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Performance Evaluation of Vegetable Oils as Novel Bio-Based Plasticizers in Polylactic Acid Films Meant for Food Packaging Applications

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Abstract

Polylactic acid (PLA) has garnered considerable attention as a food packaging material due to its biodegradable nature and significant potential as a sustainable alternative to petroleum-based plastics. However, its inherent brittleness and limited flexibility pose challenges to its widespread adoption. In the current study, vegetable oils were incorporated into PLA films as bio-plasticizers to reduce brittleness and improve flexibility. The PLA (5% (w/v) in chloroform) films were prepared using the solvent casting method. Sunflower oil, virgin coconut oil, and castor oil were used as bio-plasticizers at 0.05% (v/v) in the PLA film-forming solution, and PLA films without plasticizers and films containing glycerol, a conventional plasticizer, were used for comparison. The unplasticized PLA film exhibited brittle behaviour, with low elongation at break (EB) values of 1.17 ± 0.06% in the machine direction (MD) and 1.10 ± 0.10% in the cross direction (CD). Incorporation of virgin coconut oil (VCO) resulted in a 213% increase, or approximately a 3.13-fold improvement, in the %EB of PLA films. The Melting Enthalpy (ΔH_m) for native PLA films was 23.875 J/g, which increased in PLA film samples incorporated with vegetable oils. There was a significant increase in ΔH_m of film samples plasticized with castor oil (PLA-CO), indicating a well-packed crystalline structure and improved thermal stability. These findings highlight the potential of vegetable oils as sustainable plasticizers for PLA films, offering tailored improvements in flexibility and thermal stability. The study supports the development of eco-friendly packaging solutions, reducing reliance on conventional plasticizers with proven health risks.

Keywords: Polylactic acid (PLA), glycerol, sunflower oil, castor oil, virgin coconut oil

Introduction

Improper disposal of plastic waste leads to serious environmental issues such as soil contamination, water pollution, and air pollution. Environmental plastic waste does not readily biodegrade under natural conditions. Degradation of plastic in soil is a slow process that can take several decades to centuries to complete (Webb, Arnott, Crawford, & Ivanova, 2012). To mitigate this challenge, new bio-based polymers are being used to develop biodegradable packaging materials. Poly (lactic acid) or PLA, a popular aliphatic polyester in recent years, is produced by ring-opening polymerization (ROP) of lactic acid, which is obtained by fermenting starch-based sources. PLA is a biodegradable polymer that can be produced from renewable resources, including sugarcane, maize starch, etc., and the US Food and Drug Administration (FDA) has declared PLA to be generally recognized as safe (GRAS) (Öz, Süfer, & Sezer, 2017).

Polylactic acid is one of the most widely used biopolymers in the packaging industry and in material processing applications such as 3D printing, owing to its biodegradability and excellent processability (D'Amico et al., 2024). Depending on its stereochemistry and thermal history, PLA can have an amorphous or semi-crystalline structure in the solid state. PLA has a melting point of about 151 °C and a heat of fusion of about 21.5 J/g (Arrieta, López, Hernández, & Rayón, 2014; Ibrahim, Jollands, & Parthasarathy, 2017). Although PLA has several benefits, including high mechanical strength,

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transparency, compostability, and safety, its use is limited by its negative characteristics, which include high brittleness, low toughness, and inferior flexibility (Thuy, Duc, & Liem, 2018). Therefore, modification is required to enhance the mechanical properties of PLA. Consequently, several studies have been conducted to improve mechanical performance by the incorporation of plasticizers into polymer matrices (Ali, Awale, Fakhruldin, & Anuar, 2016).

Plasticization is a well-known technique used to reduce brittleness and enhance the softness and toughness of PLA and other brittle polymers. Plasticizers are a key group of low molecular weight, non-volatile additives extensively used in the polymer industry. Their main function is to enhance the flexibility and ease of processing of polymers by reducing the glass transition temperature (Tg), also known as the second-order transition temperature (Vieira, da Silva, dos Santos, & Beppu, 2011). Plasticizers are crucial for improving workability, flexibility, and durability of polymers, including PLA (Al-Mulla, Yunus, Bt Ibrahim, & Rahman, 2010). The incorporation of plasticizers in the manufacturing of plastic products is a well-established and long-standing practice. Plasticizers reduce intermolecular forces such as van der Waals interactions and hydrogen bonding between polymer chains, thereby increasing chain mobility, enhancing flexibility, and reducing brittleness. According to free volume theory, adding a plasticizer causes the polymer molecules to move more freely, increasing the polymer's free volume and imparting elastic properties and enhanced flexibility. However, conventional plasticizers are increasingly being scrutinized due to potential toxicity concerns, primarily associated with their tendency to migrate from the polymer matrix into the surrounding environment, food, or the human body. Moreover, most plasticizers are derived from petrochemical sources and are relatively expensive. Biobased plasticizers could be safer and cheaper alternatives to conventional plasticizers (Bocqué, Voirin, Lapinte, Caillol, & Robin, 2016).

In recent years, there has been growing interest in the use of natural plasticizers, which are valued for their low toxicity and minimal migration potential. This category includes epoxidized triglyceride vegetable oils derived from sources such as soybean oil, linseed oil, castor oil, and sunflower oil, as well as fatty acid esters (Baltacioðlu & Balköse, 1999). Vegetable oils are derived from plant sources and, due to their low cost, non-toxicity, biodegradability, and wide availability, can be effectively used as plasticizers in polymers (Garrison, Murawski, & Quirino, 2016). They exhibit low volatility, are generally colourless, and remain stable under light and heat, making them a suitable alternative to phthalate-based plasticizers (Darie-Niã et al., 2022). Castor oil, derived from the seeds of *Ricinus communis*, has been recognized for its medicinal properties since ancient times. Castor oil contains approximately 90% of a hydroxylated unsaturated fatty acid known as ricinoleic acid [(9Z, 12R)-12-hydroxyoctadec-9-enoic acid] (RA) (Kunduru, Basu, Zada, & Domb, 2015). Virgin coconut oil (VCO) is a vegetable oil rich in lauric acid, the primary saturated fatty acid in its composition. Several studies have explored the use of VCO as a plasticizer in bioplastic formulations due to its compatibility with polymer matrices and its ability to enhance flexibility (Arifin, Angelica, Nurhadi, Marta, & Nissa, 2025). Sunflower oil is another renewable and inexpensive resource, primarily composed of triacylglycerols (98–99%), with a minor fraction of phospholipids and other unsaponifiable matter, similar to most vegetable oils (D'Amico et al., 2024). With this background, the present study aims to investigate the effect of incorporating various vegetable oils such as sunflower oil, castor oil, and virgin coconut oil as plasticizers on the properties of PLA. Unlike previous studies that used higher plasticizer concentrations, this study demonstrates that even a very low concentration (0.05% v/v) of vegetable oils can effectively enhance the flexibility of PLA films, highlighting their efficiency and potential for more sustainable packaging applications (Sun, Weng, & Zhang, 2024).

Materials and Methods

A commercial grade PLA (Ingeo Biopolymer 4043D) from Nature Works LLC, USA, was used in pellet form as the base material for preparing the films. Chloroform (extra pure AR, 99.5%) was obtained from SRL Chem (Maharashtra, India). The different types of vegetable oils used as plasticizers, i.e., sunflower oil, castor oil, and virgin coconut oil, were purchased from the local market. The control plasticizer, glycerol, was procured from Himedia, India. All other chemicals used were of analytical grade.

The PLA films were prepared using the solvent casting method. A 5% (w/v) solution of PLA pellets

was prepared in chloroform, and the mixture was continuously stirred at room temperature until a completely homogeneous solution was obtained. Chloroform is a solvent that has a strong ability to dissolve PLA and produce uniform films. Although chloroform is a toxic solvent, it is expected to evaporate during the drying process, leaving minimal residual solvent in the final film. This method is widely accepted and used in PLA film preparation (Arrieta et al., 2014). A 0.05% (v/v) plasticizer was added to the PLA-chloroform film-forming solution along with continuous magnetic stirring until a homogeneous mixture was obtained. The plasticizer concentration (0.05% v/v) was selected based on prior preliminary trials and is supported by literature reports indicating that even low concentrations of plasticizers can improve polymer flexibility by enhancing chain mobility and reducing intermolecular interactions, while also minimizing phase separation and maintaining structural integrity (Vieira et al., 2011; Bocqué et al., 2016; Sun et al., 2024). After dissolution, the solution was cast into glass trays of size 26 × 26 cm. The films were dried at room temperature for 24 h and subsequently peeled off from the trays. The overall procedure of PLA film preparation by solvent casting is illustrated in Fig. 1. A total of four plasticizers (glycerol and three vegetable oils, i.e., sunflower oil, castor oil, and virgin coconut oil) were used in different batches of PLA films. The PLA films prepared without any plasticizer were kept as the control and designated as PLA-Control (P-C). The other four samples with plasticizers such as PLA-Glycerol, PLA-Sunflower oil, PLA-Castor oil, and PLA-Virgin coconut oil were labelled as P-G, P-SO, P-CO, and P-VCO, respectively.

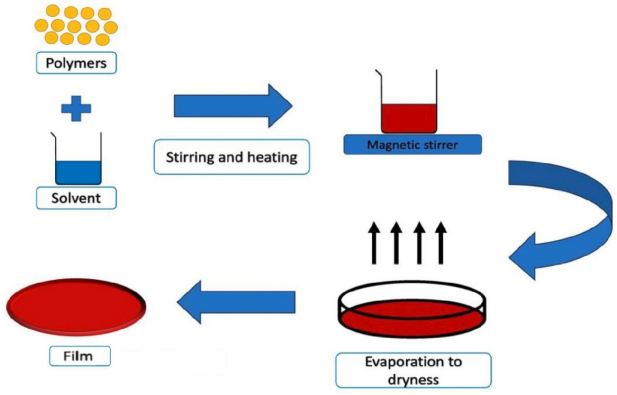

Fig. 1. Schematic representation of solvent casting method for PLA film preparation

The estimation of moisture was carried out using the oven drying technique (AOAC, 2023). The pH values were determined using a pH meter (I-Genelabserve, IG-10 pH). A 2 g sample was mixed with 10 mL of distilled water for pH estimation (Sanyang, Sapuan, Jawaid, Ishak, & Sahari, 2016).

Film specimens were placed on a white standard plate, and the colour parameters—lightness (L*) and chromaticity values a* (red–green) and b* (yellow–blue)—were measured using a colourimeter (ColorFlex EZ, HunterLab, USA). Three readings were taken from different areas of each film.

The thickness of the film was measured using a dial gauge (Mitutoyo, Japan), and the results were reported in μm. Measurements were taken at a minimum of ten random locations on each film, and the average value was calculated, which was also used for subsequent determination of tensile properties of the films.

Mechanical properties such as tensile strength, % elongation at break (ASTM D882-18), and heat seal strength (ASTM F88/F88M-15) of the PLA films were tested using standard ASTM methods employing a universal testing machine (TA Plus, Lloyd Instruments, UK) at 25 °C with a 1 kg load cell. Tensile strength was calculated by dividing the maximum load by the initial cross-sectional area of the film. Elongation at break was determined by dividing the increase in length at break by the original length of the film and multiplying by 100 (ASTM, 2018). The films were conditioned at 25 °C prior to testing. The sample width was 15 mm, with an initial grip separation of 50 mm, a testing speed of 50 mm/min, and a pre-load force of 1 N.

The thermal characteristics of the PLA blended films were analysed using a Differential Scanning Calorimeter (DSC 25, TA Instruments, USA). Film samples (approximately 10 mg) were sealed in aluminium pans. The analysis was conducted by heating the samples from 20 °C to 200 °C at a rate of 10 °C/min, followed by cooling from 200 °C to 20 °C at a rate of 1–3 °C/min under a nitrogen atmosphere. The glass transition temperature (T_g), melting temperature (T_m), and cold crystallization temperature (T_c) were recorded for each sample.

Statistical analysis was performed using SPSS software version 25.0 (SPSS Inc., Chicago, IL, USA). One-way analysis of variance (ANOVA), followed by Duncan's multiple range test was used to identify

significant differences ($p \leq 0.05$) among the properties of the PLA films. All values were expressed as mean ± standard deviation (SD) based on a minimum of three independent replicates.

Results and Discussion

The moisture content (%) of PLA films with and without plasticizers is presented in Table 1. In the present study, the moisture content of the PLA films ranged between 5.58% and 7.4%. The moisture content of PLA films varied significantly ($p \leq 0.05$) with the incorporation of different plasticizers. Among the samples, PLA-C (control, without plasticizer) showed the lowest moisture content of 5.58 ± 0.12%. Among the plasticized films, the PLA-G film had the highest moisture content of 6.74 ± 0.11%, indicating highest moisture uptake in glycerol-based PLA film. Berrío, Jiménez, and Montiel (2022) reported that the affinity of the films for water was directly related to the concentration of hydrophilic/hygroscopic plasticizers such as glycerol. The hydroxyl groups in glycerol have a strong affinity for water molecules, enabling glycerol-containing films to retain water within their matrix through hydrogen bonding. Thus, glycerol functions as a water-holding agent (Sanyang et al., 2016). According to Holm, Ndoni, and Risbo (2006), increases in humidity and temperature significantly affect the rate of moisture sorption in PLA, leading to higher moisture content over time and further influencing its performance. The incorporation of oil imparts hydrophobicity to the films, resulting in lower moisture content compared to PLA-G films. PLA-CO films had the lowest moisture content among the oil-based formulations, and there were no significant ($p \geq 0.05$) differences in the moisture content of PLA-CO (6.12 ± 0.07%) and PLA-SO (6.14 ± 0.02%) samples. The virgin coconut oil-incorporated PLA films showed slightly higher moisture content (6.46 ± 0.02%) than PLA-SO and PLA-CO samples, reflecting subtle differences in polarity and molecular interaction with the PLA matrix. The hydrophobic nature of these plasticizers is advantageous, as it enhances water resistance, reduces moisture permeability, and contributes to improved mechanical strength, making them beneficial for food packaging applications (Bolat, Necolau, Bîru, Zaharia, & Iovu, 2024).

Table 1. Moisture content and pH of PLA films

Name of the Film	Moisture (%)	pH
PLA-C	5.58 ± 0.05^{a}	4.17 ± 0.01^{a}
PLA-G	6.74 ± 0.11^{d}	4.26 ± 0.02^{b}
PLA-SO	6.14 ± 0.02^{b}	4.60 ± 0.02^{c}
PLA-CO	6.12 ± 0.07^{b}	4.83 ± 0.03^{e}
PLA-VCO	6.46 ± 0.02^{c}	4.70 ± 0.02^{d}

*PLA: Poly lactic acid, C: Control, G: Glycerol, SO: Sunflower oil, CO: Castor oil, VCO: Virgin coconut oil.
**The results are expressed as mean ± SD. The values in the same column with different superscripts are significantly different.

The pH values of the PLA films in this experiment ranged between 4.17 and 4.83, as presented in Table 1. Among the tested films, PLA-C (4.17 ± 0.01) and PLA-G (4.26 ± 0.02) exhibited the lowest pH values, indicating more acidic characteristics. PLA-SO (4.60 ± 0.02) and PLA-VCO (4.70 ± 0.02) displayed moderately higher pH values, though remaining within the acidic range. The PLA-CO film showed the highest pH (4.83 ± 0.03), indicating comparatively lower acidity. Glycerol, being more hydrophilic, increased the moisture content, which contributes to lower pH values, whereas vegetable oils, due to their higher hydrophobicity, reduced the moisture content and thereby result in comparatively higher pH values of the PLA films.

The colour values of each sample are presented in Table 2. Among all the samples, the PLA-G sample exhibited the highest lightness (L* = 34.92 ± 0.98), indicating a significantly ($p \leq 0.05$) brighter appearance compared to the control (L* = 7.89 ± 1.57) and other plasticized samples. In contrast, the PLA-SO, PLA-CO, and PLA-VCO showed lower L* values (16.30 ± 1.30 to 18.30 ± 0.78), without any significant differences ($p \geq 0.05$), reflecting their darker appearance. The a* values across all films remained negative, indicating a consistent slight greenish tint regardless of the plasticizer type. The PLA-CO film sample had a significantly higher negative a* value (-0.70 ± 0.05) in comparison to the control (-0.47 ± 0.16). However, the b* values (yellow-blue axis) of PLA film samples showed more significant ($p \leq 0.05$) variation. Among them, the PLA-CO sample displayed the most intense bluish tint (b* = –4.03 ± 0.14), followed by sunflower and coconut oil-based films. The addition of glycerol significantly ($p \leq 0.05$) increased the transparency and brightness of the film, likely due to its hydrophilic nature, enhancing dispersion and

Table 2. Colour parameters and thickness of PLA films

Film	L^*	a^*	b^*	Thickness (μm)
PLA-C	7.89 ± 1.57^a	-0.47 ± 0.16^a	-2.68 ± 0.15^b	107.7 ± 6.8^a
PLA-G	34.92 ± 0.98^c	-0.48 ± 0.02^a	-1.73 ± 0.12^a	104.7 ± 9.8^a
PLA-SO	18.30 ± 0.78^b	-0.61 ± 0.03^{ab}	-3.29 ± 0.05^b	104.6 ± 7.9^a
PLA-CO	17.13 ± 0.20^b	-0.70 ± 0.05^b	-4.03 ± 0.14^c	109.4 ± 9.9^a
PLA-VCO	16.30 ± 1.30^b	-0.59 ± 0.04^a	-3.17 ± 0.10^b	111.7 ± 14.4^a

*PLA: Poly lactic acid, C: Control, G: Glycerol, SO: Sunflower oil, CO: Castor oil, VCO: Virgin coconut oil.
**The results are expressed as mean ± SD. The values in the same column with different superscripts are significantly different.

light transmission. A previous study observed that increasing glycerol concentration improved the transparency of biodegradable films based on sugar palm (*Arenga pinnata*) starch (Sanyang et al., 2016). On the other hand, the incorporation of vegetable oils led to reduced lightness and enhanced bluish tones, possibly due to oil-induced phase separation or pigment retention during film formation. It was reported that the overall appearance of cress seed gum films became lighter with the incorporation of glycerol, indicating that addition of a plasticizer can enhance the optical properties of films (Jouki, Khazaei, Ghasemlou, & HadiNezhad, 2013). Castor oil is distinguished from other vegetable oils by its exceptionally high content of ricinoleic acid (RA) (Kunduru et al., 2015). Notably, PLA-CO films showed the strongest bluish shift, which may be attributed to the high RA content of castor oil influencing polymer interaction and surface properties. Although all the films appeared visually transparent, instrumental colour analysis demonstrated that the type of plasticizer had a significant impact on both the lightness and chromatic attributes of the PLA films.

Thickness is a crucial characteristic of biodegradable films, as it influences key packaging properties such as tensile strength, elongation at break, and permeability to water vapour and gases. In a previous study, PLA films incorporated with green tea and rosemary polyphenolic extracts were developed as active packaging for almonds and beef. The monolayer films, prepared using a Periplast single-screw extruder, had a thickness in the range of 50–60 μm (Andrade et al., 2023). In another study by Carbonell-Verdu et al. (2017), films with an average thickness of 100 μm were prepared using a cast-roll machine from plasticized PLA formulations incorporating maleinized cottonseed oil and maleinized linseed oil to enhance flexibility. In the present case, the thickness values of the five film samples were measured, and the values are shown in Table 2. The data revealed that although there were variations in the thickness across different films, these differences were not statistically significant ($p \geq 0.05$), mainly because the amount of film-forming solution poured onto the plate was adjusted to maintain a constant amount of solid for each formulation. PLA-VCO films showed the highest thickness value (111.7 ± 14.4 μm), followed by PLA-CO films (109.4 ± 9.9 μm). The higher standard deviation observed for the PLA-VCO films (111.7 ± 14.4 μm) compared to others may be attributed to minor variations in film thickness, possibly caused by non-uniform spreading during the solvent casting process and differences in oil dispersion within the polymer matrix. The control PLA film exhibited an intermediate value (107.7 ± 6.8 μm). In contrast, PLA-G (104.7 ± 9.8 μm) and PLA-SO (104.6 ± 7.9 μm) films showed the lowest thickness values among the samples. A previous study by Berrío et al. (2022) demonstrated that increasing the concentration of glycerol in chitosan/PVA films led to an increase in film thickness, attributed to glycerol's hygroscopic nature and its ability to enhance polymer chain mobility. Conversely, the incorporation of sunflower oil reduced the thickness, likely due to its influence on the polymer matrix composition, resulting in reduced crystallinity and thinner films. Furthermore, a study revealed that an increase in the thickness of PLA films often correlates with enhanced mechanical strength, which is advantageous in various sectors such as packaging and 3D printing applications (Cho, Hein, Lynn, Hla, & Tran, 2019).

Mechanical properties such as tensile strength at break (TS), elongation at break (EB) (%), and heat

Table 3. Mechanical properties of PLA films

Property	PLA-C	PLA-G	PLA-SO	PLA-CO	PLA-VCO
TS-MD (kgf)	6.71 ± 0.16^{d}	5.74 ± 0.19^{bc}	3.79 ± 0.70^{a}	5.38 ± 0.77^{b}	6.58 ± 0.21^{cd}
TS-CD (kgf)	6.35 ± 0.06^{b}	5.06 ± 0.29^{a}	4.89 ± 0.35^{a}	6.63 ± 0.69^{b}	6.07 ± 0.19^{b}
EB-MD (%)	1.17 ± 0.06^{a}	3.25 ± 0.20^{c}	2.37 ± 0.74^{b}	1.24 ± 0.12^{a}	3.66 ± 0.15^{c}
EB-CD (%)	1.10 ± 0.10^{a}	3.31 ± 0.16^{c}	1.92 ± 0.25^{b}	1.56 ± 0.22^{b}	3.16 ± 0.36^{c}
HSS-MD (kgf)	2.50 ± 0.30^{bc}	0.94 ± 0.27^{a}	1.97 ± 0.51^{ab}	1.35 ± 0.73^{ab}	3.33 ± 1.86^{c}
HSS-CD (kgf)	2.30 ± 0.25^{c}	1.24 ± 0.52^{b}	1.33 ± 0.57^{b}	0.89 ± 0.23^{ab}	0.48 ± 0.36^{a}

*PLA: Poly lactic acid, C: Control, G: Glycerol, SO: Sunflower oil, CO: Castor oil, VCO: Virgin coconut oil, TS: Tensile strength, EB: Elongation at break, HSS: Heat seal strength, MD: Machine direction, CD: Cross direction.

**The results are expressed as mean ± SD. The values in the same row with different superscripts are significantly different.

seal strength (HSS) are essential properties of a food packaging film. Tensile strength at break indicates the maximum stress a material can withstand before it permanently deforms or breaks, while elongation at break indicates the flexibility of a film and serves as an inverse measure of its brittleness. Neat PLA reportedly exhibits the typical mechanical behaviour of a brittle polymer, characterized by a low %EB value. In the present study, as shown in Table 3, the unplasticized PLA film demonstrated brittle behaviour, with low EB value of 1.17 ± 0.06 % in the machine direction (MD) and 1.10 ± 0.10 % in the cross direction (CD). In contrast, the PLA-VCO film showed the highest EB value (3.66 ± 0.15% in MD), indicating enhanced flexibility. There was a 213% increase, or approximately a 3.13-fold increase, in the %EB of PLA films upon incorporation of VCO as plasticizer. Darie-Niã et al. (2023) reported a significant increase in elongation at break of PLA-based biocomposites with the incorporation of coconut oil, supporting the enhanced flexibility observed in the present study. There was a significant improvement in the mechanical properties of PLA-based biocomposites with the incorporation of 15 wt% coconut oil (CO), leading to an 11-fold increase in EB compared to the neat PLA matrix. The PLA-G film also showed a greater %EB, with increases of 178% or 2.78-fold increase in MD and 201% or 3.01-fold increase in CD, and there was no significant difference in the EB values of PLA-VCO and PLA-G. The plasticizer, glycerol, reportedly alters the structure of the films by increasing the mobility of the polymer chains, reducing the van der Waals intermolecular forces between polymers, resulting in greater flexibility (Abraham, Soloman, & Rejini, 2016). The PLA-SO film sample exhibited moderate EB values, suggesting improved flexibility compared to the native PLA. The %EB values of PLA-CO indicated limited improvement in flexibility, compared to other vegetable oils. There were no significant ($p \leq 0.05$) differences in the EB values of PLA-CO and PLA-C in MD. In contrast, Darie-Niã et al. (2022) reported that the incorporation of natural castor oil (R) into PLA significantly enhanced its ductility, with elongation at break increasing approximately 14-fold, from 4.7% for neat PLA to 67% and 70% for PLA containing 15 wt% and 20 wt% R, respectively.

This enhancement in the flexibility of PLA films added with plasticizers was associated with a significant ($p \leq 0.05$) decrease in tensile strength at break, except for PLA-VCO. Incorporating oils into a film matrix typically reduces tensile strength (TS) but enhances elongation at break (EB) due to their plasticizing effect. Oils weaken strong polymer–polymer interactions by introducing weaker polymer–oil interactions, leading to structural discontinuities and reduced cohesion, thereby disrupting crystallinity and lowering mechanical strength (Lee, Lee, & Han, 2020). The tensile strength (TS) at break values of PLA films ranged between 3.79 ± 0.70 kgf and 6.71 ± 0.16 kgf in MD. As anticipated, the highest TS value in MD was recorded for unplasticized PLA films, and the lowest was for PLA-SO films. In line with these findings, Lee et al. (2020) reported that the incorporation of sunflower seed oil into mung bean starch-based films reduced tensile strength but enhanced water resistance. Among all the plasticized film samples, the PLA-VCO films displayed the highest %EB without any significant loss in TS.

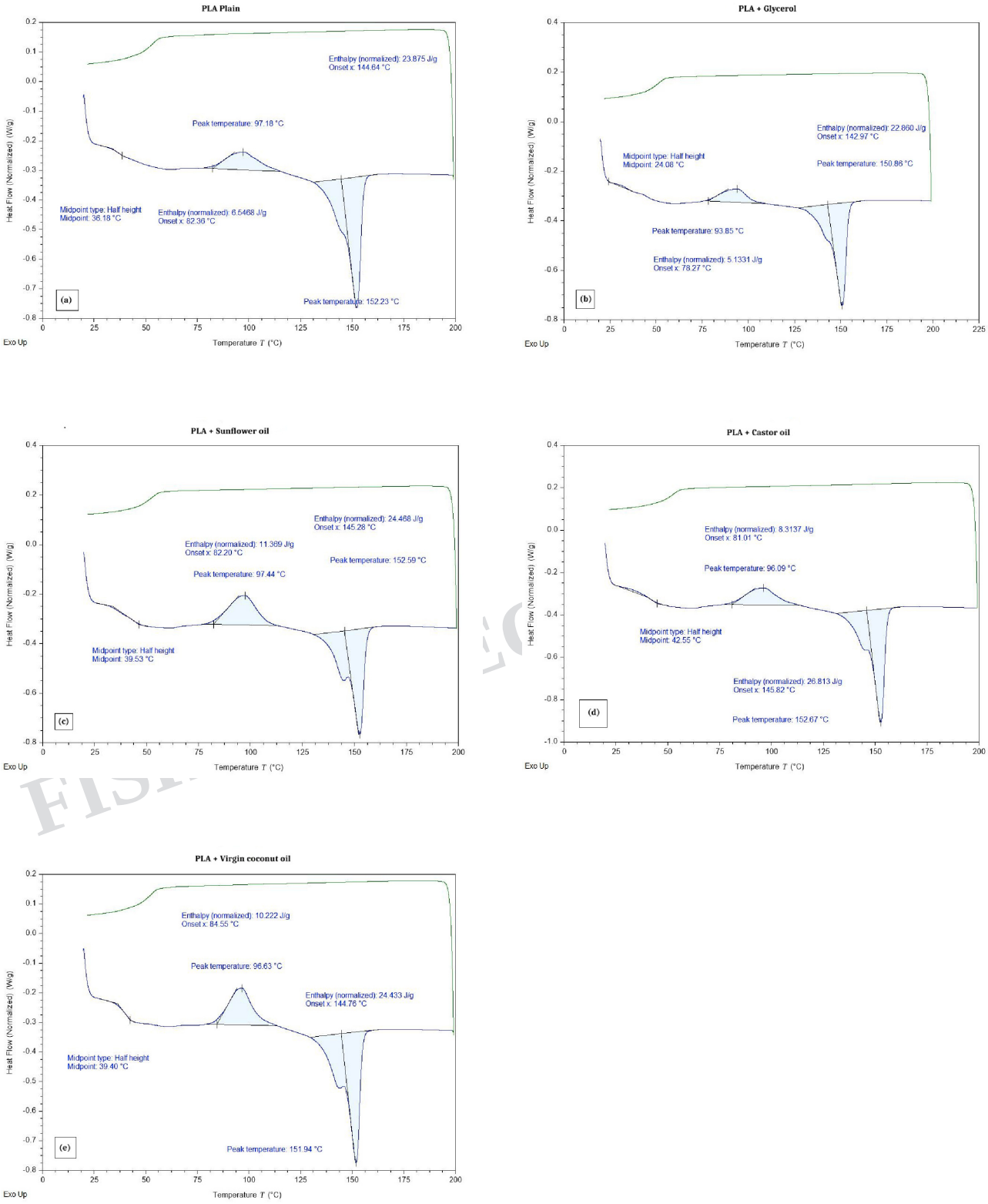

Fig. 2. DSC thermograms of PLA films: (a) PLA-Control (PLA-C), (b) PLA-Glycerol (PLA-G), (c) PLA-Sunflower oil (PLA-SO), (d) PLA-Castor oil (PLA-CO), (e) PLA-Virgin coconut oil (PLA-VCO).

There were no significant ($p \geq 0.05$) differences in the TS values of PLA-C and PLA-VCO in both MD and CD. In contrast, a previous study reported that coconut oil (7 wt %), as a non-toxic plasticizer, enhanced the flexibility of PLA films from 12% to 54%, which indicates a 350% or 4.5-fold increase in %EB in comparison to control but decreased the TS from 60 to 41 MPa (Bhasney, Patwa, Kumar, & Katiyar, 2017). Similarly, Xiao et al. (2022) reported that the incorporation of virgin coconut oil into a konjac glucomannan/agar/gum Arabic film matrix decreased tensile strength while enhancing flexibility, as indicated by increased elongation at break. The heat seal strength value of PLA films was 2.50 ± 0.30 kgf for the control sample in MD, which decreased significantly ($p \leq 0.05$) in all other plasticized film samples except PLA-VCO (3.33 ± 1.86 kgf).

The thermal transition and crystallization behaviour of PLA films were assessed using DSC analysis. The DSC thermograms of the PLA film samples are given in Fig. 2. The cold crystallization temperature (T_c) of pure PLA film was 97.18 °C, which decreased slightly in all film samples except PLA-SO. The addition of vegetable oils enhanced chain mobility, as evidenced by the decrease in crystallization temperature (T_c), indicating that the oils facilitated earlier crystallization of PLA (Bhasney et al., 2017). The melting Enthalpy (ΔH_m) for native PLA films was 23.875 J/g, which decreased in the PLA-G film sample, suggesting a lower energy requirement for melting the crystalline zones in PLA films plasticized with glycerol. In all other film samples, ΔH_m increased, and a significant increase was observed in PLA film samples plasticized with castor oil (PLA-CO), indicating a well-packed crystalline structure and improved thermal stability. The melting temperature (T_m) was 152.25 °C, which increased in PLA-SO and PLA-CO samples, and decreased in PLA-G and PLA-VCO samples. The considerable decrease in the T_m of PLA-VCO (144.76 °C) indicates a strong plasticizing effect. Bhasney et al. (2017) reported that the addition of coconut oil resulted in only a minor decrease in T_m of PLA films, indicating that the melting temperature of PLA is not significantly affected by the incorporation of coconut oil.

Polylactic acid, a thermoplastic polyester, is widely recognized for its excellent strength and biodegradability. However, its intrinsic brittleness and low elongation at break limit its broader practical applications. The incorporation of vegetable oils as bio-based plasticizers in PLA films can modify the physical, mechanical, and thermal properties, making them more suitable for biodegradable packaging applications. Sunflower oil, castor oil, and virgin coconut oil demonstrated distinct effects on film performance compared to the conventional plasticizer glycerol. Overall, the use of vegetable oils enables the development of PLA films with specific qualities while providing a sustainable substitute for petroleum-derived plasticizers. This strategy supports the creation of environmentally friendly packaging materials and contributes to global initiatives aimed at reducing plastic waste and promoting sustainable packaging. The present study focused on the mechanical, thermal, and physicochemical properties of PLA films. However, analyses such as water permeability, oxygen permeability, and morphological characterization (SEM) were not included and can be considered in future studies for a more comprehensive evaluation.

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