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Biomass and Bioenergy 210 (2026) 109061

Contents lists available at ScienceDirect

Biomass and Bioenergy

journal homepage: www.elsevier.com/locate/biombioe

Transforming nutshell waste into next-generation bioplastics for a sustainable and circular economy

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ARTICLE INFO

Keywords:
Bioplastics
Nutshells
Sustainable biomaterials
Circular economy
Agricultural waste
Green extraction

ABSTRACT

The growing concern over plastic pollution has driven the development of sustainable alternatives, such as bioplastics derived from agricultural waste. Nutshells, an abundant lignocellulosic biomass, have garnered attention for their potential in creating biodegradable plastic-based biocomposites. Traditionally considered low-value by-products used for mulch, animal bedding, or fuel, nutshells from almonds, walnuts, chestnuts, hazelnuts, peanuts, and pistachios hold untapped potential for high-value applications in sustainable biomaterials. This review explores the upcycling of nutshell waste into bioplastics, emphasizing their rich content of cellulose, lignin, and phenolic compounds and their conversion into diverse biomaterials. Innovations in extraction techniques, such as deep eutectic solvents and microwave-assisted methods, have enhanced the efficiency and sustainability of recovering these valuable components. The review examines various applications of nutshell-derived biomaterials beyond packaging, including their use in film matrices, fillers, antioxidants, antibacterial agents, UV barriers, plasticizers, biochar, and adsorbents. By transforming nutshell waste into high-value products, the nut industry can significantly contribute to a circular economy, reducing environmental impact while creating economic benefits. This comprehensive analysis highlights advancements in biorefinery processes, the potential of nutshell-derived materials in various industries, and the promising future for sustainable biomaterials sourced from agricultural waste.

1. Introduction

The global plastic pollution crisis has emerged as one of the most pressing environmental challenges of the 21st century, with an estimated 8.3 billion metric tons of plastic produced since the 1950s, much of which persists in landfills, oceans, and ecosystems [1]. Conventional plastics, derived from fossil fuels, are non-biodegradable, contributing to environmental degradation, wildlife harm, and greenhouse gas emissions during production and disposal. This crisis has spurred an urgent need for sustainable alternatives, such as bioplastics, which are derived from renewable biomass and offer biodegradability and reduced environmental impact [2]. Bioplastics, including polylactic acid (PLA) and polyhydroxyalkanoates (PHA), have gained traction as viable substitutes, but their production often relies on food-based crops, raising concerns about resource competition and sustainability [3]. Consequently, the utilization of agricultural waste as a feedstock for bioplastics has emerged as a promising strategy to address both waste management and environmental concerns.

Nutshells, the hard, protective outer layers of nuts such as almonds, walnuts, chestnuts, hazelnuts, peanuts, and pistachios, represent an abundant and underutilized resource with significant potential for bioplastic production. These shells serve critical biological roles, acting as barriers against physical damage, microbial invasion, and pest attacks, thereby safeguarding the edible seed inside. Their robust structure is attributed to a complex lignocellulosic composition, comprising cellulose (30-50%), hemicellulose (20-30%), lignin (20-30%), and phenolic

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https://doi.org/10.1016/j.biombioe.2026.109061
Received 10 December 2025; Received in revised form 1 February 2026; Accepted 1 February 2026
Available online 10 February 2026

compounds, which confer mechanical strength and bioactive properties such as antioxidant, antimicrobial, and UV-blocking capabilities [4,5]. During nut development, the ovule's surrounding tissue undergoes lignification, forming a hard shell that ensures seed survival and dissemination [6]. In the agricultural and food industries, nutshells are typically considered by-products and are discarded or used in low-value applications such as mulch, animal bedding, or fuel [7]. However, their substantial global production, driven by rising consumer demand for nutritious nuts, generates significant waste volumes, presenting both challenges and opportunities for sustainable waste management [8].

Local nut production has surged, with major producing countries contributing to large quantities of nutshell waste. In 2020, almond production reached approximately 1.3 million metric tons, primarily in the United States (California, 80% of global output), totaled 2.1 million metric tons, with shells rich in tannins and phenolics suitable for bioplastics [9]. Turkey dominates hazelnut production (~70% of the 1 million metric tons globally), with shells containing phenolic acids and flavonoids for packaging applications. Peanut production, at 45 million metric tons (led by China, India, and the United States), yields cellulose- and lignin-rich shells for nanocellulose and biochar. Pistachio production, primarily from Iran and the United States (0.9 million metric tons), provides shells with high cellulose content for advanced biomaterials [10]. Traditional disposal methods, such as landfilling or burning, pose environmental risks, including methane emissions and phenolic leachate contamination [11]. To address these challenges, the transformation of nutshell waste into valuable biomaterials is explored.

Recent advancements in biorefinery and sustainable material science have highlighted nutshells as valuable raw materials for high-value biomaterials, including bioplastics, biochar, and nanocellulose. Innovations in green extraction techniques, such as deep eutectic solvents (DES) and microwave-assisted methods, have improved the efficiency and sustainability of recovering valuable components, reducing environmental impact while increasing yield and purity [1]. For instance, cellulose from nutshells enhances the mechanical and barrier properties of bioplastic composites, while lignin offers antioxidant and UV-barrier properties [12]. These bioactive properties make nutshell-derived biomaterials particularly attractive for multifunctional packaging solutions. By transforming nutshell waste into high-value products, the nut industry can contribute to a circular economy, reducing waste and creating economic value (Fig. 1). This review aims to provide a comprehensive analysis of upcycling nutshell waste into bioplastics and other biomaterials, focusing on their chemical composition, advanced extraction methodologies, diverse applications, and environmental and economic benefits.

Despite growing interest in the valorization of nutshell, existing studies remain fragmented, often focusing on individual nut types, isolated components, or single extraction or application routes, with limited integration across composition, extraction, and end-use performance.

The novelty of this review lies in providing a unified and comparative framework that links nutshell chemical composition with green extraction strategies and their implications for bioplastic and functional biomaterial applications. This work benefits researchers by consolidating dispersed knowledge and identifying key research gaps, while offering scientists and engineers practical guidance for selecting suitable nutshell-derived components and sustainable processing routes for targeted applications. While several recent reviews have broadly addressed agricultural waste valorization for biopolymer and biomaterial applications, these studies primarily focus on diverse biomass categories or general processing strategies, without a dedicated, in-depth analysis of nutshell-derived systems. In contrast, the novelty of this review lies in its exclusive

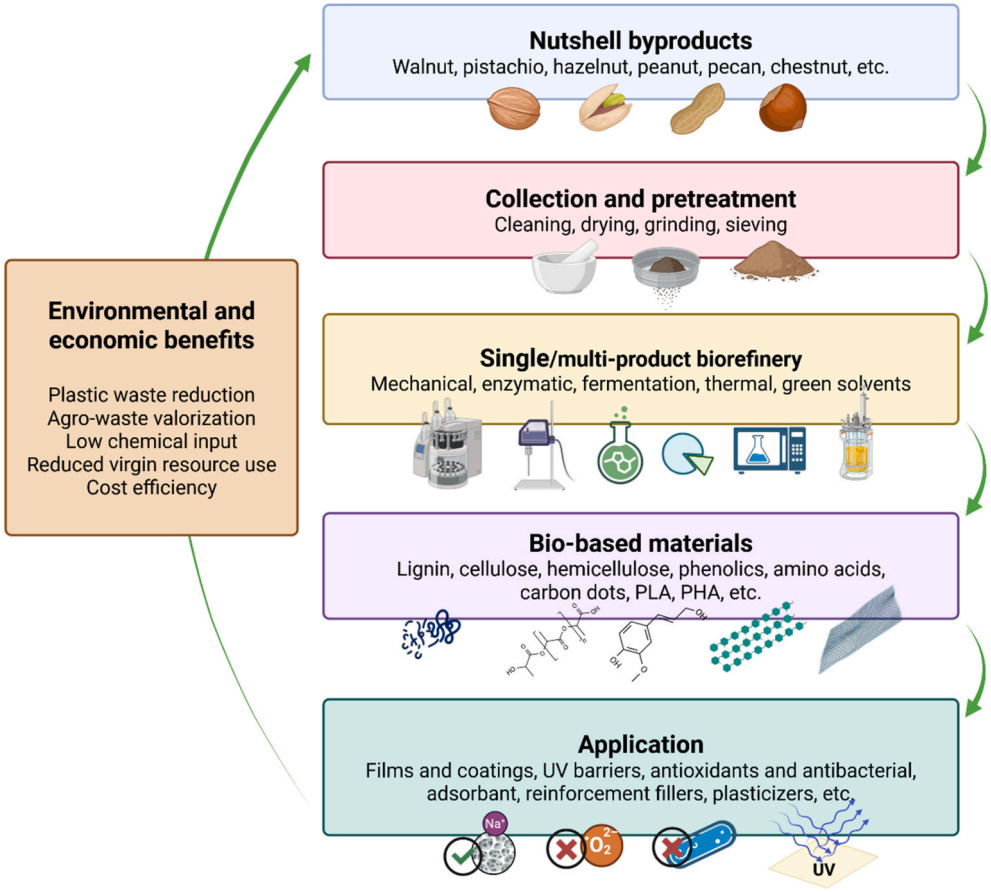

Fig. 1. From nutshell waste to sustainable bioplastics: A circular economy approach.

focus on nutshell waste as a distinct lignocellulosic feedstock, combined with a comparative evaluation of chemical composition, green extraction methodologies, and structure–property–application relationships in bioplastics and functional biomaterials. By systematically linking extraction routes to end-use performance, this work provides a level of integration and application-oriented insight that is not explicitly addressed in existing reviews on agricultural waste valorization.

This review directly aligns with several United Nations Sustainable Development Goals (SDGs), particularly SDG 12 (Responsible Consumption and Production) by promoting the valorization of agricultural waste into high-value bioplastics, SDG 13 (Climate Action) through the reduction of fossil-based plastic use and associated greenhouse gas emissions, and SDG 9 (Industry, Innovation and Infrastructure) by advancing sustainable material design and green processing technologies. In addition, the use of renewable biomass resources supports SDG 15 (Life on Land) by reducing waste and improving resource efficiency within agri-food systems.

2. Physiochemical constituents of nutshells

Nutshells, the hard protective coverings of various nuts, are composed of complex lignocellulosic materials that provide mechanical strength and protect the enclosed seeds [13]. Their physical and chemical properties, including density, hardness, thickness, and the presence of cellulose, hemicellulose, lignin, and phenolic compounds, contribute to both structural integrity and functional potential, making nutshells a valuable resource for sustainable biomaterial applications [2]. These properties vary across nut species and cultivars, influencing their suitability for applications in bioplastics, biochar, and other high-value products. Table 1 summarize the chemical composition of commonly consumed nutshells, highlighting variations in constituents and structural characteristics.

2.1. Physical properties

The physical properties of nutshells, such as density and solubility, play a crucial role in their industrial applications. Manterola-Barroso et al. (2024) reviewed the physical features of hazelnut and walnut shells, reporting real density values ranging from 1.168 to 1.19 g/cm^3 for hazelnut shells and 1.098 to 1.164 g/cm^3 for walnut shells, based on samples from various cultivars in Poland. They also noted theoretical density values of 1.155 to 1.211 g/cm^3 for hazelnut and 1.103 to 1.207 g/cm^3 for walnut shells, determined from milled byproducts [13]. Matin et al. (2017 and 2023) reported shell density values of 1.05 ± 0.09 g/cm^3 for the hazelnut cultivar Istarski duguljasti and 1.3 ± 0.21 g/cm^3 for Rimski okrugli. Regarding solubility [24,25], Pirayesh et al. (2012) found that hazelnut shells exhibited 2.0% solubility in alcohol benzene (2/1) and 50.4% in 1% NaOH, while walnut shells showed 3.2% and 35.2% solubility, respectively. Hot- and cold-water solubility ranged from 18.2 to 20.9% for hazelnut and 7.6 to 10.2% for walnut shells, indicating differences in their chemical reactivity [26].

Nutshells constitute a significant portion of the total nut weight, typically 50–60% (w/w). Manterola-Barroso et al. highlighted that hazelnut shells represent 50–60% of the whole nut weight, with morphological features including a length of 20.85 mm, a width of 19.93 mm, and a shell thickness of 1.73 mm for 12 different cultivars. For walnut shells, they reported a kernel/shell ratio leading to 50–62% nutshell yield, with thickness ranging from 1.1 to 1.7 mm depending on cultivar and location (Fig. 2) [13]. The hardness of nutshells, attributed to their lignocellulosic composition, is a key physical property. Valentini et al. reported an average penetration force of 87.89 ± 4.0 N for hazelnut shells [27], while Kabas et al. noted values of 138.05 N (longitudinal), 55.73 N (transverse), and 89.23 N (suture). For walnut shells [28]. These variations are influenced by cultivar, plantation locality, and testing orientation.

2.2. Chemical properties

Nutshells are lignocellulosic by-products primarily composed of cellulose, hemicellulose, and lignin, along with varying amounts of phenolic compounds, proteins, and lipids. The relative proportions of these constituents differ significantly across nut species, influencing their physicochemical properties and their potential for valorization in material, chemical, and energy applications. Table 1 summarizes the chemical composition of common nutshells, highlighting the variability in structural polymers and bioactive components across different nutshell types. Reported ranges for nutshell composition arise from both biological and methodological factors. Across different studies, species, and cultivars, genetic factors, growing region and climate, soil composition, and agronomic practices (irrigation, fertilization, crop load) all influence shell development and thus the relative amounts of cellulose,

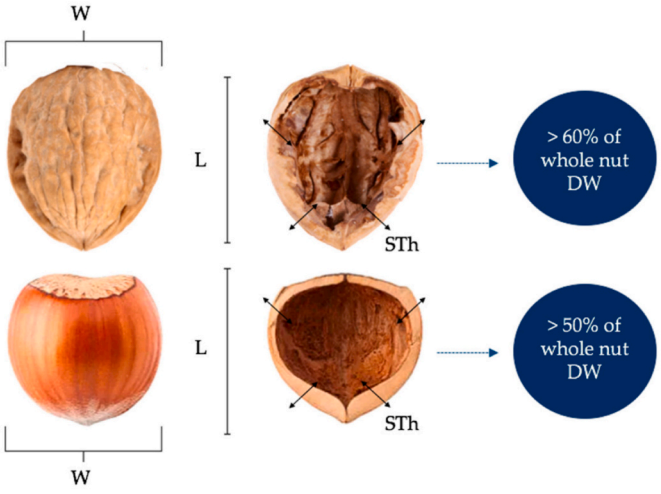

Fig. 2. Mean mass yield and morphology of the whole nut and nutshell of the hazelnut and the walnut. W: width; L: length; STh: shell thickness; DW: dry weight. Adapted with permission from Elsevier [13].

Table 1
Chemical composition of common nutshells (weight %).

Nutshell Type	Cellulose (%)	Hemicellulose (%)	Lignin (%)	Phenolics (mg GAE/g)	Proteins (%)	Lipids (%)	Ref.
Almond Shell	30.94–38.47	20–25	28–33	10–150	3–5	1–2	(14)
Walnut Shell	25–35	15–20	35–50	2.0–249.7	3–6	2–4	(15)
Pistachio Shell	Up to 47.08	23–30	20–30	50–120	2–4	1–3	(16)
Hazelnut Shell	30–40	18–25	16–38.7	9.0–274.09	2–4	2–4	(17)
Peanut Shell	30–35	6–20	28–32	10–60	5–7	1–2	(18)
Chestnut Shell	25–30	23.10–56.40	20–30	50–200	3–5	2–3	(19)
Cashew Shell	20–30	15–25	30–40	–	3–6	60–65 (Anacardic acid)	(20)
Macadamia Shell	28–34	12–18	35–40	15–50	2–3	2–4	(21)
Pecan Shell	28–36	18–24	30–38	40–110	3–5	2–3	(22)
Pine Nut Shell	35–40	20–25	25–30	25–90	2–4	3–5	(23)

hemicellulose, lignin, and extractives. In addition, post-harvest processing (drying regime, thermal treatments, oil removal, grinding/particle size) and differences in analytical protocols (extraction of extractives, lignin determination method, moisture correction) contribute substantially to the variability in published composition values [13,29].

2.2.1. Lignocellulosic composition

Lignocellulose is the primary component of nutshell composition. Lignin, a complex aromatic polymer composed of phenylpropanoid units (p-coumaryl, coniferyl, and sinapyl alcohols) linked by ether (β-O-4) and carbon-carbon (β-5) bonds, constitutes 16-50% of nutshells [1,25]. It provides rigidity, resistance to microbial degradation, and protection against oxidative stress. Walnut shells, for example, can contain up to 50% lignin, while hazelnut shells range from 16 to 38.70% [6]. Lignin's aromatic structure makes it a natural antioxidant and UV-stabilizing agent, ideal for enhancing the thermal stability and UV resistance of bioplastics [11,26]. The development of modern extraction technologies, such as DES and microwave-assisted approaches, has significantly increased the yield and efficiency of lignin extraction from nutshells [1]. Lignin extracted from nutshells has shown promise in enhancing the thermal stability and UV resistance of bioplastic formulations, providing a sustainable alternative to synthetic additives [11]. Advanced extraction methods, such as deep eutectic solvents (DES) and microwave-assisted techniques, have improved lignin recovery efficiency from nutshells like peanut (64% recovery) and chestnut shells [27,28]. These methods enable the integration of lignin into bioplastic matrices, offering sustainable alternatives to synthetic additives.

Cellulose is the most abundant organic polymer on Earth and a major component of nutshells, comprising approximately 30-50 % of their dry weight [25,26]. It consists of long chains of β(1 → 4) linked D-glucose units, forming crystalline microfibrils that provide high tensile strength and rigidity [27]. In nutshells, cellulose serves as a primary structural component, supporting the overall integrity and hardness of the shell [26]. For example, almond shells contain 30.94-38.47% cellulose, while pistachio shells can reach up to 47.08%. Cellulose extracted from nutshells, particularly almond and walnut shells, can be processed into nanocellulose through delignification (e.g., alkaline or acid hydrolysis) and purification techniques [8]. The outstanding mechanical performance of nanocellulose, characterized by high tensile strength and stiffness, makes it highly suitable for enhancing the properties of bioplastics and packaging composites [9]. Its high crystalline and surface area enhance the mechanical and barrier properties of bioplastic films, positioning nutshell-derived cellulose as a key material for sustainable biomaterials.

Hemicellulose, a heterogeneous group of branched polysaccharides, accounts for 15-50% of nutshell composition [10,28]. It consists of sugar monomers such as xylose, mannose, arabinose, glucose, and galactose, forming an amorphous structure that enhances shell flexibility and porosity [11,14]. Hemicellulose binds cellulose microfibrils to lignin, contributing to the structural matrix of nutshells. Its composition varies by nut type; for instance, chestnut shells may contain 23.10-56.40% hemicellulose, while peanut shells range from 6 to 20% [6,12]. The structure of hemicellulose varies significantly depending on the type of nut. Hemicellulose can be extracted from nutshells using mild alkaline treatments, which preserve its functional groups and structural integrity. The extracted hemicellulose can be utilized in producing biodegradable films and as a substrate for microbial fermentation [28,14]. The presence of hemicellulose in nutshells enhances their potential as a feedstock for sustainable biomaterials. Mild alkaline treatments effectively extract hemicellulose, preserving its functional groups for applications in biodegradable films and as a substrate for microbial fermentation to produce bioethanol or xylooligosaccharides [13,24]. The versatility of hemicellulose makes it a valuable component in bioplastic formulations, enhancing film flexibility and biodegradability.

2.2.2. Phenolic compounds and antioxidants

Phenolic compounds, including flavonoids, tannins, and phenolic acids (e.g., gallic acid, catechins, quercetin), are secondary metabolites in nutshells with potent antioxidants, antimicrobial, and UV-protective properties [14,15]. These compounds include flavonoids, tannins, and phenolic acids, which can be extracted using various solvent-based or green extraction methods. Phenolic compounds, such as gallic acid, catechins, and quercetin, found in nutshells like hazelnuts and walnuts, possess aromatic rings with one or more hydroxyl groups, contributing to their antioxidant properties [4,16]. Innovative extraction methods, such as subcritical water extraction and ultrasonic-assisted extraction, have been employed to recover these valuable compounds efficiently [7,17]. These compounds, abundant in chestnut (9.0-274.09 mg GAE/g) and walnut shells (2.0-249.7 mg GAE/g), are extracted using solvent-based methods, subcritical water, or ultrasonic-assisted techniques [6]. For instance, hazelnut shells yield phenolic acids and flavonoids suitable for active packaging, while chestnut shell tannins (2.5-5.2%) provide antimicrobial functionality [16]. Green extraction methods, such as DES and microwave-assisted extraction, enhance yield and purity, making phenolic compounds valuable for bioplastic films with bioactive properties [17].

2.2.3. Minor constituents

In addition to the major components, nutshells also contain proteins, lipids, and minerals that contribute to their overall chemical profile [18]. These minor constituents can be extracted and utilized in various applications, such as in the development of biodegradable films and as additives in bioplastic formulations. For instance, peanut shells are reported to contain essential amino acids and proteins that can enhance the mechanical properties of biodegradable films [8]. The diverse chemical composition of nutshells underscores their potential as a versatile, sustainable resource for a range of industrial applications. In addition to the primary components, nutshells contain minor constituents such as proteins, lipids, and minerals, which enhance their chemical versatility [18]. Peanut shells, for example, contain essential amino acids and proteins that improve the mechanical properties of biodegradable films [12]. Lipids from cashew nutshell liquid (e.g., 60-65% anacardic acid) and pectin from peanut shells (1.61% yield) offer additional functionalities for biomaterial applications [19,20]. These minor components can be extracted and incorporated into bioplastics as additives, thereby enhancing properties such as flexibility and barrier performance. Table 2 summarizes the key functional properties of the major and minor chemical constituents found in nutshells and highlights their potential applications in sustainable biomaterials. Overall, the table shows that cellulose and lignin are the dominant contributors to

Table 2
Functional properties of major chemical constituents of nutshells and their applications in biomaterials.

Constituent	Key functional properties	Applications in miomaterials	Ref.
Cellulose	High tensile strength, rigidity	Nanocellulose films, reinforcement for bioplastics	(30)
Hemicellulose	Flexibility, film-forming ability	Biodegradable films, fermentation feedstocks	(31)
Lignin	UV resistance, antioxidant activity	UV-stabilized plastics, eco-friendly adhesives, coating agents	(32)
Phenolic compounds	Antimicrobial, antioxidant, UV-shielding	Active packaging, antioxidant-rich bioactive coatings	(33)
Proteins	Plasticity, tensile reinforcement	Biopolymer blending, biodegradable film strength enhancement	(34)
Lipids	Hydrophobicity, flexibility enhancer	Water-resistant films, barrier layers, emollients in coatings	(35)
Minerals	Thermal stability, filler properties	Bio-composites, flame retardants	(36)

mechanical strength, barrier performance, and reinforcement functions in bioplastics, while minor constituents such as proteins, lipids, phenolic compounds, and pectin provide complementary functionalities, including flexibility, antioxidant activity, antimicrobial effects, and improved interfacial interactions. This combination of major and minor components underscores the multifunctional role of nutshells as versatile feedstocks for high-performance and sustainable biomaterial applications.

2.2.4. Biorefinery potential

The integrated utilization of all components of nutshells, including cellulose, hemicellulose, lignin, and phenolic compounds, can lead to the development of high value biocomposites with enhanced properties. Adopting a biorefinery approach maximizes the extraction and utilization of each constituent, minimizing waste and promoting a circular economy. Research has demonstrated the feasibility of producing multifunctional biocomposites by combining different fractions of nutshells. For example, cellulose and lignin can be co-extracted to develop bioplastics with superior mechanical and barrier properties, while phenolic compounds can provide additional antioxidant and antimicrobial functionalities [2]. The integrated utilization of cellulose, hemicellulose, lignin, and phenolic compounds through a biorefinery approach maximizes the value of nutshells while minimizing waste [21]. By combining these fractions, multifunctional biocomposites with enhanced mechanical, barrier, and bioactive properties can be developed. For example, co-extraction of cellulose and lignin from pistachio shells produces bioplastics with superior strength and UV resistance, while phenolic compounds add antioxidant and antimicrobial functionalities [8,26]. This holistic approach aligns with circular economy principles, transforming nutshell waste into high-value biomaterials for applications in packaging, agriculture, and beyond. From an integrated valorization perspective, nutshell-based biorefineries can be designed as sequential or cascading processing pathways, where each major fraction is selectively recovered and upgraded into value-added products. In a typical integrated pathway, phenolic compounds may be extracted first using green solvents or subcritical water, followed by hemicellulose recovery via hydrothermal or autohydrolysis treatments, lignin separation through organosolv or DES delignification, and final isolation of cellulose or nanocellulose for bioplastic reinforcement [37]. Such cascade biorefinery strategies enable maximal resource efficiency, reduce processing redundancy, and improve overall economic viability. Moreover, integrating multiple product streams (bioplastics, antioxidants, fibers, and functional additives) enhances process sustainability and aligns nutshell valorization with circular economy and zero-waste principles [38].

3. Extraction methodologies for nutshell-derived biomaterials

Nutshell-derived biomaterials, such as cellulose, lignin, and phenolic compounds, are extracted using a range of chemical, physical, and biological methods, each tailored to maximize yield and functionality while minimizing environmental impact (Table 3). This section details key extraction methodologies, highlighting their applications and efficiencies for various nutshell types.

Table 3 demonstrates that no single extraction approach can be considered universally optimal, as method selection strongly depends on the target compound, the required purity, and constraints associated with scale-up. Conventional solvent extraction remains attractive due to its technological maturity, scalability, and relatively low equipment cost; however, it often relies on large solvent volumes, extended processing times, and energy-intensive solvent recovery steps, which increase operational complexity and environmental burden [47]. Acid and alkaline hydrolysis can achieve high yields of cellulose and nanocellulose and are readily scalable, yet the use of corrosive chemicals, extensive washing and neutralization requirements, and the risk of cellulose degradation can substantially increase wastewater generation, salt loads, and overall processing costs [48].

Green solvent systems based on deep eutectic solvents and ionic liquids exhibit strong dissolution and fractionation capabilities and offer the advantage of solvent recyclability; nevertheless, their broader industrial adoption is frequently limited by high viscosity, challenges in efficient solvent recovery, elevated energy demand at high operating temperatures, and the current cost and availability of some ionic liquids [49]. Microwave-assisted extraction can significantly shorten processing times and, in some cases, reduce solvent consumption, but its industrial feasibility depends on reactor design, uniform energy distribution, and electricity demand, which directly influence throughput and capital investment [50]. Ultrasonic-assisted extraction is particularly suitable for thermally sensitive bioactive compounds and enables rapid extraction under mild conditions, although large-scale implementation can be constrained by reduced cavitation efficiency, energy transfer limitations, and equipment geometry at higher volumes [50].

Mechanical disruption techniques such as ball milling minimize solvent use and are effective as pretreatment steps, yet they are often energy-intensive and may induce oxidation or undesirable structural modifications, including lignin condensation, which can compromise product quality [51]. Enzymatic extraction offers high selectivity and operates under mild conditions, producing minimal toxic effluents, but enzyme costs, longer processing times, sensitivity to lignin inhibition, and the need for tightly controlled reaction conditions can limit economic feasibility at scale [52]. In practice, hybrid approaches that combine pretreatment with enzymatic, microwave, or ultrasonic techniques often offer a more balanced trade-off among efficiency, sustainability, and scalability. Overall, future process development should prioritize integrated extraction strategies that maximize solvent recyclability, minimize washing and neutralization steps, and be supported by comprehensive life cycle and techno-economic assessments to guide method selection for specific nutshell feedstocks and targeted end-use applications.

Table 3
Comparison of extraction methodologies for nutshell-derived biomaterials.

Method	Conditions	Yield	Target compounds	Applications	Environmental impact	Ref.
Solvent Extraction	45-75 °C, 30-60 min, 50% ethanol	10.6-19.58%	Phenolics, starch	Active packaging, films	Solvent-intensive, moderate	(39)
Acid/Alkaline Hydrolysis	121 °C, 60 min, NaOH/H2SO4	50-85.46% (cellulose)	Nanocellulose	Bioplastic reinforcement	Moderate, chemical use	(40)
Green Solvents (DES/IL)	150-170 °C, 45-90 min, ChCl:Oax	60-87% (cellulose)	Lignin, cellulose	Antioxidant additives	Eco-friendly, recyclable	(41)
Microwave-Assisted	66-150 °C, 2.6-10 min, DES	35.36-80% (lignin)	Polyphenols, nanocellulose	Biodegradable films	Energy-efficient, green	(42)
Ultrasonic-Assisted	30 °C, 15-50 min, 80% acetone	15-22.44 mg/g	Phenolics	Antimicrobial films	Eco-friendly, low energy	(43)
Mechanical Disruption	Ball milling, 80 °C, 15-20 min	60-80% (lignin)	Lignin, cellulose	Composite fillers	Moderate, physical impact	(44)
Enzymatic Extraction	200 °C, 5 min, 10 U enzyme	8.2-94.6% (sugars)	Xylooligosaccharides	Bioethanol, films	Eco-friendly, low waste	(45)
Supercritical Fluid	50 °C, 300 bar, 150 min, SC-CO2	82-94% (anacardic)	Anacardic acid, biochar	Nutraceuticals, adsorbents	Highly sustainable, low CO2	(46)

3.1. Chemical extraction methods

3.1.1. Solvent extraction

Numerous researchers have investigated the extraction of bioactive compounds from plants. Traditional techniques involve solid-liquid extraction using different solvents. These methods have notable disadvantages, such as lengthy extraction times and the use of relatively large amounts of organic solvents. Despite these drawbacks, traditional methods remain popular due to ongoing research to optimize their conditions and their widespread industrial applications. Fernández-Agulló and colleagues [53] Conducted a study analyzing various solvent systems, including aqueous methanol, ethanol, and water, to extract antioxidants from chestnut burs. They achieved extraction yields ranging from 12.91% to 19.58% with aqueous methanol, 11.13% to 18.38% with ethanol, and 8.54% to 17.35% using water. The highest Total Phenolic Content (TPC) was found in the aqueous methanol extract at a concentration of 50% (v/v), which yielded 17.74 to 27.69 g GAE per 100 g of extract when processed at 75 °C [53]. Similarly, the chestnut spiny burs (CSB) were subjected to a hydroalcoholic extraction process using 50% ethanol solution at 45 °C for 30 min while stirring. Following this, the solution was filtered, and the solvent was evaporated to isolate the dry extract. The yield of the extraction was found to be 10.6 ± 1.41 % (w/w) [54]. The researchers noted that employing 50% aqueous ethanol resulted in the maximum extraction yield of 11.6% w/w. In contrast, a sequential maceration process utilizing hexane, chloroform, and methanol proved to be the most effective strategy for isolating polyphenols from the alcoholic phase. Additionally, the extract obtained with methanol exhibited a considerably greater TPC of 26.42 g GAE per 100 g of extract, surpassing the ethanolic and aqueous extracts, which recorded TPC values of 20.60 and 20.26 g GAE per 100 g of extract, respectively.

In another study, Du et al. [41] conducted an extraction of tree nut shells biomass (TNSB) suitable for food and pharmaceutical applications. They utilized samples from five types of nut shells, including walnut, hazelnut, macadamia, pine nut, and pistachio, along with two eco-friendly solvents—water and ethanol—and employed cleaner room temperature green extraction (RTGE) methods. Their findings revealed that TNSB is characterized by an exceptionally high lignin concentration, with extracts derived from either solvent being abundant in phenolic compounds. This abundance leads to strong antioxidant capabilities, on par with those of vitamin C, and notable α-amylase inhibitory properties, similar to those of acarbose. Among the samples, walnut shell exhibited the highest lignin content reported to date, at 57%. The water extract from walnut shells showcased remarkable qualities, including 147.65 mg/g of total reducing sugars (TRS), a total phenolic content (TPC) of 60.49%, IC_{50} values of 0.24 mg/mL for DPPH and 0.08 mg/mL for ABTS, as well as 13.44% α-amylase inhibition. Detailed molecular analysis identified numerous phenolic compounds, such as phenol, vanillin, gallic acid, and caffeic acid, among others. These findings underscore the potential of TNSB as a prime resource for lignin-first biorefineries, positioning walnut shell as the most promising option. Additionally, extracts derived from TNSB via RTGE could be further developed as natural antioxidants, preservatives, diabetic treatments, nutraceuticals, and functional food ingredients [41].

Cardona Jimenez et al. [55] developed an economical, environmentally friendly, and easily scalable method for extraction, focusing on the analysis of seven key variables: the type of waste (full or thin pecan shells), the solvent used (double-distilled water or a 1:1 mixture of double-distilled water and ethanol), stirring or maceration techniques, milling, heating conditions, darkness, and the ratio of solvent to sample. This approach employed factorial design and Response Surface Methodology to successfully extract bioactive compounds with antioxidant properties. The extract derived from pecan nutshells contained significant amounts of phenolic compounds (166 mg gallic acid equivalents per gram of dry weight), flavonoids (90 mg catechin equivalents per gram of dry weight), and condensed tannins (189 mg catechin equivalents per gram of dry weight). These components contributed to a polymeric color proportion of 74.6% and exhibited strong antioxidant activity, including $ABTS^{+}$ radical inhibition (3665 μmol Trolox Equivalent per gram of dry weight) and iron reduction capacity (1305 μmol Trolox Equivalent per gram of dry weight). Further analysis via HPLC-ESI-MS/MS identified multiple related compounds, including catechin, gallocatechin, myricetin, dihydroquercetins, proto-anthocyanidin dimers A and B, ellagitannins, and derivatives of ellagic acid [55]. In addition, efficient solvent extraction of cardanol, a renewable phenolic for bioplastics, epoxy resins, and polyurethanes, along with cardol and 2-methyl cardol from cashew nutshell liquid provided an energy-saving, scalable alternative to conventional distillation. Using a two-step aqueous ammonia and organic solvent (hexane and ethyl acetate) process, the compounds were selectively separated, and thermodynamic modeling supported the design of a continuous, sustainable extraction system [56].

The solvent extraction has also been used to extract starch from nutshells. In research conducted by Harini et al. [57], starch was extracted from the shells of cashew nuts. The cashew shell powder was immersed in water at a 1:5 wt ratio for 3 h at a temperature of 30 °C. This process was followed by screening and re-blending it with a mixture of 70% ethanol and then sodium hydroxide. The resulting defatted starch from the cashew nut shells exhibited a significant starch concentration of 80.32%. The moisture, protein, total dietary fiber, and ash contents of the extracted starch powder were measured at 1.74%, 1.24%, 3.11%, and 4.14%, respectively. Additionally, there was a presence of 9.45% of unidentified impurities in the extracted starch. The amylose content of the starch was calculated at 26.32%, categorizing it as moderate in amylose, while the amylopectin represented 73.68% of the total starch [57].

Cashew nutshell (CNS) starch, extracted via solvent-based methods, yields a moderate amylose content of 26.32 ± 0.43%, with a thermal degradation temperature of 310 °C, suitable for thermoplastic film production. Similarly, walnut shell cellulose (WNC), obtained through mechanical and solvent processing, exhibits a high crystallinity index of 72%, with thermal degradation temperatures at 319 °C and 461 °C, making it an effective reinforcing agent for bioplastics [57].

3.1.2. Acid and alkaline hydrolysis

Acid hydrolysis is widely used to obtain nanocellulose from lignocellulosic materials. This process effectively removes the non-crystalline areas, resulting in the formation of crystalline cellulose. During hydrolysis, hydronium ions disrupt the strong hydrogen bonds within the cellulose chains, leading to variations in the breakdown rates between the amorphous and crystalline segments. Utilizing sulfuric acid enhances water dispersion of crystalline nanocellulose, as the sulfate ester forms a stabilizing electrostatic layer that improves the suspension's stability [31]. The technique of alkaline extraction has also been applied to obtain biomaterials from nutshells. The pistachio shell, after being treated with alkali, underwent acid hydrolysis as demonstrated by Movva and Kommineni [58] to extract nanocellulose. The alkali process partially degraded non-cellulosic elements, leading to a noticeable rough texture, but no significant changes were otherwise noted (Fig. 3b). The alkali treatment did not completely remove cellulose from its surrounding matrix; residual lignin served as a binding agent, facilitating connections with cellulose. Nevertheless, under intense bleaching conditions, these binding agents were removed, resulting in visible defibrillated cellulose masses (Fig. 3c). Consequently, the fiber diameter was reduced. This illustrates the roles of alkali treatment and subsequent bleaching in extracting cellulose. The acid hydrolysis enhanced the crystallinity of the cellulose fibrils by degrading the amorphous regions (Fig. 3d).

In a study by Harini et al. [57], cellulose was isolated from walnut shells. The walnut shell powder was treated with a NaOH solution, washed with distilled water, and then treated with sodium hypochlorite solution. The cellulose content in the extracted walnut shell cellulose

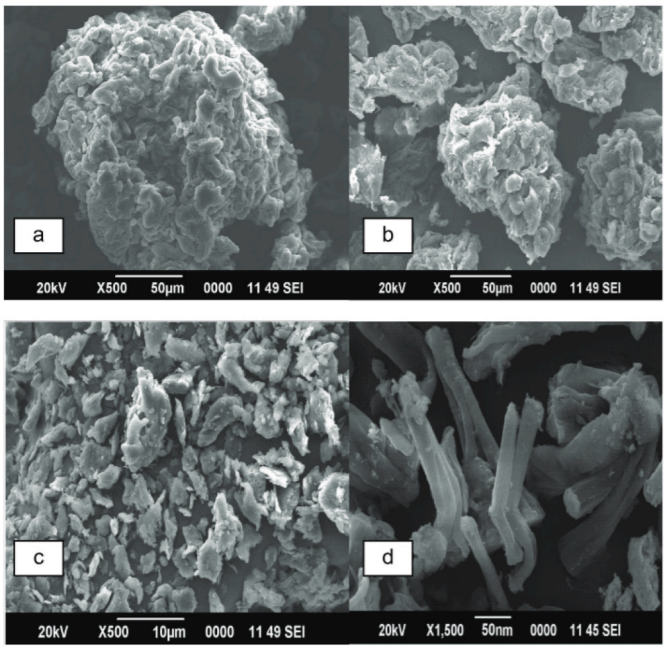

Fig. 3. SEM images of (a) untreated pistachio, (b) alkali-treated pistachio, (c) bleached pistachio, and (d) acid hydrolyzed pistachio shell [58].

was found to be quite high, at 85.46%. Additionally, the levels of protein, ash, and other undetectable impurities were measured at 0.23%, 5.53%, and 7.81%, respectively [57]. nanocellulose were extracted from the cost-effective fruit shell of *Camellia oleifera* Abel (SCOA). The process involved a series of treatments: alkali extraction, followed by hydrogen peroxide bleaching, and finally acid hydrolysis to eliminate non-cellulosic materials and isolate the CNC. The application of alkali and bleaching techniques resulted in a significant reduction of lignin and hemicelluloses in the fibers, enhancing the cellulose content of the refined SCOA to 84%. The resulting CNC features a needle-like morphology, with an average diameter and length of 6 ± 2 nm and 500 ± 100 nm, respectively. Additionally, the crystallinity index of the CNC increased to 72%, and its initial decomposition temperature rose to 230 °C [59]. An analogous process involving alkali extraction, subsequent bleaching with hydrogen peroxide, and concluding with acid hydrolysis was employed to obtain CNC from pistachio shells. It was demonstrated that typical techniques for purifying and hydrolyzing cellulose yield useable CNCs. The results indicated a yield of 50 ± 14 wt %, which was relatively high for a product derived from agricultural waste. The aspect ratio measured at 17 ± 3, the crystallinity was found to be 66%, and the surface charge density was 90 ± 12 mmol/kg [60].

In a different study, walnut shells were employed as the source material for generating purified cellulose [61]. The method for production included several processes, such as alkaline treatment and bleaching. Additionally, two types of nanocellulose were extracted from walnut shells using 2,2,6,6-tetramethylpiperidine-1-oxyl radical (TEMPO) oxidation and sulfuric acid hydrolysis, referred to as TNC and SNC, respectively. The findings indicated that walnut shells contained the least amount of cellulose, measuring 27.40%, while possessing high levels of noncellulosic substances like lignin (36.31%) and hemicelluloses (31.30%). After treating the walnut shells with NaOH, the content of lignin and hemicelluloses dropped to 30.98% and 7.6%, respectively, while the cellulose content rose to 56.6%. Post-bleaching, the cellulose concentration increased to 87.9%, achieved by eliminating leftover lignin and hemicelluloses, leading to highly purified cellulose. The microstructures of walnut shell, NaOH-treated walnut shell, bleached walnut shell, TNC, and SNC were examined using a scanning electron microscope. The SEM imagery showed that the walnut shell had a rough surface. In contrast, the NaOH-treated walnut shell displayed an irregular porous structure, attributed to the breakdown of hemicellulose and partial lignin degradation from repeated alkaline treatments. The bleached walnut shell showed a loose structure, signifying effective removal of residual lignin via an acetate buffer/$NaClO_2$ solution. Upon freeze-drying, the TNC exhibited a porous network with a lamellar configuration, resulting from strong hydrogen bonding that led to the self-assembly of nanoparticles into these structures. The aerogel produced from freeze-drying the SNC also showed a similar porous architecture.

3.1.3. Green solvents

In this context, green solvents refer to solvents designed to minimize environmental and human health impacts by exhibiting low toxicity, low volatility, high recyclability, and reduced energy and resource consumption, while maintaining high efficiency for biomass dissolution, extraction, or fractionation. Ionic liquids (ILs) and deep eutectic solvents (DESs) are attracting significant interest because of their eco-friendly properties. They offer numerous benefits, including outstanding electrical conductivity, high ionic mobility, and robust chemical and thermal stability. Additionally, they can selectively dissolve a wide range of organic and inorganic materials and are recyclable. Malaiyarasan and Ramalingam [62] successfully developed a DES comprising triethanolamine (TEA) and choline chloride (ChCl) at various molar ratios. This was done to extract cardanol from cashew nutshell liquid, utilizing isobutyl ketone and ethyl acetate as co-solvents in different molar proportions of the DES. Husanu et al. [63] developed an integrated approach to fully valorize chestnut shell waste by combining deep eutectic solvents (DES) and bio-based ionic liquids (bio-ILs). Polyphenols were first extracted using a choline chloride–oxalic acid DES, while the residual solids were subsequently treated with cholinium glycinate to separate lignin and cellulose. Spectroscopic and thermal analyses confirmed effective fractionation, and both solvents were recyclable, highlighting the process's potential for sustainable large-scale application.

Anuchi et al. [64] evaluated a low-cost protic ionic liquid, N, N, N-dimethylbutylammonium hydrogen sulfate ([DMBA][HSO_4]), for ionoSolv pretreatment of coconut husk and shell. Optimal treatment at 170 °C for 45 min achieved significant lignin removal, about 77% in husk and 82% in shell, producing cellulose-rich pulp with high glucose yields (82–89%) during enzymatic hydrolysis. The superior performance in shells was attributed to their more porous structure. Significant lignin extraction and glucose yield from these feedstocks illustrate that [DMBA][HSO_4] is a highly effective ionic liquid for processing biomass that is rich in lignin.

In another study, Carneiro et al. [65] investigated how raw peanut and chestnut shells dissolve in ILs. They successfully dissolved up to 7 wt % of raw biomasses under adjusted operational parameters. Utilizing quantitative ^{13}Cq NMR, they found that up to 87% of the cellulosic material was extracted into the IL. Regeneration processes that employed an antisolvent mixture enabled the recovery of both the cellulosic material and the IL. The overall mass balance indicated minimal losses (<8%), with recoveries of 75% for peanut shells and 95% for chestnut shells, along with over 95% recovery of the IL in both cases. These findings highlighted the significant potential of utilizing nut shells and ILs for biorefining applications, while the high recovery rates of ILs also enhanced the economic viability of the process.

3.2. Physical and mechanical extraction techniques

3.2.1. Microwave-assisted extraction

Microwave-assisted extraction has gained considerable interest across various disciplines. This method stands out due to its unique heating approach, reasonable investment requirements, and effective operation under normal atmospheric conditions. Microwaves are a form of electromagnetic radiation, characterized by oscillating electric and magnetic fields that operate within a frequency range of 0.3 to 300 GHz.

They can penetrate specific materials and interact with polar substances to produce heat. The energy produced by microwaves affects molecules through ionic conduction and dipole rotation, allowing for the selective heating of materials based on their dielectric properties [66,67]. Zhang and colleagues [68] developed a microwave-assisted enzymatic extraction (MAEE) technique to improve the yield of polyphenols from discarded peanut shells. The ideal parameters were identified as 2.6 min of irradiation, 0.81 wt% of cellulase, a pH level of 5.5, and an incubation temperature of 66 °C for 2 h. Under these conditions, the total polyphenol yield reached 1.75 ± 0.06%, surpassing yields from other extraction techniques such as heat-reflux, ultrasonic-assisted, and enzyme-assisted methods. Scanning electron microscopy revealed structural alterations in the plant material after various extraction methods, illustrating the disruptive effects of the process. Additionally, the crude extract underwent purification using NKA-9 resin, resulting in an increased polyphenol concentration of 62.73% and enhanced antioxidant and antibacterial properties.

Ahorsu et al. [69] reported a rapid and efficient delignification method for lignin-rich biomass such as walnut shells by integrating ball milling, microwave irradiation, and deep eutectic solvents (DES). Using a choline chloride–formic acid DES (1:2), lignin was selectively dissolved within 10 min at 150 °C, achieving yields of 60–80%. The recovered lignin retained chemical characteristics like milled wood lignin, confirming minimal structural alteration.

According to a study conducted by Harini and Chandra Mohan [57], nanocellulose was derived from walnut shells using a microwave-assisted extraction technique. The initial unprocessed sample contained significant levels of impurities, including hemicellulose at 10.26% and lignin at 27.19%. Consequently, the cellulose concentration in these untreated samples was found to be 42.36%. In contrast, the nanocellulose (35.36% yield) extracted from walnut shells demonstrated a much higher cellulose content of 79.24%, along with substantially reduced levels of lignin (6.18%) and hemicellulose (0.28%). The untreated powders of walnut shells had an average particle size of 840 μm. Following microwave digestion to produce nanocellulose, the particle size of this agro-waste material decreased significantly to 152 μm. This suggests that a substantial portion of the initial lignin and hemicellulose, which bind the cellulose fibers, was eliminated during the treatment, leading to a more refined form of cellulose. Microwave digestion treatment for transforming nanocellulose breaks down cellulose fibers by eliminating the lignin and hemicellulose that bind the cellulose fibrils together. This process disrupts the hydrogen bonds linking the cellulose chains, resulting in a coarse surface that enhances mechanical bonding [57].

A new method for extracting CNCs from almond shell waste using microwave assistance was created and assessed against traditional extraction techniques [70]. The evaluation employed a Box–Behnken design featuring three factors and three levels with five central points to analyze how extraction temperature, irradiation duration, and NaOH concentration affected the crystallinity index (CI). The microwave-assisted extraction yielded a CI value of 55.9 ± 0.7%, achieved in just three stages, while the conventional method, which involved five stages, had a CI of 55.5 ± 1.0%. This innovation led to a more sustainable CNC extraction process that minimizes time, solvent use, and energy requirements. Techniques such as Fourier transform infrared spectroscopy (FTIR), X-ray diffraction (XRD), atomic force microscopy (AFM), and scanning electron microscopy (SEM) confirmed the effective removal of non-cellulosic materials after chemical treatment. Transmission electron microscopy (TEM) indicated that the CNCs were spherical and averaged 21 ± 6 nm in size, showcasing their promising applications in food packaging, biomedical fields, and optoelectronic devices [70].

3.2.2. Ultrasonic-assisted extraction

Ultrasonic-assisted extraction leverages cavitation induced by ultrasonic waves to enhance solvent absorption, thereby boosting the dissolution of extractable compounds. This technique is both safe and eco-friendly, offering benefits such as reduced extraction times and a faster overall extraction process. It is commonly utilized for extracting thermally unstable compounds from plant materials. Rodrigues et al. [71] optimized ultrasound-assisted extraction of phenolic compounds from coconut shells. Using a factorial design and response surface methodology, they identified optimal conditions, 30 °C, pH 6.5, 15 min, and a 50:1 solvent-to-solid ratio, yielding 22.44 mg of phenolics per gram of biomass.

Arciello et al. [72] generated pecan nutshell extract (PNSE) using an ethanol/water mixture (6:4 v/v) in an ultrasonic bath at room temperature for half an hour. The subsequent steps included centrifugation, supernatant filtration, and ethanol removal. The residue was then freeze-dried, resulting in a red powder with a yield of 15% w/w. To prepare the PNSE solution, 0.5 g of PNSE was mixed with 10 mL of water, agitated in an ultrasonic bath, and then centrifuged. The supernatant was collected and inspected using UV–vis spectroscopy at 24-h intervals for a week to assess its stability. The concentration of PNSE in the supernatant measured 30 mg/mL. Stability tests showed that the PNSE solution remained unchanged at room temperature for at least one week, as confirmed by regular UV–vis spectroscopic analysis with no alterations in absorption spectra. Furthermore, PNSE at a concentration of 7.5 mg/mL effectively inhibited the growth of *Salmonella enterica* subsp. *enterica* ser. Typhimurium. It exhibited even greater efficacy against *Enterococcus faecalis*, with a minimum inhibitory concentration of just 1.85 mg/mL(72).

Chavan and Singhal [73] optimized ultrasound-assisted extraction of polyphenols from arecanut using acetone as the solvent. Through response surface methodology, they identified conditions that maximized polyphenol and tannin yields while reducing arecoline content. The optimized extract also showed strong antioxidant activity, confirming the efficiency of the ultrasound process. In research led by Harini and Chandra Mohan [57], nanocellulose derived from walnut shells underwent a process involving DMSO soaking and ball milling along with ultrasonic treatment to create nanocrystalline cellulose fibers by breaking down the nanocellulose using sound waves. This ultrasonic technique, conducted in a liquid medium, produces acoustic cavitation, resulting in bubble formation, expansion, and collapse. During ultrasonic treatment, localized hot spots generate shockwaves that erode cellulose fibers, breaking them along their length to form nanocellulose. The reduction in fibril diameter is attributed to the removal of amorphous regions within the semi-crystalline structure. This method produced about a 58% yield of nanocrystalline cellulose from walnut-derived nanocellulose.

3.2.3. Mechanical disruption

The ball milling technique can significantly change the inherent structure of aromatic biopolymers. Specifically, this process facilitates the creation of radical species by disrupting the β-O-4 bonds, leading to additional decomposition of the biomass [74]. Blasi et al. [75] indicated that using ball milling as a pretreatment is not ideal for the biomass of walnut and pistachio shells. This method fails to effectively separate lignin from the full tissue and causes oxidation of the material. While walnut shell oxidation can be reduced due to the strong radical scavenger activity of its extractives, pistachio shells experience partial lignin condensation due to ball milling. On the other hand, pistachio shells are an appealing resource because they can be easily delignified through mild ethanosolv techniques. This process results in uniform lignin fractions, predominantly consisting of S units (86%) and linked by β-O-4 motifs (over 60% C_9 units), making them well-suited for further chemical transformation into valuable aromatic compounds. Cheng and colleagues [76] introduced mechanochemical methods that involve ball-milling along with mild acetosolv or alkaline treatments to investigate the structural changes in lignins from *Camellia oleifera* shells. These methods notably enhance lignin yield, resulting in the extraction of high molecular weight lignins (approximately 7000 mol/g). The

lignins derived from *C. oleifera* shells are categorized as GS-type, with G units constituting 71.9% of the milled wood lignin. The combination of ball-milling with acetosolv treatment facilitates the breakdown of β-O-4 linkages. On the other hand, alkaline lignin fractions preserved a significant number of these linkages (50.67 per 100 Ar) and exhibited a low concentration of phenolic OH (1.56 mmol/g), suggesting that the ball-milling process had minimal impact on the structure of alkaline lignins (typical substructures). The method of ball milling has also been integrated with various extraction processes to obtain polyphenols and recover lignins rich in antioxidant qualities from the shells of consumable nuts [69,77].

Walnut shell powder (WSP), prepared through mechanical processing involving drying at 80 °C for 15–20 min, crushing with a mortar and pestle, grinding in a mixer, and sieving to 100 μm, serves as a lignocellulosic filler rich in cellulose and lignin. This method ensures fine particle size and uniform dispersion in starch-based composite films, enhancing mechanical and hydrophobic properties [78]. Walnut shell (WS) powder, prepared through mechanical processing (e.g., grinding) as a lignocellulosic filler, enhances the mechanical and UV-blocking properties of carboxymethyl cellulose (CMC)-based bioplastics. The fine WS particles, incorporated at 12 wt% in a one-pot casting method, contribute to high tensile strength (18.53 MPa) and water stability, demonstrating the efficacy of mechanical processing for nutshell-derived biomaterials [79].

Additionally, Pathaveerat et al. [80] investigated a hydraulic cold-pressed extraction method for cashew nutshell liquid (CNSL) from cashew nut shells, achieving a maximum yield of 35.98% at pressures of 200-275 kg/cm^2 without chemicals or high temperatures. In their study, they prepared cashew nut shells by sun-drying and grinding them into smaller pieces using a mincer, followed by compression using a custom-built hydraulic system. The hydraulic cold-pressed extraction machine consists of a compression chamber and piston driven by a hydraulic cylinder and pump, allowing controlled application of high pressure to the biomass, while a perforated pressing zone and oil collection tray enable efficient separation and recovery of CNSL under ambient, solvent-free, and low-energy conditions. This mechanical approach highlights an eco-friendly method for extracting phenolic-rich CNSL, complementing other mechanical disruption techniques (Fig. 4).

3.3. Biological and advanced extraction approaches

3.3.1. Enzymatic extraction

Enzymatic extraction offers a promising approach to generate valuable biomaterials from nutshells, utilizing the precision and effectiveness of enzymes while reducing environmental impact. On this point, Singh and colleagues [81] introduced an environmentally friendly, chemical-free sequential method that incorporates autohydrolysis, enzymatic treatment, and membrane-assisted refining to convert almond shells into low polymerization xylooligosaccharides. They studied various temperatures (180, 200, and 220 °C) and reaction durations for the autohydrolysis process. Subsequently, the autohydrolysate underwent enzymatic treatment aimed at enhancing the levels of low polymerization xylooligosaccharides. Three enzyme dosages (5, 10, and 15 U) were tested, and the ideal dosage was determined through statistical methods. The enzyme-rich solution, high in xylooligosaccharides, was then refined using membranes of 1 kDa and 250 Da to extract a concentrated product. Under optimal conditions (200 °C for 5 min), they were able to derive approximately 54.5% of xylan as oligosaccharides. However, the resulting autohydrolysate contained 3.5% (w/w of biomass) of low polymerization xylooligosaccharides, specifically xylobiose and xylotriose. By employing 10 U of the enzyme, the concentration of these low polymerization xylooligosaccharides increased to 8.2% (w/w of biomass). Ultimately, the membrane-assisted refining process successfully recovered 69.1 ± 0.1% (w/w) of the generated xylooligosaccharides [81]. In another study, Halysh et al. [82] investigated walnut shells as a viable renewable resource for biorefinery applications. Their research aimed to uncover the correlation between the chemical makeup of solid byproducts from biomass following acid treatment with H_2SO_4, alkaline treatment with NaOH, and steam explosion, and the extraction of sugars and lignin through subsequent enzymatic hydrolysis utilizing the industrial cellulase Cellic CTec2. Walnut shells contain significant amounts of cellulose (24.19%), lignin (44.63%), hemicelluloses (26.68%), and extractives (11.41%), which influence the efficiency of converting polysaccharides into sugars. The study found that alkaline treatment yielded more glucose compared to acid or steam explosion methods, achieving a maximum enzymatic cellulose conversion of 94.6%. The lignin content in these solids impedes enzymatic hydrolysis efficiency. These findings suggest effective and promising methods for utilizing walnut shells via cost-effective and straightforward chemical processes, leading to the generation of value-added products and a reduction in environmental pollution associated with traditional disposal methods such as burning, as well as decreasing reliance on external energy and materials by leveraging these renewable and economical resources [82].

Kacem et al. [83] evaluated almond shells as a feedstock for bioethanol production. The biomass was pretreated with alkali and acid, then enzymatically hydrolyzed using *Penicillium occitanis* enzymes. Process optimization improved sugar release, while laccase detoxification of the hydrolysate markedly increased fermentation efficiency, raising ethanol yield from 30% to 84%. The study highlights almond

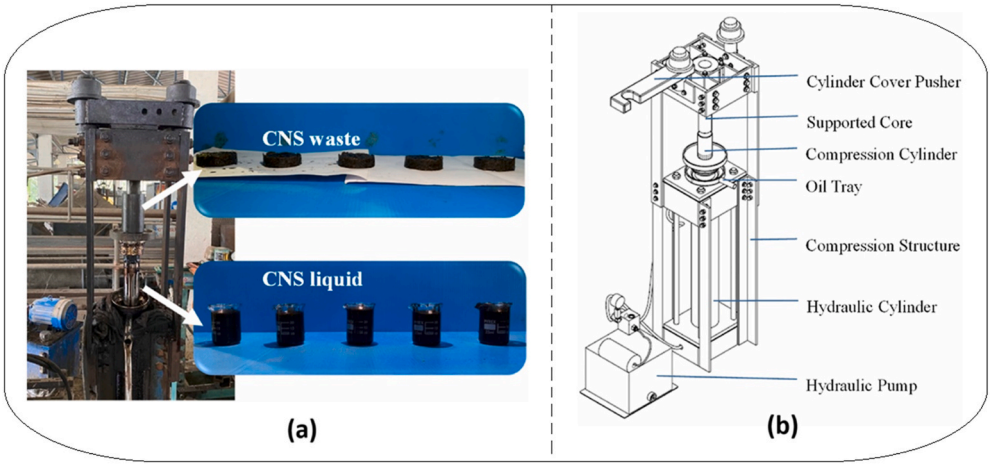

Fig. 4. (a)The liquid and waste of cashew nutshell obtained after cold pressing through the hydraulic cold-pressed extraction machine. (b) Components of the hydraulic cold-pressed extraction machine. Adapted with permission from Elsevier [80].

shells as a viable substrate for sustainable bioethanol production. In addition, Morales and colleagues [84] examined a comprehensive utilization of almond shells through a biorefinery method. They successfully extracted, isolated, and characterized the three primary components found in almond shells. The autohydrolysis technique allowed for the recovery of the hemicellulosic fraction, resulting in a liquor abundant in xylooligosaccharides (22.12 g/L). Following this, two distinct delignification methods alkaline and organosolv treatments were implemented to achieve lignin with an exceptionally high purity of around 90%, which can be utilized for various applications. The high cellulose content in the delignified materials was then processed through two approaches: one involved the production of CNCs, while the other focused on generating glucose through enzymatic hydrolysis, leaving behind a solid residue largely made up of lignin (approximately 78 wt%). Consequently, this biorefinery strategy could support a circular economy by enabling eco-friendly revaluation of all major components of almond shells.

Wang et al. [45] reported that enzymatic pretreatment using peanut shells can markedly enhance the nutritional and extraction efficiency of peanut oil. A hemicellulase–cellulase–pectinase blend (1:1:1, w/w/w) applied at 1% enzyme loading, with 7.5% peanut shell addition under mild conditions (pH 4.5, 55 °C, 3 h), boosted polyphenol levels from 25.54 mg GAE/kg to 169.84 mg GAE/kg and resveratrol from 0.15 mg/kg to 1.73 mg/kg, representing approximately 5.5- and 10.5-fold increases. The oil yield also improved substantially, rising from 57.8% to 82.5%, confirming the benefit of enzymatic and mechanical cell disruption (Fig. 5). Likewise, laccase-assisted extraction from groundnut shells combined with mechanical processing (18 h, 240 rpm, 62.6 °C drying) produced 35.2% cellulose microfibers and 29.5% lignin, illustrating an efficient and sustainable method for generating biomaterials suitable for biodegradable film production [85].

3.3.2. *Advanced extraction technologies*

Recently, the process of supercritical fluid extraction has garnered significant interest as a valuable substitute for conventional separation techniques. Cashew nutshell liquid (CNSL), commonly referred to as natural CNSL, is composed of various alkenyl phenols, with anacardic acid being the most abundant component. Owing to its potential industrial uses, Philip et al. successfully extracted anacardic acid from natural CNSL using supercritical carbon dioxide (SC-CO_2). This research determined the solubility of natural CNSL in SC-CO_2 across diverse conditions, including pressures of 100, 200, and 300 bar, temperatures of 40 and 50 °C, and CO_2 flow rates of 5, 10, and 15 g min−1. The optimal SC-CO_2 parameters identified were a temperature of 50 °C, pressure of 300 bar, and a flow rate of 5 g min−1. By using 3 g of CNSL with a solid adsorbent ratio of 1:2, it was possible to achieve a quantitative extraction of high purity anacardic acid (82% of total anacardic acid) from the crude CNSL in just 150 min. The extracted anacardic acid underwent evaluation using various spectroscopic and HPLC techniques, which confirmed its superior quality compared to the product obtained through traditional multi-step chemical processes [86].

Smith and colleagues [87] investigated the extraction of CNSL from cashew nut pericarp using SC-CO_2. Initial extractions conducted at temperatures between 40 and 60 °C, and pressures ranging from 14.7 to 29.4 MPa, resulted in low yields. However, implementing one or more intermediate depressurization stages during the extraction process significantly boosted the CNSL yield to as much as 94%. Most of the oil remained within the shell during depressurization but was captured during the following repressurization phase. The extracted CNSL displayed a distinct light brownish pink hue and showed no signs of polymerization or degradation. Utilizing the pressure profile extraction technique enhanced the potential yields of CNSL while substantially decreasing the CO_2 consumption for its extraction.

To manage dye pollution in the wastewater produced by the printing sector and to make use of walnut shell waste for economic advantages, a technology utilizing SC-CO_2 was created by Zhuang et al. [46] to generate porous biochar, which serves as a precursor for adsorption materials. Experiments were designed using various temperatures of 200, 300, and 400 °C, with durations of 20, 40, and 60 min, alongside a control group treated with N_2. Following this, KOH was applied to activate the biochar for adsorption purposes. Methylene blue dye was chosen as a standard to evaluate the adsorption potential of the biochar, and the kinetics of adsorption were analyzed using the gathered data. Findings revealed that SC-CO_2 pretreatment notably improved the efficiency and quantity of activated carbon. The specific surface area increased by 18%, while methylene blue adsorption capabilities rose by 23% when compared to the N_2 control group. Additionally, yields improved between 8% and 262%, with specific surface areas rising by 50% to 192% when compared to direct walnut shell activation. The pretreatment processes also produced phenol-rich bio-oil as a valuable by-product in the creation of biochar adsorption materials, enhancing economic potential.

Subcritical water technology has emerged as an eco-friendly approach for extracting active compounds and beneficial degradation products from waste generated by walnuts and pistachios. Čolnik et al. [88] explore the use of subcritical water extraction (SWE) to convert

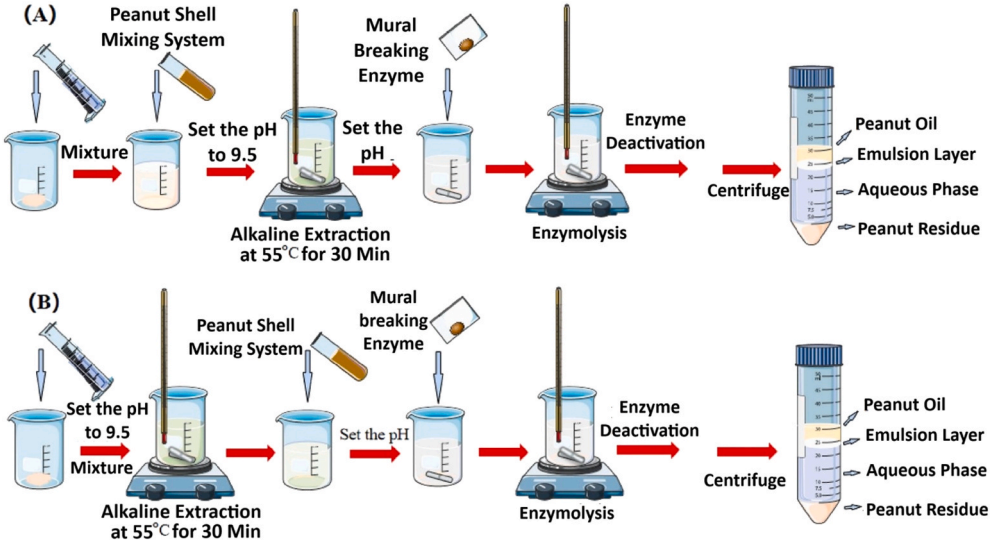

Fig. 5. Production process of peanut oil (PO) by aqueous enzymatic method (processes A and B). Adapted with permission from Elsevier [45].

pistachio and walnut shell waste into valuable components. They conducted SWE at temperatures ranging from 150 to 300 °C and reaction times of 15–60 min with a material:solvent ratio of 1:10 (g/mL), achieving extraction yields of 8–41% (Fig. 6). This study demonstrates the effective utilization of pistachio and walnut shell waste for obtaining a variety of valuable biochemicals. When employing subcritical water extraction, optimal yields were achieved for several key components. The maximum concentration of total phenols was observed in walnut shell extracts (127.08 mg GA/g) after processing at 300 °C for 15 min. Similarly, pistachio shell extracts yielded the highest levels of total flavonoids (10.18 mg QU/g) at 200 °C for 60 min, while walnut shells produced the greatest quantity of total carbohydrates (602.14 mg TCH/g) under the same conditions. The antioxidant capacity was also exceptionally high, reaching 91% in extracts from pistachio shells (300 °C, 60 min).

In comparison, conventional extraction with ethanol produced a respectable phenol content, up to 86.17 mg GA/g. However, detailed HPLC analysis identified a richer profile in the subcritical water extracts, which predominated in glucose, fructose, and their valuable degradation products: 5-hydroxymethylfurfural (5-HMF), furfural, and levulinic acid.

These findings conclude that subcritical water technology allows for

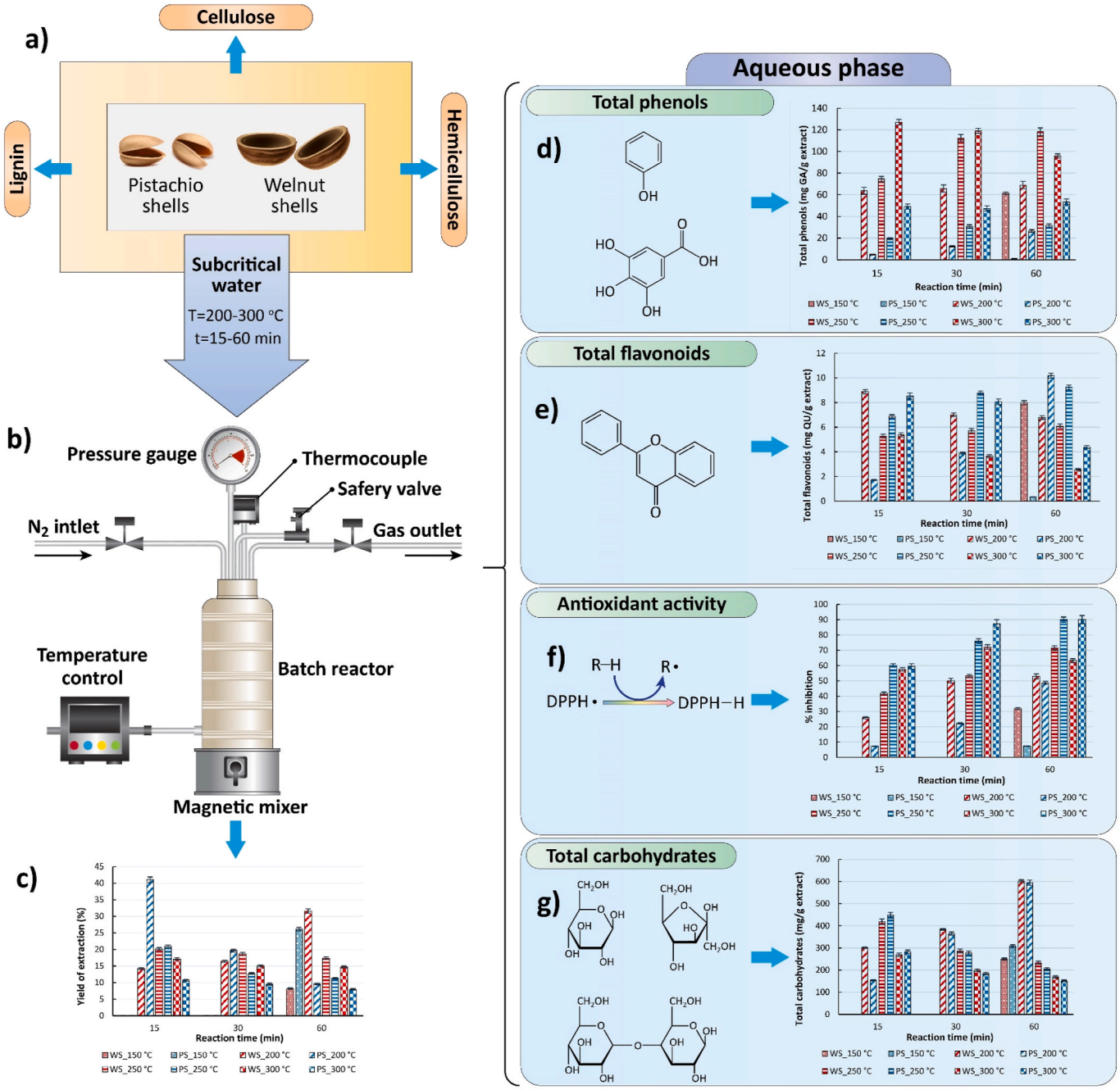

Fig. 6. (a) The Conversion of pistachio and walnut shell waste into valuable components with subcritical water (b) Scheme of apparatus for batch SWE. (c) The yield of extraction of pistachio and walnut shells (PS-pistachio shells; WS-walnut shells) (d) Content of total phenols in extracts of pistachio and walnut shells (e) Content of total flavonoids in extracts of pistachio and walnut shells (PS-pistachio shells; WS-walnut shells) (f) Antioxidant activity of pistachio and walnut shells (PS-pistachio shells; WS-walnut shells) (g) Total carbohydrate content (TCH) in the dry extract (PS: pistachio shells; WS: walnut shells). Adapted with permission from Elsevier [88].

a more thorough exploitation of biological waste materials than methods using organic solvents. It enhances the recovery of bioactive compounds like phenolics, leading to extracts with powerful antioxidant activity, while simultaneously creating valuable chemical byproducts.

4. Nutshell-derived biomaterials

Nutshells yield a diverse array of biomaterials, including cellulose, lignin, phenolic compounds, carbonaceous materials, microbial fermentation products, and other minor constituents, each with unique properties for sustainable applications. This section explores their roles in bioplastics, packaging, and other high-value products, emphasizing their functional contributions and potential in advancing a circular economy.

4.1. Cellulose and derivatives

Cellulose and its derivatives have garnered significant technological interest in sustainable packaging due to their biodegradability, renewability, biocompatibility, and the mechanical, thermal, and barrier properties they can provide to packaging materials. Various nutshells, such as those from almonds [89], walnuts [15], chestnuts [54], hazelnuts [17], peanuts [90], and pistachios [91] have been extensively used for extracting micro/nano cellulose. The quantity of cellulose in a vast range of nutshells is presented in Table 1. In this regard, cashew nutshell waste was effectively valorized into cellulose via modified low-concentration nutshell hydrolysis, yielding about 52 % with good crystallinity (≈59 %) and micro-sized particles (~0.23 μm). The extracted cellulose exhibits suitable thermal stability (~313 °C), a low density (~1.21 g/cm^3), and a well-defined crystalline structure, making it a promising sustainable raw material for bioplastic applications [92]. Nanocellulose, a popular cellulose derivative, is typically obtained through acid hydrolysis of the cellulose matrix [3]. However, more environmentally friendly methods, such as hydrothermal and microwave-assisted extraction, hydrotropic, and deep eutectic solvents (DES), have been developed for producing nanocrystalline cellulose from nutshells [62,63,69]. Görgüç et al. [93] conducted a study on different techniques for producing nanocellulose from chestnut shells using acid hydrolysis, microwave-assisted-acid hydrolysis, hydrotropic solvent, and DES (Fig. 7A). It was found that DES and microwave-assisted treatment resulted in the highest (99 %) and lowest (34 %) extraction yield, respectively. The highest crystallinity (>81 %) was obtained for the samples treated with acids, confirming that the traditional and microwave-assisted acid hydrolysis generated nanocrystalline cellulose while the hydrotropic and DES led to the production of the cellulose nanofibrils. Rheological analysis showed that all the nanocellulose samples had a gel-like behavior (storage modulus (G′) > loss modulus (G″)), while the sample treated with DES showed the highest viscosity (843.6 cp) and zeta potential (−23.7 mV). Cellulose, found in lignocellulosic biomass like nut shells, is typically surrounded by lignin and hemicellulose. Removing lignin and hemicellulose is necessary to access the cellulose. While these materials are often overlooked during cellulose extraction, there is a growing interest in fully utilizing the waste. Morales et al. [84] conducted a study on extracting nanocellulose from walnut shells using a multi-product biorefinery approach, which involved recovering hemicellulose and lignin. The process included

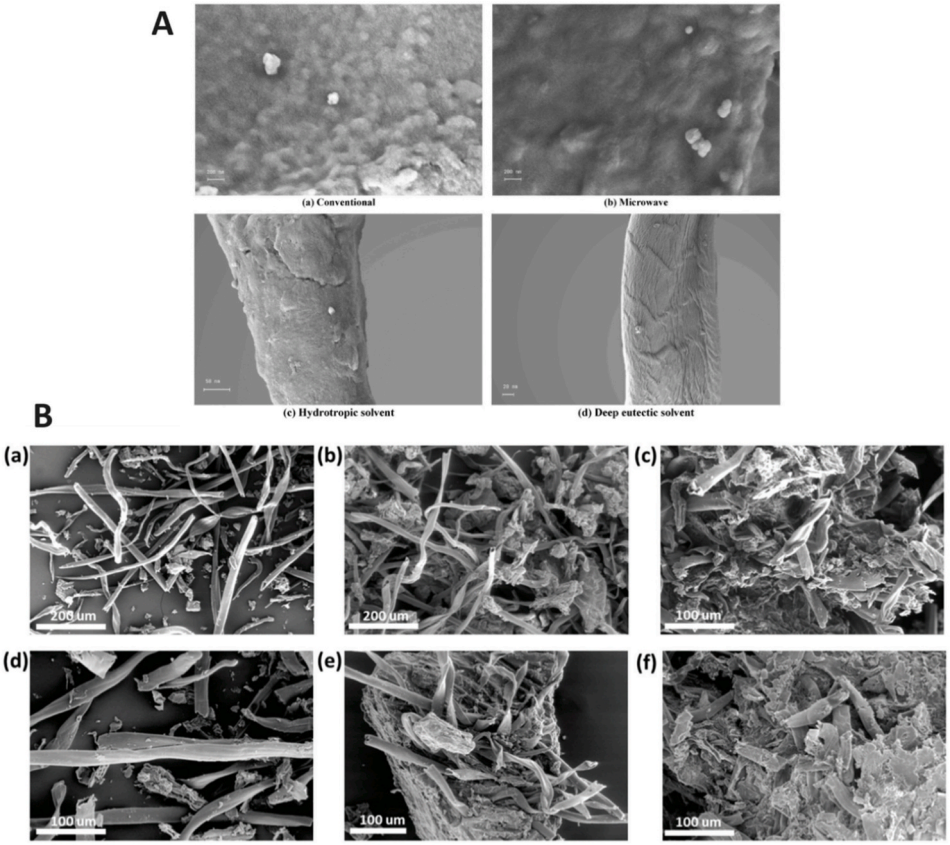

Fig. 7. (A). Morphological structure of nanocellulose extracted from chestnut shells through different techniques of conventional (acid hydrolysis) (a), microwave-assisted acid hydrolysis (b), hydrotropic solvent (choline chloride: oxalic acid) (c), and deep eutectic solvent (50% aqueous p-toluenesulfonic acid) (d) [93]. **(B).** Morphological structure of the raw chestnut shells (a) and the solid reside containing cellulose, hemicellulose, and lignin remaining after phenolic compound extraction using microwave-assisted water extraction at 65 °C, 30 min (b), microwave-assisted choline chloride-oxalic acid at 65 °C, 30 min (c), microwave-assisted choline chloride-oxalic acid dihydrate at 65 °C, 30 min (d), microwave-assisted choline chloride-oxalic acid dihydrate at 75 °C, 30 min (e), microwave-assisted choline chloride-oxalic acid dihydrate at 85 °C, 30 min (f) [96].

extracting hemicellulose using microwave-assisted and hydrothermal methods, isolating lignin with aqueous ethanol treatment, and recovering nanocellulose through bleaching and acetic acid treatment. The nano/micro cellulose derived from nutshells has been employed as nanofillers for different packaging composites. For example, the nanocellulose extracted from pistachio shells using alkaline hydrogen peroxide and subsequent hydrochloric acid hydrolysis was integrated into carrageenan films [94]. The alkaline: hydrogen peroxide ratio of 4:4.4 resulted in removing the impurities, enhancing the crystallinity, and improving the thermal properties of nanocellulose. The incorporation of nanocrystalline cellulose into the carrageenan films improved mechanical, barrier (towards oxygen and UV), and thermal properties. Similarly, the nanocellulose extracted from the almond shell was employed as a reinforcement agent in polyvinyl alcohol (PVA) composites, which could significantly improve the thermal and mechanical properties [95]. Despite the extensive use of cellulose and its derivatives from various sources in packaging materials, there is limited research on the utilization of nutshell-derived cellulose in packaging, highlighting the need for further exploration of nutshells as valuable sources of cellulose in sustainable packaging.

4.2. Lignin

Lignin is a complex organic polymer found in cell walls that contributes to the structural integrity and rigidity of materials. Nutshells have a higher lignin content (26-45 %) compared to woody biomass (14-25 %) and grass biomass (15-20 %). This naturally occurring aromatic biopolymer with a polyphenolic structure has the potential as an ingredient in packaging due to its antioxidant, antibacterial, and UV-barrier properties. Conventionally, lignin can be recovered during the cellulose extraction. For example, lignin (1.5 mg/mL) was recovered from the black liquor discarded after the alkaline treatment of peanut shells by acid precipitation during cellulose extraction [97]. Alternative methods of delignification, such as reductive catalytic fractionation (lignin-first strategy) and DES delignification, as well as utilization of intensification techniques such as ultrasound and microwave technology, are emerging in the biorefinery industry. Nishide et al. [98] explored the direct catalytic hydrogenolysis of the walnut shell using three organosolv systems (formic acid-acetic acid-water, ethanol or methanol-hydrochloric acid, and sulfuric acid-methanol-formaldehyde) for protective extraction of the lignin. The results indicated that ethanol-hydrochloric acid could sufficiently fractionate the walnut shell without protecting the lignin. However, the sulfuric acid-methanol-formaldehyde system was ideal for the extraction of lignin, leading to 64% recovery at 170 °C. Another study by Argenziano et al. [77] explored the ball milling and DES treatment (choline chloride: lactic acid, 120 °C, 24 h) to extract bioactive lignin from various nut shells, including chestnuts, hazelnuts, peanuts, pecan nuts, and pistachio. The highest recovery and extraction yield was reported for pistachio (38 and 27 %) and hazelnut shell (36 and 25 %) followed by pecan nut (34, 19 %), peanut (34, 19 %), and chestnut (27, 19 %). These studies showed that pecan nut (~0.55 mg of gallic acid equivalent (GAE)/mg of lignin) and chestnut shells-derived lignin (~0.3 mg of GAE/mg of lignin) were rich in antioxidant compounds. Additionally, the extraction of lignin from almond and walnut shells through a single delignification using alkaline or organosolv as well as a two-step biorefinery with a hydrothermal pretreatment (autohydrolysis) before delignification showed different results composition, molecular weights, and total phenolic compounds based on the source and biorefinery method. Integration of autohydrolysis in the biorefinery process has been shown to enhance the purity and molecular weight of the extracted lignin.

Despite its promising antioxidant, antimicrobial, and UV-barrier properties, the use of nutshell-derived lignin in polymer and packaging applications faces several practical challenges. Lignin purification remains a key limitation, as extraction methods can lead to significant variability in molecular weight distribution, functional group content, and residual impurities, depending on both the feedstock and delignification route. Such heterogeneity can negatively affect reproducibility and process control at larger scales [99]. In addition, the intrinsic polarity and aromatic nature of lignin often result in poor compatibility and dispersion within hydrophobic polymer matrices, potentially leading to phase separation and compromised mechanical performance. Chemical modification, compatibilizers, or blending strategies are therefore frequently required to enhance interfacial adhesion, thereby increasing processing complexity and cost [100]. Addressing these challenges through controlled fractionation, tailored surface modification, and structure–property correlations is essential to enable the broader use of nutshell-derived lignin in sustainable packaging materials.

4.3. Phenolic compounds

The increased consumer awareness regarding food safety, quality, and health issues has sparked significant research into isolating bioactive compounds. Nutshells are a promising source of bioactive compounds that can be used as additives in smart food packaging. For example, chestnut shells are found to contain polyphenols (2.5-5.2 % wt) such as tannins, phenolic acids, and flavonoids [101], while peanut shells are rich in polyphenols (428-740 mg GAE/g), flavonoids (143-568 mg QE/g), luteolin (0.25-1.12 mg/g), carotene, and isosaponaretin [90]. Hazelnut shells also contain various phenolic compounds including phenolic acids, flavonoids, tannins, diarylheptanoids, and lignans [17]. Cashew nutshell is another valuable source of phenolic compounds, with the dark liquid fraction obtained through cold pressing or thermomechanical process containing cardanol (10 %), cardol (15-20 %), anacardic acid (60-65 %), and traces of 2-methyl cardol [102]. These compounds have been found to have numerous health benefits, including reducing the risk of disease and protecting against free radicals, which can lead to conditions like cancer, stroke, inflammation, and neurodegenerative diseases [102]. The extraction process is crucial, and newer, greener extraction technologies have been developed to improve efficiency and sustainability. While traditional methods like Soxhlet extraction and maceration have been widely used for extracting bioactive compounds from natural sources, the alternative greener methods produce high-quality extracts at lower costs, addressing the limitations of traditional extraction methods. Different extraction methods, such as subcritical water, supercritical fluids, ultrasonic and microwave extraction, the utilization of biobased and green solvents, and membrane separation, have been used to extract the bioactive compounds from nutshells [88,103].

In an investigation by Nyirenda et al. [104], the extraction of cashew nutshell liquid by different solvents of petroleum ether, hexane, and ethanol yielded 24.6, 38.2, and 40.1 %, respectively. These extracts contained various phytochemicals such as alkaloids, phenols, tannins, flavonoids, terpenoids, carboxylic acids, saponins, amino acids, proteins, and carbohydrates [105]. Besides, the extraction of cashew shell liquid by supercritical CO_2 extraction yielded relatively less cashew shell liquid than the conventional solvent extraction. However, it could reduce solvent consumption without the need for further processing, such as filtration. In another study, ultrasonic-assisted DES (choline chloride: n-propanol) extraction was employed for the extraction of the ellagic acid, a valuable phenolic compound from chestnut shells. The purity of obtained ellagic acid (85.6 %) was more than double the quantity of those obtained by ethanol extraction [101]. Similarly, the impact of microwave-assisted DES extraction was investigated on the microstructure of the solid residue of the walnut shell after recovery of phenolic compounds [106]. Among the different DES containing choline chloride and different organic acids such as oxalic, malic, levulinic, citric acid, and oxalic acid dihydrate, the choline chloride-oxalic acid dihydrate system treated at 85 °C for 30 min resulted in the highest TPC. While the untreated chestnut shell had a fibrous structure with a smooth

surface, microwave-assisted extraction, whether with water or DES extraction of phenolic, changed the microstructure of chestnut shells. However, the effect of water extraction was less severe than that of DES extraction. In a similar treatment condition (65 °C for 30 min of microwave extraction), the effect of choline chloride-oxalic acid on the morphological changes of chestnut shells was more evident than choline chloride-oxalic acid chloride (Fig. 7B). However, the increasing temperature and time of extraction intensified the destruction of the remaining residue [99].

In comparison with the efficiency of Soxhlet, ultrasonic extraction, and their combination in the extraction of bioactive compounds of hazelnut shell, the highest extraction yield of 15.4 % was obtained using the combined extraction method with methanol for 16 h. The highest antioxidant activity of 0.0508 mg TE/mL and phenolic content of 0.188 mg GAE/mL resulted in using only ultrasonic extraction with methanol and Soxhlet extraction with ethanol for 8 h, respectively. Amongst different factors affecting the extraction efficiency and antioxidant capacity, the solvent type was reported as the most significant factor [107]. Conidi et al. [108] employed membrane-assisted separation (ultrafiltration, nanofiltration, and reverse osmosis) for the recovery of gallic, ellagic, and tannic acids extracted from the chestnut shell using aqueous/ethanolic solution. At the optimum condition of extraction (temperature of 50 °C and water-to-solid ratio of 9 mL/g), the total phenolic content (TPC) of the extract was 6.6 g GAE/L. The sequential membrane filtrations resulted in complete removal of suspended solids by ultrafiltration, obtaining a retentate containing 30.16 and 6.2 g/L of TPC and flavonoids, respectively enriched in tannic (1.49 g/L) and ellagic acids (3.83 g/L) by nanofiltration as well as retentate enriched in gallic acid (118.37 mg/L) obtained through reverse osmosis. In another study, microwave-assisted-water extraction of tannin from chestnut shell resulted in 25 % of extraction yield with 344 mg GAE/g of phenolic compounds and 1296 mg CE/g of condensed tannin content at the optimum condition of liquid-to-solid ratio of 50 mL/g, temperature of 107 °C and extraction time of 5 min. Table 4 summarizes the key findings of different extraction techniques to recover phenolic compounds from various nutshell biomass.

Although phenolic compounds extracted from nutshells offer strong antioxidant and antimicrobial functionality, their practical incorporation into polymer matrices presents notable challenges [114–116]. Purification of phenolic extracts is often complicated by the co-extraction of sugars, proteins, and other low-molecular-weight compounds, which can affect stability, sensory properties, and regulatory acceptance for food-contact applications [117,118]. In addition, many phenolic compounds are sensitive to heat, oxygen, and light, leading to degradation or loss of activity during polymer processing and long-term storage. Compatibility with polymer matrices also remains a critical issue, as phenolics may migrate, aggregate, or plasticize the polymer, resulting in altered mechanical or barrier properties [119]. Encapsulation strategies, controlled-release systems, and covalent or physical immobilization within polymer networks are therefore increasingly explored to improve stability, compatibility, and functional longevity in packaging applications [120].

4.4. Carbonaceous materials

Carbonaceous materials derived from biomass, such as biochar, activated carbon, and carbon dots, are gaining researchers' interest in packaging formulation. While they have been traditionally used as absorbents for pollution, they have the potential to serve as absorbents, biosensors, and fillers in biomaterial processing for packaging purposes. This can lead to reduced weight and enhanced mechanical and electrical conductivity of composites [121,122].

Biochar, produced through the direct pyrolysis of biomass under an inert atmosphere, can serve as a precursor for the development of activated carbon and carbon quantum dots. Biochar offers abundant surface functional groups, activated carbon provides a unique surface, and

Table 4
A summary of key findings on the extraction of phenolics for nutshells biomass.

Nutshells source	Extraction Method	Yield	Functionalities	Ref.
Chestnut	Ultrasonic-assisted DES (choline chloride: n-propanol) Opt.: S-to-L ratio 40 mg/mL; ultrasonic power 200 W; t 70 min	Ellagic acid: 4.64 mg/g	Antioxidant: IC50 0.309 mg/mL Antibacterial: MIC 250 μg/mL, DIZ 17.60 μg/mL	(101)
Almond	Microwave-assisted ethanol:water Opt.: T 80 °C; t 57 min; pH 8; 70% (v/v) ethanol	TPC: 5.64 mg GAE/g Flavonoids: 1.42 mg CE/g Polysaccharides: 1.59 mg glucose/g	Antioxidant: 2.82 mg AAE/g (DPPH, FRAP)	(70)
Chestnut	Solvent extraction Opt.: Ethanol 70%; T 50 °C; t 2 h Alkaline extraction Opt.: NaOH 1.68 M; L-to-S 49.86 mL/g; t 4.01 h	TPC: 9–0.54 mg GAE/g (ethanol, before/after purification) TPC: 90–0.25 mg GAE/g (alkaline, before/after purification)	Antioxidant: 90% and 80% DPPH scavenging (20 μg/mL, ethanol and alkaline) 95% and 85% ABTS scavenging (70 μg/mL, ethanol and alkaline)	(109)
Chestnut	Microwave-assisted alkaline extraction Opt.: Microwave power 800 W; t 12 min; NaOH 0.115 M	TPC: 274.09 mg GAE/g	Antioxidant: 1070.13 mg TE/g (CUPRAC), 543.94 mg TE/g (DPPH) Antibacterial: Inhibits *S. aureus* growth Pigment: Color value 41.48 (brown melanin)	(110)
Peanut	Ultrasonic-assisted solvent extraction (80% ethanol, methanol, water) Opt.: Ethanol; t 10 min	TPC: 37.9 mg GAE/g β-carotene: 53.3 mg TE/g	Antioxidant: 50.5 mg TE/g (DPPH), 21.3 mg TE/g (ABTS), 29.7 mg TE/g (FRAP)	(111)
Pistachio and Walnut	Subcritical water extraction Opt.: T 150–300 °C; t 15–60 min Organic solvents: Acetone, 50% acetone, ethanol	TPC: 127.08 mg GAE/g (walnut, T 300 °C, t 15 min) TPC: 86.17 mg GAE/g (ethanol extract) Flavonoids: 10.18 mg QUE/g (pistachio, T 200 °C, t 60 min) Carbohydrates: 602.14 mg TCH/g (walnut, T 200 °C, t 60 min)	Antioxidant: 91% DPPH scavenging (pistachio, T 300 °C, t 60 min)	(88)
Walnut	Microwave-assisted solvent extraction Opt.: 30% aqueous ethanol; t 120 s; microwave power 670 W	TPC: 249.7 mg GAE/g	Antioxidant: 668.3 μmol TE/g (ORAC), 59.20 mg TE/g (DPPH), 96.26 mg TE/g (TEAC)	(112)
Walnut	Organic solvent extraction (ethanol, acetone,	TPC: 2–17 mg GAE/g	Antioxidant: 3–28 mg TE/g (DPPH), 14000	(113)

(*continued on next page*)

Table 4 (*continued*)

Nutshells source	Extraction Method	Yield	Functionalities	Ref.
	methanol mixtures with water; T 40–60 °C; S-to-L ratio 1/20 g/mL)		μmol TE/100 g (ORAC)	

Notes: Abbreviations: T: Temperature, t: time, TPC: Total phenolic content, GAE: Gallic acid equivalent, CE: Catechin equivalent, QUE: Quercetin equivalent, TCH: Total carbohydrate, ORAC: Oxygen radical absorbance capacity, TEAC: Trolox equivalent antioxidant capacity, DPPH: 2,2-diphenyl-1-picrylhydrazyl, ABTS: 2,2′-azino-bis(3-ethylbenzothiazoline-6-sulfonic acid), FRAP: Ferric reducing antioxidant power, CUPRAC: Cupric reducing antioxidant capacity, MIC: Minimum inhibitory concentration, DIZ: Diameter of inhibition zone, AAE: Ascorbic acid equivalent, S-to-L: Solid-to-liquid ratio, L-to-S: Liquid-to-solid ratio.

carbon quantum dots offer a nanostructured material (size <10 nm) with water solubility due to hydroxyl and carboxyl groups on the surface and high quantum yield for sensing, bioactive compounds delivery among others. Despite limited research on the use of carbonaceous materials in food packaging, there is a growing interest in their potential benefits, such as acting as barriers to oxygen and moisture to prevent oxidation and microbial contamination in food products. In addition, carbon dots can improve the mechanical properties of packaging and provide antibacterial properties. Surface functionalization of quantum dots by enhancing chemical reactivity and selectivity improves sensing capabilities in smart packaging [123,124]. Nutshells, as a type of lignocellulosic biomass, show promise as a source for producing carbonaceous materials. There are several studies focused on the production of nutshell-derived carbonaceous materials in different applications. For example, walnut shells were thermally treated with Ni-metal organic framework (MOF) to obtain an efficient biochar for the extraction and detection of pesticides in wheat flour [125]. In another study, highly porous nitrogen and sulfur-doped activated carbon were produced using walnut shells via KOH activation and post-processing treatment by thioacetamide for a solid-state supercapacitor device [126]. Walnut shells have also been pretreated using supercritical CO_2 to produce biochar adsorption material to treat dye in the printing industry's wastewater [46].

Sunflower seed shells were employed to produce microporous activated carbon using a KOH activating agent to capture water vapors from humid air [127]. In this context, some studies integrated green methods to produce carbonaceous materials. Qiu et al. [128] investigated the biochar production through microwave-assisted pyrolysis of peanut shells. Biochar with micropores was first obtained at a pyrolysis temperature of 800 °C, microwave power of 500 W, and residence time of 2.0 h. However, increasing the pyrolysis temperature (700-950 °C), microwave power (350-550 W), and residence time (0.5-3 h) decreases biochar yield while increasing specific surface area. Besides, peanut shells-derived carbon dots containing Si produced through the one-pot hydrothermal method showed peroxidase-like activity, which was suggested for cysteine detection.

Regarding the carbonaceous material application in polymer processing, the biochar produced by cashew nutshells through pyrolysis at 400 °C for 3 h was utilized as the filler (10% wt) in the polyester composite. The cashew nutshells-derived biochar resulted in significant improvement in the mechanical and erosion resistance of the composite [129]. Similarly, annealed biochar derived through pyrolysis of peanut shells at 400 °C, followed by annealing at 1000-1050 °C with $Fe(NO_3)_3$ as a catalyst, was utilized for reinforcement of epoxy resin-aloe vera fiber composites. The addition of 2 % wt peanut biochar resulted in the highest tensile (160 MPa) and flexural strength (212 MPa), while the composite containing 5% peanut biochar showed the highest thermal conductivity (0.54 w/Mk) and dielectric constant (7.2). However, there is a scarcity of research on using nutshell-derived carbonaceous materials for packaging applications. Due to the high potential of these materials to meet the requirements of sustainable and smart packaging, it seems that they could lead to the expansion of smart packaging and sensors, although further studies are needed to assess the risks of their use.

Beyond their role as fillers and adsorbents, nutshell-derived carbon dots and phenolic-rich carbonaceous materials show strong potential for smart packaging applications, particularly as freshness indicators and sensing elements. Carbon dots exhibit intrinsic fluorescence, redox activity, and high surface reactivity, enabling their use for real-time detection of food spoilage markers, including pH changes, biogenic amines, and microbial metabolites [121,123]. Similarly, phenolic compounds integrated into packaging matrices can act as antioxidant or antimicrobial indicators, responding to oxidation or microbial growth through measurable optical or chemical changes. Despite these promising functionalities, current studies are largely limited to laboratory-scale demonstrations, and challenges remain regarding signal stability, sensitivity under real food storage conditions, and safe incorporation into polymer matrices [115,130]. Addressing these limitations through material stabilization strategies, controlled release systems, and validation in real packaging environments will be essential to advance nutshell-derived carbonaceous materials toward practical smart packaging applications.

4.5. Microbial fermentation products

Researchers have shown interest in bacterial fermentation products like bacterial cellulose and lactic acid for both environmental reasons and product quality. However, the high cost of fermentation media has been a limiting factor despite the high purity of bacterial products. Nutshell biomass is being considered as a potentially cost-effective media for bacterial fermentation to produce valuable biopolymers for the packaging industry. The production of fermentable sugars through acid, alkaline, or enzymatic hydrolysis can result in the generation of fermentation inhibitors. Therefore, it is important to assess the suitability of nutshell hydrolysates for fermentation before use. Lee et al. [131] utilized chestnut shell hydrolysates obtained through alkaline and enzymatic hydrolysis as a carbon source for bacterial cellulose production by *Gluconacetobacter xylinus* ATCC 53524. The chestnut hydrolysates, containing 40 g/L of glucose and 1.9 g/L of acetic acid, led to a 1.3-fold increase in bacterial cellulose concentration (16.7 g/L) compared to the control group. Chestnut shells have also been investigated for lactic acid production, which is a monomer of polylactic acid, a promising biobased polymer for food packaging [36]. In this regard, a three-step process involving alkaline (KOH) pretreatment, enzymatic saccharification, and lactic acid fermentation was conducted to produce lactic acid. The alkaline pretreatment effectively removed lignin as a fermentation inhibitor and increased the cellulose content. Fermentation with *Lacticaseibacillus rhamnosus* resulted in 21, 23, and 27 % improvements in lactic acid production, glucose consumption, and cell growth compared to the control group. Similarly, chestnut waste (including shells, leaves, burrs, and pruned material) was treated with diluted sulfuric acid (3% w/v and 130 °C) to solubilize the fermentable sugars for lactic acid production. Although acid hydrolysis generated inhibitory factors like furan derivatives, phenolic compounds, and aliphatic acids, activated charcoal (5 % w/v) was able to remove about 95 % of the color. Fermentation of chestnut waste hydrolysates using *Lactobacillus plantarum, Lactobacillus pentosus*, and *Lactococcus lactis* produced lactic acids with yields of 0.89, 0.92, and 0.83 g/L h, respectively. Due to the rich source of cellulose and hemicellulose that provide fermentable monomers, the valorization of nut shells into biopolymers such as PHA and PLA with performances competitive with synthetic polymers will significantly contribute to the expansion and commercialization of sustainable packaging.

4.6. Other biomaterials

Nutshells contain other valuable compounds that can be beneficial in the food and packaging industry, such as polysaccharides, proteins, minerals, vitamins, and lipids. Research has shown that pistachio shells are rich in protein, minerals, antioxidants, and vitamins. The oil extracted from pistachio shells contains significant amounts of α-pinene and α-terpineol [132,133]. Nutshells also serve as a potential source of bioactive amino acids. For example, in the study by Imran et al. [111], peanut shells composed of 4 % of protein and 2.18 mg/g (dry basis) of essential amino acids including valine (0.55 mg/g), leucine (0.41 mg/g), isoleucine (0.39 mg/g), lysine (0.27 mg/g), phenylalanine (0.20 mg/g), histidine (0.18 mg/g), threonine (0.14 mg/g), and methionine (0.04 mg/g). Whereas the total non-essential amino acids contained 3.26 mg/g (dry basis), mostly including proline (0.85 mg/g), alanine (0.76 mg/g), aspartic acid (0.68 mg/g), and arginine (0.56 mg/g). Pectin was extracted from peanut shells through a combination of thermal treatment (70-90 °C for 2 h) and ultrasonic-assisted extraction (40kHz/1500 W, 10-30 min) [134]. Pectin extracted at 80 °C with ultrasound treatment for 10 min had the highest pectin extraction yield of 1.61 % with 88.64 % esterification degree and 52.12 % galacturonic acid, like commercial pectin. In another study, different approaches of enzymatic, microwave, and pulsed electric field extraction were compared for the extraction of soluble dietary fiber from peanut shells [45]. Dietary fibers extracted using pulsed electric field and microwave exhibited superior properties compared to enzymatic extraction, including a more complex structure and higher thermal stability. Dietary fibers extracted using a pulsed electric field showed the lowest molecular weight, strongest gelation properties, oil and water holding capacity, glucose and cholesterol adsorption capacity, as well as pancreatic lipase activity inhibition.

In another study by Cebin et al. [135] hemicellulose (xylooligosaccharides) was produced from walnut shells through alkaline treatment for delignification. This process resulted in a significant increase in the extraction of xylan, which was further hydrolyzed into xylotriose, xylobiose, and xylose using GH11 *endo-xylanase*. However, despite their valuable dietary fiber content, nutshells have not been fully exploited. Therefore, it is suggested that due to the importance of polysaccharides in the development of biodegradable packaging, these biomaterials should be considered from the perspective of nutshells biorefinery.

5. Applications of nutshells-derived biomaterials

Nutshells have emerged as valuable raw materials for a wide range of industrial applications, including sustainable packaging, polymer reinforcement, environmental remediation, and the manufacture of lignocellulosic panels. Their rich lignocellulosic composition confers mechanical strength, durability, and biodegradability, making them well-suited for the development of eco-friendly materials. For example, Fig. 8 illustrates the successful valorization of cashew nutshell residues into sustainable particleboards using a bio-based phenolic resin derived from cashew nutshell liquid (CNSL). The resulting panels exhibited satisfactory mechanical performance, with modulus of elasticity values ranging from 707 to 1866 MPa and modulus of rupture values ranging from 6.4 to 14.3 MPa. The incorporation of sugarcane bagasse further improved mechanical properties, while the CNSL-based resin enabled low formaldehyde emissions (E1 class) and 20–30% biodegradability within 30 days [102].

Beyond structural and panel applications, nutshell-derived biomaterials are increasingly being explored for biodegradable films and food packaging systems, where their functional molecules can play multiple roles as polymer matrices, reinforcements, and active additives.

This section discusses the utilization of nutshell-derived molecules as key components in biodegradable films, including polymer matrices, reinforcing fillers, functional additives (such as antioxidants, antibacterial agents, and UV-barrier agents), plasticizers, and biochar. These applications demonstrate the potential of nutshell-based biomaterials to enhance the performance of sustainable packaging while supporting eco-friendly material development and a circular economy. Table 5 summarizes the physicochemical, functional, and biological properties of biodegradable composites incorporating nutshell-derived biomaterials for food packaging applications.

5.1. Film matrix

The production of biodegradable plastics is possible using a variety of renewable agricultural resources including plant-based polysaccharides, proteins, lipids, and polyesters. Among these, polysaccharides such as cellulose and starch are an important focus in the research of bioplastics since they are widely available in nature [113,153,154]. Nutshells have been widely used to develop biocomposite films in the form of intact powders or after the extraction of their main biopolymers. In this regard, Harini et al. [57] explored the development of thermoplastic starch films made from cashew nut shells (CNS) and reinforced with walnut shell cellulose (WNC), and the impact of extracts from *Punica granatum* peels on their antimicrobial properties. The researchers found that CNS starch has a moderate amylose content of 26.32 % and a thermal degradation temperature of 310 °C. The 2 %

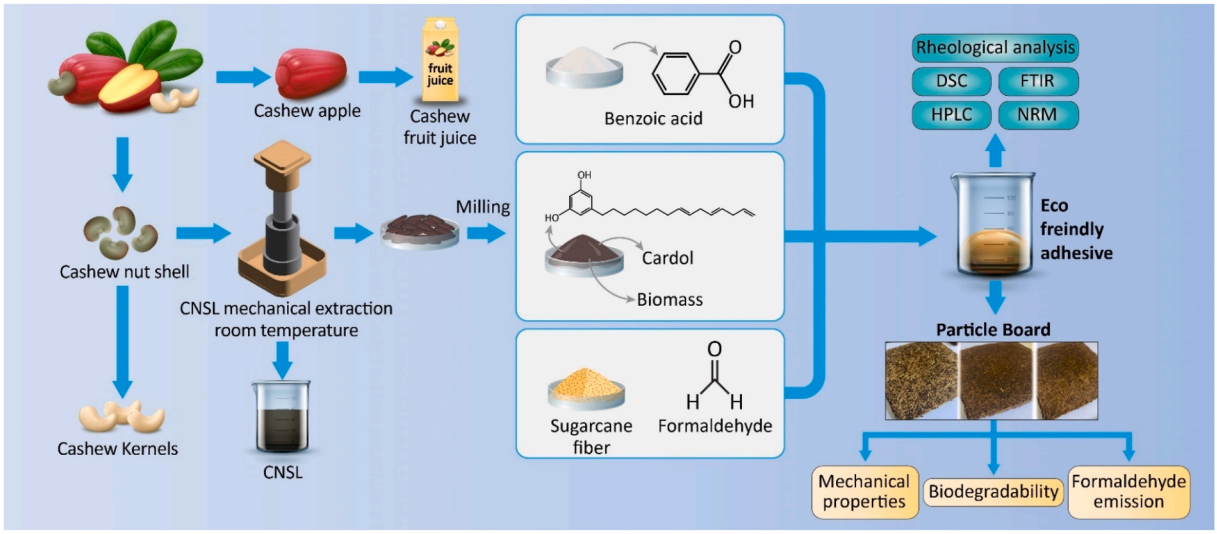

Fig. 8. Proposed particleboard production process from cashew nutshells. Reproduced from Ref. [102] with permission.

Table 5
Physicochemical, functional, and biological studies of composites containing nutshells derived biomaterials in food packaging.

Nutshell-derived biomaterial	Treatment	Loading	Polymer Matrix	Results	References
Pecan nutshell (PNS)	Ethanol extraction, ball milling (30–120 min)	50% w/w	Itaconic anhydride-grafted Poly(lactic acid) (MPLA)	Enhanced crystallinity (30% increase vs. neat MPLA) improved thermal-oxidative stability and maintained mechanical properties. Thermal annealing increased HDT up to 130 °C. PNS promoted biodegradation by increasing water uptake and surface erosion during soil burial.	(136)
Pecan nutshell (PNS)	Chloroform extraction (Fatty acids removed)	5, 7.5% w/w	Poly(lactic acid) (PLA)	Increased tensile and flexural modulus and impact strength compared to neat PLA, especially with treated PNS. Improved adhesion (SEM) with treated PNS but reduced thermal stability with higher PNS content. FTIR confirmed the removal of carbonyl and methyl groups in treated PNS. Suitable for rigid packaging.	(137)
Pecan nutshell extract	Ethanol extraction	~0.20% w/w	Whey Protein	Films with pecan nutshell extract had reduced flexibility (lower elongation at break, higher Young's modulus) but increased tensile strength.	(72)
Ground nutshell (GNS) Powder	Mechanical (Grinding, assumed)	0–25% w/w	Poly(vinyl alcohol) (PVA)	Increased crystallinity index and tensile strength (26.6 MPa to 37.8 MPa at 25% GNS). Enhanced thermal stability (degradation temperature from 287 °C to 337 °C). Decreased elongation at break (49.7% to 2.1% at 25% GNS). FTIR confirmed complete GNS dispersion. Suitable for packaging applications.	(138)
Walnut shell powder	None	2–8% w/w	Poly(lactic acid) (PLA)	Tensile strength decreased (vs. neat PLA) but increased with higher filler load (87.29 MPa at 8% vs. 63.73 MPa at 2%). Flexural strength followed a similar trend (106.5 MPa neat PLA to 87.29 MPa at 8%). Walnut shells increased impact strength by 67%.	(139)
Almond shell powder (ASP)	Mechanical (Grinding, sieving, drying at 80 °C)	35–50% w/w	Poly(butylene succinate-co-butylene adipate) (PBSA)	Increased HDT (~24%), flexural strength (125%), and tensile/flexural moduli (368–385%) with 50% ASP and 5% MA-g-PBSA compatibilizer. Reduced complex viscosity (~65% at 0.1 s^{-1}). Enhanced crystallinity and interfacial adhesion (SEM). Suitable for rigid single-use packaging.	(140)
Walnut shell powder (WSP)	Mechanical (Drying, grinding, sieving)	5–10% w/w (assumed)	Starch	Enhanced water resistance (42.8% TSM vs. 56% plain starch), hydrophobicity (contact angle 46.63° vs. 29.8°), crystallinity (19.12% increase), and hardness. Biodegradation reduced to 30.30% (vs. 65.78% plain starch). FTIR confirmed uniform WSP dispersion.	(78)
Walnut shell powder (WSP)	Mechanical (Grinding, assumed)	12% w/w	Carboxymethyl Cellulose (CMC)	Tensile strength 18.53 MPa, fracture strain 78.11%, water stability (no decomposition after 7 days), UV-blocking (99.22% UVA, 99.95% UVB), 82.23% optical transmittance, and 100% biodegradation in 2 days (rainy conditions). Recyclable under alkaline conditions.	(79)
Peanut shell powder (PSP)	Mechanical (Grinding, assumed)	5–15% w/w	Polyamide 6 (PA6)	Maintained matrix density and crystallinity up to 15% PSP. Structural stability confirmed by FTIR, XRD, DSC, TGA, and MEB. Crystallinity decreased slightly at 20% PSP, suggesting structural changes at higher loadings.	(90)
Cashew nut Shell powder (CNS)	Mechanical (Grinding, assumed)	20% w/w	Poly(vinyl alcohol) (PVA)	Tensile strength increased to 22 MPa (130% increase), tensile modulus to 264 MPa, thermal stability to 215 °C (43% increase), and 93% bacterial growth reduction. Enhanced biodegradability and cellulose content confirmed by XRD.	(141)
Almond shell powder (ASP)	Mechanical (Grinding, assumed)	5–30% w/w	Starch	Increased Young's modulus, reduced impact strength and elongation at break. Brownish aesthetics suitable for toys and packaging. Filler content had greater impact than particle size (<0.05 mm to 0.125–0.250 mm).	(89)
Torrefied almond shell (TAS)	Torrefaction, mechanical (Grinding, sieving)	0–50% w/w	Polypropylene-Polyethylene (PP-PE) blend	Increased flexural and tensile moduli but decreased strength, elongation, and toughness with higher TAS loading. HDT increased by 10 °C at 30% loading with smaller particles (100–120 mesh). SEM confirmed good adhesion. Crystallinity unaffected by particle size or loading.	(142)
Peanut shell powder (PSP) and cellulose microfiber (CMF)	Laccase enzyme treatment, mechanical (Grinding)	Not specified	Agar	CMF films showed higher thickness, tensile strength, and water solubility than PSP and agar films. CMF and PSP films had higher opacity than agar films. PSP films showed less weight loss than CMF in soil burial due to lignin. Enhanced biodegradability is suitable for food packaging.	(85)
Peanut shell cellulose	Acid hydrolysis, mechanical (Grinding)	5–20% w/w	Poly(vinyl alcohol) (PVA)/Polyvinyl Pyrrolidone (PVP)	Higher cellulose content (20%) increased acid absorption and biodegradability. FTIR confirmed good interaction in PVA/PVP/cellulose blends. Antimicrobial activity higher against Gram-negative (*E. coli*) than Gram-positive (*B. subtilis*) bacteria.	(143)

(*continued on next page*)

Table 5 (*continued*)

Nutshell-derived biomaterial	Treatment	Loading	Polymer Matrix	Results	References
Defatted cashew nut shell starch	Wet milling	Not specified	None (Starch as matrix)	Isolated starch (85.01% yield) with high amylopectin (75.35%) and crystallinity. Bonded resins detected on granules. Suitable as renewable polymeric material for packaging due to high crystallinity and low cost.	(144)
Pistachio shell hemicellulose	Chemical extraction	35.93% w/w (hemicellulose/ gelatin ratio)	Gelatin	Reduced tensile strength (16.64 MPa vs. 20.41 MPa) and water solubility (39.21% vs. 49.57%), increased elongation at break (4.34x), and enhanced water vapor permeability and soil degradation compared to gelatin films. Suitable for edible packaging.	(145)
Hazelnut husk flour	Mechanical (Grinding, assumed)	10–40% w/w	Poly(lactic acid) (PLA)	Tensile strength decreased from 33 MPa to 26.3 MPa at 10% and 6.33 MPa at 20–40% loading due to weak interfacial bonding and husk agglomeration. Thermal stability improved with higher filler content.	(146)
Modified pistachio shell (PS)	Alkaline/ultrasound treatment, mechanical (Grinding)	Not specified	Starch	Increased crystallinity index (31.41% to 46.91%), storage modulus (198 MPa to 721 MPa) due to enhanced O•C•O and O•C bonds. SEM showed etching and cavities, improving compatibility with starch. Enhanced rigidity for packaging applications.	(91)
Hazelnut shell powder (HSP)	Mechanical (Grinding, assumed)	Not specified	PLA/PBSA blend	Modest increase in melt viscosity with ESO/malic acid chain extender at low HSP loading. Crystallinity varied by HSP size (reduced for fine HSP, increased for coarse). Tensile properties unaffected by chain extenders, dependent on HSP size.	(147)
Almond shell flour (ASF)	Mechanical (Grinding, assumed)	10–30% w/w	Poly(hydroxybutyrate-co-hexanoate) (PHBH)	Increased stiffness with higher ASF content, reduced tensile and impact strength. OLA (10–20 phr) improved ductile properties but not plasticization. Higher CLTE and storage modulus with ASF. Brittle at 30% ASF, suitable for injection molding.	(148)
Almond shell extract (ASE)	Microwave-assisted ethanol extraction	Not specified	None (Extract as additive)	High TPC (5.64 mg GAE g^{-1}), flavonoids (1.42 mg CE g^{-1}), polysaccharides (1.59 mg glucose g^{-1}), and antioxidant capacity (2.82 mg AAE g^{-1}). Suitable as active additive for antioxidant packaging.	(70)
Almond shell powder (ASP)	Mechanical (Grinding, assumed)	10, 25% w/w	INZEA F2® (Polyester blend)	Increased elastic modulus at 25% ASP. MLO and Joncryl improved compatibility at 10% but showed gaps at 25% (FESEM). Reduced thermal stability, 50% disintegration at 25% ASP after 90 days under composting.	(149)
Pistachio shell extract	Microwave-assisted ethanol extraction	Not specified	None (Extract as additive)	High TPC, flavonoids, and hydrolysable tannins (e.g., gallic acid, pentagalloylglucose, kaempferol) in fractions Fr4–Fr6. High antioxidant activity (DPPH, TEAC, ORAC) for food and healthcare applications.	(150)
Macadamia nutshell powder	Mechanical (Grinding, assumed)	Not specified	Poly(lactic acid) (PLA)	Increased water absorption with higher powder content due to hydrophilic nature. Counter-rotating processing showed reduced weight loss with higher powder content; co-rotating showed consistent biodegradation. Slower biodegradation than pure PLA.	(9)
Almond shell powder (ASP)	Mechanical (Grinding, assumed)	30% w/w	Starch (Mater-Bi DI01A)	Increased flexural modulus, reduced impact strength and tensile/flexural strength. No significant differences across almond varieties (Desmayo Rojo, Largueta, Marcona, Mollar, commercial mix). Suitable for toys and packaging.	(151)
Almond shell powder (ASP)	Mechanical (Grinding, assumed)	20% w/w	Thermoplastic starch (TPS)	Progressive degradation in mechanical, thermal, and aesthetic properties after three reprocessing cycles. Feasible to reprocess at least three times without virgin material. ELO improved compatibility and ductility.	(152)

WNC-reinforced CNS starch films exhibited a desirable oxygen transfer rate and mechanical and physical properties, with thermal degradation temperatures ranging from 298 to 302 °C. Furthermore, hydrophilic compounds extracted from pomegranate peel were incorporated into 2% walnut nanocellulose, reinforced starch films, enhancing their functional performance for active packaging.

Similarly, Girgin et al. developed edible walnut shell films plasticized with glycerol or sorbitol and enriched with *Aloe vera* gel and lemon essential oil [155]. Adding lemon essential oil (LEO) and *Aloe vera* gel (AVG) significantly affected the physical properties of the films. Moisture content dropped in most samples, except for those with 20% sorbitol, where it rose sharply. Due to the hydrophobic nature of LEO, water vapor permeability increased, while glycerol-plasticized films showed lower solubility compared to those with sorbitol. The inclusion of LEO and AVG also thickened the films and caused greater color variation, particularly in glycerol-based formulations. Júnior et al. [156] developed a cardanol–formaldehyde resin biocomposites by reinforcing with both untreated and alkali-treated bamboo fibers. The fibers were modified using NaOH (5–10%) and $NaClO/H_2O$ bleaching, then incorporated into the resin via hand lay-up. Alkali treatment enhanced fiber–matrix bonding, leading to improved tensile strength, modulus, and elongation. Thermogravimetric and XRD analyses confirmed higher thermal stability, while biodegradation tests showed increased microbial breakdown due to the removal of lignin and hemicellulose. In another study, Long et al. [157] utilized *Camellia oleifera* shells (COS), a tea oil byproduct rich in hemicellulose, to produce high-performance bio-based films via citric acid treatment (15%, 100 °C, 24 h). Partial hydrolysis increased surface reactivity and hydrogen bonding among holocellulose fibrils, enhancing the films' mechanical and thermal properties. The resulting COS holocellulose (COSH) films exhibited excellent strength, transparency, and biodegradability. Mechanical blending further improved performance, with tensile strength and

Young's modulus reaching 123.5 and 5265 MPa, respectively. Complete soil degradation confirmed their sustainable and durable nature. The resulting edible films exhibited suitable physical and barrier properties, making them promise for food packaging applications. Nutshell-derived biomolecules such as starch and cellulose have strong potential as matrices for producing bio-based thermoplastic films. Cruz et al. (2024) further demonstrated that incorporating walnut shell powder into pectin-based composites increased film thickness (0.49–0.57 mm), opacity (100%), and elongation at break (32–36%), yielding eco-friendly packaging materials with improved barrier performance [158]. Qian et al. [159] reported that adding 12 wt% walnut shell powder to CMC-based bioplastics significantly improved their performance. The composites exhibited higher tensile strength (18.5 MPa), strong water resistance (remaining stable after seven days), and excellent UV-shielding efficiency (blocking over 99% of UVA and UVB). Moreover, their recyclability in alkaline environments highlights their suitability for sustainable packaging applications [79]. Polyvinyl alcohol (PVA) biocomposite films reinforced with 20% cashew nut shell powder (CNS), fabricated via solution casting, exhibited significant improvements in tensile strength (22 MPa, 130% increase), tensile modulus (264 MPa), thermal stability (43% increase at 215 °C), and antibacterial activity (93% bacterial growth reduction), making them promising for eco-friendly packaging [141]. Starch-based biocomposites with 5–30 wt% almond shell powder (ASP) of varying particle sizes [89].

5.2. Fillers

While biopolymers are widely used for food packaging purposes, their inherent permeability to gases and vapors makes them unsuitable for standalone use. To address this issue, researchers have been exploring the development of compatible fillers to create polymeric composites with enhanced properties. Nanofillers have been found to be particularly effective in improving mechanical properties and overcoming barriers to gas and water vapor [160,161]. Among the various by-products of the agro-food industry, nutshells and biowastes can be used as renewable and cost-efficient structural fillers in polymer composites.

In this regard, pecan shell-derived fillers have been utilized in a poly (lactic acid) (PLA) matrix by extrusion [162]. Increasing pecan shell-derived fillers content (5 and 7.5 wt%) altered color and increased water absorption, while also reducing tensile strength and ductility due to the rigid, lignocellulosic nature of the filler. Thermal stability was slightly enhanced, and morphological analysis confirmed generally good dispersion with some particle agglomeration at higher loadings. Defatting treatment had a limited effect on overall performance, likely due to the relatively low filler content used. The by-product obtained after ethanolic extraction of diethyl ether extract from hazelnut skin residue has been utilized to reinforce PLA and poly(propylene) (PP). This fraction increased the storage modulus of PLA and PP up to 30 and 20 % (with 30 wt % filler content), respectively. Increasing filler content (from 10 to 30 wt%) increased oxygen permeability in both polymers, particularly in polypropylene. This was attributed to poor interfacial adhesion and to the relatively large filler particle size observed by SEM, which resulted in interfacial voids and diffusion pathways. In PLA composites, however, selected hazelnut skin fillers exhibited an unexpected barrier effect at low loadings, indicating that improved dispersion and partial compatibility can enhance gas barrier properties [163]. Agustin-Salazar et al. [164] reinforced PLA biocomposites with pecan nutshell fibers and their main fractions holocellulose (HC) and acid-insoluble lignin (AIL). Both nutshell fibers and HC acted as nucleating agents, helping to regulate PLA's physical ageing and enhance its viscoelastic behavior by limiting molecular mobility. Flexural testing revealed a 25% increase in modulus with HC, while AIL improved ductility, raising stress and strain at break by 55 and 65%, respectively. Although interfacial bonding remained weak, reducing resilience, the study demonstrated the strong potential of nutshell-derived fillers for improving PLA-based biocomposites.

In PHBV-based composites reinforced with nutshell and other agricultural waste fillers, composite performance was strongly influenced by filler type, loading, and interfacial interactions. According to Mazur et al. [165], thermal stability decreased when pre-treated natural fillers were added to the 3-hydroxybutyrate-co-3-hydroxyvalerate (PHBV) polymer, resulting in a lower degradation temperature. Among the three different agricultural wastes added at 15 wt% each (nettle fibers, walnut shell flour, and pinecone flour), walnut shells exhibited the least reduction in thermal stability. This indicated that their addition to PHBV was more effective compared to the other fillers. Although the strength properties of each composite were reduced due to voids and poor adhesion resulting from the incorporation of additives, there was an observable increase in hydrolytic degradation rate. This led to a gradual reduction in tensile strength over time, which could be advantageous for disposable applications. In a different study, it was found that incorporating various amounts of walnut shells (10-40 wt%) with a particle size of less than 100 μm into PLA influenced mechanical performance through its effect on dispersion and stress transfer within the matrix. SEM micrographs showed that at higher walnut shell contents, the increased filler led to more brittle behavior and reduced impact resistance. This is consistent with larger and poorly dispersed particles acting as stress concentrators and interrupting load transfer in the PLA matrix. On the other hand, the flexural properties, especially flexural strength, improved, reaching 107.5 MPa with the 10 wt% addition of walnut shells, compared to 98 MPa for pure PLA. This indicates enhanced lateral deformation. Impact strength also increased with the addition of walnut shells and continued to improve up to 10 wt%, possibly due to the interaction between PLA and walnut shells, which can withstand high-impact loads [166].

Zaaba et al. examined PLA/thermoplastic starch composites reinforced with peanut shell powder using compression molding. Composites containing up to 40 wt% filler showed reduced tensile strength and elongation due to filler agglomeration and poor dispersion, though stiffness increased with higher filler loading. The study also found that processing sequence affected performance, with the addition of thermoplastic starch to the PLA–filler mix yielding the most balanced mechanical properties [167].

Nutshell-derived fillers have also shown potential in paper making industry. Pistachio shell particles, valorized as renewable fillers in paperboard packaging, with particle sizes ground to a median diameter of about 6.9 μm and most particles kept below the fiber network pore size (less than ~24.4 μm). These fillers disperse well in aqueous systems, show better retention than ground calcium carbonate, and, when having a narrow particle size distribution, exhibit comparable tensile and burst strength, while offering cost reduction and improved sustainability by replacing mineral fillers with agricultural waste [168–170].

The surface chemistry of fillers strongly affects polymer-filler adhesion. Polar –OH groups on lignocellulosic fillers interact poorly with non-polar polymers, resulting in weak interfaces, poor dispersion, and agglomeration. Surface modifications such as chemical treatments, silane coupling agents, or reactive compatibilizers like maleic anhydride–grafted polymers have improved interfacial bonding by forming covalent or strong interactions with the filler. This enhanced adhesion increases mechanical strength, reduces defects, and improves the overall performance of polymer composites [171]. In this regard, Rojas-Lema et al. [171] developed poly(butylene succinate) grafted with maleic anhydride to improve the bonding between the polymer and pistachio shell flour as filler. The composites containing 5–30 wt% fillers were fabricated through injection molding. Increasing the loadings enhanced stiffness but reduced tensile strength and elongation due to poor interfacial bonding and filler agglomeration. SEM images confirmed void formation, which was minimized with compatibilizer addition. Thermal analysis showed unchanged melting temperature but higher crystallinity at greater filler contents. The compatibilized composites exhibited delayed degradation, lower water absorption, and improved structural

uniformity [171]. Peanut shell powder, incorporated at 5–15% by weight in PA6 biocomposites, enhanced structural and thermal stability without affecting matrix density, as shown by Oulidi et al. [90]. Compatibility confirmed by FTIR and MEB analyses supports its use as a sustainable filler in biocomposites.

5.3. Functional additives

5.3.1. Antioxidants

Nutshells-derived antioxidants can be added to biopolymer-based film for the dual purpose of stabilization and functionalization. This technique is especially useful in active food packaging, which is a significant sector in the food industry that helps prevent or prolong the oxidation process of food, thus preserving its quality for a longer time [172]. In line with this, Moccia et al. [173] investigated the potential of pecan nutshell extract (PNSE) as a natural antioxidant additive for active food packaging. The hydroalcoholic extract, rich in condensed tannins, exhibited strong antioxidant capacity ($EC_{50} = 0.004$ mg/mL in the DPPH assay). When incorporated into PLA films (up to 10 wt%) via solvent casting, PNSE maintained the films' mechanical integrity. It also provided significant antioxidant activity achieving over 60% DPPH reduction at 3 wt% loading. Films containing 10 wt% PNSE also delayed browning in apple smoothies by approximately 30%.

Similarly, Agustin-Salazar et al. [136] evaluated PNSE as a natural antioxidant for improving the stability of PLA and polyethylene (PE) films. The hydroalcoholic extract, rich in 26 phenolic compounds, mainly proanthocyanidins, exhibited strong radical-scavenging activity. In addition, PNSE enhanced both thermal and photo-oxidative resistance, showing high compatibility with PLA and superior UV protection for PE by neutralizing peroxy radicals and limiting photolytic degradation. Economic analysis further demonstrated that large-scale production of PNSE is both technically viable and cost-effective, supporting its potential as a sustainable polymer stabilizer. Nutshels has also shown efficient antioxidant activity in protein and polysaccharide-based films. In this regards, Arciello et al. [72] demonstrated that PNSE effectively enhanced the antioxidant characteristics of the whey protein-based films. It, showed a Trolox equivalent value that was 2.4 times greater than that of the control film after 4 h (0.6 ± 0.1 μg/mg versus 0.27 ± 0.02 μg/mg). Additionally, Esposito et al. [54] developed bioactive films made from pullulan, infused with polyphenol-rich extracts derived from chestnut spiny burs (CSB, *Castanea sativa* Miller) and roasted hazelnut skins (RHS, *Corylus avellana* L.). Each extract was isolated through a hydroalcoholic process and then mixed with the filmogenic polysaccharide pullulan at concentrations of 1, 5, and 10% (wt). The pullulan-based films demonstrated significant antioxidant properties, with an approximate DPPH scavenging activity of 94% achieved with only 1% of the hydroalcoholic extract (equivalent to192 μg of extract per cm^2 of film). Additionally, peanut shell flavonoids, rich in antioxidants like luteolin and eriodictyol, significantly improved the mechanical strength, barrier properties, and antioxidant activity of sodium alginate–carrageenan films. They also reduced lipid oxidation and microbial growth in chilled pork. Films containing 0.45 % peanut shell flavonoids delayed increases in total viable count, pH, and volatile nitrogen. This extended the shelf life of chilled pork from 6 to 12 days [174].

The findings suggest that the utilization of antioxidants obtained from nutshells in polymer films can serve as an effective and cost-efficient method for stabilizing the films in various applications, including active food packaging.

5.3.2. Antibacterials

Food spoilage and deterioration are typically caused by microorganisms such as Campylobacter, Salmonella, *Yersinia enterocolitica*, *Escherichia coli*, and *Listeria monocytogenes*, which grow on food surfaces. To combat this, active packaging materials have been developed to extend the shelf life of food products. Antimicrobial films are one of the most promising active packaging materials, which can be created by incorporating synthetic or natural antimicrobial agents into films or directly coating them on food. Natural antimicrobial agents are particularly appealing due to their relative safety and ease of availability and have been developed for this purpose [175]. The antimicrobial activities of several composite polymers/films containing nutshell-derived biomaterials against foodborne pathogens have been reported. Pullulan-based films infused with polyphenolic-rich extracts from chestnut spiny burs and roasted hazelnut skins showed antibacterial activity against *Staphylococcus aureus*. The antibacterial effectiveness increased with higher extract concentrations. The film containing 10 % roasted hazelnut skin extract achieved the strongest effect, reducing bacterial counts by 4 log CFU/mL [54]. In a similar manner, Leon-Bejarano et al., showcased the antibacterial capabilities of octenyl succinic anhydride (OSA)-PNSEand OSS-hazelnut skin extract (HSE) films against *S. epidermidis*, *S. aureus*, *P. aeruginosa*, and *K. pneumoniae* (in descending order of effectiveness), particularly in a liquid environment [176]. Furthermore, Mahanta and Joardar found that PVA films containing CNS exhibited a noteworthy 93% decrease in bacterial growth, indicating the strong antibacterial properties of CNS [177].

In a study conducted by Arciello et al. [72], the antimicrobial effect of PNSE-loaded whey protein on the growth of foodborne bacteria, *Enterococcus faecalis* and *Salmonella enterica* subsp. *enterica* ser. *Typhimurium*, was investigated. The results showed that after 24 h at 37 °C, bacterial growth was only inhibited in the presence of PNSE (Fig. 9a). The film containing PNSE exhibited a strong inhibition of bacterial growth, with almost no colonies detected underneath it, while numerous bacterial colonies were observed under the control film. This suggests that the antimicrobial substances in the film act directly on bacterial cells in contact with the film, inhibiting their growth. This particular outcome has been linked to PNSE capacity to oxidize microbial cell membranes, bind to crucial metal ions, or obstruct extracellular enzymes. To further explore the effectiveness of the PNSE-functionalized films against bacteria, confocal laser scanning microscopy analyses were conducted. The addition of PNSE significantly improved the antibacterial properties of the films, as shown in Fig. 9b. Notably, the presence of PNSE led to a decrease in biofilm volume for both bacterial strains tested. Since both strains had the ability to form biofilms, the results suggest that whey protein/PNSE film forming solution can hinder biofilm formation by preventing cell adhesion. This effect may be linked to specific active compounds present in PNSE, such as tannins.

The findings provide a foundation for the potential implementation of biopolymer-based films that are functionalized with nutshell biomaterials. These films can serve as a novel and eco-friendly alternative for enhancing the shelf-life of food products.

5.3.3. UV-barriers

One of the most intriguing focuses for the packaging industry is the development of UV-protection films that rely on renewable materials extracted from natural sources. Lignin and phenolics are the main UV-barrier compounds found in nutshells (Table 2). In line with this, Halloub et al. [178] developed intelligent food packaging films using modified lignin and CNCs extracted from the Argania nutshell. These films were designed to protect food products from UV light and maintain their freshness for a longer period. The researchers found that adding 2 % of CNCs to the films increased their tensile strength and flexibility while also improving their optical properties. However, higher concentrations of CNCs resulted in aggregation and reduced mechanical properties. The lignin film reinforced with 2% of CNCs showed the best results in terms of mechanical properties and UV protection. In fact, it significantly enhanced the shelf-life of red cabbage, demonstrating the UV-light protection efficacy of this film in food packaging applications. In another study, Esposito et al. fabricated bioactive films using pullulan (PL), combined with extracts from chestnut spiny burs (CSB) and roasted hazelnut skins (RHS). The films containing 10% hydroalcoholic extracts showed excellent UV-absorbing properties across the UVC (100–280 nm), UVB (280–320 nm), and UVA (320–400 nm) regions. The

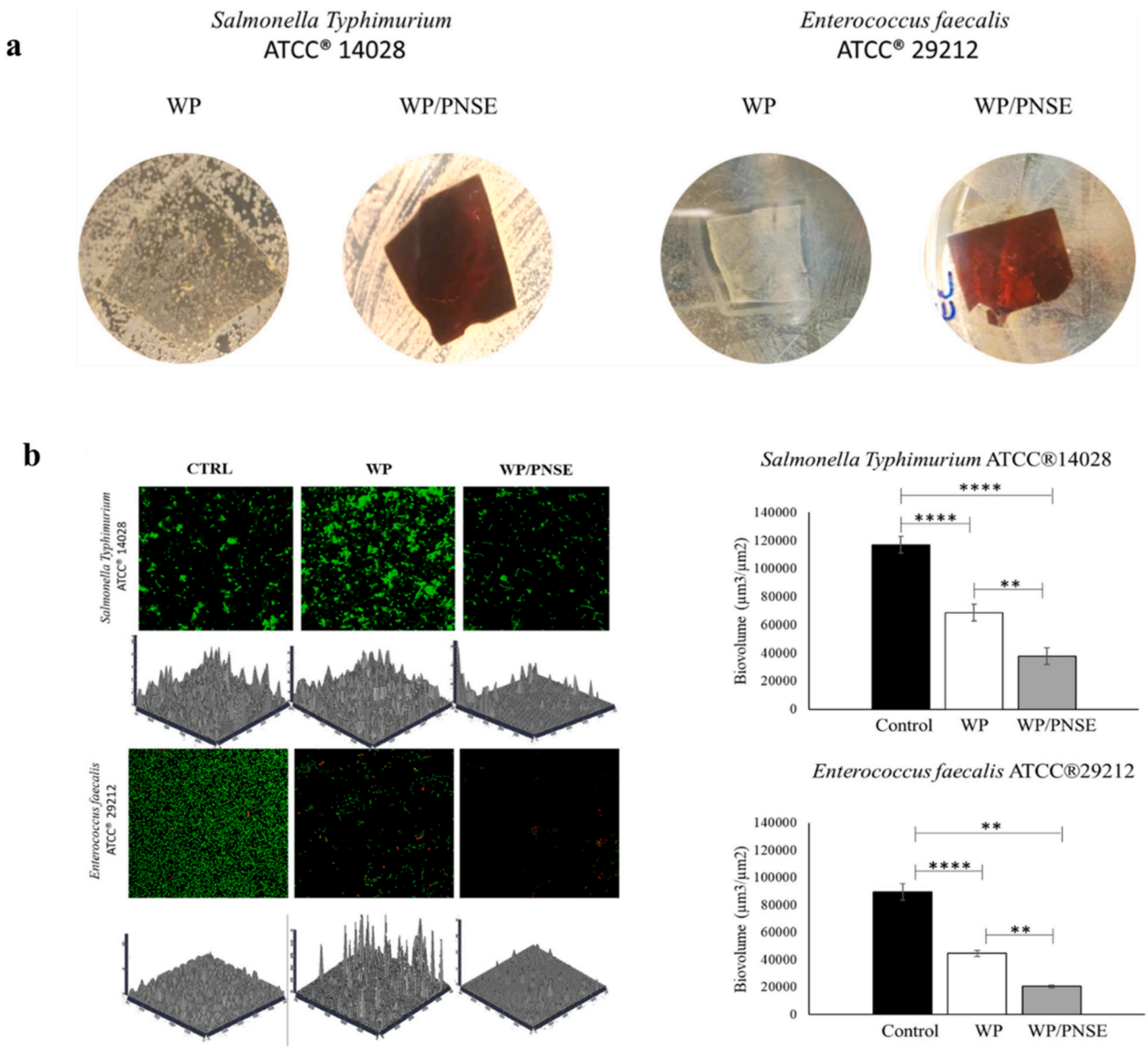

Fig. 9. (a) Evaluation of the antimicrobial activity of whey protein (WP) films functionalized with pecan nutshells extract (PNSE). The antimicrobial activity of the films was tested by assessing the growth of *Salmonella enterica* subsp. *enterica* ser. Typhimurium ATCC® 14028 and *Enterococcus faecalis* ATCC® 29212 in direct contact with agar surface. (b) Analysis of bacterial cell adhesion on functionalized coverslips using confocal laser scanning microscopy. The effects of film forming solution on the attachment of *Salmonella enterica* subsp. *enterica* ser. Typhimurium (ATCC® 14028) and *Enterococcus faecalis* (ATCC® 29212) bacterial cells were evaluated. Images were captured under consistent conditions in at least three independent experiments. Scale bar 10 µm. Statistical significance was observed for control versus treated samples and WP vs WP/PNSE sample, indicated by $^{**}p < 0.01$, $^{***}p < 0.001$, or $^{****}p < 0.0001$ [72].

PL/CSB_10% film had transmittance values below 0.4%, while the PL/RHS_10% film showed transmittance values of 2.5%. These properties are essential for light-sensitive food products that are prone to photo-oxidation. Leon-Bejarano et al. [176] conducted a study where they produced films using octenyl succinate starch (OSS) containing either PNSE or hazelnut skin extract. The control film did not absorb light in the 200–800 nm wavelength range, indicating that light passed through it completely. On the other hand, the OSS films with PNSE and hazelnut skin extract exhibited absorption peaks within the 250–300 nm range.The strength of these peaks increased as the concentration of the extracts increased. The enhancement of the UV-barrier properties of the films was attributed to phenolic compounds and pigments. In another study, Battegazzore et al. [163] evaluated the degradation of PP in the presence of hazelnut skin extract (N) and cocoa by-products extract (C) through photo-oxidation. The type of UV-stabilizer was found to strongly influence the oxidation induction time of PP when N or C were added to the polymer matrix. N was observed to significantly increase PP oxidation induction time (+30%), while its photo-oxidation rate remained nearly constant. However, the presence of C did not have a significant effect on PP photo-stability. This could be attributed to the limited thermal stability of C, as evidenced by thermogravimetry, and the differences in phenolic compounds and flavonoids between N and C, as indicated by UV spectroscopy. Therefore, biomolecules derived from nutshells have the potential to serve as effective UV absorbers. They can protect light-sensitive food products from photo-oxidation, thereby prolonging their shelf life. These films can be incorporated into antioxidant-active packaging systems as secondary antioxidants to enhance their effectiveness.

5.4. Plasticizer

According to IUPAC, a plasticizer is a substance added to polymers to improve flexibility and processability. With the growing shift toward bio-based plastics, demand for sustainable plasticizers has increased.

Cardanol, a low-cost byproduct of the cashew nut industry, has emerged as a promising candidate. Studies have shown that covalently linking cardanol to PVC via click chemistry yields an internal plasticizer with a lower glass transition temperature, good thermal stability, and minimal migration. Further work revealed that unmodified cardanol is ineffective as a plasticizer; however, its acetylated and epoxidized derivatives display good solubility and performance comparable to traditional DEHP-based systems. Despite this, ageing tests indicated faster migration due to residual unreacted cardanol, slightly compromising long-term stability [179]. In this regard, esters synthesized from cashew nutshell liquid exhibit excellent mechanical properties (tensile strength 14 MPa, elongation 449 %) and thermal stability, with lower gelation temperatures and good chemical resistance. Using the natural mixture avoids the time-consuming and energy-intensive separation of pure cardanol or cardol. It also provides additional benefits, such as UV-blocking up to 375 nm, making cashew nutshell liquid-based plasticizers suitable for flexible PVC products and packaging applications [180].

Additionally, hazelnut skin extract has been explored as a natural plasticizer for PLA film, showing partial plasticization based on thermal and mechanical analyses [163]. Similarly, Leon-Bejarano et al. [176] reported that incorporating PNSE or hazelnut skin extract (HSE) showed plasticizing effect in octenyl succinate starch (OSS) films by reducing tensile strength and stiffness. The decrease in puncture resistance and Young's modulus indicated that phenolic compounds from the extracts disrupted the polymer matrix, enhancing flexibility through a plasticizing effect.

5.5. Biochar

Biochar is a carbon-rich material produced through the pyrolysis of organic biomass under limited oxygen conditions [181]. It has gained significant attention for its diverse applications, including soil amendment, water treatment, and reinforcement in polymer composites [182,183]. Nutshells, an abundant agricultural waste, are particularly promising feedstocks for biochar production due to their high lignocellulosic content. Biochar can also be produced from nutshell residues left after biomaterial extraction in cascade extraction and processing systems. Biochar derived from nutshells involves the thermal decomposition of nutshell biomass in an oxygen-limited environment, resulting in a stable, carbon-rich product [184]. The process enhances the physical and chemical properties of biomass, making it suitable for various applications. Recent advancements in pyrolysis techniques, including traditional slow pyrolysis, fast pyrolysis, and gasification, have improved the yield and quality of nutshell-derived biochar. Additionally, biochar can be activated using physical or chemical methods to enhance its adsorption capacity and surface area [185]. Derived from nutshells, biochar exhibits unique properties that make it an ideal material for sustainable practices. The pyrolysis process, typically conducted at temperatures ranging from 300 to 700 °C, converts these shells into biochar, retaining a high carbon content and porous structure [186–188]. Nutshell-derived biochar possesses several beneficial properties. Its high surface area and porosity enhance its ability to adsorb contaminants. Additionally, the presence of functional groups like hydroxyl, carboxyl, and phenolic groups increases its reactivity. Furthermore, it contains essential minerals such as potassium, calcium, and magnesium, and its high stability makes it effective for long-term carbon sequestration [189]. These properties make nutshell-derived biochar highly versatile for various applications [23].

Sundarakannan et al. [190] investigated the mechanical properties of biochar derived from cashew nutshell waste (CNSL) and its reinforcement in polymer matrix composites. The biochar was produced through pyrolysis at 500 °C for 1 h, followed by ball-milling to achieve a uniform particle size. Different weight percentages (5, 10, and 15 %) of biochar were mixed with unsaturated polyester resin to fabricate the composites. Mechanical testing revealed that the composite with 10 % biochar exhibited the highest tensile strength (41 % improvement), impact strength (37 % improvement), and hardness (21 % improvement). The improvements were attributed to the homogeneous dispersion of biochar particles, which act as barriers to crack propagation and enhance load transfer within the polymer matrix. Similarly, Ramraji et al. studied hybrid polymer composites reinforced with alkaline-treated flax fibers and almond shell biochar. The biochar produced via slow pyrolysis (at 600 °C for 2 h) showed significant improvements in tensile strength (36.5 %), bending strength (39.4 %), and impact strength (99.75 %) of the composite. This observation correlated with improved interfacial bonding and efficient load transfer within the matrix. Beyond polymer composites, biochar's role in environmental applications is noteworthy. Leichtweis et al. [191] focused on a biochar-ZnO composite derived from pecan nutshells for the sequential removal of Reactive Red 97 (RR97) dye by adsorption and photocatalysis. Produced through mechanical mixing and pyrolysis at 650 °C, the N20Z composite (20 % ZnO) demonstrated the highest efficiency, degrading 100 % of the dye in 67 min. This performance was attributed to reduced electron-hole recombination, decreased band gap energy, and increased surface area. This study highlights biochar's versatility in water treatment applications. The biochar, characterized by its high surface area, porous structure, and abundant functional groups, effectively adsorbs heavy metals (e.g., Pb, Cd, Cu) from contaminated water, resulting in purified water output. Dao et al. explored another aspect of biochar's environmental utility by investigating activated biochar from macadamia nutshells for removing Cu^{2+} and Zn^{2+} from aqueous solutions. Produced through carbonization at 350 °C and chemically activated with K_2CO_3, H_3PO_4, and H_2O_2, the biochar showed significant removal efficiencies. Among them, H_3PO_4-activated biochar achieved 95.92 % for Cu^{2+} and 67.41 % for Zn^{2+}. The enhanced adsorption performance was attributed to the increased surface area and functional groups on the biochar surface, providing numerous active sites for metal ion adsorption [192]. Expanding on heavy metal remediation, Qiu et al. [128] examined walnut shell-derived biochar for cadmium (Cd) remediation in contaminated soils. Produced through slow pyrolysis at 500 °C under nitrogen, the biochar effectively reduced Cd mobility. The enhanced stability is attributed to mechanisms such as physical isolation, formation of precipitates, and complexes on the biochar surface. These findings suggest that walnut shell-derived biochar can effectively be used as a cadmium sorbent for soil remediation, improving soil quality and reducing cadmium toxicity. Similarly, Bakly et al. [193] investigated macadamia nutshell biochar (MBC) for nitrate removal from aqueous solutions. Produced through slow pyrolysis at 900 °C and 1000 °C, MBC prepared at 1000 °C exhibited better nitrate removal efficiency due to higher crystallinity and rough texture. It achieved 0.11 mg/g nitrate removal at a concentration of 15 mg/L and a flow rate of 2 mL/min. This illustrates biochar's potential in water purification. In agricultural applications, Bhaduri et al. [194] studied peanut shell biochar to restore carbon and microbial activity in salt-induced soils. Produced through pyrolysis at 300 °C for 2 h, the highest application rate (10 %) increased soil organic carbon by 47-106 % and total organic carbon by up to 32 %. The mechanism involves biochar's porous structure and functional groups, which enhanced nutrient retention, microbial habitat, and enzyme stabilization. This collectively improved soil fertility and microbial activity under saline conditions.

Further illustrating biochar's agricultural benefits, Yao et al. [195] evaluated peanut shell biochar in saline-sodic paddy fields within 24 months. Produced at 350-550 °C for 4 h and applied at various rates, the biochar improved soil nutrients, enzyme activities, and grain yield (7.06 %). The mechanisms include improved nutrient retention and availability, enhanced microbial activity, and better soil structure, reducing salinity stress and promoting plant growth. The study by Xu et al. [196] examined the effects of peanut shell biochar on soil properties and peanut kernel quality in a red Ferrosol. The biochar was produced via pyrolysis at 550 °C, yielded a very alkaline product with high total carbon and nitrogen content. The experiment applied peanut shell

biochar at a rate of 9.2 t/ha under field conditions alongside varying irrigation and fertilization regimes. The application of biochar improved soil total carbon, total nitrogen, and nutrient availability, including potassium and zinc, which were higher in biochar treatments. These improvements led to an increased proportion of high-value peanut kernels, particularly the 'jumbo' grade. The biochar's porous structure and high nutrient content enhanced soil fertility and nutrient retention, leading to better crop quality. The mechanism involves biochar's ability to improve soil organic carbon, enhance nutrient availability, and provide a stable form of carbon that benefits long-term soil health. Another study investigated the use of pistachio shell-based biochar (PSB) as a soil amendment to enhance the growth of bell pepper (*Capsicum annuum L.*). The biochar is produced from pistachio shells through slow pyrolysis at temperatures of 450 °C, 550 °C, and 650 °C for 2 h in a nitrogen atmosphere. The application of PSB in soil was evaluated at different ratios (0 %, 1 %, and 2 % w/w) under greenhouse conditions. The results demonstrated that the addition of PSB improved soil properties, such as increasing cation exchange capacity (CEC) by up to 8 %, organic matter (OM) and organic carbon (OC) by 100-200 %, and total nitrogen (TN) by 35 %. Moreover, the addition of 1 % PSB significantly enhanced plant height, chlorophyll content, and flower and fruit yield, while 2 % PSB resulted in superior fruit weight and length. The enhanced performance is attributed to PSB's ability to improve soil fertility through nutrient retention, increased water-holding capacity, and modification of soil structure. Together, these effects promote better plant growth and productivity [197]. In dye removal applications, Zazycki et al. [198] produced biochar from pecan nutshells via pyrolysis at 800 °C, resulting in a micro/mesoporous structure with a surface area of 93 m^2/g. The biochar achieved 85 % removal efficiency for Reactive Red 141 dye under acidic conditions. The enhanced adsorption performance was attributed to the biochar's high surface area, porous structure, and the formation of strong chemical bonds between the dye and the biochar surface. Fristak et al. [199] investigated magnesium composite/walnut shell-derived biochar for arsenic and phosphorus sorption in sorption applications. Produced by slow pyrolysis at 500 °C and modified with 8 % $MgCl_2$, the biochar significantly enhanced sorption capacities. It showed a 10-fold increase for arsenic and a 20-fold increase for phosphorus, attributed to increased anion exchange capacity and new sorption sites. The study by Zhou et al. [200] investigates the preparation of magnetic biochar from macadamia nutshells pretreated with $FeCl_3$-assisted mechanochemical activation for the adsorption of heavy metals (Cu^{2+} and Pb^{2+}). The macadamia nutshells were pretreated with $FeCl_3$ and mechanically activated before undergoing solvent-free pyrolysis at 400 °C. The resulting biochar exhibited a serrated lamellar structure with abundant functional groups, leading to outstanding adsorption performance. The magnetic biochar (MCA-BC) achieved maximum adsorption capacities of 339.78 mg/g for Cu^{2+} and 200.44 mg/g for Pb^{2+} based on Langmuir isotherm models. The biochar could be recycled five times while maintaining a 70 % removal rate. The adsorption mechanisms included pore filling, electrostatic adsorption, complexation, and coordination reactions. The unique properties of MCA-BC, such as its large specific surface area and magnetic characteristics, make it a promising material for treating heavy metal-contaminated wastewater. For energy storage applications, Altinci and Demir reported a green approach for synthesizing biocarbon from pistachio shells using $Na_2S_2O_3$ for supercapacitor electrodes. Produced via hydrothermal carbonization and chemical activation at 800 °C, the biocarbon exhibited a high specific surface area of 775 m^2/g and significant nitrogen/sulfur doping. The electrodes showed a specific capacitance of 166 F/g with 96 % retention over 5000 cycles, demonstrating efficient and sustainable energy storage potential.

The nutshell-derived biochar has demonstrated considerable potential across various applications, including polymer reinforcement, water treatment, soil remediation, and energy storage. The diverse methodologies for biochar production and activation enable the tailoring of its properties to specific applications, making it a versatile and sustainable material.

6. Environmental and economic impacts

The valorization of nutshell waste into biomaterials addresses significant environmental challenges while offering economic opportunities. This section examines the environmental issues associated with nutshell disposal, the sustainability benefits of bioplastics and biochar, the economic feasibility of advanced extraction methods, and the role of biorefineries in promoting a circular economy.

6.1. Environmental challenges

Nutshell waste, if not managed properly, poses significant environmental challenges. Landfilling nutshells contributes to landfill overcrowding, with over 600,000 metric tons of almond shells alone generated annually in the United States [201]. Decomposition in landfills produces methane, a potent greenhouse gas, exacerbating climate change. Additionally, phenolic compounds in nutshells, such as those in chestnut and walnut shells (9.0-274.09 mg GAE/g and 2.0-249.7 mg GAE/g, respectively), can leach into soil and water, causing contamination and toxicity to ecosystems [202]. These risks highlight the need for sustainable waste management strategies to mitigate environmental harm [201].

6.2. Sustainability benefits

Converting nutshells into bioplastics and biofuels significantly reduces the carbon footprint compared to conventional plastics and fossil fuels. For instance, nutshell-derived PLA films and bioethanol from almond shell hydrolysates (54.5% xylan yield) offer biodegradable alternatives with lower greenhouse gas emissions [201]. Biochar from cashew and walnut shells sequester carbon, with cashew nutshell biochar enhancing soil carbon storage by up to 30% while improving nutrient retention. These applications reduce reliance on non-renewable resources and support sustainable waste valorization, aligning with global decarbonization goals [190]. Comparing the environmental burden of processing techniques, the environmental impact assessment of producing natural tannin-based coagulants from chestnut shells showed that microwave-assisted extraction was more environmentally friendly than solid-liquid extraction for producing 100 g of condensed tannins. The freeze-drying step was the main hotspot, responsible for the largest environmental burden due to high electricity consumption but using renewable energy could reduce this impact by approximately 90 %. Overall, tannin-based coagulants effectively treated 1 cubic meter of river water to residual turbidity below 1 NTU, while providing sustainable advantages such as non-toxicity, no residual metal contamination, and competitive cost-performance compared to conventional aluminum- and iron-based coagulants [203].

6.3. Economic feasibility

Techno-economic assessment (TEA) on cellulose production from walnut shell showed that pure nanocellulose extracted from walnut shell using nitric acid treatment was cost-effective, yielding a net profit of 9.02 $/kg due to its high purity and crystallinity, despite higher operating costs. In contrast, cellulose extracted with the alkaline-peroxide method was not pure and resulted in a negative profit. The work demonstrates that valorizing walnut shell into high-purity cellulose can be economically viable, while simultaneously producing a magnetic cellulose-derived biochar with excellent iodine adsorption for wastewater treatment [204]. Similarly, TEA on the biorefinery of pistachio shell showed that producing nanocellulose, with a yield of 32–41 % and a crystallinity index of 86 %, and microcrystalline nitrocellulose, with a crystallinity index of 79 % and a nitrogen content of 10.81 %, was economically feasible. The process involved a rapid, recyclable nitric

acid treatment to isolate pure nanocellulose, followed by a facile synthesis to convert it into nitrocellulose. In contrast, biochar production, with a yield of 24–28 % and a higher heating value of 24–28 MJ per kilogram via pyrolysis, was not profitable due to low competitiveness with coal [205].

Advanced extraction methods, such as deep eutectic solvents (DES) and microwave-assisted extraction (MAE), enhance the economic feasibility of nutshell valorization by increasing product yields, reducing energy consumption, and lowering production costs. DES, like choline chloride:oxalic acid for chestnut shells, achieves high phenolic yields (4.64 mg/g ellagic acid) at lower costs than traditional solvents [159]. In the study on microwave-assisted pretreatment of lignocellulose, combining microwave heating with deep eutectic solvents increased total product yield by 8.53 % compared to conventional heating, while reducing energy demand by 5.82 % and decreasing environmental impacts such as global warming and fossil resource scarcity by 4-6 %. Economically, the total annualized cost per ton of product decreased from 3400 $ to 3194 $ when using microwave-assisted pretreatment. This demonstrates that integrating microwave-assisted extraction with solvent-based pretreatment not only intensifies the process for higher efficiency but also improves both economic and environmental performance, making biomass valorization, such as nutshell processing, more sustainable and cost-effective [206]. Additionally, in the study on cellulose nanocrystal production from oil palm empty fruit bunches, DES recovery was shown to significantly reduce production costs per kilogram (from $4.75 to $3.56) and improve economic indicators, while also lowering environmental impacts, including global warming potential [207].

MAE reduces extraction time and energy use, as seen with peanut shell phenolics (3.79 g GAE/100 g). To support this, in the seaweed-protein study, microwave-assisted extraction achieved the highest recovery (50 %) and lowest carbon footprint (10.8 kg CO_2 per kg protein) compared to ultrasound-assisted extraction, enzyme-assisted extraction, pH-shifting, and a hybrid pH-shifting + ultrasound method [208]. Similarly, microwave-assisted pyrolysis of sugarcane bagasse for biochar production showed positive investment feasibility, with a payback time of 7.33 years, a maximum return on investment of 16.83 %, and a minimum selling price of 0.89 USD/kg [209]. The global bioplastics market, projected to reach $44 billion by 2027, underscores the potential for high-value products like nanocellulose and biochar from nutshells, creating revenue streams for the nut industry. These cost-effective methods make nutshell-derived biomaterials commercially viable [210].

6.4. Circular economy

Integrated biorefinery approaches transform nutshell waste into a range of high-value products, minimizing waste and maximizing resource efficiency. For example, co-extraction of cellulose, lignin, and phenolics from pistachio shells produces bioplastics with enhanced mechanical and barrier properties, while biochar from walnut shells supports water treatment and soil amendment [211]. This holistic utilization reduces landfill dependency, recycles agricultural by-products, and generates economic value, aligning with circular economic principles. By integrating extraction and application processes, biorefineries ensure sustainable resource use and environmental benefits [201].

7. Challenges and future directions

The transformation of nutshell waste into high-value biomaterials offers significant potential for sustainable material science, but several challenges must be addressed to enable widespread adoption. This section explores technical hurdles, regulatory and standardization needs, emerging innovations, and future prospects for nutshell-derived biomaterials, emphasizing their integration into a circular economy.

7.1. Technical challenges

The variability in nutshell composition, such as differences in cellulose (30-50%), lignin (20-30%), and phenolic content (e.g., 9.0-274.09 mg GAE/g for chestnut shells) across almond, walnut, and pistachio shells, complicates standardized processing. This heterogeneity requires tailored extraction methods, increasing complexity and costs [201]. High processing costs, particularly for energy-intensive methods like acid hydrolysis (e.g., 85.46% cellulose yield from pistachio shells) and ball milling, posing economic barriers. Scalability issues further hinder industrial adoption, as processes like enzymatic extraction (e.g., 54.5% xylan yield from almond shells) are often limited to lab-scale due to high enzyme costs and long processing times. Addressing these challenges requires optimized, cost-effective, and scalable technologies to ensure consistent biomaterial quality [212]. Key research gaps remain in translating laboratory protocols into robust, industrial workflows. Future studies should prioritize scalable process intensification and techno-economic optimization, alongside reliable quality control strategies that can accommodate feedstock variability while maintaining consistent biomaterial performance.

7.2. Regulatory and standardization needs

For nutshell-derived bioplastics to be used in food-grade packaging, stringent regulatory requirements must be met, including safety assessments for migration of phenolic compounds (e.g., chestnut shell tannins, 2.5-5.2%) and other bioactives into food. Compliance with standards like those set by the FDA or EU regulations (e.g., Regulation (EC) No 1935/2004) is critical to ensure consumer safety and market acceptance [213]. The lack of standardized protocols for characterizing nutshell-derived biomaterials, such as nanocellulose or biochar, complicates quality control and certification. Developing standardized testing methods and regulatory frameworks for bio-based materials will facilitate their commercialization and integration into mainstream markets. A major research gap is the standardization of nutshell-derived biomaterials for commercial use. There is a clear need for harmonized characterization protocols, reference materials, and agreed performance benchmarks for products such as nanocellulose, lignin fractions, and biochar, including specifications for purity, particle size distribution, functional group content, batch-to-batch variability, and food-contact safety. Establishing these standards will strengthen certification pathways and enable reliable scale-up and market adoption.

7.3. Innovations

Emerging technologies are enhancing the efficiency and sustainability of nutshell valorization. Deep eutectic solvents (DES), such as choline chloride:oxalic acid for chestnut shell phenolics (4.64 mg/g ellagic acid), offer eco-friendly alternatives to traditional solvents, reducing environmental impact. Supercritical CO_2 extraction, achieving 82% anacardic acid from cashew nutshell liquid, provides high-purity yields for plasticizers and antimicrobials [87]. Novel applications, such as smart packaging sensors using carbon dots from pistachio shells for real-time spoilage detection, are gaining traction [214]. Additionally, microwave-assisted extraction (e.g., 3.79 g GAE/100 g phenolics from peanut shells) and ultrasonic-assisted methods (e.g., 22.44 mg/g phenolics from coconut shells) improve yield and reduce energy use, paving the way for scalable, sustainable processing.

7.4. Future prospects

The integration of nutshell-derived biomaterials into circular economy models offers significant potential. Biorefinery approaches that co-extract cellulose, lignin, and phenolics (e.g., from pistachio shells for bioplastics and biochar) maximize resource efficiency and minimize waste [215]. Policy support, such as subsidies for green technologies or

incentives for agricultural waste valorization, can accelerate commercialization [201]. Expanding applications, such as using walnut shell biochar for heavy metal adsorption in water treatment or chestnut shell tannins in antimicrobial films, can drive market growth [15]. Collaborative efforts between researchers, industry, and policymakers are essential to scale up production, standardize processes, and promote nutshell-derived biomaterials as sustainable alternatives to conventional plastics. **Beyond laboratory validation, long-term stability and biodegradation under real-world conditions represent critical gaps. Future work should evaluate nutshell-derived bioplastics and active additives under realistic storage, transport, and end-of-life scenarios, including temperature and humidity cycling, UV exposure, mechanical stress, and contact with real foods, and should report standardized biodegradation outcomes in soil, marine, home-composting, and industrial composting environments. In parallel, the practical integration into existing nut-processing infrastructure remains underexplored. Research should focus on low-disruption valorization pathways that can be embedded into current shell-handling streams, including on-site preprocessing, modular extraction units, solvent and water recycling loops, and logistics models that leverage existing collection and drying operations. Demonstrating compatible process integration will be essential for economic feasibility and rapid industrial uptake.**

8. Conclusion

Nutshell-derived bioplastics offer a sustainable solution by utilizing agricultural waste (e.g., almond, walnut, chestnut shells) rich in cellulose (30-50%), lignin (20-30%), and phenolics (e.g., 9.0-274.09 mg GAE/g in chestnut shells) to produce biodegradable films, nanocellulose, biochar, and antimicrobial additives for packaging, water treatment, and soil amendment. Green extraction methods like deep eutectic solvents and microwave-assisted techniques enhance efficiency and sustainability. Environmentally, nutshell valorization reduces landfill waste (e.g., 600,000 metric tons of almond shells annually), methane emissions, and phenolic contamination, while lowering the carbon footprint compared to conventional plastics. Economically, cost-effective methods and the growing bioplastics market ($44 billion by 2027) create revenue opportunities. Integrated biorefineries align with circular economy principles, but challenges like composition variability and processing costs require ongoing research, regulatory support, and innovations (e.g., smart packaging sensors) to drive industry adoption and scalability for a greener future.

CRediT authorship contribution statement

Shima Jafarzadeh: Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Project administration, Investigation. **Zeinab Qazanfarzadeh:** Writing – review & editing, Writing – original draft, Visualization, Validation, Investigation. **Nazila Oladzadabbasabadi:** Writing – original draft, Visualization, Validation, Investigation. **Minoo Naebe:** Writing – review & editing, Visualization, Validation, Supervision. **Colin J. Barrow:** Writing – review & editing, Visualization, Validation, Supervision.

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