CHAPTER 2

REVIEW OF LITERATURE

2.1. Cellulose

In nature, cellulose is the most predominant polymer, plays a critical role in the structural rigidity of plant cell walls and is produced in vast quantities annually. This biodegradable, non-toxic, and stable biopolymer comes from a variety of natural sources, such as bacteria, plants, and animals, making it a versatile material with wide-ranging industrial applications (Gupta et al., 2019). Its unique properties—such as biocompatibility and hydrophilicity—have made cellulose a crucial component in fields like pharmaceuticals, engineering, and medicine, where it is used in antimicrobial and hemostatic agents. By 2026, the worldwide cellulose market is expected to have grown from USD 219.53 billion to USD 305.08 billion, driven by applications in biotechnological and industrial sectors (Farooq et al., 2024).

Recent advancements have also highlighted cellulose's potential in nanotechnology, where acidic hydrolysis is used to isolate crystalline cellulose nanoparticles or nanofibers, which exhibit high rigidity and stability. These nanostructures are particularly valuable in applications such as cosmeceuticals, drug delivery systems, packaging, and food additives (Hemmati et al., 2018). Cellulose's ability to form derivatives like fibers, ethers, and microcrystalline cellulose further enhances its industrial versatility. The market for cellulose is expanding through both top-down (plant-derived) and bottom-up (bacterial-derived) approaches, with the Asia Pacific region generating significant revenue, approximately USD 7.4 billion in 2015 (Naomi et al., 2020). This underscores cellulose's essential role as a sustainable and efficient bio-based material in the modern economy.

2.2. Nanocellulose

Nanocellulose (NC), a biodegradable and sustainable material, holds great promise as a renewable resource for advanced materials due to its high cellulose content and environmental benefits. Agricultural waste from crops like banana, pineapple, and sugarcane, offers abundant sources of cellulose, with waste products such as banana rachis, pineapple leaves, and sugarcane bagasse containing up to 83% cellulose. Nanocellulose, with its exceptional properties such as rigidity, stability, and cost-effectiveness, has been used in a variety of fields, such as drug delivery, packaging,

food additives, and cosmeceuticals. Extensive studies have concentrated on isolating NC as cellulose nanofibers (CNF) and nanocrystals (CNC) from these lignocellulosic materials, typically through sulfuric acid hydrolysis (Moreno et al., 2018). This process requires precise control of factors like temperature and chemical concentration to optimize yield and prevent degradation.

Nanocellulose, derived from plant sources, offers remarkable characteristics including high aspect ratio, surface area, and beneficial rheological behavior, making it highly suitable for use in the food industry. It’s non-cytotoxic and non-genotoxic nature has enabled its application in three main areas: as an essential food ingredient, a food stabilising agent, and in food packaging (Serpa et al., 2016). NC is widely employed in food packaging due to its exceptional air and oxygen barrier properties, protecting food from external contamination. Additionally, it is used as a stabilizer in various food products like sauces, soups, and desserts, helping maintain texture and quality (Winuprasith & Suphantharika, 2015). Studies have also demonstrated its role as a dietary fiber, contributing to improved health outcomes such as reduced risks of chronic diseases and better blood sugar regulation. These versatile applications highlight NC’s growing significance in the global food sector (Gupta & Shukla, 2020).

2.3. Mechanical treatment applied to nanocellulose

2.3.1. Ultrasonication

Ultrasound-assisted extraction has gained significant attention as a greener and more efficient method for isolating NC, driven by rising consumer demand for natural and sustainable products. Compared to conventional methods, ultrasound enhances extraction by reducing processing time, minimizing the use of harmful chemicals, and improving both yield and the properties of NC. This technology works by utilizing high-intensity sound waves to create cavitation phenomena, where the implosion of microbubbles increases mass transfer and extraction efficiency (Silva et al., 2017). Despite its promising advantages, challenges such as high energy requirements, equipment design, and process optimization hinder its large-scale application. However, ultrasound's potential to revolutionize NC extraction, through improved control of parameters like intensity and frequency, makes it a focal point for future developments in bio-based material production (Tao & Sun, 2015).

Ultrasound plays a pivotal role in surface modification by facilitating important mechanisms including heat and mass transfer, defibrillation, homogenization, and deagglomeration. Through acoustic cavitation, ultrasonication improves defibrillation by disrupting hydrogen bonds within the cellulose network, breaking down micron-sized fibers into nanofibers using ultrasonic energy within

the range of 10–100 kJ/mol (Tischer et al., 2010). This mechanical impact accelerates the conversion of cellulose into nanofibers, significantly enhancing the extraction process. In chemical treatments, ultrasonication further boosts efficiency by intensifying mass and heat transfer (Fu et al., 2020). The implosion of microbubbles generates localized shear forces, turbulence, and mixing, which reduce diffusion barriers and allow solvents to penetrate deeper into the cellulose matrix. This improvement is particularly valuable for large-scale production, as it increases extraction productivity and effectiveness (Low et al., 2022).

2.3.2. High pressure homogenization

A mechanical method such as high-pressure homogenization (HPH), which uses high shear forces to break cellulose fibres along their longitudinal axis, can be used to extract cellulose. Several mechanical techniques are frequently employed to extract NC, such as steam explosion, ball milling, cryocrushing, microfluidization, and ultrasonication, but HPH is particularly effective due to its high yield and efficiency (George & Sabapathi, 2015; Yu et al., 2021). In HPH, cellulose microfibrils are broken down into nanoscale dimensions by impact and shear forces created when cellulose slurry is pushed through a vessel at high pressure and velocity (Khalil et al., 2014). The advantages of HPH include high productivity and improved fibrillation with repeated pressure cycles. For example, Wang et al. (2013) extracted NC from cotton using HPH at 80 MPa for 30 cycles, producing fibrils with a diameter of approximately 20 nm, though the process reduced thermal stability and crystallinity. Similarly, Wu et al. (2021) extracted NC from okara using HPH at 140 MPa for three cycles, producing fibrils with a diameter of 0.23 µm and a crystallinity index of 70%. In addition to mechanical methods, HPH is often combined with chemical treatments like acid hydrolysis to further enhance extraction efficiency. For instance, Karina et al. (2020) used a combination of acid hydrolysis (60 % sulphuric acid) and HPH at 20 MPa for 15 cycles to extract NC from sugar palm fibres. This process boosted crystallinity while preserving the chemical structure of the cellulose. NC can be effectively extracted from a variety of agricultural residues, such as oil palm empty fruit bunch fibres, sugar palm fibres, and eucalyptus wood pulp, due to this combination of processes (Tian et al., 2016; Ismail et al., 2023).

2.4. Characterization of nanocellulose

Characterization of NC is crucial to understanding its structural, morphological, and chemical properties, which directly impact its functionality in various applications. Methods like X-ray diffraction (XRD) is employed to examine the crystallinity of NC, while transmission electron

microscopy (TEM), field emission scanning electron microscopy (FESEM), and scanning electron microscopy (SEM) offer detailed images of its surface morphology and nanoscale structure. Atomic force microscopy (AFM) allows for three-dimensional surface profiling at the atomic level. Spectroscopy techniques, such as UV-visible spectroscopy (UV-Vis) for optical properties and Functional groups can be identified and the existence or absence of particular chemical bonds can be verified using the use of Fourier-transform infrared spectroscopy (FTIR). Thermal properties are evaluated using thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC), while dynamic light scattering (DLS) is used to determine particle size distribution in suspensions. Together, these techniques offer a comprehensive understanding of NC's shape, surface characteristics, crystallinity, and molecular structure, confirming its suitability for use in composites and other advanced material applications (Varshney et al., 2021).

2.5. Food application of nanocellulose

Initially introduced as a food additive in the 1980s, NC is now being used in various roles such as stabilizing agents, emulsifiers, thickeners, and fat replacers. Additionally, it serves as a carrier for bioactive compounds, enhancing the delivery of nutraceuticals such as omega-3 fatty acids, vitamins, and antioxidants (Shishir et al., 2018; Perumal et al., 2021). Additionally, NC contributes significantly to food preservation by being used in coatings and films that increase shelf life by reducing microbial spoilage and inhibiting oxygen and water vapor permeability (Perumal et al., 2019). The use of NC as a reinforcing filler improves the mechanical and thermal properties of packaging materials, making it ideal for extending food shelf life. Moreover, NC's application in encapsulation technologies enhances the bioavailability and stability of bioactive substances, providing regulated release in particular environments, such as the gastrointestinal tract. With these diverse applications, NC continues to show promise in the development of functional foods and innovative packaging solutions in the food sector (Perumal et al., 2022).

2.6. Pickering emulsion

Pickering emulsions are a particular kind of emulsion that is stabilized by solid particles or soft colloidal particles, such as microgels and hydrogels that adsorb at the oil-water interface. Unlike conventional emulsions stabilized by surfactants, these emulsions provide exceptional physical stability because solid-like particles adsorb irreversibly at the interface, forming close-packed layers that prevent coalescence, flocculation, and Ostwald ripening (Mwangi et al., 2020). Pickering emulsions have gained significant attention in industries such as food, pharmaceuticals, and

cosmetics, due to its excellent stability, adjustable interface characteristics, and structural adaptability. Given growing concerns over the safety of synthetic stabilizers, food-grade biopolymers such as cellulose, chitosan, and plant and milk proteins are increasingly favoured as natural, biocompatible stabilizers in these systems (Yi et al., 2021). **Fig. 2.1** illustrates the formation of Pickering emulsion stabilized with nanocellulose.

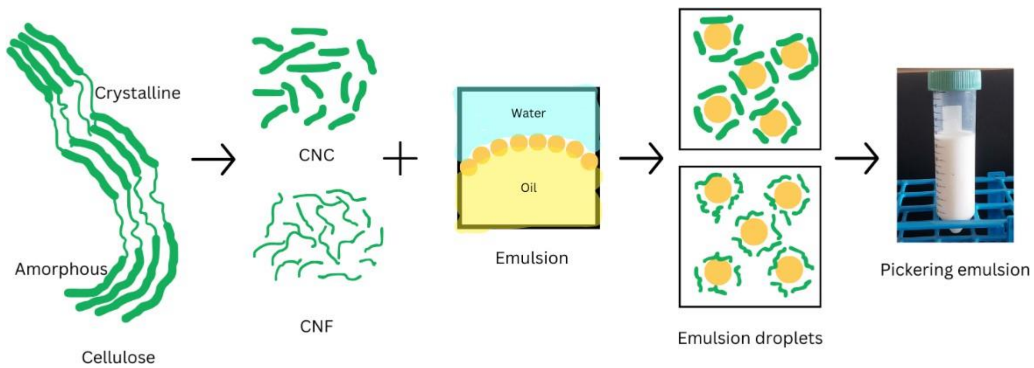

Fig. 2.1. Formation of Pickering emulsion.

For encapsulating lipophilic bioactive substances like β-carotene, a carotenoid with potent antioxidant qualities and high pro-vitamin A activity, Pickering emulsions have shown potential as a delivery method. β-carotene is extensively utilized in both food and pharmaceutical industries, but its hydrophobic nature, low oxidative stability, and limited bioavailability present significant challenges (Boonlao et al., 2022). To address these issues, β-carotene is encapsulated in lipid-based systems, like oil-in-water (O/W) Pickering emulsions, which offer enhanced chemical stability and improved bioavailability. Solid particles in Pickering emulsions create a barrier that shields the oil droplets from degradation and coalescence, making them a suitable option for encapsulating and stabilizing bioactive compounds like β-carotene.

2.6.1. Stabilization mechanisms of nanocellulose

The stabilization of emulsions by NC is attributed due to its capacity to create a steric barrier that stops droplets from coalescing at the oil-water interface by irreversible adsorption and enhances the physical stability of the emulsion. This irreversible adsorption results in the formation of a protective layer of cellulose nanoparticles, which inhibits droplet aggregation and stabilizes the emulsion over time, even under harsh conditions like freeze-thaw cycles. NC also exhibits low toxicity compared to synthetic surfactants and other nanoparticles, making it a safer and more sustainable option for foodand pharmaceutical applications (Dickinson, 2010). Along with its stabilizing qualities, NC also helps to increased viscosity of the emulsion system, which further slows down sedimentation and creaming—the phenomena where oil droplets move under gravity to form a concentrated layer. This is particularly useful in food emulsions, where stability during storage is critical. The NC structure, whether spherical, rod-like, or nanofibrous, can adhere to or get adsorbed at the interface of the phases, facilitating the formation of either oil-in-water or water-in-oil emulsions (Cunha et al., 2014; Guo et al., 2017). Moreover, the conductive attributes and porosity of NC add further value to its role in nanoemulsions, as they can influence factors such as emulsion conductivity and encapsulation efficiency for nutraceuticals.

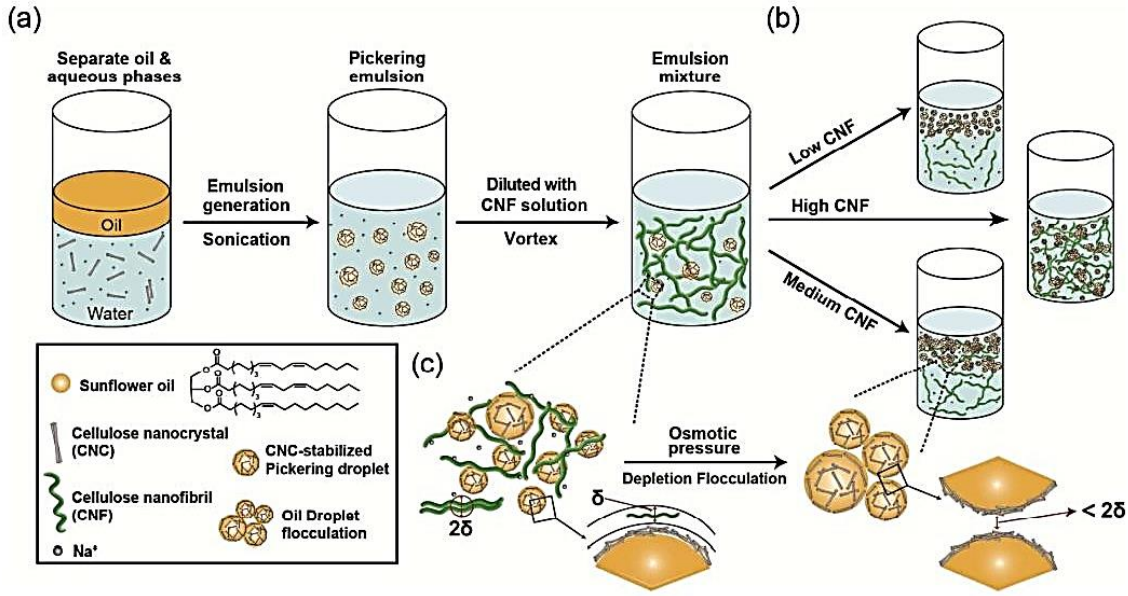

Fig. 2.2. Diagrammatic representation of (a) the addition of CNF after the emulsification process to produce stable CNC-based Pickering emulsions. (b) At varying CNF concentrations, three stabilization regimes are displayed. (c) Non-adsorbed CNF reaches the critical flocculation concentration, indicating depletion flocculation of oil droplets in the CNC-stabilized Pickering emulsions. In this method, the size of a single CNF or CNF flocs in the aqueous phase is indicated by the symbol δ (Bai et al., 2018).

Stable emulsions with low oil concentrations were successfully developed by combining two types of nanocelluloses: cellulose nanocrystals (CNC) and cellulose nanofibrils (CNF). This innovative approach offers an environmentally sustainable route for creating multiscale materials sourced from renewable resources (Bai et al., 2018). CNC and CNF work synergistically to stabilize the Pickering emulsions, with each component contributing unique structural and functional properties that

reinforce emulsion stability. **Fig. 2.2** illustrates the formation stages of these emulsions and the underlying stabilization mechanisms, where CNC provides rigid structural support to the oil-water interface, while CNF enhances viscosity, reducing oil droplet movement and preventing coalescence for prolonged stability.

NC has also proven effective for encapsulating and delivering various bioactive compounds, including micronutrients, polyunsaturated fatty acids, polyphenols, and flavours, improving their bioavailability and protecting them from degradation

(Saffarionpour, 2020). The use of NC in Pickering emulsions for food packaging and nutraceutical delivery aligns with the growing demand for natural, biocompatible, and biodegradable stabilizers in food and pharmaceutical formulations. The ability of NC to stabilize emulsions without synthetic surfactants, combined with its favourable safety profile and functional versatility, makes it a promising candidate for advanced emulsion systems.

2.6.2. Impact of Pickering emulsion on β-carotene bioaccessibility

Nanocellulose-stabilized Pickering emulsions are a promising strategy for enhancing the bioaccessibility of β-carotene, primarily by improving its stability and delivery. Many fruits and vegetables include the lipophilic compound β-carotene, which is very susceptible to chemical degradation and has low bioavailability because of its poor water solubility and sensitivity to environmental parameters including heat, light, and oxygen (Jiang & Hsieh, 2013). Encapsulating β- carotene in oil droplets within Pickering emulsions, stabilized by NC, offers a robust solution to these challenges (Binks et al., 2007). NC forms a strong steric barrier around the oil droplets, protecting β- carotene from oxidative damage and degradation (Wei et al., 2020). This barrier enhances the chemical stability of β-carotene and ensures that its bioactive properties are preserved during storage and processing. Moreover, NC-stabilized emulsions provide a more controlled release of β-carotene in the gastrointestinal tract, improving its absorption by epithelial cells (Winuprasith et al., 2018).

While NC is an insoluble dietary fiber and may interact with digestive components like bile salts, phospholipids, and calcium, potentially affecting lipid digestion, studies show that these interactions do not negatively impact the bioaccessibility of β-carotene (Borba et al., 2019). Instead, NC helps to stabilise the emulsion to prevent oil droplets from coalescence and ensuring the efficient delivery of β-carotene during digestion. Their ability to protect β-carotene from degradation, facilitate its controlled release, and improve its absorption makes these emulsions a valuable tool in the design of food systems aimed at increasing the intake of this important bioactive compound (Fitri et al., 2022).

2.6.3. Health benefits and nutraceutical applications of β-carotene

β-carotene is a crucial provitamin A carotenoid present in green, yellow, red, and orange fruits and vegetables, and a lower risk of chronic illnesses like cancer, cardiovascular disease, and macular degeneration has been associated to its dietary intake (Rao et al., 2013). However, the bioavailability of beta-carotene is inconsistent and comparatively low, which limits its effectiveness in promoting health. Several factors contribute to this low bioavailability, including its poor solubility in both water and oil, its entrapment within the food matrix, and its conjugated double bond structure, which makes it susceptible to oxidation. These factors prevent the gastrointestinal system (GIT) from releasing β-carotene, prevent its incorporation into mixed micelles, and reduce its absorption (Boon et al., 2010).

To address these challenges, encapsulating β-carotene in nanoemulsions (NEs) has emerged as an effective strategy. NEs can protect β-carotene from oxidative degradation, enhancing its stability and bioavailability. For example, the encapsulation of β-carotene in krill oil NEs has been demonstrated to enhance its bioaccessibility and oxidative stability after spray-drying and freeze-drying processes (El-Messery et al., 2020). Similarly, encapsulating polyunsaturated fatty acids (PUFAs), such as those in fish oil, in NEs has demonstrated significantly higher uptake rates in the small intestine, which enhances cholesterol-lowering and anti-inflammatory properties (Dey et al., 2019). These results demonstrate how effective NEs could be as nutraceutical carriers, improving the bioavailability of β-carotene and promoting better health outcomes, such as cholesterol reduction, weight control, and liver fat reduction.

2.6.4. Application of Pickering emulsion in mayonnaise

Mayonnaise is a traditional oil-in-water emulsion that is usually made up of 3–5% vinegar, 6–20% egg yolk, and 65–80% oil. The emulsifying qualities of egg yolk, which lower surface tension and improve emulsion stability, are essential to the stability and structure of mayonnaise. However, concerns over the high cholesterol and saturated fatty acid content of egg yolk, along with the risk of microbial contamination by *Salmonella enteritidis*, have prompted the search for alternative emulsifiers (Lu et al., 2021; Xiong et al., 1999). In recent years, the demand for egg yolk-free mayonnaise has grown, driven by consumer preferences for veganism and healthier options. To meet these demands, studies are focusing on novel emulsifiers, such as plant-based proteins and gums that can replicate the quality and texture of traditional egg-based mayonnaise (Liu et al., 2018).

Pickering emulsions, which utilize solid particles for stabilization instead of conventional surfactants, have shown promise in enhancing the stability and performance of egg yolk-free mayonnaise. These emulsions provide better resistance to coalescence and Ostwald ripening, along with smaller droplet size distributions compared to conventional emulsions (Akcicek et al., 2022). Furthermore, Pickering emulsions employing protein-polysaccharide mixtures offer enhanced stability against droplet coalescence and aggregation, making them ideal for incorporating and delivering probiotics, nutraceuticals, and essential oils (Wang et al., 2020). Research has shown that various plant protein particles and polysaccharides can serve as effective alternative stabilizers for mayonnaise-like emulsions, significantly impacting their stability, viscosity, texture, and rheology (Hosseini et al., 2020; Liu et al., 2018). Despite the food science community's considerable interest, research focused on the practical application of edible Pickering emulsions in the food industry remains limited (Akhtar & Masoodi, 2022).

Additionally, novel approaches have been put up to improve the stability and solubility of emulsion- based gels by using oils high in omega fatty acids, including walnut oil (Zhu et al., 2018). Amphiphilic biopolymers, particularly combinations of proteins and polysaccharides, can form three-dimensional network structures that stabilize oil-in-water emulsions by adsorbing at the oil droplets (Liu et al., 2015). Improved emulsification properties and environmental friendliness result from the electrostatic interactions between negatively charged polysaccharides and positively charged proteins (Gentile, 2020).

2.7. Hydrogel

Hydrogels are intriguing polymeric materials characterized by crosslinked hydrophilic networks, demonstrating significant potential across various fields, particularly in pharmaceuticals and biomedicine, including applications in tissue engineering for artificial cartilage and tendons, as well as drug delivery systems (Dash et al., 2013). These materials can be defined through both rheological and structural perspectives. From a rheological standpoint, based on its storage modulus (G') and loss tangent (tan δ) within a particular angular frequency range, a gel is characterised as a material that cannot flow. An aqueous solution (hydrogel), air (aerogels), or oil (oleogels) can all be used to create a gel, which is structurally composed of a three-dimensional network interconnected by crosslinks (Cao

& Mezzenga, 2020; Nishinari, 2009).

Hydrogels made of natural polymers have attracted a lot of attention since they are renewable, biocompatibility, and biodegradability (Bao et al., 2019). However, conventional natural hydrogels often exhibit limitations in mechanical strength and brittleness due to restricted molecular motion of their biopolymer chains, hindering their practical applications (Tang et al., 2019). Recent advancements have focused on enhancing the mechanical properties of these hydrogels, particularly by incorporating cellulose, the most prevalent biomaterial on the planet. These cellulose nanomaterials, including nanocrystals and nanofibrils, exhibit remarkable mechanical strength, optical transparency, and biodegradability, positioning them as promising reinforcements for the development of high-performance nanocomposites (Zhao et al., 2019). Various methods have been explored for isolating cellulose nanofibers, with a focus on maximizing their reinforcing effects while addressing challenges related to crystallinity and energy demands in processing (Liu et al., 2020).

The properties of gels are significantly influenced by the functional groups of the polymers and the nature of the crosslinks that maintain the network, which in turn affects water retention and overall gel stability. The two primary forms of food hydrogels can be distinguished by the sort of crosslinks they include: physical gels formed through non-covalent interactions and chemical gels created via covalent bonding (Cao & Mezzenga, 2020; Hu et al., 2019). Additionally, food hydrogels are classified according to their polymer composition into single, mixed, and filled (composite) hydrogels, each offering unique insights into the gelation mechanisms and rheological properties of individual biopolymers or their combinations (Khalesi et al., 2021).

2.7.1. Polysaccharide /protein-based hydrogels

Protein and polysaccharide hydrogels have gained significant attention in recent years due to their ability to form complex structures through both covalent and non-covalent interactions. These interactions include hydrophobic, electrostatic, hydrogen bonding, and Van der Waals forces, and chemical cross-linking reactions like the Maillard reaction—enable the creation of binary hydrogels that combine the unique properties of proteins and polysaccharides (Wijaya et al., 2017). Research has shown that these mixed hydrogels can enhance the structural and functional properties of their individual components, resulting in improved mechanical strength, water retention, and stability. For example, studies have demonstrated that the incorporation of κ-carrageenan into casein gels significantly increases their storage modulus and thermal denaturation temperature, while also promoting a more compact and stable network structure (Zhao et al., 2020). Similarly, polysaccharides like konjac gum and gellan gum are known to improve the soy protein gels' rheological and textural qualities, increasing their capacity to retain water, and lowering gelling temperatures (Ozel et al., 2020). Building on this, He et al. (2024) investigated the effects of cellulose nanofibers (CNF) and cellulose nanocrystals (CNC) on soy protein isolate (SPI)-konjac glucomannan hydrogels. CNC (1.0%) improved gel strength, while CNF (0.75%) enhanced elasticity by reinforcing the honeycomb-like structure through synergistic molecular interactions, highlighting their potential

for tailoring hydrogel properties. The development of composite hydrogels from proteins and polysaccharides is not limited to plant-based applications; meat proteins combined with cellulose have also demonstrated enhanced functional characteristics (Zhang et al., 2018). These innovations highlight the versatility and potential of protein/polysaccharide hydrogels in various food and pharmaceutical applications, providing a foundation for further exploration and development in this field (Liu et al., 2021).

2.7.2. Influence of formulation on hydrogel

Recent consumer trends have increasingly favoured reduced or low-fat products due to their association with lower risks of coronary heart diseases and obesity. As a result, natural resource- based biopolymers have drawn a lot of interest due to their accessibility, affordability, and health advantages, which include lowered cholesterol and a decreased risk of cardiovascular disease and dyslipidaemia (Cengiz & Gokoglu, 2005; Scholten et al., 2014). Among these biopolymers, polysaccharides and plant proteins are important components that affect food texture and flavour (Gupta et al., 2022). Low viscosity and mechanical strength limit soy protein isolate (SPI), a highly concentrated plant protein with a protein level of around 90%. However, SPI has advantageous properties such swelling, solubility, foaming, and gelling (Nishinari et al., 2014). SPI is commonly processed into emulsions and gels, allowing for modulation of its properties through combinations with hydrophilic and hydrophobic materials (Song et al., 2011).

Research has highlighted the use of SPI in various applications, including edible and packaging films, filtration membranes, adhesives, tissue engineering, and drug delivery, as well as its role as a food supplement (Reddy & Yang, 2011).

By altering the rheological characteristics of the aqueous media and interacting with protein molecules, the addition of polysaccharides has a considerable impact on the emulsion stability of SPI (Wang et al., 2011; Pan et al., 2015). Combining SPI with polysaccharides can create a fat-like taste with fewer calories, making it suitable for food products designed to mimic or replace fat (Corredig et al., 2011). However, fat removal may negatively impact the structure, texture, and flavour of foods. Cellulose, a semi-rigid polysaccharide recognized as dietary fiber, is frequently used in protein- polysaccharide complexes for its numerous physicochemical and physiological benefits, including water retention, emulsion stabilization, and cholesterol reduction (Schmitt & Turgeon, 2011; Wu et al., 2009). Derivatives of cellulose, such as hydroxypropyl methylcellulose (HPMC) and carboxymethyl cellulose (CMC), are utilized as fat substitutes in various food products, while microcrystalline cellulose (MCC) has demonstrated effectiveness in low-fat applications, such as hamburgers and sausages (Gibis et al., 2015; Wang et al., 2013).

Derived from lignocellulosic sources, cellulose nanofiber (CNF) has special qualities like strength, biocompatibility, biodegradability, low density, and high aspect ratio. Because of its capacity to function as a self-assembly agent and rheological modifier, CNF is a desirable substitute for conventional cellulose derivatives in the creation of novel biopolymer foods or composites (Chen et al., 2014; Khalil et al., 2012). The addition of CNF to SPI can change the mixes' physicochemical characteristics, which are affected by external variables such as temperature, pH, the ratio of SPI to CNF, total solid content, and ionic strength (Sun et al., 2015). The textural qualities and dynamic features of SPI–CNF combinations must be carefully managed to ensure that product quality meets consumer expectations (Patel, 2020).

2.7.3. Entrapment of β-carotene in hydrogels

Because bioactive substances like β-carotene are hydrophobic, incorporating them into food products presents a number of difficulties. These bioactive ingredients frequently exhibit poor oral bioavailability, restricted compatibility with hydrophilic gels, and low solubility in fluid-like conditions (Liang et al., 2013; McClements, 2015). As a result of their poor absorption by the human body, their potential health advantages might not be fully realized. Encapsulation strategies have been developed to entrap bioactive components in delivery systems including solid lipid nanoparticles, emulsions, nanoemulsions, micelles, or microemulsions in order to overcome these restrictions (Singh et al., 2009; Troncoso et al., 2012a). These systems improve the solubility, stability, and bioaccessibility of hydrophobic bioactives, enabling their incorporation into food products.

For highly lipophilic compounds like carotenoids, including β-carotene, bioaccessibility can be significantly enhanced by co-ingestion with triacylglycerols (Hou et al., 2007). Triacylglycerols are broken down by pancreatic and stomach lipases, releasing monoacylglycerols and free fatty acids that combine with phospholipids and bile acids to create mixed micelles (Lo & Tso, 2009). These micelles solubilize and transport hydrophobic compounds like β-carotene to epithelial cells, facilitating their absorption (Wang et al., 2012). β-carotene, a major dietary carotenoid with high pro-vitamin A activity, is abundant in many fruits, vegetables, and animal products (Johnson, 2002). Fortification in functional foods is a perfect fit for it because its consumption has been associated with lower risks of chronic diseases (Gutierrez et al., 2013). Entrapping β-carotene in hydrogels can enhance its stability and bioavailability, providing a promising approach for its incorporation into food matrices (Mun et al., 2015).

The rising incidence of chronic diseases, such as diabetes, hypertension, and cancer, has prompted food manufacturers to add bioactive components, such as vitamins, minerals, and nutraceuticals, to their products (Cushen et al., 2012). However, incorporating these incorporating bioactive ingredients into functional food items presents challenges, particularly for highly lipophilic molecules like β- carotene (Liang et al., 2013). These compounds often exhibit low oral bioavailability, weak water solubility, and chemical instability, making their integration into foods difficult (Reboul, 2013). To overcome these issues, food-grade delivery systems are needed to encapsulate, protect, and release these bioactives in a wide range of commercial food products. Many food items, including dips, dressings, sauces, and desserts, have gel-like qualities and are frequently made using food-grade proteins or polysaccharides to form three-dimensional networks in water. This gelation provides viscoelastic properties, making hydrogels an attractive medium for fortifying foods with bioactives. It is possible to integrate emulsion-based delivery methods into these hydrogels to entrap lipophilic nutraceuticals like β-carotene. For the development of successful functional food products, it is still essential to comprehend how the encapsulation of bioactive components within hydrogels affects their bioaccessibility, specifically their release from the food matrix and incorporation into mixed micelles under gastrointestinal conditions.

2.7.4. Functional properties of nanocellulose/protein hydrogels

Nanocellulose/protein hydrogels are promising delivery systems for bioactive compounds due to their three-dimensional water-filled network structure. This structure shields the bioactive substances that are encapsulated from external factors like oxygen, heat, light, and enzymes, thus enhancing their stability, water solubility, and bioavailability (Pateiro et al., 2021). Hydrogels can be modified to regulate bioactive release at specific locations, such as in the intestine, improving nutrient absorption.

Carotenoids, natural pigments with antioxidant, anti-aging, and immune-boosting properties, have low chemical stability and water solubility (Wani et al., 2016). Hydrogels have been developed to enhance carotenoid delivery. For example, β-carotene-loaded hydrogels formed with protein-coated fat droplets offer better bioaccessibility and stability than emulsions. Additionally, during food preparation, casein hydrogels shield β-carotene from oxidation (Wani et al., 2016). Lycopene and lutein have shown improved chemical stability and water solubility when encapsulated in hydrogels (Koop et al., 2022). Moreover, astaxanthin-loaded hydrogels enhance both UV-light and storage stability (Xu et al., 2023).

Food hydrogels are frequently utilized in dietary supplements and functional foods to enhance the dispersion and bioavailability of bioactives (Li et al., 2022). However, key challenges remain, such as understanding the methods by which dietary ingredients and hydrogels interact, exploring their digestion and absorption in the human body, and ensuring their safety in practical applications. Hydrogels hold potential for developing specialized foods for health conditions like obesity, diabetes, and cardiovascular diseases (Liu et al., 2021).

2.7.5. Hydrogel and its application in ice-cream

Ice cream is a dairy food that is heavy in fat, sugar, and calories (Hashemi et al., 2015), with milk fat playing a crucial role in its texture, flavour, and overall mouthfeel. Excessive consumption of sugar and ice cream fat can cause long-term health problems like type 2 diabetes and obesity (Di Nicolantonio et al., 2016). This has led to growing demand for reduced sugar and fat ice cream products to promote healthier alternatives without compromising quality. Thus, the ice cream industry is increasingly focusing on meeting consumer demand for healthier products with good quality and sensory characteristics. Various formulations have been developed to reduce fat and sugar content, such as incorporating dietary fibers, probiotics, proteins, and gels. Additionally, lowering the calorie density of ice cream through air incorporation into the food matrix has been shown to decrease energy intake, slow gastric emptying, and enhance satiety (Nooshkam et al., 2023).

A growing number of reduced-fat ice cream products with enhanced fatty acid profiles are being designed using hydrogels, especially emulsion hydrogels. These hydrogels address health issues associated with solid fats and act as vehicles for the delivery of bioactive substances such proteins, vitamins, minerals, and antioxidants (Ashfaq et al., 2024). Emulsion gels also allow for the creation of food products with specific textures and nutritional properties to cater to targeted populations. As consumer demand for healthier food options rises, food industries are fortifying products with micronutrients, many of which are sensitive to environmental conditions and have low bioavailability. Emulsion hydrogels enhance the stability and functionality of these ingredients (Lu et al., 2020). For instance, β-carotene-loaded whey protein emulsion gels improve the light and heat stability of the nutrient, while curcumin encapsulated in emulsion gels retains significantly more of its bioactivity under light exposure (Lv et al., 2022). Lycopene-loaded emulsion gels made from whey protein and sodium alginate also demonstrate improved stability and bioavailability (Liu et al., 2022).

In dairy-based products like ice cream and yoghurt, hydrogels act as fat replacers and texture enhancers, providing the desired consistency and mouthfeel without high fat content. For example, emulsion gels made from whey protein have been used to substitute fat in low-fat yoghurt, improving its texture, water-holding capacity, and stability (Li et al., 2022). Similarly, Pickering emulsion gels stabilized by pea protein microgels or bacterial cellulose nanofibers have successfully substituted fat in ice cream, maintaining the product's texture, structural stability, and melting behavior (Qin X et al., 2024).

2.8. Aerogels

Aerogels are a unique kind of nanostructured substance that is created by extracting the pore fluid from a gel and has unique physical properties (Garcia-Gonzalez et al., 2021). These materials are solid, usually in the mesoporous range, with open porosity and low bulk density. Aerogels are materials formed involves substituting gas for the liquid in a gel, creating structures with consistent pore diameters, high surface area, and porosity (Montes & Maleki, 2020). A material must have large specific surface areas (150 m^2/g or more) and high porosity (95–99.99%) in order to be classified as an aerogel. Aerogel networks consist of loosely packed nanometric fibers or particles, which contribute to their remarkable thermal insulation, soundproofing, and high loading capacities. These properties have driven their application in industries such as aerospace, construction, and petrochemicals, while recent research has expanded their potential uses in environmental and biomedical fields, as well as in packaging and functional food (Lehtonen et al., 2020; Plazzotta et al., 2018; Selmer et al., 2019).

Biopolymers, primarily derived from plants and animals, are the predominant nanomaterials used in aerogel manufacturing due to their abundance, renewable sources, low toxicity, biocompatibility, and biodegradability. Among these, plant biopolymers are favored for their availability and sustainability. Cellulose is extensively used in aerogel production. Due to their unique properties and versatility, NC have been explored for a wide range of potential applications, demonstrating their significance in the development of innovative aerogels (Khalil et al., 2020).

The food business and scientific community have shown a great deal of interest in food-grade aerogels because of its remarkable qualities, which include high specific surface area, nanoporous structure, ultra-low density, and superior mechanical and thermal insulation (Santos et al., 2020; Ubeyitogullari & Ciftci, 2020). Consumable aerogels' potential in food applications is still mainly unrealised, despite their substantial research on medication delivery and pharmaceutical uses (Garcia-Gonzalez et al., 2011; Veronovski et al., 2014). The production of food-grade aerogels with Generally Recognised as Safe (GRAS) status using polysaccharides, proteins, and seed mucilages has been the focus of recent advancements (Kleemann et al., 2020; Wang et al., 2019). For food-grade aerogels, polysaccharides such as cellulose, κ-carrageenan, pectin, starch, alginate, and konjac glucomannan are frequently utilized (Wang et al., 2018).

Understanding the effects of different production parameters, such as the type and concentration of the precursor, the gelation process, the use of crosslinkers and surfactants, the drying method, and other important factors, is crucial for efficiently controlling and fabricating the structural properties of food-grade aerogels. The ultimate structural properties of each type of food aerogel are significantly influenced by these parameters (Kleemann et al., 2018; Wang et al., 2019). For the tailored manufacturing of food-grade aerogels, it is therefore vital to conduct additional research focused on optimizing these critical factors.

2.8.1. Nanocellulose aerogels as sustainable packaging materials

Nanocellulose-based aerogels have gained significant attention due to their outstanding properties and versatile applications. These aerogels, derived from renewable NC sources, exhibit excellent performance attributes such a high specific surface area, high porosity, and low density, along with superior mechanical and thermal properties (Chen et al., 2021). The synthesis of NC aerogels involves several key steps, including the preparation of the NC precursor and the selection of appropriate drying technologies. Their mechanical performance is noteworthy, as they display compressive strength, Young’s modulus, and flexibility that surpass those of traditional silica aerogels, which tend to be brittle and easily break under pressure (Jiang et al., 2014). The compressive behavior of NC aerogels can be categorized into three distinct stages: a plastic stiffening effect at high strain, a nonlinear plastic deformation plateau at moderate strain, and an initial linear elastic area at low strain. As the content of NC increases, the stability and density of the aerogel improve, leading to enhanced compressive strength (Peng et al., 2022). Moreover, the relationship between relative density and Young’s modulus is well-defined, indicating that modest increases in aerogel density can further enhance the modulus due to strong hydrogen bonding within the cellulose chains (Jiang et al., 2014). Overall, the unique structural characteristics of NC aerogels, including their honeycomb-like open- cell architecture, make them attractive options for a variety of uses (Levoine & Bergstrom, 2017).

Packaging materials serve multiple critical functions, primarily aimed at protecting products from dirt, bacteria, light, moisture, gases, mechanical stress, and temperature changes (Robertson, 2010). These substances are chosen for their ability to protect while simultaneously serving functions like product presentation, transportation, serving, containing, and informing customers. Primary packaging, also known as consumer packaging, secondary packaging, which contains several primary packages, and tertiary packaging, which is used for effective storage and transportation, are the different categories of packing. Additionally, packaging can include active components that absorb or release useful substances to prolong shelf life, or clever features that offer information about product quality and shelf life (Dobrucka & Przekop, 2019).

Environmental and financial effects, such as the sustainability of raw materials and recycling pathways, are vital criteria for packaging material selection. The user experience and convenience also significantly influence the packaging materials' market potential (Michaloudis & Dann, 2017). Aerogels stand out in food packaging due to their unique porous structure, this leads to a large specific surface area and low weight, providing options for thermal insulation, mechanical protection, and active packaging that can release or adsorb particular molecules (Ghafar et al., 2017). Aerogels' porous architecture dictates their mechanical characteristics, and reinforcing elements like fibres or nanoparticles can increase their strength. To illustrate its potential for protecting packed food during handling and transportation, a chitin-based aerogel weighing 60 mg and having an apparent volume of about 5.6 cm^3 was able to sustain a 100 g object without deforming (Yan et al., 2020).

Moisture can significantly affect the properties of bio-based materials, but techniques such as repeated freezing and thawing after mechanical grinding can produce dimensionally stable aerogels from cellulose fibers that retain their shape and size when in contact with moist substances (Khlebnikov et al., 2020). This stability makes them particularly useful for packaging various food items. Bio-based thermal insulators, like aerogels, could serve as sustainable alternatives to expanded polystyrene in cold storage for items such as fish or hot contents like ready-made meals (Mikkonen et al., 2013). For instance, sol-gel-prepared pectin-TiO_2 nanocomposite aerogels showed thermal conductivities between

0.022 and 0.025 W/m·K, which is less than that of air (0.024 to 0.032 W/m·K), with further optimizations yielding even lower thermal conductivity values (Groult & Budtova, 2018). The lightweight and thermal insulation properties of these aerogels make them particularly appealing for specialized food service applications, such as in aircraft meal services or space missions, where fuel efficiency is paramount. Active aerogel components have also been developed to increase the fresh produce's shelf life by using in situ manufacturing and volatile chemical release like hexanal to inhibit ethylene production and microbial growth. This approach has been shown to reduce mold growth in blueberries and maintain the firmness of cherry tomatoes when packaged with hexanal-releasing active components compared to control samples (Lehtonen et al., 2020).

2.8.2. Novel aerogel delivery systems for improved food packaging application

Despite a growing body of research on impregnated aerogels, there is limited information regarding their ability to alter how the encapsulated components perform and remain stable. Existing studies indicate that aerogels can effectively protect sensitive compounds. For instance, research by De Oliveira et al. (2020) demonstrated that entrapping plant extracts within cellulose aerogels significantly preserved their antioxidant activity. The aerogel coating plays a crucial role in reducing the oxygen susceptibility of loaded oils. According to Ahmadi et al. (2016), fish oil encapsulated in zein-coated whey protein aerogels showed peroxide levels that were almost 60% lower than those of uncoated oil, highlighting the protective capacity of the aerogel matrix.

It is also thought that the special physical characteristics of aerogels improve the bioavailability of molecules that are included. As an illustration, it was discovered that the in vitro bioavailability of phytosterols loaded into starch aerogels was substantially greater (35%) than that of crude phytosterols (3%). (Ubeyitogullari et al., 2019). Additionally, the in vitro bioavailability of these phytosterol-loaded aerogels was three times higher than that of free phytosterols when they were incorporated into food items like puddings and granola bars. The decreased crystallinity of the phytosterols trapped in the aerogels was the cause of this rise.

Unlike aerogels based on polysaccharides, which typically dissolve readily in water, protein-based aerogels exhibit greater resistance to swelling and digestion. This resistance stems from substantial protein denaturation during hydrogel formation, followed by contraction of the protein backbone during drying, which enhances protein-protein interactions (Tang et al., 2014). Consequently, the water insolubility of protein aerogels leads to a delayed release of loaded molecules. For example, protein aerogel-encapsulated fish oil was mostly released during intestinal digestion, with very little release taking place during oral and stomach digestion (Kleemann et al., 2020). Overall, bioactive compound enriched aerogels demonstrate significant potential for improving the bioavailability and stability of sensitive compounds in food applications.

The food industry is actively exploring new technologies to develop packaging that not only protects food from external factors but also provides health benefits to consumers. Food packaging is increasingly being seen as a dual-purpose solution, capable of shielding food from microbial contamination and oxidation while simultaneously acting as a means of delivering bioactive substances. However, during food processing, the bioactivity of natural compounds often diminishes, reducing the nutritional value of many products (Oliveira et al., 2020). To address this, aerogels have emerged as a promising technology for both food preservation and regulated release of bioactive substances. These aerogels, which can be composed of organic materials like cellulose, pectin, and starch or inorganic materials like silica, have drawn interest because of their capacity to encapsulate

and release beneficial compounds in food applications (He et al., 2019). Their unique mesoporous structure enhances their suitability for this purpose, positioning aerogels as an innovative solution in food packaging (Phanthong et al., 2018).

2.8.3. Enhancing food preservation with bioactive compounds in aerogels

Food losses, waste, and foodborne outbreaks are significant concerns for the food industry and society, with packaging playing a vital role in addressing these challenges. Packaging not only protects food from outside factors and maintains its quality, but it can also enhance its nutritional value by incorporating health-promoting compounds, such as antioxidants (Candia et al., 2019). This is especially important for meat packaging, where moisture control is often achieved with absorbent pads. Ideal absorbent pads for meat preservation would also possess antioxidant properties to lower the oxidation of polyunsaturated fatty acids, thereby maintaining meat quality during storage (Otoni et al., 2016).

Traditional absorbent pads are typically made from synthetic materials, like polyethylene, and contain substances to inhibit bacterial growth. However, the environmental impact of synthetic plastics has prompted the food packaging industry to seek sustainable alternatives (Gonzalez et al., 2023). Biopolymers and natural extracts from lignocellulosic biomass present promising solutions for creating eco-friendly packaging. Plant-derived biopolymers, such as cellulose and NC, have been utilized in biocomposite films, while hemicelluloses and lignin have been used as organic food packaging materials (Kumar et al., 2022). Recent studies explored the possibility towards using natural biopolymers to create absorbent pads. For example, the aerogels made by Fonseca et al. (2021) from corn starch and pinhao coat extract showed significant water absorption and contained phenolic substances. Similarly, cellulose and NC extracted from yerba-mate were used by de Oliveira et al. (2020) to create bioactive aerogels, demonstrating desirable properties for food packaging applications. These bioactive aerogels offer a sustainable approach to food preservation, enhancing both product longevity and environmental sustainability.

2.8.4. Application of aerogel in food freshness indicator

According to Khan et al. (2023), conventional techniques for determining the freshness and spoiling of food, like chemical analyses, microbiological counting, and sensory assessments, are frequently expensive, specialized, and difficult to adopt for real-time monitoring throughout the supply chain. Intelligent sensing materials, on the other hand, have drawn a lot of interest because of their low cost, great sensitivity, and quick reaction times. These sensors help producers and consumers monitor and detect changes inside and outside food packaging (Kuswandi et al., 2022).

A particular kind of intelligent indicators is on-package (such as barcode labels, RFID, freshness, ripeness, and time-temperature indicators) and off-package sensors (e.g., electrochemical, optical, and electronic noses) (Taherkhani et al., 2020). Of these, freshness indicators are the most extensively studied, often made from responsive pigments or dyes like curcumin and anthocyanins embedded in polymer matrices (Zheng et al., 2023). Recent research has utilized anthocyanins extracted from food industry by-products, such as purple sweet potatoes, red cabbage, blueberries, and black carrot extracts, to assess the freshness of protein-based foods (de Oliveira Filho et al., 2021). Grape pomace, an important source of anthocyanins is a by-product of the wine industry, particularly from red grape skins, which display visible color changes from pink to blue in response to pH variations, making them effective for monitoring food freshness (de Souza Mesquita et al., 2023).

Biomaterials with high porosity, absorption capacity and high surface area, are well-suited for developing freshness indicators due to their responsiveness and sensitivity to environmental changes. In particular, aerogels, with their high porosity (80–99.8%), light 3D structure, low density, and large surface area, are excellent candidates for porous packaging materials (Sen et al., 2022). Traditionally, aerogels were made from plastics like polyaniline or inorganic materials like SiO_2, but recent research has focused on more sustainable, biomass-derived polysaccharides (Wei et al., 2021). In order to create intelligent and active packaging substances, such as intelligent colorimetric aerogels that have been used to track the freshness of products like chicken and prawns, cellulose-based aerogels have garnered a lot of interest (Zia et al., 2021).

Cellulose, derived from plants and microorganisms, is a common material in food packaging, and its polymerization degree, crystallinity, and fiber size influence aerogel properties. One challenge in aerogel production is the hornification of cellulose, which makes it stiff and brittle. To address this, co-polymers are frequently added, such as carbohydrate gums or soluble cellulose derivatives (Gomez-Maldonado et al., 2022). A novel approach used salep, a stabilizer obtained from the tubers of *Orchis maculata*, as a co-polymer to stabilize cellulose aerogels (Sofiane & Wafa, 2018). Glucomannan, a water-soluble polysaccharide present in salep, enhances the functionality of these materials (Yasar & Bozdogan, 2018). Approximately one-third of food is lost in the supply chain, mostly as a result of perishable goods spoiling, and the growth of e-commerce has increased the demand for long-distance food delivery (Ishangulyyev et al., 2019). To reduce this waste, efficient preservation methods are crucial. Intelligent active packaging (IAP) is an emerging technology that helps extend food shelf life. When food begins to degrade, IAP frequently uses pH indicators to show a colour shift and can release bacteriostatic agents to preserve freshness (Du et al., 2023). Film-based IAPs are the most common form, with many studies embedding pH-responsive natural pigments like curcumin, anthocyanin, and betaine, alongside antimicrobial agents like nisin and thyme essential oil, into the film matrix. These agents are usually embedded through hydrogen bonding, but strong bonds can inhibit their release, limiting their effectiveness (Abedi-Firoozjah et al., 2023; Qin et al., 2024). Additionally, the solubility of most film-based IAPs makes them prone to dissolution under high-humidity and high-temperature conditions, reducing color sensitivity and freshness accuracy (Ezati & Rhim, 2020).

Aerogels are solid materials with pores that are created using sol-gel techniques and gas-phase drying, offer an alternative to traditional film-based IAP. Their large surface area, low density, and mechanical properties make them ideal for food packaging, as they can adsorb biogenic amines and release active ingredients. Due to these properties, aerogels are gaining attention in IAP applications (Selvasekaran & Chidambaram, 2022). For example, Smart aerogels have been produced to monitor the freshness of ground beef using cellulose taken from grape stems, salep as a co-polymer, and anthocyanin from red grapes (Mirmoeini et al., 2023). Similarly, a smart label using red beet pigment and an Arabic-based gelatin/gum aerogel was created to monitor shrimp freshness (B. Li, Chen et al., 2024). However, no reports have yet been published on aerogel-based IAPs that combine spoilage detection with freshness preservation.