A Review on Ultra- and Nanofiltration/Diafiltration Processes of the Food-Oriented Agro-industrial

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Abstracts: This review was conducted to implement the potential use of ultra- and nanofiltration as a separation technique accompanied by a diafiltration (DF) technique. We start by introducing the principle of pressure-driven membrane-based separation process and DF process in general, and selected applications of ultra- and nanofiltration/diafiltration techniques on the food-oriented agro-industrial products in particular, such as nanofiltration/diafiltration (NF/DF) technique of mung beans autolysate, ultrafiltration/diafiltration (UF/DF) technique of forest honey products, and ultrafiltration/diafiltration (UF/DF) technique of fish gelatin. We also describe the potential benefits as green and sustainable and the drawbacks of membrane-based separation process. Both relevant information and data related to this review were obtained from various sources originating from international organizations (FAO and UNEP, United States Standards for Grades of Extracted Honey, USA and National Honey Board (NHB), USA), review of various scientific articles and scientific literature, handbook, and the selected result of experiment activity. We hope this review manuscript is useful for further research into membrane technology implementations in food-oriented agro-industrial products and in a wider range of applications.

Keywords: Diafiltration (DF), Ultrafiltration (UF), Nanofiltration (NF), Food-Oriented Agro-Industrial.

1. INTRODUCTION

1.1. Background

Indonesia is endowed with tremendous potential in terms of abundant natural resources that are closely associated with the food-oriented agro-industry. The agro-industrial sector is a subset of the manufacturing sector and plays a vital role in transforming raw agricultural materials and intermediate products derived from agriculture, forestry, and fishery (AFF) into value-added goods that significantly contribute to economic growth. Therefore, Indonesia must maximize effective management in these sectors to improve the national economy and welfare of the community. The significance of the agro-industry is evident in its substantial contribution to the country’s Gross Domestic Product (GDP), accounting for 13.28% in 2021. This places the sector as the second biggest contributor to Indonesia's GDP, following the manufacturing industry with 19.25% in 2021. One possible strategy to improve the welfare of the food-oriented agro-industry is to focus on developing food-oriented agro-industrial products [1;2]. The agricultural sector, being the main source of food-oriented activities, serves as the backbone of the agricultural economy and plays a role in national economic development. Furthermore, its contribution to the Gross Domestic Product (GDP) is significant, and it serves as a major employment provider, thereby increasing people’s income and livelihoods. Indonesia, known as the largest agricultural country, has the second-largest biodiversity land area globally, following Brazil. This is exemplified by the diverse range of commodities, including food crops, horticulture, plantations, and farms that have long served as a source of food and social revenue [3]. The forestry sector encompasses the management of trees and other vegetation within the natural forests consisting of primary and secondary types. Forests are crucial for biodiversity conservation, as they provide habitats for food and medicinal plants. These areas play a role in carbon capture, storage, and climate change mitigation. Previous studies have shown that forests offer a wide array of natural resources to meet human needs along with abundant biodiversity [4]. This abundance facilitates the production of diverse goods, including timber and non-timber forest products (NTFPs) (also known as non-wood forest products). Although both timber and NTFPs are considered renewable or sustainable, they exhibit distinct characteristics and contrasting attributes. In Indonesia, the fishery sector plays an important role in ensuring national food security. Due to the coastal nature of several communities, fish is an important component of the people's diet. As a natural resource, the fishing sector is recognized as a potential area for economic growth and a strategic issue in sustainable fishery activities in Indonesia. Furthermore, it is renowned as one of the globally recognized food industry sectors [5]. The forestry sector encompasses the management of trees and other vegetation within the natural forests consisting of primary and secondary types. Forests are crucial for biodiversity conservation, as they provide habitats for food and medicinal plants. These areas play a role in carbon capture, storage, and climate change mitigation. Previous studies have shown that forests offer a wide array of natural resources to meet human needs along with abundant biodiversity. This abundance facilitates the production of diverse goods, including timber and non-timber forest products (NTFPs) (also known as non-wood forest products). Although both timber and NTFPs are considered renewable or sustainable, they exhibit distinct characteristics and contrasting attributes. In Indonesia, the fishery sector plays an important role in ensuring national food security. Due to the coastal nature of several communities, fish is an important component of the people's diet. As a natural resource, the fishing sector is recognized as a potential area for economic growth and a strategic issue in sustainable fishery activities in Indonesia. Furthermore, it is renowned as one of the globally recognized food industry sectors [6]. To ensure the production of convenient agro-industrial products, it is important to establish a continuous and streamlined process. Therefore, Indonesia must focus on exploring its agro-industrial product and resources by implementing the separation and purification process science and technology. This approach can lead to increased added value, the adoption of environmentally-friendly practices, and a reduction in global energy consumption. Furthermore, recent developments in advanced polymer manipulation, particularly in membrane design, have provided new possibilities for effective, practical, and useful separation unit processes and unit operations in chemical engineering. The use of separation process technology has been widely implemented to fulfill the demand for these commodities while achieving the desired quantity and quality [7].

In recent decades, membrane processes have emerged as the forefront technology for separation, purification, and concentration procedures. Pressure-driven membrane separation processes, including Ultrafiltration (UF) and Nanofiltration (NF), have been widely implemented in various industries, such as food and beverage, biotechnology, pharmaceutical, agriculture, and others [8]. Materials used in these technologies are often chemically inert and selected based on their suitability for specific separation processes. Furthermore, these materials possess a wide range of desirable features, including excellent chemical and thermal stability, durability, bacterial resistance, back-flushing capability, cleaning and biological stability, as well as superior mechanical strength and flexibility. Over time, several membrane separation techniques have been developed, progressing from conventional methods to advanced approaches, such as ultra-and nanofiltration/ diafiltration-based processes [9].

1.2. Objective

This manuscript aims to provide an overview of the principles underlying pressure-driven membrane-based-separation processes, with a specific focus on selected applications in the food-oriented agro-industrial sector. These applications often involved the utilization of the diafiltration (DF) technique. This review discussed the nanofiltration/ diafiltration (NF/DF) technique of mung beans autolysate, as well as the ultrafiltration/diafiltration of forest honey products and fish gelatin. Information and data supporting this review were obtained from various relevant sources, including international organizations, such as FAO, UNEP, the United States Standards for Grades of Extracted Honey, and the National Honey Board (NHB), USA. Several scientific articles, literature, handbooks, and selected experimental results were also consulted. This review also discussed the potential benefits and limitations of the membrane-based separation process.

2. BASIC PRINCIPLES OF MEMBRANE

2.1. Pressure-Driven Membrane Separation Process

The term ‘membrane’ referred to the thin layer of selective semi-permeable material with the thickness of the top active layer being approximately less than 1.0 μm [10]. The membrane employed a semi-permeable barrier, which served as a selective filter, and relied on an applied pressure to facilitate the separation of the target components from a liquid solution. Furthermore, it preferentially blocked one or more desired and target components on the surface, allowing them to be retained in what is known as the retentate or concentrate. Meanwhile, the membrane allowed other materials to pass freely as the permeate or filtrate. Depending on the specific process, the target products could either be found in the permeate, retentate, or both. The membrane's ability to selectively allow components to pass through was influenced by various factors, including pore size or molecular weight cut-off (MWCO), physical/chemical characteristics, the material being sieved, and trans-membrane pressure (TMP) [11]. Although all membranes were rated for specific pore sizes, the ratings were an unreliable measure of filter efficacy as they differed from one product and manufacturer to another. Based on the main ranges of separation, the processes could be classified into Microfiltration (MF, pore size of 0.1 – 10 µm, TMP of 0.2 – 5 bar), Ultrafiltration (UF, 0.01 – 0.1 µm, 1 – 10 bar), Nanofiltration (NF, 0.001 – 0.01 µm, 5 – 20 bar), and Reverse Osmosis (0.0001 – 0.001 µm, 10 – 150 bar) [12;13]. The UF membrane referred to a pressure-driven separation process that removed particles larger than the pore size through a sieving mechanism, separating particles from solution based on their MWCOs. Furthermore, it typically separated and concentrated macromolecular solutions with MWCO range of 1,000 – 1,000,000 Dalton (Da.) due to small pore sizes. This indicated that all substances larger than a membrane's MWCO, such as macromolecules (proteins, carbohydrates, and fat), were kept on the surface, while others smaller than the MWCO (sugars, salt minerals, and water) could easily pass through [14;15]. NF membrane referred to a pressure-driven separation process operating based on size (sieving effect) and charge (Donnan effect) with separation capabilities between UF and RO membranes. NF has nanoscale pores with a size typically ranging from 0.001 – 0.01 µm and corresponding to the MWCO range of 200 – 1000 Da. The process was often used to reject small molecules in dissolved organic matter with a molecular weight (MW) ranging from 150 – 250 Da. Furthermore, it had good selectivity for divalent ions (Ca2+, Mg2+), and could be used to separate low MW solutes (glucose, saccharides, amino acid, peptide), with demineralization activity. Based on previous studies, some of its advantages included high molecular rejections when compared to UF and low operating pressures in comparison to RO. NF separation technique was commonly used for demineralization and concentration of thermosensitive aqueous solutions [16;17].

2.2. Diafiltration (DF)

Another common application of UF and NF membranes, which utilized cross-flow filtration, was in the production of diafiltration (DF) frames. Furthermore, DF referred to a term used to achieve two main objectives, namely maximizing the yield of a macro solute solution (product yield) and replacing membrane-permeable molecules (micro solutes, small molecules, salts, undesirable impurities) in the initial product solution (feed) by adding a fresh diluent based on differences in molecular size. The fresh diluent inflow (solute-free pure, fresh water) was added into the feed solution tank at a flow rate approximately equal to the permeate outflow. At least one impurity with a particle size smaller than the membrane's pores was continuously washed out through a selective active membrane known as permeate. The most predominant parameter during the DF process was the number of DF volumes (ND), which was calculated by dividing the total collected permeate volume by the constant original product solution (feed) or retentate volume. The component concentration in the starting solution was an essential parameter because it affected the volume of the product to be diafiltered (low concentration, greater volume) as well as the permeate flow rate (higher concentrations, lower permeate flow rate). The permeate stream leaving the system was known as flux expressed as the volume of permeate per square meter of membrane area per time. The requirement for an effective separation was the utilization of an appropriate membrane with a high rejection for the macro solute and a low rejection for the micro solute based on their molecular size to obtain a pure solution [18;19;20;21]. DF mode in the product solution is shown in Figure 1 [22].

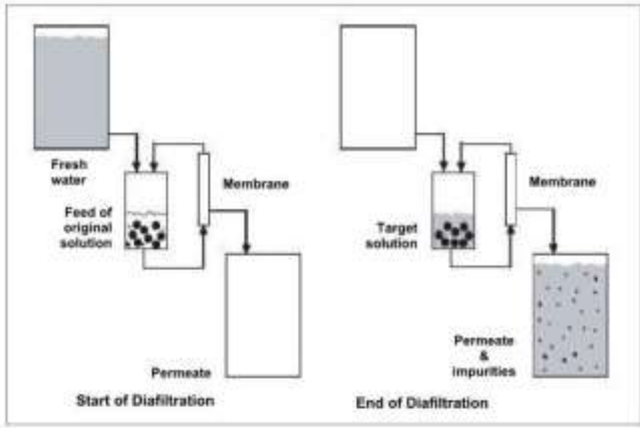

Figure. 1. Diafiltration methods in product solution (Adapted from ref. [22]).

2.3. Application of Ultra-and Nanofiltration/Diafiltration Processes

The pressure-driven membrane process as unit operations and processes in chemical engineering represented a unique separation techniques, such as separation, fractionation, purification, and/or concentration a wide range of liquids. To achieve these purposes, ultra- and nanofiltration membranes were often implemented in a process known as Diafiltration (DF). Depending on the separation target, DF could be combined with ultrafiltration (UF) or nanofiltration (NF) membranes. Furthermore, DF was extensively used in the food and beverage, pharmaceutical, nutraceutical, and biotechnological industries for separating multi-component process streams. One of the selected important applications discussed in this review was Ultra- and Nanofiltration/Diafiltration-Based Processes of Food-Oriented Agro-industrial Products [23]. Application of Ultra- and Nanofiltration/Diafiltration Techniques had substantially improved the food-oriented agro-industrial process. The membrane-based separation methods in the product of food-oriented agro-industrial, including agriculture (beans), forestry (honey), and fishery (gelatin), are discussed below.

2.3.1. Nanofiltration/Diafiltration (NF/DF) technique of mung beans autolysate

Mung bean (*Vigna radiata* (L.) R. Wilczek var. *radiata*) was a significant dietary legume crop with great potential for improving human nutrition for thousands of years. The mung bean was a useful and valuable component of prepared food systems because it was an excellent source of carbohydrates, iron, and zinc, as well as digestible protein and other essential minerals. Furthermore, vegetable or plant protein could contribute a much higher percentage of human nutrition in the future, thereby decreasing the consumption of animal protein products for health and economic reasons**.** Prepared mung bean was commonly consumed in East Asian (China, Korea, Japan, and Taiwan) and South-East Asian (Vietnam, Thailand, Malaysia, and Indonesia) countries [24].

One of the mung bean varieties that had been produced involved autolysis through brine fermentation with the addition of *Rhizopus* sp.-PL19 and *Aspergillus* sp.-K3 at pH 5.5 and 50 °C for 16 hours. Autolysis was often performed through the activity of the protease enzyme to boost savory (umami) fraction content, such as L-Glutamic acid. The umami flavor was distinct, with savory, brothy, or meaty characteristics. The salt gradually transformed into a culture medium, and the rate depended on several variables, including mung bean variety, mung bean/salt/inoculum proportion, and type and concentration of inoculum [25]. The savory portion in autolysate was separated using a 0.2 µm microfiltration (MF) membrane, yielding permeate and retentate. The NF membrane was then used to concentrate the MF permeate to produce NF retentate. (concentrate). Concentrate-Aspergillus sp.-K3 (Autolysate B) was a lighter-colored liquid compared to concentrate *Rhizopus s*p.-PL19 (Autolysate A). Furthermore, autolysate B was a murky yellow liquid with an umami and salty flavor. This liquid color was related to the autolysis process condition and inoculum types in which the presence of heat produced brown-colored pigment due to the Maillard reaction between monosaccharide and amino acids. Autolysate A and B were introduced as feed to NF/DF mode to remove part of the salt and partly caramel-colored pigment to meet the growing demand for healthy food. The need for highly valuable end products with low-salt content and light-colored liquid provided new opportunities to apply membrane technologies [26;27].

The NF/DF mode was carried out by filling the feed of Autolysate A into the feed tank to approximately one-third of its maximum capacity (3.5 L). The content in the feed tank was subsequently pumped tangentially using a fixed pump motor frequency (20 Hz, cross-flow rate of 7.5 L/min.) through a pre-filtration in 200 μm filter, heat exchanger system. The content was also pumped into a flat sheet NF membrane module (plate & frame) mounted vertically under constant room temperature, cross-flow rate of 7.5 L/min, and TMP of 20 bar for 5 minutes until the permeate contained colorless liquid. Autolysate A was then recirculated to the feed tank to keep an effectively constant concentration. DF-NF mode was performed in the continuous mode, where the permeate flux was compensated by an equal input of RO water (32 mL/min.). During the DF/NF mode, pure water (RO water) was introduced into the feed suspension tank, while the permeate was removed from the suspension tank through the same membrane surface. In this case, the retentate (feed) micro solutes progressively decreased and were removed as permeate flux. This experiment involved the addition of RO fresh water to different retentate (feed) volumes (N_D) at various ratios. Retentate and permeate were regularly sampled and recorded to analyze their salt, N-amino, and L-Glutamic Acid concentrations. Furthermore, the same procedure was conducted for Autolysate B. At the end of each process, the membranes were thoroughly flushed with RO water. The membranes were then cleaned using a 4% NaOH solution before storing them in 1 % Sodium azide till the subsequent run [25;28].

The experiment result showed that the NF-DF mode of both Autolysate A and B could reduce the optimal salt concentration at N_D 0.2 (20%) and N_D 0.2 (4.76%), respectively. High N_D gave a high salt reduction in retentate (concentrate). The optimum N_D (0.2) showed permeate as a side product with 70 and 51.23 L/m^2.h fluxes, 0 and 0.1325% salt concentration, 3.5 and 1.4 mg/mL N-amino, and 0.6375 and 0.3188% L-Glutamic acid (total protein). Meanwhile, the optimum N_D (0.2) gave the composition of Autolysate A and Autolysate B, including 0.106 and 0.53% salt, 7 and 7 mg/mL N-amino, and 0.5063 and 0.8437% L-Glutamic Acid (as a savory fraction) (total protein). At the best N_D (0.2), there was a decrease in the L-Glutamic Acid present in the retentate (concentrate) of Autolysate A (15%). Compared to the feed, there was an increase of L-Glutamic Acid in the retentate (concentrate) of Autolysate B (0.5%). Concentrate-*Rhizopus* sp.-PL19 (Autolysate A) as feed (F_a), retentate (R_a) and permeate (P_a) at the best N_D (0.2), and concentrate-*Aspergillus* sp.-K3 (Autolysate B) as feed (F_b), retentate (R_b), and permeate (P_b) at the best N_D (0.2) is visualized in Figure 2 [26].

(F_a) (R_a) (P_a) (F_b) (R_b) (P_b)

Figure 2. Concentrate-*Rhizopus* sp.-PL19 (Autolysate A) as feed (F_a) in NF/DF mode, retentate (R_a) and permeate (P_a), and concentrate-*Aspergillus* sp.-K3 (Autolysate B) as feed (F_b) in NF/DF mode, retentate (R_b), and permeate (P_b) (Adapted from ref. [26]).

2.3.2. Ultrafiltration/Diafiltration (UF/DF) technique of forest honey product

Non-timber forest products (NTFPs) played a significant role in tropical forests by providing two main products apart from timber. Several important NTFPs were categorized as healthcare medicinal plants and raw materials for pharmaceutical products. Some of them also served as raw material for other plant-based products, including foods (edible forest leaves and fruits, mushrooms, sago, palm sugar, forest tuber), food additives (forest spices and herbs), various resins (damar), oils, gaharu (a resin-impregnated fragrant wood), and animal-based materials, such as bushmeat from a wild animal, hides, skins and trophies, edible bird nests (saliva from the nests of swiftlets), and forest honey and beeswax. Among these products, forest honey was the major NTFPs for the generation of significant income [29].

Forest honey was classified as honey naturally produced in the forest by wild bees, particularly *Apisdorsata.* These bees sucked nectar from flowers and stored it in the beehive attached to trees. Forest honey was produced in almost every province in Indonesia, where they were often collected and labeled based on their geographical origins, such as Tesso Nilo (Riau), Gunung Kerinci National Park (Jambi, Sumatera), Pelawan Tourism Forest, Namang in Bangka Tengah and Belitung Island (Bangka Belitung), Ujung Kulon National Park and Panaitan Island (Banten), Odeng honey (West Java), Sumbawa Sub District (West Nusa Tenggara), Muntis Mountain Conservation (South Central Timor) and Maumere (Flores Timor, East Nusa Tenggara), Sentarum Lake National Park (Kapuas Hulu, West Kalimantan), and Tulak Tallu village (North Luwu, South Sulawesi) [30].

Honey was a naturally sweet substance in the form of a viscous liquid produced by bees (*Apis mellifera* L., *Apis dorsata*) and collected from plant flower extracts (floral nectar) or other parts of plants (extra or different floral) or excretion of plant-sucking insects or the living parts of plants (honeydew). Furthermore, its aroma characteristics were based on the floral sources providing nectar and pollen. Several reports had shown that it had favorable organoleptic properties with various benefits for human health. Honey had been considered a non-medicinal product, but it could have a supportive application in preventing diseases as a consumable food [31]. The color, taste, fragrance and flavor, and composition of a particular variety could vary due to several factors, including the season, the botanical origin of the flower, the bee and tree species, the nectar provider, geographical factors, and region, various ecological and environmental conditions, harvest method, processing techniques, storage condition, beekeeping practice, packaging, and storage time. The honey types could be classified into three general groups, namely monofloral, poly floral, and honeydew honey (known as wild honey) [32]. The color of liquid honey varied from colorless and clear (like water) to dark amber or black, where all the colors were shades of yellow and amber [33]. Monofloral [34], poly floral [35], and forest honey [36] are visualized in Figure 3.

(a) (b) (c)

Figure 3. (a) monofloral or flower honey (Adapted from [34]), (b) polyfloral honey (Adapted from ref. [35]), and (c) forest honey (Adapted from ref. [36]).

Honey was mainly composed of carbohydrates in the form of 38.2% fructose, 31.0% glucose, 17.1% water, 7.2% maltose, 4.2% trisaccharides, 1.5% sucrose, and 0.5% minerals. Furthermore, it contained different valuable minor nutrients, including vitamins, enzymes, antioxidants, flavoring organic compounds, free amino acids, and numerous volatile compounds, such as flavonoids, wax, ash, pollen, propolis, and trace elements. The main element found in honey was potassium along with others, such as calcium, chlorine, copper, iron, magnesium, phosphorus, sodium, and zinc. Maintaining a balance between sugar and water was critical in determining its quality. The aroma of honey was one of its most typical features and was often the main selection criteria used by consumers. Dark-colored honey had been reported to contain more phenolic acid derivatives but fewer flavonoids compared to light-colored variants [37].

According to USDA Grading Standards and instead of conventional thermal processing methods, the primary aims of UF with suitable membrane types and MWCO were frequently coupled with a diafiltration (UF-DF) mode involving the use of a UF membrane of 15,000 Da. Honey was very viscous and must be diluted by adding water before the process. The application of UF-DF mode on honey involved the addition of fresh and pure water at a flow rate equal to the permeate flow rate generated, filtering it under high operation pressure at the molecular level, and removing the water. This mode could be implemented to remove sugar (glucose and fructose) partially from honey solution (in permeate) with simultaneous rejection of proteins and elimination of unwanted extraneous substances, such as microorganisms, fine particles, pollen grains, air bubbles, and other materials normally found in suspension (in retentate) [38]. UF membrane of 15,000 Da could be used to completely remove the microorganisms present in honey, rendering it a microbiologically safe ingredient. The process could successfully remove enzymes (water-soluble proteins) and yeast cells from the product, leading to improved stability. Furthermore, the process was often operated under constant pressure and temperature. UF/DF mode was carried out to produce clear and brilliantly transparent honey that remained in the liquid state for a much longer time compared to the unfiltered variant. The benefits of the UF membrane process of honey included the absence of cloudiness, sedimentation, granulation, uniformity, and translucency in the product, as well as reduced viscosity, sterile, and consistent quality characteristics. The results obtained from the applications of UF showed that ultrafiltered honey could be used in gel formulations, cosmetics, and pharmaceutical preparations, as a sweetener in tea or coffee, fruit beverages, and food [39;40].

2.3.3. Ultrafiltration/Diafiltration (UF/DF) technique of fish gelatin

As one of the land-oriented maritime countries, Indonesia possessed an abundant wealth of marine natural products (MNPs). National economic growth and research development were fostered by the abundant utilization of various marine-based resources, including fishes, mollusks, crustaceans, micro and macroalgae (seaweeds), and mangroves, which frequently served as sources for valuable marine natural products (MNPs). Furthermore, fish was considered a main source of protein due to the presence of several essential amino acids in their muscles. Based on previous studies, the composition of fish included 64% meat and 36% by-products. The by-products represented 17% heads, 8% skin, 5% viscera, 4% bones, and 2% fins. The skin and bone generated by the fish-processing industry had the potential to be used as a source of functional ingredients, such as gelatin [41].

Gelatin was considered a high-value functional protein found in nature and was often produced from the controlled partial hydrolysis of the fibrous native collagen ($C_{102}H_{149}N_{31}O_{38}$) found in animal bones, skin, cartilage, tendons, and connective tissues. Furthermore, its physical properties included solid substance, colorless or slightly yellow to white, translucent, shiny, tasteless, and odorless. This protein could be in the form of flakes, powders, granules, and sheets. Gelatin, a heterogeneous mixture of fibrous, denatured, biodegradable, hot water-soluble, and cold water-insoluble protein, had molecular weights in the range of 80,000 to 250,000 Dalton (Da.), with 88% protein, 10% moisture, and 1-2% salt. In dry form, it contained approximately 98 – 99% of protein. Gelatin had been reported to have the ability to swell, soften and adsorb approximately 5-10 times its water weight to form a gel. Based on previous studies, gelatin had a molecular formula of $C_{102}H_{151}N_{31}O_{39}$ [42;43;44].

The production of gelatin from fish processing by-products, particularly skin, and bone, was an interesting topic that had gained a lot of attention. The use of skins and bones of fish as a source of this protein could minimize waste and provide value-added fishery products due to its properties, qualities, and relatively harmless natural characteristics. Furthermore, the valorization strategy of fish by-products through novel product development could lead to more sustainable utilization of marine resources, with higher economic benefits [45].

In the preparation of gelatin, the addition of dilute acid or base to animal collagen caused cross-cutting of protein bonds, where the structure became broken, and the pieces dissolved in water. These pieces of water-soluble protein chains were referred to as gelatin. In principle, its production from an acid-treated precursor was known as Type A. Meanwhile, the process of making gelatin derived from an alkali-treated process was known as Type B gelatin. The difference between these two processes was in the immersion process. Because of its less covalently cross-linked collagen, type A gelatin was the most often reported kind made from fish skin material, and acidic treatment was thought to be the best approach for this raw material. Type B gelatin was often produced through an alkaline-based process. The acid process was economically preferable compared to the alkaline process because the immersion in the acidic type was relatively shorter. The production of gelatin from the skins and bones of fish using type A was carried out conventionally through three main steps. The first step was the preparation of raw materials, followed by the conversion of collagen to gelatin, with subsequent purification, drying, and powdering [46;47].

The first step comprised washing or cleaning fish skins and bones with cold water and boiling water for 1-2 minutes to remove all superfluous material. Subsequently, fat (degreasing) was removed from the skin and bone tissue through immersion in water at the optimum temperature (between the melting point of fat and the coagulation temperature of bone albumin). The optimum degreasing process involved the use of a temperature of 32-80 ºC to produce optimum fat solubility. The second step was conducted by cutting the samples into small sizes, treating them with diluted alkali solution, and washing them with an ample amount of water to eliminate any excess alkali solution, continuing the rinsing process until the wash water reached a predominantly neutral state. The process was continued by ministering the neutralized fish skins and bones with a citric acid solution to weaken, swell the collagen structure, and convert the collagen into gelatin. The samples were then washed with cold water until the solution was substantially neutral. Subsequently, a hot water extraction process was employed at around 55 °C to dissolve partially degraded collagen materials. The extraction was also carried out to control the molecular weight distribution of polypeptide chains, the amino acid composition, and the functional properties of the gelatin. A series of processes in the second step was used to produce the aqueous solubilized gelatin broths, which were not pure because they were within a complex mixture of several components containing significant amounts of moisture, salts, low fat, and other proteins. In the third step, extracted gelatin (dilute gelatin broth) was filtered, deionized, and concentrated by a cross-flow ultrafiltration (UF) membrane, followed by drying with a moisture content of approximately 10% and powdering. One of the main drawbacks limiting the application of fish gelatin for industrial use was its dark color. The color depends on the nature of the raw material extracted, irrespective of the process used. However, several studies had shown that fish gelatin products did not influence other functional properties [48].

According to previous studies, optionally, UF membrane separation could successfully concentrate mammalian gelatin solutions. This membrane technology presented a promising approach for processing fish gelatin broth. Furthermore, it served as the initial concentration step and facilitated the complete desalting of the broth through ultrafiltration/diafiltration (UF/DF) mode before proceeding to evaporation and drying. UF membrane systems produced higher-quality gelatin, making it easier to meet blending specifications. The performances of productivity, protein yield, and desalting capability were similar to those reported with mammalian gelatin liquors [49]. The application of the UF/DF mode could facilitate the separation of a broth into a retentate with a high concentration of high MW solutes and a permeate by adding solute-free pure, fresh water. The permeate produced from the process was nearly 100% free of high MW solutes. During the constant-volume UF/DF mode, an equivalent volume of solute-free, pure water was introduced into the fish gelatin broth feed, matching the volume of permeate extracted. This ensured that the final volume remained unchanged, aligning with the initial feed volume. Therefore, the UF/DF mode primarily functioned as a pre-concentration stage to obtain similar concentrations of low MW components in retentate and permeate. The DF stage was often carried out to purify the retentate by adding a DF liquid. The process was then followed by a final concentration step, where the concentration of high MW solutes in the retentate was maximized and the undesired micro solutes (calcium and salt) were discarded in the permeate. Due to the above investigations, UF/DF mode was expected to be an effective way to produce fish gelatins with high quality. Fish gelatin solution produced through UF/DF mode was referred to as concentrate, which was subsequently chilled and dried to a moisture content of approximately 10%. After drying, the sample was ground to get the required particle size and then blended to obtain the powdered form, which was subjected to quality control [50]. The dominant amino acids found included glycine (20.9%), proline (12.6%), glutamic acid (11.6%), hydroxyproline (10.5%), and arginine (8.9%) [51]. Various gelatin granular sizes are visualized in Figure 4 [52].

Figure 4. Various gelatin granular sizes (Adapted from ref. [52]).

Gelatin was considered a polypeptide (biopolymer) applied widely in food and non-food products, such as pharmaceuticals, vitamin capsules, cosmetics, photographic films, paper and paint products, and diverse technical uses. As a water-soluble polymer, it was often used as clarifying agent (beer, wine, vinegar), gel former (confectionery, gelled desserts), stabilizer (chocolate milk, yogurt), protective colloid (ice creams), a binding agent (meat rolls, cheeses, dairy products), film former (coating for fruits, meats), thickener (sauces, puddings, jellies, syrups), emulsifier (cream soups, sauces, meat pastes) in the food industry [53;54]. Several reports had shown that it was an important fibrous protein possessing numerous applications due to its unique functional properties. In the medical and pharmaceutical industry, it was used for the encapsulation of different drug products, tableting, and sugar-coated pills. Gelatin was employed in the healthy and cosmetic field for maintaining joint and bone health, preventing osteoporosis, promoting hair growth, improving nail strength and growth, and enhancing moisturizer in skincare, lotions, and face-masks, toothpaste, body lotions, shampoos, sunscreens. Furthermore, it had photographic (roll film material, X-ray picture), and technical (match industry, adhesives, paper manufacturing) applications [55]. The use of gelatin in the manufacture of various applications, such as (a) clarification of beverages and juices, (b) confectionary, (c) desserts), (d) tablet coating, (e) soft elastic gelatin capsules, (f) typical hard capsule gelatin, (g) gelatin for photographic use, (h) matches are shown in Figure 5 [56].

(a) (b) (c) (d) (e) (f) (g) (h)

Figure 5. The use of fish gelatin in the various applications : (a) clarification of beverages and juices, (b) confectionary, (c) desserts, (d) tablet coating, (e) soft elastic gelatin capsules, (f) typical hard capsule gelatin, (g) gelatin for photographic use, (h) matches (Adapted from ref. [56]).

3. ADVANTAGES AND DISADVANTAGES

Pressure-driven membrane processes, such as ultrafiltration (UF) and nanofiltration (NF), were consolidated systems to separate, fractionate, purify, and/or concentrate target compounds in food-oriented agro-industrial products. The basic properties of membrane separation systems showed several advantages, including easy and simple operations, scalability, enhanced product quality, reduced processing time, decreased waste disposal, and low operation cost (economic feasibility) [57;58]. Furthermore, membrane separation processes could be used as alternatives to traditional techniques or as a novel technology to process food-oriented agro-industrial products. The low energy requirement (environmental friendliness) was related to the absence of a phase involving the transformations of solution (nutritional value close to or better than that of a fresh product in native form) during the membrane process with mild operating conditions (low temperature and pressure) to minimize thermal damage and product degradation on thermo-sensitive products. Membrane separations were effective technology, specifically for permeated water from certain processes, which could be reused in production activities sustainably [59;60].

The major problems affecting membrane separation processes were the fouling of the membrane and concentration polarization (CP). Based on the terminology introduced by IUPAC, fouling was a process leading to loss of performance due to reversible or irreversible accumulation of suspended or dissolved particles, colloidal ions, or solute molecules on the external surface, and/or within pores of the wall. The accumulation caused a reduction in the pore size or a blockage, thereby increasing the transmembrane pressure (TMP) required to operate the process. The stabilized or final flux was more important for long-term operations compared to the initial flux. The term CP described the build-up of a boundary layer of highly concentrated solutes nearest to the membrane surface. CP caused an increase in resistance to solvent transport, leading to a change in membrane performance in terms of permeate flux (productivity) and purity (selectivity). This could create a need to increase the TMP required to operate the process, leading to increased processing time and reduced membrane life. Several studies had reported that CP served as a precursor of membrane fouling, creating severe problems in the long process time range [61;62;63]. This problem could be minimized or prevented by carefully selecting membrane properties (pore size, hydrophilicity) and filtration conditions (TMP, Flow rate). Membrane processes were also limited by higher solids content in fluid, the osmotic pressure of the retained component, and high viscosity, which caused difficulty in pumping the retentate [64;65].

CONCLUSIONS

In conclusion, the recovery of high added-value components from various food-oriented agro-industrial products in agriculture, forest, and fishery (AFF) sectors using Ultra- and Nanofiltration/Diafiltration Modes was presented in this review. The membrane separation technology used in the unit process and unit operation in chemical engineering could be classified as an athermal technology. Furthermore, it was particularly suitable for extracting valuable and desired components from sustainable food-oriented agro-industrial products. This technology offered high separation capacity and selectivity while consuming low energy, aligning with an environmentally friendly concept. The implementation allowed for optimum recovery of the target product and minimizing waste, adhering to the concept of Zero Liquid Discharge (ZLD). The ZLD concept provided an opportunity to implement membrane process technologies for food-oriented agro-industrial products. During this DF mode, the new solution phase (new buffer) continuously replaced the old buffer in the retentate, maintained product concentration and volume of the ultrafiltered or nano-filtered feed solution, removed impurities, influenced the concentration of solutes, controlled the separation process, concentrated the ultrafiltered or nano filtered feed or retentate due to removal of permeate. Due to the high costs, there was a strong motivation to apply more energy-saving processes in membrane technologies. These separation processes were considered green technology due to the non-use of chemicals and additives, thereby improving the environment and human health. Future developments on separation, fractionation, concentration, and purification processes implementing ultra- and nanofiltration/diafiltration modes were directed toward a wide range of applications to get desired compounds from various nutraceutical-oriented products to achieve more benefits. Several strategies had also been developed to mitigate the rate and extent of fouling. The application of a prefiltration technique was sometimes helpful in removing large particulate matter. The popularization and implementation of new technologies in a broad range of applications depended more on the basic study of new materials and the transition from laboratory and pilot scale studies to semi-industrial scales.

REFERENCES

[1] N. S. Wisnujati and M. Patiung, “As the Agriculture, Forestry and Fisheries Sector Still as a Potential in the Prosperity of Indonesian Society,” Agricultural Socio-Economics Journal, vol. XX, no. 4, pp. 319, 2020. DOI: http://dx.doi.org/10.21776/ub.agrise.2020.020.4.7.

[2] Maulida Nur Isnaini, Agustono, and Umi Barokah, “Identification of Agriculture, Forestry, and Fisheries Sectors in Mitigating Income Inequality in the Former Residencies of Madiun,” Agriecobis (Journal of Agricultural Socioeconomics and Business), vol. 5, no. 1, pp. 83–856, March 2022.

[3] D. Kartikawati, Darsono and M. T. Sundari, “The role of agriculture, forestry and fishery sector in the development of Malinau District (location quotient and shift share approach),” IOP Conf. Series: Earth and Environmental Science, 314, 012077, 2019, pp. 1. Doi:10.1088/1755-1315/314/1/012077.

[4] FAO and UNEP, “The State of the World’s Forests 2020 : Forests, Biodiversity and People,” Rome, pp. 3-6, 2020. https://doi.org/10.4060/ca8642en.

[5] Thanh Sang Dinh and Thi Van Pham, “Non-Timber Forest Products in Cat Tien: Local Use and Sustainable Management,” Jurnal Manajemen Hutan Tropika, vol. 29, no. 1, pp. 79, April 2023. DOI: 10.7226/jtfm.29.1.79.

[6] R. K. Isfani, S. Muhammad, A. Jamal, M. Nasir, and J. Saputra, “Supply Chain Strategy for Developing the Fishery Industry in Aceh Province, Indonesia,” International Journal of Supply Chain Management, vol. 9, no. 3, pp. 41, June 2020.

[7] Papaioannou, E.H., Mazzei, R., Bazzarelli, F., Piacentini, E., Giannakopoulos, V., Roberts, M.R., Giorno, L., “Agri-Food Industry Waste as Resource of Chemicals: The Role of Membrane Technology in Their Sustainable Recycling,” Sustainability, vol. 14, no. 1483, pp. 5, 27 January 2022. https://doi.org/10.3390/su14031483. MDPI.

[8] Wang, X., Feng, H., Muhetaer, H., Peng, Z., Qiu, P., Li, W., and Li, Z., “Studies on the Separation and Purification of the Caulis sinomenii Extract Solution Using Microfiltration and Ultrafiltration,” Separations, vol. 8, no. 185, pp. 1–2, 14 Oct. 2021. https://doi.org/10.3390/separations8100185. MDPI.

[9] S. Purwayantie and W. B. Sediawan, “Membrane Separation for Recovery of Umami Compounds: Review of Current and Recently Developed Membranes,” Journal of Environmental Treatment Techniques, vol. 8, Issue 1, pp. 390, 2020.

[10] Ibraheem, B.M., Aani, S.A., Alsarayreh, A.A., Alsalhy, Q.F., Salih, I.K., "Forward Osmosis Membrane: Review of Fabrication, Modification, Challenges and Potential," Membranes, vol. 13, no. 379, pp. 3, 26 March 2023. https://doi.org/10.3390/membranes13040379. MDPI.

[11] Antónia Teresa Zorro Nobre Macedo, Joana Filipa Oliveira Monteiro, David José Chaveiro da Silva Azedo, Elizabeth da Costa Neves Fernandes de Almeida Duarte, and Carlos Dias Pereira, "Membrane Applications for Lactose Recovering," pp. 2, 11 Nov. 2020. DOI: http://dx.doi.org/10.5772/intechopen.92135. IntechOpen.

[12] Md. N. Pervez, A. Mahboubi, C. Uwineza, T. Zarra, V. Belgiorno, V. Naddeo, and M. J. Taherzadeh, "Factors influencing pressure-driven membrane-assisted volatile fatty acids recovery and purification : A Review," Science of the Total Environment, 817, 152993, pp. 10, 2022. http://dx.doi.org/10.1016/j/.scitotenv.2022.152993.

[13] M. G. Eloffy, Dina M. El-Sherif, Mohamed Abouzid, Mohamed Abd Elkodous, Hamada S. El-nakhas, Rawia F. Sadek, Mohamed A. Ghorab, Abdulaziz Al-Anazi, and Gharieb S. El-Sayyad, "Proposed approaches for corona viruses elimination from wastewater: Membrane techniques and nanotechnology solutions," Nanotechnology Reviews, 11, pp. 10, 2022. https://doi.org/10.1515/ntrev-2022-0001.

[14] Charis M. Galanakis. "Separation of Functional Molecules in Food by Membrane Technology," Academic Press, The Boulevard, Langfond Lane, Kidlington, Oxford OX5 1GB UK,, 2019, pp. 6–7.

[15] Macedo, A., Azedo, D., Duarte, E., Pereira, C, "Valorization of Goat Cheese Whey through an Integrated Process of Ultrafiltration and Nanofiltration," Membranes, vol. 11, no. 477, pp. 2, 2021. https://doi.org/10.3390/membranes11070477.

[16] Rehmana, Y. D. Ahdaba, and J. H. Lienhard V., "Monovalent selective electrodialysis: Modelling multi-ionic transport across selective membranes," Water Research, 199, pp. 3, 12 May 2021. https://doi.org/10.1016/j.watres.2021.117171.

[17] Diksha Yadav, Swapnali Hazarika, and Pravin G. Ingole, "Recent development in nanofiltration (NF) membranes and their diversified applications", Emergent Materials, 23 Sept. 2021. Springer.

[18] Laura Rolinger, Jurgen Hubbuch, and Matthias Rudt, "Monitoring of ultra- and diafiltration processes by Kalman-filtered Raman measurements," Analytical and Bioanalytical Chemistry, 415, pp. 841–854, 18 Jan. 2023. https://doi.org/10.1007/s00216-022-04477-7.

[19] Zoheb Khan, Xiaoyan Long, Eoin Casey, Denis Dowling, and Steven Ferguson, "Development of continuous spatially distributed diafiltration unit operations," The Royal Society of Chemistry 2023. DOI: 10.1039/d3re00013c.

[20] Raak, N., Co Skun, Ö., and Corredig, M., "Transmembrane Pressure during Micro- and Diafiltration of Milk Affects the Release of Non-Sedimentable Caseins," Foods, 12, 2234, pp. 1–2, 1 June 2023. https://doi.org/10.3390/ foods12112234.

[21] Ruijie Tan and Matthias Franzreb, "Continuous ultrafiltration/diafiltration using a 3D-printed two membrane single pass module," Biotechnology and Bioengineering, pp. 1–8, 22 November 2019. DOI: 10.1002/bit.27233.

[22] Cytiva, "Handbook of Cross Flow Filtration Method," CY14739, pp. 59, 24 Feb. 2021

[23] Elorm Obotey Ezugbe and Sudesh Rathilal, "Membrane Technologies in Wastewater Treatment: A Review," Membrane, 10, 89, pp. 3, 30 April 2020. Doi:10.3390/membranes10050089. MDPI

[24] Temesgen Dinsa, Urgaya Balcha, Fayisa Benya, and Mihratu Fufa, "Performance evaluation of improved mung bean (Vigna radiata (L.) Wilczek) varieties at low moisture areas East Shewa, Oromia, Ethiopia," Journal of Scientific Agriculture, 6, pp. 1, Feb. 23, 2022. Doi: 10.25081/jsa.2022.v6.7289.

[25] Sun, W., Shahrajabian, M.H., Lin, M., "Research Progress of Fermented Functional Foods and Protein Factory-Microbial Fermentation Technology," Fermentation, 8, 688, pp. 12, 2022. https://doi.org/10.3390/fermentation8120688. MDPI.

[26] Aspiyanto, A. Susilowati and H. Melanie, "Reducing Salt from Autolysate of Fermented Mung Bean (Phaseolus radiatus) using Diafiltration-Nanofiltration (DN-NF) Mode for Quality Improvement of Savory Flavor Product," in Procedia Chemistry, 16, pp. 58–65, 2015. Doi:10.10.1016/j.proche.2015.12.022.

[27] Gifuni, L. Lavenant, J. Pruvost, and A. Masse, "Recovery of microalgal protein by three-steps membrane filtration: Advancements and feasibility," Algal Research, 51, 102082, 2020. https://doi.org/10.1016/j.algal.2020.102082.

[28] O. J. Olsen, "Operating Manual of DSS LabUnit M20," Danish Separation System AS, DK-4900 Nakskov, Denmark, Jan. 2000.

[29] Md. Habibur Rahman, Bishwajit Roy, and Md. Shahidul Islam, "Contribution of non-timber forest products to the livelihoods of the forest-dependent communities around the Khadimnagar National Park in northeastern Bangladesh," Regional Sustainability, 2, pp. 281, 29 Nov. 2021. https://doi.org/10.1016/j.regsus.2021.11.001.

[30] Y. Riswahyuli, A. Rohman, F. M. C. S. Setyabudi, and S. Raharjo, "Characterization of Indonesia wild honey and its potential for authentication and origin distinction," Food Research, vol. 4, no. 5, pp. 1671, 7 June 2020. https://doi.org/10.26656/fr.2017.4(5).105.

[31] Guerrero, M.C.S., Varghese, A., Conlu, M.T., and San Jose, D., "Non-Timber Forest Products Exchange Programme Asia (NTFP-EP Asia)," ASEAN Guidelines for Sustainable Harvest and Resource Management Protocols for Selected Non-Timber Forest Products (NTFPs), NTFP-EP Asia, Quezon City, Philippines, 2020.

[32] M. W. Warui, L. Hansted, M. Gikungu, J. Mburu, G. Kironchi, and A. S. Bosselmann, “Characterization of Kenyan Honeys Based on Their Physicochemical Properties, Botanical and Geographical Origin,” International Journal of Food Science, vol. 2019, Article ID 2932509, pp. 1, 2019. https://doi.org/10.1155/2019/2932509. Hindawi.

[33] T. E. Oskouei and M. Najafi, “Traditional and Modern Uses of Natural Honey in Human Diseases : A Review,” Iranian Journal of Basic Medical Sciences, 16, pp. 732, 2013.

[34] M. Lixandru. Properties and Benefits of Monofloral Honey, Nature Word, May 1, 2017. Available: https://www.natureword.com/properties-and-benefits-of-monofloral-honey/.

[35] M. Lixandru. Properties and Benefits of Polyfloral Honey, Nature Word, April 19, 2017. Available: https://www.natureword.com/properties-and-benefits-of-polyfloral-honey/.

[36] Nelson Honey. Honeydew honey, Nature, NCDI, New Zealand. Available: https://beeswiki.com/honeydew-honey/.

[37] H. Blackiston. The composition of Honey, 2021. Available: https://www.dummies.com/home-garden/hobby-farming /beekeeping/the-composition-of-honey/.

[38] W. T. Manley. United States Standards for Grades of Extracted Honey, The fifth issue, Washington D.C., Effective date May 23, 1985, pp.2.

[39] Itoh, S., Yoshioka, K., Terakawa, M., Sekiguchi, Y., Kokubo, K.I., Watanabe, A. The Use of Ultrafiltration Membrane Treated Honey in Food Processing, J. Jap. Soc. Food Sci. Technol. (Nippon Shokuhin Kagaku Kogaku Kaishi), 46, pp. 293–302, 1999. DOI: 10.3136/NSKKK.46.293. Corpus ID: 95605429.

[40] NHB. Consumer Attitudes & Usage Report, National Honey Board (NHB), USA, August 2019, pp.137. Available: https://honey.com/images/files/NHB-AAU-2019-Report.pdf.

[41] F. J. Marti-Quijal, F. Remize, G. Meca, E. Ferrer, M. J. Ruiz, and F. J. Barba, “Fermentation in fish and by-products processing: an overview of current research and future prospects,” Current Opinion in Food Science, vol. 31, pp. 9–16, Feb. 2020. https://doi.org/10.1016/j.cofs.2019.08.001.

[42] S. Calixto, N. Ganzherli, S. Gulyaev, and S. F. Gerstenmaier, “Gelatin as a Photosensitive Material,” Molecules, 23, 2064, pp. 2, 17 August 2018. doi:10.3390/molecules23082064.

[43] Andrey Baryshnikov, Svetlana Derkach, Nadezhda Shumskaya, Kira Rysakova, Ivan Lyzhov, and Yuri Ripak, “Collagen Isolation from Arctic Marine Organisms and Their Industrial Processing Wastes,’’ in International applied research conference : Biological Resources Development and Environmental Management, KnE Life Sciences, pp. 295–296, 2020. DOI 10.18502/kls.v5i1.6073.

[44] Behrooz Jannat, Kazem Ghorbani, Somaye Kouchaki, Naficeh Sadeghi, Ehsan Eslamifarsani, Faride Rabbani, and Samira Beyramysoltan, “Distinguishing tissue origin of bovine gelatin in processed products using LC/MS technique in combination with chemometrics tools,” Food Chemistry, 319, 126302, 28 Jan. 2020. Available: https://doi.org/10.1016/j.foodchem.2020.126302.

[45] Javier Enrione, Cielo Char, Marzena Pepczynska, Cristina Padilla, Adrian González-Muñoz, Yusser Olguín, Claudia Quinzio, Laura Iturriaga, and Paulo Díaz-Calderón, “Rheological and Structural Study of Salmon Gelatin with Controlled Molecular Weight,” Polymers, 12, 1587, pp. 1–2, 17 July 2020. Doi:10.3390/polym12071587.

[46] W. C. Shiao, T. C. Wu, C. H. Kuo, Y. H. Tsai, M. L. Tsai, Y. H. Hong, and C. Y. Huang, “Physicochemical and Antioxidant Properties of Gelatin and Gelatin Hydrolysates Obtained from Extrusion-Pretreated Fish (Oreochromis sp.) Scales,” Marine Drugs, 19, 275, pp. 2, 14 May 2021. https://doi.org/ 10.3390/md19050275.

[47] Mala Nurilmala, Heny Suryamarevita, Hanifah Husein Hizbullah, Agoes M. Jacoeb, and Yoshihiro Ochiai, “Fish skin as a biomaterial for halal collagen and gelatin,” Saudi Journal of Biological Sciences, 29, pp. 1104, 2022. https://doi.org/10.1016/j.sjbs.2021.09.056.

[48] M. Abd Elgadir, M. E. S. Mirghani and A. Adam, “Fish gelatin and its applications in selected pharmaceutical aspects as alternative source to pork gelatin,” Journal of Food, Agriculture & Environment, vol. 11, Issue 1, pp. 74 – 75, January 2013.

[49] A. Simon, L. Vandanjon, G. Levesque, and P. Bourseau, “Concentration and desalination of fish gelatin by ultrafiltration and continuous diafiltration processes,” Desalination, 144, pp. 314, 2002.

[50] GMIA. Gelatin Handbook, Gelatin Manufacturers Institute of America, pp. 5–6, Member 2019.

[51] Weishardt, “Fish skin gelatin,” 2018. Available : https://www.weishardt.com/products/gelatine/fish-gelatine/.

[52] Xiamen Gelken Gelatin Co., Ltd., Fujian, China, 2020. Available in https://m.made-in-china.com/product/60-Mesh-Halal-Food-Grade-Gelatin-Beef-Gelatin-898161135.html.

[53] Yoni Atma and Hisworo Ramdhani, “Gelatin extraction from the indigenous Pangasius catfish bone using pineapple liquid waste,” Indonesian Journal of Biotechnology, vol. 22, no. 2, pp. 85, 2017. DOI 10.22146/ijbiotech.32472.

[54] Petr Mrázek, Pavel Mokrejš, Robert Gál, Jana Orsavová, “Chicken Skin Gelatine as an Alternative to Pork and Beef Gelatines,” Potravinarstvo Slovak Journal of Food Sciences, vol. 13, no. 1, pp. 224–225, 5 Feb. 2019. Doi: https://doi.org/10.5219/1022.

[55] Noor, N.Q.I.M., Razali, R.S., Ismail, N.K., Ramli, R.A., Razali, U.H.M., Bahauddin, A.R., Zaharudin, N., Rozzamri, A., Bakar, J., and Shaarani, S.M., "Application of Green Technology in Gelatin Extraction: A Review," Processes, 9, 2227, pp. 1–4, 2021. https://doi.org/10.3390/pr9122227.

[56] Sharif R., "Gelatin : Switch back to Halal : A Mini Review," PSM Biological Research, vol. 4, issue 2, pp. 66, 29 April 2019.

[57] Hao Zhang, Jianquan Luo, Lulu Liu, Xiangrong Chen, and Yinhua Wan, "Green production of sugar by membrane technology: How far is it from industrialization," Green Chemical Engineering, 2, pp. 29, 2021. https://doi.org/10.1016/j.gce.2020.11.006.

[58] Kabay N., Shirazi M.M.A., Güler E., and Bryjak M, "Grand Challenges in Membrane Modules and Processes," Frontiers in Membrane Science and Technology, vol. 1, Article 913597, pp. 1 – 2, 18 May 2022. Doi: 10.3389/frmst.2022.913597.

[59] Enny Ratnaningsih, Reynard Reynard, Khoiruddin Khoiruddin, I Gede Wenten, and Ramaraj Boopathy, "Recent Advancements of UF-Based Separation for Selective Enrichment of Proteins and Bioactive Peptides—A Review," Applied Science, 11, 1078, pp. 2, 2021. https://doi.org/10.3390/ app11031078. MDPI.

[60] Talukder, M.E., Alam, F., Mishu, M.M.R., Pervez, M.N., Song, H., Russo, F., Galiano, F., Stylios, G.K., Figoli, A., and Naddeo, V., "Sustainable Membrane Technologies for By-Product Separation of Non-Pharmaceutical Common Compounds," Water, 14, 4072, pp. 3, 2022. https://doi.org/10.3390/w14244072.

[61] Maria Norberta de Pinho and Miguel Minhalma, "Introduction in Membrane Technologies," in Separation of Functional Molecules in Food by Membrane Technology, Charis M. Galanakis, Ed. Academic Press, 2019, pp. 15

[62] Herehau N. Blais, Karin Schroën, and John T. Tobin, "A review of multistage membrane filtration approaches for enhanced efficiency during concentration and fractionation of milk and whey," International Journal of Dairy Technology, vol. 75, no. 4, pp. 749, Nov. 2022. Doi: 10.1111/1471-0307.12884.

[63] Alfredo Cassano, Carmela Conidi, René Ruby-Figueroa, and Roberto Castro-Muñoz, "Nanofiltration and Tight Ultrafiltration Membranes for the Recovery of Polyphenols from Agro-Food By-Products," International Journal of Molecular Sciences, 9, 351, pp. 10, 2018. Doi:10.3390/ijms19020351.

[64] Macedo, A., Azedo, D., Duarte, E., Pereira, C., "Valorization of Goat Cheese Whey through an Integrated Process of Ultrafiltration and Nanofiltration," Membranes, 11, 477, pp. 1, 2021. https://doi.org/10.3390/membranes11070477.

[65] El Batouti, M., Alharby, N.F., Elewa, M.M., "Review of New Approaches for Fouling Mitigation in Membrane Separation Processes in Water Treatment Applications," Separations, vol. 9, no. 1, pp. 1–2, 2022. https://doi.org/10.3390/separations9010001.

[66] Jam, F.A., Khan, T.I., Zaidi, B., & Muzaffar, S.M. (2011). Political Skills Moderates the Relationship between Perception of Organizational Politics and Job Outcomes.

[67]] Khan, T. I., Akbar, A., Jam, F. A., & Saeed, M. M. (2016). A time-lagged study of the relationship between big five personality and ethical ideology. *Ethics & Behavior*, *26*(6), 488-506.

[68] Khan, S., Jam, F. A., Shahbaz, M., & Mamun, M. A. (2018). Electricity consumption, economic growth and trade openness in Kazakhstan: evidence from cointegration and causality. *OPEC Energy Review*, *42*(3), 224-243.

DOI: https://doi.org/10.15379/ijmst.v10i3.1576