REVIEW

A review on marine collagen: sources, extraction methods, colloids properties, and food applications

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Abstract

The growing interest in valorizing industrial by-products has led researchers to focus on exploring diferent sources and optimizing collagen extraction conditions over the past decade. While bovine hide, cattle bones, pork, and pig skins remain the most abundant collagen sources, there is a growing trend in the industrial utilization of collagen from non-mammalian species. This review explores alternative marine collagen sources and summarizes emerging trends in collagen recovery from marine sources, with a particular focus on environmentally friendly methods. Additionally, this review covers the colloidal structure-forming properties of marine collagens, including foam, flm, gel, and emulsion formation. It also highlights the potential and important applications of marine collagen in various food products. Based on the currently reported marine sources, collagens extracted from fsh, jellyfsh, and sea cucumbers were found to have the highest yield and mostly comprised type-I collagen, while crustaceans and mollusks yielded lower percentages of collagen. Traditional extraction techniques isolate collagen based on acetic acid and pepsin treatment, but they come with drawbacks such as being time-consuming, causing sample destruction, and using solvents. Conversely, marine collagen extracted using conventional methods assisted with ultrasonication resulted in higher yields and strengthened the triple-stranded helical structures. Recently, an increasing number of new applications have been found in the food industry for marine collagens, such as biodegradable flm-forming materials, colloid stabilizers, foaming agents, and micro-encapsulating agents. Furthermore, collagen is a modern foodstuf and is extensively used in the beverage, dairy, and meat industries to increase the stability, consistency, and elasticity of products.

Keywords Marine collagen, Jellyfsh, Sea cucumber, Colloid stabilizer, Edible flm, Dairy product

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1 Introduction

Collagen, a group of structural proteins in the extracellular matrix with a fbrillar arrangement, contributes to the integrity and mechanics of bio-tissues [[1\]](#page-22-0). Found ubiquitously in living organisms, collagen maintains conserved forms in gene and amino acid sequences, notably in the triple-helix structure. Collagen can also be very abundant, especially in mammals, constituting up to 30% of total proteins and serving as vital components in tissues such as skin, bone, and cartilage [[2\]](#page-22-1). Currently, 29

types of collagen have been identifed, each difering in bio-physiological properties, morphological structure, amino acid sequence, and distribution [\[1](#page-22-0)]. Among the 29 identifed collagen types, type I is the most common, characterized by a triple-helix structure $[2]$ $[2]$. The collagen molecule, called tropocollagen, is structurally organized in a lengthy amino acid sequence, consisting of three domains, *i.e*., –COOH non-triple-helix terminal (C-terminal), triple-helix, and $-NH₂$ non-triple-helix terminal (N-terminal), primarily stabilized by hydrogen bonds [[3\]](#page-22-2). These \sim 300 nm long to \sim 1.5 nm diameter collagen molecules self-assemble into collagen fbrils (diameter \sim 100 nm), and thousands (\sim 4000) of these fibrils can intertwine to create collagen fbers, which further pack and crosslink to generate diverse hierarchical and robust bulk collagen materials (Fig. [1\)](#page-2-0) $[3]$ $[3]$. The hydrogen and covalent bonds stabilizing the collagen triple helix are broken during the extraction process, resulting in a polypeptide mixture called gelatin, *i.e*., collagen partially degrades to form gelatin. Gelatin, produced via partial hydrolysis of collagen, has molecular weights ranging from 15 to 250 kDa $[4]$ $[4]$ $[4]$. Two types of gelatin, type A and B, can be obtained based on pre-treatment methods like alkaline or acid conditions [\[5\]](#page-22-4). Enzyme pre-treatment targeting specifc labile peptide bonds is also involved in gelatin production. The final product comprises polypeptides of varied conformations and sizes due to diverse pre-treatment and extraction procedures. It may include higher molecular weight fractions, such as β-chains (covalently linked α-chains dimers), γ-chains (covalently linked α-chain trimmers), and microgels with higher orders [[5\]](#page-22-4). Despite its denatured state, gelatin retains the same amino acid composition as collagen [[4\]](#page-22-3).

Collagen derived from land-based animals, such as pigs, cattles, and chickens, fnds extensive applications in tissue repair, medical devices, pharmaceuticals, and the food industry. Despite its widespread use, challenges arise, including potential immune reactions, limited availability, high costs, and substantial land requirements for land-based animals [[6\]](#page-22-5). Concerns about infectious diseases, like transmissible spongiform encephalopathy, bovine spongiform encephalopathy, foot and mouth disease, have limited the use of collagen from these sources. Religious prohibitions also impact a substantial global population. Conversely, marine collagen exhibits physicochemical characteristics similar to mammalian collagen, with distinct advantages such as a lower denaturation temperature, lower denaturation weight, reduced risk of disease transmission, simpler extraction methods, and minimal infammatory responses [[7\]](#page-22-6). Marine collagen molecules consist of three α -chains (α 1- α 1- α 2) of about 1000 amino acid residues or multi-peptide α-chains, forming a stable triple helix structure arranged vertically and bilaterally in a periodic fiber structure. The high stability of the characteristic triple helix region in marine collagen mirrors its similarity in quaternary structure

Fig. 1 Multi-hierarchical structure of type I collagen

and essential amino acid composition to terrestrial mammalian collagen [\[8\]](#page-22-7). Despite this similarity, the marine environment's higher complexity and diversity result in varied marine collagen structures infuenced by species, origin, growth cycle, season, environment, and other factors. As a consequence, there are subtle diferences in the structure and composition of marine collagen compared to collagen from terrestrial animals.

The global demand for functional proteins is projected to reach \$7.98 billion by 2026, experiencing a 6.93% annual growth rate from 2019 to 2026 [\[7](#page-22-6)]. Collagen, a crucial macromolecule derived from marine vertebrates and invertebrates, is widely utilized in the pharmaceuticals, food, and healthcare industries. The increased demand for collagen and its hydrolyzed gelatin is evident in various sectors, including food packaging, beverages, dairy products, meat-based items, drug delivery systems, and food supplements. Recent data indicates a 9.4% growth in the global collagen market from 2015 to 2023, with a projected market value of \$9.37 billion by 2023, up from \$4.13 billion in 2014, primarily sourced from marine origins [[1,](#page-22-0) [9](#page-22-8)].

According to current research reports, collagen extraction and separation methods from various marine sources involve acidic, enzymatic, ultrasound-assisted, and physical-aided techniques [\[10](#page-22-9)[–12](#page-22-10)]. Researchers have also introduced environmentally friendly processes, such as extrusion-hydro-extraction [[13](#page-22-11)], supercritical fuid extraction [[14](#page-22-12)] and deep eutectic solvent extraction [[15](#page-22-13)] to enhance yield and reduce pollution. The distribution of collagen fbers, binding tightness, and cross-linking degree with other components impact separation difficulty, extraction rate, purity, and structural integrity $[16, 16]$ $[16, 16]$ [17\]](#page-22-15).

To date, no review article has comprehensively summarized and discussed the diverse marine collagen sources, including sea cucumbers, mollusks, marine sponges, fsh, jellyfsh, and crustaceans. Addressing this gap, the current review provides a comprehensive exploration of prevalent novel marine collagen sources, established and developing methods employed in their extraction, and the associated challenges. Additionally, this review aims to provide an updated and detailed overview of the colloidal structure-forming properties of marine collagens, such as gels, flms, foams, and emulsions. Subsequently, the paper delves into various applications of marine collagen, encompassing food packaging materials, beverages, dairy products, supplements, and meat-based items.

2 Marine collagen sources

Collagens from various marine sources, including, sea cucumbers, mollusks, sponges, crustaceans, jellyfsh, and particularly fsh, have been extracted and characterized to varying extents. These sources exhibit significant variations in their physicochemical properties (Table [1](#page-4-0)).

2.1 Sea cucumbers

Sea cucumbers, classifed as invertebrates in the *Holothuroidea* class within the phylum *Echinodermata*, are traditional food sources in Korea, Japan, China, and parts of Southeast Asia [\[47](#page-23-0)]. Often referred to as gamat or bêche-de-mer, these marine invertebrates comprise over 1250 species globally, many of which are not only edible but also possess signifcant nutritional and bioactive functions $[48]$ $[48]$. The nutritional content and chemical composition of sea cucumbers may vary based on the species and growing environment. For example, *Holothuria arenicola* sea cucumbers from the Bndar-e-lengeh coast in southern Iran showed protein, moisture, ash, crude fber, and fat percentages of 24.4%, 69.5%, 10.9%, 2.29%, and 2.9%, respectively, while *Holothuria parva* sea cucumbers had percentages of 17.6%, 67.8%, 32.7%, 1.97%, and 2.4% [\[49\]](#page-23-2).

The primary edible part of the sea cucumber is its body wall, where collagen constitutes approximately 70% of total protein, plays a crucial role $[47]$ $[47]$. The sea cucumber body wall is a unique mutable collagenous tissue, fabricated from basic structural components such as collagen, proteoglycan and glycoprotein. These elements assemble into collagen fbrils, microfbrils, and collagen fbers, with insoluble collagen fbrils dominating the majority of the total body wall proteins [[48\]](#page-23-1). Collagen fbers, encircled by a microfbrillar network, contribute to organizational stability and provide a long-range restoring force. Senadheera, Dave [[3\]](#page-22-2) investigated the structures and biomechanics of collagen's microfbrillar network derived from the sea cucumber (*Cucumaria frondosa*), observing morphological characteristics similar to vertebrate fbrillin microfbrils.

The collagen in sea cucumbers is characterized by symmetrically spindle-shaped and short collagen fbrils in echinoderms. Molecularly, these fbrils are bipolar and associate with surface-bound proteoglycans. Internal covalent crosslinks, which are similar to mammalian collagen, contribute to collagen stabilization. Solubilized collagen from the body wall of the sea cucumber exhibits a distinctive triple helix structure built by three homologous $α1$ chains as $(α1)3$ and is notably rich in glutamic acid [\[50](#page-23-3)]. Tian, Xue [\[47](#page-23-0)] employed bioinformatic methods and proteomic techniques to study the constituents within sea cucumber collagen. Phylogenetic analyses of collagen sequences indicated that these sequences did not align with typical collagen branches. The authors established that the complex and heterogeneous nature of sea cucumber collagen warrants thorough investigations.

Table 1 Physicochemical properties of collagens derived from marine sources

Table 1 (continued)

Sea cucumbers predominantly contain type I collagen, playing a crucial role in the food quality, particularly the textural properties, of sea cucumbers and their processed products (*e.g*., microwave-dried, freeze-dried, salt-dried, boiled-dried, and ready-to-eat products). Previous studies, including *Acaudina leucoprocta* [[32\]](#page-23-6), *Stichopus monotuberculatus* [\[51](#page-23-19)], *Stichopus japonicus* [\[50\]](#page-23-3), *Holothuria parva* [[35\]](#page-23-9), *Stichopus vastus* [[33\]](#page-23-7), and *Parastichopus californicus* [[31\]](#page-23-5), focusing on sea cucumber autolysis and processing activities such as soaking, boiling, drying, and rehydration attribute changes in food properties to sea cucumber collagen. In addition, rheological properties of sea cucumber collagen, indicative of its molecular structure and chain conformation, signifcantly impact the physical characteristics of sea cucumber products. Liu, Oliveira [[31\]](#page-23-5) found that the reduced rheological properties of collagen from the giant red sea cucumber (*Parastichopus californicus*) compared to calf skin collagen, was ascribed to a lower imino acid residue content in both connective tissue and skin (15.3% and 14.2%, respectively). These values were even lower than those observed in cod or walleye pollock. Additionally, sea cucumber collagen exhibited lower thermal stability than terrestrial animal collagen. For example, the calculated T_d values for collagen from the connective tissue and skin of the giant red sea cucumber were 17.9 and 18.5 °C [\[31](#page-23-5)], respectively, markedly lower by about 19.1 and 18.5 °C than the T_d value of porcine skin collagen (T_d=37.0 °C) [[47\]](#page-23-0).

2.2 Crustaceans

Crustaceans, which belong to the Phylum *Arthropoda*, encompass a varied range of marine invertebrates, such as shrimp, mantis shrimp, lobsters, crabs, prawns, krills, crayfsh, ostracods, and copepods. Crustaceans are globally produced in large quantities, with an annual value of over \$57 billion, equating to 23% of the total global aquaculture market. Notably, according to the 2021 China Fishery Yearbook, China alone recorded a global shrimp production of approximately 2.69 million tons in 2021 [\[52](#page-23-20)]. Several large crustaceans, such as lobsters, shrimp, crabs, and prawns, are consumed as food due to their excellent nutritional characteristics, including high unsaturated fatty acids, protein, and essential microelements (*e.g.*, Mg²⁺, Ca²⁺, Zn²⁺) [[52\]](#page-23-20). Additionally, the processing of crustacean seafood (crabs, lobsters, shrimp, and prawns) generates substantial waste in the form of carapace and head [\[40](#page-23-14)]. Approximately 60–85% of lobster, 60–70% of crab, 65–70% of crayfsh, 70–75% of krill, and 60–80% of shrimp are discarded during processing $[53]$ $[53]$. These by-products can be separated into their chemical components: 15–40% chitin, 20–40% protein, and 20–50% calcium carbonate [[54](#page-23-22)].

Crustacean muscles primarily consist of myofbrillar proteins, with sarcoplasmic proteins also present, while collagen constitutes only a small fraction of the total protein content [[55\]](#page-23-23). For instance, Hiransuchalert, Oonwiset [[41\]](#page-23-15) isolated collagen from the muscles of four diferent mantis shrimp species: *Odontodactylus cultrifer, Erugosquilla woodmasoni, Harpiosquilla harpax,* and *Miyakella nepa*. The collagen exhibited type I characteristics similar to vertebrate muscles, despite low content percentages ranging from 0.015% to 0.4878%. Noteworthy collagen content variations may exist among diferent crustaceans along with diferences in collagen types. Li, Han [[56\]](#page-23-24) found that the collagen concentration in the skeletal muscles of three crustacean species (spiny lobster, feshy prawn, and giant river prawn) ranged from 2.4 to 2.6% of the total protein. In muscle tissues, collagen content showed variations, such as 5.9–6.2% in squilla (*quilla Oratosquilla oratoria*), 3.4–3.6% in crayfsh (*Procambarus clarkia*), 2.6–2.9% in prawn (*Penaeus japonicas*),

2.5–2.7% in lobster (*Panulirus Iongipes*), and 1.1–2.2% in the shrimp (*Pandalus borealis*). For thoracic and pereiopod muscles of four crab species (*Chionoecetes opilio, Erimacrus isenbeckii, Portunus trituberculatus,* and *Charybdis japonica*), the collagen content varied from 0.2 to 0.8% [\[57\]](#page-23-27).

2.3 Marine sponges

Marine sponges or poriferans, belonging to the phylum *Porifera*, constitute a diverse group of flter-feeding benthic invertebrates, predominantly inhabiting saltwater but occasionally found in freshwater (around 220 species) [[58](#page-23-28)]. Unlike other animal groups, sponges exhibit a simple organizational structure devoid of real organs and tissues. They are composed of gelatinous internal tissue (mesohyl) surrounded by a layer of epithelial cells, including pinacocytes and choanocytes. These cells are integrated into a complex 3-D matrix network rich in collagen, commonly referred to as spongin or spongin-like collagen [[45\]](#page-23-25). Spongin, a collagenous protein found in the exoskeleton of certain sponges, forms a complex fbrous network that provides fexural rigidity to the sponge. However, its precise and complete molecular composition remains unclear due to the extensive diversity within this sponge family, posing a signifcant challenge for future elucidation [\[58\]](#page-23-28). For example, Araújo, de Souza [[44\]](#page-23-18) reported a fbrillar structure in spongin-like collagen from *Chondrilla caribensis*, contrasting with the nodular/ particulate aggregate structure observed in the sponginlike collagen from *Aplysina fulva*. Tziveleka, Ioannou [[46](#page-23-26)] suggested that collagen extracted from the demosponges *Suberites carnosus* and *Axinella cannabina* can be differentiated based on various characteristics, including thermal behavior, solubility, amino acid composition, isoelectric point, and microscopic observations. The study's fndings indicated that *Axinella cannabina* had intercellular collagen, insoluble collagen, and spongin-like collagen content of 3.0%, 12.6%, and 42.8%, respectively, while for *Suberites carnosus*, they were 1.9%, 5.0%, and 21.9%.

Despite the identifcation of around 15,000 sponge species, only a few dozen have been explored for collagen extraction and characterization, highlighting the largely untapped biotechnological potential of marine sponges [[58\]](#page-23-28). While sponge collagen holds promise due to its unique physicochemical properties, its industrial/largescale production faces two key obstacles: (1) spongederived collagen typically exhibits lower thermal stability compared to that of homeothermic mammals and birds, given the poikilothermic nature of sponges living in lower temperatures than the human body $[46]$ $[46]$. (2) Despite the growing demand for sponge-derived natural products, there is a scarcity of sustainable and economically viable culture methods to generate substantial and stable sponge biomasses [\[58](#page-23-28)].

2.4 Jellyfsh

Jellyfish, also known as 'medusae,' have been consumed as a traditional food source in numerous Asian countries, notably in Japan and China, for over 1700 years due to their nutritional and pharmacological value [\[59](#page-23-29)]. Global jellyfsh productions now surpass 800,000 tons/ year, exceeding catches of popular seafood such as lobsters, clams, and mussels. Despite the existence of over 1400 jellyfsh species worldwide, only 23 species have been extensively explored as a sustainable, collagenrich food source [\[59\]](#page-23-29). With a richness in collagenous protein and minerals, jellyfsh's low-fat and low-calorie content makes them as an ideal, healthy seafood commodity. De Rinaldis, Leone [\[60](#page-23-30)] highlighted jellyfsh as an untapped marine collagen resource. Jellyfsh collagen exhibits potential advantages over terrestrial animal collagens, including lower infammatory and immunogenic responses, as well as fewer biological toxins and contaminants [[59](#page-23-29)]. León-Campos, Claudio-Rizo [[61](#page-23-31)] reported the high biocompatibility, low potential for transmitting zoonotic diseases to humans, and low allergic response (requiring caution for consumers with fsh allergies) associated with jellyfish collagen. The invertebrate nature of jellyfsh suggests the production of unique and commercially appealing products with novel physicochemical and functional properties.

Fresh jellyfsh exhibit a proximate composition that differs slightly among species, primarily comprising minerals (∼1–2%), moisture (∼95–97%), and crude protein (∼1–3%) with a signifcant portion of the protein up to 75% being collagen $[62]$ $[62]$. The types of jellyfish collagen, akin to other marine and mammalian collagens, vary across species, and no two species share identical collagen makeup. Commonly, types I and II collagen are identifed, with occasional examples of types III, IV, and V. Contrary to conventional collagen-type nomenclature, Smith, Domingos [\[63](#page-23-33)] suggested that jellyfsh collagen, particularly from invertebrates, deviates due to negligible calcifed tissues, a high collagen-to-insoluble extracts ratio, and similarities in function and morphology to mammalian collagen. Hoyer et al. (2014) reported that the jellyfsh collagen in S. *meleagris* shares similarities with vertebrate collagen type II, as indicated by solubility properties, salting-out concentration, molecular mobility, a high hydroxylysine content, a highly hygroscopic nature, and the absence of disulfde bonds. Despite low sequence similarities to vertebrate collagen, invertebrate collagen from diferent jellyfsh species can exhibit unique physical and functional properties [[63\]](#page-23-33).

2.5 Mollusca

Mollusca, a diverse phylum containing over 120,000 species such as the squids (*Doryteuthis singhalensis*), mussels (*Mytilus chilensis*), scallops (Patinopecten yessoensis), oysters (*Crassostrea gigas*), and clams (*Meretrix meretrix*), displays significant morphological, ecological, and chemical diversity [[10\]](#page-22-9). Distributed across tropical seas, temperate waters, and polar regions, mollusk species can be diferentiated based on the size and shape of their bodies. Mollusks are protein-rich, with 80% of their fleshy material suitable for human consumption $[64]$ $[64]$ $[64]$. The edible portions of mollusks are characterized by high water content, while their dry mass consists of proteins and micro/macro minerals, but is low in fats. For example, Wu, Guo [\[42](#page-23-16)] found that the surf clam shell (*Coelomactra antiquata*) had moisture, protein, carbohydrate, and ash contents of 82.46%, 11.56%, 3.05%, and 2.38%, respectively, with a fat content of only 0.55%. Certain parts of mollusks have received special attention due to their successful use in collagen extraction. For example, the muscle and mantle of the bivalve mollusk *Anadara broughtonii* contain 6.8% and 7.2% collagen, respectively. Similarly, *Mactra chinensis* showed 7.1% and 7.9% collagen content, as highlighted by $[64]$.

Studies have shown that the bio-physiological properties, morphological structure, and molecular composition of mollusk collagen have not been fully elucidated owing to the great diversity within this family of mollusks. Vallejos, González [\[10](#page-22-9)] provided the frst comprehensive proteomic description under industrial conditions of the Chilean mussel (*Mytilus chilensis*), a species recognized for its favorable characteristics in collagen production. They observed high concentrations of proline, hydroxyproline, and glycine in Chilean mussels collagen, characterized by $β$ and $γ$ bands, indicating signifcant cross-linking in the telopeptide region. Multiple studies have reported the potential availability of type I collagen in cephalopods [[65\]](#page-23-35), squid [[65](#page-23-35)], clam Shell [\[42](#page-23-16)], and byssus [[10](#page-22-9)], indicating their suitability as potential raw material sources for food applications. Veeruraj, Arumugam [\[65](#page-23-35)] identifed type I collagen in squid (*Doryteuthis singhalensis*) with α1 and α2 chains, showing thermal denaturation temperatures of 34.80–35.70 °C and a glycine residue range of 32.8–33.2%. This highlights the potential of squid skin collagen as a thermostable alternative for commercial use.

2.6 Fish

Fish is globally recognized as a nutritious and soughtafter food. Currently, over 33,600 fsh species have been identifed and classifed into four groups based on their habitats: ice-water, cold-water, warm-water, and hot-water. During the production of fish meat, various byproducts like scales, bones, cartilages, and skin are often discarded as waste (about 7.3 million tons/year) by fsh processing industries, contributing to environmental pollution $[66]$ $[66]$. Approximately 30% of fish bone comprises organic collagen, with 60–70% constituting inorganic substances, mainly calcium phosphate and hydroxyapatite [[66](#page-23-36)]. Fish collagen has specifc amino acid compositions, including low concentrations of hydroxyproline, proline, and glycine, resulting in a lower denaturing temperature compared to mammalian collagen. According to Caruso [[7](#page-22-6)], fsh collagens from skin, bone, cartilage, and scales surpass porcine or bovine collagen in bioavailability, demonstrating greater absorption efficiency (up to 1.5) times) and quicker bloodstream circulation due to their low molecular weight and small particle size.

Fish collagen's structural characteristics have been delineated via amino acid analysis and physicochemical characterization. Research on the imino acids of collagen from various fsh species indicates substantial diferences in terms of proline and hydroxyproline imino rings, imparting conformational stability and imposing rigid constraints on rotational movement along the N-Cα bond in the backbone. Cruz-López, Rodríguez-Morales [[6\]](#page-22-5) reported that the imino acid content in the skin of gulf corvina (*Cynoscion othonopterus*) (16.8%) was lower than the skin of giant croaker (19.1%) [[67](#page-23-37)], but similar to that from pacifc cod (*Gadus macrocephalus*) (15.9%) [[68\]](#page-23-38), bighead carp $(16.5%)$ [[24\]](#page-22-22), and grass carp $(16.6%)$ [[69\]](#page-24-0). Differences between imino acid profiles and structures are probably due to factors such as fsh species, tissues, origin, habitats, growth cycle, extraction methods, and other factors [[69\]](#page-24-0).

The thermal stability of fish collagen can vary based on the relative abundance of imino acids (proline and hydroxyproline), which closely tied to the species' living conditions and body temperature [[68](#page-23-38)]. Fish collagen, infuenced by colder habitats, typically displays lower imino acid levels and thermal stability compared to collagen from warm-blooded land animals. For instance, chicken sternal cartilage collagen exhibited a higher imino acid content (23.2%) and a greater denaturation temperature (32.8 °C) than shark cartilage collagen 15.6% and 16.8 °C, respectively [[70](#page-24-1), [71\]](#page-24-2). Despite similar imino acid levels to land animals, sturgeon collagen displays a lower denaturation temperature. However, the challenges lie in the low denaturation temperature (25– 30 °C for most fsh species) and diverse composition of fsh collagen, hindering its applications. While strict comparisons are challenging due to methodological differences across studies, the denaturation temperatures of collagens in deep-sea redfsh, inhabiting an ocean temperature of 3–8 °C, resembled those of cold-water fsh like chum salmon (19.4 °C), Argentine hake (10 °C),

Baltic cod (15 °C), and Alaska Pollack (16.8 °C) [[24,](#page-22-22) [68](#page-23-38), [69,](#page-24-0) [71\]](#page-24-2). Notably, these temperatures were substantially lower than those observed in temperate and tropical fish species, including common Nile perch (36.5 °C) , ayu (29.7 °C), Japanese seabass (26.5 °C), skipjack tuna (29.7 °C), eel (29.3 °C), and mackerel (26.1 °C) [\[72\]](#page-24-3).

3 Extraction methods

Due to collagen's robust structure and its low solubility in water, two key steps are involved for extraction: (a) pretreatment of raw materials; and (b) collagen extraction. Pretreatment phase involves cleaning, dehydration, degreasing, and decalcifcation of the raw material. Four commonly employed degreasing methods include combined salt solution, enzymatic digestion and lipase degreasing, degreasing solution usage, and organic solution extraction [\[23](#page-22-21)]. Non-collagenous proteins, fats, and pigments may be present in some samples, and they are typically removed at this stage using NaOH, alcohols (ethanol or butyl-alcohol), and oxygen peroxide. Additionally, demineralization of the raw material with HCl or EDTA before extraction enhances the efficiency of collagen extraction from mineral-rich body parts, such as cartilage [[3\]](#page-22-2). Diferent methods designed for collagen extraction from marine sources are discussed below:

3.1 Acidic extraction

Collagen proteins, predominantly types I, II, III, and V, are fbrous and exhibit lower solubility in aqueous mediums than in acidic mediums, and therefore, are typically extracted through acidic treatments following the removal or degradation of noncollagenous molecules $[23]$ $[23]$. The mechanism involves enhanced repulsive forces between tropocollagen molecules under acidic conditions, leading to the solubilisation of less cross-linked collagens, resulting in acid-soluble collagen. This process breaks down various amino acids within the sample, achieving the dissolution of non-covalent intramolecular and intermolecular bonds through non-selective chemical hydrolysis of collagen chains. Typically, 0.5 M acetic acid is commonly employed because it attains the highest yield (90%), while diluted organic (chloroacetic, citric and lactic) and inorganic (hydrochloric) acids result in a relatively low yield $(\leq 20\%)$ [\[22\]](#page-22-20). These acidic solutions offer a notable advantage by efectively disrupting Schif bases and ionic bonds between molecules at low acidic concentrations $[22, 23]$ $[22, 23]$ $[22, 23]$ $[22, 23]$ $[22, 23]$. The extractability of collagens depends on marine age and tissue type, with extraction factors including temperature, treatment duration, acid concentration, and the ratio of acidic solution to raw material [[73\]](#page-24-4). The key drawback of this extraction method is that it is time-consuming (Fig. [2](#page-10-0)), requiring 2–4 days and has a relatively low yield [[23](#page-22-21)].

The acidic extraction solution, ranging from 0.5 to 1 M , facilitates cleavage of inter- and intramolecular crosslinks without compromising the structure of collagen chains. Arumugam, Sharma [\[74](#page-24-5)] investigated the impact of acetic acid concentration (0.2–1 M) on the extraction of collagen from sole fsh skin, keeping other variables constant. The collagen yield exhibited a gradual increase with acetic acid concentration, reaching a peak at 0.54 M with a maximum yield of 19.27 mg/g. However, concentrations beyond 0.6 M resulted in a decrease in collagen yield. In a study on Baltic cod *(Gadus morhua*) skin, Skierka and Sadowska [[16\]](#page-22-14) reported a higher recovery of collagen using acetic acid (90%), followed by citric acid (60%) and hydrochloric acid (18%). Conversely, Tan and Chang [[73\]](#page-24-4) found that HCl-assisted extraction produced the highest collagen yield (42.36%), followed by acetic acid extraction (39.45%). The discrepancy may result from varying pH values in the two studies; the former at pH 2.4 and the latter at pH 0.87. Under lower pH conditions, amine groups bond with anions (Cl−), decreasing electrostatic repulsion between charged groups. This leads to tightened collagen fbers, diminishing their water bonding ability and reducing collagen solubility. Additionally, despite equal total H concentration, the ionized H^+ in the solutions differed.

3.2 Enzymatic extraction

Traditional collagen extraction involves acidic solutions, but collagens from diferent sources may not fully dissolve in these mediums, resulting in the retention of intermolecular crosslinks [[75\]](#page-24-6). To enhance collagen yield, enzymatic extraction methods prefer the use of enzymes like pepsin, papain, trypsin, and various collagenases under specifc environmental conditions and pH levels [[67\]](#page-23-37). Pepsin, widely used for seafood collagen extraction, is employed alone or with varying acetic acid concentrations, resulting in pepsin-soluble collagens [\[71](#page-24-2)]. Enzymatic extraction is preferred for increased efficiency, reduced collagen antigenicity, and preservation of the collagen triple helical structures. Maintaining a low temperature (4–10 °C) during pepsin use is crucial due to its sensitivity to higher temperatures (above 60 °C), risking self-digestion and deactivation [[24\]](#page-22-22).

Using pepsin has been found to increase extraction yields and decrease the duration of collagen extraction from giant croaker (*Nibea japonica*) [[67\]](#page-23-37). The results showed that the extraction yield increased from 66.35 to 79.93% by increasing the pepsin level from 800 to 1200 U/g, and triple-helical structure of collagen was largely intact through hydrogen bonding, suggesting insignifcant occurrence of protein breakdown. Most recently, attention has been focused on the use of protease extracted from proteolytic bacteria. For instance,

Fig. 2 The advantages and disadvantages of the diferent extraction methods

Ahmed, Getachew [[76\]](#page-24-7) reported that proteases isolated from *Bacillus cereus* (FRCY9-2) and *Bacillus cereus* (FORC005) were employed in extracting collagen from bigeye tuna (*Tunnus obesus*) skin, resulting in yields of 177.2 and 188 g/kg, respectively, compared to using acid alone (134.5 g/kg).

3.3 Deep eutectic solvent extraction

Deep eutectic solvents, defned as mixtures of two or more components showing a signifcant melting point depression at a specifc composition, transform into liquids at room temperature. These solvents, formed through the interaction between a hydrogen-bond acceptor and a hydrogen-bond donor, have gained prominence as green and sustainable alternatives to conventional industrial processes $[15]$ $[15]$. The complexing agent, usually a hydrogen-bond donor, interacts with the halide anion, increasing its efective size and reducing the anion interaction with the cation, resulting in mixture melting points far below those of individual components.

Particular interest arises when these deep eutectic solvents are composed of natural components (urea, oxalic acid, ethylene glycol, and choline chloride), making the method particularly suitable for extracting volatile aromatic and phenolic compounds, metals, and collagen proteins [[77\]](#page-24-8). Notably, they offer advantages such as costefective, numerous combinations, biodegradability and low toxicity [[77](#page-24-8)].

Batista, Fernández [[15](#page-22-13)] introduced a green and sustainable collagen extraction methodology from blue shark by means of a deep eutectic solvent comprising xylitol:citric acid:water at a 1:1:10 molar ratio. This approach resulted in isolating over 21% of the protein content from blue shark skin, surpassing conventional methods by 2.5 times, without the need for raw material pre-treatment and decreasing the procedure time from 96 to 1 h. Among the six deep eutectic solvents studied by Bai, Wei [[78\]](#page-24-9), the choline chloride and oxalic acid combination demonstrated remarkable efficacy, achieving an extraction efficiency 90% for cod skins. Furthermore, elevating

the oxalic acid quantity resulted in a corresponding increase in free protons within the solution, which, in turn, enhanced interactions with collagen helices, thereby facilitating the leaching of peptides.

3.4 Supercritical fuid extraction

Supercritical fuid extraction has emerged an alternative to traditional solvent extraction by utilizing supercritical fuids, generally carbon dioxide, to extract bioactive compounds from various marine sources, such as microalgae, brown algae, and green seaweed. These fluids, existing above critical pressure and temperature, demonstrate gas-like compressibility, lower density, and potent solvating capabilities similar to liquids. Their high diffusivity facilitates easy penetration into solid materials, enabling dissolution and accessibility [[79](#page-24-10)]. Supercritical fluid extraction offers various advantages over traditional extraction methods, including increased extraction yields, enhanced selectivity, improved fractionation capabilities, and a reduced environmental impact.

Silva, Barros [[14\]](#page-22-12) explored the application of supercritical fuid extraction at pressures of 10, 30, and 50 bar for 3 h, using CO_2 -acidified water for collagen extraction from demosponge (*Chondrosia reniformis*). The study demonstrated a 30% increase in collagen recovery at 10 bar pressure compared to traditional enzymatic/ acidic methods. Similarly, a study by Sousa, Martins [[79](#page-24-10)] utilized supercritical fuid extraction for collagen extraction from Atlantic cod (*Gadus morhua*) skin. The extraction, conducted with CO_2 -acidified water at 37 °C and 50 bar for 3 h, resulted in a 13.8% collagen yield. Notably, this method proved significantly more efficient than the conventional approach, which requires nearly 200 h to extract only 10.9% collagen from the same fsh species.

3.5 Extrusion‑hydro‑extraction

Collagen in fsh scales is tightly bound to hydroxyapatite, posing challenges in separation. The extrusion–hydro extraction process utilizes extrusion to break the strong linkage between collagen and hydroxyapatite, easing collagen release through water extraction from fsh scales [13]. This method provides benefits such as continuous production, minimal labor, reduced waste, high yield, straightforward operation, and versatility in product outcomes. Huang, Kuo [[13](#page-22-11)] introduced a novel extrusion–hydro-extraction method for collagen extraction from tilapia fish scales. Their findings demonstrated that the high pressures reached during extrusion resulted in collagen yields ranging from 7.5 to 12.3%, which was 2–3 times higher compared to non-extruded scale samples. Moreover, the amino acid profles, and moisture absorption and retention properties of collagens obtained through this process closely resembled those of collagen isolated by conventional methods.

3.6 Physical‑aided extraction

Physical-aided collagen extraction methods contribute to enhanced solubilization and tissue homogeneity, significantly improving the efficiency and yield of collagen extraction within a limited timeframe compared to traditional acidic and enzymatic methods [\[79](#page-24-10)]. Khong et al. $[9]$ $[9]$ reported a new technique to isolate collagen from marine sources, combining acidic treatment with a sequence of mechanical and physical processes, including adjusting pH, mixing, sonication as well as homogenization. Khong, Yusoff $[11]$ $[11]$ $[11]$ employed these techniques to extract collagen from jellyfsh (*Acromitus hardenbergi*) oral arms and bells. The physical-aided processes led to a remarkable increase in extraction efficiency, with nearly a fve times improvement from oral arms and a seven times enhancement from bells compared to acid-assisted extraction. Additionally, there was a two times increase compared to pepsin-assisted extraction. Kuwahara [[80\]](#page-24-11) suggested that the combination of acetic acid and ultrafne bubbles of various gases (ozone, carbon dioxide, and oxygen) enhances both the yield and quality of collagen extraction from tilapia scales. The most effective method was using $CO₂$ ultrafine bubbles in a 0.1 M acetic acid solution, resulting in a noteworthy 1.58% collagen yield after 5 h of aeration. Araújo, de Souza [\[44](#page-23-18)] introduced a specifc procedure for extracting spongin-like collagen from marine sponges (*Chondrosia reniformis*), involving a mildly basic solution with a chaotropic agent. The procedure included treating sponges with a 100 mM Tris–HCl bufer (8 M urea, 100 mM 2-mercaptoethanol, 10 mM EDTA, and pH 9) for 24 h at room temperature. Subsequently, sponge collagens were solubilized, separated through centrifugation, and further precipitated by decreasing the pH to 4 with the addition of acetic acid, then freeze-dried for preservation of the collagen.

Ultrasonic extraction, utilizing high-intensity sound waves beyond the human hearing limit (20 kHz– 10 MHz), has gained signifcant interest in the food industry owing to its non-toxic, eco-friendly, and cost-efective nature $[81]$ $[81]$. This method relies on high-frequency sound waves creating low- and high-pressure regions, disrupting cell walls, and boosting acidic and enzymatic treatment, thereby substantially decreasing extraction time compared to conventional methods $[81]$. The technique enhances extraction efficiency at lower temperatures, aiding in extracting temperature-sensitive proteins with higher yield and minimal damage. Researchers optimized ultrasound efectiveness by adjusting its amplitude, frequency, propagation cycle (discontinuous or continuous), device nominal power, and system geometry, including

probe dimensions. Ultrasonic treatment at 200–750 W, amplitude 20–100%, 20–35 kHz, and pulsation 2/2–20/20 s takes about 10–30 min or even 0–24 h. For instance, 300 W ultrasound power with 25 min exposure improved the collagen recovery rate from yellowfn tuna skin to 57.06% [[82\]](#page-24-13).

Ali, Kishimura $[83]$ $[83]$ investigated the efficiency of ultrasonication in extracting pepsin- and acid-soluble collagens from golden carp (*Probarbus Jullieni*). The study's fndings revealed that applying ultrasonication at 80% amplitude significantly improved the extraction efficiency, resulting in yields of 81.53% for pepsin-soluble collagen and 94.88% for acid-soluble collagen from the golden carp skin, compared to conventional method yields of 51.90% and 79.27%, respectively. The study suggested that ultrasonication strengthened the triplestranded helical structures through interchain hydrogen bonding. Kim, Kim [\[84\]](#page-24-15) reported the improved collagen yield through ultrasonication from sea bass skin (*Lateolabrax japonicus*) compared to traditional extraction, with no alteration in collagen components.

Despite its promise for yield improvement, highintensity ultrasound might be destructive. After applying ultrasonication with an intensity of 11.35 kW \times cm^{−2} for 36 min, Kim et al. [[77\]](#page-24-8) found that the secondary structure of the collagen was disordered. Additionally, long-term ultrasonication apparently led to destroy α-chain in collagen from sea bass skin. Petcharat, Benjakul [[85\]](#page-24-16) studied the impact of ultrasonication on collagen extraction from clown featherback (*Chitala ornata*) skin, varying amplitudes (20–80%) and durations (10–30 min), observing an enhanced collagen yield from 27.18 to 57.35%. However, ultrasound-induced protein degradation led to reduced hydroxyproline level and compromised collagen purity, especially evident with higher amplitudes and longer durations [[84\]](#page-24-15). Thus, maintaining optimal amplitude and extraction time is crucial for harnessing ultrasound's potential to enhance both quantity and quality of collagen.

3.7 Salt solubilization extraction

Collagen extraction from marine sources using saline solutions (*e.g*., citrates, phosphates, Tris–HCl, or sodium chloride) is known as salt-soluble collagen. Despite its potential, this technique is rarely employed due to collagen's limited solubility in saline solutions. Liang, Wang [[75\]](#page-24-6) reported the successful extraction of salt-soluble collagen from the cartilage of Amur sturgeon (*Acipenser schrenckii*) using NaCl (0.45 M at a ratio of 1:1000 w/v) with constant stirring for 24 h. Apart from diferences in molecular structure, amino acid profle, and thermal stability, there were remarkable diferences in yields using salt-soluble collagen (2.18%), acid-solubilized collagen (27.04%), and pepsin-solubilized collagen (55.92%).

4 Collagen‑based colloidal structures

Marine collagen is ideal for thickening, texturizing, and gel production due to its high water absorption capacity. Additionally, it exhibits interesting surface characteristics, such as emulsifcation, foaming, and flm-forming properties. These attributes may be attributed to charged groups and hydrophobic/hydrophilic elements in the protein side chains. The source and extraction method signifcantly impact these functional attributes of collagen.

4.1 Emulsions

An emulsion is a dispersion of small liquid droplets in another immiscible liquid, stabilized by surface-active emulsifiers $[86]$ $[86]$. These emulsifiers, such as marine collagen-based ones, fall into two categories: soluble molecules and insoluble aggregates. Soluble collagen, which is rich in hydrophobic and hydrophilic amino acids, enhances emulsion stability by lowering interfacial tension and inducing electrostatic repulsion between droplets. This effect is measured by emulsifying activity and stability indices [[81\]](#page-24-12). For instance, the emulsifying activity and stability indices of type II collagen from softshelled turtle calipash were lower than those of collagen from chicken sternal cartilage at pH 7, while the results at pH 4 or 10 are opposite [\[81,](#page-24-12) [87\]](#page-24-18). Kulkarni, Maniyar [[88\]](#page-24-19) observed that collagen from fsh (*Cirrhinus mrigala*) scales exhibited good emulsifying potential with an emulsion activity index and emulsion stability index of 21.49 m^2 g⁻¹ and 15.67 min, respectively. Shaik, Asrul Effendi [[89\]](#page-24-20) revealed that collagen extracted from the *sharpnose stingray'*s skin using acetic acid (15.01 min) exhibited signifcantly higher emulsifying stability compared to that extracted with hydrochloric acid (12.91 min). This difference is likely attributed to changes in the collagen's triple helix structure during emulsion formation, thereby modifying protein surface hydrophobicity and partially infuencing its emulsifying properties. As an insoluble protein, collagen is generally considered an inefective emulsifer, but it can embed in the oil–water interfaces, forming Pickering emulsions stabilized by solid particles [[90\]](#page-24-21). For that purpose, processing treatments can convert insoluble collagen into soluble forms like gelatin, which acts as a surface-active agent, reducing interfacial tension or improving droplet zeta-potential.

Dey, Kadharbasha [\[91](#page-24-22)] isolated collagen type I (110– 120 kDa) from seven fsh by-products, yielding 9.15% to 92.38%. It was found that collagen hydrolysate from Tilapia bones and Pacu skin showed over 80% solubility across a wide pH range, 92–96% moisture retention, zeta potential>50 mV, and emulsifcation activity 53–70

 $\mathrm{m^2\ g^{-1}}$ with stability lasting 62–85 min. This collagen improved drug emulsion stability by 14 times, revealing a polyproline-II conformation forming a quasifbrillar network. Surface activity relied on small size, varied hydrophilic/lipophilic ratios, hydroxyproline abundance, and peptide assembly at the emulsion interface, forming a mimic-helix-based quasifbrillar network for optimal orientation and interaction with multiple phases. Razali, Zainol [\[92\]](#page-24-23) utilized fsh scale collagen to stabilize a water-in-virgin coconut oil emulsion, demonstrating excellent physical stability without separation layers, making it suitable for skin application. They found that the addition of collagen slightly reduced emulsion droplet size and resulted in shear-thinning behavior, aligning well with the requirements for topical application.

4.2 Foams

Foam, characterized as a two-phase system containing air bubbles dispersed in a continuous liquid/solid phase, is a fundamental component of various food products, including smoothies, desserts, whipped cream, ice cream, carbonated beverages, meringues, marshmallows, and mousses [[1](#page-22-0)]. Foam structures are typically stabilized by alcohols, fats, proteins, and surfactants. Their halflife stability, lasting 50 min or more, makes them especially valuable in the food industry for preserving the desired appearance, color, and texture of aerated products. Marine collagen, with a hydrophilic-hydrophobic amphipathic structure, can quickly migrate and adsorb to the air–water interface through difusion and resetting, improving interfacial rheological viscoelasticity and forming cohesive three-dimensional gels for gas packing $[81]$ $[81]$. These proteins effectively reduce interfacial tension, creating protective flms around air bubbles [[20\]](#page-22-18).

Furthermore, foaming capacity and stability of collagen proteins depend on the physical properties of the formed flm [[81\]](#page-24-12). For instance, conformational aggregation and fexibility of the protein can create higher interfacial activity and a thicker interfacial layer, increasing viscosity and facilitating a multilayer cohesive protein flm at the interface [\[93](#page-24-24)]. Moreover, macromolecular collagen proteins consist of covalently bonded amino acid residues, resulting in a rare thermodynamic rupture of the bubble membrane compared to that caused by small molecules. For instance, Chen, Li [\[94\]](#page-24-25) reported that the foam stability values of acid- and pepsin-soluble collagens, extracted from red stingray skin, ranged from 12.50 to 72.0%, and 5.32 to 61.85%, respectively, which were higher than those of basmati rice protein concentrate (0.65–2.50%) and casein (0.17–0.54%). In addition, foaming capacity of acid- and pepsin-soluble collagens ranged from 47.62 to 146.67%, and 71.43 to 151.67%, respectively, which exceeded those of rice bran protein concentrate

(5.2–10.03%), casein (3.95–10.15%), and scallop gonad protein isolates (25–90%) [\[91\]](#page-24-22). However, collagen proteins, while widely used, may lack the necessary foam capacity or stability to meet the stringent demands of the evolving food industry. For instance, adjusting the pH to the isoelectric point reduces foaming capacity for acidand pepsin-soluble collagens extracted from rainbow trout (*Oncorhynchus mykiss*), rendering "soluble" collagen insoluble and hindering the necessary protein-water interaction for foaming [\[20\]](#page-22-18). Zou, Wang [[81](#page-24-12)] reported that soft-shelled turtle (*Pelodiscus sinensis*) collagen had the highest foam capacity and foam stability values at pH 10, followed by pH 4 and 7. Changes in protein fexibility and exposure of hydrophobic groups in alkaline conditions could induce foam formation and stability changes.

4.3 Films

Marine collagen is employed to fabricate edible and biodegradable flms, providing both economic advantages and environmental protection $[93]$ $[93]$ $[93]$. These collagen-based flms are typically fabricated through casting or extrusion of colloids aqueous dispersions containing collagen fbers [[95\]](#page-24-26). Pure collagen-based flms generally exhibit a uniform and continuous structure characterized by smooth surfaces without holes or cracks, which can be attributed to the good compatibility between biopolymers. The distinctive triple-helical structure of collagen is noted for enhancing flm density, resulting in superior mechanical properties compared to other biopolymer flms. For instance, 5 mg/mL collagen showed the tensile strength of 88.46 MPa, while 70 mg/mL soy protein isolate, 50 mg/mL gelatin, 25 mg/mL chitosan, and 10 mg/mL κ-carrageenan exhibited 13.6 MPa, 22.42 MPa, 1.76 MPa, and 22.59 MPa, respectively [[96–](#page-24-27)[99](#page-24-28)]. Moreover, various pretreatments may be applied to collagen, infuencing its structure and the characteristics of the resulting flm. Xu, Wei [\[95](#page-24-26)] reported that flms cast from collagen fber dispersion pretreated at temperatures above 39 °C exhibit reduced tensile strength, but increased water resistance properties. Acid swelling within the pH range of 1.5–4.0, with pH 3 being optimal, enhances the mechanical and thermodynamic properties of the flms by maximizing the swelling ratio of collagen fbers. Ma, Teng [[100](#page-24-29)] employed high-pressure homogenization to fabricate micro/nano collagen fbers, suggesting that smaller fber sizes lead to improved mechanical strength and water resistance in collagen flms, with minimal impact on thermal stability. Ahmad, Nirmal [\[101\]](#page-24-30) prepared biodegradable flms from acid-solubilized collagen extracted from starry triggerfsh, which showed the highest contact angle, elastic modulus, and tensile strength, but the lowest water vapor permeability and elongation at break compared with the pepsin-solubilized collagen film. The

former flm exhibited a heat-stable mass residue of 30.9% (w/w) in the temperature range from 50 to 600 °C, while the latter one showed 14.3% (w/w), indicating robust protein–protein interactions within the flm network.

The film-forming properties of collagen can also be enhanced by combination with proteins/polysaccharides since protein-polysaccharide complexation efectively improves the mechanical and functional properties (Table [2\)](#page-14-0). Liang, Feng [[102](#page-24-31)] designed composite flms from fish scale collagen and polyvinyl alcohol containing varying amounts of potassium sorbate, and found the improved elongation at break, tensile strength, and antimicrobial properties. According to Nuerjiang, Bai [[9\]](#page-22-8), compared to a pure fsh collagen flms, the Young's modulus, tensile strength, and elongation at break of collagen-gelatin composite flms enhanced by 79.2%, 37.1%, and 34.4%, respectively, while the water vapor permeability reduced by 51.5%. Ghosh, Grosvenor [[17\]](#page-22-15) found that *spongia* collagens exhibited hydrophilic properties, and flms made solely from these collagens were mechanically fragile, disintegrating instantly when wet. However, blending chitosan with *Spongia* collagens at a ratio of 30:70 (dry mass basis) not only enhanced mechanical properties but also improved structural integrity. This improvement was further intensified by the addition of organic cross-linkers, such as glyceraldehyde and genipin, as evidenced by stress–strain spectra, microscopic images, and mass swelling in water. In a study by Sionkowska, Kozłowska [[103](#page-24-32)], UV-irradiation of fsh scale (Esox lucius) collagen flm altered the surface free energy and contact angle due to the decrease in amino acids present. Also, the addition of Ag⁺, Ca^{2+} , and Fe^{3+} proved to be an efective in improving the functional and mechanical properties of collagen-based flms [\[100,](#page-24-29) [104](#page-24-33)].

In recent years, the incorporation of Pickering emulsion into collagen-based flms has emerged as an innovative hydrophobic modifcation technique, suggesting great potential for introducing novel functionalities and enhancing packaging material characteristics [\[112,](#page-25-0) [113](#page-25-1)]. Pickering emulsion excels in transporting hydrophobic bioactive ingredients or essential oils, improving their compatibility with hydrophilic matrices and consequently

enhancing film physical properties $[112]$. The schematic diagram in Fig. [3](#page-15-0) illustrates the fabrication process of Pickering emulsion-loaded collagen-based flm. In a study by Ran, Zheng [[113](#page-25-1)], cinnamon essential oil-loaded Pickering emulsions were fabricated using solid particles soy protein isolate/chitosan. These particles irreversibly adsorbed onto the oil through hydrophobic interactions, electrostatic interactions, van der Waals forces, and hydrogen bonding. The resulting Pickering emulsions were incorporated into the collagen flm-forming solution, which not only increased the water barrier properties, but also improved UV-blocking properties of the collagen film. The incorporation of Pickering emulsion into collagen flms increased water contact angle, water vapor permeability, and elongation at break from 81.87° to 105.37°, 1.34 to $4.60 \text{ g} \times \text{m}^{-1} \times \text{s}^{-1} \times \text{Pa}^{-1}$, and 9.22 to 36.85%, respectively, while decreased the tensile strength from 93.43 to 41.77 MPa. Furthermore, this integration enhanced the flm's antimicrobial and antioxidant activities while improving thermal stability.

4.4 Gels

The gel formation of marine collagen referred to a continuous process, which includes the self-assembly of collagen through self-aggregation and intermolecular forces, micro-fbrils produced via fbrillogenesis by collagen (4–8 molecules), and collagen fbrils formed from these micro-fbrils [\[1](#page-22-0)]. Generally, during the nucleation phase,

the size and number of microfbrils formed play a crucial role in shaping the fnal structure of self-assembled collagen fibrils. These factors considerably effect the features of the collagen gel networks, such as uniformity of pore distribution, size, and fber density [\[114\]](#page-25-3). For instance, Jiang, Wang [\[115](#page-25-4)] established that ultrasonic treatment reduced the diameter and uniformity of grass carp skin collagen fbrils by enhancing microfber number and homogeneity during nucleation. This results in collagen gels with more heterogeneous pore structures and larger pore sizes. In addition, Xu, Wei [[95\]](#page-24-26) employed ultraviolet radiation at low temperatures to induce collagen gel formation. They observed molecular degradation and crosslinking, which produced collagen fbrils with increased "branches," promoting fbril intertwining. Notably, the amino acid content and characteristics of marine collagen difer from mammalian collagen, giving rise to unique features in the self-assembly of marine collagen [\[116](#page-25-5)]. For instance, Bao, Sun [[117\]](#page-25-6) observed that higher amounts of hydroxyproline and cysteine induced more cross-linking, resulting in a more homogeneous structure, for example, tilapia collagen gel showed higher mechanical properties and elasticity than porcine collagen gel [\[118\]](#page-25-7).

Marine collagen gels can be induced by altering the temperature, pH, or ionic conditions. When collagen solutions are vigorously heated, the stabilizing bonds of collagen's triple helical structures break, resulting in a disordered conformation. Typically, collagen proteins

Pickering emulsion loaded collagen solution Homogenization Oil ∔ **Pickering emulsion** Mixing Drying **Particle solution Dissolution Pickering emulsion loaded** Collagen powder **Collagen solution** collagen film

Fig. 3 Schematic diagram of the formation process of Pickering emulsion-loaded collagen-based flm

denature in the temperature range of 53–63 °C, likely involving the initial breakage of hydrogen bonds, followed by the loss of collagen fibril and molecule contraction [\[116\]](#page-25-5). Shi, Tian [[119\]](#page-25-8) demonstrated that varying pH (5.0–8.0) afects the gel formation of golden pompano collagen, leading to an increased collagenous fbril number and diameter with rising pH. Moreover, collagen gel networks exhibit reduced fbril diameters and pore spaces as temperature increases from 4 to 37 °C. Tian, Ren [[118](#page-25-7)] reported that tilapia skin collagen gels develop a denser fbril network with higher NaCl concentrations in simulated body fluid. This is attributed to chloride ions neutralizing collagen molecule surface charges, reducing inter-molecular repulsion, and promoting the formation of compactly packed collagen fbril aggregate gels with D-periodicity structures.

Generally, collagen gels produced from native marine sources are not strong enough to meet the requirements of diferent food applications [\[120,](#page-25-9) [121](#page-25-10)]. One possible way to manipulate the properties of a low-gelling collagen to achieve greater viscoelasticity and mechanical strength than mammalian collagen is to induce cross-linking through physical, chemical, or enzymatic actions [[120](#page-25-9), [122](#page-25-11), [123](#page-25-12)]. However, the use of chemical cross-linking agents (*e.g*., carbodiimide, glutaraldehyde, and formaldehyde) has been reported as a means of reinforcing collagen structures, but they are potentially toxic [\[120](#page-25-9)]. On the other hand, the formation of collagen-based composite gels, such as combinations of collagen/protein or collagen/polyphenol, not only enhances the oxidative stability of gels but also maintains their structural stability and provides mechanical properties. The formation of networks in collagen-myosin composite gels involves various interactions, including ionic bonds, disulfde bonds, hydrogen bonds, hydrophobic interactions, and elec-trostatic interactions (Fig. [4](#page-16-0)) $[123-125]$ $[123-125]$ $[123-125]$. Zhao, Lu $[125]$ $[125]$ $[125]$ reported that collagen type I, with increased water solubility and a higher concentration of charged amino acids compared to type II, efficiently integrates with surimi myofibrillar proteins. This integration promotes heightened exposure of protein functional domains, induces signifcant conformational changes in myosin, and fosters stronger chemical forces among proteins. Consequently, these improvements accelerate the gelation rate, resulting in enhanced water holding capacity and superior textural profles in the collagen-myosin composite gels. Recently, An, Duan [[124\]](#page-25-14) introduced a cross-linking technique to boost tilapia collagen fbril density using chlorogenic acid

Fig. 4 Proposed collagen-myosin interaction model to illustrate diferent chemical forces in collagen-based gel

and procyanidin, which resulted in a roughly threefold increase in enzymatic resistance, mechanical properties, and water absorption and retention capacity of the collagen gel, while thermal stability decreased. In addition, polyphenol cross-linking granted better antioxidant activity to the gel, especially procyanidin, resulting in higher DPPH radical scavenging and antibacterial activity, while chlorogenic acid showed a higher Fe (II) chelation ratio.

5 Applications of marine collagen in food systems

In this section, we explored the potential applications of marine collagen in the food industry, considering its enhanced texture, mechanical properties, as well as antibacterial and antioxidant activities. The schematic diagram illustrating the simulated application of marine collagen is presented in Fig. [5](#page-17-0).

5.1 Meat products

Many processed meat products, such as sausages, patties, and burgers, include ingredients to enhance their functional properties. Collagen proteins, especially those derived from marine sources, are frequently added to comminuted meat recipes to boost stability through improved gel structure and texture. While collagen has limited nutritive value concerning essential amino acids, several authors suggested that substituting 10–20% of the total meat protein content with collagen could enhance emulsifying stability, resilience, hardness, and water-binding capacity in processed meat products [[126\]](#page-25-15). According to Zhao, Lu $[125]$ $[125]$ $[125]$, adding 9% cod skin collagen to surimi gel enhances the formation of ionic bonds, disulfide, and hydrogen bonds. This results in the highest gel strength of 554 g × cm and the highest waterholding capacity of 74.66%, which could be attributed to more charged amino acids intertwining with surimi

Fig. 5 An overview of food applications of marine collagens

myofbrillar proteins. Ibrahim, Ismail‐Fitry [\[126\]](#page-25-15) demonstrated that partially substituting bufalo fat with fsh collagen hydrolysates (ranging from 2.5% to 10%) in buffalo patties afected cooking loss, water-holding capacity, and pH value of the cooked patties. Sousa, Fragoso [[127\]](#page-25-16) replaced pork backfat with hydrolyzed collagen in Frankfurt-type sausages, fnding that 50% collagen addition resulted in improved aroma, protein profle, oxidative stability, texture properties, and water-holding capacity compared to sausages prepared from whole pork meat. Prestes, Carneiro [\[128](#page-25-17)] reported a higher waterholding capacity (92.8–98.8%) in turkey ham processed with added collagen, guar gum, and starch, attributing it to the synergistic efect of biopolymers for retaining water. Ham, Hwang [[129\]](#page-25-18) investigated the replacement of fat with combinations of collagen and dietary fbers in the fermented sausage products, and found the increased moisture, protein, and ash contents but decreased fat content.

5.2 Dairy products

Dairy products currently constitute approximately 40% of functional foods, aligning with consumer preferences that extend beyond essential nutrition to focus on wellbeing and disease prevention. Probiotics and prebiotics are notably key ingredients in these dairy products [\[130](#page-25-19)]. However, recently reported bioactive compounds, such as marine collagen, are gaining attention for their use as emulsifers in various products like ice cream, chocolate, yogurt, and butter. Apart from providing functional properties, marine collagen enhances the microbiological, rheological, and physicochemical aspects of these products (Table [3\)](#page-19-0). These foods can be classified into two emulsion-based categories: oil-in-water-based foods (*e.g*., chocolate sauce) and water-in-oil-based foods (*e.g*., butter) [[91\]](#page-24-22).

Dey, Kadharbasha [[91\]](#page-24-22) reported that collagen hydrolysates preserved the emulsion properties of butter and chocolate sauce, leading to a signifcant extension of their shelf life. Moreover, no cytotoxic efects were observed on leukocytes and Vero cells. Shori, Tin [[141](#page-25-20)] found that the presence of fsh collagen in *Codonopsis pilosula*yogurt improved O-phthaldialdehyde peptide concentrations and scored positively in sensory evaluations. Additionally, Shori, Baba [\[142](#page-25-21)] investigated the effect of fsh collagen on cheddar cheese (*Allium sativum*) in terms of acidity and proteolysis of milk protein during a 60-day ripening period. The results revealed that fish collagen increased the free amino acid content in cheddar cheese from 1.66 to 2.47 mM/L (Leucine equivalent), suggesting a signifcant increase in the proteolysis of milk protein in the presence of fsh collagen. Shori, Yong [[143](#page-25-22)] also observed the highest peptide content in plant extract-based cheeses (*Illicium verum-, Psidium guajava-, or Curcuma longa*) by adding fsh collagen. In another development, Bhagwat and Dandge [[130\]](#page-25-19) created a milk-based food product, paneer, by incorporating collagen extracted from fish scales. The resulting paneer contained a higher concentration of moisture and protein, contributing to favorable sensorial and textural attributes.

5.3 Drinks

The integration of collagen into beverages has been found to provide various benefts, driving the growing demand for products such as soy collagen, cocoa collagen, and cappuccino collagen. Collagen inclusion in these drinks supports the body's natural ability to generate fatty tissues and can stimulate bodily mechanisms, contributing to the strengthening of tissues and reducing skin wrinkles. Hydrolyzed collagen, due to its low viscosity and high water solubility, is commonly added to beverages to enhance their functional and nutritional properties without causing technological issues in production. Bilek and Bayram [[144\]](#page-25-23) prepared new functional drinks from white grape, apple, and orange juice blends containing hydrolyzed fsh collagen. Results indicated that fruit juice drinks formulated with a substitution of up to 2.5% collagen increased the protein content of the product from 0.56 to 2.22–2.48 g/100 mL. In vitro bioavailability results showed that apple (90.71%) and orange (95.37%) drinks exhibited higher bioavailability (5–14%) compared to white grape juice blends (1.43%). The drinks also varied widely in terms of total phenolic content (86.93–117.43 mg GAE/100 mL), ascorbic acid content (81.39–113.5 mg/100 mL), and antioxidative results (104–127 µmol TEAC/100 mL). Sae-leaw, Aluko [[145](#page-25-24)] investigated the impact of salmon scale ossein-derived collagen on the sensory, antioxidant, and physicochemical aspects of chrysanthemum tea over a 6-month storage period. The research revealed that incorporating collagen into the tea led to elevated α-amino group content, pH, turbidity, and browning index. Additionally, it yielded a beverage with antioxidant properties, extending its shelf life to 6 months at 30 °C.

5.4 Food packaging materials

Marine collagen, renowned for its native triple helices and fbril networks, fnds extensive application in the packaging of meat, poultry, and seafood products, particularly as artifcial casing due to its sustainable, biodegradable, and biocompatible nature (Table [4](#page-20-0)) [\[146](#page-25-25)]. Collagen-based packaging materials serve as excellent barriers to moisture and oxygen due to their tightly packed, ordered covalent-bonded network structure. Additionally, collagen packaging materials exhibit

Table 3 Food applications of marine collagen-based packaging materials

Table 3 (continued)

Collagen source	Collagen type & concentration	Other compounds	Packaged food	Key findings	References
Blue shark	Type &0.8%	Chitosan	Red porgy fillets	Thiobarbituric acid, pH value, drip loss, and sen- sory evaluation scores were improved	[134]

Table 4 Applications of marine collagen in dairy products

enhanced mechanical properties, including adsorption, adhesion, solubility, transparency, and favorable sensory and organoleptic properties [\[146](#page-25-25)].

Bhuimbar, Bhagwat [\[132](#page-25-27)] utilized thick black ruf skin collagen to develop collagen-chitosan-based antibacterial food packaging flms with pomegranate peel extract, proving efective against water vapor permeability and foodborne pathogens. In another study, Song, Liu [\[131\]](#page-25-26) characterized tilapia fsh skin collagen for developing active food packaging materials to preserve tilapia muscle. It was found that collagen/zein electrospun flm encapsulating diferent concentrations of gallic acid inhibited tilapia muscle quality deterioration, prolonging the shelf life of fsh muscle for at least two days. Wang, Hu [\[133](#page-25-28)] reported that collagenlysozyme coatings (4% collagen+0.5% lysozyme, w/v) on fresh-salmon fllets (*Salmo salar*) inhibited bacterial

growth, reduced weight loss, and improved texture. Ben Slimane and Sadok [[2\]](#page-22-1) highlighted the effectiveness of a bioflm prepared from cartilaginous fsh (*Mustelus mustelus*) collagen and chitosan, demonstrating superior UV barrier properties and antioxidant activity compared to chitosan alone. This biofilm shows promise as a green bioactive flm for preserving nutraceutical products. Nicklas, Schatton [[155\]](#page-26-0) formulated an aqueous gastro-resistant coating using freeze-dried sponge collagen at a concentration of 15% (w/w) as the film-forming agent. The findings indicated that tablets coated with this collagen successfully resisted 0.1 M HCl for over 2 h while disintegrating within 10 min in a phosphate buffer solution with a pH of 6.8. These coated tablets exhibited favorable mechanical properties and maintained their enteric characteristics for a minimum of 6 months during storage.

When employed in sausage casings, marine collagen provides several advantages over natural casings, including uniformity, sanitation, and fexibility, making it recognized as one of the most promising casings. Its unique ability to shrink and stretch allows it to mimic the expansion and contraction of flesh batter during continuous processing $[8]$ $[8]$. This characteristic not only improves shelf life but also reduces cooking shrinkage, enhances juiciness, and minimizes elastic stretch after heat treatment. Additionally, marine collagen fnds extensive application in producing edible casings for preserving meat products, such as salami and snack sticks. Regardless its numerous benefts, collagen casings face certain challenges, including bending, folding, and stretching during shirring, resulting in increased friction. Their low mechanical strength may lead to ruptures during stufing, and the casings may easily rupture during sausage flling, often due to excessive local resistance [[8\]](#page-22-7). Nevertheless, researchers have explored solutions to these challenges. The addition of aldehydes $[156]$ $[156]$, metal ions [[104\]](#page-24-33), keratin [\[157](#page-26-3)], and carbodiimide $[158]$ to collagen casings has been shown to increase their mechanical properties, addressing issues related to deformation and rupture during processing. For instance, Chen, Liu [[156\]](#page-26-2) reported that glutaraldehyde improved the tensile strength and Young's modulus of collagen casings, resulting in enhanced deformation resistance and mechanical strength.

5.5 Food supplements

Marine collagen serves as a food supplement, contributing to lean muscle growth, joint repair, accelerated recovery, and improved cardiovascular performance. The demand for collagen supplements, driven by their myriad benefts, has soared in sports nutrition [[159\]](#page-26-5). Notably, collagen has demonstrated efectiveness in treating rheumatoid arthritis, with studies revealing signifcant reductions in pain, morning stifness, and joint swelling [[160\]](#page-26-6). Collagen is recommended as a dietary supplement for the treatment of hand, knee, and hip osteoarthritis in the United States [[161\]](#page-26-7). As a food supplement, the daily collagen recommended dosage ranges from 800 to 1200 mg [[161](#page-26-7)]. In sports nutrition products for athletes, sardine scales collagen, available in various forms such as powdered and liquid supplements, multicomponent bars, and chewing marmalade, is valued for its rich amino acid composition $[162]$ $[162]$. Shark cartilage supplements have also gained attention, claimed to be natural antiinfammatory and anti-carcinogenic agents that relieve arthritis and reduce joint pain, thanks to their chondroitin and glycosaminoglycan contents [\[163](#page-26-9)]. Furthermore, marine collagen peptide derived from fsh waste has been

utilized as a fortifer in biscuits, contributing to elevated protein and antioxidant content [[161\]](#page-26-7).

6 Conclusions and future perspectives

Marine collagen is rapidly gaining global acceptance for its interesting functional and physicochemical properties, with vast potential in various food applications, including packaging materials, beverages, dairy products, food supplements, and meat-based items. This review has discussed the current state of knowledge on diverse marine collagen sources, including sea cucumbers, mollusks, marine sponges, fsh, jellyfsh, and crustaceans, emphasizing low-cost collagen extraction based on various studies. Fish and jellyfsh notably yield predominantly type I collagen, while crustacean muscle collagen constitutes a minor fraction (0.5–1%) of total protein content. The prevalent method involves solvent extraction with acetic acid, yet acidic treatments are impractical for marine sponge collagens due to their insolubility. Some studies have explored enzymatic extraction, showing signifcant potential for high collagen yield. While emerging technologies like extrusion-hydro-extraction, supercritical fuid extraction, and deep eutectic solvent extraction hold promise, further research is needed for their practical application in efficiently and cost-effectively extracting collagen proteins from marine sources. Therefore, it is recommended that research efforts should be focused on the development of environmentally friendly and costefective methods for extracting collagen from diverse marine sources.

Marine collagen, when utilized in its native form, has demonstrated the capability to enhance both biodegradability and the antioxidative/antimicrobial properties of flms, especially those incorporating Pickering emulsions. However, challenges such as brittleness, low mechanical strength, high water solubility, and increased water vapor permeability are associated with marine collagen flms. A major drawback, particularly with collagen sourced from cold-water organisms, is its lower denaturation temperature, afecting both processing conditions and flm properties. While techniques like crosslinking, blending, or chemical modifcations can improve thermal stability, exploring collagens from warm water marine species with superior thermal stability offers a more sustainable approach. This could facilitate collagen processing under milder conditions, reducing the necessity for extensive modifcations to achieve thermal stability in food packaging flms. Consequently, ongoing research and development are essential to establish packaging alternatives that can compete with currently employed petroleum-based polymers.

Marine collagen-based novel functional food ingredients contain nutritional benefts, including non-essential

and essential amino acids, enhancing the quality of various food products. Studies suggest that incorporating collagenous proteins can improve technological and sensory attributes in comminuted meat products undergoing fat replacement with dietary fber. Additionally, they serve as texturing agents and natural antioxidants, potentially reducing reliance on chemical preservatives and meeting consumer demands for safer and environmentally friendly food products.

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Author contributions

SF: Investigation, Conceptualization, Writing—original draft. MIA and: Designed fgures, UA: Data curation. SZ: Designed tables, YL and CS: Contributed to manuscript revision. HZ: Conceptualization, Project administration, Supervision, Writing—review & editing, Funding acquisition.

Availability of data and materials

The authors declare that all the data supporting the fndings of this study are available within the article.

Declarations

Competing interests

Hui Zhang serves as the Associate Editor-in-Chief of Collagen and Leather, and he was not involved in the editorial review, or the decision to publish this article. All authors declare that there are no competing interests.

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