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Current and Future Application for Pressure-Driven Membrane Separation Technology in the Marine-based Resources Processing: A Review

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Abstract: Nowadays, membranes are a superior engineering technology, and their utilization has been extensively applied in the desalination of seawater and brackish water, water purification, food and beverage, pharmaceutical, biotechnology. The fast growth of innovation and development of membrane science and technologies has resolved the emergence of pressure-based membrane separation technology as a valuable, viable, and non-thermal separation unit operation in chemical engineering. This review focuses on a state-of-the-art implementation in membrane separation technology to various product commodities in the marine-based resources, particularly on the fundamentals of pressure-driven membrane separation processes, formation of thin-film composite (TFC) membrane, classification of membrane configurations, the applications of the pressure-driven membrane, benefits, and drawbacks, and prospects. In this review, pressure-driven polymeric membrane separation techniques including microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and hyperfiltration (HF), or reverse osmosis (RO) can be used to explore and implement marine-based resources, such as freshwater from seawater or brackish, surimi by-product, fish gelatin, fish sauce (moromi), and algae. Exploration of potential marine-based resources using a membrane-based separation process has great significance. The application of pressure-driven membrane separation technology on marine-based resources has a novelty to be applied in Indonesia. This review gives an overview of a sustainable approach to convert and recover marine-based resources into valuable products to achieve green and environmental-friendly based technology.

Keywords: pressure-driven membrane, non-thermal separation, permeate, retentate, marine-based resources.

混合資源和相互作用模式對偏遠地區可再生能源管理的影響

摘要：如今，膜是一項突出的工程技術，其利用已廣泛應用於海水和微鹹水淡化、水淨化、食品飲料、製藥、生物技術等領域。膜科學技術創新和發展的快速增長，解決了壓力膜分離技術作為化學工程中一種有價值的、可行的、非熱分離單元操作的出現。本綜述側重於對海洋資源中各種產品商品的膜分離技術的最先進實施，特別是壓力驅動膜分離過程的基本原理、薄膜複合(TFC)膜的形成，膜結構的分類，壓力驅動膜的應用，優點和缺點，以及未來的前景。在這篇綜述中，壓力驅動的聚合物膜分離技術包括微濾 (中频)、超濾 (超濾)、納濾 (NF) 和超濾 (高频)

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或反滲透(反滲透)可用於探索和實施海洋資源，例如來自海水或鹹水的淡水、魚糜副產品、魚明膠、魚露(莫罗米)和藻類。使用基於膜的分離過程探索潛在的海洋資源具有重要意義。壓力驅動膜分離技術在海洋資源上的應用在印度尼西亞具有創新性。本綜述概述了將海洋資源轉化和回收為有價值的產品的可持續方法，以實現基於綠色和環保的技術。

关键词：压力驱动膜、非热分离、渗透、滞留、海洋资源。

1. Introduction

Nowadays, human beings are beset by global problems for fundamental development challenges, such as high population growth, fast economic progress, the severe shortages of adequate water, food supplies and security, and energy, substantial degradation of natural resources, advancement in industrialization, degradation of environmental quality, global climate change, degradation of the ocean, and hunger and poverty [1]. Indonesia is the world’s largest archipelagic state with roughly 17,000 islands spread over nearly 6,500 km from east to west sprawling between Asia and Australia continents and stretching out between the Indian and Pacific Oceans. This area is recognized as the global center of marine biodiversity incorporating all or parts of six nations – Indonesia, Malaysia, the Philippines, Papua New Guinea, Solomon Islands, and Timor-Leste. Indonesia has a total maritime area of around 5,800,000 km^2, a total land area of more than 1,919,300 km^2, and the longest coastline (the second-longest coastline in the world after Canada) 95,181 km. [2]. Besides, Indonesia is one of the land-oriented maritime countries with ocean ecosystems in the world possessing a tremendous richness of marine natural products (MNPs). MNPs are derived from various marine-based resources, such as fishes, shellfish, mollusks, univalves, cephalopods, crustaceans, marine cyanobacteria, dinoflagellates, sponges, soft corals, micro-and macroalgae (seaweeds), and mangroves, which contribute to economic and research development [3]. Indonesia is the world’s second-largest producer of marine-based commodities producer in the world after China, with wild capture fisheries (6.412.068 tons), aqua/marine capture (6.981.505 tons), and carrageenan seaweeds productions (9.918.455 tons) in 2019. If managed well, fisheries are natural assets that provide a flow of present and future economic returns. According to the FAO (2018), global fish production amounted to 177.8 million tons in 2019, and it is expected to reach 196.3 million tons by 2028 [4].

Indonesia has not developed in exploring both biological and non-biological maritime sectors with substantial economic potential yet. These products require continuity to be suitable products to consume and increase the added value of the products. Therefore, Indonesia should give extra attention to manage and explore its marine-based resources by implementing separation and purification technology. It will improve, maintain, and develop the quality of human life, public welfare, country prosperity, and environmental health and reduce global energy use. Recent technological advances in polymer manipulation as membrane architecture have created a new opportunity for efficient, viable, and valuable separation unit processes and unit operation in chemical engineering as non-thermal and green technology with environmentally-friendly concepts [5].

The emergence of polymeric membrane-based technology plays a vital role in marine-based resource processing, which will probably also become even more critical in the future. Utilizing polymer membrane-based technology to explore marine sources represents one of the potential challenges. Membrane-based technology has been applied in tailor-made marine products to increase added value in scientific, innovative research and development and in new environmentally friendly and clean (green) downstream technology to contribute to Indonesia’s economic growth. However, appropriate separation methods should be considered to improve and increase the added value of the marine-based sources in contributing to nutrition and economic development. As a result, guaranteeing high selectivity, high productivity, and high resolution to fulfill the separation and purification technology is still a limiting factor [6].

This review summarizes the recent and future developments of membrane-based separation process technologies employing pressure-driven force, such as Microfiltration (MF), Ultrafiltration (UF), Nanofiltration (NF), and Hyperfiltration (HF) or Reverse Osmosis (RO). Furthermore, this review aims to apply pressure-driven membrane technologies in the marine-based products areas, including desalination and purification of seawater and brackish water, surimi by-product, fish gelatin, fish sauce, and algae. Moreover, it is related to pressure-driven membrane separation processes, thin-film composite (TFC) membrane, membrane configuration, and benefits and drawbacks of applying membrane technology. Moreover, the information in this review is also given on the fundamentals of pressure-driven membrane separation processes, formation of thin-film composite (TFC) membrane, classification of membrane configurations, benefits and drawbacks, and prospects.

2. Fundamental Pressure-Driven Membrane Separation Processes

The word membrane comes from the Latin word “membrane,” which means thin skin. The membrane has been extended to describes a flexible thin-film sheet with finer mesh or smaller pores. It acts as a selective boundary between two phases to separate physically among tiny and ultrafine particles, even molecules of different sizes and other particular active semi-permeable properties. A feed stream contacting the membrane’s surface is divided into a permeate and a retentate in a pressure-driven membrane separation process. The pressure difference between the feed stream and the permeate sides under atmospheric pressure acts as the driving force to transport the solvent through the membrane to control the desired hydrodynamic flow and wanted components. The selected products can either be in permeate, retentate, or both streams, which differ in their characteristics and composition of components [7].

The application of a driving force across the membrane will control the transfer rate of solutes or solvents through a membrane matrix that provides a basis for classifying membrane-based separation processes, such as the differences in pressure, temperature, concentration, and electrical field. Meanwhile, the size and shape of the solute molecules will control the rate of solute rejection. The pressure-driven membrane, namely MF, UF, NF, and HF or RO, is applied in chemical engineering separation unit operation. The pores size of a typical membrane is so tiny that pores can only be measured in the scale of Angstroms (0.1 nm or 0.0001 μm). Consequently, the operation pressure difference must force and transport components in the feed stream via the selective active semi-permeable membrane. The pressure applied differs depending on the ability of pores size of the semi-permeable membranes. As a result, the pore sizes (or molecular weight cut-off, MWCO in terms NF and HF or RO) of membranes decrease in the order from MF to HF or RO, and operational TMP values increase from MF to UF to NF to HF or RO [8].

Based on pores size, operation pressure and mechanism of separation, the membrane processes can be classified as MF (0.1 – 10 μm, 1 – 6.2 bar, sieving), UF (0.01 – 0.1 μm/2,000 – up to 500,000 Da., 1 – 10 bar, sieving/size exclusion/particle capture), NF (approximately 0.001 μm/200 – 2,000 Da., 20 – 40 bar, sieving/size exclusion/size-selective/ion-selective sieving), and HF or RO (0.1 – 1 nm/< 200 Da., 30 – 100 bar, diffusion-solution). MF membrane is applied to separate colloidal or fine particles, bacteria, microorganisms, individual algae cells, exopolysaccharides (EPS), yeast cells, turbidity, and haze in the approximate linear dimensions in the range of 0.02 μm – 10.0 μm or particles with molecular weights (MWs) over 200,000 Dalton (Da.) from suspensions [9]. In the UF membrane, the water (pure solvent), sugars, inorganic salts, and small molecules pass through the pores within the membrane as they permeate. Meanwhile, macromolecules (proteins and peptides), colloidally dispersed substances, biomolecules, and enzymes range from 300 to 500.000 Da. They are retained on the membrane surface as retentate (concentrate) [10]. NF is a beautiful pressure-driven membrane separation process that has been characterized into a situation with performance (separation capabilities) between UF and RO and exhibits features of both. If the solvent flows through the selective active semi-permeable membrane (polymer matrix) by convective flow through pores, it is stated as UF.

Meanwhile, if solvent flows through the polymer matrix (membrane) by diffusive, it is expressed as HF or RO. Due to their structural features, the separation mechanism of NF membranes involves separating aqueous solutions of monovalent anion salts and some di- and multivalent anion salts, amino acids, more significant organic compounds (sugar) and dextran with MW in the range of 200 – 1,000 Da. Besides, NF membranes have a dense active layer that contains charged functional groups on their surface. Consequently, NF membranes selectivity for multivalents ions is not based only on the sieve effect (size exclusion, size-selective, steric hindrance) but also on ion-selective due to electrical interactions between the charged (Donnan exclusion) NF membrane and the ions in the aqueous solutions. HF or RO membrane enables to separate, retain, and reject most ionic solids whose size is less than 0.1 – 5 nm or particles with below 350 Da. up to 99% of salts from water.

Regarding HF or RO, this technique uses a dense semi-permeable membrane, highly permeable to water (pure solvent) and highly impermeable to microorganisms, colloids, dissolved salts, and organics. It means that RO processes work against chemical potential difference (osmotic pressure). For this reason, the operation TMP applied in HF or RO usually is much higher than the osmotic pressure on a solution in contact with a semi-permeable membrane [11].

3. Formation of Thin-Film Composite (TFC) Membrane

The development of functional polymeric membrane and membrane structures in implementing membrane-based separation technology suited with high functionality (permeability) and good separation capability (selectivity) for the desired substances is an urgent and essential requirement for a successful separation membrane. The permeability is inversely proportional to the thickness of the ultrathin membrane as a selective active layer. Both properties can be combined as a multilayer thin-film composite (TFC) membrane. TFC membrane commonly consists of an ultra-thin, less porous, highly selective surface layer on top as a particular active layer (Polyamide, 0.1 – 0.3 µm in thickness) cast on top of microporous support layer (finger-like structure or porous, highly permeable) as polysulphone or polyethersulphone layer with 20 – 50 µm in thickness and reinforcing fabric (non-woven web) layer for supporting the physical structure (100 – 200 nm in thickness) to form the actual semi-permeable membrane. This layer has to maintain its integrity and mechanical stability under operation. The microporous support layer formed will provide the structural integrity required to support the top, ultrathin dense layer. It supports the layer as a cushioning spacer between the membrane and fibers of the reinforcing fabric. The non-woven web layer serves only to keep the selective layer providing the required mechanical stability for the whole membrane structure to operate and withstand under high-pressure compression and temperatures and does not affect membrane transport properties [12]. Until now, most functional polymer membranes is dominated by TFC membranes, particularly polyamide membranes due to membranes physiochemical properties, such as functionality (permeability, selectivity), and applicability (mechanical, chemical, and biological stability), hydrophilicity, thermal resistance, resistance to pH changes, and chemical inertness. Schematic of reaction mechanism and preparation of TFC membrane of polyamide with high membrane performance [13], the structure of asymmetric membrane with three layers stated as TFC membrane [14], and products of flat sheet TFC membrane [15] are illustrated in Figures 1A, 1B and 1C, respectively.

4. Classification of Membrane Configuration

Pressure-driven membrane separation technology has developed concerning the functional membrane polymer materials and appropriate modules. The vast majority of separation membranes are based on functional polymers (polysulphone, polyethersulphone, cellulose acetate, polyamides, fluoropolymer). According to the geometric shape, position in space, and the amount of membrane surface to feed solution flow and permeate applied, membranes can be configured commonly into four main types of modules, such as flat sheet (plate-and-frame), spiral wound, tubular, and hollow fiber [16].

The flat sheet (plate-and-frame) module is the most straightforward configuration consisting of two endplates, a support plate with a flat sheet membrane on both sides, and suitable spacers. A plate-and-frame stack is assembled by the number of membrane sets required for a given membrane area installed with sealing rings and two endplates. The permeate that passes through the membrane is collected in permeate spacer; meanwhile, the retentate leaves the module by the other side of the feed spacer. The benefits are linked to the fact that the user can easily modify or replace the membranes according to the solutions filtered. The most common configuration of a membrane module in NF or HF (RO) membrane is a spiral wound module. This module based on membranes structure consist of a flat sheet membrane, feed spacers, and permeate spacers forming the membrane layers. Further, the membrane layers are wrapped together with feed and permeate channel-spacers of woven materials around the perforated central collection tube, creating the spiral shape that permits the feed liquid to pass while permeating moves toward the center and into the product tube. As a result, the feed flows axially within the cylindrical module parallel to the central collection tube, whereas the permeate flows radially towards the main collection tube. Therefore, the need for a significant pretreatment is recommended [17]. Tubular membranes modules are tube-like structures assembled inside porous steel, ceramic or plastic tubes with an internal diameter larger than 1 cm in general. A single module comprises several tubes that can be arranged with a gasket or assembled in a monolithic module, a particular type of ceramic module. Tubular membrane modules consist of a minimum of two tubes, in which the inner tube is the membrane tube, and the outer tube is the shell. The feed solution is pumped inside tubes, while the permeate flows through the porous wall and is collected in the module housing [18]. Hollow fiber modules consist of thousands of long, porous bundles of hollow fibers ranging from as little as 50 µm to over 500 µm outer diameter assembled in place in a pressurized PVC shell. To ensure the sealing of the module, fibers and the housing are generally glued by an epoxide. Hollow fiber modules can have a shell-side feed configuration where the feed passes along the outside of the fibers and exits the fiber ends. Hollow fiber modules can also be used in a bore-side feed configuration where the feed is circulated through the fibers [19]. Hollow fiber filtration works on the same principle as tubular and capillary configurations but utilizes a small tube diameter, allowing for flexibility. They are only used when the feed stream is relatively clean or has received a very effective pretreatment (seawater desalination) due to a high fouling tendency [17]. Configuration modules of a flat sheet (plate-and-frame), spiral wound, tubular, and hollow fiber were illustrated in Figure 2 [18]. The desirable characteristics of a membrane configuration are compactness, uniform velocity distribution, high degree of turbulence at the retentate side to minimize fouling and promote mass transfer, easy cleaning and maintenance, and low cost per unit membrane area. In addition, three main parameters characteristics attribute to a membrane's economic utility for a particular application. The first is permeability (membrane flux) which dictates the amount of membrane (productivity) required or the capital investments. The second is its selectivity, which directly affects the degree of separation, recovery, and purity, and indirectly affects membrane area. The third is the life of the membrane, maintenance, and replacement cost [19].

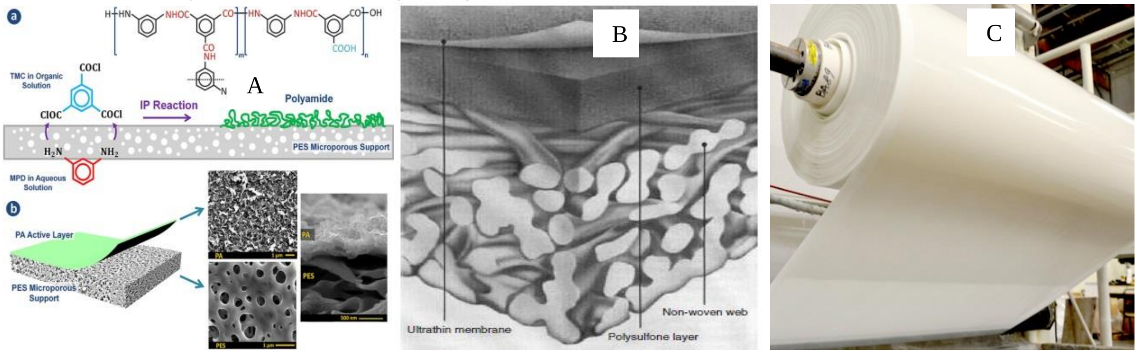

Fig. 1 [A] (a) Reaction mechanism and (b) preparation of polyamide thin film composite (TFC) membrane [13], [B] structure of asymmetric membrane with 3 layers as TFC [14], and [C] products of flat sheet TFC membrane [15]

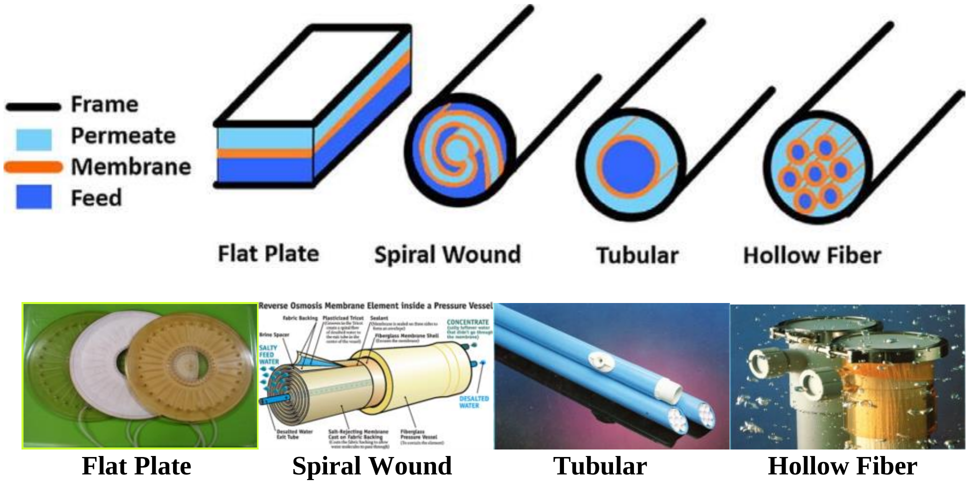

Fig. 2 Classification of membranes configuration for flat sheet, spiral wound, tubular and hollow fiber or Geometric shape and configuration of membranes module [18]

5. Application of Pressure-Driven Membrane Technology

Membrane separation technology occupying an essential place among separation technologies can overcome many drawbacks of conventional separation technologies and stands out as alternatives to traditional processes for the chemical, pharmaceutical, biotechnological, and food industries [20]. The pressure-driven membrane separation technology can be implemented to achieve the different purposes of the separation process, such as clarification, purification, fractionation, concentration, due to their adaptability and selectivity. The main product of membrane separation systems may be permeate (purification) or retentate (concentration). Therefore, a breakthrough in exploring potential marine-based resources through membrane-based separation technologies is vital and sustainable for future processes. It converts marine-based resources into valuable products and recovers useful and target products to achieve green and environmentally friendly beneficial economic [13]. The recent developments and application of the pressure-driven membrane processes of MF, UF, NF, and HF (RO) in molecular separation, clarification, fractionation, or concentration in marine-based products are distinguished in practice, including desalination of seawater and brackish water, recovery of valuable components from the by-product of surimi wash-water, fish sauce (moromi), fish gelatin, and algae.

5.1. Desalination of Seawater and Brackish Water

Water is the source of life and an essential element for human existence, and good health maintenance is consumed daily. Therefore, the quality of water is vital for human beings. In recent years, water has become essential for us to think about effective water use and protect water from pollution. The primary need and availability of clean, fresh water sources are constantly becoming a significant problem for ships (cruise ships, megayachts, research and supply vessels, military vessels during the long voyage, offshore drilling platforms). All ships are installed with a freshwater production unit, which produces large amounts of fresh and clean potable water from seawater or, in some cases, from brackish water that meets the hygiene standards. The core of the freshwater production unit is HF (RO) technology which converts seawater or salty water into fresh, clean drinking water through semi-permeable membranes by applying high operation pressure. Seawater is pumped through the membranes system for separating a solution containing dissolved materials into two streams. An aqueous feed stream containing these materials comes into contact with one side of a semi-permeable membrane. When a sufficient level of pressure is applied to this solution, molecules of pure solvent (water) are forced through the membrane. Since the dissolved materials do not pass through the membrane, a separation occurs. The result is potable freshwater consumed by passengers and employees on board. With the HF (RO) membrane technology, microorganisms and germs are held back with the salt molecules. Generally, membrane technology applied in ships is a complete and integrated system, including pretreatment, the membrane system (desalination), and post-treatment. The unique aspect of using HF (RO) technology in the military is that a standard design must own the ability to treat and handle all potential water sources without any modification to the system. The driving principle behind the military sea-based water program is that sea-based military forces of Indonesia must be able to locate, purify, store and distribute from any available potential source to drinking, cooking, washing, shower and water closet, laundry, and even running other necessary machinery. They all use freshwater as a cooling medium for routine sail activities through the standard Reverse Osmosis Water Purification Unit (ROWPU) system. The raw water may be fresh, brackish or seawater, that may be highly polluted, turbid, or contaminated by nuclear, biological, chemical (NBC) warfare agents.

Drinking water supplies must meet the stringent field water supply standards established by the military services and must be approved by military medical authorities [21]. Cruise ships, megayachts, research and supply vessels, a military vessel, and offshore drilling platforms have to provide an adequate supply of safe water for drinking, washing, preparing food, supplying recreational water, such as pools and spas, fire control, dishwashers, laundry, air conditioning, boilers, deck washing, toilets, and refrigeration. Baruna Jaya VIII, one of the research ships owned by the Indonesian Institute of Sciences (LIPI), has applied Seawater Reverse Osmosis (SWRO) technique as a mobile water purification unit. This technique can convert seawater to freshwater with 6 – 7 m^3/day to meet laboratory and consumption needs every day. Launched in 2017, the Naval Vessel of the Republic of Indonesia named with the prefix KRI Bima Suci (945) as gradually replacing KRI Dewa Ruci is the Indonesian Navy's newest and largest sail training ship for navy cadet during a long expedition. Seawater reverse osmosis systems are built for highly inconsumable water sources like the sea, removing enormous amounts of salt and other contaminants from them. By rejecting salts, particulates, and other pollutants from carrying on, water molecules are only permitted freely through the membrane's tiny pores. Fresh and purified water emerges on the permeate side and is ready to be collected, whereas salt, particulates, and other contaminants are discharged into a drain from the concentrate side. Providing clean water that is sufficient in quantity, acceptable, and sustainable is vital in supporting operational activities in offshore drilling platforms. The availability of clean and fresh drinking water with a conductivity of < 30 µS/cm (permeate) is essential to have a safe and carefree stay aboard ships and on offshore platforms [22].

5.2. Recovery of Valuable Components from the By-Product of Surimi Wash-Water

Oceans possess diversified and exploited food resources (fishes, seaweeds, crustaceans, mollusks). Sea products can be eaten fresh, frozen, dried, or transformed. One major fish transformation consists of a washed fish pulp with added cryoprotection expressed as the "surimi base." Surimi originating from the word "sir-ree-mee," is a Japanese word that refers to unique seafood products. The process comprises sorting and cleaning, heading/gutting, washing, deboning/ mincing, washing, refining, dewatering, mixing and stabilizing with cryoprotectant to prevent the denaturation and aggregation of protein in natural concentrated fish paste during storage, shaping/packaging, freezing, cold storage, and frozen surimi. Surimi can be formed into various shapes and block to resemble artificial seafood, such as crabs, shrimps, lobsters, and other shellfish known as Frozen Surimi [23].

The surimi manufacture requires vast amounts of freshwater for sorting and cleaning, heading/gutting and washing, refining, and dewatering. Surimi wash-water with a high concentration of organic substances, particularly water-soluble protein (0.5 – 2.3% protein), composed of sarcoplasmic proteins with small amounts of myofibrillar proteins, lipids, mineral and pigment, and fine particles of water-insoluble myofibrillar proteins. The surimi wash-water is often discarded into the plant’s waste stream to prevent environmental problems and loss of valuable components, especially soluble proteins with good functional and nutritional properties that could be fractionated and recovered. Surimi manufacturing produces various by-products, such as viscera (15 – 30%), fish heads (14 – 20%), fish skeletal (17%), skin and bone (8 – 10%), wash water (14 – 16%), and refiner discharge (4 – 8%). The significant components of fish mince/surimi processing by-products are enzyme, lipid, and protein in viscera, extractives, meat and bone-in fish skeletal, collagen and mineral in skin and bone, soluble protein in surimi wash water, and collagen in refiner discharges [24].

Fish industrial wastewaters in general and surimi wash water in particular show low protein concentration that makes classical processes of protein recovery economically not feasible. An alternative to methods like evaporation or spray-drying is pressure-driven membrane separation using nanofiltration (NF). NF, a present state of the art in membrane processing in fishery and surimi manufacture, offers considerable potential to be an integral part of a clean technology process. In general, the application of NF could be classified into three areas, including removing monovalent ions from wastewater, separating between ions with different valencies, and separating low- and high MW compounds. Fish and surimi wash-water, regarded as a pollutant, contain water-soluble protein compounds (average of 6 g of soluble protein/L). Thus, NF emerges as one of the pressure-driven membrane separations applying to push the surimi wash-water across membrane pores by cross-flow. Proteins with a molecular weight more prominent than the pore size are retained by the membrane and concentrated without involving a phase change due to sensitive biological substances (proteins, enzymes, hormones) and using no heat.

Meanwhile, water and particles with sizes smaller than the NF membrane pores pass freely through it. By removing water and smaller particles, solids more extensive than the NF membrane pores are recovered. Thus, functional properties of recovered soluble and insoluble proteins are preserved and present utilization and new product opportunities for surimi processing. Therefore, it seems that MF, UF, and NF can be considered powerful techniques to purify, fractionate and concentrate in the recovery of specific proteins, respectively, and reduce environmental problems [25].

5.3. Fish Sauce (Moromi)

Consumption of fermented food has contributed to several ethnic and geographical identities since the prehistoric period. Fermentation releases a range of flavourful taste compounds and aroma substances. However, the fermentation of marine-based products is still quite limited. Therefore, viable efforts to develop current research trends and future challenges associated with their various marine-based fermented products are performed. Fermentation of marine-based product sources, such as fish, shellfish, and mollusks, is one of the different fermented products that are often considered to lead to some of the most flavourful products in cuisines worldwide [26].

In a standard fermentation, fish sauce is a protein hydrolysate generally obtained by mixing three parts of anchovies (fish or krill) materials with one piece of sea salt and allowing the mixture to stand overnight. The fish are then placed in a large vat and covered with concentrated brine causing salt to pull water from the fish cells by osmosis. Endogenous proteinases induce protein hydrolysis in fish muscle and digestive tract and proteinases produced by halophilic bacteria. Fermentation under natural conditions degrades the fish protein spontaneously to produce liquid containing a large number of solid soluble (savory or umami amino acids, nucleotides) by proteinases enzyme for ten months if incubated in the sun or 12 months if in the shade, causing hydrolysis. Various volatile compounds are formed during fermentation, including acids, carbonyls, nitrogen-containing compounds, and sulfur-containing compounds. They are believed to be responsible for the distinct aroma and the delicious taste of the fish sauce. When the fermentation is completed, the fish sauce is filtered through a cloth into a large jar to separate filtrate and residue. This filtrate constitutes fish sauce as fermented broth, a translucent amber-colored liquid with 8 - 14% digested proteins and about 25% salt. The most critical taste sensation of fish sauce is salty and umami taste and particular aroma of ammoniacal, meaty, and cheesy odor attributes. Among the sensory qualities, the distinct smell is the crucial attribute reflecting fish sauce quality. Fish sauces contain about 20 g/L of nitrogen, of which 80% is in the form of amino acids. In addition, fish sauce is also a rich source of essential amino acids, especially lysine. Fish sauce is an excellent source of vitamin B12 and many minerals such as sodium (Na), calcium (Ca), magnesium (Mg), iron (Fe), manganese (Mn), and phosphorus (P) [27].

The result of fish sauce fermentation tends to dilute and contain a complex liquid (bacteria, bacteria debris, proteins, polysaccharides, amino acids, and inorganic salts), substrate components. By-products are often labile, heat-sensitive, and degradable compounds. Therefore, the separation method must be carried out under mild conditions, no phase change, the absence of additives, and low energy consumption. There are possible reasons to consider implementing pressure-based membrane separation technologies (MF, UF, NF, and HF or RO) as an alternative to recover desired components from fish sauce [9].

The implementation of the purification and separation methods of fish sauce emphasized on microfiltration membrane. The aim is to recover valuable and target compounds with better uniformity level of particles size as permeate (protein, umami flavor, salt) with smaller MW solutes ranging approximately 500 – 2500 than pores size of MF membrane. In addition, it can provide products with superior clarity, maintain most of the flavor and nutrition substances, remove the microorganisms to stop the fermentation, stabilize the fish sauce, and improve the visual appearance of permeate. Meanwhile, unwanted compounds as by-products of fermentation with larger MW solutes than pores size of MF membrane will be retained or rejected on the membrane surface as retentate (concentrate). Fish sauce is a vital source of delicious liquid condiments and a staple ingredient in various cuisines cultures and is often synergistic. Fish sauce is familiar in Southeast Asia and the coastal regions of East Asia, such as China, Japan, Korea, and Taiwan, and other parts of the globe, to turn abundant small marine life into a nutritious, stable, and flavorful food resource. Fish sauce is known as fish ketchup (Indonesia), budu (Malaysia), bagoong (Philippines), nam pla (Thailand), and Phú Quốc, nuöc mâm and Phan Thiết (Vietnam), bya yay and ngapi (Myanmar), teuk tre (Cambodia), padaek (Laos). In China, fish sauce is called yúlù (fish dew) and yeesu, gyoshō (uoshō) or fish soy sauce and eojang (Korea), aekjeot made of fermented anchovies (the Korean Peninsula), eoganjang made of fermented godori (young chub mackerels) (Jeju Island, South Korea), shottsuru (Japan), colombo-cure (Pakistan and India), pissala (France), garos (Greece), and garum (Italian). Raw materials (Krill, small fry of fish) in preparation of fish sauce and examples of fish sauce products from Thailand (Mai Nam Pla), South Korea (Myeolchi-aekjeot) and Vietnam (Nước chắm) are displayed in Figures 3a, 3b, 3c, and 3d, respectively [28].

Figure 3. (a). Krill (small fry of fish), (b). Mai Nam Pla (Fish sauce, Thailand), (c). Myeolchi-aekjeot (anchovy sauce, South Korea), and (d). Nước chắm (fish sauce, Vietnam) [28]

5.4. Fish Gelatin

Gelatin or gelatine (from Latin: *gelatus* meaning “stiff” or “frozen”) is a heterogeneous mixture of denatured, biodegradable, and water-soluble polypeptides and protein, natural macromolecule, and pseudo-plastic materials having properties of translucent or transparency, colorless or slightly yellow substance, odorless, tasteless and colloidal flavorless, and gummy (when moist) and brittle (when dry). Gelatin has average molecular weights (MWs) ranging between 20 and 250 kDa and a molecule formula of C102H151N31O39. It consists of approximately 88% of protein, 10% of moisture, and 1 – 2% of salt and can retain more than 50 times its weight of water within its gel structure [29]. According to the technical aspect, gelatin is a water-soluble protein prepared from collagen extraction present in various animal by-products. The enormous sources of gelatin come from porcine skins and bones prohibited in both Judaism and Islam communities. In contrast, gelatin from cattle is unaccepted in Hinduism communities as animal by-products from slaughterhouses. The competitive market data indicates the increased preference and demand for fish gelatin as an alternative to mammals because of socio-cultural, religious aspects [30].

Fish, chicken, and bovine slaughtered according to Islamic teachings have been looked upon as a possible alternative to gelatin produced from halal sources. One practical option to prepare gelatin is fish processing by-products, such as skin and fins, backbone and thorns, and fish that are damaged or inedible or unsuitable for human consumption or further processing. These by-products are mostly discarded without recovery intention and have a considerable negative ecological impact. Therefore, the valorization of fish processing by-products could positively impact the economic viability of the fishing industry through the production of value-added ingredients attained using practical technological answers. In addition, fish processing by-products could be a novel source of value-added compounds like proteins (gelatin), peptides, and amino acids. The data show that fish by-products produce muscle 49%, head 18%, backbone 10%, viscera 7%, skin 6%, and off-cuts 10% of the entire body weight, respectively [31].

The preparation of gelatin from fish skins comprises cleaning the skins with cold water to remove all extra material substantially and cut into small sizes and treating with dilute alkali solution. Plenty of water is used to remove excess alkali solution until the washing water is substantially neutral. Then, the fish skins are added with the citric acid solution. Next, the citric acid-treated fish skin is washed with cold water until tepid. The neutralized citric acid-treated fish skin is then extracted with hot water at approximately 55 °C to solubilize partially degraded collagen materials and influence the amino acid composition and molecular weight distribution of polypeptide chains' functional properties the gelatin. Optionally, the aqueous solubilized gelatin solution (the gelatin broths) was filtered through a cellulose/diatomaceous earth plate & frame filter and de-ionized (demineralized) using a cationic-anionic resin bed to remove impurities. The concentrated solution was then ultrafiltered to remove any unwanted substances and eliminate impurities more minor than the nominal cut-off of the membrane and sterilized (280 – 290 °F, 8 – 12 seconds) to get approximately 15 – 35% of concentration. Then, the process was finally chilled, dried, grounded, blended to obtain gelatin powder [32]. Although fish gelatin mainly consists of protein, it is still not pure. Instead, it is present within a complex mixture of many components containing significant amounts of moisture, salts, small amounts of fat, and other proteins. However, according to experimental studies, its chemical composition largely depends on the source of the collagen-containing material, the pretreatment, and extraction conditions. Besides, one of the main drawbacks of fish gelatin limiting its applicability for industrial use is often the dark color caused by the raw materials from which it is extracted. However, it does not influence other functional properties [33].

According to few works, UF membrane technology is applied in fish gelatin by concentrating the gelatin broth and desalting in the ultrafiltration - diafiltration (UF-DF) mode before evaporation and drying. The performances are similar to those reported with mammalian gelatin liquors in terms of productivity, protein yield, and desalting capability [34]. By applying the UF-DF method, a fish gelatin broth can be separated into a concentrate with a high concentration of high molecular weight solutes and a permeate, nearly 100% free of high molecular weight solutes new solvent or buffer. In constant-volume UF-DF, the volume of added solvent to the feed material during membrane filtration is equal to the volume of permeate removed, making the final volume of fish gelatin broth the same as the starting volume. As a result, the function of UF-DF is as a preconcentration stage achieving similar concentrations of low molecular weight components in the retentate and permeate, a diafiltration stage to purify retentate by adding a diafiltration liquid. A final concentration stage is to maximize the concentration of high molecular weight solutes in the retentate, and the undesired microsolutes are discarded in the permeate. Due to the above investigations, it is expected that UF-DF may be an effective way to produce fish gelatins with upper quality [35]. Five dominant amino acids in the amino acid composition of fish skin gelatin include glycine (20.9%), proline (12.6%), glutamic acid (11.6%), hydroxyproline (10.5%), and arginine (8.9%). Fish Transforming Industry (FTI) (a) [36], edible fish skin gelatin granule (Food Grade) (b) [37], and fish gelatin powder (c & d) [38] are illustrated in Figures 4a, 4b, 4c, and 4d, respectively.

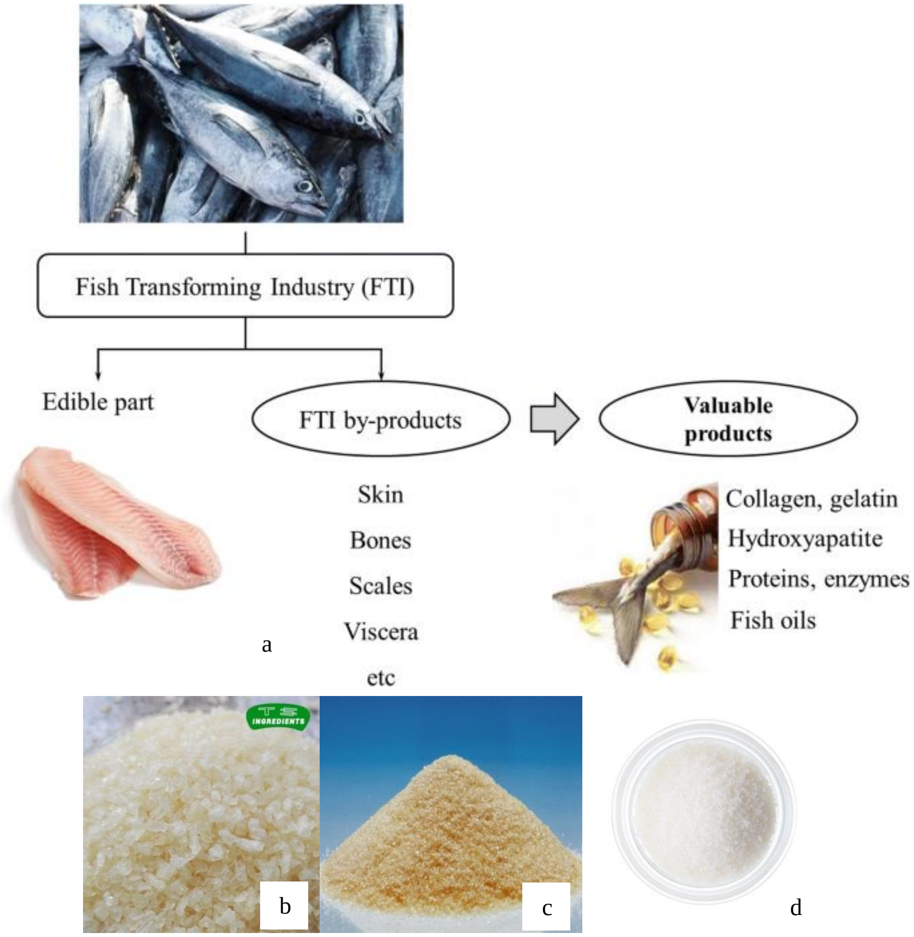

Fig. 4 (a) Fish transforming industry (FTI) [36], (b) edible fish skin gelatin granule (Food Grade) [37], (c) and (d) fish gelatin powder [38]

In food, medical and pharmaceutical, and cosmetic products, halal gelatin, such as fish gelatin, is a concern of the Muslim community [39]. In particular, fish gelatin possesses a melt-in-the-mouth unique functional property in the range of approximately 10 – 30 °C resulting in the melting of gelatin gels in the mouth. Therefore, it is suitable for a wide range of applications in food. In the food applications, gelatin is used as gel former (confectionery), protective colloid (ice cream), a binding agent (cheeses), clarifying agents (fruit juice), thickener (sauces, soups, puddings, dairy products), emulsifier (sauces, cream soups, meat pastes, dairy products), process aid (microencapsulation of colors), stabilizer (chocolate milk, cream fillings, yogurt, icings), an adhesive agent (candy). On the other hand, the most significant proportion of gelatin in the medical and pharmaceutical industry is mainly used for hard and soft gelatin capsules (soft gels), tableting, tablet coating, granulation, and microencapsulation. In health and cosmetic applications, gelatin is mainly implemented to maintain joint and bone health, prevent osteoporosis, promote hair growth, improve nail strength and development, and improve moisturizer in skincare, lotions, and face masks toothpaste, face creams, body lotions, shampoos, and sunscreens. In addition, gelatin is applied as a component in a photographic developer during the processing of the exposed film material (roll film, X-ray picture) and other technical applications (match industry, adhesives, paper manufacturing, box making, paint, printing process) [40].

5.5. Algae

Algae, referred to as aquatic “plants,” ranges from unicellular cyanobacteria to complex multicellular forms and generally possess chlorophyll. They can be classified into two groups, microalgae, and macroalgae (commonly known as seaweeds). Although micro-and macroalgae have common characteristics, such as photosynthesis as unicellular sunlight-driven organisms, they differ in various ways from their sizes to phylogeny [41]. Microalgae cells have a small size range between 5 and 50 µm. They are negatively charged and form stable suspensions in the culture media. Their negative surface charges prevent aggregation from maintaining its stability in the dispersed state [42]. More than 50,000 different types of microalgal species can survive, grow and reproduce fastly, and breed with short cycles in almost all spectrums of ecological habitats ranging from freshwater (lakes, ponds, and rivers) and brackish water to seawater even industrial/domestic wastewater. They can tolerate a wide range of temperatures, salinities, and pH values in various environmental conditions and contaminants. They require only water, light intensity, and CO2 to grow alone or in symbiosis with other organisms. However, only 30,000 species have been studied. Algae are broadly classified as Rhodophyta (red algae), Phaeophyta (brown algae), and Chlorophyta (green algae). Depending on the species, algae biomasses are rich in proteins, amino acids, lipids, carbohydrates, vitamins (A, B, C), trace elements (copper, iron, zinc), pigments, and nutritionally valuable components various biologically active substances. Microalgae-based biomasses are considered promising, attractive, economical sources of renewable, sustainable production of nutraceuticals, pharmaceuticals and vitamins, food and food supplements, fertilizer, animal/aquaculture feeds, feedstock, and bioplastics biofuel (non-food products), cosmetics, pigments, and medicine [43].

The membrane separation process is one of the physical separation methods, which has long been applied in different stages of microalgae upstream process and downstream process (biorefinery). The upstream process includes cultivation and biomass harvest; meanwhile, the downstream process (biorefinery) draws applications relevant to algae production. First, microalgae culture can be conducted in photobioreactors and open ponds through various methods, such as batch, semi continues, and continues. Next, the harvesting process is performed after microalgal biomass concentration generally achieves ~10^7 cells/mL. Techniques currently used in microalga harvesting to produce microalgae concentrate are conventional methods, such as filtration, centrifugation, electromagnetic separation, chemical flocculation, flotation, and sedimentation, or a combination of these processes. Both these techniques have a major limiting factor due to the usage of chemicals, and they require a sufficiently long time to get desired microalgae concentration. In line with an advance in separation technology, the application of membrane technology to concentrate and harvest microalgae cells starts to be implemented by removing large volumes of water. One of the most rational membrane-based separation technologies to harvest by focusing on various micro-sized microalgae cells is ultrafiltration (UF). The capability of appropriate and suitable UF membrane technique can harvest with the efficiency of ~99% cells of culture to obtain a thick paste of algal biomass as concentrate via desired solid-liquid separation. While the particle-free permeates as the growth liquid media, leaving the algae to concentrate biomass can be reused and recycled to culturing microalgae.

On the other hand, drawbacks to the UF membrane in harvesting and concentrating microalgae is a flux decline due to fouling on the membrane surface and into the membrane matrix. Therefore it requires frequent change of new membrane so that this drawback might contribute significantly to its processing cost. However, UF based on the size-exclusion method may be helpful and scalable for microalgae harvesting as problems in membrane fouling can be minimized or prevented [44]. Microalgae cultivation process, microalgae dewatering, and protein fractionation using membrane-based separation technology are illustrated in Figure 5 [43].

6. Benefits and Drawbacks

The potential benefits of pressure-driven membrane technology as separation unit operations in chemical engineering instead of other conventional unit operations and unit processes in chemical engineering are related to the unique separation principle, such as the transport selectivity of the membrane. Pressure-driven membrane separation technology consumes lower energy (energy for the pumps) associated with the absence of phase change in solution (athermal) during the process to prevent the deterioration of processed material. Meanwhile, a sufficient amount of thermal energy is required to evaporate the water mass in the evaporation process, which implies energy-consuming phase changes to affect the final product's physical and chemical characteristics. Another benefit of the pressure-driven membrane is completing the process at low, ambient, and high temperatures to concentrate heat-sensitive products and maintain and preserve their natural properties. It also raises the final product's yield, improves desired product quality without heat and chemical treatments, and eliminates fewer amounts of remaining wastes or unwanted components due to a negative impact on product quality. In addition, pressure-based membrane separation systems are environmental-friendly, easily upscaling and downscaling, efficiently implementing, simple integration into existing unit separation processes (hybrid processes). Furthermore, it has lower capital, lower installation operating and maintenance costs, and more reliable performance. Pressure-based membrane separation systems related to competitiveness and economic consideration give a compact and modular system, which occupies less space than conventional treatment systems. To remove bacteria, cold pasteurization or sterilization employing appropriate and suitable MF membranes instead of thermal treatment like sterilization are more economical in terms of energy consumption and shelf-life extension of products to create a green image for the processing procedure [45].

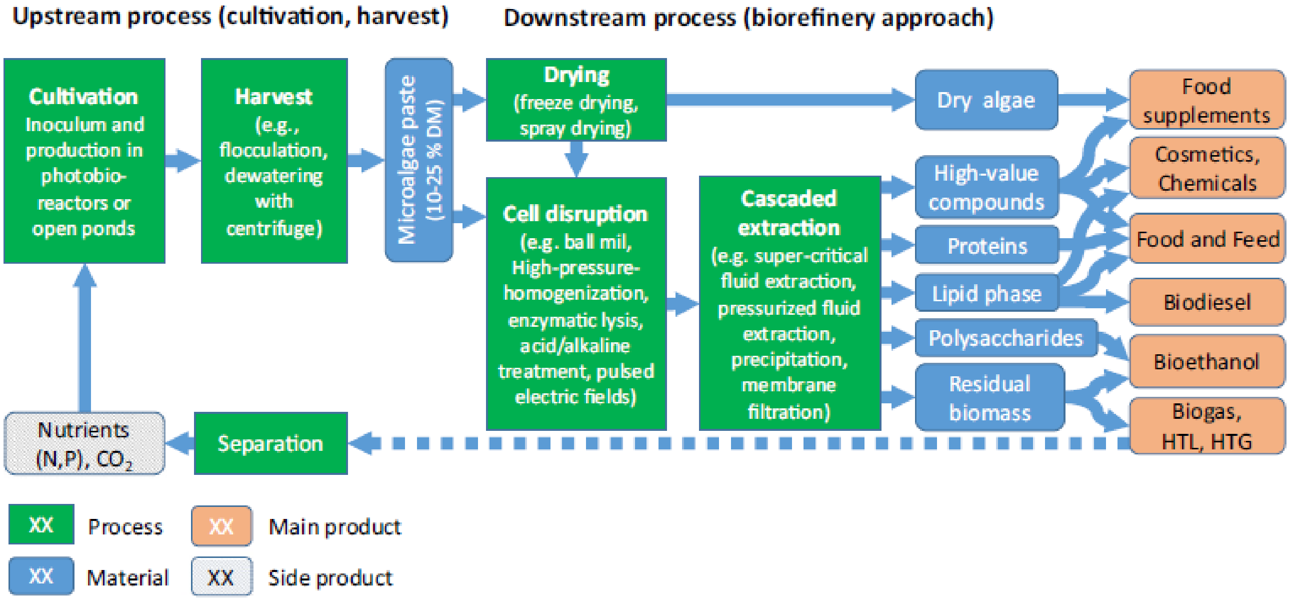

Fig. 5 Integrated fuel and food production with microalgae [43]

The sugar industry requires efficient methods in clarifying the raw sugar cane juice to improve the quality of the clarified juice and reduce or eliminate chemicals (lime). The mud produced in a conventional clarifier is sent to a rotary vacuum filter to separate dirt and filtrate containing the sugar. However, the filtrate from the rotary vacuum filter contains impurities, such as small particles of bagasse, soluble substances (protein, polysaccharides), a small number of flavonoids, polyphenolics, and organic acids present in the juice. These impurities contributed to the dark brown color of the liquid, which will not introduce the evaporation station directly and should be returned to the conventional clarifier. In sugar mills, determining the production of juice of consistently high clarity, light, and color through the clarification process is challenging. The membrane separation process promises sugar cane juice with superior quality, better transparency, lower viscosity, and noticeable color removal. Ultrafiltration of clarified sugar cane juice can be performed employing a polymeric membrane separation system in a flat sheet or spiral wound modules. The permeate from the UF membrane experiences an increase of 1.5 – 3 units of juice purity compared to the rise of 0.5 – 1 unit being achieved in the liming-sulphation process [46].

Advances in separation-based science and technology are critical to developing sustainable major chemical processes, including distillation, drying, evaporation, extraction, adsorption, absorption, membranes, crystallization, and physical property-based operations (floatation, screening). Decrease and energy consumption efficiency in chemical separation processes was identified as one of the grand challenges in sustainability in chemical separation. Distillation, drying, and evaporation are several examples of high energy-intensive processes that are well established in the separation process. They are thermally driven based on the heats-of-vaporization of the components and account for 49%, 11%, and 20% of the separations energy consumption. Meanwhile, non-thermal separation processes, such as extraction, adsorption, absorption, membranes, crystallization, and physical-based operations tend to require low energy consumption and improve product quality to realize any significant reductions in the energy intensity of the chemical separation processes due to the heat of vaporization of at least on the component. However, none of these processes accounts for more than 3% of separations energy consumption. Figure 6 is illustrated examples of thermal separation processes (high energy use) and non-thermal separation processes (low energy use) [47]. On the other hand, one of the critical drawbacks to the pressure-driven membrane process is limited by both higher solids concentration associated with the viscosity and the osmotic pressure in the concentration process. The concentration process aims to reduce water content, minimize storage, transport, and distribution costs, and improve and preserve product stability (nutritional, aroma, flavor, freshness) by decreasing water activity. Low mass transfer rates obtained with full macromolecules and the high viscosity make the retentate’s pumping difficult. Gelatine is a protein that is produced from bone and tissue from slaughterhouses. By UF, gelatin is possibly concentrated up to about 10 % of total solids. Above these values, the pressure-driven membrane process becomes technically and economically unfeasible [48].

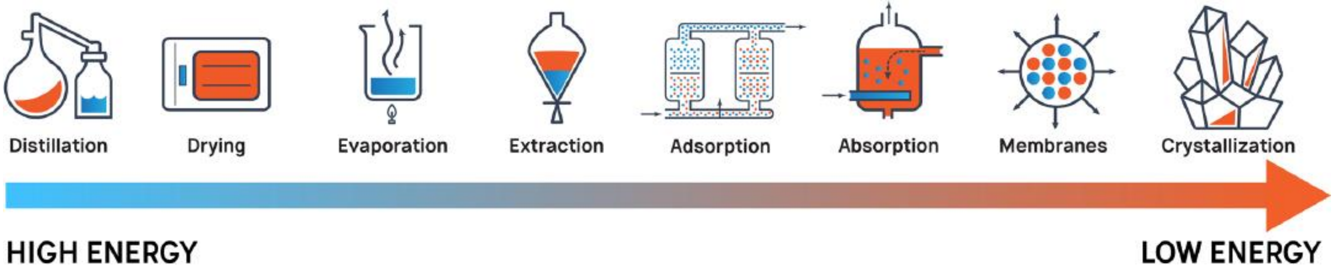

Fig. 6 Examples of thermal separation processes (high energy use) and non-thermal separation processes (low energy use) [47]

Although pressure-driven membrane separation processes provide numerous benefits for marine-based resource processes, the current primary challenges associated with membrane separation technology are fouling and concentration polarization. Fouling refers to the irreversible accumulation of particles on the membrane surface or into the membrane pores. Fouling can cause various phenomena that can modify separation behaviors, such as permeability and selectivity. In this case, the permeate flux can only be re-established by interrupting the process for membrane cleaning. Since membrane cleaning involves both operation and maintenance costs, it affects the operational performance of the membrane (dropping in permeability as productivity, changing in selectivity), shortens the membrane lifetime, increases energy requirements, and alters permeate quality. Thus, fouling is often a severe problem that causes premature replacement of membranes in various industries [49]. Fouling can be caused by multiple colloidal particles, inorganic particles (minerals), microorganisms (biofilms, bacteria, viruses) or macromolecules (proteins, carbohydrates, fats), salts (monovalent or multivalent), etc. However, this undesired phenomenon affecting productivity as well as membrane selectivity can be related to the nature of the feed (pH, concentration, ionic strength, component interactions), the nature of the membrane (pore size, pore size distribution, porosity, hydrophobicity, roughness, charge), and operation conditions (temperature, transmembrane pressure/TMP, cross-flow velocity). Although there is no universal definition, concentration polarization can be described as the reversible formation of dissolved or suspended particles near the membrane surface. This phenomenon can usually be controlled by adjusting hydraulic parameters (speed of recirculation, pressure) to re-establish the permeate flux [5].

Many authors have studied and assessed the mechanisms of classic membrane fouling, classified as complete pore blocking (a) occurs when particles are more prominent than the membrane's pore size due to a complete pore obstruction using sealing (blocking) due to denaturation. Internal (standard) pore blocking (b) occurs when particles penetrate inside the membrane on the pore walls. The resulting smaller size of pores of the membrane, the membrane's permeability is reduced. In this case, membrane resistance increases as a consequence of pore size reduction. Partial (intermediate) pore blocking (c) takes place when solid particles or macromolecules that at any time reach an open-pore might seal it. Finally, cake filtration (d) happens when particles or macromolecules which do not enter the pores form a cake layer on the top membrane surface. The overall resistance is composed of a cake layer resistance and a membrane resistance which causes the thickness to increase. The formation of the cake layer occurs as starting in the construction of concentration polarization [50].

7. Conclusions and Future Perspectives

As one of the maritime countries with ocean ecosystems, Indonesia should explore and develop marine-based resources. This matter is expected to contribute to the national economic growth sector and the country's prosperity. The separation and purification strategies adopted by chemical engineering sometimes follow the conventional processes due to operation familiarity for several decades. Implementing pressure-driven membrane separation technologies in the sector of marine-based sources, reclamation, recycle, and reuse of wastewater produced by marine-by products have attracted attention and have addressed the pressing worldwide concerns. Future developments from separation and purification techniques using membrane-based process technology are directed toward using various marine-based products and sources or by-products to obtain value-added goods. The continuity of separation and purification techniques depends on many factors, including higher selectivity, energy efficiency, economic viability, environmental safety and compatibility, and sustainability. According to the concept of zero waste, innovative green technologies that are environmentally benign have significant advantages over conventional methods. Among other benefits, these techniques are increasingly implemented at an industrial scale because of their low energy consumption, separation process at lower and ambient temperatures, and in most cases, without phase changes on the organoleptic properties of the valuable compounds. However, membrane fouling limits the implementation of the pressure-driven membrane separation processes. Still, this technology preserves and enhances the quality and process efficiency, minimizing functional properties losses of the desired compounds and processing by-products to obtain value-added goods explored from marine by-products and sources.

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