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Maximizing crustaceans (shrimp, crab, and lobster) by-products value for optimum valorization practices: A comparative review of their active ingredients, extraction, bioprocesses and applications

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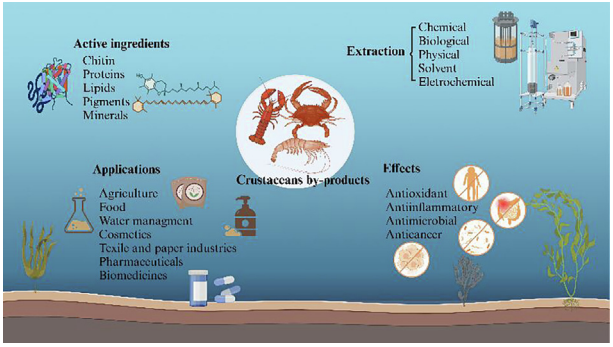
HIGHLIGHTS

- This review addresses the important bioactive ingredients recovered from different major crustacean's by-products.
- A comprehensive application of crustacean's by-products in various fields is presented.
- Nowadays, many newly developed techniques have been applied in crustacean's by-products recovery.
- A combination of innovative extraction techniques with industrially applicable technologies can efficiently recover these valuable components.

GRAPHICAL ABSTRACT



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ABSTRACT

Background: The processing of the three major crustaceans (shrimp, lobster, and crab) is associated with inevitable by-products, high waste disposal costs, environmental and human health issues, loss of multiple biomaterials (chitin, protein hydrolysates, lipids, astaxanthin and minerals). Nowadays, these bioresources are underutilized owing to the lack of effective and standardized technologies to convert these materials into valued industrial forms.

Aim of review: This review aims to provide a holistic overview of the various bioactive ingredients and applications within major crustaceans by-products. This review aims to compare various extraction methods in crustaceans by-products, which will aid identify a more workable platform to minimize waste disposal and maximize its value for best valorization practices.

Key scientific concepts of review: The fully integrated applications (agriculture, food, cosmetics, pharmaceuticals, paper industries, etc.) of multiple biomaterials from crustaceans by-products are presented. The pros and cons of the various extraction methods, including chemical (acid and alkali), bioprocesses (enzymatic or fermentation), physical (microwave, ultrasound, hot water and carbonic acid process), solvent (ionic liquids, deep eutectic solvents, EDTA) and electrochemistry are detailed. The rapid development of corresponding biotechnological attempts present a simple, fast, effective, clean, and controllable bioprocess for the comprehensive utilization of crustacean waste that has yet to be applied at an industrial level.

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One feasible way for best valorization practices is to combine innovative extraction techniques with industrially applicable technologies to efficiently recover these valuable components.

Contents

Introduction

Crustaceans are segmented invertebrates and mainly encompass shrimps, crabs and lobsters, which are harvested for food and then processed at a large scale for export being favored sea food dishes worldwide [1]. According to FAO 2020 statistics, the global aquaculture production of crustaceans exceeded 9.3 billion tons annually, accounting for *ca.* $69.3 billion, with shrimps production ranking first, followed by lobsters and crabs [2]. The unavoidable growth of the world's population, along with the growing consumer demand, will lead to a dramatic increase in global crustacean waste (mainly cephalothoraxes, shells and exoskeletons) in the coming decades, which makes up to 50% to 70% of the original weight [3]. An estimated 6 to 8 million tons of crustacean waste are generated annually across the globe [4]. After processing, ca. 45% to 60% of whole shrimp become by-products; the percentage varies depending on the species, area, and processing techniques [5]. In addition, lobster processing globally generates more than 50,000 tons of by-products, with disposal costs estimated at $7.5 million annually for the industry as a whole [6]. The same is true for the by-products generated during crab processing [7]. Currently, residues disposed of as municipal solid waste end up in landfill or in the sea; dumping costs can be as high as $150/ton, compared to the market price of $100–120/ton for this waste [8]. Therefore, the unscientific disposal of crustaceans' by-products (CBPs) can cause huge disposal costs, lead to serious pollution of the marine environment, create risks to human health [9]. More importantly, these CBPs encompass a plethora of precious bioactive ingredients with huge market potential [10]. The residual biomaterials from CBPs discarded in aquatic products processing encompass about 20%–40% of protein, 20%–50% of mineral salts (mainly $CaCO_3$), 15%–40% chitin, together with several minor components including lipids, pigments (such as astaxanthin) and other minerals, depending on the origin, season, species, age, among other factors (Fig. 1) [6,10].

Notably, extraction of the biomaterials from CBPs and using them directly or after additional processing may be a strategy to reduce waste and create valuable compounds with exceptional biological properties that might be used in a variety of fields (Fig. 2) [11,12]. In addition to producing large economic benefits, the effective use of CBPs would also aid to solve waste management issues related to the crustacean industry. Traditionally, CBPs are used to produce bioactive ingredients with the aid of extensive chemical methods [13–15]. These methods are against the current green chemistry trend that aims to avoid unnecessary creation of hazardous effluent, which is problematic from both economic and environmental standpoints. Therefore, to efficiently valorize CBPs into high value-added products, appropriate methods with minimum environmental impact is urgently needed as an alternative for chemical methods [16]. Recently, within the blue biotech era, several biotechnological and novel extraction methods have been used, such as enzymatic hydrolysis and fermentation [17,18], microwave-assisted [19], ultrasound-assisted [20,21], ionic liquid extraction [22] and natural deep eutectic solvent [23]. These biotechnological methods will help the sustainability requirement and are anticipated to become the standard in the future.

Despite the fact that there are numerous studies on active ingredients, extraction, and applications of marine by-products [12,24], the extraction and application of the active ingredients of the three primary CBPs, shrimp, crab, and lobster, are rarely documented and compared in a single study. Besides, the amount of CBPs used for large-scale production is still rather limited in comparison to the tons produced, mainly due to the fact that there are few effective and standardized methods for converting these resources into a marketable form. Therefore, this study provides the first holistic comparative overview of the bioactive components, their currently available extraction methods, industrial applications of the three main CBP sources, and provides proposals for future large-scale production of their active ingredients. We thoroughly searched PubMed and Web of Science (from 2010 to June 2023) for published research. These key terms were used ("Crustacean" AND "Shrimp" AND "Lobster" AND "Crab" AND "by-products" AND "Chitin" AND "Protien hydrolysates" AND "Lipids" AND "Astaxanthin"). Finally, we mentioned 200 papers in our exhaustive review based on the titles and abstracts of the search results.

Chitin and its derivatives (chitosan and chitooligosaccharides)

The basic properties of chitin and its derivatives

Chitin $(C_8H_{13}O_5N)n$, the world's second largest polymer after cellulose, is a copolymer comprised of *N*-acetyl-D-glucosamine units joined by β-(1–4) glycosidic bonds (Fig. 3A) [25]. Chitin is a rigid, inelastic, white or yellow nitrogenous polysaccharide that is generated at a pace of about 100 billion tons annually [26]. Three polymorphic forms of chitin are found in nature: α-chitin (anti-

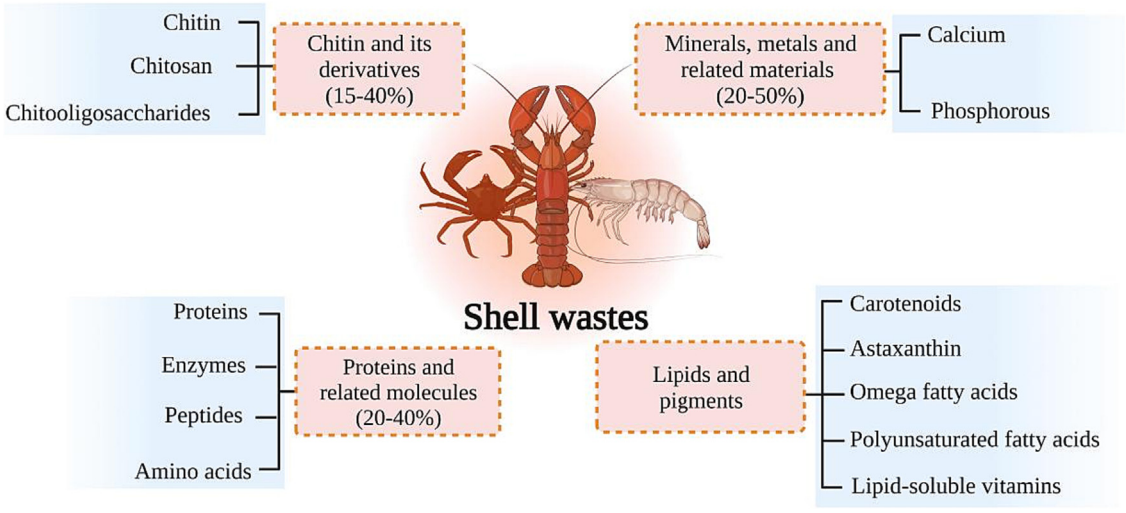

Fig. 1. Main active ingredients from three main crustaceanś by-products.

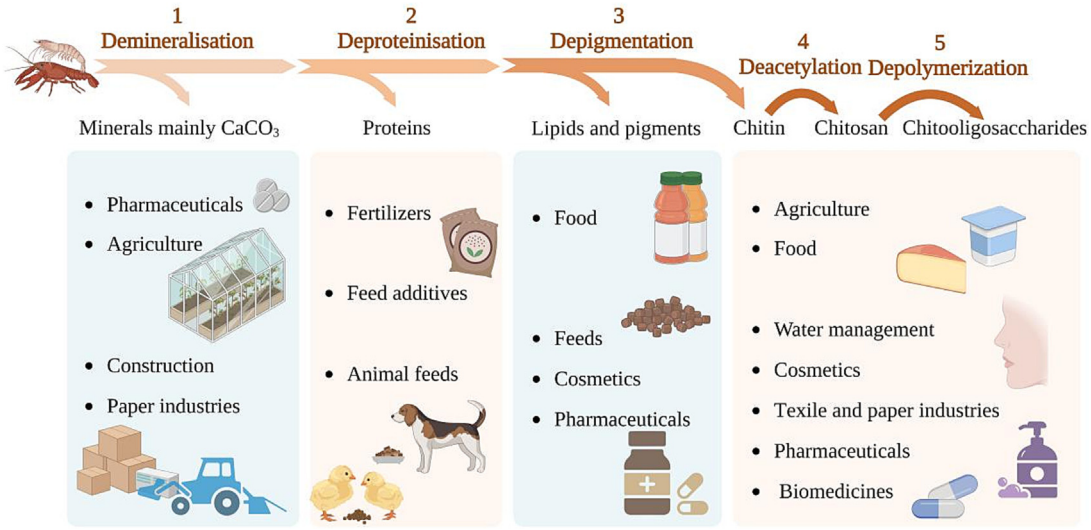

Fig. 2. The comprehensive applications of crustaceanś by-products in various fields.

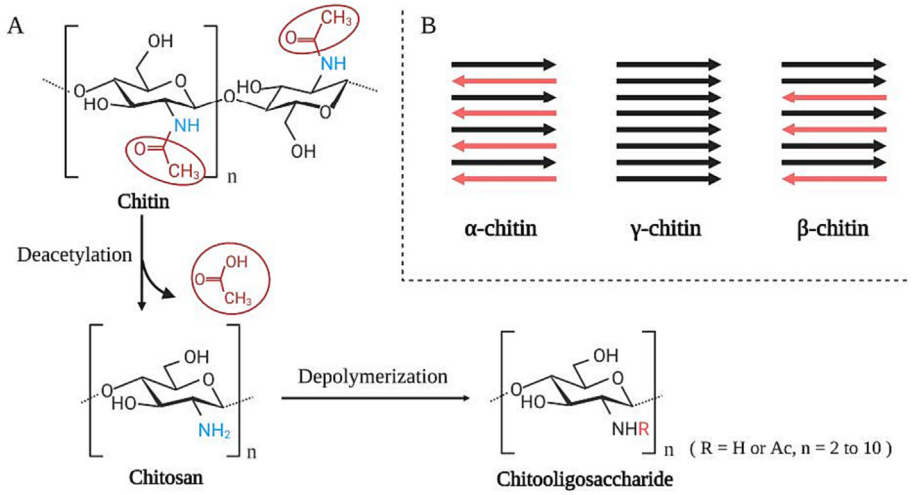

Fig. 3. The chemical structure of chitin, chitosan and chitooligosaccharides (A) and schematic diagram of chitin's crysalline structure (B).

parallel arrangements), β-chitin (parallel arrangements), and γ-chitin (mix α of and β arrangements) (Fig. 3B) [27], with α-chitin being the most prevalent form and responsible for the polymer's rigidity in crustacean shells [28]. Due to the potent intermolecular hydrogen bonding, chitin is extremely hydrophobic and insoluble in water as well as the majority of organic solvents, while a few solvents are capable of dissolving it [29]. Crustacean shells are a natural source of commercial chitin, with various sources yielding different levels of chitin from shrimp (14%–30%), lobster (16%–23%), crab (14%–28%), depending on species, organism, nutrition status, processing methods and other factors [1,10].

Chitosan $(C_6H_{11}O_5N)n$, the only naturally occurring cationic polysaccharide, is a well-known *N*-deacetylated derivative produced by partial chemical or enzymatic chitin deacetylation (Fig. 3A) [30]. Therefore, chitosan encompasses copolymers of repeating units of D-glucosamine (GlcN, deacetylated units) and *N*-acetyl-D-glucosamine (GlcNAc, acetylated units). Additionally, the GlcN/GlcNAc ratio serves as a broad indicator to distinguish chitin from chitosan. In order to be classified as 'chitosan', chitin must be at least 50% deacetylated and therefore contain at least 50% GlcN, which affects the chemical properties of chitosan (e.g. solubility, viscosity, flexibility, tensile strength). Chitosan is insoluble at neutral and basic pH levels but soluble in aqueous acid (pH between 2 and 6) [31]. In addition, the amino and hydroxyl groups of chitosan affect its biological properties (e.g. non-toxicity, bioavailability, biocompatibility, biodegradability, hemocompatibility, mucoadhesiveness, antioxidants and adsorption enhancers), expanding the range of chitosan applications [32,33].

Chitooligosaccharides (COS), also known as chitosan oligomers or chitooligomers, are chitosan depolymerized products produced by acid hydrolysis (Fig. 3A) [34]. Chitosan classified as COS has an average molecular weight of less than 3900 Da and a degree of polymerization of less than 20 [35]. COS is easier to work with and more suitable for large-scale industrial applications than chitosan because of its short chain length, low molecular weight, low viscosity, and high solubility [36]. Excellent biological characteristics of COS have been described, including anticancer, antitumor, antibacterial, cholesterol-lowering and immuno-enhancing activities, warranting for its inclusion in health applications [37].

Chemical extraction of chitin and its derivatives

The various crustacean shells represent major sources of chitin, in which the compact matrices of chitin fibers are interlaced with

proteins and strengthened by the deposition of minerals (calcium carbonate) and carotenoids (mostly astaxanthin) [38]. Hence, current industrial methods for chitin and its derivatives isolation rely on chemical processes involving: (1) grinding into fine power; (2) demineralization (DM); (3) deproteinisation (DP); (4) depigmentation; (5) deacetylation to form chitosan; (6) depolymerization to form COS [39]. Chitin can be produced using a variety of methods, among which chemical and biological processes are the two most common ones.

The most common method for demineralizing shells is acid treatment (including HCl, HCOOH, HNO_3, CH_3COOH, H_2SO_4), which removes minerals like calcium carbonate and calcium phosphate by heating the extraction temperature above 100 °C for a longer period of time. HCl is the preferred solvent for extracting mineral elements among these acids [40]. Different shell types, extraction times, temperatures, and acid concentrations affect the demineralization's characteristics in different ways. For example, shrimp shell is thinner, making its chitin separation efficiencies higher, while the yield is less and the quality is lower compared with lobsters and crabs. Harsh acid treatment may lead to modification, such as depolymerization and deacetylation of chitin. Mild acids, including formic acid, acetic acid, citric acid, sulfurous acid, are good solutions to the fore-mentioned problems, while the recovered chitins resulted in high residual ash content [41].

Demineralized shells are deproteinated utilizing an alkali treatment with widely available chemicals like NaOH, KOH, Na_2CO_3, $NaHCO_3$, Na_2SO_4, $NaHSO_4$, Na_3PO_4 etc. Among these alkalis, NaOH is the preferential solvent with its concentration ranging from 0.125 to 5.0 M, coupled with different temperatures (up to 160℃) and extraction time (from few minutes to few days) [41]. The chemical links between protein and chitin are cleaved during deproteination. However, prolonged exposure with strong alkali causes chitin to become depolymerized and deacetylated.

Colored chitin is produced during the demineralization and deproteination of CBPs. The final stage in producing colorless chitin for consumer preferences is depigmentation. To remove pigments, including carotenoids, chemical solvents (acetone, ethanol) and strong oxidants (H_2O_2, NaOCl, and $KMnO_4$) are often used for 10 to 20 min before being dried for 2 h at ambient temperature [2].

Conventional chemical process of converting chitin to chitosan in concentrated NaOH at preferably high temperatures is widely used for large-scale production due to its low cost and short processing time, although this poses significant environmental concerns, low reproducibility and high energy consumption [14,42]. Glycerol can be used as a reaction solvent for the conversion of chitin to chitosan, with the advantage of not only recycling glycerol but also reducing the alkali concentration required for the deacetylation reaction, thus reducing the environmental impact of the process [43].

Last but not least, COS is produced by by depolymerization of chitin or chitosan with chemical acids as HCl, HNO_2, H_3PO_4, etc. [44]. Chitosan's molecular weight can be reduced through the usage of free radicals. Chitosan's viscosity can be reduced using H_2O_2 or potassium persulfate degradation [35]. Investigations into the degradation of chitin or chitosan by lactic, formic, and trichloroacetic acids were also conducted. These treatments did, however, yield secondary compounds that are challenging to remove due to the complexity of the reaction process.

The chemical treatment for chitin extraction is the most commonly used technique commercially nowadays for its short processing time, while it holds several disadvantages: (i) negative impact on the physico-chemical properties of chitin; (ii) environmentally unfriendly; (iii) higher solvent consumption; (iv) the extracted protein and minerals are damaged for no longer appropriate applications in human foods [45]. Therefore, biocatalysis and biotransformation using enzymes and microorganisms offer an alternate method to extract chitin from CBPs without risking the harsh acid and alkali damage.

Bioprocesses of chitin and its derivatives

Bioprocesses with enzymes and microorganisms can also be used in chitin recovery including: (1) enzymatic demineralization; (2) enzymatic or fermentation deproteinisation [46]. Other procedures such as decoloration largely remain the same. A comprehensive comparison between chemical extraction and bioprocess of chitin and its derivatives is shown in Fig. 4 highlighting each advantages and limitations.

Fermentation processing method

Microorganism-based fermentation is an intriguing new technique for chitin extraction that provides an alternative to the harsher chemical processes. Fermentation is envisioned as one of the most ecologically friendly, safe, adaptive technologically, and economically feasible alternative approaches [39]. The fermentation of CBPs can be carried out by selected strains of bacteria that produce lactic acid (LA) and protease [47]. During fermentation, the LA produced reacts with calcium carbonate present in the chitinous fraction, resulting in the formation of calcium lactate, which can be removed after precipitation. In addition, the low pH also inhibits the growth of spoilage microorganisms [40]. The deproteination of CBPs occurs mainly by extracellular proteases produced by the added LA bacteria [48,49]. The efficiency of LA fermentation depends on many factors, mainly the differences between microbial species, the added carbon source, the initial pH, the temperature and the duration of fermentation [18]. A variety of microorganisms and methods suggested for CBP biofermentation were summarized [2,46,50].

Non-LA bacteria, which produce proteases with high proteolytic activity, have been reported to be used in the fermentation of CBPs as well [51]. However, not all non-LA bacteria create enough organic acid to dissolve calcium carbonate [48]. In some cases, co-fermentation with acid-producing bacteria or two-step fermentation has been used [49,52]. Similar to LA fermentation, many factors have been reported to influence the fermentation process, i.e. crustacean shell concentration, added glucose concentration, incubation time and inoculum size [53].

Enzymatic extraction

Enzymes are advantageous as they minimize or generate near-zero wastes by transforming crustacean wastes into value-added products [54]. Enzymatic hydrolysis is regarded as a mild process that create high-yielding products without compromising nutritional quality by, for example, degrading amino acids as seen in chemical hydrolysis. Enzymatic deproteination is accomplished with the use of proteolytic organisms or commercial proteases such as alcalase, papain, pepsin, pancreatin, trypsin and others, among which alcalase is the most common commercial protease to eliminate proteins from CBPs [55]. Because commercial proteolytic enzymes are rather expensive, using crude extracted proteases produced from bacteria and marine animal viscera for chitin recovery has received great interest in studies during the last few decades especially if to be applied at commercial level [56]. Numerous parameters, including enzyme: substrate ratio, temperature, and incubation period, might have a considerable impact on protein hydrolysis [57]. Although enzymatic extraction is a significantly faster and more convenient technique to extract chitin from CBPs with high deproteination efficiency, demineralization with organic or inorganic acid is still required to achieve highly purified chitin.

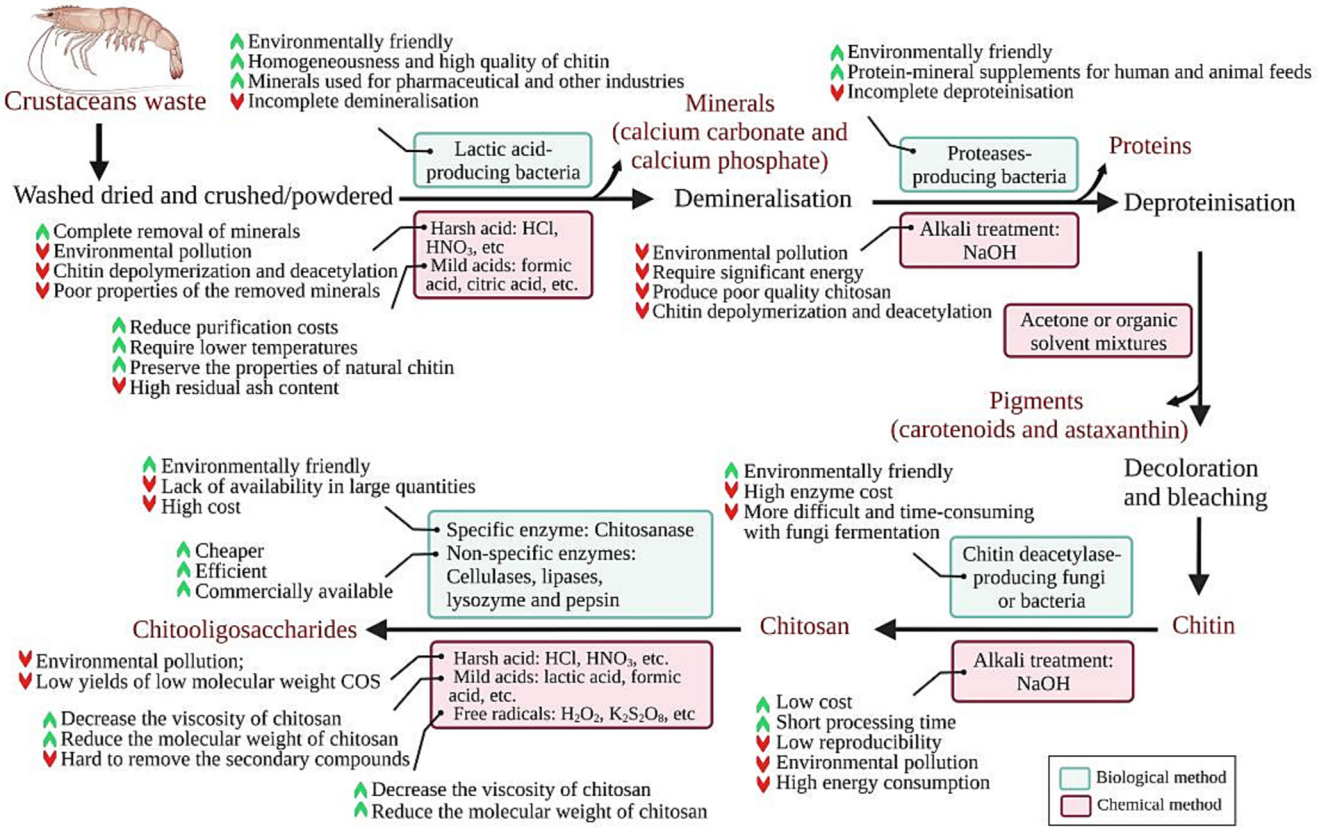

Fig. 4. A comparison of the chemical and biological processes used to remove chitin from crustacean by-products.

Chitin deacetylase from enzyme-producing fungi and bacteria can be used to produce chitosan, with fungi being more difficult and time-consuming to culture in industrial-scale fermentation systems [58,59]. Specific chitosanases and non-specific enzymes from bacterial, fungal, mammalian, and plant sources, such as cellulases, lipases, lysozyme, hemicellulases, pectinases, and pronases, can generate COS [60]. Specific chitosanases are restricted in use due to their high price and little availability in large quantities, whereas these non-specific enzymes are less expensive, more readily available and more efficient for industrial use.

It should be noted that the enzymatic method is less efficient than chemical method, with *ca.* 5% to 10% of residual protein usually still associated with the separated chitin, which must then be treated with additional alkali to obtain more purified chitin under milder conditions [41]. In general, the ultimate yield and quality of the recovered chitin are unaffected by the order of demineralization and deproteination during chemical processing [38]. However, if enzymatic deproteination is carried out first, minerals present in CBPs may lessen the effectiveness of the proteases. Therefore, demineralization should therefore be done first during enzymatic extraction.

The advantages of biological extraction include: (i) environmental safety; (ii) ease of manipulation; (iii) negligible solvent consumption; (iv) higher reproducibility in less time; (v) high-quality solubilized minerals and proteins that can be employed in human and animal feed. However, due to concerns with incomplete deproteination, lengthy incubation times, and low-quality chitin depending on the kind of microorganism utilized in fermentation, biological extraction is only suitable for laboratory scale studies. As a result, chemical extraction of waste from crustaceans' shells is still more practiced [61].

Physical assisted extraction of chitin and its derivatives

To improve chitin quality and reduce manufacturing cost, physical assisted methods can be applied before, after or during the demineralization and deproteination process as a more environmentally friendly option. Common physical assisted techniques include microwave assisted extraction (MAE) [62], ultrasound assisted extraction (UAE) [63], and HOW-CA process [64]. A comparison of the advantages of the physically assisted extraction methods is shown in Fig. 5.

Microwave assisted extraction

Among numerous extraction methods, microwave has been found to be successful as a green, economical and sustainable method to assist in the demineralization of chemical and enzymatic chitin extraction [65]. Microwave heating has been shown to have a great potential to accelerate chemical reactions, boost reaction yield, improve product purity and properties compared to conventional heating [66–68]. Microwave heating is primarily accomplished through two mechanisms: (i) dipolar polarization and (ii) ionic conduction [69]. High yields of chitin were produced using MAE and the lactic acid demineralization procedure with barely any remaining minerals (0.2%) [70]. Besides, microwave technology can break down chitin and chitosan into low molecular weight chitosan or chito-oligomers applied in a number of fields [71,72]. For example, the microwave-intensified lobster shell protein hydrolysate exhibits outstanding functions in terms of solubility, emulsification, foaming, water absorption, oil absorption, and nutritional value for food applications [73]. Besides, the degree of deacetylation and the molecular mass of the resulting chitosan

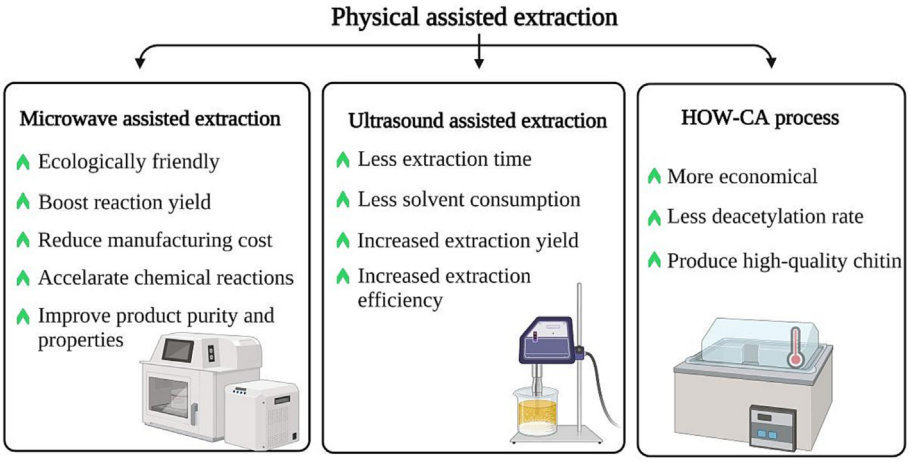

Fig. 5. A comparison of the advantages of microwave assisted extraction, ultrasound assisted extraction and HOW-CA process.

are affected by different microwave working settings such as reaction time, solvent concentration, and solid-to-liquid ratio.

Ultrasound assisted extraction

UAE is a physical extraction method that uses ultrasonic waves to break up the material's cells and release the target compounds more quickly and effectively [74]. Due to depolymerization of macromolecules, dissociation of covalent connections in polymer chains, and dispersion of aggregates, the cavitation effect of ultrasound enhances the solubility of protein coupled with chitin [75]. UAE increases the efficiency of chitin extraction, reduces extraction time and avoids the requirement for high temperatures [76]. Currently, UAE is a technology that has been successfully used to improve the speed of many extraction procedures [63,77].

HOW-CA process

The HOW-CA technique, in which HOW stands for Hot Water and CA refers for Carbonic Acid, is another successful and established method for extracting chitin [78]. In order to reduce expenses and waste production, the HOW-CA method only uses water and CO_2 as reagents, with hot water being used to denature proteins and carbonic acid being used to dissolve minerals. At high temperatures, proteins are partially hydrolyzed and completely dissolved in water. Then, calcium carbonate is dissolved in an aqueous solution while being under pressure from CO_2 at room temperature. When CO_2 is released, calcium carbonate re-precipitates, allowing for calcium carbonate recovery through filtration. To reduce capital expenditures, water and CO_2 can both be recycled in a semi-batch fashion. Using method modeling, techno-economic analysis and life cycle evaluation, the HOW-CA approach was found to be more cost-effective, economic and ecological than traditional chemical methods [64]. Furthermore, the HOW-CA process generates high quality chitin (90%) with less deacetylation, making it suitable for a larger scale production.

Solvent extraction of chitin and its derivatives

Ionic liquids (ILs)

ILs are gaining an increasing interest from researchers due to their excellent thermal and chemical stability, high conductivity, and potent solubilizing capacity for various organic or inorganic solvents [79]. Compared to traditional organic solvents, they increase selectivity and reduce environmental impact. ILs typically contain particular organic cations, and organic or inorganic anions that, under appropriate conditions can dissolve chitin without causing the polymers to break down during extraction. Imidazoles, morpholines, pyrroles, quaternary ammonium salts, quaternary phosphonium salts, and other compounds are examples of common ILs [80]. For instance, chitin was produced from the crab shell by 1-allyl-3-methylimidazolium bromide ([AMIM][Br]), with a yield of 12.6% and acetylation of 93% [81]. Another novel ammonium-based ionic liquid was employed to recycle 13.4 % of the medium molecular weight chitin found in shrimp shell waste [82]. Although ILs have a strong solubilization capacity for chitin, they are still controversial owing to their inherent toxicity and non-degradability [83]. Therefore, the numerous applications of ILs are severely constrained by their hazardous properties.

Deep eutectic solvents (DESs)

Recently, there has been increasing reports on DESs, an alternative for ILs, as a green solvent for utilization in chitin and its derivatives. DESs have features comparable to ILs, however they are more favorable owing to their biodegradability, low cost, and simple manufacturing procedure [84]. Crustacean shell waste was subjected to the choline chloride-based DESs for chitin separation [85]. Furthermore, DES was also applied in the production of chitin and chitosan film fabrication, chitosan nanomaterials [86,87]. Chitin films that have been DES-plasticized exhibited remarkable performance, including greater elasticity and decreased tensile strength. However, the poor biocompatibility of DES components restricts their usage in the food and pharmaceutical industries. Thus, a superior alternative, natural deep eutectic solvents (NADESs), which are composed of biological metabolites such as amino acids, choline and sugar, was proposed [88]. NADESs is superior to DES for its biodegradability, sustainability, low toxicity and preparation cost because of its natural composition [89]. Additionally, the NADESs method is not only more environmentally friendly and efficient, but also produces chitin with a chemical structure comparable to that of chitin produced using conventional acid/alkali processes [90,91].

Ethylenediaminetetraacetic acid (EDTA)

As an alternative to hydrochloric acid, EDTA is a metal chelator that can demineralize chitin [92]. The chitin molecular chain is not significantly affected by this chemical procedure, and EDTA can be readily recovered or recycled. This process produced much higher levels of chitin than the enzymatic method, but less than the chemical method. Reaction time was much lowered concurrent with a decrease in pollution hazards.

Electrochemistry of chitin and derivatives

Along with green solvents, electrochemistry - a field that has been less investigated- is also gaining increasing popularity. In a study, shrimp shells were electrolytically treated with 1% acidic and alkaline water at 20 V for 6 h [93]. After complete removal of minerals and proteins, 19.5% of the extraction ratio was recovered. Chitin extracted using electrochemical techniques has approximately the same physicochemical characteristics as chitin extracted using conventional chemical techniques. The electrochemical extraction method uses little reagent and is effective. It may be a safer option than chemical ones for the environment. The preparation method is though still in its infancy, with needed work to ascertain the ideal ratio of chemical reagents to produce products with high levels of purity and chitin yield [94].

Comparison and proposal of chitin extraction methods

Overall, chitin is often extracted using single or combination of chemical, biological and physical techniques, with physical techniques commonly utilized to aid extraction. In recent years, numerous innovative extraction techniques have evolved to overcome the potential shortcomings of acid-base procedures, including electrochemical technology and green solvents (ILs, DESs, EDTA). All chitin extraction methods and their characteristics are listed in Fig. 6. According to the 'blue economy' and 'circular economy' in modern societies, the extraction process for chitin would limit its applications. The review of chitin extraction methods has shown potential deficiencies. For example, chemical extraction methods are very economical and efficient, and are widely used in industry. Recovered chitin can be used to develop sorbents and batteries for environmental and energy applications, but is not suited for use in the production of related food products due to residual acids and bases. Eco-friendly biological methods meet the requirements of green development, but are less efficient and can be combined with chemical techniques aimed at increasing efficiency with minimal chemical treatment. Therefore, combining chemical, biological and physical techniques or investigating more cost-effective, environmentally friendly extraction methods can help expand the industrial production of chitin, and raise the standard of the final product. Besides, the development of large-scale studies for chitin recovery of various chemical or biotechnology methods is also still necessary.

Application of chitin and its derivatives

Chitin and its derivatives are rich bioresources with nontoxicity, non-allergenicity, renewability, sustainability, high biodegradability, and biocompatibility and possess many biological properties, including antioxidant, antimicrobial, antifungal, anti-coagulant, antitumor, anti-cancer, cholesterol-lowering properties and bioadhesiveness [95]. Therefore, they are accordingly applied in a wide range fields, such as agriculture, food, wastewater management, cosmetics, textile and paper industry, pharmaceuticals, biomedicine (Fig. 7) [96]. According to the statistics of the global chitin market, the reported net value of the chitin market was USD 36 million in 2019 and is projected to reach USD 53 million by 2024 [97].

Agriculture

Chitin and its derivatives with interesting antimicrobial and eliciting properties are anticipated to exert significant positive impacts upon their application in agriculture [98]. (1) Crop growth. Chitosan and its derivatives are effective in improving seed germination, promoting crop growth, increasing crop yield and improving quality [99,100]. (2) Effectiveness against pest and

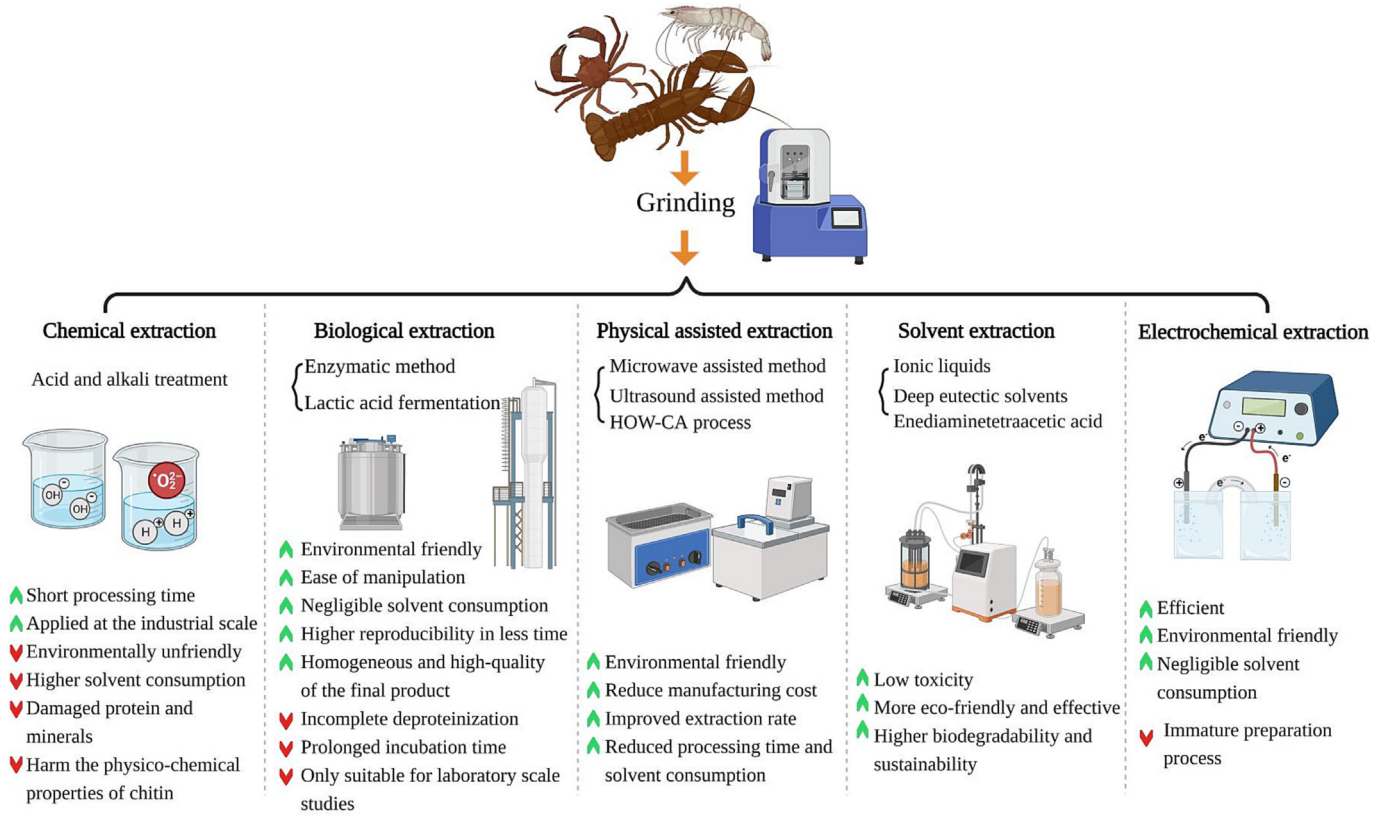

Fig. 6. A comparison of the advantages and disadvantages of the different chitin extraction techniques.

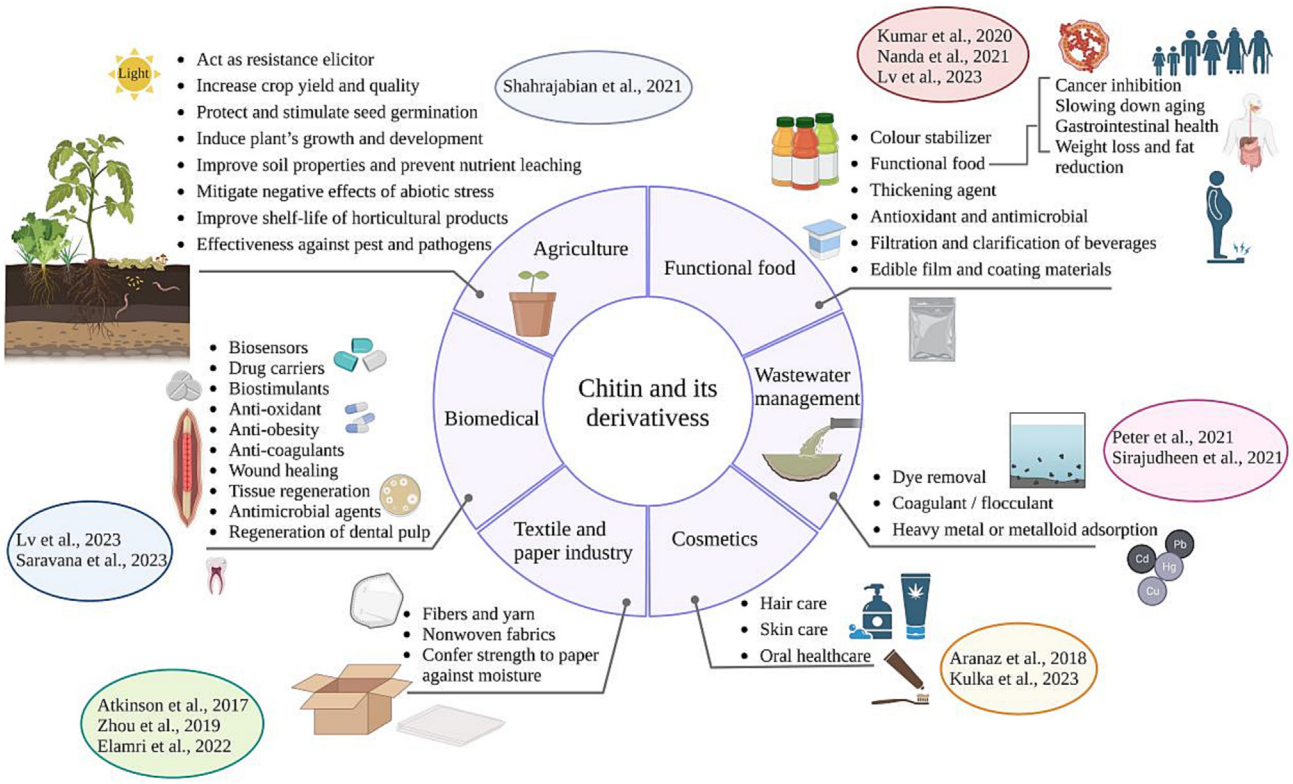

Fig. 7. A comprehensive overview of chitin applications and its derivatives.

pathogens, e.g. fungi, bacteria, nematodes, insects and viruses [101,102]. (3) Crop defense. The application of chitosan and its derivatives induces the production of various resistant substances such as lignin, resistance proteins and guaifenesin in plants, minimizing the damage caused to crops by stresses of adversity [103]. (4) Fertilizer and soil amendments. Chitin's low C/N ratio and high nitrogen concentration can be used to promote crop development and microbial activity in the soil [104]. (5) Fruit and vegetable preservatives. Fresh fruit and vegetables can be coated with chitosan to prevent bacterial and pathogen deterioration, extend shelf life, maintain quality, and reduce water loss [105,106].

Food

Chitosan has strong antioxidant activity and is recommended as a food preservative [107,108]. Besides,[108] chitosan possesses antibacterial qualities that guard against microbial food spoilage, off flavors and extend the shelf life of the foods [109–111]. The internal molecules of microorganisms seep out when the positive charges in chitinous materials interact with the negative charges of bacterial cell walls, which is assumed to be the reason for their antibacterial activity [27]. Active edible or biodegradable packaging has also been developed using chitosan's antimicrobial characteristics [112] Chitosan added to food can confer specific functional effects that contribute to human health, such as weight loss and fat reduction, gastrointestinal health, age-delaying, cancer inhibition [92,113].

Wastewater management

For decades, non-toxic and biodegradable chitosan and chitosan have been utilized as coagulating agents, cheating polymers, or bio-absorbents in water treatment due to their strong absorbance, chelating, and affinity capabilities [114,115]. Due to their polycationic properties, they can agglomerate and precipitate at neutral or alkaline pH. Moreover, the polymer long chain may enhance contact with the contaminated medium [116]. As a result, chitosan has potnetial advantage compared with other polysaccharides (such as cellulose or starch) in that its chemical structure permits specialized alterations to design polymers for particular applications. Reactive groups have the capacity to combine several chemicals to form composites. Instead, the cationic charge can successfully neutralize and flocculate the anionic suspended colloidal particles, lowering the levels of turbidity, chlorides, and chemical oxygen demand in wastewaters [117]. Chitosan is used as a coagulant/flocculent for contaminated wastewaters, in heavy metal or metalloid adsorption (Cu(II), Zn(II), Pb(II), Cd(II), Fe(III), Cr(III), etc.) [118,119], and to remove dyes from industrial wastewater as well as other organic pollutants like organic oxidized, organochloride pesticides, or fatty and oil impurities [120].

Cosmetics

Chitosans alongside their derivatives can be produced with various chain lengths and distinctive properties for their use in cosmetics, including skin care (face and body creams, make-up, lotions and nail lacquers), hair care (hair additions, hair colorant, hair spray and shampoos), and oral care (toothpaste, oral hygiene agent, mouthwashes and chewing gum) [121]. The majority of chitosan products cannot enter the skin due to their exceptionally high molecular weight, which is a significant advantage that makes them suitable for skin care. They can either be employed in solid form or dissolved in aqueous solutions. Chitosan has special properties that are employed in cosmetics, such as its ability to produce foam, retain moisture, be antistatic, bacteriostatic, fungistatic, and release bioactive compounds under controlled conditions [122]. Chitosan is also widely used in cosmetic formulations because it interacts well with other ingredients such glucose, starch, oils, lipids, waxes, acids, saccharose, polyols, nonionic emulsifiers, and nonionic water-soluble gums [105].

Textile and paper industry

In recent years, due to the similarity of chitosan molecule to cellulose, water-solubility and bioadhesiveness with their positive charges, chitin and its derivatives can be used in texture and paper industry, for both internal and surface applications [123,124]. In the textile sector, they are commonly used in a wide range of products, such as chitosan nonwoven fabrics, chitosan fibers and yarn, pretreatment of textiles, dyeing, printing, and functional fabric finishing [125]. In the papermaking industry, chitin and its derivatives can be used for improving the wet and dry strength of paper [126,127], showcasing the compatibility of chitosan with paper stock components, and demonstrating its capacity to function as a retention and drainage additive, or as a dye fixative in the production of colored paper [128]. In the meantime, research is being conducted on chitosan's intrinsic antibacterial characteristics and its capacity to form films for prospective uses in papermaking, establishing the groundwork for creating useful papers like greaseproof paper and antibacterial paper [129]. Chitosan is posed as an interesting and feasible option for producing high-value, ecologically friendly paper. However, the economics of chitosan's widespread applicability in the paper industry have not yet been considered [130].

Biomedical applications

Chitin and its derivatives have gained more recent attention for use in pharmaceuticals and biomedicines due to their biocompatibility, biodegradability, and lack of toxicity [50]. The Food and Drug Administration (FDA) announced that the usage of chitosan in foods and drugs is considered safe, devoid of contaminants, and has good adsorption and moisturizing properties [131]. They also exhibit several biological and physiological traits with proven health benefits. These polymers alongside their derivatives, for instance, exhibit antioxidant, antibacterial, anticancer, immunestimulating, hypocholesterolemic, hypoglycemic, ACE inhibitor, and anticoagulant properties [132]. In the biomedical field, chitin and its derivatives are usually used for drug delivery, bone regeneration, blood cholesterol control, tissue engineering, wound healings and enzyme immobilization [132–136]. Given their wettability, mechanical stability, flexibility, optical clarity, transparency, gas permeability, and immunological compatibility, chitin and its derivatives are particularly well-suited for use in the production of contact lenses [137,138].

Crustaceans' proteins and related molecules

The properties of protein hydrolysates in CBPs

Protein, which can account for up to 40% of the total wast weight depending on processing methods and species, is another component found in the waste stream that has potential nutritional value and function [139]. Shrimps and crabs have comparable protein content, while lobsters exhibit lower levels [140]. Crustacean proteins are rich in non-protein nitrogen (amino acids, peptides and nucleotides) and their richness in arginine, glutamic acid, glycine and alanine can be used as dietary supplements for humans or animals [141]. In addition, the amino acid composition of crustacean proteins is similar to that of red meat proteins [1]. Collectively, the nutritional value of crustacean protein is comparable or superior to that of soy protein, milk protein and red meat protein due to its optimal amino acids composition [3,10].

Bioprocesses for isolating protein from crustaceans

Chemical extraction

The protein found in CBPs is strongly linked to chitin and minerals. Similar to chitin, the method of recovering proteins from CBPs involves deproteination. The most commonly used DP strategy is chemical hydrolysis using acidic or alkaline reagents because of its low cost, rapidness and high protein recovery. Despite being a common and easy-to-use method, chemical extraction has some negative effects such as environmental pollution, decreased nutritional quality, and a poor final product value [45].

Fermentation process method

Studies demonstrated that biotechnology based methods are more safer, cleaner, greener, environmentally friendly and cost effective process as they maintain the physical and functional integrity of the ingredients of interest [142,143]. Autolysis, enzymatic processes, and fermentation have all been highlighted as three biotechnological techniques [144,145]. The crustacean's natural digestive enzymes are used in autolytic method for protein extraction. For instance, shrimp heads or residues can be used as a source of protein via an autolytic method [146]. The effectiveness of protein recovery is increased by shrimp-crab *endo*-enzymes and autolysis, which also produced important small peptides for fish nutrition [147].

Enzymatic extraction

In the enzymatic hydrolysis method, enzymes such as Protamex, Flavourzyme and mainly Alcalase are frequently used [148,149]. In addition to separating proteins and peptides, enzymes can also hydrolyze proteins into free amino acids, making them more accessible, depending on the degree of hydrolysis [31]. Enzymatic hydrolysis is the most widespread biotechnological technique for releasing bioactive peptides from proteins in CBPs because of the mild and controlled conditions, minimizing the risk of adverse reactions [150]. Moreover, enzymatic extraction is widely used in the pharmaceutical and food industries due to the absence of harmful chemical residues. Precision is provided by this technique, which also enhances the end product's physical, chemical, and organoleptic qualities while preserving its nutritional content [151,152]. However, this method still has several drawbacks, including lower yields, taste problems, general economic viability, and a lack of uniform hydrolysates.

Isoelectric solubilization/precipitation (ISP)

The more innovative, ecological and economical techniques, such as ISP, are necessitated to recover nutritious and functional protein isolate from CBPs [153]. The ISP method depends on altering protein solubility by adjusting pH, which changes the net electrical charge of proteins [154]. When CBPs proteins are dissolved, they are dispersed from lipids and other insoluble substances like bones, skins, etc. The approximate composition of protein isolates obtained by ISP processing changes depending on protein source, pH and the type of acid and base utilized during ISP. This approach not only results in high yield protein recovery but also in high-quality protein with increased nutritional and functional value [155]. ISP technique has been utilized to recycle fish protein in laboratory and pilot size applications due to its ease of use and speed [154]. Compared with traditional methods, ISP method has significant advantages, and has great nutraceutical food application potential in protein recovery from CBPs.

The application of protein hydrolysates in CBPs

Crustacean proteins, which are highly delicious, palatable, and nutritious, are also used in pharmaceuticals, cosmetics, human and animal nutrition, and the treatment of diabetes, cancer, hypertension, inflammation, and neurodegenerative illnesses [149,156,157]. For example, protein hydrolysates that were extracted from the by-products of lobsters showed outstanding emulsifying properties [158]. Hydrolyzed lobster by-product protein could be employed as flavor enhancers in a variety of designed food products [3], improving water-binding or lowering lipidemic effects of animal protein [73]. Additionally, protein hydrolysates represent a rich source of nitrogen for microorganisms in their growth environment [159,160]. Currently, these proteins are not extensively used as they are destroyed during the recovery process, but the World Bank estimates that if waste from South East Asia were simply made into protein meals, the market value would be over $100 million annually [10].

Crustaceans' lipids and pigments

The properties of crustaceans' lipids

Crustacean shells are abundant in omega fatty acids, polyunsaturated fatty acids (PUFAs) and lipid-soluble vitamins (A, D, E, K) that vary in context to species, gender, weather and environment [161]. Eicosapentaenoic acid (EPA) and docosahexaenoic acid (DHA), which are well known for their medicinal and nutraceutical applications, were the two primary omega-3 fatty acids in PUFAs and are found at higher levels in specific organs, i.e., liver and pancreas in shrimps, the head of lobsters, and the gonads of crabs [162,163]. In comparison with other crustaceans (crabs and shrimps), lobsters have the highest lipid level, with different composition in various lobster body areas [164]. A comparison of lipid content between crustaceans showed that crabs have a significantly lower proportion of lipids in edible tissue than shrimps [165].

The properties of crustaceans' pigments

The fat-soluble pigments carotenoids, including zeaxanthin, lycopene, β-carotene, lutein and astaxanthin, the oxidized form of carotenoids, provide crustaceans with their characteristic pink-orange colour [166]. Astaxanthin ($C_{40}H_{52}O_4$) is the most prevalent carotenoid in crustacean shells, accounting for ca. 75%–95% of total pigment concentration [166]. Crustaceans astaxanthin levels vary according to the species, season, and other environmental circumstances. Generally, they are twice as high in lobster by-products than in shrimp, but still lower than in crab [3]. Red fat-soluble pigment astaxanthin requires FDA permission before it may be used as a food colorant [167]. Most of the astaxanthin used in commercial products is created synthetically. As a result, discarded CBPs provide a significant potential supply of natural astaxanthin. Astaxanthin can be found in free form or as complex proteins called carotenoproteins in crustaceans. Astaxanthin has 10 fold higher antioxidant activity than zeaxanthin, lutein, canthaxanthin, and β-carotene, and 100 fold higher antioxidant activity than α-tocopherol [3].

Extraction methods of lipids and pigments

Solvent extraction

Solvent extraction for recovering lipids and astaxanthin has always been the most prevalent method considering their lipophilic nature [168]. Different solvent types are crucial to the extraction process and yields. Solvent selection has been based on factors such as solvent toxicity, solvent polarity, handling risk, ease of solvent removal, etc. [169]. Acetone, ethanol, ethyl acetate, hexane and isopropanol are a few organic solvents that have been approved for use in the food industry. Acetone is recommended as the most suitable astaxanthin extraction agent among those investigated because of its structure's abundance of carbonyl groups, many of which are quite comparable to those found in astaxanthin [170]. Moreover, the effects of single organic solvents and mixed solvents (isopropyl alcohol: hexane = 1:1) on astaxanthin extraction were also studied, and results showed that the higher extraction rate was achieved with mixed solvents. Other organic solvents including alcohol and vegetable oils are used for safer extraction compared with traditional solvent extraction [171].

Solvent extraction is considered expensive, time-consuming, hazardous and environmental unfriendly. A poorer yield is obtained and huge amounts of solvent are consumed when the wrong solvent is used. Besides, the abundance of organic solvents damages the structure of astaxanthin and reduces its stability. The toxicity and flammability of solvents and the difficulty of removing traces of solvents from the final product have prompted the development of alternative extraction procedures that are green and environmentally friendly.

Supercritical fluid extraction (SFE)

SFE uses supercritical fluids, primarily CO_2 under supercritical conditions, to extract biologically active molecules from solid or liquid materials, overcoming the drawbacks of conventional solvent extraction [172]. SFE is a quick and effective extraction technique without the use of potentially harmful organic solvents. Another significant benefit is the simplicity of solvent separation following extraction because CO_2 turns into a gas at ambient temperature [173]. Since supercritical CO_2 (SC-CO_2) is an inert, affordable, nonflammable, nontoxic, and usually regarded as safe solvent, it is perfect for usage in the food business (GRAS). Since SFE does not require high temperatures for processing, it is especially employed to extract heat-sensitive substances like carotenoids and lipids. The SC-CO_2 method has been widely used in shrimp by-products, lobster livers and crab shell waste [161,174]. However, the cost of adopting SFE technology and its economic viability on an industrial scale are two important factors that limit the use of SFE in the food industry.

Pulsed electric field (PEF) extraction

PEF, a non-thermal method, has been widely employed in the food industry for biomaterial extraction [175]. PEF has a minimal operating cost and barely raises the samples' temperature [176]. The PEF device consists of three main parts: a high voltage pulse generator that produces high voltage pulses using high capacity capacitors, a treatment chamber that contains the sample to be treated and two electrodes, and an oscilloscope that records the wave of applied pulses. PEF technology can replace conventional extraction techniques to boost the extraction yields of bioactive compounds whether used alone or in combination with other extraction procedures as an intensification pre-treatment [177]. For example, setting electric field intensity at 4–16 kV cm^{-1}, PEF pretreatment of the Pacific white shrimp cephalothorax boosted the extraction rate of lipids by 61.3% [178].

Other novel extraction methods

In addition to SFE and PEF, other novel methods that have recently been applied to the extraction of lipids and carotenoids from CBPs include UAE, MAE, enzyme-assisted extraction, high-pressure processing method and microbial fermentation [170]. UAE is increasingly popular due to various benefits such as

Table 1
Advantages and disadvantages of the lipids and pigments extraction methods.

Technology	Characteristics	Advantages	Disadvantages
Conventional solvent extraction	Widely used technique; Uses a variety of organic solvents	No need for any special experimental setup; Economical of all methods	Longer extraction times; Lower extraction yields; Higher solvent consumption; Loss of active compounds
Supercritical fluid extraction	High diffusivity of supercritical fluids facilitate extraction of bioactive compounds	Short extraction time; Negligible solvent consumption; High yield and purity of obtained products	Difficult to optimize conditions; High capital cost
Pulsed electric field extraction	A non-thermal technique that involves passing a high-voltage electric current through a sample that is positioned between two electrodes for a very short time	Less time and energy consumption; Mostly suitable for thermolabile compounds; High yields for carotenoids extraction	Not suitable for low moisture products due to low conductivity; High capital cost; Limited extraction of lipophilic compounds
Ultrasonic assisted extraction	Utilization of ultrasonic waves to generate cell disintegration and the cavitation effect, allowing for simple solvent penetration	Higher extraction yield or rate; Increase the yield of lipids; Enhancing yield extraction of heat-sensitive components	Scale-up to industrial applications still needs to be explored and optimized; Can lead to degradative processes such as lipid oxidation and hydrolysis

increased extraction yield, decreased solvent usage, faster extraction rates, improved repeatability, simplicity of scaling-up, and higher purity of final products [178]. UAE of Pacific white shrimp cephalothorax increased lipid and carotenoid extraction yield by almost twofold [179]. MAE has been extensively used to extract valuable components. Shrimp cephalothorax was microwave-pretreated, then the output of shrimp oil and carotenoids was increased [180]. Enzyme-assisted extraction can recover lipids and carotenoids, e.g. proteases with the ability to hydrolyse complex molecules and cell membranes, releasing the target substance during the extraction process. Alcalase was used to increase shrimp lipid output as a result of protein hydrolysis of shrimp by-products [181]. A comparison of the advantages and disadvantages of lipids and pigments extraction methods is shown in Table 1 [180,182].

The application of lipids in CBPs

The high bioavailability of lipids makes them ideal for application as novel and beneficial dietary supplement, flavor enhancers, food ingredient and oil supplement [183]. Research shows that the intake of food rich in polyunsaturated acids has a certain proportion to the low mortality rate of cardiovascular complications such as stroke, owing to their capacity to control dyslipidemia, mainly cholesterol, obesity control and anti-inflammatory activities [184]. The fatty acids found in crustaceans also have the characteristics of preventing and treating cognitive impairment, nutrition, and brain development, especially in children and elderly people [185]. They are also widely used as dietary supplements for the treatment of CNS disorders such as Alzheimer's disease, Parkinson's disease's disease and schizophrenia, memory decline, and resistance to oxidative stress [186]. In addition, omega-3 is used to treat cancer because it has significant anti-tumor effects at high concentrations [187].

The application of pigments in CBPs

In our bodies, these carotenoids are regarded as the source of vitamin A [188]. Carotenoids' antioxidant properties make them valuable commercially, as they can be used to enhance health rather than only as food coloring or feed additives [189]. Astaxanthin has been used in cosmetic, food and pharmaceutical industries because of its superior antioxidant activity [170,190]. Particularly, astaxanthin has demonstrated potential for improving human health and for the prevention and treatment of a number of illnesses (anti-cancer, anti-inflammatory, anti-diabetic, anti-lipid peroxidation, cardioprotective, neuroprotective, etc.) [191]. Nowadays, astaxanthin products can be purchased in the market in the following forms: biomass, capsule, cream, energy drink, oil and granulated powder, soft gel, syrup and tablet [167]. As a result, astaxanthin is a high-value product being increasingly sold as a functional food component, with prices ranging from US$3,000 to US$12,000 per kilogram [192]. The demand for it on the international market is rising and is anticipated to reach $2.57 billion by 2025 [193].

Crustaceans' minerals composition

The properties of crustaceans' minerals

Crustaceans shell wastes encompass several minerals including calcium, phosphorus, magnesium, nitrogen etc., which vary in level between the different species and demineralization methods. Different forms of calcium salts can be obtained using different acids under appropriate temperature, time and acid concentration [194]. The predominant minerals in crustacean's shell is calcium carbonate as calcite, amorphous calcium carbonate, and tricalcium phosphate as hydroxyapatite [195]. Calcium carbonate is mainly generated from geological sources, which has the advantage of being a very productive source, but it also has some disadvantages, such as the presence of heavy metals that are difficult to remove. As a result, this biological calcium carbonate source is more acceptable for human consumption. In addition, investigations based on Fourier Transform Infrared Spectroscopy (FTIR) and Thermogravimetric Analysis (TGA) have shown that crab shells contain more calcium carbonate than lobster and shrimp shells [196]. The market price for coarsely ground calcium carbonate is ca. $60–66 per ton, but the value of calcium carbonate processed into ultra-fine particles can reach $14,000.

Extraction methods of minerals

Chemical extraction

Demineralizing the raw material is necessary in order to recover the minerals. Since the carbonate combines with the acidic substance to create the precipitation of minerals, this technique is typically carried out with acids, most commonly HCl. Depending on the reaction variables, such as the temperature, pH, time, and

amount of acids used, the amount of minerals produced through DM by acids ranges from 69.4 to 100%.

Biotechnology extraction

The fermentation of crustaceans' shells typically involves the use of microorganisms like *Lactobacillus sp*, which can produce lactic acid and enzymes. However, just like in the chemical technique, the lactic acid produced by these organisms combines with the calcium carbonate contained in the shells to form calcium lactate, which is easily removed by washing [144]. The quantity, reaction time, species of the inoculated organism, environmental variables including pH and temperature, as well as the presence of carbon sources i.e., glucose being the most prevalent carbon source- all contribute to the efficacy of this procedure [70].

The application of minerals

Minerals of CBPs (mainly calcium carbonate) can be used in the manufacture of pigments, fillers, soil conditioners, rubber, plastics and in the construction, paper, pharmaceutical and agricultural industries [10,197]. Besides, calcium carbonate can be utilized as a terrestrial and aquaculture fertilizer, and its nanoparticles can be used in phytopharmaceuticals or insecticides due to their environmental friendliness, outperforming pyrethroids, organophosphates, and spinosad [198]. Calcium oxide, which is generated from calcium carbonate, acts as a catalyst in the generation of biodiesel [168].

Additionally, phosphates are present in comparatively high concentrations in CBPs. Protein stabilizers, emulsifiers, antioxidants, texture enhancers, and microbiological control agents are a few of the examples that phosphates contribute to food processing [199]. To increase the extraction of bioactive substances, phosphates are utilized as additives [200]. Phosphates are used by the processing industry, primarily in seafood, to alter the structural configuration of tissues. These residues also contain sodium-bound phosphates that are used in medicine, such as monosodium and disodium phosphate that is used as a laxative in drugs.

Conclusions and future directions

The global crustacean processing industry generates large amounts of by-products that are currently discarded or underutilized, resulting in high disposal costs. Meanwhile, many valuable biological resources, such as chitin, proteins and lipids, are wasted, creating an environmental burden. This multifaceted review discusses in a comparative manner the high-value compounds in different crustaceans and their main applications in agriculture, food, water treatment, nutrition, cosmetics, paper, textile, pharmaceuticals and biomedicine, with comparison among different crustaceans for best source of each targeted chemical. In comparison among the three crustaceans, shrimp had the highest content of chitin, lobster had the lowest protein content but the highest lipid content, and crab had the highest astaxanthin content.

Crustacean shell wastes are still underutilized despite their potential value, and more studies are needed before using them in any of the aforementioned applications. The need to commercialize many of these applications must also be supported across many industries with considering of upscaling laboratory procedures to meet industrial needed. In addition, it summarizes various extraction methods such as chemical, biological, physical, solvent, SFE, ISP and PEF for the recovery of various active ingredients highlighting their advantages and limitations to aid future users decide on optimal technologies. The rapid development of biotechnology applications present a fast, effective, clean and controllable bioprocess for the comprehensive utilization of crustacean waste that has yet to be applied at an industrial level. One feasible way for best valorization practices is to combine innovative extraction techniques with industrially applicable technologies to efficiently recover these valuable components from such biowastes.

Thus, the development of simplified processes based on existing extraction methods, combined with the economic recovery of these valuable components using industrially applicable technologies, would be a practical solution to maximize the use of these marine by-products. Using this approach, CBPs could become economically more profitable than the traditional sources of contamination and cost. In conclusion, there is great potential for converting waste/biomass from crustaceans processing into unique bio-based products, which can in turn aid to alleviate environmental pressures and landfill/disposal problems. The blue bio economy will undoubtedly help the crustacean industry move towards a more green economy and a globally sustainable future through the recycling and reuse of these wastes in the feed, food, pharmaceutical and other industries.

Ethical statement

No ethical approval is needed.

CRediT authorship contribution statement

Zuying Zhang: Conceptualization, Data curation, Formal analysis, Writing – original draft. **Zhenmin Ma:** Data curation, Formal analysis. **Lili Song:** Supervision, Funding acquisition. **Mohamed A. Farag:** Supervision, Funding acquisition, Project administration, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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