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Recent advances in different fabrication techniques to formulate biodegradable plastic and their effects on functional properties of the film: A mini-review

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Abstract

Current global markets and companies are focusing on and trying to explore their productivity in terms of sustainable biodegradable packaging materials, implementing flexible and standardized technology to fabricate packaging materials due to increasing consumer demand. In this mini-review, we look into the state of the art in the packaging industry in light of recent advancements in the field of smart fabrication technology including solvent casting, spin coating, dip coating, electrospinning technique, phase immersion, melt and blown extrusion, how the techniques are correlates with fabrication conditions, physio-mechanical properties and also their positive and negative impact on the film properties and their improvements for food packaging. The following and related topics are shortly reviewed and presented in the correct order.

Keywords: Biodegradable plastic, fabrication techniques, physical-mechanical properties

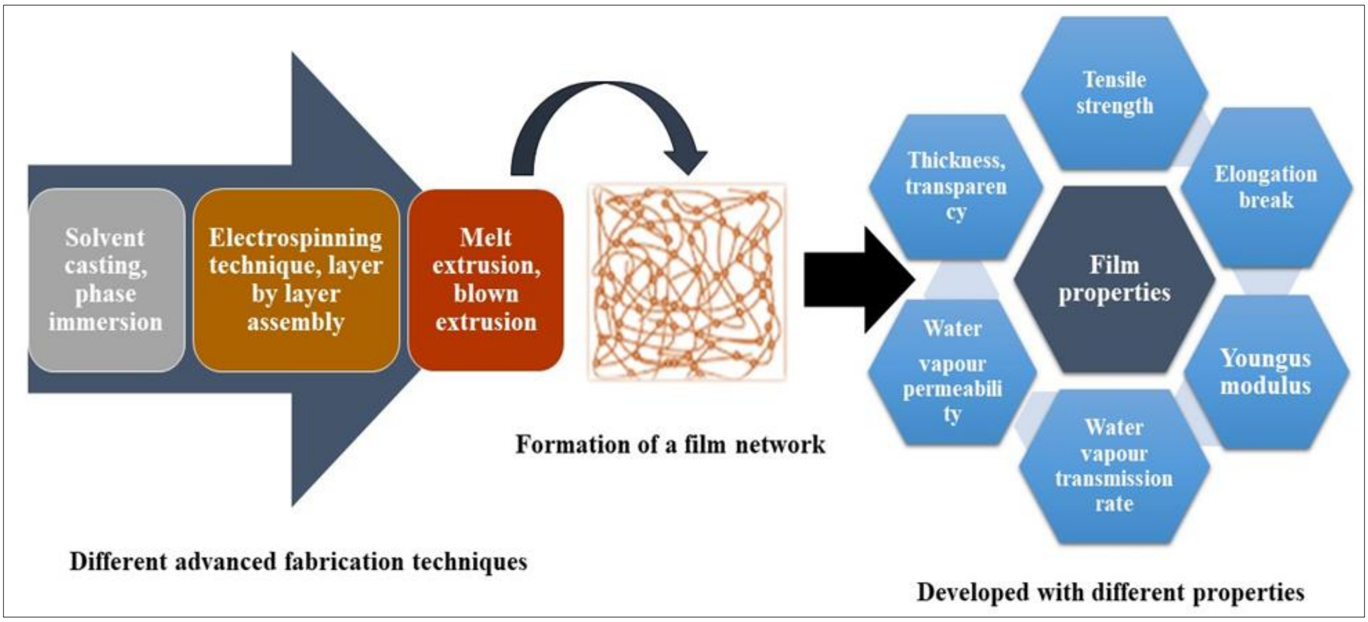
1. Introduction

Biodegradable packaging has been directed toward the development of technologies for the generation of packaging with biodegradable materials which can serve as substitutes for conventional packaging. Biodegradable polymers are reliable sources of edible packaging materials with excellent renewability, biodegradability, and bio-compatibility as well as antioxidant and antimicrobial activities (Rosales *et al*., 2022) [1]. Packaging films developed from different petroleum-based polymers have various disadvantages such as releasing micro-plastic, and a high carbon footprint (Ali *et al*., 2023) [2]. To mitigate these problems, consumer demands are increasing for alternative materials with similar functionalities and characteristics for sustainable packaging applications.

Recently, green plastic demand is growing for producing high-quality packaging film with better properties and formulated by using advanced fabrication techniques to ensure better quality and texture. The solvent casting method is one of the most common and popular methods to produce polysaccharide-based biodegradable plastic. Electrospinning, layer-by-layer assembly, phase immersion, extrusion, and injection molding are some additional novel synthesis techniques adopted by industries to produce biodegradable plastic (Bertuzzi *et al*., 2016) [3]. Previous research work indicated that biopolymer-based thin film developed with different characteristics and functional properties depending on the selection of material and fabrication techniques (Liu *et al*., 2021) [4]. Spin coating or spin casting is a dynamic approach that conveys more stress to the prepared film. As per the previous study, solvents can produce more stress in the film, and the previous work has shown that the solvents evaporate from the thin film more slowly in flow coating, the solvent evaporation from the film solids provides a dynamic structure to the film (Kim *et al*., 2018) [5].

The fabrication techniques used for biopolymer-based films and coatings with their functional properties. Recent advances in fabrication techniques and their applications are also reviewed. The electrodynamic approach is a new technique for biopolymer coating which is directly fabricated with the food surface and acts as a moisture barrier and decreases the respiration rate by modifying the outer environment of the product (Versino *et al*., 2023) [6]. Thin film sheets are developed with lower thickness whereas, the coating is directly formed on the surface of the product, which can be ready to consume by the customer, or applied on the film to enhance mechanical, and optical properties which can serve as packaging material (Feurer *et al*., 2017) [7].

Recent advances in sustainable plastic, developments can be appropriately categorized into innovations in the manufacturing of disposable plastic. However, many challenges are faced by those attempting to reproduce plastic to become environment friendly, innovation of green plastic has provided broad changes in producing plastic (Chia *et al*., 2020) [8]. The development of the bioplastic industry is changing the pathway in history, business, and scientific research areas that benefit those which supported sustainable plastics. In the future, biodegradable plastic introduced to a larger area in the modern environmental movement and manufacturing industry. So, this review emphasized the current potential of film synthesis techniques and their effects on the functional properties of biodegradable film.



2. Recent advances in different fabrication techniques

2.1 Solvent casting method

Solvent casting (SCM) is one of the traditional, popular, and lab-scale methods as well as an industrially applicable technique to fabricate bio-composite film. Although SCM is a very old technique, currently multilayer and electrospinning are widely used techniques to fabricate polysaccharide-based film.

Solvent casting is a more appealing technique to synthesize biopolymer-based films with exceptionally high-quality requirements. The key fundaments of this synthesis method included the bio-polymer properly dissolved into the volatile solution, the viscous solution should be obtained with minimum solid content after homogenization and the formation of the film easily release from the casting support (Lizundia *et al*., 2020) [9]. The advantages of the solvent casting method included thickness distribution with uniformity, high optical density, provides better quality than extrusion. However, this method leads to more disadvantages over advantages including a long time for drying, film Shrinkage after drying, brittleness, non-uniform drug distribution, and various storage conditions which are not desirable for the film. The “rippling effect” is one of the disadvantages of this method which leads to the problem of shear stress of the film and also leads to the lack of homogeneity. Sometimes over-drying is one of the causes of bubble formation which is also known as “nucleate boiling” (Lizundia *et al*., 2020) [9]. To overcome these drawbacks, plasticizers can be added according to polymer solution which reduced the cohesive intermolecular forces into the polymer networks which resulted in reduced tensile strength and induced flexibility over the films (Tan *et al*., 2022) [11].

2.2 Electrospinning or electrodynamic method

The electrospinning technique is an industrial technique that recently gained popularity among all fabrication techniques. This method depends on several parameters including temperature, the effect of the voltage applied, the effect of solution flow rate, the effect of temperature, the concentration of polymer, the viscosity of the solution, the distance between the needle to the collector, and the diameter of the needle (Yadav *et al*., 2019) [12]. This technique required a syringe that acts as a pump, high electric voltage with a positive electrode, and ground a collector connected to the ground or second electrode. The beginning of the electrospinning phase by applying electrical charges to the polymer solution through the syringe needle, triggering charges in turn into a conical shape (known as Taylor's cone) as the electric field increases (Li *et al*., 2021) [13]. However, at the endpoint, ultrafine nanofibers emerge from the needle in the shape of a cone and are collected by a spinning collector. The electric charges provide an unstable condition to the liquid medium. A reciprocal repulsion force opposes a surface tension at the same time allowing the liquid solution to flow toward the electric field. The solution droplet changes into a spherical jet shape which makes this technique more favorable over the solvent casting method and phase immersion method due to its extreme surface area to volume ratio as well as a maximum number of inter/intra fibrous pores (Yadav *et al*., 2019) [12]. Electrospinning is a time-consuming technique that takes more than 24 hours along with drying for only fabrication and the dried product is obtained as film. This method is more advantageous than the solvent casting method. As compared to solvent casting, obtained film fabricated under the electrospinning method was observed with better properties including a smooth surface, improved flexibility, viscoelastic behavior, and moderately strong (Fei *et al*., 2017) [15].

2.3 Phase immersion casting technique

Phase inversion casting is a mostly introduced fabrication technique used as a coating as well as for bio-film formulation. In the phase immersion method, the liquid (polymer and solvent) solution is mainly transformed into a solid state due to the molecular exchange of solvent and non-solvent components and precipitation takes place which resulted in obtained film after washing and drying (Gao *et al*., 2023) [16]. The phenomenon in the entire process is known as “liquid-liquid demixing”. Liquid-liquid demixing factors provide a significant impact on the properties of the obtained solid film such as the porosity of the protective film can be controlled by this method (Kim *et al*., 2023) [17]. The advantages of this technique such as low-cost energy, less time required for film formation, and ease of operation, as well as this method also offers a good potential for scale-up in future applications.

2.4 Layer-by-layer assembly techniques

Layer-by-layer assembly technique is another popular technique to fabricate film which is also a simple, popular, and industrially acceptable method that depends on several factors including electrostatic interactions, H-H bonding, covalent bonding, non-hydrophilic attraction, or entanglements between molecules (Zhang *et al*., 2019) [18]. The interactions of polymers depend upon the behavior of substances that helps to produce films. Generally, this process includes the sequential deposition of numerous film-forming materials onto a surface to form layered films or coatings. This is one of the most popular techniques to fabricate the film by using electrostatic interaction between positive and negative layers (Tian *et al*., 2019) [19]. This technique immerses a substrate which leads to a negatively charged solution having positively charged substances, causing these substances to be attracted to the surface of the substrate. Following the removal of the excess solution, the positively charged substrate immersed inside another solution leads to negatively charged substances, creating a new layer with the help of these substances. This process can be repeated several times to create layer-by-layer films (Zhang *et al*., 2019; Lipton *et al*., 2020) [18, 20]. Layer-by-layer technique leads with a range of potential advantages over other fabrication methods such as film developed by altering their physio-chemical and structural characteristics (strength, thickness) by selecting different types and numbers of film-forming material. The technique is non-complicated and inexpensive and does not require any specialized equipment (Lipton *et al*., 2020) [20].

2.5 Extrusion technique

Co-extrusion technique was designed for making multilayer films that have more than two extruders and feeding with a common die (Scarfato *et al*., 2017) [21]. The number of extruders depends upon the number of different materials comprising the coextruded film. Three-layer co-extrusion consists of a barrier core and requires only two extruders for the formation of two layers (Dziadowiec *et al*., 2023) [22]. The film extrusion die is attached to the adapter. A good die ensures complete melt flow with smoothness and clarity. Most of film extrusion dies are divided into heating zones and die heaters which are automatically controlled (Morris *et al*., 2022) [23]. In blown film extrusion, the melt enters the round die through the bottom. The melt is passing through the spiral grooves around the surface of the mandrel which are present inside the die and extruded through the circular die. The melt distribution can be improved by increasing the number of spiral grooves (Ghosh *et al*., 2016) [24]. Most of the blown film dies for polyethylene material which vertically positioned to push the tube upward and the dies extruded downward. The gap between the mandrel and the die ring ranges from 0.5 to 3 mm (Li *et al*., 2020) [25]. In some dies, the opening system can be changed by moving the mandrel lengthwise in the die. The opening of the die can be changed by moving the mandrel lengthwise in a die. Most of the die requires changes in the mandrels by adjusting the gap of the die. A wider ring increases the output easily, but it may make the gauge and frost line which makes more difficult due to the uneven flow. It is also a trend to promote film while the film is drawn down to a gauge of fewer than 0.5 mils (Li *et al*., 2020; Chachlioutaki *et al*., 2020) [25, 26].

2.5.1 Cast film and blown film extrusion

There are two basic methods currently used in industries for making biodegradable packaging such as cast film extrusion and blown film extrusion. In these methods, firstly the resin is melted by heat and creates pressure inside the barrel of an extruder, and provides force to the melt material through the narrow slit (the slit may be straight or circular in nature) in a die. The resulting thin film forms a sheet or tube which is generally called as blown film (Ghosh *et al*., 2016; Li *et al*., 2020) [24, 25]. The prepared film comes out of the die, gets cooled, and then rolled up on a core. There are some conditions that are directly related to film properties such as temperature, cooling, screw speed, gauge control, and blow-up ratio. The barrel heat temperature in cast film extrusion is generally higher than the blown film extrusion (140 to 190 °C) (Li *et al*., 2020) [25]. It is very vital to keep melt temperature should be uniform across the die width. Screw speed control is one of the major machine factors for thin film extrusion (Phothisarattana *et al*., 2022) [27]. However, to form the best film with desirable properties, the highest extrusion temperature permits film cooling as well as the maintenance of uniform flat film. The higher speed of the screw without pressure and temperature provides flat film machine operation and comparatively better output from the extruder. Although there are no connections between the die and film gauge in the thin film forming film gauges generally require large openings such as for film (around 20 mils or 0.5 mm). The hopper needs to be kept completely full of resin while the extruder is working (Heremans *et al*., 2016) [28]. The type of packaging material being extruded and whether blown or flat film is being produced have a significant impact on screw speed, which is measured in revolutions per minute (rpm) (Li *et al*., 2020) [25]. Generally, the amount of internal heat and output produced increases with the increase of screw speed. Less external heating is required when there is more interior heat.

3. Different fabrication techniques affects film properties

The physical properties of ternary blend-based films have recently been improved through physical and chemical alterations, which appear to be quite successful. Tensile strength (TS), water vapour permeability (WVP), water uptake ratio (WUR), barrier qualities, and solubility ratio are among the main physical and mechanical characteristics of the packaging film. The functional properties of biodegradable film such as thermal, optical, mechanical, and structural properties are influenced by the conformation mode of polymeric chains whereas film leads with unorganized structure and their chain interactions closely related to the degree of space-occupancy by the polymeric chain coils (Mir *et al*., 2018) [29]. On the other side, if those exist with organized form, then they can be capable of stable association into compact chain networks. However, the structural confirmation of the polymeric chain is almost prominent in the area of the kinetics of water solubility, water vapour permeability, and tensile strength formed by films by such biopolymers (Basiak *et al*., 2018) [30]. The previous study investigated the electrospinning method over the solvent-casting method for the synthesis of polymeric sheets with increased plasticity and flexibility. The systematic evaluation hypothesizes that the mechanical properties and flexibility of electrospinning polymer fiber can be influenced by fabrication techniques (Ghosal *et al*., 2018) [31]. The morphological structures of an average biopolymer-based film from solvent casting, developed with irregular surface morphology characterized by the absence of fibers and the presence of crystals due to the slow evaporation process. In contrast, electrospinning provided membranes with aligned and non-aligned nano-sized fibers (Deshmukh *et al*., 2022) [32]. Previous research emphasizes that the mechanical properties of electrospinning nanofibers depend on the direction of the fiber alignment within the membrane. Electrospun random membranes collected from static collectors have lower tensile strength while the rotating mandrel as a collector produced random membranes with higher tensile strength (Ghosal *et al*., 2018) [31]. The previous study concluded that most of the research enhances the plasticity of the film by using the electrospinning technique (Kargarzadeh *et al*., 2018) [33]. Some literature revealed the mechanical properties of polymeric films which are modified by many folds via the electrospinning process. However, the electrospinning process enhances flexibility, smoothness, softness, and moderately strong thin polymer films. In contrast, membranes are cast in a solvent and have semi-crystalline domains that are flexible in an electrospun matrix but brittle and rigid in a solvent-casting film (Ghosal *et al*., 2018) [31].

During the extrusion technique, the starch granules of the starch-based biofilm were molten and physically broken down into small fragments under the action of shear and thermal energy. The film developed with the extrusion technique was uniform and not completely transparent (Pimparade *et al*., 2017) [34]. Previous studies concluded that starch-based extruded film developed with high flexibility without brittleness after drying. However, the higher temperature of the extrusion technique must affect the physical properties of the film (Choi *et al*., 2023) [35]. As per previous literature, the thickness of the film reduced with the induced temperature because of the viscosity of the plasticizer melt and biopolymer, indirectly sensed by the higher torque requirement and higher temperature at 110 °C and 120 °C (Coltelli *et al*., 2020) [36]. In the case of the glass transition temperature of the extruded film reduces the intermolecular hydrogen bonding between the chains due to the increased mobility of polymer chains, facilitating an easy transition from a glassy to a rubbery state (Harnkarnsujarit *et al*., 2017) [37]. On the other hand, the water vapour permeability (WVP) of the extruded films was induced and the permeation rate was affected by the extrusion temperature and the concentration ratio of plasticizers (Dang *et al*., 2021) [38].

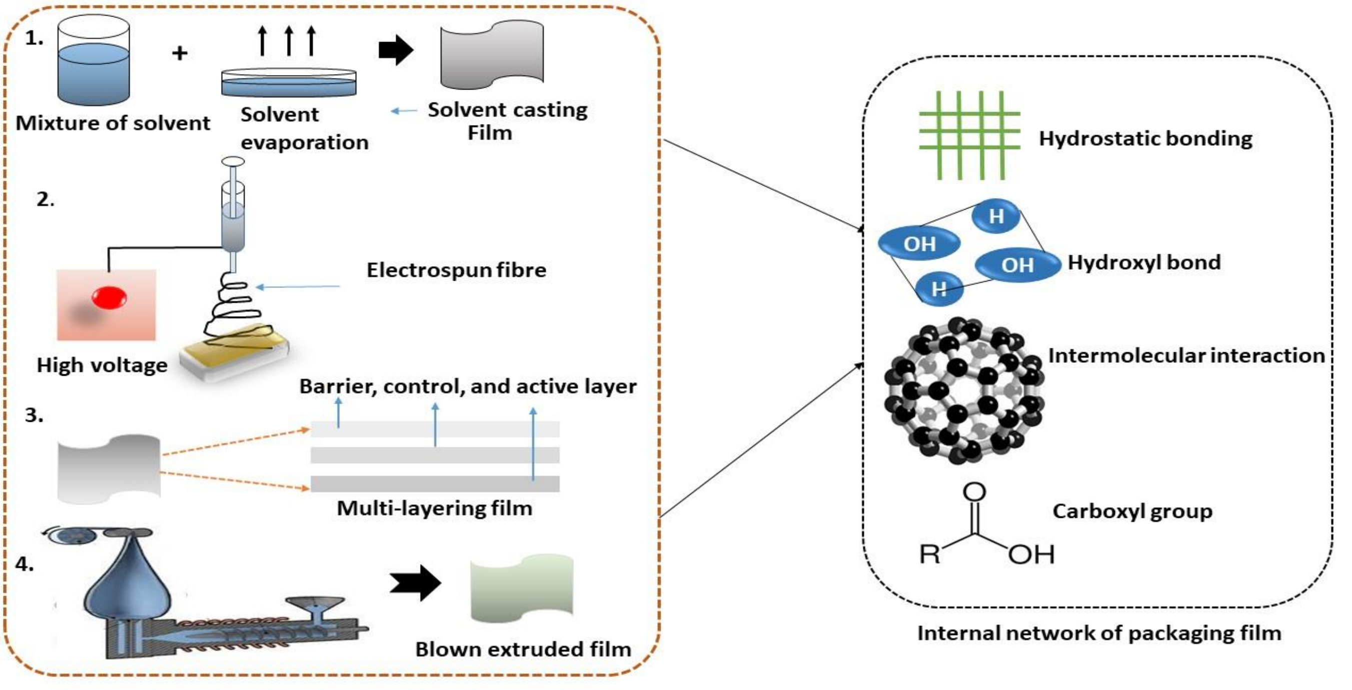


Fig 1: Different advanced film synthesis techniques and their effect on internal properties

Table 1: Film properties affected by biopolymers and fabrication techniques

Film formulation	Fabrication technique	Physio-mechanical properties	References
Chitosan/Konjac glucomannan/tragacanth gum	Tri layer film Solvent casting film	Good oxygen and water barrier property Excellent thermal Stability and UV- shielding Properties great degradability	(Mu *et al*., 2023) [39]
Alginate/Carboxy methyl Cellulose/ starch	Ternary blend Solvent casting method	Developed film with smooth surface higher tensile strength and decreased elongation break Increased UV, oxygen, and water vapour permeability	(Ramakrishnan *et al*., 2023) [40]
Guar gum/ chitosan	Binary blend Solution casting method	Developed with higher contact angle and hydrophobic in nature Developed with lower Water vapour Permeability without Cross -linked solution Water-repellent nature	(Rahman *et al*., 2021) [41]
PVA/ starch/ pectin	Ternary blend Solution casting Method	Increased biodegradability improve the tensile strength, elongation break, and provides sustainable packaging properties	(Lal *et al*., 2020) [42]
Chitosan/ GG/ polyvinyl alcohol	Ternary blend Solution casting Method	Swelling properties shows hydrophilic nature of the film shows excellent drug releasing properties film membranes are structurally developed with porosity.	(Iqbal *et al*., 2019) [43]
Starch/ LDPE/ chitin	Ternary blend Shear Deformation in a Rotor disperser	Enhanced Biodegradability high mechanical characteristics increased crystallinity	(Rogovina *et al*., 2015) [4]

4. Future trends and Conclusion

The design and production of biodegradable packaging is a currently growing interest to replace conventional plastics to reduce the negative impacts on the environment associated with their fabrication technique and disposability. This review focused on the recent advanced techniques to fabricate biopolymer-based film networks and how the technology affects film properties. Additionally, the basic mechanisms which controlled the released packaging and the positive impact of the synthesis techniques on the mechanical and other properties of films and coatings are reviewed.

Currently, biodegradable packaging has been successfully designed, characterized, and applied in research laboratories. Moreover, many factors are still necessary to be addressed before the widespread commercial success of the packaging film, including demonstrating their efficacy in real-life applications, developing large-scale economic manufacturing processes, and establishing appropriate government regulations to gain consumer trust and acceptance.

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