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A Study Review on Mechanical Properties of Polylactic Acid (PLA) with Plasticizer Vegetable Based and Composite Material

Ernie Suryati Jefri[a], Mohd Hafidzal Mohd Hanafi[a,*], Nurul Hanim Razak[a], Muhammad Zaidan Abdul Manaf[a], Fahmi Asyadi Md Yusof[b], Syazwan Hanani Meriam Suhaimy[c], Grasianto[d]

[a]Centre for Advanced Research on Energy (CARe), Faculty of Mechanical Engineering and Technnology, Universiti Teknikal Malaysia Melaka (UTeM), Hang Tuah Jaya, 76100 Durian Tunggal, Melaka, Malaysia
[b]Universiti Kuala Lumpur, Malaysian Institute Of Chemical And Bioengineering Technology, Universiti Kuala Lumpur, 78000, Alor Gajah, Melaka, Malaysia
[c]Department of Physics and Chemistry, Faculty of Applied Sciences and Technology, Universiti Tun Hussein Onn Malaysia, Johor, Malaysia
[d]Department of Chemistry, Faculty of Science and Data Analytics, Institut Teknologi Sepuluh Nopember (ITS), Surabaya, 60111, Indonesia
hafidzal@utem.edu.my

Polylactic Acid (PLA) has received considerable attention worldwide due to its ability to act as biodegradable material that has a lot of similarity towards petrochemically derived products. However, PLA still experience drawbacks such as high brittleness and low toughness that require further research. These limitations can be addressed through plasticization, polymer blending and crosslinking mechanism. The incorporation of vegetable oil-based plasticizer in PLA has been found to enhance its mechanical properties due to high lubricity, high index viscosity and good solvency characteristic. These contributes to increase in ductility, tensile strength, stability and toughness. In parallel, addition of composite materials further strengthens PLA by redistributing stress from polymer matrix to reinforcing phases and improving interfacial adhesion. This review emphasis on the effect of plasticizer and composite material in modifying PLA mechanical properties focusing on tensile strength, flexural strength and elongation at break, while other aspects are reserved for future studies. The aim is to advance PLA as a sustainable material with performance comparable to petrochemically derived product. Plasticizer that being discussed in this review are epoxidized jatropha oil (EJO), epoxidized chia seed oil (ECO), epoxidized soybean oil (ESO), epoxidized palm oil (EPO) and epoxidized waste cooking oil (EWCO). Meanwhile, composite material reviewed are kenaf fibre, zinc oxide nanoparticles (ZnO NPs), and 3-aminophenylboronic acid (APBA). The results showed that EPO promotes stronger hydrogen bonding within the PLA matrix due to higher oxirane oxygen content, thereby increase the tensile strength efficiently. The study also revealed that APBA helps to form borate ester linkage through interaction between boric acid groups and hydroxyl groups in PLA, that contributes to higher elongation at break. ZnO NPs additions introduce new functional properties which is antibacterial traits, while increasing tensile strength. The ability of epoxy group in EWCO to insert themselves between PLA chains helps to reduce van der Waals and hydrogen bonding, that leads to increase in flexural strength. This paper present findings on the effect of vegetable oil-based plasticizer and composite material towards PLA mechanical performance for wider applications as a sustainable product.

1. Introduction

The global escalation of plastic waste, particularly from single-use items such as bottle caps, straw and plastic bottles has sparked widespread environmental concern. Traditional polymers like polyethylene (PE), polypropylene (PP) and polyethylene terephthalate (PET) are derived from petrochemicals and non-biodegradable materials, which leads to long-term ecological damage and waste management challenges. The

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growing accumulation of plastic waste poses a serious global challenge, affecting ecosystems, public health, and economic stability. As such, there is growing urgency to shift towards sustainable alternatives that can reduce environmental impact and reliance on fossil resources, which has propelled research into biodegradable polymers as a viable solution. Among biodegradable polymers, polylactic acid (PLA) has gained significant attention due to its favourable biodegradability, mechanical strength and biocompatibility (Xie et al., 2022). It is derived from renewable sources such as corn, cassava, sugarcane and sugar beet pulp which promotes sustainability and support growth in agricultural sector. However, PLA suffers from intrinsic brittleness, low elongation at break, low toughness and low tensile elongation (Giita Silverajah et al., 2012). These limitations stem from PLA's rigid backbone and strong intermolecular forces, which restrict chain mobility and hinder its use in flexible, impact-resistant application. These drawbacks are influenced by moisture, which degrades the mechanical properties of PLA by breaking down its polymer chains and reducing the intermolecular forces, thereby lowering the material's resistance to stress-induced deformation. PLA is also affected by temperature, which decreases its molecular weight and mechanical properties. At low temperatures it behaves in a brittle manner, while at higher temperatures its stress–strain response resembles that of thermoplastics, showing reduced ultimate strength but increased elongation at break (Hedayati et al., 2024). However, previous studies have largely overlooked the influence of different plasticizer types on the mechanical performance of PLA composites, particularly concerning their compatibility and long-term stability. This gap hinders the development of optimized, sustainable PLA composites.

Despite that, PLA continues to show great potential as numerous researches has been conducted to address its limitations by using various mechanism such as plasticization, polymer blending and crosslinking. Plasticization occurs when plasticizer molecules diffuse into the PLA matrix, reducing intermolecular forces and friction. This enhancement of free volume and chain mobility allows the polymer chains to move more freely, leading to increase in flexibility, elongation at break and ductility. It often interpreted through three dominant models of plasticization as shown in Figure 1b. Then, polymer blending involves physically combining two or more polymers to produce new materials. Using melt or solution blending, the process often results in a two-phase or partially miscible system, governed by thermodynamic and kinetic factors. In such blends, second polymer serves as a soft, flexible domain within the brittle PLA matrix, thereby increasing PLA toughness, impact resistance and elongation at break. For crosslinking mechanism, it involves physical links between two or more molecules to create new network structure with increased stability, improved heat resistance and strength. The mechanism is different based on the type of bonds formed where chemical reactions results in covalent bond and physical interaction results in hydrogen bonds. Covalent bonds form stable chemical compounds while hydrogen bonds form weaker electrostatic forces between hydrogen atom and electronegative atom.

In the present study, modified vegetable oils received positive attention as a renewable source of plasticizer for polymers and PLA composites due to their potential as an additive. It possessed high lubricity, high index viscosity and good solvency, which reported can enhance PLA mechanical properties (Kamarudin et al., 2018). High lubricity characteristic found to weaken intermolecular friction between PLA chains, thereby increasing ductility, elongation at break and overall flexibility. This effect must be observed closely as excessive lubricity leads to weaker interfacial bonding which affects PLA tensile strength and modulus. Then, high index viscosity in vegetable oils provides stability across wide temperature range. This contributes to consistent processing behaviour when incorporated into PLA, ensuring better performance at elevated temperatures. Additionally, vegetable oil-based plasticizers exhibit good solvency that promote strong compatibility with PLA matrix. This decreases phase separation and improve stress transfer within the material, resulting to increase in toughness and flexibility.

Besides, incorporating composite materials such as natural fibres, nanofillers and inorganic particles have also been shown to significantly enhance PLA mechanical properties through several reinforcement mechanisms. One of the main mechanisms used is stress transfer where the external load is redistributed from weaker PLA matrix to the stronger reinforcing phase, leading to improved tensile and flexural strength (Kamarudin et al., 2018). Another important factor is interfacial bonding between filler and polymer that act as strong adhesion. It can restrict slippage which resulted in increased stiffness and structure stability. In addition, the development of filler networks within PLA matrix acts as barriers to crack propagation, which increase the toughness, ductility and impact resistance (Xie et al., 2022).

Moreover, the generous availability of modified vegetable oils around the world such as jatropha, chia seed, soybean and palm oil make it accessible and possibly cheaper to be a plasticizer. Also, it promotes better contributions towards the environment and is more sustainable. For composite materials, it can be derived from different sources. The natural options include kenaf, hemp and jute, while synthetic and inorganic origin obtained from nano clays, zinc oxide and carbon-based nanofillers. Each type offers different properties which benefits PLA performance. As shown in Figure 1a, plasticizers are categorized into external and internal where external is a stable compound that physically integrated into the polymer matrix, constituting the prevailing method of plasticization. In addition, external plasticizers categorized into primary have strong interaction with polymer

matrix, while secondary can increase viscosity and surface lubricity. Then, for secondary external plasticizers, extenders can be used to partially replace primary plasticizers which are typically expensive. Meanwhile, internal plasticization relies on monomers copolymerized with polymer to reduce the glass transition temperature (Tg) (Sun et al., 2024). Figure 1b shows the major classification of plasticizers and three dominant theoretical models of plasticization which are lubrication theory that refers to the ability of a substance to reduce friction and wear between contacting surface during motion. Then, gel theory explains the polymer system transforms from liquid to gel state through the formation of cross-linked network. For free-volume theory, it describes the physical behaviour of polymers that is influenced by the unoccupied space between molecular chain. Therefore, these classifications and models emphasis the role of plasticizer and composite material in modifying polymer behaviour in PLA, where it influences the mechanical properties. Thus, the purpose of this review paper is to discuss the effects of vegetable oil-based plasticizer and composite material towards the mechanical properties of PLA that focused on tensile strength, elongation at break and flexural strength, while other aspects are reserved for future studies. The novelty of this review lies in its dual focus on vegetable oil-based plasticizers and composite materials, presenting a comparative analysis across different bio-based modifiers. Unlike prior reviews, this work integrates recent findings (2021–2024), classifies plasticizers based on their interaction mechanisms and mechanical outcomes against specific additive types. This aims to support both academic researchers and industry practitioners in developing high-performance, sustainable PLA materials.

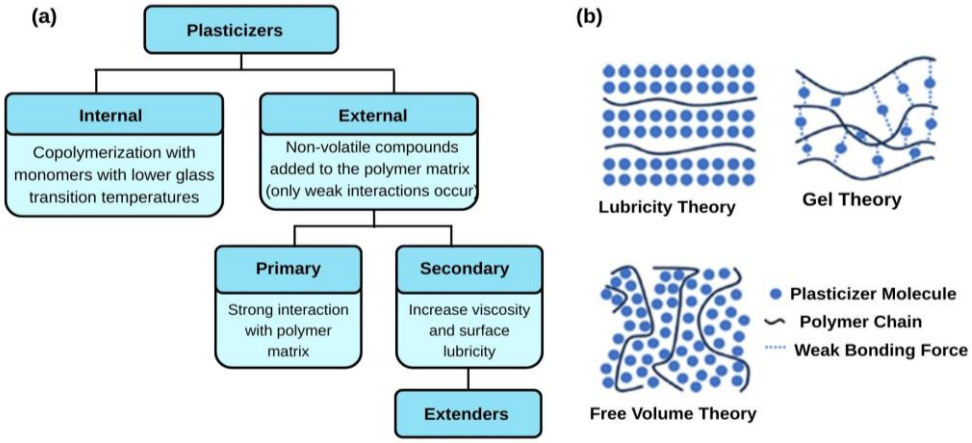

Figure 1: The major classification of plasticizers (a) and three dominant theoretical models of plasticization (b). Adapted from (Sun et al., 2024)

2. Mechanical Properties

Studies combining PLA with vegetable oil-based plasticizer and composite materials that been made throughout the years presents results as shown in Table 1 below that depicts the mechanical properties of PLA from literature. There are five types of vegetable oils that been reviewed, which are Epoxidized Jatropha Oil (EJO), Epoxidized Chia Seed Oil (ECO), Epoxidized Soybean Oil (ESO), Epoxidized Palm Oil (EPO), and Epoxidized Waste Cooking Oil (EWCO). In addition, the composite materials discussed are Kenaf Fibre (KFT), Zinc Oxide Nanoparticles (ZnO NPs) and 3-aminophenylboronic acid (APBA), where it can be categorized based on their origin and structural characteristics. KFT is a natural fibre originates from renewable plant sources that can improve tensile strength and reduce environmental impact. Then, ZnO NPs classified as inorganic nanofiller which promotes thermal stability and antibacterial properties, while APBA belongs to organic small-molecule modifier that contains boronic acid group that forms strong interfacial bonding with PLA through hydrogen bonding. There are various types of PLA used in these reviewed studies, including grade of 2003D, Ingeo 4043D, 4042D, 4032D, 2 % D-Lactide and 3001D. The choice of PLA influenced the results of the research since it contains different molecular weight, stereoisomer composition and additives that applicable for different applications such as packaging, 3D printing, biomedical applications and sustainable products. The molecular weight determines the length of polymer chains in PLA, where increased weight contains more chain entanglements that increase tensile strength, toughness and thermal resistance of PLA. Then, stereoisomer composition is the ratio of L-lactide and D-lactide from lactic acid that controls the crystallinity of PLA. By increasing L-lactide content, the crystallinity of PLA also increases which leads to increased stiffness, thermal stability and slower degradation. Meanwhile, increasing D-lactide will decrease the crystallinity and makes PLA becoming more amorphous resulting in increased flexibility and transparency but lower heat resistance.

Additionally, certain grades of PLA contain additives such as nucleating agents, plasticizers and stabilizers which are tailored to fulfil specific properties. The example includes impact modifiers and flow enhancers that used for packaging and 3D printings. For biomedical and consumer applications, it is important to choose suitable grades of PLA as the degradation products of PLA are known to reduce local pH, accelerate degradation and induce inflammatory reactions (Gorrasi and Pantani, 2013).
Moreover, the blending ratio with PLA for plasticizers is around 1 wt% to 10 wt%, while composite material is between 3 wt% to 40 wt%. Table 1 shows the mechanical properties of tensile strength, elongation at break and flexural strength. All the literature review did not undergo the same types of experimental test due to different priorities by the researchers. Thus, resulting in inconsistent data of the mechanical properties summarized in Table 1.

Table 1: Mechanical properties for EJO/KFT, ECO, ESO/ZnO, EPO, ESO/APBA, and EWCO with PLA

Authors	PLA Grade	Vegetable Based Plasticizer	Composite Material	Blending Ratio (wt%)	Sample Characterization		
					Tensile Strength (MPa)	Elongation at Break (%)	Flexural Strength (MPa)
(Kamarudin et al., 2018)	2003D	Epoxidized Jatropha Oil (EJO)	Kenaf Fibre (KFT)	PLA (57) EJO (3) KFT (40)	36	-	-
		Standard Method			ASTM D638-10		
(Dominguez-candela et al., 2021)	2003D	Epoxidized Chia Seed Oil (ECO)	-	PLA (92.5) ECO (7.5)	35	56	-
		Standard Method			ISO 527		
(Ghoroghi et al., 2024)	Ingeo 4043D	Epoxidized Soybean Oil (ESO)	Zinc Oxide Nanoparticles (ZnO NPs)	PLA (85) ESO (10) ZnO (5)	36	33-37	-
		Standard Method			ASTM D882-12		
(Giita Silverajah et al., 2012)	4042D	Epoxidized Palm Oil (EPO)	-	PLA (99) EPO (1)	60	-	-
		Standard Method			ASTM D638		
(Xie et al., 2022)	4032D, 2 % D-Lactide	Epoxidized Soybean Oil (ESO)	3-aminophenylboronic acid (APBA)	PLA (87) ESO (10) APBA (3)	40-43	130-190	-
		Standard Method			ASTM D638		
(Omar et al., 2023)	3001D	Epoxidized Waste Cooking Oil (EWCO)	-	PLA (97.5 EWCO (2.5)	-	-	6.7

The tensile strength for PLA blends was evaluated following standard testing methods established by American Society for Testing and Materials (ASTM) and the International Organization for Standardization (ISO). The PLA/EJO/KFT blends followed ASTM D638-10 and both PLA/EPO, PLA/ESO/APBA were tested according to ASTM D638. For PLA/ESO/ZnO, the test conducted using ASTM D882-12, and PLA/ECO comply with ISO 527. However, the standard used for PLA/EWCO blends is not specified as the tensile test is not being done on the research. The results obtained from reviewed research shows that PLA/ESO/APBA exhibits higher tensile strength compared to PLA/EJO/KFT and PLA/ESO/ZnO, with values of 43 MPa versus 36 MPa. Then, PLA/EPO posed significantly higher tensile strength than PLA/ECO with values of 60 MPa versus 35 MPa. The tensile strength is influenced by the presence of epoxy group in vegetable-based plasticizer that interacts with hydroxyl groups present in PLA, this interaction decreases the intermolecular forces between the atoms which consequently increase its ductile property (Dominguez-candela et al., 2021). Epoxy group in the vegetable oil-based plasticizer is highly reactive as it can undergo ring-opening reactions with nucleophiles such as hydroxyl group. Its interaction with PLA matrix resulted to the decrease in stiffness, which makes polymer chains become more mobile and flexible that makes it capable of plastic deformation under stress, thereby increase the ductility

and elongation at break. Besides, presence of composite material APBA in PLA/ESO blends exhibits higher tensile strength compared to PLA/ESO/ZnO. This enhancement is attributed to the chemical interactions between boric acid groups in APBA and hydroxyl groups present in PLA, which increase the yield strength of polymer. These interactions facilitate the formation of borate ester linkages, that act as crosslinking points within the polymer matrix. The resulting network structure increases rigidity and resistance of the material to deformation. Thus, the weakness of marked deterioration of mechanical properties during PLA melting process in the presence of amino group-containing compounds can be overcome (Xie et al., 2022). Also, addition of APBA plays a significant role in the mechanical properties of the blends, where it improves the tensile strength by influencing the crystallization behavior of the polymer. The borate ester linkage in the blend acts as a nucleating agent to enhance the formation of crystalline region that leads to increase in tensile strength. Besides, higher ZnO content provides increased tensile modulus and antibacterial properties towards the nanocomposites (Ghoroghi et al., 2024). In contrast, PLA/ESO/ZnO blends did not exhibit the same level of enhancement in tensile test compared to PLA/ESO/APBA blend. This is because ZnO nanoparticles are less effective at forming crosslinking network within PLA matrix due to lack of boric acid groups that are available in APBA. Moreover, Table 1 indicates that the highest tensile strength between the research reviewed is from PLA/EPO blends. According to the researchers, the optimum EPO loading that can enhanced mechanical and thermal properties of PLA is 1 wt% (Giita Silverajah et al., 2012). Based on the result obtained, the statement is proven since the tensile strength of PLA/EPO blends shows significantly higher results between this reviewed research. This slightly higher tensile strength contributed by the interaction between polymer-plasticizer that contains higher oxirane oxygen content (OOC) from the EPO, which indicates the epoxy group present in the plasticizer (Giita Silverajah et al., 2012). Additionally, OOC represents polar portion apart from carbonyl group of carboxylic ester functionality that is important for good compatibility. It acts as the stabilizer of polymers by reacting with acidic or unstable sites that promotes better thermal and oxidative stability. Also, the higher contents of OOC provides stronger interaction between hydrogen bonds in the PLA. To ensure excellent compatibility between plasticizer and polymer, the plasticizer used should be in polar structural component (Giita Silverajah et al., 2012).

Furthermore, Table 1 also states the result for elongation at break for PLA/ECO, PLA/ESO/ZnO, and PLA/ESO/APBA with value of 56 %, 33 %-37 %, and 130 %-190 %. The elongation at break is influenced by the free volume of the blends, where when it is full of plasticizer, a decrease of elongation at break occurs due to the free volume reduction (Dominguez-candela et al., 2021). From results, the remarkably higher elongation at break of PLA/ESO/APBA blends contributed by plasticization of ESO, where it interacts with PLA matrix through hydrogen bonding and weak polar interactions. This reduces intermolecular forces and increases the free volume matrix which allows polymer chains to move freely, thereby increasing chain mobility and plastic deformation. Also, APBA helps to form borate ester linkage through interaction between boric acid groups and hydroxyl groups in PLA. These dynamic crosslinking points provide balance for rigidity and flexibility, which allows stretching and recovery that contributes to high elongation at break properties. The combination of ESO and APBA in PLA matrix promotes chain mobility while preventing excessive deformation, which effectively enhances ductility while maintaining tensile strength. On the other hand, PLA/ECO/ZnO chemical interactions in the matrix did not form crosslinks and mainly acted as rigid fillers. This restricts chain mobility which decreases the elongation at break, resulting in lower result. For PLA/ECO, the lack of borate esters in the blend shows decrease in elongation at break due to limited ability of the matrix to stretch. Thus, PLA/ESO/APBA experienced high elongation at break rather than PLA/ESO/ZnO and PLA/ECO.

Then, flexural test results on PLA/EWCO blends are 6.7 MPa that indicates the ability of EWCO plasticizer to decrease the intermolecular interactions, increasing the extensibility and flexibility of the PLA (Omar et al., 2023). Epoxy group in EWCO plasticizers tend to interpose themselves between PLA chains to reduce van der Waals and hydrogen bonding. This interposition weakens the intermolecular force, allowing movement, rotation and sliding easier for PLA chains. As result, the polymer can deform easily without failure which contributes to high flexural strength.

Incorporating vegetable oil-based plasticizer into PLA was found to improve its mechanical performance and versatility where it can increase PLA chain mobility, ductility, tensile strength, elongation at break and flexural strength. Similarly, composite materials addition can further reinforce the polymer to increase tensile strength and stiffness. However, excessive use of plasticizers and composite material can be detrimental as they may cause phase separation and decreasing cohesion in the polymer matrix. This influences mechanical stability and durability of the PLA.

3. Conclusions

This paper reviewed the mechanical properties of PLA with vegetable oil-based plasticizer and composite material. The novelty of this review lies in its dual focus on vegetable oil-based plasticizers and composite

materials, presenting a comprehensive review regarding improvement in tensile strength, elongation at break and flexural strength, which can improve PLA mechanical performance. ZnO was shown to contribute antibacterial properties and increased tensile strength. Then, APBA contains boric acid group that influencing mechanical properties by chemically interacting with hydroxyl groups in PLA, which increases yield strength of the polymer. Besides, it found that EWCO can decrease the intermolecular interactions between the atoms, thereby increasing the flexural strength and flexibility of PLA composition. Additionally, EPO can improve tensile strength of PLA due to its higher OOC that provides stronger hydrogen bonds between PLA. Furthermore, to achieve good compatibility between PLA, plasticizers are recommended to be in polar structural component, which improved the interfacial adhesion, miscibility and mechanical performance. Further studies also need to be done on EPO since it possessed high potential in enhancing mechanical properties of PLA. From these enhancements, potential industrial applications include substituting mold components used for encapsulating and protecting semiconductor devices on printed circuit boards. Those are often made by using epoxy resin which are harmful towards living things and environment, as it did not break down naturally and can contribute to environmental pollution. Currently, epoxy-resin molding compounds (EMCs) are widely used to encapsulate integrated circuits, transistors and other components due to their high adhesion, chemical resistance and thermal stability. On the other hand, PLA provides greener alternatives, and less toxic compared to EMCs. For functionality concerns, PLA blends with plasticizer and composites modifications found to be able to achieve increase in mechanical properties. Thus, it is possible to make them suitable for electronic packaging specifications. Based on the observations, more research work is needed to improve the mechanical properties of PLA, to address the problems and difficulties discussed and further develop bio composites.

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