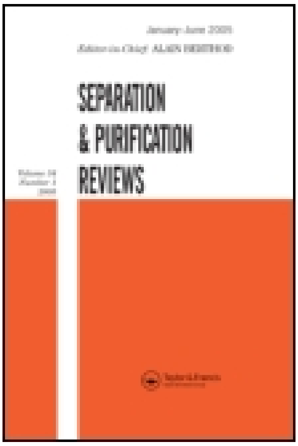

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Purification of Aquacultural Water: Conventional and New Membrane-based Techniques

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Removing solids is an essential task when recirculating water an aquaculture system. Dissolved solids production directly from particulate solids as well as by fish is a function of time. These contaminants can indirectly affect the fish both biologically and physically. The flaws of conventional water treatment on seawater aquaculture systems are reviewed in this paper. Then a new technology for membrane processes is described to remove fine particles and dissolved matter in addition to performing gas transfer.

KEYWORDS *Membrane, seawater, fine particles, dissolved matters, ammonia*

INTRODUCTION

Fish are a large source of protein in the human diet (1). Fish are also the cheapest protein-based food. Therefore, fish that are used as food must be healthy and toxin-free. However, pollution of the sea, lakes and rivers by industries is getting worse. Therefore, fish husbandry is currently boosted by building recirculating aquaculture systems (RAS). An aquaculture system operates as a batch system in which water culturing is reused by circulating through more than one mechanical treatment unit sequentially, and thickened solid waste is regularly discharged.

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Solids retained in an aquaculture system are a major problem for fish growth and health. The main source of solids comes from faeces and fish feed waste. These solids are present in particulate and dissolved forms. Particulate solids have been conventionally removed by sedimentation; however, a major portion of solids with a low density close to that of water do not settle. Hence, they remain suspended in water. Suspended solids can be effectively removed by a filter. About 60 μm or greater of suspended solids are often removed by a microscreen, which is a popular filter utilised in the aquaculture industry. However, conventional treatment systems have mostly ignored fine particle (<60 μm) removal that have contributed more to dissolved matter production, such as ammonia-nitrogen, dissolved carbon and phosphorus.

The relatively larger solids undergo leaching, thus the dissolved matter increases over time. Biological treatment is a well-known technique to remove dissolved matter in a freshwater aquaculture system. However, bacteria may lose their microbial function in saline wastewater resulting from osmotic effects. Pathogen nourishment, local eutrophication and the conventional biofiltration may deplete the water's dissolved oxygen content. Therefore, removing the fine particles efficiently to prevent water quality reduction is one of the goals of this review paper. The paper also introduces a useful membrane technology for ammonia removal and water oxygenation.

CONVENTIONAL WATER TREATMENT IN AN INTENSIVE AQUACULTURE SYSTEM

The sources of the organic materials and nutrients in an RAS are feed wastage, faecal solids and detached bacterial flocs. According to Wu (2), organic materials composed of 80–88% carbon in seawater RAS as feed are lost into the environment via uneaten feed, undigested feed residues and respiration. For nutrients, about 85% of phosphorus and 52–95% of nitrogen in the feed may be lost into the environment through excretion, feed wastage and faecal production. The faecal production in RAS is about 26–46% of the ingested feed (3), whereas the uneaten feed and waste excretion are about 11–38% of the applied feed (4–6). However, the characteristics of the metabolic and non-metabolic wastes depend on several factors, such as feed quantity, feed quality, feeding techniques, water make-up flow rate, fish culture tank hydrology and fish density (7).

Nutrient content is characterised as total nitrogen (TN) and total phosphorus (TP) in an RAS. The TN can be organic or inorganic in both particulate and dissolved forms, whereas TP is the sum of soluble reactive phosphorus, soluble organic phosphorus and particulate phosphorus. Solid particles retained in the culture tank may undergo hydrolysis and leaching to produce dissolved fractions. About 68–93% of TN and 16–70% of TP have been reported to be dissolved solids in the system (7). Some of the algal blooms increase along with dissolved nutrient concentration increases. Negative impacts on aquatic environments, such as severe reduction in water quality and fish welfare, may occur.

A Cornell-type dual drain culture tank, which is commonly used in intensive RAS, was principally designed to be self-cleaning (8–12). The volume capacity of the tank ranges from 1 to 150 m^3. This tank has a sidewall drain and a bottom-centre drain. The tank is a circular tank with a conical base where larger-sized solids generated by the fish are collected. About 78–93% of the flow containing low concentrations of solids is discharged from the sidewall outlet, while approximately 7–22% of the discharged flow with a high solid content is drained via the bottom-centre outlet (8,9). The flow with a high solid content is transported into Device I, as shown in Figure 1. Device I as listed in Table 1 concentrates and then removes the solids from the RAS as sludge. The suspended solids in the liquid mixed with the sidewall effluent are then removed by Device II. However, the fine solids as well as dissolved matters may still escape the treatments. Therefore, advanced treatments (Device III and Device IV) are then introduced to minimise the fine solids and dissolved matters to an acceptable level.

Removal of Settleable and Suspended Solids

Sedimentation and filtration are solid separation techniques in conventional aquaculture systems. Much of the literature is devoted to the sedimentation,

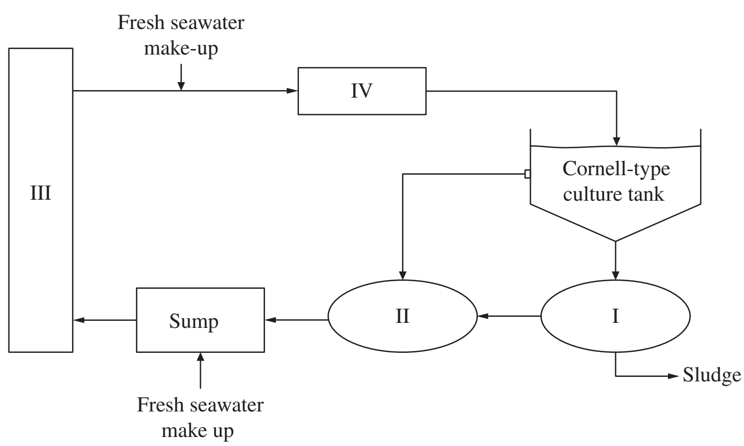

FIGURE 1 The recirculating aquaculture system conventionally reuses the treated water. A Cornell-type dual drain culture tank is designed to be self-cleaning. The high solid content is drained through the bottom-centre outlet into Device I; whereas the low solid content is discharged via the sidewall outlet into Device II. The advanced treatments by Device III and Device IV are introduced to minimise the fine solids and dissolved matters that escape the primary treatments.

TABLE 1 List of the conventional unit operations for Device I, II, III and IV, accordingly, in the recirculating aquaculture system based on Figure 1

Device	Unit operations
I	Sedimentation basin (13), stationary bowed screen (14), triple standpipe sump (9, 15), swirl separator (8, 12, 16), radial flow clarifier (8, 10), gravity thickening tank (17)
II	Trickling filter (13), fluidized bed reactor (13), microscreen drum filter (8, 9, 15, 17), propeller-wash bead filter (10, 12)
III	Pump sump (15), combination of pump sump and fluidized sand biofilter (8), fluidized bed sand filter (9, 12, 17)
IV	Air-stripping column and *low head oxygenator (8, 9, 15)

*Low head oxygenator is the most common used; the others oxygen absorbers are packed column, spray tower, U-tubes, sealed column, oxygenation cones and oxygen aspirators (18).

in which Cripps and Bergheim (7) described the principle of sedimentation in detail. The density of settleable suspended solids is greater than that of water; hence, the solids can settle and be separated from the culture water. The denser solids (1,150 kg/m^3) are composed mainly of the wheat-related heavy cellulose portion in the feed, which is digestible, whereas the lighter solids (1,050 kg/m^3) consist of fine particles as well as mucus-based flocs in the 3–5 μm range (19). Stokes' Law states that the settling velocity in a sedimentation basin is determined by the viscosity of the water and the diameter of the particles. Sedimentation includes thickening (20) and clarifying, both of which are gravitationally based processes. The primary function of thickening is to increase the concentration of a relatively large quantity of suspended solids, whereas that of clarifying is to remove a dilute slurry of fine particles to produce a clear liquid effluent. Settling velocity curves are useful information when selecting a solid removal device (21) as well as when estimating the theoretical solid removal efficiency (22).

A hydrocyclone, which is commonly known as a swirl separator, is used to remove the high specific gravity solids especially from water that is discharged directly from the culture tank (8, 12, 16). A swirl separator is a circular tank with a conical bottom section. Wastewater enters the separator tangentially, and the water spins around the central axis of the tank, generating a whirlpool effect. Inertial forces exerted on the solid particles due to the relatively slow circular velocity of the water at the whirlpool region allow the particles to settle easily. However, the particles are separated more by the effects of gravity rather than the inertial forces (23). A sufficient pressure drop in the swirl separator is required to force the solids into a discharged pipe. Gravity sedimentation is not able to concentrate all of the low specific density of fine particles (1.005–1.050), which are slightly denser than the water. These fine particles remain suspended in water, so further treatment with microscreen filtration is required.

A radial-flow clarifier is similar to a swirl separator physically, but the flow hydraulic is nothing like that of the swirl separator. A radial-flow clarifier operates by injecting wastewater into the centre of the tank. The water flows outward away from the centre, and the water velocity decreases progressively. An injector of water inlet is designed to minimise turbulent or mixing effects on the inflowing water, which can cause solids to re-suspend in the water. The larger particulate solids may settle along the radial paths in low water velocity injection. The radial-flow clarifier could minimise the amount of solids that re-suspend but has higher removal of settleable solids compared to the swirl separator (8). Moreover, the clarifier could reduce the mean particle diameter in the effluent (10) and solid loading on a microscreen filter as well as reducing the need for backwashing (8).

A rotary drum microscreen is a well known mechanised filtration unit for suspended solid removal from water (24–27). The drum is a cylindrical body that is entirely covered by a screen. A fine mesh screen is used to hold back the particles but still allows clear water to pass through. The pore size range of the mesh is usually from 60 to 200 μm (7). Wastewater enters axially at the open side of the drum and exits radially through the screen due to the effects of gravity. The level of the wastewater increases as progressive screen clogging occurs. When a sensor level is reached, a backwashing operation will begin on the screen. The backwash effluent that has a high solid concentration is treated prior to being discharged into the environment (9,28,29).

Fluidised sand filtration has recently been commercially applied to large scale aquaculture systems because filter construction only requires a low cost reactor vessel, a plumbing system and sand. Sand filtration is defined as the process of separating solid particles from water that passes through porous sand media; the water fills the pores between the sand media particles. Fine particles in the water will clog the pores or attach to the sand media surface. The particles may consist of nitrogenous, phosphorus and carbonaceous particulates. The wastewater from a culture tank is pre-filtered by Device I and II before it enters the fluidised sand filter. A biological process takes place in the filter via a process known as bio-filtration. Pfeiffer et al. (12) constructed a fluidised silica sand filter that was able to remove over 65% of 23–55 μm particles. In the same study, the concentration of 55–105 μm particles removed was found to have increased to about 96%, which is similar to the observation reported by Davidson et al. (17) in which the removal of 10–20 μm particles increased about 55–60%. These results are due to biofilm sloughing. In an aquaculture application, however, the fluidised sand filters are most suitable for dissolved solid removal rather than for particulate solid removal from water (30).

Removal of Dissolved Solids

Suspended fine particles (i.e., sizes of <60 μm) and dissolved components that escaped from Device II (i.e., a microscreen or any other filters) are further treated by either chemical or biological agents (31). Rueter and Johnson (32) reported that chemical oxidation by ozonation has improved suspended solids removal up to 55%, a figure which depended on wastewater hardness and the initial suspended solid concentration. Summerfelt et al. (33) found that 35% of the mean concentration of total suspended solid was reduced by ozonation. Krumins et al. (34) reported the total organic carbon, turbidity and total ammonia nitrogen were also significantly reduced by ozone treatment. However, Tango and Gagnon (35) found that bromate and other brominated compounds, which are the byproducts of ozonation used in marine RAS, are significantly harmful to fish health and carcinogenic to humans. Therefore, biological processes are more popular for the treatment of suspended fine particles and dissolved solids.

Biological filtration is most often utilised for treatment of dissolved solids in the aquaculture industry. Aerobic bacteria can oxidise biodegradable organic matter, ammonia and nitrite-nitrogen (17, 36–39), whereas anaerobic bacteria can reduce nitrate-nitrogen (40, 41), phosphorus (42, 43) and sulphate (44, 45) by denitrification (46). Generally, biofilters are classified into two systems: suspended growth (47) and attached growth. A suspended growth system is when the substrates, such as dissolved organic carbon, ammonia and nitrite-nitrogen, are mixed with suspended microbes in fish culture tanks. In an attached growth system, the substrates are transported from a fish culture tank to sequential treatment units. The attached growth units can be emerged, packed, expandable and expanded reactors, as shown in Table 2.

Dissolved Gas Control

Dissolved carbon dioxide (DCO_2) is the product of fish respiration, and the amount of gas is greatly increased in water after biofiltration from the aerobic activity of bacteria. According to a stochiometric estimation (54), nitrifiers produce about 5.9 mg/L of DCO_2 by consuming 1 mg/L of total ammonia-nitrogen (TAN) and 4.6 mg/L of DO_2, whereas heterotrophic microbes utilise 1 mg/L of DO_2 to produce about 1.38 mg/L of DCO_2. In the same study, the production of DCO_2 by a fluidised sand biofilter was experimentally

TABLE 2 The reactors of attached growth systems in which the substrates are transported from a culture tank to sequential treatment units

Reactor	Example
Emerged	Rotating biological contactor (48), trickling filter (13, 49)
Packed	Plastic packed bed, submerged rock, shell filter
Expandable	Floating bead bioclarifier (51), foam filter (50, 52), upflow sand filter (13, 17, 53)
Expanded	Fluidized sand filter (12), downflow microbead, moving bed filter

measured to be 37% of the total DCO_2 produced within their salmonid RAS. However, extensive studies of DCO_2 production in seawater RAS are scarce.

Management of dissolved gases, especially dissolved oxygen (DO_2) and dissolved carbon dioxide (DCO_2), is a challenging task for intensive RAS. Gas transfer equipment strips DCO_2 as well as dissolved nitrogen (DN_2) from the bio-filtered water prior to oxygenation. Surface aerators and air-stripping packed columns are more effective in DCO_2 removal than ceramic diffusers (18) because they form water droplets to move through the air rather than air bubbling in the water. A blower is mounted to inject the air directly into a packed column. Removal efficiency of DCO_2 increases as the ratio of volumetric air to water loading rate increases (55). Packing in the column breaks up the water flow that is countercurrent to the air stream to increase the gas transfer rate. A target concentration of DCO_2 in the effluent is achieved by injecting NaOH into the column where the DCO_2 is selectively removed by chemical reaction (56).

The degassed water from the packed column with very low concentrations of DO_2 is not suitable for fish culturing. Therefore, pure oxygen must be added to the water for maintaining the minimum level at 5 mg DO_2/L (57), especially when the fish density in a system is being increased. A spray tower is utilised in aquaculture as a tool for oxygenation. The spray tower operates at atmospheric pressure in which the degassed water is directed via a spray nozzle mounted at the top of a sealed chamber that receives pure oxygen. Gas-phase axial dispersion can be seen with an insufficient pressure drop along the axis of the tower, nozzle spraying force and the force of bulk tower gas recirculation (58). This dispersion reduces the water-oxygen contact area to levels as low as in plug-flow system. However, increases of hydraulic loading and tower height could improve the spray tower's performance (59). Watten et al. (56) modified a spray tower by incorporating a packed column for DCO_2 desorption that excluded the blower systems and reduced the pumping cost.

A low head oxygenator (LHO) is another oxygen absorber that is most often utilised in intensive aquaculture. An LHO consists of distribution plates in a vertical position that establish about 5–10 rectangular chambers. The degassed water from the air-stripping column flows into a collection trough and is distributed as droplets through a perforated plate into the rectangular chambers by gravity. Pure oxygen is injected into the side of outermost chamber and flows through the individual chambers serially in a direction normal to the fallen water droplets. Gas transfer may occur when the oxygen stream contacts the water droplets. However, oxygen absorption is mostly due to splashing and subsequent bubble entrainment when the water droplets fall into the receiving pool of water (60). The bubbles penetrate the receiving pool more deeply as the diameter of holes on the perforated plate become larger (61), thereby allowing more oxygen to be absorbed in the water.

FINE PARTICLE ACCUMULATION AND AMMONIA-NITROGEN PRODUCTION

Commercial fish feeds composed of about 35–50% crude protein, 7–30% crude fat, 2–5% crude fibre, 9% ash, 2% added minerals, about 15 g/kg TP and about 70 g/kg TN are usually used in intensive aquaculture system. The fish excrete faecal solids as waste products of metabolism from the anus. These solids as well as uneaten feed in an RAS may damage and irritate the fish gills by fouling. Solid buildup in an RAS may exert stress on and reduce the immune system of the cultured fish, which can cause disease outbreaks. High concentrations of suspended solids provide an environment that may harbour and nourish pathogens. The biological oxygen demand (BOD) is increased by the respiration of the pathogens and excessive eutrophication effect. Dead organic algae matter may also increase the turbidity of water. An increase of the BOD reduces the amount of dissolved oxygen that is required for fish and causes an acidic environment that is harmful to fish growth.

The formation of fine particles from larger particles in an RAS is a function of time. Therefore, the larger particles (>250 μm) should be removed as rapidly and efficiently as possible to protect aquatic species from diseases and minimise production costs. In addition to the particles leaching, the larger particles that reside and accumulate in the system will be broken down into fine particles due to the pumping effect (62) and water turbulent forces (63). Removal of the fine particles is costly because it is difficult to separate them from the filter meshes. A large quantity of backwash water is required for a filter mesh as small as 40 μm, thus filter meshes that are ≥70 μm are preferable (31). However, Chen et al. (64) showed that >95% of the suspended particles in an RAS were <20 μm. Couturier et al. (65) developed a mathematical model that expresses the rate of fine particle accumulation, dC_{fine}/dt, in an RAS:

$$V \cdot \frac{dC_{\text{fine}}}{dt} = P_{\text{fine}} + Q_{\text{in}}\left(C_{\text{fine,in}} - C_{\text{fine}}\right) \tag{1}$$

where V, Q_{in} and $C_{\text{fine,in}}$ are total water volume, makeup water flow rate and concentration of fine particles in the makeup water, respectively. The mass production of fine particles P_{fine} is defined as:

$$P_{\text{fine}} = (1 - \eta) f_{\text{TSS}} F \tag{2}$$

where η, f_{TSS} and F are the fraction of total waste solid captured by the solid removal equipment, mass of waste solids produced per unit mass of feed, and feed rate, respectively.

Dissolved ammonia-nitrogen increases as a result of particle mineralisation and leaching, which can deteriorate the culture's water quality. The production of ammonia-nitrogen, P_{Ammonia}, in an aquaculture system is estimated as (29):

$$P_{\text{Ammonia}} = 0.092F \cdot f_{\text{protein}} \tag{3}$$

where f_{protein} is the protein fraction in the feed. Fish excrete unionised ammonia directly from their gills in water. Ammonia excretion by fish depends on the species, body weight, water temperature, water salinity and feeding, as well as the size of the rations (66–70).

TOXICITY OF AMMONIA IN AN AQUACULTURE SYSTEM

Ammonia or total ammonia-nitrogen (TAN) exists as unionised ammonia, NH_3, and ionised ammonia, $NH_4{}^+$, in water. NH_3 is more toxic to fish than $NH_4{}^+$ because it is non-polar and readily soluble in the lipids of biological membranes (71). Seawater species have been reported to be more sensitive to ammonia toxicity than freshwater species, and the average of mean acute toxicity values for 32 freshwater species is 2.79 mg NH_3/l compared with 1.86 mg NH_3/l for 17 seawater species (72). The concentration of ammonia in the blood plasma of the fish (73–75) and mortality rate (76,77) increase as the ambient ammonia level in the water increases. Reddy-Lopata et al. (78) reported that the growth of juvenile abalone was inhibited when they were chronically exposed to sub-lethal free NH_3 levels. The required energy for ammonia detoxification leads to a reduction of fish growth (79,80).

The ammonia equilibrium in water is affected by environmental parameters, which include water pH, temperature, dissolved oxygen and salinity. Dissolved DCO_2 is the second most toxic excreted metabolic waste, and it can acidify the water, leading to a reduction in the proportion of NH_3 (81). Stripping the gas from water may elevate the water pH as well as the lethal concentration of NH_3 (76). Low water temperature can increase tolerance of fish to ammonia (77), and high levels of dissolved oxygen also reduce ammonia toxicity (82,83). Interestingly, Serafini et al. (84) found that a lethal concentration of 50% mortality (LC_{50}) of dissolved oxygen varied from 4.02 to 5.02 mg/L at 0.927 mg NH_3/L. Additionally, NH_3 might impair the supply of oxygen to tissues. An increase in salinity leads to a lower proportion of NH_3 (85). However, the effect of salinity on ammonia toxicity is relatively less compared to the effect of the water pH. NH_3 can be calculated as (86):

$$NH_3(\%) = \frac{100}{1 + 10^{(\mathrm{p}Ka - \mathrm{p}H)}} \tag{4}$$

where the p*Ka* is defined as (87):

$$\log Ka = -0.467 + 0.00113 \cdot S + 2887.9/T \quad (5)$$

where S and T are the seawater salinity and temperature, respectively.

THE EFFECT OF SALINITY ON BIOLOGICAL TREATMENT

Activated sludge microbes suffer a plasmolysis effect in an environment with a high salt content (>1% salt) and become inactive. High salinity strongly inhibits aerobic (88–91) and anaerobic biological treatments (92, 93). Sakairi et al. (94) reported that immobilised nitrification in seawater is about six times less than that in freshwater. Significant inhibition of an activated sludge process for fish processing saline wastewater treatment has been found at levels of NaCl over 4% (95). The nitrification rate decreases about 20% in 5% salt when compared to salt-free wastewater (96). Interestingly, ammonia oxidisers are more sensitive to the stress of salt than nitrite oxidisers (97). A significant reduction in both nitrification and denitrification was observed at salt concentrations above 2% (98). Denitrification decreased markedly as salinity increased from 0 to 10% (99). Denitrification by entrapped microbial cell immobilisation processes decreases at levels higher than 20 g NaCl/L (100). However, salt inhibition of denitrification is greater than that of nitrification at levels higher than 20 g NaCl/L (101).

Salt-tolerant microbes were recently tested for saline wastewater treatment. For instance, yeast has been found to be more efficient in chemical oxygen demand (COD) and total nitrogen removal than bacterial cultures for high salinity conditions, i.e., >25 gNaCl/l (102). The nutrient (COD, NH_4-N and PO_4-P) removal efficiency by halophiles, such as the *Halobacter halobium*, is higher than by *Halobacter*-free for concentrations between 0 and 6% NaCl (103). Aerobic and anaerobic treatments by *Halobacter halobium* (104, 105) and *Halanaerobium lacusrosei* (106), respectively, are inhibited at salt levels greater than 3%. However, removal efficiency may rely slightly upon the composition of the wastewater. Organic matter (C, N and P) removal by a sequencing batch reactor is affected much more by the changes in salinity than changes in hydraulic retention time and organic loading rate (107). On the contrary, COD removal by a rotating biodisc contactor is more sensitive to changes in the ratio of disc surface area to wastewater flow rate (A/Q) and feed COD compared to salinity (108). Aspé et al. (44) showed that the temperature effect was smaller for the saline wastewater treatment than for the freshwater treatment.

MEMBRANE-BASED TREATMENT

Membrane Bio-filtration

Membrane technology has been used as an alternative to fluidised sand and active carbon filtration beds. In membrane bio-filtration, a bio-reactor is integrated into a membrane module system, which is referred to as a membrane bio-reactor (MBR). Microbes degrade the substrates by forming a biofilm that can be mobile (109) or attached/immobilised (110) in a bio–reactor. In a mobile biofilm reactor, high-density supports are used to hold the biofilm in wastewater. The membrane, either in a flat/plate (109) or hollow fibre (111), provides an area for the biomass to attach. The nitrifying microbes are autotrophs, chemolithotrophs and obligate aerobes. Autotrophic nitrifiers fix and reduce inorganic carbon. Denitrifying microbes are chemotrophs; heterotrophs utilise organic electron donors, whereas autotrophs consume hydrogen and reduce sulphur as electron donors.

The applications of MBR have recently received increased attention, such as in drinking water production (112–114) and urban wastewater (115), synthetic (116–119) and saline wastewater (120) treatments. Sharrer et al. (121), however, observed that as salinity increased to 8 parts per thousand, there was no nitrogen removal in MBR, even though it successfully removed >99% of TSS. Salt concentration in wastewater is an important parameter to optimise high retention MBR (122). The acclimation stage of nitrification seemed to stop in marine or seawater systems due to the changes in salinity (123). High salinity significantly affects the physical and biochemical properties of activated sludge, which could increase the microbial products and lower the membrane's permeability (124).

Membrane Filtration

In membrane filtration, a membrane acts as a semipermeable barrier to control the rate of transportation of various molecules between two liquid phases. Pressure-driven membrane processes are the most commonly used and utilise hydraulic pressure to force water molecules through the membranes. Larger particles are retained and concentrated in the feedwater. The water that passes through the membrane is known as permeate, while the retained particles are called the rejection. The membrane process includes microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO). Figure 2 illustrates the performance of the membrane process for a range of sizes of selected constituents in water. Today, membrane technology is often applied in dairy production (125), juice production (126, 127), drinking water production (128, 129), biotechnology (130–131), for example. Using membrane technology for wastewater treatment is being investigated for waters such as oily wastewater (132–134), acidic wastewater (134), dairy wastewater (135), and textile dyehouse

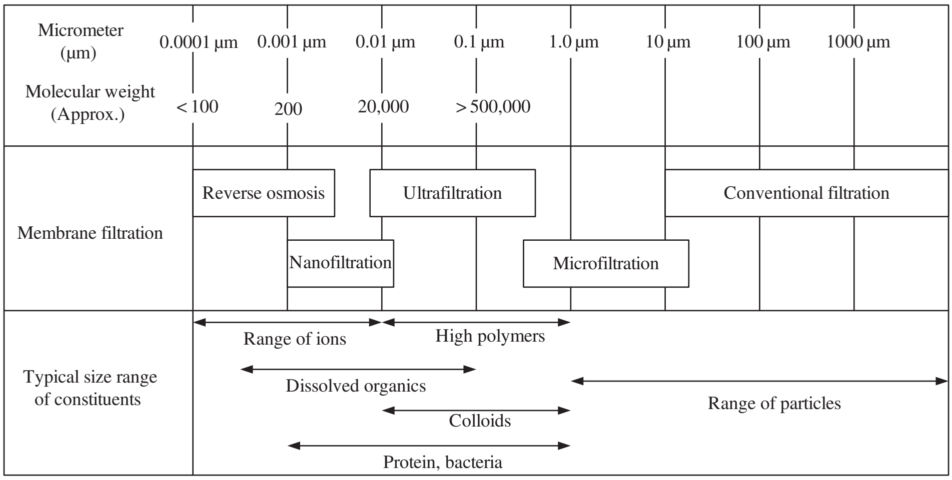

FIGURE 2 Selection of a filtration process relies on the sizes or molecular weights of the target components. The membrane separations include microfiltration, ultrafiltration, nanofiltration and reverse osmosis remove the suspended solids that escape from the conventional filters.

wastewater (136). However, the application of membrane technology in aquaculture wastewater treatment is limited.

In this paper, we attempt to introduce the potential use of membrane technology without biological consideration in seawater aquaculture water treatment, especially for fine particle (<20 μm) removal. The viscid matter present in an RAS is comprised of protein-polysaccharide complexes. It is a mucus produced by fish and is able to bind to the lighter fine particles to form micro-flocs (19). Micro- and ultra-membrane filtrations can be used to remove these micro-flocs and smaller sized particles by ultrafiltration. The membrane can be made from either polymers or ceramics. Given the limited pore size and porosity as well as high membrane resistance of its symmetric structure, Figure 3 shows why asymmetric membranes should be widely used for most applications. Wastewater flows perpendicular to a membrane surface in a dead-end microfiltration module, and only clear water (permeate) can leave the module, as illustrated in Figures 3a and 3c, whereas in a cross-flow system, as shown in Figure 3b and 3d, wastewater flows parallel to the membrane surface where both clear water and particles (rejection) streams leave the system (137). The particles form a layer of cake on membrane surface, which acts as a barrier to reduce the clear water flux in a dead-end system. An ultra-membrane is mostly operated using a cross-flow system to prevent the particles from clogging the relatively smaller pore sizes compared to the micro-membrane.

Bottino et al. (128) were able to remove total suspended solids (TSS) and microorganisms from Brugneto lake water completely with a 0.2 μm ceramic membrane. Guo et al. (129) successfully reduced the turbidity >98%

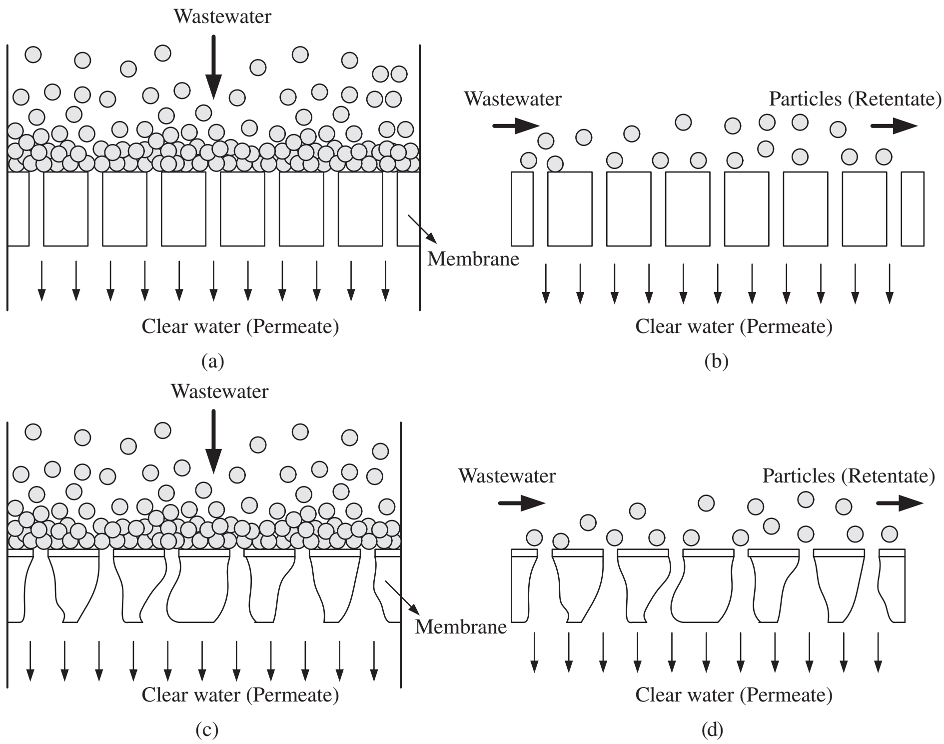

FIGURE 3 Symmetric membranes (a, b) give low amount of clear water since the membrane resistance is high as compared to asymmetric membranes (c, d). Wastewater flows normal in direction to a membrane surface in a dead end module (a, c) and only the clear water leaves the module. In a cross-flow module, wastewater tangentially flows to the membrane surface where both clear water and accumulated particles streams leave the module (b, d) (137).

from surface water that originated from Luan River and Huang River with a 4–6 nm polymeric membrane. Cross-flow filtration by a 0.5 μm ceramic membrane has reduced about 90% of suspended solids from acidic wastewater from a vegetable oil refinery (134). Orecki et al. (138) used cross-flow nanofiltration to treat the surface water collected from Miedwie Lake, from which about 94% of colour and total organic compounds and 86% turbidity were removed. A 96% removal of total phosphorus (∼80% in solid form) and about 86% removal of total ammonia from discharge aquacultural water in a sand filter using an ultra-low pressure asymmetric polymeric membrane were reported (139). Viadero and Noblet (140) utilised dead-end filtration with a 0.05 μm membrane to successfully remove >94% of TSS and 76% of BOD from synthetic aquacultural process water.

Colloids are suspended fine particles below 10 μm in size that remain continuously in motion as a result of electrostatic negatively charges. The colloidal materials in wastewater cause membrane fouling by forming a cake on the membrane surface, which results in clear water flux reduction. Bowen and Jenner (141) theoretically described membrane filtration models of colloids and fine particles by considering membrane fouling. For potable water production, Kaiya et al. (142) reported that the total organic carbon and manganese were the major fouling materials in Kasumigaura Lake, which were used as a raw water source. A hybrid process to reduce membrane fouling, combining a conventional treatment process and membrane filtration, has been recently developed. However, the water treatment performance of a hybrid system is determined by the particle size distribution in the water and the membrane pore sizes. A hybrid system of sand filtration-membrane filtration resulted in lower water quality (143) but higher membrane fouling (144) compared to coagulation-membrane filtration. Coagulation shows significantly improved permeate flux by forming larger particles from fine particle aggregation (145). Coagulants have positive charges that neutralise the negatively charged particles where the particles start to collide and agglomerate. The small membrane surface area is occupied by the larger particles, which allows a large free volume for permeate to pass through, leading to a high permeate flux. A hybrid system of coagulation-microfiltration performs better than a single system for particle (146–148) and virus (149) removals.

The hybrid process of coagulation-membrane filtration can remove particles efficiently, which is strongly influenced by the coagulant type and the membrane characteristics. The use of the right coagulants can protect the membranes from fouling inside the membrane pores and prevent the formation of the cake layer (150). Konieczny et al. (151) reported that 2.4 g Fe/m^3 of iron trichloride ($FeCl_3$) formed the particles of flocs that could penetrate the pores inside the structure of ultrafiltration membrane, which reduced the water flux. Kabsch-Korbutowicz (152) revealed that 3.59 g Al/m^3 of sodium aluminate ($NaAlO_2$) coagulant increased the pH of the water and led to the dissociation of organic matter. The smaller organic matter particulates can block the membrane pores. There have been no reports of the sizes of flocs formed by the $FeCl_3$ and $NaAlO_2$ coagulants at the amount that can cause pore blockage and membrane fouling. Nevertheless, Mijatović et al. (153) have recommended using a nanofiltration membrane with a molecular weight cut-off at 300 D rather than the ultrafiltration membranes to remove the low molecular weight organic matter.

Polyaluminium chloride (PACl) has been reported by many researchers to perform better than the conventional Al-based coagulants at particle removal under certain conditions (152, 154, 155). The flocs accumulate and form a layer of cake on the membrane surface where the porosity of the cake determines the clear water flux. The removal of organic particles relies on the different Al species in the PACl coagulant (156). The size of the flocs formed by the monomeric species Al_a is the largest compared to those formed by the intermediate polymer Al_b and solid species Al_c (157–159). As a result, the monomer alum coagulant increases the cake permeability by forming a highly porous cake on a microfiltration membrane (157, 159), which allows more water to be transported through the membrane and prevents pore blockage and fouling after rinsing the membrane.

The properties of a membrane material play an important role in adsorptive fouling and organic retention. Kabsch-Korbutowicz (154) reported that in a weak acidic solution, more hydrophobic organic matter tends to adsorb on the membrane surface. When dealing with a basic solution, the membrane preferentially adsorbs hydrated anions (colloids), thus reducing the membrane pore size and causing a decline in the water flux. In the same study, the organic removal efficiency of the relatively hydrophilic regenerated cellulose membrane was lower than that of the relatively hydrophobic polyethersulfone membrane. The denser structure in addition to the more hydrophobic membrane has been reported to remove organic matter efficiently (152, 160). However, no study has been conducted on the organophobic effect of the membrane materials on the adsorptive fouling and organic removal. Adsorption is the significant mechanism for the fouling by colloids (148, 160, 161), in which a more organophobic membrane rather than a more hydrophobic membrane may be more appropriate to describe the fouling. Dispersive adhesion is the mechanism of adsorption due to the attraction between the two materials by van der Waals forces. By using a more organophobic and hydrophilic membrane, the interaction between the organic particles and the membrane is weaker. The organic particles tend to be repelled from the membrane surface, but the water is allowed to flow through the membrane. The organophobic membranes appear more hydrophilic when the membranes are wetted. For instance, polytetrafluoroethylene and polyvinylidene fluoride are respectively, highly organophobic and partially organophobic membrane materials that might minimise the adsorptive fouling.

Critical flux is the highest stabilised permeate flux and corresponding pressure transmembrane that can overcome the additional resistance of reversible filtration. There is no particle accumulation on the membrane surface below the point of critical flux where fouling is negligible in this regime (162). The Hagen-Poiseuille model states that the operating permeate flux,J, below the critical point is pressure dependent:

$$J = \frac{\varepsilon_{\mathrm{m}} \rho d_{\mathrm{pore}}^2 \Delta P}{32 \mu \delta_{\mathrm{m}}} \quad (6)$$

where ϵ_{m} is the membrane surface porosity, ρ is the density of permeate, d_{pore} is mean pore diameter, ΔP is the transmembrane pressure, μ is the absolute viscosity of the permeate and δ_{m} is the membrane thickness. Irreversible fouling occurs at specific points on the membrane as the operating permeate flux exceeds the critical point. A cake layer due to particle deposition on the membrane surface is formed over time. At this point, the operating permeate flux is defined by Darcy's Law:

$$J = \frac{\Delta P}{\mu\,(R_m + R_c)} \tag{7}$$

where R_c and R_m are resistances of the cake layer and membrane, respectively. Membrane filtration reaches a pseudo-stationary condition as wastewater is continuously supplied into the system. The flux in this regime is called a limiting flux, in which heavy fouling occurs in the membrane system. An increase in the transmembrane pressure does not significantly enhance the flux. The thin-film model may be used to describe the flux (140):

$$J = \frac{D}{\delta_{bl}} \rho ln \left(\frac{C_m - C_p}{C_b - C_p} \right) \tag{8}$$

where D is the solute diffusion coefficient, δ_{bl} is the solute boundary layer thickness, C_m is the solute concentration at the membrane surface, C_p is the solute concentration in the permeate and C_b is the solute concentration in the bulk.

Several major types of fluid instability methods used to increase permeate flux and fouling prevention have been discussed by Al-Bastaki and Abbas (163). Unfortunately, the use of backpulsing (164, 165) and Taylor vortices principles have been found to be unfavourable because their industrial application is limited by the need for extra equipment and their high energy consumption, which would increase the capital and operating costs. Vortices principle installation without moving parts has been explored to improve the membrane filtration performance. Dynamic mixing by vortices in a curved channel may carry away particles instead of depositing them on the channel surface (166). Dean vortices, also known as secondary flows, which are centrifugal instabilities that appear in curved hollow fibre membranes, as illustrated in Figure 4, have been reported to improve the limiting flux in ultrafiltration compared to conventional straight membranes (167–170). The geometry of a curved membrane can be helically coiled, twisted/woven or sinusoidal/meander-shaped. Woven hollow fibres are more favourable because of lower shear stress and higher packing density compared to a helical geometry hollow fibre (171). Another approach to generate secondary flow is to use stamped or corrugated tubular ceramic membranes (172–176) and to place inserts in the membranes (177).

Membrane Gas Separation

Separation of a gas-liquid mixture by a semipermeable membrane is new to the aquaculture industry. This type of membrane allows one or more components of the mixture to transport through more readily than the others.

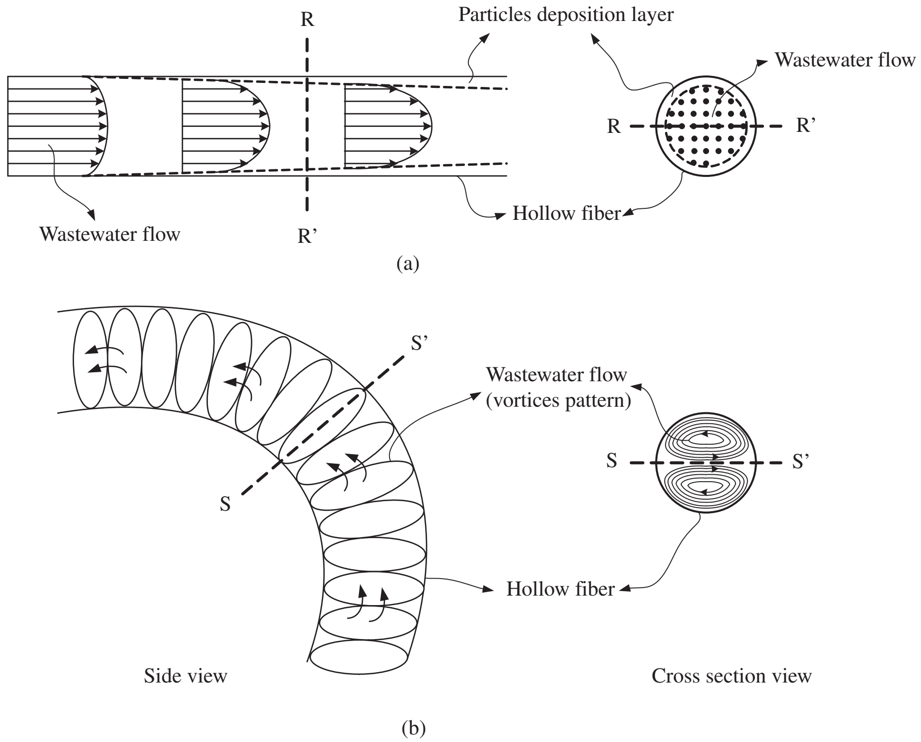

FIGURE 4 (a) Particles accumulate on the surface of a straight hollow fibre membrane. Fouling may occur from below the point of critical flux to a pseudo-stationary condition when wastewater continuously flows into the hollow fibre. (b) Dean vortices in a curved hollow fibre carry away the particles rather than depositing them on the surface fibre (166–170).

The membrane can be porous or dense, in which case the design selection relies on the volatility and solubility of the target gaseous components. The partial pressure gradient is the typical driving force of gas transportation. Mass transportation occurs when the partial pressure of a dissolved gas in a feed solution (upstream) is higher than that of permeate side (downstream). In addition, transportation might vary according to membrane selectivity, structures and flow orientation.

Organophilic pervaporation is the most promising technology for volatile organic compound (VOC) removals (178–182). Pervaporation is defined as a separation process in which one or more components of a liquid mixture can be selectively removed because of its higher affinity for the membrane and diffusivity through a membrane. A very low pressure is maintained on the permeate side to remove the permeable component(s). However, the transportation of VOCs through the membrane is relatively independent of the permeate pressure (183). Furthermore, the flux reduction of the VOCs can be reduced by increasing the permeate pressure (184). The concentration polarisation effect can reduce membrane selectivity and permeation flux for organics (185). Using Dean vortices can reduce the polarisation layer and greatly improve mass transfer in pervaporation (186).

Membrane distillation is a separation process that only uses microporous and hydrophobic membranes where liquid is not allowed to wet the pores (187). Membrane distillation uses a partial pressure gradient between the two ends of the membrane pores as the driving force for mass transfer. The partial pressure gradient is maintained by a temperature gradient whereby the temperature of the feed solution is raised. Therefore, the most volatile component in the feed vaporises and will selectively transport through the gas-filled pores. The partial pressure downstream is kept lower by a condensation process (188–190), by a gas sweeping (191, 192) and by using a vacuum (193). Absorption is sometimes integrated into direct contact membrane distillation to reduce partial pressure in permeates and increase mass transportation, i.e., sulphuric acid on the permeate side for dissolved ammonia removal (194, 195). Membrane distillation has been widely applied in low-concentration-dissolved gas treatments; these applications include the removal of alcohols and acetone by air gap membrane distillation (188, 189) and volatile organic compounds (196), including dissolved ammonia (197), by vacuum membrane distillation.

A gas-liquid membrane contactor brings a gas phase into contact directly with a liquid phase by absorption without the one phase dispersing to the other. Absorption can be facilitated by a chemical reaction. In the case of dissolved gas removal from an aqueous stream, the aqueous stream flows on one side of a hydrophobic membrane, and an absorbent flows on the other side. The hydrophobic membrane does not allow the liquid to enter the gas-filled pores until the pressure of the aqueous side exceeds the critical liquid penetration pressure. The membrane acts as physical support for the gas-liquid interface whereby mass transfer occurs between the phases. The driving force of the mass transfer is the partial pressure gradient, which is more due to the solubility of a solute in an absorbent than its volatility (198). The application of membrane contactors has been widely investigated for the removal of dissolved ammonia (199–203) by acidic absorbents, and cyanide (204) and iodine (199) by basic absorbents.

Using transverse flow to transfer oxygen with 100% efficiency using a low power input by a membrane contactor has been done successfully (205). A bundle of sealed-end hydrophobic fibres or composite membranes (206) are filled with pure oxygen at a pressure below the bubbling point and immersed into the water. Gradients of pressure and density allow the oxygen to diffuse through a gas-filled pore and create a plume vortex that results from aerodynamic effects in the water, as illustrated in Figure 5. The diffused oxygen is dissolved directly into the water without forming bubbles. However, Ahmed and Semmens (207) stated that the partial pressure

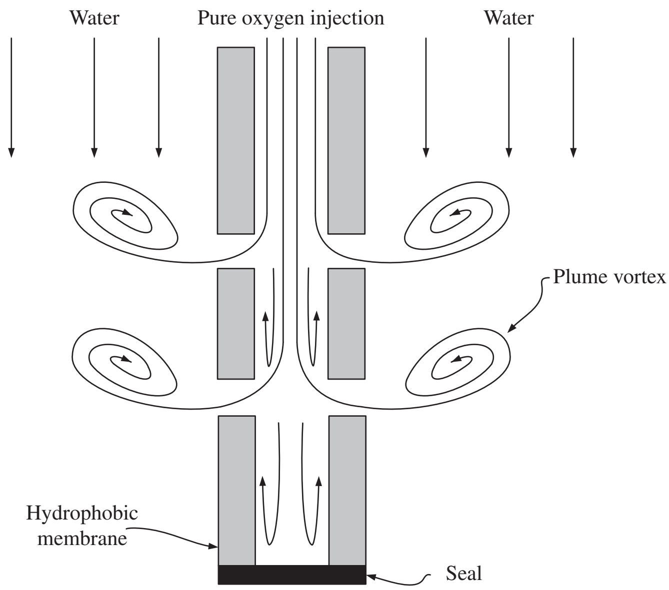

FIGURE 5 Water oxygenation by transverse flow technique requires a low power input by using a membrane contactor which is essential for aquaculture application (198). The gradients of pressure and density allow the oxygen to diffuse via a gas-filled pore and create an aerodynamic effect in the water. This could transfer oxygen with 100% efficiency (205, 206).

of oxygen might change along the fibre's length due to back diffusion of nitrogen gas from the water into the fibre. A higher mass transfer could be achieved at a low water flow rate when the flow is in a perpendicular orientation with the fibre (208, 209). Moulin et al. (210) showed that the water oxygenation performed better when Dean vortices were integrated with the transverse flow. It is noteworthy that water oxygenation by a low power input is essential for aquaculture because the fish that require high DO_2, such as trout, salmon and other species, are troubled by conventional aerators that have a high power input (198).

SUMMARY: ADVANTAGES OF MEMBRANE SEPARATION OVER CONVENTIONAL TECHNIQUES

Fish excrete faecal solids and unionised ammonia as waste products of metabolism directly from their anus and gills, respectively, in water. The uneaten feed and metabolism wastes accumulate in the RAS and may become a threat to fish health and welfare. Unionised ammonia is highly toxic to the fish. The organic particles retained in the fish culturing system undergo leaching or hydrolysis to produce finer particles and dissolved matter, which can be classified into organic carbon (C), phosphorus (P) and nitrogen (N). C, P and N are the sources of life for algae and pathogens in the RAS. Dissolved oxygen decreases over time due to fish respiration, excessive eutrophication and the growth of pathogens, which in turn increases carbon dioxide in water. For fish culturing, the dissolved oxygen level in the water should never fall below 5 mg/L; otherwise, the fish will suffer stress and die. Therefore, high quality water in an aquaculture system requires an engineered technology. Various techniques have been introduced and applied in aquacultural water purification such as settling systems, clarifiers, mechanical filters, ozonation and biological treatment. However, these conventional techniques show imperfect performance due to their disadvantages of sludge production, high energy consumption and requiring a number of mechanical parts and frequent maintenance. Hence, the membrane-based technique is introduced to complement the flaws of the conventional techniques. Table 3 shows the advantages of membrane processes over the conventional techniques in aquacultural water purification.

CONCLUSIONS

Solids in an aquaculture system come from uneaten feed and the faecal solids produced by the fish. Solids that are larger than 60 μm are conventionally removed by microscreens. However, research on fine particle (<60 μm) management and controlling dissolved ammonia production from particle leaching deserves further inquiry. Unionised ammonia is highly toxic to fish, and this form of ammonia is excreted directly from the fish gills into the water. A seawater recirculating aquaculture system should be managed carefully because seawater species are slightly more sensitive to ammonia than freshwater species. Microbial activities in ammonia reduction may also encounter the stress of salinity changes. Membrane technology is therefore proposed in this paper specifically to remove fine particles as efficiently as possible to achieve and maintain the ammonia concentration at a safe level. Dean vortices and transverse flow can prevent membrane fouling and perform well in water oxygenation, respectively. The economic feasibility of membrane technology integrated with a seawater recirculating aquaculture system still requires evaluation.

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TABLE 3 Summary of the flaws of conventional water treatment and the advantages of membrane-based technique over the conventional techniques in aquacultural water purification

Conventional techniques	Membrane-based techniques
Microscreen: • The filter meshes ≥60 μm are commonly used in the intensive RAS (7); thus, the solids <60 μm remain in the fish culturing system. • Pumping and turbulent forces stimulate the mineralisation and leaching of particles (62, 63) thus increasing the concentration of finer particles of <60 μm in the system. Fluidised sand filter: • Could increase the 10–105 μm particles (sludge) due to biofilm sloughing (12, 17). • Oxidising the dissolved solids is a time consuming process. • Aerobic microbes consume dissolved oxygen in completing the process. • For marine RAS, the microbes become inactive when the wastewater contains >1% salt (88–101). • Salt-tolerant microbes give inconsistent performance because the removal efficiency depends on the composition of wastewater (107, 108). Chemical treatment (ozonation): • The removal efficiency is limited by the wastewater hardness and the initial suspended solid concentration (32). • It produces byproducts when used in marine RAS. These byproducts are bromate and brominated compounds are harmful to fish and carcinogenic to humans (35). Spray tower: • Gas-phase axial dispersion reduces the water-oxygen contact area. • Preventing dispersion requires a sufficient pressure drop along the axis of the tower, nozzle spraying force and bulk gas recirculation force (58), which requires a relatively high pumping energy. Low head oxygenator: • The oxygen absorption is inconsistent because the efficiency relies on splashing and the depth of bubble penetration in the receiving pool of water (60).	General advantages: • A membrane system requires a relatively low pressure and energy and thus reduces the operating cost. • Fewer mechanical parts required. • Simple construction, low corrosion and easy maintenance. • Small footprint. Liquid-liquid and liquid-solid separations: • Separation depends on molecular sieving principle rather than properties of wastewater. • The membranes in micro-, ultra- and nano-filtration and reverse osmosis could remove the fine particles <10 μm (Figure 2). • No biological process; thus, no sludge production, a low dissolved oxygen reduction and a faster process suitable for seawater RAS. • No carcinogenic byproducts are produced. • Hybrid processes of coagulation-membrane filtration can improve the particle removal (146–148). • The coagulation process can be excluded when using the membrane materials with a weak interaction with the target components or applying the Dean vortices principle (167–170). Gas-liquid separations: • Pervaporation and membrane contactors can selectively remove the volatile organic compounds that give odours to the water. • Membrane distillation is the fastest process in gas-liquid membrane separations. • Uses low temperature and energy. • Produces a relatively high quality of water. Oxygenation by membrane contactors: • Could transfer oxygen into water with 100% efficiency by transverse flow (205, 206). • Using a low power input (205). • Oxygenation is improved when incorporated with Dean vortices principle (210).

NOMENCLATURE

C concentration (mol/dm^3)
d diameter (m)
D diffusion coefficient (m^2/s)
f mass of waste solids per unit mass of feed (-)
F mass flow rate (kg/s)
J operating permeate flux (kg/m^2.s)
P mass production rate (kg/s)
ΔP transmembrane pressure (Pa)
Q volumetric flow rate (m^3/s)
R resistance (m^2/kg)
S salinity (% or part per thousand)
T temperature (K)
V volume (m^3)

Greek Letters

δ thickness (m)
ϵ porosity (-)
η fraction of total waste solid (-)
ρ density (kg/m^3)
μ viscosity (Pa.s)

Subscripts

b bulk
c cake layer
bl boundary layer
fine fine particles
m membrane
p permeate

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