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Legume seeds for the development of biodegradable food packaging film: A review

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Abstract

Food packaging plays a crucial role in any food processing organization, and the increasing awareness of consumers about the impact of packaging material on food quality and the environment makes the packaging material even more significant. The development of novel biopolymers with improved functioning, gaseous barrier abilities, and prolonged shelf life, with minimal impact on the environment, will be the ultimate solution for food packaging industry. Underutilized legumes, which are the forgotten gems embedded with high nutritional packages and their cost effectiveness, can be an alternative to synthetic polymers. The application of legumes as edible coatings can significantly enhances their economic value. Edible coatings and films derived from legumes can improve the keeping quality of food products by serving as barriers to oxygen, moisture, and solute transfer. Further incorporation of bioactive components such as antimicrobials and antioxidants can enhance the functionality of these films. The development of such eco-friendly packaging solutions aligns with growing consumer demand. Furthermore, the use of underutilized legumes in packaging applications could stimulate local agricultural economies and promote crop diversification.

Keywords: Legumes, biopolymer, food, packaging, biodegradable, shelf-life

Introduction

The use of synthetic polymers to pack and carry goods has revolutionized the packaging industry worldwide. Currently, dependency on polymer packaging materials has become inevitable, primarily because of their high versatility and price. According to the OECD (2022) [25], approximately 40% of global plastic waste is attributed to packaging, with a substantial portion originating from the three regions that produce the most plastic waste: the United States, Europe, and China. Hence, it is worrying that they affect human existence and the environment, contributing significantly to the issue of single-use plastics because of their difficulty in reuse and recycling. There has been a notable increase in the research exploration of new environmentally friendly packaging materials, which are intended to serve as alternatives to traditional synthetic plastics (Liminana et al., 2018) [18].

Microplastics, formed by the breakdown of larger plastic items, have been found in various food chains and human blood, raising concerns about their potential health impacts. There is growing interest in the creation of biodegradable packaging materials. These materials are made from renewable resources, such as plant-based natural polyfibers. Biodegradable polymeric films have been found to be suitable alternatives and have received worldwide attention because they are abundantly available in nature and can be

extracted from renewable resources. Biodegradable films can be prepared from proteins, lipids, and polysaccharides, which can be extracted from various renewable resources such as agricultural waste, seaweed, and underutilized crops (Zhang *et al.*, 2016) [47].

Numerous scholarly articles have been published on the application of proteins as packaging materials, highlighting the significant interest in utilizing proteins for the production of biodegradable films. Protein polymers are easy to handle, and films derived from proteins are distinguished by their non-toxic nature and excellent barrier properties against flavorings, oxygen, and lipids (Rojas-Lemaetal., 2021) [30]. These films have also been utilized as carriers to deliver antioxidant- and antimicrobial-rich formulations (Purewal et al., 2023) [29]. Protein films can also supplement the nutritional value of foods. (Gennadios & Weller, 1990) [9]. These films exhibit better mechanical properties than polysaccharide- and fat-based films because of their high intermolecular binding capacity, which can form strong bonds at numerous linkages (Bourtoom, 2009) [2]. However, protein films have several drawbacks compared to their synthetic counterparts, such as poor mechanical properties, poor barrier properties, and high moisture sensitivity due to their hydrophilic nature. Researchers have developed various methods to improve the mechanical strength of protein films by incorporating

reinforcing materials such as nanoparticles, crosslinking polymer chains (Fan *et al.*, 2022) [8], combining them with hydrophobic polymers (Shellhammer & Krochta, 1997) [38], or using physical methods such as irradiation treatment (Lacroix *et al.*, 2002) [15].

Legume grains are frequently referred to as "the poor man's meat," as most of the population relies heavily on them to fulfill vegan protein requirements (Roy et al., 2017) [32]. The Fabaceae or Leguminosae family encompasses 800 genera and 20,000 species, making it the third-largest family in this category. Traditionally, cultivated in specific regions by indigenous communities and they have not received the research and commercial attention to same extent as that of major crops like soy and common beans. The legumes included Vignaradiata (mung bean), Macrotyloma uniflorum (horse gram), Psophocarpus tetragonolobus (winged bean), Vigna subterranean (Bambara groundnut), Canavalia gladiate (sword bean), Canavalia ensiformis (jack bean), and viciafaba (broad bean/faba bean). However, some legumes are considered weeds in cereal crops. whereas others are significant grain crops (Stagnari et al., 2017) [41]. In a global effort to combat hunger and malnutrition, these underutilized legumes are highly advantageous because of their resilience and hardiness in adverse climates. Their enhanced resistance to climate change and disease positions has made them a potentially transformative crop for the successive green revolution in agriculture. Underutilized legumes are a nutritious and sustainable food source that can enhance food security. stimulate economic growth, and alleviate poverty in various regions worldwide.

Despite their nutritional value (as a source of fiber, complex carbohydrates, protein, and vitamins), suitability as a crude material for the development of bio-based products, and socioeconomic advantages (Rojas-Lema *et al.*, 2021) [30]. Many leguminous plants are underutilized, mainly because of their high dependency on a few popular legume varieties in the global market (Samal *et al.*, 2023) [35]. The main objective of the present study was to identify underutilized leguminous plants and review the different legumes used in the manufacturing of biodegradable packaging films.

Underutilized leguminous crops

The legume family, also referred to as Fabaceae or Leguminaceae, ranks third globally in terms of total population. Legumes are cultivated across a diverse range of environments, including deserts, woodlands, alpine regions, aquatic areas, and African rainforests in the Amazon (Samal *et al.*, 2023) [35]. Despite their resilience to various climates and the nutritional benefits they offer, such as increased protein and essential nutrients, only a limited number of legumes, including peas, soybeans, and certain types of beans, are widely consumed. Approximately 700 genera and 20,000 species have the potential to be developed and integrated into mainstream agriculture for domestication and human utilization (Minde *et al.*, 2021) [21].

Legume based packaging material Faba Beans

faba bean (*Vicia faba* L.) is distinguished by its affordability and rich in protein content, which can reach up to 28% protein (dry weight). This makes it a good raw material

source for protein extraction, particularly for use in films in the food packaging industry (Samaei *et al.*, 2020) ^[34]. Faba bean protein-based films typically exhibit reduced thickness; however, the incorporation of glycerol enhances their tensile strength and water-vapor barrier properties. Hopkins *et al.* (2019) ^[11] reported that faba bean protein films with a 50% glycerol content demonstrated superior mechanical strength and opacity (Table 1), as well as reduced water permeability, compared to protein films derived from other legumes such as lupins, peas, soybeans, and lentils.

Montalvo-Paquini et al. (2014) [23] analyzed the impact of varying pH levels and plasticizer content in film-forming solutions on the physical and chemical properties of faba bean protein films. Their findings indicated that alterations in pH did not influence film thickness. Nonetheless, at an alkaline pH, the water vapor permeability diminished. In a similar study, Saremnezhad et al. (2011) [36] showed that the tensile strength increased at pH 12 in films based on faba bean protein with 40% glycerol, whereas water vapor permeability declined under the same conditions. In addition, films based on faba bean protein isolate with carrageenan achieved better compatibility between macromolecules through pH modification and denaturation temperature of the protein, allowing its reactive sites to interact with carrageenan and form stable structures (Rojas et al., 2021) [30]. These studies highlight the potential of faba bean protein films as promising alternatives to other legume-based films in packaging applications. Manipulation of pH and glycerol content appears to be crucial for optimizing the physical and chemical properties of these films. Furthermore, incorporation of carrageenan and strategic modification of the protein structure can lead to enhanced film stability and performance.

Pea

Pea proteins and starch can be used to develop edible packaging (Van Soest et al. 2002) [44]. Pea proteins are primarily globulins with a small proportion of albumins. Globulins constitute approximately 80% of the total proteins in peas and are storage proteins found in the cotyledons (Choi and Han 2001) [5]. Choi and Han (2001, 2002) [5, 6] established that films produced from both pea-protein concentrate (PPC) and pea-protein isolate (PPI), which had not undergone heat treatment, showed considerably lower tensile strengths compared to those subjected to heat treatment at 90°C for 25 min and durations ranging from 5 to 50 min, respectively. The observed increase in tensile strength can be attributed to the higher concentration of PPC and lower concentration of glycerol utilized (Table 1). The tensile elongation of the pea protein films was influenced by the heat treatment. Choi and Han (2002) [6] investigated the impact of heat treatment duration (0-50 min) at 90°C on the tensile elongation of PPI films and found that tensile elongation was increased after 20 min of heat treatment, indicating that some degree of heat treatment is essential for elongation. Additionally, pH has been shown to significantly affect tensile elongation; in pea protein films examined at pH levels of 7, 9, and 11, tensile elongation was maximum in films formed at pH 11 and lowest at pH 7 (Kowalczyk, Gustaw, Świeca, & Baraniak, 2014) [13]. Giosafatto et al. (2018) [10] used grass pea (Lathyrus sativus

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L.) flour as a biopolymer source to develop edible packaging films. The protein was structurally modified using microbial transglutaminase, an enzyme capable of catalyzing isopeptide bonds between glutamine and lysine. In their study, optical analysis demonstrated that films cast in the presence of microbial enzymes exhibited greater transparency than untreated films. Scanning electron microscopy further revealed that the enzyme-modified films possessed a more compact and homogeneous structure. Additionally. the incorporation of transglutaminase facilitates the production of films with enhanced mechanical resistance. The enzyme-modified films also displayed improved barrier properties with reduced water vapor permeability compared to the untreated films. This improvement in the film characteristics can be attributed to the cross-linking effect of microbial transglutaminase, which fortifies the protein network within the film matrix. These findings suggest that the application of microbial transglutaminase in edible packaging film production may result in more durable and effective food packaging solutions.

Winged bean

Winged bean (Psophocarpus tetragonolobus), also referred to as Goa bean, Asparagus pea, and winged pea, is commonly known as the 'One Species Supermarket' due to its utility as a green pods, leaves, tuberous roots, and both immature and mature seeds. The leaves are consumed similarly to spinach, the tubers are eaten either raw or processed, and the seeds are utilized in various forms of processed food. Poeloengasih and Marseno (2003) [28] developed edible films using the protein fractions of winged bean seeds. Their findings indicated that increasing the concentration of the protein fraction of winged bean seeds resulted in a decrease in the water vapor transmission rate and elongation while increasing the thickness and tensile strength. Conversely, increasing the concentration of palmitic acid decreased the water vapor transmission rate and tensile strength. The optimized film, comprising 1% (w/v) tapioca, 2.5% (w/v) protein fraction of winged bean seeds, 1% (w/v) sorbitol, and 8% (w/w polymer) palmitic acid, was used to wrap fresh-cut apples, demonstrating a significant effect on reducing weight loss and browning during storage. Sian and Ishak (1990) [40] investigated the impact of various pH levels on the formation and rehydration capacity of winged bean and soybean proteinlipid films. They discovered that film formation was feasible only within a definite pH range. Films prepared at higher pH values between 7.5 and 11 contained higher proportions of proteins, carbohydrates, and ash, but lower proportions of fat, compared to those prepared at pH between 6.7 and 2.0. The winged and soybean films exhibited distinct rehydration patterns; however, both tended to absorb more water at higher pH values. In terms of film formation efficiency, protein content, and rehydration capacity, the soybean film was found to be superior to the winged bean film.

Soybean

Soybean-derived packaging films are typically manufactured using soymilk, a water-based soybean extract. These films are generally produced from soy protein isolates (SPIs) and their structures are established through disulfide,

hydrophobic, and hydrogen bonding. Research has indicated that SPI packaging films are effective barriers to oxygen and lipid permeation. However, their hydrophilic nature renders them less effective at preventing water passage (Park et al. 2013) [26]. SPI is primarily used to create film-forming solutions (Prakash and Mishra 2023). PIAs the most prevalent plant protein, is a refined product obtained from soybean oil extraction and consists of a mixture of albumins and globulins (F. Hassan, Mu, and Yang 2024). Additionally, SPI demonstrates significant stability under high-temperature and humid (F. Hassan, Mu, and Yang 2024). Its gel-forming capability is beneficial for developing suitable structures for composite films that include lipids and bioactive substances such as antioxidants and antimicrobial agents (A. Kumar, Hasan, et al. 2022). Various methods have been explored to enhance the water barrier properties of SPI films, including the incorporation of hydrophobic compounds and cross-linking agents. These modifications were aimed at decreasing the hydrophilicity of the films and improving their moisture resistance. Researchers have also examined the use of nanocomposites and blending them with other biopolymers to further boost the overall performance of SPI-based films.

Cho et al. (2010) [4] developed an edible film pouch designed to serve as an oxygen barrier by laminating a heatsealable corn zein layer onto a soy protein isolate film, specifically for packaging olive oil condiments with instant noodles. This study evaluated the mechanical, barrier, and physical properties of corn zein/sov protein isolate bilayer films and assessed the oxidative stability of olive oil stored in these pouches under dry and moderately humid conditions. The addition of a corn zein film layer to the soy protein isolate film enhanced the tensile strength and water resistance, although it resulted in decreased elongation at break and reduced oxygen-barrier properties. The superior oxygen barrier capability of the soy protein isolate bilayer films contributed to a reduction in the oxidative rancidity of olive oil packaged in these films compared with those packaged in NY/mLLDPE films. Li et al. (2021) [17] investigated the effects of varying concentrations of ferulic acid between 0 and 8.0% w/w) as both an antioxidant and a cross-linking agent in soy protein isolate/cellulose nanofiber/ferulic acid composite films. The mechanical and barrier properties and antioxidant activity of these films were assessed using scanning electron microscopy, Fouriertransform infrared spectroscopy, and X-ray diffraction. The results indicated that films containing 4.0% ferulic acid exhibited reductions in elongation at break, oxygen permeability, and water vapor permeability of 69.76%, 77.55%, and 90.91%, respectively. Sun *et al.* (2024) [42] examined soy protein isolate films incorporating quercetingrafted dialdehyde starches. The physical, structural, and functional characteristics of the prepared composite films were evaluated. These findings demonstrated that quercetingrafted dialdehyde starch formed hydrogen bonds with the SPI matrix, thereby enhancing the structural properties of the films. The incorporation of quercetin-grafted dialdehyde starch improved the light-blocking ability, hydrophobicity, tensile strength, thermal stability, and elongation at break as well as the antioxidant and antibacterial properties of the films.

Mung Bean

Mung beans (Vigna radiata L.) contain approximately 25-28% protein and are a source of bioactive compounds such as flavonoids, phenolic acids, organic acids, and polysaccharides (Moghadam *et al.* 2020) [22]. It is a starchrich food, and mung bean starch (MBS) has a high amylose content ranging from 30-45% (Deshmukh *et al.* 2022) [7]. MBS forms dense and cohesive film networks because of its high amylose content. This high amylose content contributes to the formation of a transparent dried product, distinguishing it from the other starches.

The linear structure of amylose promotes interchain hydrogen bonding more effectively than the branched structure of amylopectin. The relatively linear and long backbone of amylose chains, combined with the close proximity of starch chains with high amylose content, supports the development of cohesive film networks during the drying process. Consequently, films made from amylose are denser and stronger than those made from amylopectin (Rompothi *et al.* 2017) [31]. Films containing higher levels of amylose typically exhibit a higher glass transition temperature, tensile strength, and modulus of elasticity while showing lower elongation compared to films with lower amylose content.

Cheappimolchai et al. (2000) [3] assessed the feasibility of using mung bean protein as a base material for the production of biodegradable films in comparison to soy protein. The protein from mungbean seeds was extracted through acid precipitation at pH 4.5, resulting in an isolated mungbean protein with a content of approximately 82%. Films were prepared by dissolving either mung bean or soybean proteins in water at pH 7.0 and casting the solution onto a Teflon plate. This study investigated the impact of varying the glycerol concentration on three physical properties of the film: percentage elongation, tensile strength, and water vapor permeability. The findings indicated that as the glycerol concentration increased, the tensile strength of the films decreased, whereas both the percentage elongation and water vapor permeability increased (Table 1). The authors concluded that mungbean protein was a viable raw material for the production of biodegradable films.

Roy et al. (2020) [33] investigated the starch derived from mung beans, focusing on the production and properties of nano-starch. The size of the nano-starch particles was 141.772 nm (average). Mungbean starch granules were smooth and oval, without cracks, whereas nano-starch exhibited aggregation and a rough surface. Films were formed with varying concentrations of nano starch (0.5 to 10%) using a solution casting method. The native starch film had average thickness, moisture content of 0.040 mm, 8.03%, and $5.982 \times 10-3$ g/s, respectively, while water solubility and burst strength were 38.49%, and 868.49 g, respectively. The addition of 0.5 to 10.0% nano starch improved the film thickness from 0.043 to 0.063mm, burst strength from 943.56 to 1265 g, and moisture content 6.09 to 4.80%, water vapor transmission rate from $5.558 \times 10-3$ to $3.364 \times 10-3$ g/s), and solubility from 37.99 to 34.11.

Yao *et al.* (2022) ^[45] conducted an investigated into the hydrophilicity of mungbean starch films using the natural cross-linker citric acid at concentrations from 1to15% to assess the physicochemical and barrier properties of

mungbean starch films. Compared to mungbean starch films without citric acid, the incorporation of citric acid at levels of 1-3% resulted in increased Young's modulus, tensile strength, and heat seal strength, whereas water solubility, water contact angle, break elongation, transparency, water vapor permeability, and oxygen permeability decreased. However, further increases in citric acid concentration did not yield additional improvements. The FTIR spectra revealed an absorption peak at 1721 per cm for citric acid-cross-linked mungbean starch films, confirming the formation of an ester carbonyl bond between the hydroxyl and carboxylic groups. Overall, cross-linking with 1-3% citric acid reduced the hydrophilicity and enhanced the barrier properties of mungbean starch films.

Kidney beans

Kidney beans, which contain 20%-30% protein, are an important bean among the different beans (Shevkani and Singh, 2015) [39]. On a dry weight basis, kidney beans usually contain 20-30% protein (Sathe, 2002) [37]. Recently, there has been considerable interest in the storage proteins of this bean because of their exceptional properties, such as emulsification, protein solubility, and ability to form gels when exposed to heat (Ma et al. 2013) [20]. In 2013, Ma et al. developed a peelable and self-supporting film using kidney bean protein isolate and chitosan. The kidney bean protein film alone demonstrated a typical stiffness. However, the combination of kidney bean protein and chitosan resulted in films that were less rigid and significantly more flexible. characterized by reduced elastic modulus, storage modulus, and glass transition temperature. The addition of chitosan improves the surface hydrophobicity of the films. Tang et al. (2009) investigated the film-forming properties of kidney bean protein isolates at a neutral pH. Their findings indicated that chitosan films exhibited considerably lower tensile strength and elongation at break than soy protein films.

Locust Bean Gums (LBGs)

Locust bean gum (LBG), derived from the seeds of the carob tree (Ceratonia siliqua), is a galactomannan polysaccharide known for its excellent film-forming, biodegradable, and rheological properties, making it a promising candidate for biodegradable packaging applications (Yuan et al., 2020; Petitjean and Isasi, 2022) [46, ^{27]}. Its potential as a packaging material arises from its ability to form films independently or in combination with other biopolymers, such as proteins, lipids, and polysaccharides (Yuan *et al.*, 2020) [46]. These LBG-based films can enhance the shelf life and safety of food products owing to their barrier properties against gases and moisture and can also act as edible coatings (Yuan et al., 2020) [46]. Inherent physicochemical properties of LBG, such as its solubility and rheological behavior in aqueous solutions, coupled with its ability to synergize with other polymers, further extends its functionality and application range. This can lead to enhanced mechanical properties and improved performance, offering a viable alternative to conventional petroleum-based packaging materials (Petitjean & Isasi, 2022) [27]. Furthermore, the polysaccharide nature of locust bean gum ensures that the products manufactured using it are both biodegradable and non-toxic, aligning with the

growing demand for environmentally friendly and sustainable packaging solutions (Tukenmez Emre *et al.*, 2025) [43]. The most significant characteristic of this polysaccharide polymer is its ability to exhibit high viscosity in aqueous solutions over a wide range of pH levels and temperatures (Aydinli and Tutas 2000) [11]. The exceptional biodegradable nature, rheological attributes, and film-forming capabilities of LBG make it well suited for packaging film development (Yuan *et al.* 2020) [46]. It can create viscous solutions, even at low concentrations, and generate compact and dense films and coatings with favorable mechanical strength and water vapor barrier characteristics (Mostafavi *et al.* 2016) [24].

In practical applications, LBG can be chemically modified to improve its properties. For example, carboxymethylation of LBG enhances its swelling properties and

biodegradability, and when combined with polyvinyl alcohol, it can create interpenetrating polymer networks that are useful for controlled drug delivery (Kaity & Ghosh, 2013) [12]. This modification demonstrates versatility and potential of LBG for broader applications, including biodegradable packaging.

Moreover, blending LBG with other gums, such as gellan gum, to form double-network hydrogels can impart benefits such as self-healing and pH-responsive shape memory properties, making such materials attractive for an array of applications beyond packaging, including biomedicine and soft robotics (Lv *et al.*, 2019)^[19]. Overall, LBG's versatility, biodegradability, and safety make it an excellent choice for developing innovative and sustainable packaging materials that can mitigate the environmental impact associated with traditional packaging methods.

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Legume films	Additives	Tensile strength	WVP	Reference
Faba beans	50% glycerol	10.0 MPa	0.56 g.mm/m ² .h.k.Pa	Hopkins et al., 2019 [11]
Pea protein	50% glycerol	8.00 Mpa	-	Hopkins et al., 2019 [11]
Pea protein concentrate (heat denatured film)	50% glycerol	0.692 ± 0.073Mpa	7.42±0.69 g.mm/m².h.k.Pa	Choi & Han, 2001 [5]
Pea protein concentrate (Native film)	50% glycerol	0.317±0.024Mpa	-	Choi & Han, 2001 [5]
Soy protein	50% glycerol	7.4 Mpa	-	Hopkins et al., 2019 [11]
Soya protein isolate	1.5% glycerol W/V	$7.36 \pm 0.42 \text{ Mpa}$	4.39 ± 0.59 g.mm/m ² .h.k.Pa	Kunte et al., 1997 [14]
Mung bean	1% glycerol W/W	0.466 Mpa	0.810 ×10 ⁻¹⁰ g/m.s.Pa	Cheappimolchai et al., 2000 [3]
Kidney bean protein isolate	4% glycerol w/w	7.2 MPa	4.3-4.7 g.mm/m2.d.k.Pa	Shevkani & Singh, 2015 [39]
lentil protein film	50% glycerol	4.1 MPa	-	Hopkins <i>et al.</i> , 2019 [11]

Table 1: Physical and mechanical properties of legume based films

Conclusion

Legume-based biodegradable packaging demonstrate significant potential as sustainable alternatives for extending product shelf life and maintaining product quality. The rich composition of legumes, including proteins, polysaccharides, fibers, and bioactive compounds, contributes to the favorable mechanical and functional properties of packaging films. Utilizing underutilized legumes aligns with sustainability goals, promotes food security, and reduces environmental impacts. However, challenges persist, including limited research underutilized legumes, knowledge gaps in nutritional content and agronomic requirements, and policy and investment barriers that favor major crops. To fully leverage the potential of legume-based biodegradable packaging, future efforts should focus on increasing research, developing supportive policies, conducting comprehensive assessments, and exploring diverse applications for various product categories. Addressing these issues can contribute to the advancement of sustainable packaging solutions and promote the utilization of underutilized legumes in the food industry.

Conflict of Interest

The authors have declared that no competing interests exist.

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