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Plasticization of Poly (lactic acid) Using Epoxidized Palm Stearin

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Abstract. Polylactic acid (PLA) is a promising biodegradable alternative to petroleum-based polymers, yet its industrial applications are hindered by inherent brittleness and low elongation at break. This study aims to enhance PLA's mechanical performance by investigating the effects of epoxidized palm stearin (EPS) as a bio-based plasticizer and starch as a reinforcing filler. EPS was selected as a sustainable, reactive additive because its epoxy groups interact with PLA, thereby improving chain mobility and flexibility. Starch was incorporated as a natural filler because it is cost-effective, renewable, and can act as a reinforcing agent to mitigate the loss of stiffness and toughness often caused by excessive plasticization. The primary objective of the study was to evaluate the thermal and mechanical properties of these blends to determine the optimal composition for balanced flexibility and strength. Blends containing 5–20 wt.% EPS and a fixed 5 wt.% starch were prepared via melt blending. Thermal analysis via DSC revealed that increasing EPS content successfully reduced the glass transition temperature (T_g) and cold crystallization temperature (T_{cc}), confirming improved molecular mobility. Mechanically, the PLA/EPS20/St5 blend achieved the highest elongation at break at 15.65%, while the addition of starch consistently improved impact strength across all EPS-containing blends. Although excessive EPS loading eventually reduced tensile strength, the results demonstrate that the combination of EPS and starch offers a viable, eco-friendly strategy to tailor PLA properties for industrial use.

Keywords: Biopolymer, Epoxidized Palm Stearin (EPS), Plasticizer, Polylactic Acid (PLA), Starch

INTRODUCTION

Petroleum-based polymers such as polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), polyethylene terephthalate (PET), and biaxially oriented polypropylene (BOPP) are widely used in manufacturing a wide range of polymeric products (Yue *et al.,* 2024). These materials are used across various sectors, including engineering applications, advanced technologies, and commodity goods. Most of these materials are non-degradable since they are largely derived from fossil fuels. These products contribute significantly to environmental pollution due to their persistence in the ecosystem when discarded (Vega-Baudrit, 2019). Consequently, there is a growing need for sustainability to address these challenges, leading to extensive research into the development of biodegradable polymers derived from

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renewable resources.

Biodegradable polymers have attracted significant attention due to their environmental friendliness and low toxicity, offering clear advantages for both consumers and the environment (Moshood *et al.,* 2022). Most of these materials are designed to decompose naturally, as they can be synthesized from various renewable resources, such as natural polymers (agro-polymers), microbial polymers (synthesized via microbial fermentation), and synthetic polymers (derived from renewable bio-derived monomers) (Ali *et al.,* 2016).

PLA is an alternative to petrochemical polymers due to its mechanical characteristics, such as Young's modulus and tensile strength, which are comparable to those of nylon or polyethylene terephthalate (PET) (Chieng *et al.,* 2014). PLA with the chemical formula $(C_3H_4O_2)_n$ (Figure 1) belongs to the family of aliphatic polyesters and is commonly synthesized from a-hydroxy acids (Garlotta, 2001). As an extensive property, PLA is suitable for various industries such as packaging, textiles, automotive, and clinical applications (Hussain *et al.,* 2024).

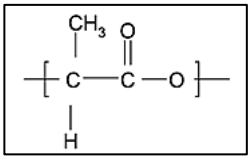


Fig. 1: Structure of the PLA

Despite the many attractive properties of PLA, such as high modulus, tensile strength, biodegradability, and processability, it also exhibits poor characteristics, including low elongation at break, low toughness, and high brittleness, which restrict its use in various applications (Ramezani Dana & Ebrahimi, 2023). PLA has an elongation at break of less than 10%, indicating that it can fracture under tensile stress with minimal deformation (Lin & Qu, 2019). This low elongation indicates a brittle nature, as the material tends to break rather than stretch significantly before failure, making it unsuitable for applications that require flexibility or impact absorption without breaking.

Therefore, many studies have been conducted to improve mechanical properties, and better meet end-user demands. To address the limitations in the flexibility and mechanical properties of PLA, various approaches have been developed, including blending PLA with other polymers such as polybutylene adipate terephthalate, polycaprolactone, or polybutylene succinate (X. Li *et al.,* 2022). Additionally, incorporating plasticizers and fibers into the polymer matrix has been employed to enhance further the mechanical properties of PLA (Sanivada *et al.,* 2020). Additives derived from various vegetable oils through epoxidation have emerged as promising and sustainable alternative plasticizers, stabilizers, and modifiers for polymers, coatings, and resins. It is created by introducing epoxy groups into the chemical structure of vegetable oil, providing a reactive functional group that acts as a plasticizer in PLA.

Commonly, the use of epoxidized vegetable oils as plasticizers is limited to concentrations of approximately 1% to 10% (Omar *et al.,* 2021). For instance, the thermal properties of PLA blended with epoxidized sunflower oil (ESO) have been studied, showing a decrease in the glass transition temperature (T_g) from 54°C to 51°C with the addition of 3 wt.% of ESO. The T_g continued to decrease gradually with increasing ESO content alongside a similar reduction in the cold crystallization temperature (T_{cc}) (Bouti et al., 2022), indicating that the addition of ESO improves PLA chain mobility.

Moreover, the tensile properties of PLA blended with epoxidized palm oil (EPO) have significantly improved flexibility. In addition, the elongation at break of PLA increases from 5.3% to 114.4% with 5 wt.% of EPO due to the introduction of a plasticizer to a polymer matrix by disrupting extensive intermolecular interactions, which reduces the brittleness (Bond Tee *et al.,* 2016).

In addition to plasticization, incorporating natural fillers such as starch has shown great promise for enhancing the mechanical properties of PLA (Tan *et al.,* 2022). Potato starch is increasingly explored as a filler in polymer blends due to its renewable, biodegradable, and cost-effective nature. In polymer blends, potato starch has been used in various polymers, such as polypropylene, PBAT (polybutylene adipate terephthalate, polyethylene, and polylactic acid (PLA) (Gupta *et al*., 2008; Knitter & Dobrzyńska-Mizera, 2015; Vanin *et al.,* 2025; Weng *et al.,* 2025). The application of potato starch in PLA blends offers high compatibility, improves mechanical properties, and enhances biodegradability (Chauhan *et al.,* 2021; Licea-Saucedo *et al.,* 2024; Vanin *et al.,* 2025).

While plasticization improves the flexibility of PLA, starch can act as a natural filler, increasing tensile strength and toughness, especially when plasticizers are used in excess (Awale *et al.,* 2018). This improvement is due to the rigid structure of starch and potential for hydrogen bonding with the PLA matrix, which enhances interfacial adhesion. The study has reported that adding 5 wt.% starch to PLA/20 wt.% EPO has significantly improved the elongation-at-break and impact strength compared to the PLA/5 wt.% EPO. The elongation at break increased from 15.7% to 1321% as the amount of EPO increased from 5 wt.% to 20 wt.%. Meanwhile, the impact strength increased from 50 J/m to 80 J/m (Awale *et al.,* 2018). These findings highlight the potential of combining starch with epoxidized vegetable oil to improve both the stiffness and ductility of PLA composites.

Thus, this study investigates the effect of epoxidized palm stearin (EPS) as a plasticizer on the properties of PLA, as well as the influence of 5 wt.% starch content as a natural filler in enhancing the mechanical properties of PLS/EPS blends. The research also aims to determine the optimal blend composition of PLA/EPS/starch that achieves a balanced improvement in both flexibility and mechanical performance.

MATERIALS AND METHODS

Polylactic acid (PLA, extrusion grade) was obtained from Xiamen Keyuan Plastic Co., Ltd. in pellet form with a density of1240 kg/m^3 and a melt flow index (MFI) of 9 g/10 min. This semi-crystalline PLA grade contains 1.4 wt.% D-lactide. Epoxidized palm stearin and potato starch were obtained from the Polymer Laboratory at Universiti Teknologi MARA.

Blends Preparation

The preparation of poly(lactic) acid (PLA) epoxidized palm stearin (EPS), and starch blends was carried out by drying the PLA granules in a vacuum oven (Model: Taisite VO-24D) at 60°C for 5 hours to remove any residual moisture. The dried PLA granules, EPS, and starch were then weighed according to the blend ratios specified in Table 1. The composition was based on previous work on the potential application of PLA/palm stearin (PS) blends (Aiman *et al*., 2022). The premixed materials were processed by melt blending using an internal mixer (Haake Rheomix OS

Thermo Scientific) at 160°C and a screw speed of 50 rpm for 10 minutes. After mixing, the resulting blends were crushed with a compact crusher to obtain tiny pellets. The pellets were stored in a glass desiccator to prevent oxidation and moisture absorption, ensuring their quality for further analysis.

Table 1: Composition and ratio of (PLA, EPS and Starch) in the PLA/EPS/starch blend.

Blend ID	Composition Ratio (wt.%)		
	PLA	EPS	Starch
PLA	100	0	0
PLA/EPS5/St5	90	5	5
PLA/EPS10/St5	85	10	5
PLA/EPS15/St5	80	15	5
PLA/EPS20/St5	75	20	5

Characterization

Tensile Testing

The mechanical properties of the PLA/EPS/starch blends were evaluated using tensile testing. Dumbbell-shaped specimens were prepared using a hot-press molding technique in accordance with ASTM D638 Type I specifications. The processing involved pre-heating the samples at 165°C for 2 minutes, followed by 5 minutes of full heating. The samples were then cooled in two stages: first to 70°C over 7 minutes, then to 45°C over 8 minutes. The resulting specimens had a thickness of approximately 3 mm and a total length of 165 mm. The tensile test was carried out using Tinius Olson's Universal Tensile Machine (Model: H50KT) equipped with a 1 kN load cell. The tests were conducted at a crosshead speed of 10mm/min at room temperature 23°C.

Impact Strength Testing

The impact strength of PLA/EPS/starch blends was determined using the Izod notched impact test in accordance with ASTM D256 (Awale *et al*., 2018). The test was performed on a Tinius Olsen Model Impact 503 pendulum impact tester. Specimens were prepared with the following standard dimensions: 64 mm length, 12.7 mm width, and 10.2 mm thickness. A V-notch was introduced to each specimen with a depth of 10.2 mm and a notch radius of 2.5 mm. The pendulum energy used during testing was 2.75 J with a specified drop height of 610 ± 2 mm. The impact strength was calculated by measuring the energy absorbed during the fracture and normalizing it over the cross-sectional area at the notch.

Differential Scanning Calorimetry

The glass transition temperature (T_g), melting temperature (T_m), and cold crystallization temperature (T_{cc}) of the composites were measured using a differential scanning calorimeter (Mettler Toledo/DSC1). Approximately 0.5 g (±0.05 g) of each sample was weighed and pre-sealed into a 40 µl aluminum pan. Thermal analysis was performed using a heat-cool-heat cycle. The procedure involves heating the sample from 25°C to 200°C, cooling it to -50°C, and reheating it to 200°C. All at a consistent heating rate of 10°C/min under a nitrogen atmosphere. Neat PLA was analyzed under the same conditions for comparison purposes.

RESULTS AND DISCUSSION

Sample Appearance

The appearance, thermal and mechanical behavior of PLA and PLA/EPS/starch blends were investigated to evaluate the effect of epoxidized palm stearin (EPS) and 5 wt.% starch on the structural and functional properties of the PLA matrix.

Figure 2 shows five specimens of PLA

and PLA/EPS blends containing 5 wt.% starch. The first sample (neat PLA) appears smooth and semi-transparent, while the subsequent samples become progressively more opaque and darker in color. This visual change reflects the increasing presence of EPS in the blend.

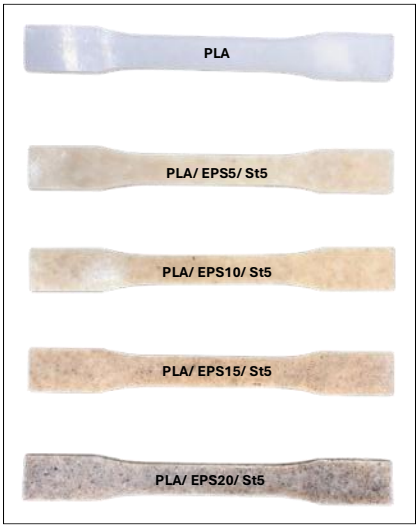


Fig. 2: PLA and PLA/EPS/Starch blend specimens with varying EPS content.

Thermal Properties

Differential Scanning Calorimetry (DSC)

Thermal properties of neat PLA, PLA/EPS/Starch blends with varying EPS content are shown in Figure 3 and summarized in Table 2. The thermal properties derived from the analysis include the glass transition temperature (T_g), cold crystallization temperature (T_{cc}), melting temperature (T_m), enthalpy of fusion ΔH_m, and degree of crystallinity (X_c). The T_g was determined from the onset temperature of the glass transition, while T_{cc} and T_m were taken from the peak maxima in the DSC curves.

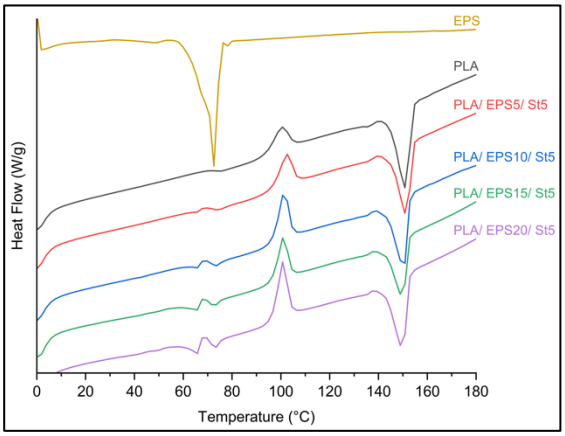


Fig. 3: Heating curve of differential scanning calorimetry (DSC) thermograms of PLA and PLA/EPS/Starch blends.

As shown in Table 2, the neat PLA exhibited a T_g of 73.54°C, while the T_g of the PLA/EPS/starch blends decreased with increasing EPS content to 67.73°C in the PLA/EPS20/St5 blend. This indicates that the material became flexible at a lower temperature due to the plasticizing effect of EPS, which increases the mobility of PLA chains (Saini *et al.*, 2024). The decrease in T_g is a characteristic of plasticization, in which the plasticizer disrupts intermolecular interactions between polymer chains, allowing them to move more freely (Mascia *et al.*, 2020). As a result, the polymer transitions from a rigid, glassy state to a more flexible, rubber state at a lower temperature. A similar

Table 2: Thermal properties of PLA and PLA/EPS/Starch blend.

Blend ID	T_g (°C)	T_{cc} (°C)	T_{m1} (°C)	T_{m2} (°C)	ΔH_m (J/g)	X_c (%)
PLA	73.54	105.67	0.00	150.82	12.99	13.96
PLA/ EPS5/St5	71.60	105.62	65.80	150.84	11.16	13.34
PLA/ EPS10/St5	69.76	103.70	65.78	150.85	11.16	14.09
PLA/ EPS15/St5	69.66	101.71	65.75	148.91	10.88	14.63
PLA/ EPS20/St5	67.73	100.72	63.86	148.90	63.86	11.82

reduction in T_g has been reported in PLA blends with other epoxidized oils such as epoxidized palm oil (EPO) and with starch. The addition of 20 wt.% EPO and 5 wt.% starch reduce the T_g from 59.10°C to 56.10°C (Awale *et al.,* 2018). Enhanced chain mobility reduces the temperature required for the polymer to undergo its glass transition (Qian *et al.,* 2019).

Therefore, the observed decrease in T_g with increasing EPS content indicates that the PLA chains become less rigid and more flexible.

The cold crystallization temperature (T_{cc}) also decreased with increasing EPS content. T_{cc} refers to the temperature at which a polymer transitions from its amorphous state to a crystalline state during cooling. From Table 2, the PLA showed a T_{cc} of 105.67°C, which remained unchanged for PLA/EPS5/St5 but decreased to 100.72°C for PLA/EPS20/St5. The reduction is attributed to the plasticizing effect of EPS, which increases chain mobility and alters the polymer's crystallization kinetics (S. Li *et al.,* 2021). Although increased molecular motion generally promotes crystallization, it can also delay the onset of crystallite formation by disrupting regular chain alignment, thereby slightly reducing T_{cc} (Qu *et al.,* 2023). Lower crystallization temperatures typically indicate a more amorphous polymer structure, which results in greater flexibility and reduced brittleness (Qu *et al.,* 2023).

The melting temperature (T_{m2}) also decreased slightly upon incorporation of EPS, indicating that the presence of the plasticizer influenced melting behavior. This occurs because plasticizers weaken intermolecular forces and reduce crystallinity, allowing polymer chains to melt at lower temperatures (Mascia *et al.,* 2020). As shown in Table 2 and Figure 3, the neat PLA exhibited a single melting peak at 150.82°C. With the incorporation of EPS, the PLS/EPS/starch blends exhibited two melting peaks, with T_{m1} in the range of 65.80°C to 63.86°C and T_{m2} around 150.84°C to 148.90°C. The appearance of T_{m1} corresponds to the melting of EPS, which occurs at 72.54°C and 72.16°C. It shows that EPS melts before interacting with the PLA matrix during heating. As EPS diffuses into molten PLA, it disrupts the polymer chains and reduces PLA crystallinity. This effect is reflected in the observed decrease in T_{m2}, indicating that the material becomes less rigid and more flexible at lower temperatures. This observation is further supported by the reduction in T_g values across all blends, indicating increased chain mobility and confirming the miscibility of EPS with PLA.

The minor shift in T_{m2} suggests that while EPS altered chain mobility and crystallization kinetics, it did not significantly disrupt the crystalline phase of PLA. The crystals formed in the presence of EPS retained comparable thermal stability, indicating that the primary crystalline structure of PLA was largely preserved despite the increase in amorphous regions (Mondragón-Herrera *et al.,* 2024). Previous studies reported no major shift in T_m upon the addition of ESO and EPO, confirming that these plasticizers influence mobility without changing the crystalline phase of PLA (Bond Tee *et al.,* 2016).

Mechanical Properties

Izod Impact Strength Test

The impact strength of neat PLA and PLA/EPS blends at various percentages, with the addition of 5 wt.% starch, is presented in Figure 4.

Neat PLA exhibits the highest impact strength at 11.50 J/m, which decreases with the incorporation of EPS. As the EPS content

increases, the impact strength of the blends decreases. This trend suggests a reduction in toughness due to the plasticizing effect of EPS, which weakens intermolecular interactions within the PLA matrix (Schyns & Shaver, 2021). As EPS content increases, the matrix becomes softer, which makes it less capable of withstanding sudden impact forces.

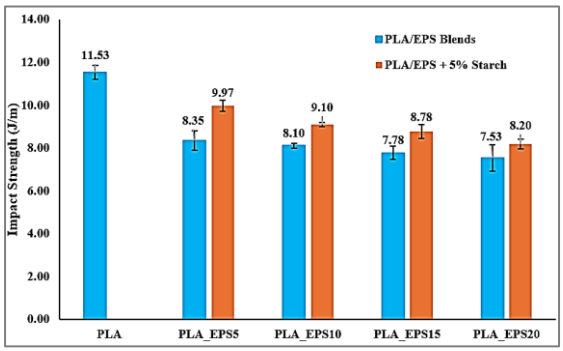


Fig. 4: Impact strength of PLA and PLA/EPS Blends with and without 5 wt.% Starch.

The addition of 5 wt.% starch improves in impact strength across all PLA/EPS blends. The addition of starch resulted in increases in impact strength of approximately 19.4% (PLA/EPS5), 12.3% (PLA/EPS10), 12.9% (PLA/EPS15), and 8.9% (PLA/EPS20) compared to EPS blends. Starch acts as a reinforcing filler that helps compensate for the reduction in toughness caused by EPS's the plasticizing effect (Tan *et al.,* 2022). It enhances energy absorption during impact and improves bonding between the filler and the PLA matrix. As a result, the overall structure becomes more resistant to sudden forces. This leads to a consistent increase in impact strength even though EPS alone tends to reduce it. These findings suggest that starch can help balance the softening effects of EPS and improve the mechanical strength of PLA.

This observation is supported by DSC analysis, which showed a decrease in T_g and T_{cc}. These thermal changes increased molecular motion and shifted the material toward a more amorphous structure, both of which reduce impact resistance (Perez-Nakai *et al.,* 2023). The incorporation of starch thus appears to mitigate some of the structural compromises introduced by EPS, leading to a more balanced material in terms of flexibility and toughness.

Young's Modulus

The Young's modulus values of neat PLA and PLA/EPS blends containing 5 wt.% starch are shown in Figure 5.

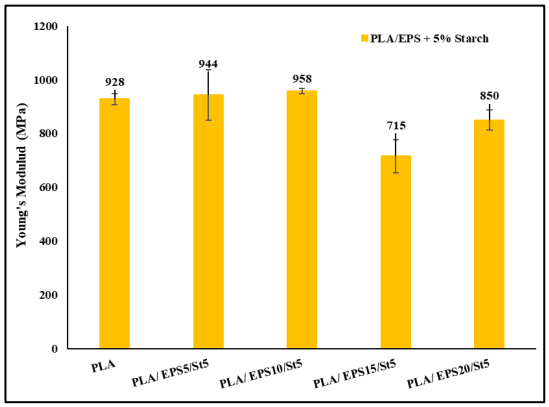


Fig. 5: Young's modulus of PLA and PLA/EPS blends with 5 wt.% starch.

Neat PLA exhibited a modulus of 928 MPa, indicating its relatively stiff and brittle nature. At lower EPS loadings, PLA/EPS5/St5 and PLA/EPS10/St5, the modulus remained stable or increased slightly, which indicates that small amounts of EPS did not significantly affect the overall stiffness of the PLA matrix. This may be due to limited plasticizer interaction at low concentrations, which allows the matrix to retain its stiffness (Awale *et al.,* 2018). However, a reduction in modulus was observed in PLA/EPS15/St5, with the lowest value of 715 MPa. This indicates a significant loss of stiffness and increased material flexibility, attributable to the plasticizing effect of EPS, which increases

the free volume between polymer chains and reduces the intermolecular forces that contribute to rigidity (Eslami *et al.,* 2023).

Nevertheless, PLA/EPS20/St5 showed a slight recovery in modulus compared to PLA/EPS15/St5. This could be due to improved dispersion or better interaction of EPS and starch within the matrix at higher concentrations. Overall, the results demonstrate that while EPS enhances ductility, excessive plasticizer loading can compromise the material's stiffness. The addition of 5 wt.% starch may help mitigate some of this softening by providing mild reinforcement, maintaining mechanical stability while allowing for improved flexibility.

Tensile Strength

The tensile strength of neat PLA and PLA/EPS blends containing 5 wt.% starch is presented in Figure 6.

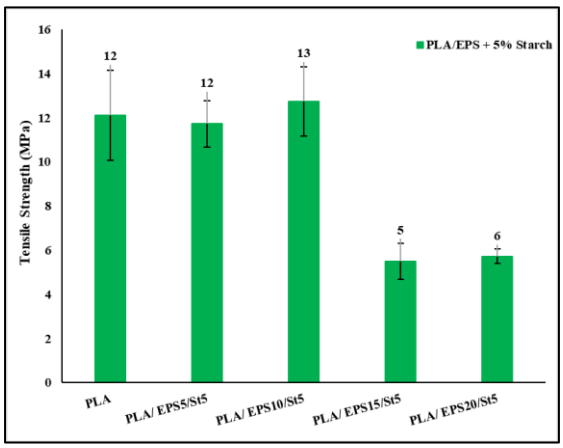


Fig. 6: Tensile strength of PLA and PLA/EPS blends with 5 wt.% starch.

Neat PLA exhibited a tensile strength of 12 MPa. With the addition of EPS, the tensile strength of PLA/EPS5/St5 showed no significant difference compared to neat PLA, whereas the PLA/EPS10/St5 had the highest tensile strength. This observation aligns with the high Young's modulus values, which indicate the material remained stiff and structurally rigid at these compositions. Therefore, the low EPS content did not substantially change the polymer structure as the plasticizer was not present in sufficient quantities to disrupt the intermolecular interactions within the PLA matrix.

However, a sharp decline in tensile strength was observed at higher EPS concentrations. The PLA/EPS15/St5 and PLA/EPS20/St5 recorded significantly lower tensile strength values of 5 MPa and 6 MPa, respectively. The reduction in both tensile strength and rigidity of PLA with increasing EPS content can be explained by the disruption of polymer-polymer interactions, which weaken the material's structural integrity (Augustine O. Ayeni *et al.,* 2022; Lazaro-Hdez *et al.,* 2025). This possibility is due to the diffusion of plasticizer in the polymer matrix (Wypych, 2023). As EPS content increases, more of these connections in the polymer matrix are disrupted, softening the matrix and reducing tensile strength (Mastalygina & Aleksanyan, 2024).

This behavior is consistent with the thermal analysis, in which the decrease in T_g reflects improved chain mobility. As polymer chains become more flexible, they may be less capable of withstanding tensile loads, leading to lower tensile strength at higher EPS content. However, this increased flexibility is beneficial to elongation at break. Overall, the results confirm that EPS enhances ductility but may reduce tensile strength at high concentrations, and that starch helps maintain mechanical strength only within a limited range of EPS concentrations.

Elongation at Break

The elongation at break of PLA and PLA/EPS blends containing 5 wt.% starch is shown in Figure 7.

Neat PLA exhibited an elongation of 8.01%. With the incorporation of EPS, the elongation at break initially decreased slightly up to PLA/EPS15. However, a significant increase was observed in PLA/EPS20, which reached the highest elongation at break value at 15.65%. This trend aligns with the reduction in Young's modulus and tensile strength, where, at higher EPS content, the material became less stiff and more flexible. The increased flexibility and softening effect due to plasticization allows the material to deform more before failure, with the addition of 5 wt.% starch to the PLA/EPS20, the elongation at break has increased significantly to 15.65% from 7.01% in PLA/EPS5. This trend is comparable to the results reported by Awale *et al.* (2018), where the elongation at break for PLA/EPO20wt.% increased significantly to 1321% from 15.7% when 5 wt.% starch was added to the blend.

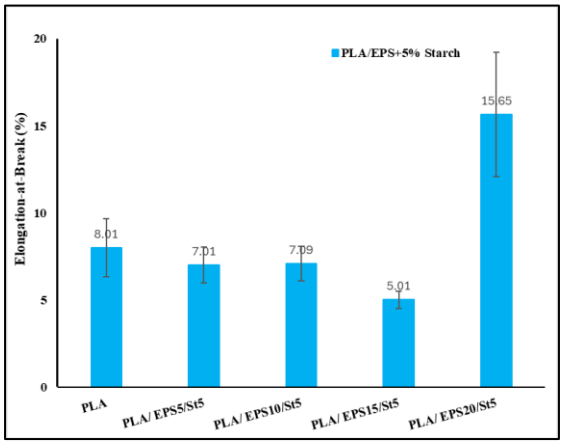


Fig. 7: Elongation at break of PLA and PLA/EPS blends containing 5 wt.% starch.

EPS contains oxirane (epoxy) groups that act as polar components, significantly enhancing its interactions with the hydroxyl and carboxyl groups present in PLA (Chen *et al.,* 2019). At the same time, the non-polar fatty acid chains allow EPS to blend effectively with non-polar regions of the polymer. Thus, combining both polar and non-polar characteristics enables EPS to interact within the PLA matrix, weakening the existing polymer-polymer interactions and increasing molecular mobility. As a result, it can reduce intermolecular forces between polymer chains, allowing them to move freely (Awale *et al.,* 2018).

Thus, the results indicate that EPS acts as an effective plasticizer, particularly at higher concentrations, where it significantly enhances PLA ductility.

CONCLUSIONS

This study demonstrates that epoxidized palm stearin (EPS) is an effective bio-based plasticizer that enhances the flexibility of Polylactic acid (PLA). The incorporation of EPS significantly improved the chain mobility of the PLA matrix, as evidenced by a consistent reduction in the glass transition temperature (T_g) and cold crystallization temperature (T_{cc}) across all blends. Mechanically, the PLA/EPS20/St5 blend exhibited the highest elongation at break at 15.65%, nearly doubling the ductility of neat PLA.

Furthermore, the addition of 5 wt.% starch as a natural filler proved instrumental in reinforcing the PLA/EPS matrix. Starch served as a reinforcing agent, helping mitigate the loss of stiffness and impact strength typically associated with excessive plasticization. Specifically, the inclusion of starch led to a notable increase in impact strength across all EPS-containing blends compared with those without starch, effectively mitigating the softening effects of the plasticizer.

However, the study also reveals that while EPS enhances ductility, excessive loading (above 15%) can lead to a decline in tensile strength and Young's modulus due to the disruption of polymer-polymer interactions

and matrix softening. Ultimately, the combination of EPS and starch offers a balanced approach to tailoring the mechanical and thermal properties of PLA, overcoming its inherent brittleness. These findings support the use of sustainable, palm-based additives in the development of high-performance, environmentally friendly biopolymer blends for industrial applications.

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AUTHOR CONTRIBUTION

Each author contributed to the research idea and design of this work. Muhammad Nazmi Mohd Pauzi wrote the first draft of the manuscript and conducted the experimental work hands-on. Nur Azrini Ramlee and Rahimi Mohd Shariff provided and revised the analysis and discussion for the final manuscript. All authors contributed to and approved the final version.

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