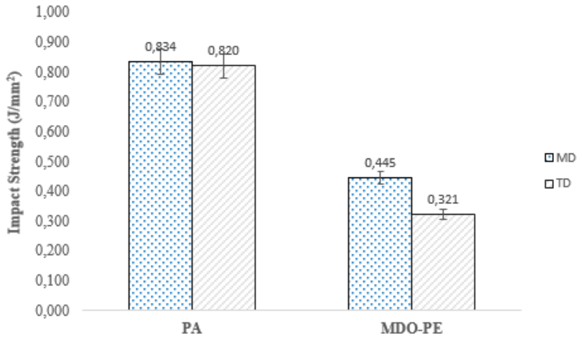


Figure 2 Comparison of MDO-PE and PA films data: Impact test.

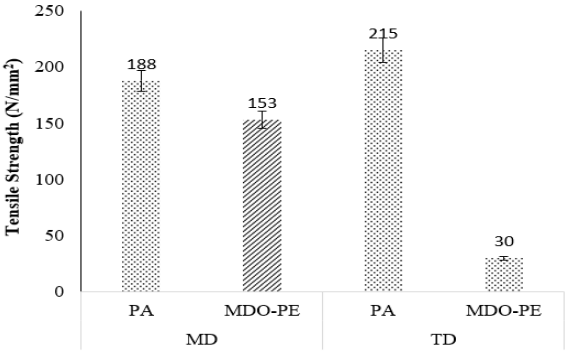


Figure 3 Comparison of MDO-PE and PA films data: Tensile strength.

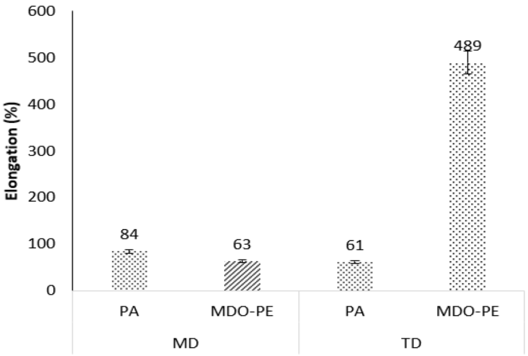


Figure 4 Comparison of MDO-PE and PA films data: Elongation

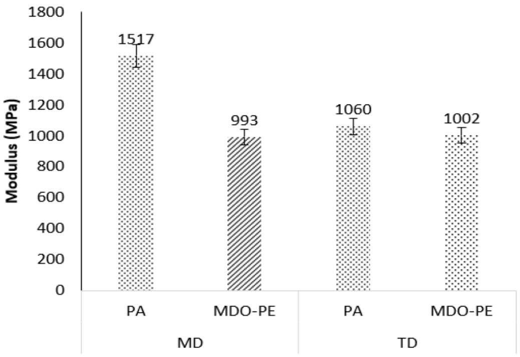
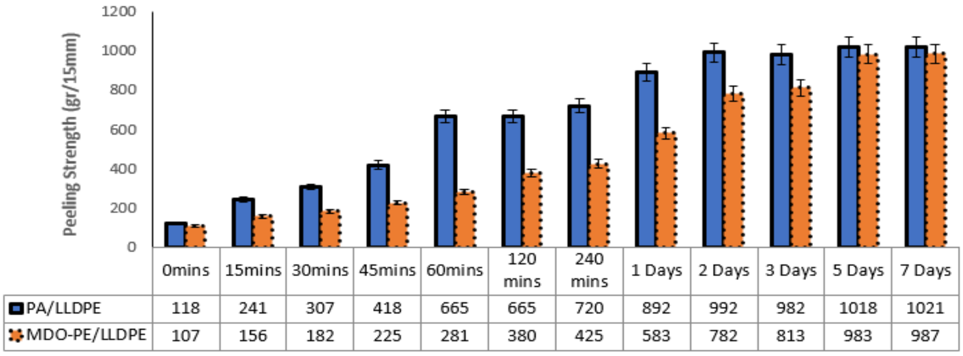


Figure 5 Comparison of MDO-PE and PA films data: Modulus.

The mechanical properties data of the films in **Figures 3** - **5** show the influence of direction, precisely the more pronounced anisotropic properties in MDO-PE samples, especially for tensile strength and elongation (see **Figures 3** and **4**). Meanwhile, the Young's modulus values of PA films differ by about 33 %, with the highest value exhibited by PA films in the MD. In contrast, for MDO-PE films, the influence of anisotropic properties is not prominent, nearly the same in all directions. The dissimilarities between PA and MDO-PE films arise because these 2 polymer films possess distinct characteristics attributable to their unique chemical compositions and production processes [4]. PA is characterized by an abundance of amide functional groups [5]. Within these functional groups, the C = O bond exhibits pronounced polarity, primarily arising from the disparity in electronegativity between carbon (C) and oxygen (O), giving rise to a charge distribution of C = O. Simultaneously, the N - H bond is subject to polarization, with a corresponding charge distribution in the N - H bond. As a result, these polarized functional groups facilitate the establishment of hydrogen bonds between the PA chains. Hydrogen bonds are usually seen as relatively weak interactions, but their profusion, due to the numerous amide groups in the PA chains [9], creates robust intermolecular peeling together. Consequently, the overall strength and durability of the material are significantly enhanced [9,17,18]. Molecular alignment in MDO polymer molecules has pronounced alignment along the machine direction [18,19].

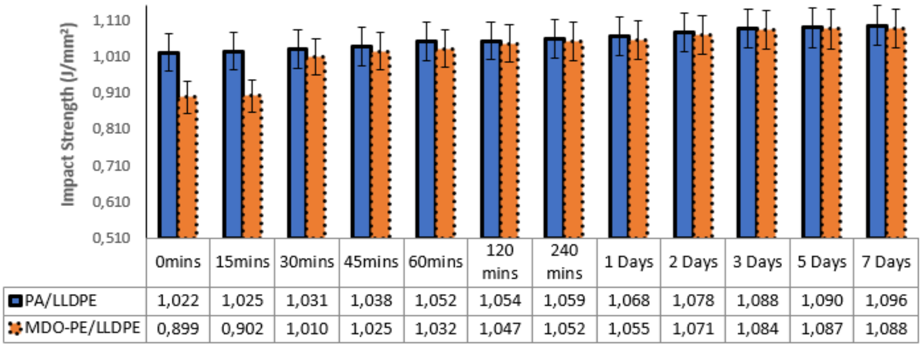
Effect of the peeling strength on the laminated films MDO-PE/LLDPE and PA/LLDPE

A comparison analysis of peel strength is presented in **Figure 6** between PA/LLDPE and MDO-PE/LLDPE, evaluated at specific aging intervals from 0 min to 7 days. Evaluating peel strength is essential for understanding the intrinsic properties of materials, especially in applications requiring robust adherence or cohesion.



	0mins	15mins	30mins	45mins	60mins	120 mins	240 mins	1 Days	2 Days	3 Days	5 Days	7 Days
■ PA/LLDPE	118	241	307	418	665	665	720	892	992	982	1018	1021
✱ MDO-PE/LLDPE	107	156	182	225	281	380	425	583	782	813	983	987

Figure 6 Peeling strength vs. aging time.



	0mins	15mins	30mins	45mins	60mins	120 mins	240 mins	1 Days	2 Days	3 Days	5 Days	7 Days
■ PA/LLDPE	1,022	1,025	1,031	1,038	1,052	1,054	1,059	1,068	1,078	1,088	1,090	1,096
✱ MDO-PE/LLDPE	0,899	0,902	1,010	1,025	1,032	1,047	1,052	1,055	1,071	1,084	1,087	1,088

Figure 7 Impact strength vs. aging time.

Variations in peeling strength tests, resulting from differences in testing methods or conditions for MDO-PE/LLDPE and PA/LLDPE laminated films, can significantly impact the overall impact strength of the laminated structure. A strong interlayer bond, reflected in enhanced peel strength, is crucial for effectively dispersing and absorbing impact forces, thereby substantially improving the material's resistance to impacts. In practical applications, laminated films with inadequate peel strength are susceptible to delamination, leading to layer separation and compromising structural integrity, ultimately reducing their ability to withstand impact forces.

At the commencement of the study, PA/LLDPE consistently demonstrated superior peeling strength relative to MDO-PE/LLDPE at all sampled time points. An initial reading marked 118 gr/15 mm for PA/LLDPE and 107 gr/15 mm for MDO-PE/LLDPE, delineating a distinct variance. As the aging process advanced, an augmentation in peeling strength was discernible for both materials. Nonetheless, the acceleration and uniformity of this enhancement presented variations between the 2 samples. During the preliminary 60 min, PA/LLDPE experienced a pronounced surge in peeling strength, escalating from 118 to 662 gr/15 mm. The growth trajectory leveled somewhat in the subsequent phase beyond the 60-minute threshold, registering a slight enhancement to 665 gr/15 mm at 120 min. This modest ascent persisted, attaining a peak of 1,021 by the conclusion of the 7-day duration. This trend intimates that PA/LLDPE might be characterized by swift initial peeling, which subsequently transitions to a stabilization phase.

Conversely, the peeling trajectory for MDO-PE/LLDPE manifested a more uniform yet gradual ascent. The peeling strength metric transitioned from 107 to 281 gr/15 mm within the initial hour. Following this phase, a methodical elevation was evident, reaching a 987 gr/15 mm zenith by the 7-day mark. Intriguingly, even with its reduced onset value and tempered growth velocity, the systematic progression of MDO-PE/LLDPE drew it proximate to the values exhibited by PA/LLDPE by the study's culmination.

Focusing initially on impact strength, shown in **Figure 7**, a clear progressive trend is evident. For the PA/LLDPE polymer, the baseline measurement was 1,022 J/mm^2, escalating to 1,096 J/mm^2 by the conclusion of the experimental phase. This consistent rise suggests an augmented resistance of the material to external mechanical perturbations. The MDO-PE/LLDPE sample demonstrated a similar trajectory in a parallel vein. There was a notable increment with an onset value of 0,899 J/mm^2, culminating at 1,088 J/mm^2 by 7 day, underscoring the polymer's augmented mechanical endurance over the duration.

The pattern highlights the material's increasing ability for improved intermolecular interactions. According to the data, the observed phenomenon in laminates indicates that peeling and impact strength improves over time due to the cross-linking process involving the formation of chemical bonds between polymer chains. This process can be likened to constructing bridges between the chains, significantly boosting the material's overall strength. Over time, these cross-links become more abundant and sturdier. Its is heightened cross-linking diminishes the mobility of polymer chains, making the material more challenging and more resistant to both peeling and impact forces [20,21].

A dataset in **Table 2** provides information on the mechanical properties of 2 LLDPE samples: LLDPE C_8 with n-octane and LLDPE C_6 with n-hexane. Crucial mechanical metrics have been quantified and compared: Impact strength, tensile strength, elongation, and modulus. The intrinsic density of each sample plays a significant role in the discernment of the exhibited behaviors.

Table 2 Summary of the mechanical properties of LLDPE film, with references and samples of LLDPE film.

Name	Impact strength (J/mm^2)	Tensile strength (N/mm^2)	Elongation (%)	Modulus (MPa)
LLDPE C_8, n-octane	1,385	33	689	162
LLDPE C_6, n-hexane	1,585	40	682	122

The sample of LLDPE C_6 demonstrates more outstanding toughness with an impact strength of 1,585 J/mm^2, significantly outperforming the C_8 sample, which has an impact strength of 1,385 J/mm^2. LLDPE C_6 is more resistant to impacts or breaks. Moreover, LLDPE C_6 exhibits a slightly lower density, and coupled with distinct physical characteristics employed in its production, this factor may contribute to its increased toughness and seems to have conferred improved energy absorption capabilities. Higher tensile strength in the LLDPE C_6 could suggest an altered molecular orientation or a more uniform crystalline arrangement, likely influenced by the density. Elongation percentages, representing the material's ductility, are relatively close for both samples. This marginal difference suggests that the intrinsic molecular chain mobility and entanglement are relatively unaffected by the density variations, retaining their inherent ductile nature [17,22]. The observed reduction in the modulus for the LLDPE C_6 sample suggests that it is a more flexible and less rigid material. A lower degree of crystallinity is likely responsible for this characteristic. Differential Scanning Calorimetry (DSC) data could support this inference, revealing that LLDPE C_6 exhibits lower crystallinity than LLDPE C_8. A polymer's crystallinity significantly impacts its mechanical properties, with lower crystallinity typically resulting in increased material flexibility.

Thermal properties of LLDPE

The thermograms of LLDPE C_6 and LLDPE C_8 are compared in **Figures 8** and **9**, from which some thermal properties can be calculated. Such thermal properties are presented in **Table 3**. The crystallinity percentage calculated using the crystallinity percentage formula is included [23,25]. In the first heating cycle, the melting peak of LLDPE C_6 occurs at 113 °C, while LLDPE C_8 exhibits a melting peak at 122 °C. Correspondingly, the percentage of crystallinity (% X_C) for LLDPE C_8 is significantly higher at 93.3 %, compared to 78.5 % for LLDPE C_6. In the second heating cycle, LLDPE C_6 again shows a melting peak at 117 °C with an increased % X_C of 75.8 %, whereas LLDPE C_8 presents a slightly reduced % X_C of 87.6 %. These observations indicate that LLDPE C_8 has a considerably higher degree of crystallinity than LLDPE C_6.

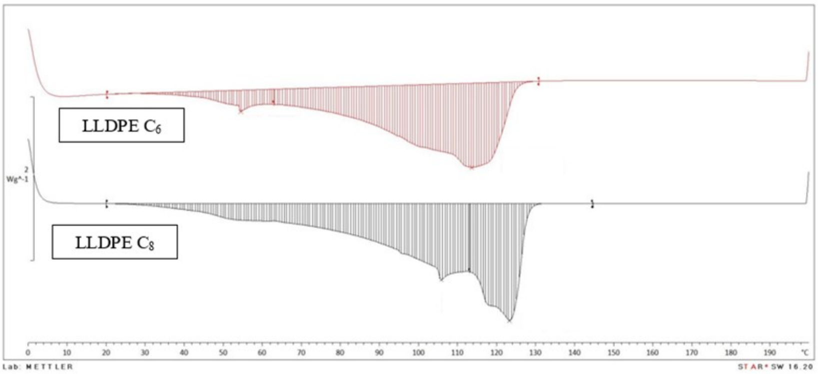


Figure 8 DSC analysis for LLDPE C_8 & C_6: 1st heating cycle.

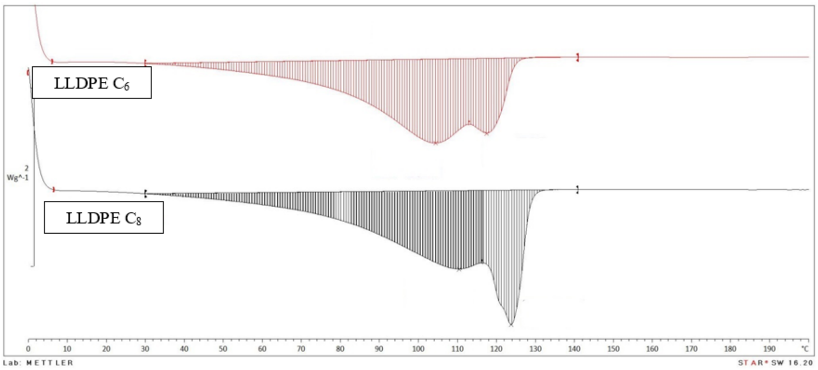


Figure 9 DSC analysis for LLDPE C_8 & C_6: 2nd heating cycle.

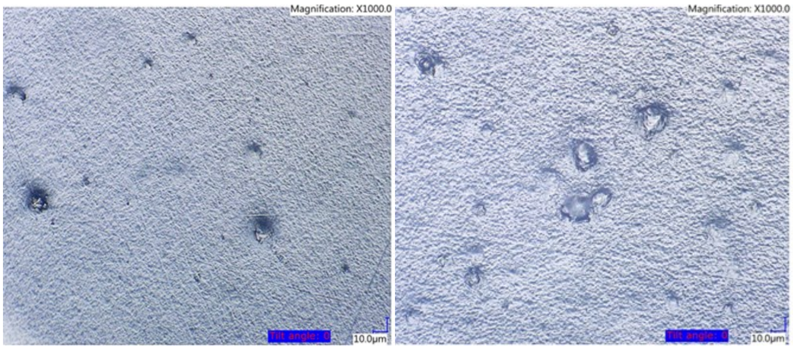
Table 3 LLDPE C_8 & LLDPE C_6 film thermal characteristics, as determined by DSC analysis.

	First Heating Peak Data			Second Heating Peak Data		
	T_m (°C)	H_m (J/g)	X_c (%)	T_m (°C)	H_m (J/g)	X_c (%)
LLDPE C_8	122	273,38	93,3	123	256,88	87,6
LLDPE C_6	113	230,25	78,5	117	222,13	75,8

This difference in crystallinity is reflected in the mechanical properties of the materials. LLDPE C_8 demonstrates higher elongation and modulus values, making its toughness more outstanding than LLDPE C_6. These findings have significant implications for selecting and applying these materials in various industrial contexts where specific mechanical properties are essential for performance.

Morphological characterization of LLDPE

Figures 10(a) - 10(b) show the surface morphology of LLDPE films, which indicate distinct morphological features in the 2 samples: LLDPE C_8 10(a) and LLDPE C_6 10(b). Both samples exhibit a consistent surface texture marked by infrequent scattered spherical inclusions.



(a) (b)

Figure 10 Surface morphology of (a) LLDPE C_8 and (b) LLDPE C_6.

Effect of mechanical strength laminated film for MDO-PE/LLDPE and PA/LLDPE

The analysis was also focused on the laminated films between MDO-PE with LLDPE C_6 and PA with LLDPE C_8. The mechanical effects observed show a significant decrease in tensile strength, elongation, and modulus for these laminated films, as illustrated in **Figures 11 - 13**. It is due to the breaking failure in the laminate, which decreased values and delamination of layers in both PA/LLDPE and MDO-PE/LLDPE. Though the focus on interfacial peeling is central to addressing adhesion challenges, the inherent mechanical qualities of the polymers cannot be overlooked when considering lamination success. Laminating 2 materials with differing mechanical characteristics can result in uneven stress distribution. For example, a pairing of 2 polymers: one is characterized by its elasticity and another by its stiffness. When exposed to stress, the more elastic polymer will likely elongate to a greater extent than its stiffer counterpart. Such variability in response can lead the stiffer polymer to absorb an inordinate share of the exerted force, rendering it prone to early failure. In summation, the lamination of polymers holds significant promise for advancing material attributes. Nonetheless, an in-depth evaluation of interfacial adhesion and the polymer's innate mechanical characteristics is essential. The development of superior laminated polymer laminates becomes attainable through judicious material choices and fine-tuning of lamination procedures.

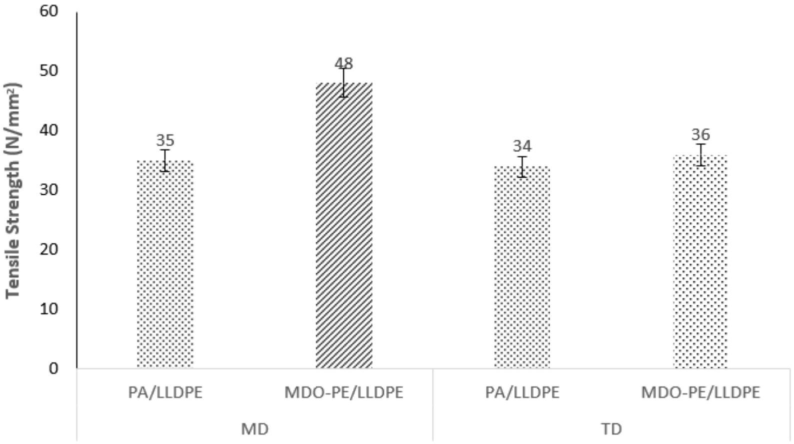


Figure 11 Comparison of tensile strength: PA/LLDPE vs. MDO-PE/LLDPE in MD/TD directions.

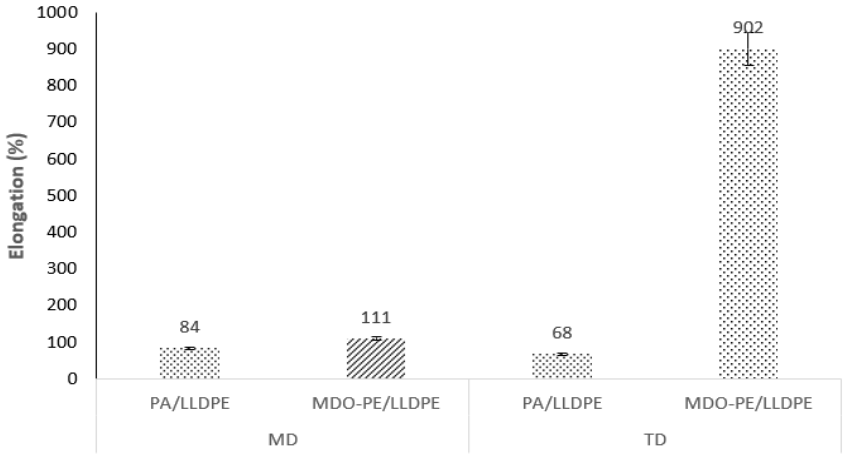


Figure 12 Comparison of elongation: PA/LLDPE vs. MDO-PE/LLDPE in MD/TD directions.

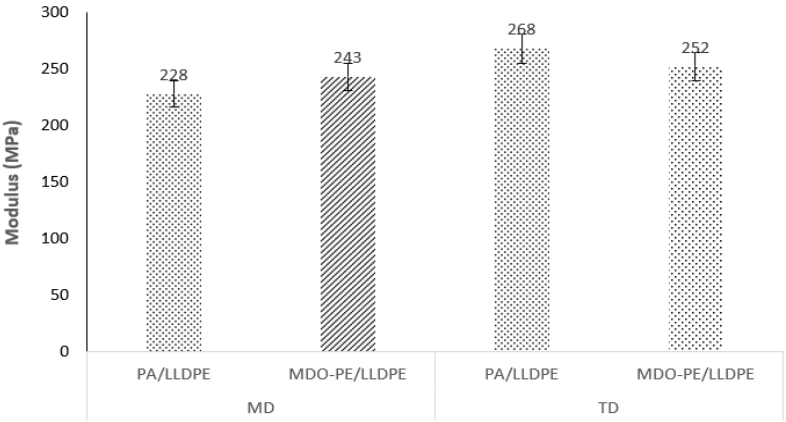


Figure 13 Comparison of modulus: PA/LLDPE vs. MDO-PE/LLDPE in MD/TD directions.

In the MD orientation, PA/LLDPE shows a modulus of 228 MPa, whereas MDO-PE/LLDPE has a slightly higher modulus of 243 MPa. It indicates that MDO-PE/LLDPE is stiffer or more resistant to deformation along the MD when compared to PA/LLDPE. In the TD orientation, the modulus for PA/LLDPE increases significantly to 258 MPa, suggesting that this material is stiffer in the TD than in the MD. Conversely, MDO-PE/LLDPE shows a slight decrease in the modulus to 252 MPa in the TD, which is still higher than PA/LLDPE in the MD but shows that MDO-PE/LLDPE is less anisotropic than PA/LLDPE in terms of stiffness.

In evaluating the mechanical robustness and adhesion characteristics of laminated film, a systematic comparison was conducted between PA/LLDPE and MDO-PE/LLDPE materials. The subsequent section presents a tabulated summary of the quantitative findings. This data elucidates the inherent mechanical properties and the potential applicability of each laminate type under various stress conditions. These findings are crucial for determining the suitability of each film type for specific industrial applications. The detailed analysis, as shown in **Table 4**, underpins the discussion on the material selection for optimized packaging solutions.

Table 4 Comparative performance analysis of PA/LLDPE and MDO-PE/LLDPE laminates in key mechanical tests.

Research parameter	PA/LLDPE	MDO-PE/LLDPE	Details & Observation
Initial peeling strength	118 g/15 mm	107 g/15 mm	Initial measurements indicate a slightly higher peeling strength for PA/LLDPE. This parameter is crucial for understanding the initial adhesive properties of the laminates.
Peeling strength at 7 days	1,021 g/15 mm	987 g/15 mm	Both materials demonstrate increased peeling strength over 7 days, indicative of enduring adhesive interactions, with PA/LLDPE maintaining a slight advantage.
Impact strength (MD)	0.834 J/mm²	0.445 J/mm²	The pronounced impact strength in the machine direction for PA/LLDPE may be attributed to its superior energy dissipation capabilities under dynamic loading conditions.
Impact strength (TD)	0.820 J/mm²	0.321 J/mm²	Enhanced impact strength in the transverse direction for PA/LLDPE underscores its isotropic mechanical behavior, which is desirable for multidirectional stress environments.
Laminates film strength	Influenced by the combination of polymers	Influenced by the combination of polymers	The polymer constituents significantly influence the laminate's intrinsic mechanical strength, underscoring the synergy in laminated material.
Overall assessment	Strong initial adhesion, optimal for rapid-application contexts	Potential for long-term adhesive stability, indicative of durability	The findings suggest that while PA/LLDPE is optimized for applications where immediate adhesion is critical, MDO-PE/LLDPE shows promise for longevity and sustained performance applications.

Conclusions

Based on the results and depth analysis of the mechanical and peel strength characteristics of MDO-PE/LLDPE and PA/LLDPE laminate structures, the following conclusions can be drawn:

1) PA vs. MDO-PE Films: Due to their chemical constitution, they demonstrate superior mechanical strength over MDO-PE films. This strength is attributed to the abundant amide groups facilitating robust hydrogen bonds.

2) Peeling Strength in Laminated Films: An evaluation of MDO-PE/LLDPE and PA/LLDPE reveals PA/LLDPE's superior initial peeling. However, MDO-PE/LLDPE's consistent peeling growth narrows this gap, suggesting its potential for sustained adhesive progression.

3) LLDPE Dynamics: Comparing LLDPE C_8 with LLDPE C_6 underscores density's role in shaping mechanical traits. LLDPE Sample C_6, with its altered density, offers improved impact and tensile strength but displays a reduced modulus and elongation.

4) Laminates Film Strength: Their constituent layers dictate the laminated film's mechanical attributes. The observed discrepancies in strength, leading to potential structural challenges, highlight the intricacies of combining polymers with varied mechanical properties.

In a comprehensive evaluation, the MDO-PE/LLDPE laminate structure emerges as a potential alternative to the PA/LLDPE laminate, primarily due to its nearly comparable impact strength. As the industry prioritizes sustainable and cost-effective packaging solutions, MDO-PE/LLDPE laminate films present promising advantages. However, a thorough understanding and rigorous validation are necessary to realize these benefits fully.

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