## 1. Introduction

People often fail to realize that we live in a society where nearly everything is made from plastic-from the fabric of our clothes to our glasses and even the cars we drive. Most of this plastic is derived from oil, consuming 10% of the global oil supply, contributing to global warming, and taking over 1,000 years to degrade. The significance of the packaging system and its diverse functions has been on the rise in recent decades. COVID-19 has played a major role in economic development and food systems worldwide having a decisive impact on public health (Kochanska et al., 2021). The packaging industry has gained significant importance during the pandemic and is expected to continue growing in popularity among consumers in the near future. Food packaging plays a crucial role in ensuring product quality, safety, and shelf-life (Barbero, 2020). The primary purpose of food packaging is to safeguard products from alterations in their surroundings, particularly changes in gases, ambient temperature, and relative humidity. The significance of packaging functions continues to increase, leading to more innovative and creative packaging solutions. Owing to the increasing consumer demand for easy food management and prolonged shelf life in recent years (Ahmed et al., 2022; Rani et al., 2024), there has been a notable rise in the significance of scientific publications and research discussing innovative technologies in food packaging, particularly those associated with active and intelligent packaging (Wyrwa and Barska, 2017). Active packaging incorporates additives that interact with the food or its environment to extend quality and safety (Barska and Wyrwa, 2017). It can be categorized into non-migratory systems (example: oxygen scavengers) and active-release systems (example: antimicrobial packaging) (Yildrim et al., 2018). This technology represents a shift from passive to active protection, allowing intentional modification of the package environment to prolong shelflife and enhance food safety while maintaining quality (Janjarasskul and Suppakul, 2018). Recent advancements include the development of sustainable food packaging that focuses on using renewable, recyclable, or recycled materials while ensuring functionality and food safety (Nilsen-Nyygaard et al., 2021).

Biopolymers derived from plants and animals, including proteins, lipids, and polysaccharides, as well as other bioplastics generated by microbial synthesis, have been utilized to create environmentally acceptable food packaging materials that hold great promise for usage as carriers of functional chemicals (Niaounakis, 2015). Packaging materials derived from biopolymers possess unique characteristics such as abundant availability, renewability, and biodegradability (Mangaraj et al., 2019). Notably, there is a projected significant growth in the global market for bio-packaging materials in the upcoming years (Bajpai, 2019). The forecast indicates that the production capacity of biobased packaging is anticipated to increase from 2.11 million tons in 2019 to approximately 2.43 million tons by 2024 (Amir et al., 2022). Starch is a promising biopolymer for various applications, including bioplastics production, due to its abundance, renewability, and biodegradability (Gamage et al., 2022). It's composed of amylose and amylopectin, with amylopectin significantly influencing its properties. Starch's hydroxyl groups make it hydrophilic and modifiable, but also vulnerable to moisture (Kumari and Sit, 2023). Various starches (potato, cassava, corn, sago) have been studied for active packaging. Amylose content and crystallinity affect starch's properties and film-forming ability. Starch-based packaging can degrade in humid environments, which aids in its biodegradability but may limit some applications (Niranjana and Prashantha, 2018).

Proteins are considered biopolymers, as they are composed of monomeric units (amino acids) that are covalently bonded in chains to form larger molecules (Cozzone, 2002). Proteins as biopolymers have complex structural conformations that enhance their interaction and binding capabilities. This results in films with superior mechanical properties compared to other sources (Gupta and Nayak, 2015). Casein, in particular, is desirable due to its high nutritional value, water solubility, and emulsification capabilities for film formation (Khan et al., 2021). The distribution of polar amino acids in caseins creates excellent barriers against oxygen and nonpolar molecules, making them effective for protecting products susceptible to oxidation (Ranadheera et al., 2016). These properties make protein-based films, especially those derived from caseins, promising candidates for biodegradable packaging applications. Fiber-based packaging is an effective solution to reduce the environmental impact of plastic packaging. It aligns with circular economy principles due to its recyclability and biodegradability (Reichert et al., 2020). Cellulose, the most abundant natural and renewable polymer, is a viable alternative to fossil-based materials (Magalhaes et al., 2023). Natural fibers can be used as reinforcement in

composites, providing mechanical strength. While they have higher moisture absorption and limited thermal stability compared to synthetic fibers, their recyclability and degradability make them crucial for addressing environmental concerns (Elfaleh et al., 2023). Surface modification techniques show promise in mitigating these limitations, making fiber-based packaging an increasingly attractive option for sustainable packaging solutions (Ranadheera et al., 2016).

Biodegradable packaging materials can break down into environmentally friendly products like CO<sub>2</sub>, water, and compost (Roohi et al., 2018). Active packaging systems, developed since the 1980s, use plant and food extracts to create both active and biodegradable materials. Bio-based polymers have drawbacks compared to synthetic ones, such as humidity-dependent properties and potential microbial contamination (Stuppa et al., 2023). These limitations can be addressed by incorporating additives like plasticizers, antioxidants, and antimicrobial agents (Moeini et al., 2021). Essential oils are widely used in fruit and vegetable packaging to enhance preservation and control microbial growth (Perumal et al., 2022). Essential oils (EO) are natural substances, known as safe compounds by the Food and Drug Administration, that have stood out as excellent antibacterial and antifungal agents due to the presence of alcohols, phenols, terpenes, esters, and other bioactive compounds (Gavahian et al., 2020). They can improve structural attributes and physical properties when added to biopolymeric films. However, essential oils pose challenges such as strong odors, low solubility, high volatility, and potential impacts on food taste (Perumal et al., 2022). Despite these challenges, their incorporation into packaging materials can significantly enhance antimicrobial properties while minimizing direct contact with food (Khaneghah et al., 2018). The research centered on packaging for fresh fruits, particularly grapes, due to their short shelf life and the rising concerns about foodborne outbreaks. Fresh fruits have become a key target for active packaging research and commercial applications. While active packaging technologies offer numerous benefits in improving product quality and extending shelf life (Yildrim et al., 2018), this study specifically focused on developing and exploring the potential application of new packaging material for fresh grapes. This targeted approach aims to address the unique challenges associated with grape storage and safety.

Active food packaging, incorporating ingredients to enhance safety and shelf life, offers a promising solution to both food safety and waste reduction. This innovation aligns

with the Sustainable Development Goal of achieving zero hunger by ensuring safer food and reducing waste (Yildrim et al., 2018). Despite the rapid evolution of active packaging technologies, their commercial adoption remains limited, representing only about 1% of the overall packaging landscape in the United States (Barbero, 2020). However, recent publications emphasize the importance of active packaging for food safety during transportation and storage. While significant progress has been made in biobased polymer packaging, several research gaps remain to be addressed. The use of composite or blended biobased polymers and fiber reinforcements represents an emerging area that has not been fully explored. These novel approaches could potentially overcome existing limitations and enhance the performance of biopolymer-based packaging materials. A key challenge in the field is the availability and cost-effectiveness of biopolymers (Gowthaman et al., 2021). The high cost of raw materials for biopolymer-based films hinders their commercialization and widespread adoption (Khalid and Arif, 2022). Research efforts should focus on optimizing the composition of biopolymer composites to balance cost and effectiveness, potentially making these materials more economically viable for large-scale production. Another critical area requiring attention is more research on the improvement of water resistance and mechanical properties of biopolymer-based films (Cui et al., 2023). Developing biopolymer-based composites with stronger mechanical properties could significantly enhance their competitiveness in the packaging industry. Furthermore, safety concerns surrounding biopolymer-based packaging materials need to be thoroughly investigated. Potential toxicity and allergic reactions associated with certain biopolymers, such as protein-based and polysaccharide-based polymers, necessitate further biosafety studies (Ghosh et al., 2020). Additionally, research should focus on studying the migration of additives from these materials into food to ensure food safety. The ongoing research aims to create a cost-effective and simpler way of producing films to promote the wider adoption of eco-friendly packaged material in the food industry. Addressing these research gaps could lead to significant advancements in sustainable packaging solutions, making biopolymer-based materials a more viable alternative to conventional synthetic packaging. The present study thereby focuses on developing an active biodegradable packaging system for application in food products using natural extracts, modification of biopolymers, and natural fiber reinforcement. However, limited research exists on the combined effects of these factors in developing high-performance, active, biodegradable materials. The main effort will be on the suitably modified blending of biopolymers i.e.,

starch, protein, and fibers for the formation of the film. The developed film will be utilized for packaging grapes along with a shelf-life study.

The pandemic increased single-use material consumption, prompting the development of active food packaging films (Eraslan et al., 2023). Biobased polymers with active components are needed to meet eco-conscious market demands and extend product shelf life. Additionally, integrating performance and safety in packaging design allows agriculture and food industries to develop safer, eco-friendly systems (Moustafa et al., 2019). A variety of biopolymers derived from renewable sources and food byproducts including polysaccharides like starch, proteins such as casein, and their blends have been employed in film production, often reinforced with natural fibers (Roy and Rhim, 2022). These biopolymers hold immense potential as substitutes for traditional plastics due to their non-toxicity, compatibility with biological systems, and rapid degradation. However, some of the drawbacks of using pure biopolymers as food packaging materials include poor mechanical (e.g., low tensile strength), barrier properties, and thermal properties (Eraslan et al., 2023). The incorporation of natural active agents into bio-films offers significant advantages, such as lower antimicrobial concentrations and slower diffusion speeds compared to synthetic counterparts (Calva-Estrada et al., 2019; Dash et al., 2024). The study aims to develop a biodegradable film consisting of a biopolymer matrix at different proportions of starch and protein reinforced with natural fiber particles, exhibiting many improved properties compared to single-base biopolymers. The reinforcement of natural fibers would improve the mechanical strength and barrier properties of the developed biofilm. In addition, essential oils could be the potential carrier of antioxidant and antimicrobial agents, therefore incorporation of active agents into packaging material may enable retaining food quality and enhance the shelf life of the packaged product. The development of biodegradable polymeric materials and active antimicrobial/antioxidant packaging could be applied to protect packaged food products (Vieira et al., 2022). The goal is to create packaging that enhances food quality, reduces post-harvest losses, and supports sustainability across various commodities. The developed film has the potential to preserve grapes and uphold their quality for up to 8 days under normal conditions due to its effective water and moisture barrier properties, which are hypothesized to inhibit microbial growth and maintain product freshness.

The whole work is divided into the following objectives to achieve the aim of the present thesis:

- To prepare biodegradable film from modified starch and protein, and their blends
- To isolate fibers from natural sources and prepare fiber-reinforced composite films of starch/protein blends
- To incorporate natural active ingredients for development of active films of starch/protein/fiber blends
- To study the application of the developed films for packaging of food materials

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