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Breaking Boundaries in Biodegradable Packaging: A Comprehensive Review on Magnetic Alignment of Iron-Cellulose in PLA

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ABSTRACT

This review paper explores the transformative potential of incorporating iron-coated cellulose into polylactic acid (PLA) composite films, presenting a comprehensive analysis of the advancements, implications, and challenges associated with this innovative approach. The introduction establishes the context, emphasizing the growing significance of sustainable packaging and the unique properties offered by biopolymers. The subsequent sections delve into the synthesis and fabrication methods, emphasizing the pivotal role of iron-coated cellulose in enhancing the mechanical, magnetic, and barrier properties of PLA nanocomposites. The review discusses in detail the magnetic alignment techniques employed, elucidating their impact on particle distribution and alignment within the PLA matrix. The exploration of magnetic field application reveals intricate relationships with curing times, emphasizing the dynamic interplay between magnetic alignment, curing processes, and particle distribution. The mechanical properties section further underscores the positive influence of magnetic alignment on tensile strength, stiffness, and dimensional stability, offering promising avenues for oriented structures in structural and functional materials. Expanding the scope to water barrier properties, the review investigates the effects of iron-coated cellulose on moisture absorption, revealing nuanced interactions that enhance the water barrier characteristics of the nanocomposites. Contact angle measurements provide insights into the surface properties, with the study uncovering how magnetic alignment contributes to improved hydrophobicity, thereby resisting water absorption and enhancing the effectiveness of these materials in packaging applications. The implications for sustainable packaging constitute a critical aspect of the review, shedding light on the environmental benefits and challenges associated with implementing magnetic alignment on a larger scale. The optimized material usage, renewable nature of iron-coated cellulose, and potential reduction in waste align with sustainability goals. However, challenges such as specialized equipment requirements and disposal considerations are also discussed, providing a balanced perspective. The paper concludes by summarizing the key advancements achieved through the incorporation of iron-coated cellulose into PLA composite films. It highlights the potential of these nanocomposites for future sustainable packaging, emphasizing their robust mechanical properties, magnetic functionalities, and enhanced water barrier characteristics. The conclusions underscore the collaborative effects of cellulose and iron coating, envisioning a future where sustainable packaging not only meets but surpasses industry standards. In essence, this review paper serves as a comprehensive guide, consolidating knowledge and insights to pave the way for future research and industry practices in the realm of sustainable and enhanced biopolymer packaging.

Keywords: Magnetic Alignment; Biopolymer Packaging; Iron-Coated Cellulose.

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1. INTRODUCTION

The demand for biodegradable packaging has experienced a noteworthy surge in response to escalating environmental concerns and a heightened awareness of the ecological impact associated with traditional plastic packaging. This surge reflects a dynamic response to the ongoing environmental crisis, with biodegradable materials emerging as an alternative to conventional plastics. Advancements in biopolymer research, notably the prominence of Poly-lactic acid (PLA), signify a conscientious shift towards sustainable packaging practices [1]. Concurrently, the plastic industry in Malaysia, like those in many developing nations, grapples with a surge in plastic waste, contributing significantly to environmental pollution. According to a data report published by SWCorp Malaysia, Malaysia generated 38 142 tonnes of waste daily in 2018 as illustrated in Figure 1 [2]. A represented from SWCorp Malaysia also mentioned that the latest statistics showed that plastic was 20% of total waste. This issue can alter habitats and natural processes, reducing ecosystem and directly affecting millions of people's live hood. In response to this challenge, new research has emerged to reduce the negative impact of synthetic polymers, particularly plastics derived from petrochemicals, on the environment and finite petroleum resources [3].

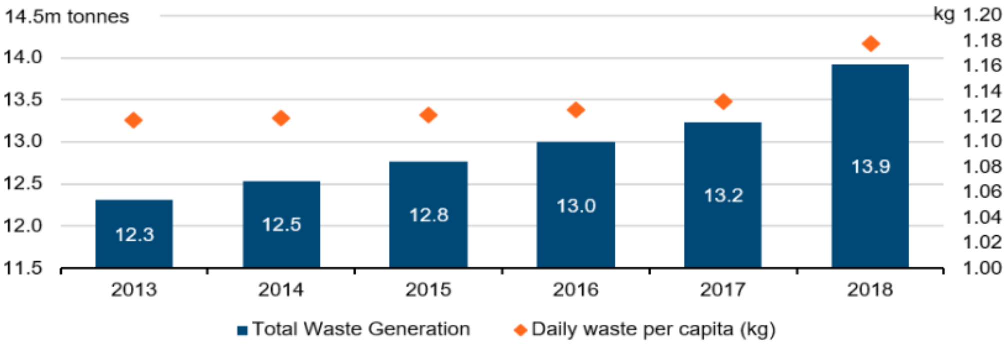


Figure 1. Waste generated in Malaysia (2013-2018)

Thus, this review aims to contribute to the discourse on biodegradable packaging, focusing on the advancements in PLA composites. Specifically, the study explores the innovative approach of magnetic alignment of iron-cellulose to address existing challenges in PLA packaging and enhance its mechanical and barrier properties [4, 5]. The two-fold objectives of this paper are to provide a comprehensive overview of the role of biopolymers, particularly PLA, in mitigating environmental concerns and to investigate the potential benefits of magnetic alignment as a reinforcing filler in PLA composites [6]. By meticulously examining existing literature and presenting experimental findings, this paper sheds light on the implications of magnetic alignment for sustainable and improved biopolymer packaging [7]. In the context of Malaysia and other developing nations, the plastic industry has inadvertently contributed to substantial environmental pollution [8, 9]. To combat this, the study highlights the potential of biopolymers, derived from renewable resources like corn and sugarcane, as a sustainable alternative to conventional petroleum-based plastics [10, 11]. Specifically, materials such as PLA, PHA, and starch-based polymers offer reduced carbon footprints and contribute to waste reduction, being compostable and minimizing environmental pollution [12, 13]. The low density, cost-effectiveness, rigidity, and enhanced plasticity of PLA make it particularly promising in addressing the burgeoning issue of plastic waste.

Furthermore, advancements in PLA composites are crucial due to the inherent brittleness and low barrier properties of PLA [14, 15]. Researchers have explored various strategies, including blending PLA with reinforcement materials such as cellulose, lignin, and PHA [16, 17]. While these advancements have shown promise, the poor alignment of reinforcing fillers, particularly cellulose, has presented challenges affecting mechanical properties [18, 19]. Consequently, the study delves into the investigation of incorporating iron-coated cellulose to overcome these limitations [20, 21]. This advancement enhances the performance and functionality of PLA-based materials, leading to improved barrier and mechanical properties. The continuous improvement and innovation in PLA composites contribute to the overall sustainability of biopolymer packaging, providing enhanced properties and functionality while reducing reliance on non-renewable resources. Ultimately, these advancements can drive the adoption of biopolymer packaging, contributing to a more sustainable approach in packaging practices.

1.1 Biopolymers and Packaging Challenges

The escalating environmental concerns stemming from the extensive use of traditional plastic packaging have propelled the demand for sustainable alternatives, particularly biodegradable materials. In response to this global imperative, various studies and statistical data underscore the urgent need for adopting eco-friendly packaging solutions. According to recent statistics [13, 22], the proliferation of plastics derived from petrochemicals, coal, and natural gas has led to a substantial ecological impact. This includes the depletion of petroleum resources and the alarming accumulation of plastic waste, with post-consumer plastic waste generation estimated to exceed 1 million tonnes in certain regions [23]. As a result, the environmental challenges posed by conventional plastics have become a pressing issue that necessitates immediate attention and innovative solutions.

Biopolymer is considered a promising solution to the environmental problem. According to Samir et al., [24], biodegradable polymers are materials that can be naturally degraded by the action of microorganisms, producing eco-friendly and useful substances such as carbon dioxide (CO_2) and methane (CH_4). These types of polymers can decompose in the soil without affecting the environment. Thus, biopolymers are an alternative to materials made from fossil resources because they can be used in almost everything, from packaging to durable disposable products [25]. Biodegradable polymers are divided into two groups, as shown in Figure 2.

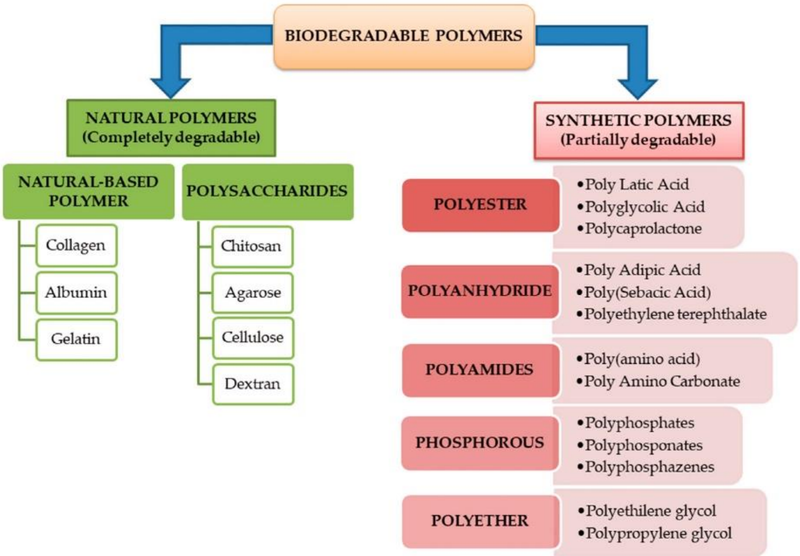


Figure 2. Classification of biodegradable polymers [26]

Natural biopolymers are derived from renewable sources consisting of microbial animal, and plant sources, while synthetic biopolymers are man-made polymers produced from renewable resources. Most natural-based polymers are fully biodegradable, while the synthetic polymers and their blends are not. However, aliphatic polymers such as polyhydroxy butyrate (PHB), polycaprolactone (PCL), and polylactic acid (PLA) are of special interest because they can generate metabolites upon degradation. In the family of biodegradable synthetic polymers, polylactic acid (PLA) appears to be one of the most attractive for film applications in agriculture and as a packaging material due to its good biodegradability, availability, and good mechanical properties. Polylactic acid (PLA) is a renewable thermoplastic polymer matrix and an aliphatic polyester produced from lactic acid and is a key component in the production of biodegradable products, making it one of the most promising biopolymers [10, 11]. Lactic acid is produced by fermentation under controlled conditions from carbohydrate sources such as corn starch or sugarcane, making the process sustainable and renewable. Among biopolymers, PLA has higher thermal stability, making it suitable for automobile and packaging applications that require long-term use. Although biodegradability can help reduce plastic waste due to the greenhouse effect, bio-based sustainable materials known as bioplastics are currently considered the way to go and could be an alternative in the future when petroleum resources are depleted. There is growing interest in the use of PLA applications that require biodegradable disposable materials which include improvements of physical, thermal, barrier and mechanical properties. Outline challenges in PLA packaging, setting the stage for the magnetic alignment solution.

However, PLA's brittleness and low crystallization rate have limited its advantages in industry, especially in the packaging industry. According to Outline et al., [27], PLA is a brittle polymer with an elongation at break of less than 10%, which makes it unsuitable for demanding mechanical performance. Therefore, PLA is unsuitable for packaging applications that require plastic deformation at higher loads. The weakness of polylactic acid (PLA), such as brittleness, causes some researchers to have some ideas for modifications of this polymer. According to Rasal et al.,[28] , the surface modification of PLA has been attempted to improve toughness, roughness and to introduce reactive groups. There are many ways to modify this polymer, such as the inclusion of plasticizer and the most popular method is introducing reinforcing filler into the biodegradable polymer to produce a composite material [28, 29].

1.2 Reinforcing Fillers and Magnetic Alignment

In general, the addition of filler materials into a polymer matrix is a common research study undertaken by various researchers. When fillers are added to the matrix, they render composites with a better surface finish retarding the formation of coarse structure and getting better mechanical properties which are impossible to obtain with coarse structure [30]. Fillers can be divided into two types which are organic and inorganic fillers (see Table 1). The addition of fillers in the polymer matrix, such as PLA, improves the properties, enhances process ability, and reduces material cost. Usually, filler particles are added to the polymer matrix in micro and nano-sized. Thus, the applications of PLA polymer as a matrix in natural fiber-reinforced composites have drawn intensive research interest.

Table 1. Difference between organic and inorganic filler

Organic filler	Inorganic filler
Cellulose	Silica
Chitin	Alumina
Lignin	Calcium carbonate
Starch	Titanium dioxide

Natural fibers such as cellulose, bamboo, starch, and straw as reinforcing agents or fillers in polymer composites have attracted substantial attention. Cellulose fibers-reinforced polymer composites have received much attention because of their low density, low cost, non-abrasiveness, fire resistance, lack of toxicity, and biodegradable properties compared to other inorganic reinforcing fillers [31]. Cellulose molecules are made up of glucose units linked together in long chains (β-1,4 glycoside linkages bind the repeating units of D-anhydro glucose$C_6C_{10}O_5$), linked together in microfibrils. The hydrogen bonding in cellulose determines its crystallinity which controls the physical properties of natural fibers. This is the primary component that gives them resilience, stiffness, and stability. Cellulose is a promising candidate to revise non-renewable and non-biodegradable packaging materials due to its particular properties such as biodegradability and low cost [32, 33]. In addition to their basic properties, they are highly available and present reliable barrier and mechanical properties. Including active packaging is highly desirable in the food packaging industry to maintain the quality and extend the shelf life of packaged

The study aligns seamlessly with established research, such as the work by Norazlina et al., [34], emphasizing the transformative impact of surface modifications on the hydrophilicity and hydrophobicity of fibers. The specific modification of MCC using iron oxide nanoparticles (NPs) emerges as a crucial factor in influencing contact angles, showcasing the effectiveness of these modifications in tailoring the surface properties of the nanocomposites foods. The use of biopolymer incorporated with cellulose as a packaging material could expand active packaging applications in the food packaging industry.

1.3 Explore the concept of reinforcing fillers in polymers

Reinforcing fillers are integral components that play a pivotal role in augmenting the mechanical, physical, thermal, and barrier properties of polymers, thereby enhancing their overall performance and versatility across diverse applications [35, 36]. These fillers, commonly existing in particle or fiber forms, are strategically incorporated into the polymer matrix to fortify and strengthen the material. The addition of reinforcing fillers serves the purpose of mitigating inherent limitations associated with polymers, including issues such as low stiffness, poor stability, and inadequate strength.

Traditionally, a spectrum of organic fillers, exemplified by cellulose, chitin, lignin, and starch, alongside inorganic counterparts like silica, alumina, calcium carbonate, and titanium dioxide, are employed to illustrate the diversity in polymer matrices. Noteworthy among these is cellulose, derived from natural fibers, which stands out for its commendable properties, including low density, cost-effectiveness, fire resistance, lack of toxicity, and biodegradability [37, 38]. The application of cellulose as a reinforcement agent finds prominence in packaging, owing to its ability to distribute stress and enhance load-bearing capabilities. Beyond mechanical enhancements, reinforcing fillers contribute significantly to improving the thermal and barrier properties of polymers [30, 39]. Integration of fillers with high thermal conductivity, such as metal oxides or carbon-based materials, facilitates superior heat dissipation and resistance to temperature changes—an imperative consideration in applications where thermal stability is paramount. Furthermore, fillers with low permeability, exemplified by certain nanoparticles, act as effective barriers against gases and moisture [40]. This quality is particularly valuable in packaging applications, where the preservation of product quality and extension of shelf life are of paramount importance.

In summation, the concept of reinforcing fillers in polymers serves as a multifaceted strategy, providing avenues to tailor materials with augmented mechanical strength, improved thermal conductivity, and enhanced barrier properties. The diverse array of fillers, ranging from traditional organic to inorganic variants, presents a rich landscape for optimizing polymer matrices across various industrial applications.

1.4 Introduce the idea of magnetic alignment and its potential benefit.

The main challenge in mixing reinforcing fillers, such as cellulose, with thermoplastics like PLA, is the limited dispersion of cellulose in the polymer matrices [41, 42]. The polar surface of cellulose leads to poor hydrophilicity, dispersibility in non-polar solvents, and compatibility with hydrophobic matrices. These factors not only impede the preparation of reinforced polymer composites but also result in weak interfacial bonding. To address this issue, it becomes imperative to enhance the hydrophobicity of cellulose, aiming to improve its dispersibility in non-polar solvents or compatibility with hydrophobic matrices [41]. The poor dispersion of microcrystalline cellulose (MCC) in the PLA matrix, as highlighted by Mohamad Haafiz et al., [43], leads to a decrease in tensile strength and elongation at the break of the PLA/MCC composite. This inefficiency arises from the hydrophilic nature of MCC conflicting with the hydrophobic nature of thermoplastics like PLA, hindering effective interaction and dispersion throughout polymer matrices.

The application of magnetic fields to enhance the dispersion and alignment of fillers in composite materials has recently garnered significant attention. Magnetic alignment, involving the orientation of particles or materials using magnetic fields, presents a fascinating concept. By applying a magnetic field to a magnetic particle, these particles can align themselves in a preferred direction, resulting in a collectively oriented structure. This approach proves particularly beneficial for addressing dispersion challenges associated with fillers that tend to agglomerate or self-attract, such as nanofillers. While magnetic alignment has been successfully employed for nanotubes with considerable magnetic properties [44], its application to MCC is hindered by the latter's non-magnetic properties. However, the introduction of metal nanoparticles with magnetic properties, particularly through surface modification of MCC, stands out as an innovative idea. Successful incorporation of magnetic nanoparticles into MCC enhances its magnetic susceptibility, supporting the production of MCC-reinforced polymer composites with improved dispersion and suitable filler alignment at both micro and macro levels. Moreover, the incorporation of a magnetic field for improving the dispersion and alignment of cellulose in polymer matrices offers a potential solution to the challenges associated with cellulose-reinforced polymer composites. Decoration with iron oxide nanoparticles has shown promise in enhancing alignment, conductivity, and stability of films due to their magnetic character [45]. This magnetic character can be harnessed through proper surface coating of cellulose with ferromagnetic particles like iron, nickel, and cobalt, enabling their dispersion into suitable solvents to form homogenous suspensions known as ferrofluids [46]. This novel technique opens avenues for aligning cellulose fibers magnetically during the solvent casting of polylactic acid (PLA) for advanced packaging applications. Figure 3 illustrates the magnetic dipole moments in magnetic materials, emphasizing the principles behind ferromagnetic, paramagnetic, anti-ferromagnetic, and diamagnetic behavior. The alignment achieved through the application of an external magnetic field (H) can significantly impact the properties of the resulting composite materials, as discussed by several researchers [47, 48]. In conclusion, the integration of magnetic alignment techniques into the preparation of polymer composites represents a promising avenue for overcoming challenges related to filler dispersion and alignment, thereby enhancing the overall performance of the resulting materials.

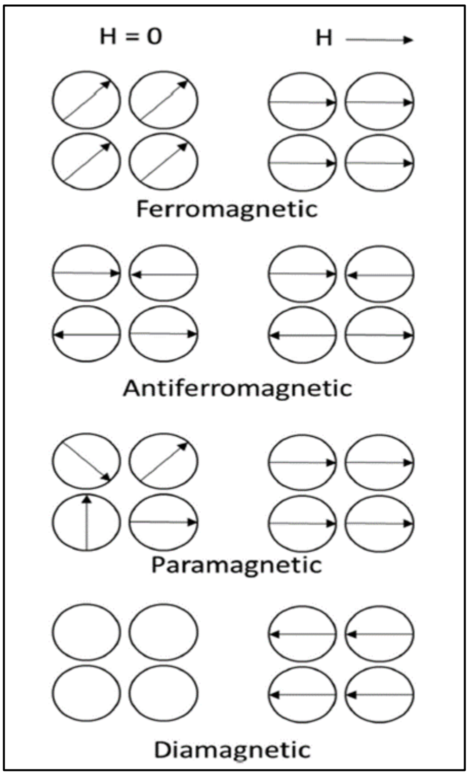


Figure 3. Illustration of the magnetic dipole moments in magnetic materials with and without an external magnetic field (H)

Alignment, defined as the process of rotating and moving particles to orient them in a consistent direction, plays a crucial role in influencing the properties of cellulosic materials. Innovations are essential to enhance or create diverse properties in the final products of cellulosic materials [49, 50].The alignment of cellulose significantly impacts the performance of cellulose fiber-based materials, bridging the gap between the nanoscale and macroscale and allowing the incorporation of improved nanoscale properties into high-performance macroscale materials.

Despite its significance, the alignment and dispersion of cellulose pose major challenges. To overcome these limitations and expand the utility of cellulose materials, a strategic approach involves combining them with ferromagnetic particles containing iron oxide nanoparticles (NPs) [51]. In this study, iron (III) nitrate ($Fe(NO_3)_3.9H_2O$) is chosen for its cost-effectiveness and lower toxicity. Three primary methods are employed for cellulose particle alignment: shear-induced alignment in a fluid flow field, electric field-induced orientation, and magnetic field-induced orientation [52, 53]. The magnetic field proves useful for processing weak magnetic materials, exerting a force on particles to align them based on their magnetic nature.

Microcrystalline cellulose (MCC), generally considered an antimagnetic material with magnetic anisotropy, demonstrates the ability to align under magnetic fields [51]. This alignment is pivotal for producing oriented structures in structural and functional materials, enhancing their properties [7]. In the presence of a magnetic field, particles align their net moment and easy magnetization direction with the field orientation, allowing free rotation. Research by Nagarajan et al.,[48], highlights how external magnetic fields in the manufacturing process led to parts with high permeability. Similarly, Song et al., [54] utilizes an electromagnetic particle alignment configuration in an inkjet printing-based additive manufacturing process. Kokkinis et al., [55], demonstrate local control of particle alignment using a low external magnetic field and a rotating neodymium permanent magnet.

While magnetic fields have been utilized for particle alignment, there is a notable gap in research on the alignment of natural fibers in polymer resin using a magnetic field for packaging purposes. Therefore, this proposed research introduces a novel technique involving the coating of cellulose with iron oxide nanoparticles. This technique makes cellulose fibers magnetically responsive, and an optimal magnetic field is applied to align the fibers during the solvent casting of polylactic acid (PLA). Figure 4 illustrates the magnetic effect on the deposition of ferromagnetic particles on polymer resins. Without a magnetic field, the net magnetization is zero, as individual particles are randomly oriented and cancel each other's field. However, upon applying a magnetic field, the particles align in the direction of the magnetic field, presenting magnetic techniques as a promising means to align reinforcing fillers in polymer composite films.

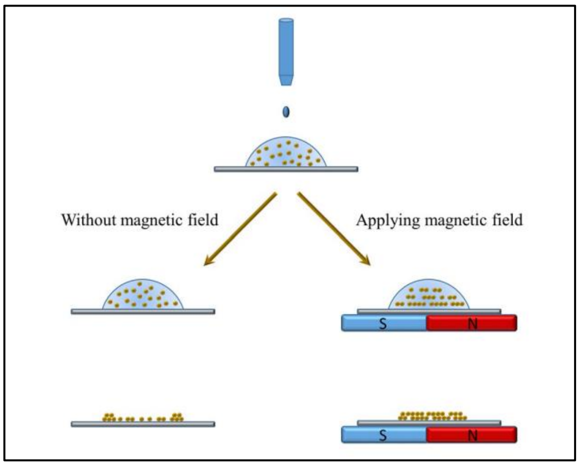


Figure 4. Schematic illustration of magnetic field effect on ferromagnetic particle deposition in polymer resin

2. METHODOLOGY & CHARACTERIZATION TECHNIQUE

The methodology for preparing the iron-modified cellulose and incorporating it into PLA involves a systematic two-step process. Firstly, cellulose fibers are modified by coating them with iron oxide nanoparticles. This is achieved through a controlled deposition process, ensuring uniform coverage of the cellulose surface. The synthesis of iron oxide nanoparticles is a crucial step for better control of particle size, distribution, and morphology. Various metals, metal oxides, sulphides, polymers, and composite nanoparticles can be prepared using different synthesis techniques, with chemical and physical methods being the primary approaches [56]. Physical synthesis, involving the use of mechanical equipment to break down ferrous material, includes vacuum evaporation, laser heating evaporation, electron beam irradiation, and sputtering. On the other hand, chemical methods, like the chemical reduction method, are considered easy and relatively inexpensive [57].

Key parameters and conditions are critical in both steps of the methodology. The temperature and duration of the iron coating process influence the thickness and distribution of the iron oxide layer on cellulose fibers. Achieving uniform dispersion and alignment of the modified cellulose within the PLA matrix is a notable challenge due to the hydrophilic nature of cellulose. Moreover, during solvent casting, factors such as solvent type, concentration, and casting conditions impact the dispersion and alignment of modified cellulose within the PLA matrix. Precise control over these parameters is essential to achieve reproducibility and consistency in the fabrication of the composite material.

To prepare the pure PLA film, the solvent casting method is chosen due to its simplicity and ease of control in producing PLA film with balanced properties [58, 59]. Firstly, 5 g of PLA is mixed in 100 ml chloroform ($CHCl_3$) and rapidly agitated at room temperature until completely dissolved. The solution is then poured into a glass petri dish and dried at room temperature for 24 hours. The resulting PLA film is carefully removed from the casting surface for further analysis.

2.1 Characterization Techniques for Analyzing Modified Cellulose/PLA Composite

The analysis of modified cellulose/PLA composite films encompasses a diverse array of techniques aimed at assessing their structural, thermal, mechanical, and morphological properties. These techniques play a crucial role in comprehensively understanding the characteristics and performance of the composite material.

Scanning Electron Microscopy with Energy Dispersive X-ray Spectroscopy (SEM-EDX): SEM is employed to scrutinize the surface morphology and microstructure of the composite, offering high-resolution images that unveil the dispersion of cellulose particles within the PLA matrix and the interfacial interaction [60, 61]. Complementarily, the EDX technique analyzes the elemental composition of the sample, providing insights into the presence of iron-coated cellulose.

Fourier Transform Infrared Spectroscopy (FTIR): FTIR is utilized to identify chemical functional groups and analyze molecular interactions within the cellulose/PLA composite, contributing to a detailed understanding of the chemical composition and bonding within the material [62, 63].

Thermogravimetric Analysis (TGA): TGA is instrumental in investigating the thermal stability and degradation behaviour of the composite, offering valuable insights into its thermal characteristics [64].

Magnetic Alignment: This technique involves capturing images before and after applying an external magnetic field to PLA suspension using a High-Speed Camera Microscope (HSCM) as illustrated in Figure 5. In this study, particles are dispersed in PLA suspension to form the Fe-coated cellulose/PLA nanocomposite film. The alignment of magnetic particles is achieved by placing a permanent magnet underneath the substrate to force the particles to align with the externally applied magnetic field.

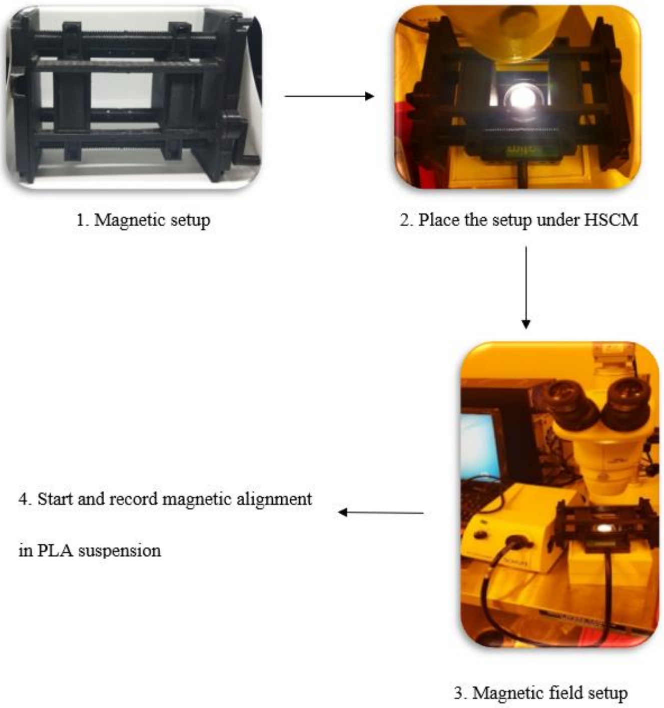


Figure 5. Magnetic alignment setup using high speed camera microscope (HSCM)

Mechanical Testing: Tensile, flexural, and impact testing are conducted to evaluate the mechanical properties of modified cellulose/PLA composites. The stress and strain of the films are measured using a texture analyzer, specifically a Shimadzu universal tensile tester, following ASTM D882-02 standard methods [65-67].

Water Absorption: This test assesses the moisture absorption capacity of modified cellulose. The swelling test or water absorption analysis is conducted following ASTM D 570.

Contact Angle Measurement: To corroborate the results of water absorption, contact angle measurements are performed to evaluate the wettability of the film and determine its hydrophilicity and hydrophobicity. This is achieved using the sessile drop method [68, 69].

The rationale behind the selection of these techniques lies in their ability to offer detailed information about different aspects of the modified cellulose/PLA composite. SEM allows visual inspection of filler dispersion and alignment, while FTIR provides chemical information about interactions within the composite. Benchmarking against standard PLA or other biodegradable materials is integral, offering a comparative analysis to evaluate the composite's performance against established materials and providing insights into potential advantages or areas for refinement. This comprehensive approach enhances the significance of the research in the broader context of biodegradable materials.

2.2 Magnetic Alignment:

Image analysis of iron cellulose/PLA composites subjected to a constant distance of separation between an external magnet demonstrated a comparison in alignment with increasing magnetization time before curing (see Figures 6 and 7). The microstructure was observed before and after the application of an external magnetic field at different curing times. Without an applied magnetic field, various particle deposition patterns exhibited random movement due to zero net magnetization. However, under the influence of an external magnetic field, the particles aligned with the magnetic flux (MF). The findings align with those of Al-Milaji et al., [70], emphasizing the significant impact of an external magnetic field on the deposition of colloidal ferromagnetic particles. These particles were compelled to chain and assemble with their easy magnetization directions along the external magnetic field vector, as reported by Al-Milaji et al., [70]. Additionally, Kimura & Kimura, [52] highlighted the feasibility of achieving alignment and patterning of cellulose fibers using a magnetic field modulator.

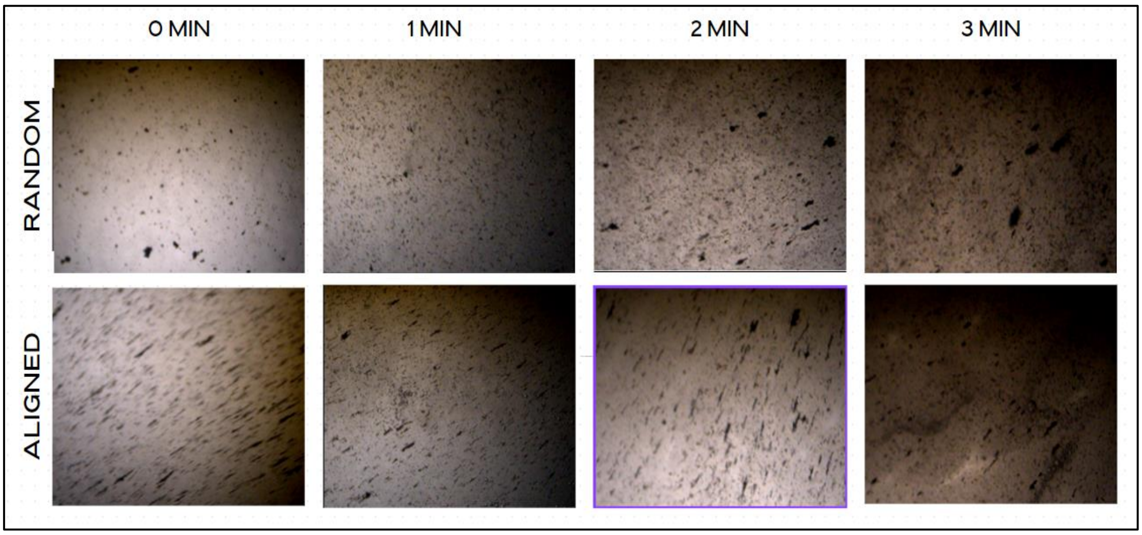


Figure 6. Image of 2.5 % iron cellulose in PLA suspension before and after applying magnetic field

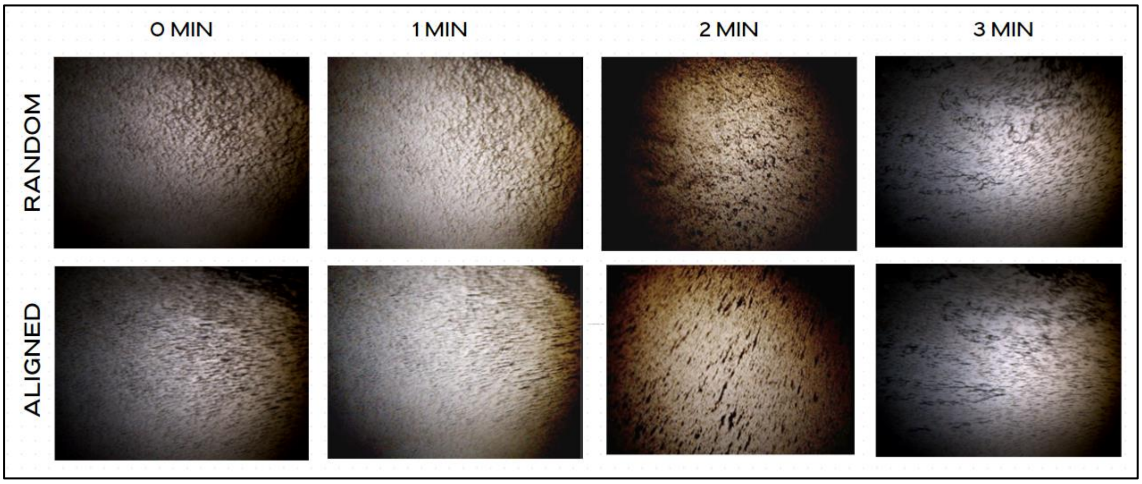


Figure 7. Image of 5.0 % iron cellulose in PLA suspension before and after applying magnetic field.

The study delved into the application of maximum magnetic fields to align metal phase particles, presenting a novel technique for producing oriented structures in structural and functional materials with improved properties. However, the challenge arises with increased curing time, leading to the immediate curing of polymer resins and affecting particle movement. The particles do not have sufficient time for full alignment, as evidenced by observations at different pre-curing times, including 3 min, 2 min, 1 min, and 0 min (no pre-curing). The increase in magnetization time in these magnetic fields is attributed to the non-uniform distribution of iron cellulose particles in PLA suspension, potentially caused by the formation of chains of particles and premature suspension curing. Optimal alignment quality was achieved at a curing time of 0 min and 1 minute, emphasizing the critical role of precise control over curing parameters in maximizing the benefits of magnetic alignment. This intricate analysis is drawn from existing literature, providing a comprehensive understanding of the magnetic alignment of iron-cellulose in PLA within the context of the broader review.

2.3 Mechanical Properties:

The extensive evaluation of Fe-coated cellulose/PLA nanocomposites extends to their mechanical properties, shedding light on their performance in structural applications. The stiffness of microcrystalline cellulose (MCC) uniformly dispersed in the PLA matrix emerges as a critical factor influencing the tensile strength of composite films, aligning with findings from previous studies [71]. However, the present review nuances these observations, highlighting the impact of Fe-coated cellulose on the overall mechanical behaviour of PLA composites.

In line with expectations, the graph in Figure 8 distinctly depicts a significant increase in tensile strength as the iron concentration in the composite film rises, reaching its peak at 5.0%. This enhancement is attributed to the inherent stiffness of iron cellulose, coupled with the presence of immobilized or partially immobilized polymer phases and the superior surface toughness of modified MCC. A nuanced comparison with FeNp/MCC/PLA composite films reveals a minor decrease in tensile strength, signalling potential challenges in the interfacial adhesion between PLA matrix and the reinforcing filler, as articulated by [71].

Delving into the nuances of the magnetic field application, the study explores various pre-curing times, ranging from 3 minutes to no pre-curing. Surprisingly, the optimum alignment quality is achieved at curing times of 0 minutes and 1 minute, challenging conventional expectations. This underscores the dynamic interplay between magnetic alignment, curing time, and particle distribution in PLA suspension. The magnetic field's role in particle alignment is evident, but the intricate dance between curing time and magnetic alignment adds a layer of complexity to the mechanical behaviour of the composites.

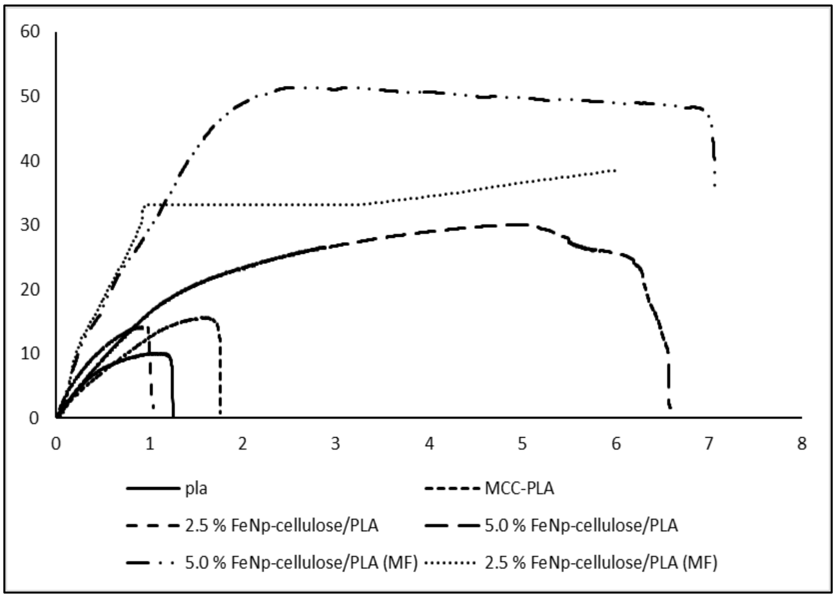


Figure 8. Tensile strength analysis between PLA and modified PLA

Furthermore, the mechanical testing protocol involves not only tensile strength analysis but also includes flexural and impact testing. This holistic approach aligns with the overarching goal of evaluating Fe-coated cellulose/PLA nanocomposites for diverse applications. The findings corroborate the multifaceted nature of these materials, showcasing promising mechanical properties that extend beyond simple tensile strength considerations. In synthesizing these mechanical property insights, the review not only consolidates existing knowledge but also contributes nuanced perspectives on the interplay of factors influencing the mechanical behaviour of Fe-coated cellulose/PLA nanocomposites. This nuanced understanding is vital for guiding future research directions and optimizing the utility of these materials in practical applications.

In general, the mechanical performance of composite films is influenced by factors such as filler compatibility, aspect ratios, and orientations [71]. The study concludes that the stiffness of MCC uniformly dispersed in the PLA matrix improves the tensile strength of composite films. However, the decrease in stress for MCC-PLA films may occur due to MCC aggregation in the PLA matrix, emphasizing the importance of compatibility between the polymer matrix and cellulose, as noted by Haafiz et al., [72]. As the content of reinforcing fillers, such as iron-coated cellulose, increases, the micro-spaces between fillers expand, leading to nanofiller agglomeration and a subsequent decrease in mechanical strength. The study indicates that the mechanical properties of Fe-coated cellulose/PLA nanocomposites, especially tensile strength, are significantly enhanced with increasing iron concentration. However, a slight decrease compared to FeNp/MCC/PLA composite films suggests potential challenges in interfacial adhesion between PLA matrix and reinforcing filler, in line with observations by [71].

Despite the potential drawbacks, the focus of the study is to evaluate the effect of a magnetic field on Fe-coated cellulose/PLA nanocomposites. The application of a magnetic field results in a clear enhancement in strength and yield, attributed to better dispersion and alignment of iron cellulose in PLA suspension. This aligns with literature stating that fiber orientation significantly affects tensile strengths. Magnetic films are acknowledged for their mechanical as well as magnetic properties, emphasizing their value in diverse applications.

2.4 Water barrier

The investigation into water barrier properties adds a crucial dimension to the comprehensive understanding of Fe-coated cellulose/PLA nanocomposites within the context of a review paper. The interplay between hydrophilicity and hydrophobicity significantly influences their potential applications, particularly in packaging. The findings from this study, along with existing literature, illuminate the intricate relationship between modified microcrystalline cellulose (MCC) and polylactic acid (PLA) in affecting moisture absorption.

As highlighted by the study, the incorporation of modified MCC into the PLA matrix brings forth notable variations in moisture absorption behaviour. The water absorption test, depicted in Figure 1.8, illustrates the impact of iron cellulose on the hydrophilicity/hydrophobicity nature of PLA. The slow water absorption of pure PLA, attributed to its hydrophobic nature [73, 74], undergoes a noticeable increase with the addition of MCC due to its inherent hydrophilic character [75]. The hydroxyl groups in cellulose interact with water molecules, encouraging water intake [76]. However, the rate of water absorption is mitigated with increasing concentrations of modified cellulose, showcasing the effectiveness of the iron oxide nanoparticles (NPs) in enhancing water barrier properties. Figure 9 presents a comparative analysis of water solubility for different composite films, confirming the low water absorption rate with increasing modified cellulose. The 2.5% and 5.0% concentrations of iron-coated cellulose, especially after applying a magnetic field, emerge as the most effective films with lower water barrier properties. The reinforcement of fillers in these concentrations showcases strong interaction within the polymer matrix, enhancing the barrier to the diffusion of water molecules into the PLA film matrix. In addition to moisture absorption, the study incorporates contact angle measurements as a critical parameter to confirm the hydrophilicity and hydrophobicity of the films.

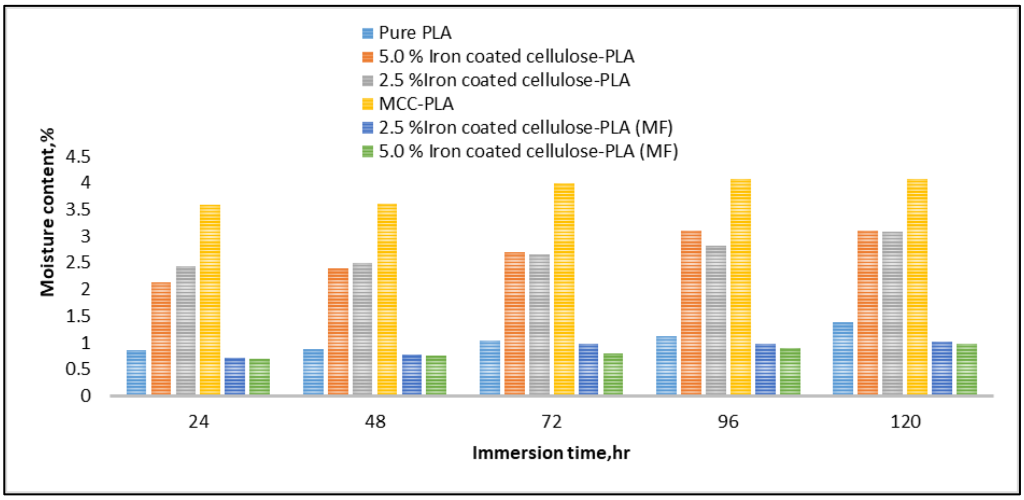


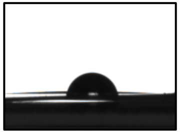
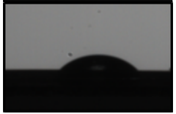
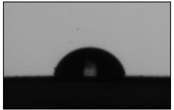
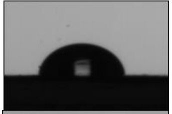
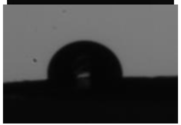
Figure 9. Water solubility of composites film for 120 hr

2.5 Contact Angle Measurement

The incorporation of contact angle measurements in the review of Fe-coated cellulose/PLA nanocomposites significantly enhances the comprehensive understanding of these materials, especially in the context of their applications in packaging. The exploration of hydrophilicity and hydrophobicity through contact angle measurements provides valuable insights into the surface characteristics, complementing the broader analysis presented in this review. By integrating findings from the study and aligning them with existing literature, a more nuanced perspective emerges regarding the intricate interplay between modified microcrystalline cellulose (MCC) and polylactic acid (PLA) in shaping the surface properties of these nanocomposites.

The thorough analysis of contact angle measurements, detailed in Table 1.1, sheds light on the water contact angles for different composite films. The inherent hydrophobic nature of pure PLA, reflected in a contact angle of 79.64^0, undergoes a significant shift with the addition of modified cellulose, indicating enhanced hydrophobicity. This change is attributed to the increased surface roughness resulting from the incorporation of the modified filler.

Table 2. Values of contact wetting angles of samples surfaces

Sample	Contact Angle (CA),^{0}	Images
Pure PLA	79.64	
PLA/MCC 1 wt %	67.65	
PLA/MCC 3 wt %	66.85	
PLA/MCC 5 wt %	64.35	
PLA/Fe-MCC 2.5 %	78.87	
PLA/Fe- MCC 5.0 %	79.97	
PLA/Fe-MCC 2.5 % (MF)	81.48	
PLA/Fe- MCC 5.0 % (MF)	84.91	

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Furthermore, the investigation into contact angles provides practical insights into the water resistance of Fe-coated cellulose/PLA nanocomposites. The results indicate that the 2.5% and 5.0% concentrations of iron-coated cellulose, especially after applying a magnetic field, exhibit lower water barrier properties. This outcome is linked to the strong interaction within the polymer matrix, enhancing the barrier against water diffusion into the PLA film matrix. Contact angle measurements serve as a tangible metric for assessing the success of these nanocomposites in achieving the desired water repellence.

2.6 Implications for Sustainable Packaging

The exploration of magnetic alignment within the context of sustainable packaging is a critical dimension of this review, shedding light on both the potential environmental benefits and challenges associated with the implementation of this innovative technique. The integration of magnetic alignment in the creation of Fe-coated cellulose/PLA nanocomposites aligns seamlessly with the overarching objective of advancing sustainable practices within packaging materials. The precision achieved through magnetic alignment plays a central role in enhancing the structural and mechanical properties of these nanocomposites, thereby contributing to their overall sustainability.

A significant environmental advantage lies in the optimized material usage facilitated by magnetic alignment. This technique ensures a controlled and uniform distribution of iron cellulose particles within the PLA matrix, resulting in improved mechanical performance. Beyond enhancing functionality, this precision reduces material waste, aligning closely with sustainability principles. Additionally, the incorporation of Fe-coated cellulose, derived from renewable and biodegradable sources, further enhances the eco-friendly profile of the nanocomposites, offering an environmentally conscious alternative for traditional packaging materials.

Nevertheless, the transition to magnetic alignment on a larger scale presents its set of challenges. The requirement for specialized equipment and controlled manufacturing processes may pose logistical and economic hurdles for widespread adoption. Scaling up production while maintaining cost-effectiveness and minimizing energy consumption becomes a critical consideration. Furthermore, thoughtful disposal and end-of-life management of Fe-coated cellulose/PLA nanocomposites are crucial to ensuring that the benefits of sustainable packaging are not compromised by potential environmental impacts associated with these materials.

An additional consideration is the necessity for comprehensive life cycle assessments to evaluate the overall sustainability of magnetic alignment in packaging. Examining environmental impacts throughout the entire life cycle, from raw material extraction to end-of-life disposal, will provide a holistic understanding of the ecological footprint of these nanocomposites. In conclusion, while magnetic alignment holds promise for sustainable and enhanced biopolymer packaging, its widespread adoption necessitates a careful examination of both its environmental benefits and potential challenges. By addressing these implications, this review paper contributes significantly to the ongoing discourse on sustainable packaging practices, guiding future research and industry practices towards more environmentally friendly solutions.

3. CONCLUSION

The review underscores the significant advancements achieved through the incorporation of iron-coated cellulose into PLA composite films, illuminating a promising trajectory for sustainable packaging. The addition of modified cellulose has demonstrated tangible improvements in mechanical strength and barrier properties, setting the stage for enhanced biopolymer packaging solutions. The collaborative effect of aligned cellulose fibres and the magnetic property of iron presents a multifaceted enhancement of material properties. Looking forward, these strides in biopolymer packaging, particularly the integration of iron-coated cellulose into PLA composite films, hold tremendous promise for the future of sustainable packaging practices. The presence of iron-coated cellulose serves as a robust reinforcing agent within the PLA matrix, fortifying its mechanical properties. The cellulose fibres contribute strength and stiffness, complemented by the iron coating that optimizes dispersion and alignment. This combined approach yields biopolymer packaging films with elevated tensile strength, enhanced impact resistance, and overall durability. Furthermore, the introduction of magnetic properties through iron coating opens up innovative possibilities, such as magnetic field alignment. Harnessing magnetic fields allows for controlled particle alignment within biopolymers, presenting a strategy with the potential to further elevate mechanical and barrier properties in packaging materials. This strategic alignment offers prospects for improved preservation of packaged products, extending shelf life, and concurrently reducing waste. In conclusion, the reviewed advancements in incorporating iron-coated cellulose into PLA composite films not only mark a pivotal moment in material science but also chart a promising course for the evolution of sustainable packaging. By amalgamating the insights from both conclusions, the paper contributes a holistic understanding of the subject, guiding future research and industry practices toward more resilient and environmentally friendly packaging solutions.

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