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Review

Research Progress and Prospects of Marine Oily Wastewater Treatment: A Review

Meiling Han [1,*], Jin Zhang [2], Wen Chu [3], Jiahao Chen [1] and Gongfu Zhou [1]

1 College of Navigation, Dalian Maritime University, Dalian 116026, China; chenjiahao_0202@163.com (J.C.); zhougongfudlmu@163.com (G.Z.)
2 College of Environmental Sciences and Engineering, Dalian Maritime University, Dalian 116026, China; zhangjin@dlmu.edu.cn
3 ACRE Coking & Engineering Consulting Corporation, MCC, Dalian 116085, China; chuwen4881@hotmail.com
* Correspondence: ml_han@dlmu.edu.cn

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Abstract: Oily wastewater from shipping waste and marine accidents have seriously polluted the marine environment and brought great harm to human production and health. With the increasing awareness of environmental protection, the treatment of marine oily wastewater has attracted extensive attention from the international community. Marine oily wastewater has various forms and complex components, so its treatment technology faces great challenges. Sources, types, supervision, and treatment of marine oily wastewater are introduced in this paper. The research progress of marine and ship's oily wastewater treatment technologies in recent years are reviewed from the perspectives of physical treatment, chemical treatment, biological treatment, and combined treatment, respectively. Principles and characteristics of all kinds of technologies were analyzed. In addition, this paper shows that multiple processing technologies used in combination for the purpose of high efficiency, environmental protection, economy, and energy conservation are the future development trend.

Keywords: marine; oily wastewater; filtration; membrane; adsorption; electrochemical; biological; combined method

1. Introduction

With the rapid development of the global shipping industry, marine oil pollution caused by illegal discharge of sewage, oil spill accidents, and offshore oil drilling platforms has become an increasingly serious environmental and safety problem [1–3]. On the one hand, oil products contain volatile organic compounds (VOCs), polycyclic aromatic hydrocarbons (PAHs), and other toxic and harmful substances, which can affect the reproduction and growth of marine organisms [4,5]. The large amount of oily wastewaters can reduce the production and diversity of marine animals and plants. Toxic compounds in oily wastewater can cause ecological disturbances, including alteration of the aquatic community structure and food chains. For example, carcinogens and toxins accumulated at the bottom of the food chain and passed down to human consumption could cause great harm to human health. Furthermore, toxic compounds also have various adverse impacts on the surrounding environment, such as air pollution caused by the evaporation of oil and hydrocarbon contents to the atmosphere. In addition, they can affect groundwater, seawater, or drinking water. On the other hand, a large amount of oil spilled on the sea surface may cause fires and affect shipping safety [6]. The reasons for such a situation are not only the weak safety awareness of the crew, improper supervision, and other management loopholes, but also the lack of mature and economic methods for the treatment of marine oily wastewater. Therefore, in order to reduce the discharge of oily sewage from ships and reduce the pollution of marine environment, the technologies of marine oily wastewater treatment should be developed while the supervision of ships is strengthened.

In recent years, the research of marine oily wastewater treatment technology has made progress by leaps and bounds by scientists all over the world. The objective of this paper is to review the currently developed methods for marine oily wastewater treatment from physical treatment, chemical treatment, and biological treatment, respectively. The progress of applications of combined methods in marine oily wastewater treatment is introduced in this review. Aiming at the existing problems, future development of marine oily wastewater treatment technology is also forecasted. Figure 1 presents the graphical abstract of the review.

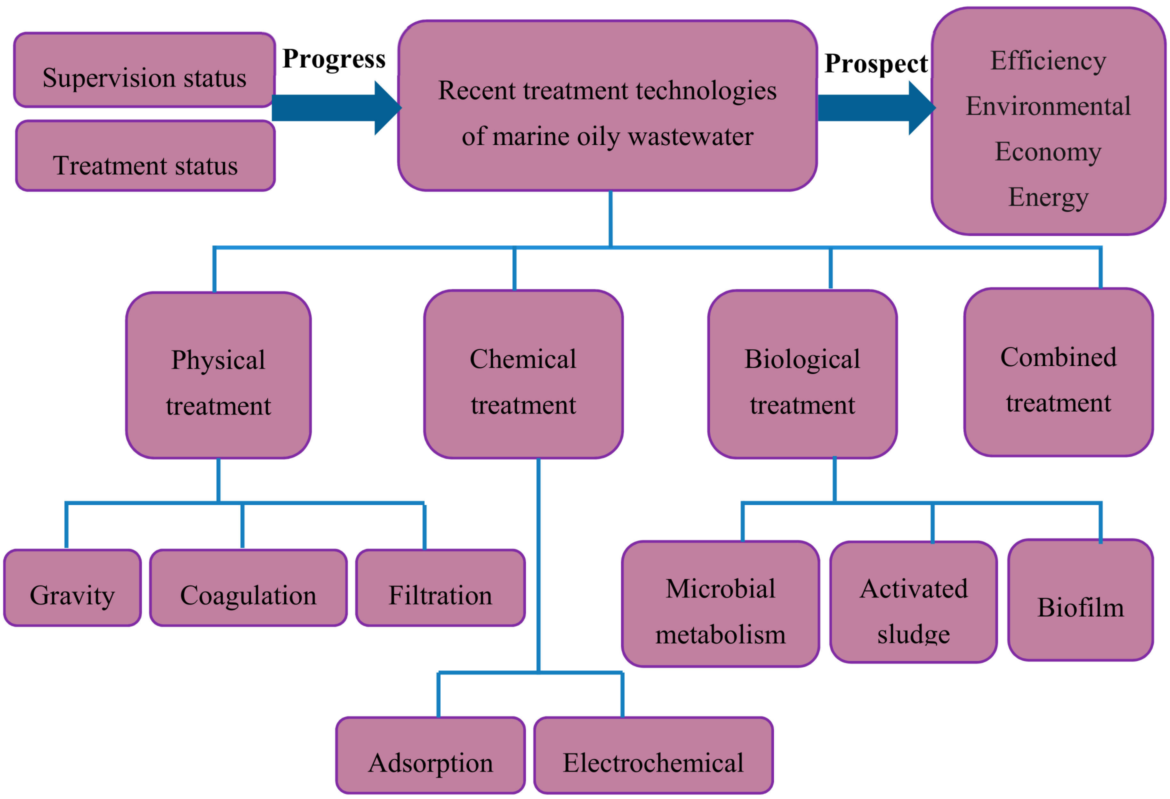

Figure 1. Graphical abstract of the review.

2. Supervision and Treatment Status of Marine Oily Wastewater

After being discharged into the sea, a ship's oily wastewater will cause pollution to the water body and seriously damage the marine ecological environment system. The oil mixed in water can be fats, hydrocarbons, hazardous metal ions, detergents, surfactants, and petroleum products, such as crude oil, diesel oil, gasoline, lubricant, and kerosene. Hydrocarbons, including benzene, toluene, ethylbenzene, and xylene, are known for their persistence in the environment. The physical/chemical properties of marine oily wastewater are salty, alkaline, indecomposable, and serious emulsification, which will present four states in the water, namely, floating oil (>100 μm), dispersed oil (10–100 μm), emulsified oil (0.1–10 μm) and dissolved oil (<0.1 μm). There are two main ways marine oil pollution is caused by ship transportation and offshore operations: accidental discharge and operational discharge. Accidental discharge refers to the overflow of crude oil, fuel oil and lubricating oil into the sea after ship accidents, such as collision, grounding, and hitting rocks by oil tankers [7]. Figure 2 demonstrates two of the most renowned oil spills in history [8]. Accidental oil spill pollution has a long enduring time, wide spread, and is difficult to treat. In order to reduce the probability of marine accidents effectively, the international community has strengthened maritime security regulations by issuing many conventions, laws, and regulations, such as OILPOL1954, CLC1992, MARPOL73/78, and OPRC1990. At present, the main methods to deal with the accidental discharge of oil wastewater include oil containment booms [9], combustion [10], oil dispersants [11], microbial degradation [12], adsorption method [13], and so on.

(a)

(b)

Figure 2. Oil spills and clean-up efforts: (**a**) Atlantic Empress (1979); (**b**) Exxon Valdez Oil spill clean-up efforts (1989) [8].

Operational discharge mainly refers to bilge water, tank washing water, and ballast water, which involves oil wastewater collected by port waste reception facilities. In practice, there will be some problems, such as defects of supervision, discharge of untreated oil sewage, insufficient utilization of port oil sewage reception facilities [14], and unclear whereabouts of oil wastewater after receiving. In order to effectively control oily wastewater discharge from ships, the relevant international conventions and domestic laws and regulations have created increasingly stringent restrictions [15]. The MARPOL 73/78 convention clearly stipulates that the amount of oil discharged into the sea from ships should be less than 15 mg/L, which varies by sea area. MEPC.60 (33), passed in 1992, has made it mandatory that ships should be equipped with a ship oil/water separator to deal with the ship's oil wastewater. Then, MEPC.107 (49) added the experimental requirements and test methods for emulsified oil treatment, and also put forward new requirements for ship oil/water separators. China's latest national standard GB 3552-2018 also strictly illustrates that oil sewage generated by ships should meet the specific requirements and discharge limit of 15 mg/L or discharge into port waste reception facilities [16].

Treatment methods for operational discharge of oily wastewater include shipboard treatment and shore treatment. For shipboard handling, limited to the size of the equipment and maintenance costs, the phase separation technique is mainly adopted in oil/water separation technology. The method has a good effect on dealing with floating oil and dispersed oil, but no obvious effect on the handling of emulsified oil and dissolved oil. With respect to the shore-based approach, the treatment methods are varied and the treatment effect is better than that of the shipboard approach as a whole, since the treatment methods of domestic and industrial oily wastewater can be served as reference. The shore-based approach is often used in the treatment of oily sewage collected by the port waste reception facilities. However, due to its characteristics, marine oily wastewater is usually of a complex composition, high in salt, high in oil content and serious emulsification, and there are certain bottlenecks in the treatment technology.

3. Recent Treatment Technologies of Marine Oily Wastewater

Due to different oil content and morphology, different sources and types of oily wastewater have different treatment methods. Recent treatment technologies of marine oily wastewater mainly include physical treatment, chemical treatment, biological treatment, and combined treatment.

3.1. Physical Treatment

3.1.1. Gravity Separation Method

As the most basic method for marine oil sewage treatment, the gravity separation method is carried out by using the density difference between oil and water. The greater the density difference between the oil and water, the better the separation effect. Although the structure of a gravity separation device is simple, it has the disadvantages of having a large area, limited separation capacity, complex operation and management, and poor treatment effect on emulsified oil [17]. In order to meet the requirements of the convention, this method is usually used as a pretreatment or primary treatment. Currently, the gravity separation method is used in the first stage of a ship oil/water separator, which is mainly for the treatment of floating oil and dispersed oil.

3.1.2. Coagulation Separation Method

Coagulation separation is a method of separating oil and water by increasing the chance of collision of small oil particles. When the oil droplets gradually gather to be large enough, they will float on their own buoyancy. The cyclone technique can be used in the coagulation separation method to make oily sewage enter the cylinder along the tangential direction at a high speed along the cylinder wall under pressure, and form a high-speed rotating motion in the cylinder to generate centrifugal force, so that the floating oil and dispersed oil can quickly gather in the center of the cyclone to achieve the separation [18]. At present, most ship oil/water separation equipment adopts a gravity-coagulation combination.

3.1.3. Filtration Separation Method

Filtration separation allows oil to pass through a layer of porous media that traps oil droplets and other particles in the sewage and removes the water. Recently, reports on the filtration separation of oily wastewater have mainly focused on the research and development of filtration separation materials, including mesh [19], porous hydrogel [20], aerogel [21], textile [22], membrane [23], and so on. Table 1 shows the treatment efficiencies obtained in some works by using filtration separation method for removal of pollutants from marine oily wastewater.

Table 1. Removal efficiencies of pollutants from marine oily wastewater by filtration separation method.

Filtration Separation Material	Source of Oily Wastewater	Experiment Result	References
multilayer-coated mesh substrates	Simulated oil-seawater	completely preventing bulk water	[19]
SA-NFC aerogel	Synthetic oil-seawater mixtures	99.65% separation efficiency; 40 cycles reusability	[24]
GO-ALG aerogel	Synthetic oil-seawater mixtures	>90% separation efficiency; at least 40 cycles reusability	[25]
NFC-CS aerogel	Synthetic oil-seawater mixtures	>90% separation efficiency; at least 40 cycles reusability	[26]
DLC coated cotton textile	Synthetic oily wastewater with diesel oil and crude oil	>99% separation efficiency	[27]
integrated UF/RO membrane	Synthetic oily bilge water	<10 mg/L oil and no SS during UF process; 70% TOC, 90% cations, P_2O_5 and sulfate anion during RO process	[28]
UF process with FP100 membrane	Oily bilge wastewater from harbor wastewater treatment facility Miedzyodrze (Szczecin, Poland)	0.9–1.1 mg/L oil; 0.08-0.26 NTU	[29]

Kratochvil et al. [19] attempted to prepare covalent cross-linked nanoporous polymer superhydrophobic and superoleophilic substrates (wire meshes) for the treatment of marine oily wastewater, which could allow oil to pass through while completely preventing bulk water. Li et al. [24] investigated the capability of filtration separation in the treatment of marine oily wastewater from oil spill by using a robust salt-tolerant superoleophobic aerogel as filtration material. The green aerogel is prepared by freeze-drying of sodium alginate (SA)-nanofibrillated cellulose (NFC) using Ca^{2+} ions as crosslinking agent, as shown in Figure 3. The results showed that the aerogel could separate oil–seawater mixtures with high separation efficiency (up to 99.65%) and good reusability (at least 40 cycles). Subsequently, the research group [25,26] used freeze drying and ion crosslinking methods to prepare aerogels by incorporating graphene oxide (GO) into alginate (ALG) matrix or incorporating nanofibrillated cellulose (NFC) into a chitosan (CS) matrix, which also achieved good results. The facile fabrication process combined with the excellent separation performance makes it promising for practical applications in marine environments.

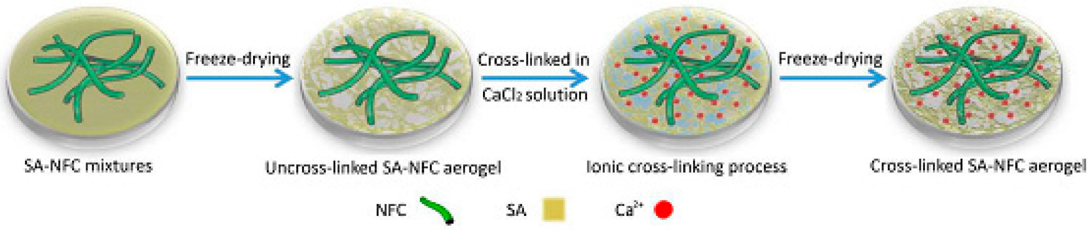

Figure 3. The preparation of cross-linked sodium alginate (SA)-nanofibrillated cellulose (NFC) aerogel [24].

Cortese et al. [27] described a convenient approach to fabricate cotton textiles with a hydrophilic coating for the treatment of wastewater from marine spilt oil, showing both superhydrophobic and superoleophilic properties. The surfaces are successfully prepared by one-step growth of diamond like carbon film onto the textile through plasma enhanced chemical vapor deposition method, as schematically shown in Figure 4. The results exhibited a highly-controllable, energy-efficient oil/water separation with high separation efficiency.

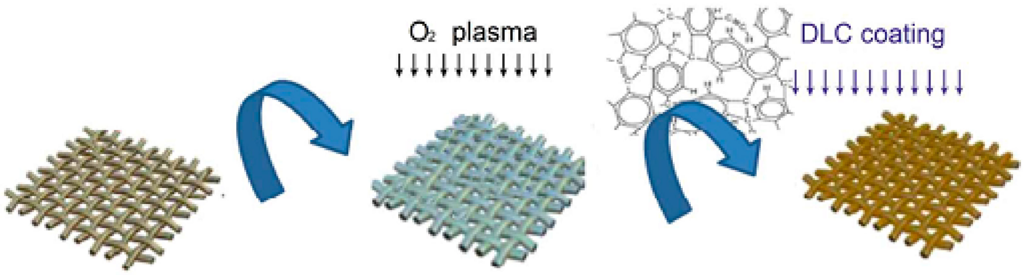

Figure 4. The formation of a superhydrophobic surface on cotton based on oxygen plasma pretreatment and DLC coating [27].

Membrane separation is the physical process of selectively separating the oil or water in the oil-water mixture by using the microporous structure on the membrane surface. Depending on the different pore diameters, it can be divided into microfiltration (MF) membrane, ultrafiltration (UF) membrane, nanofiltration (NF) membrane, and reverse osmosis (RO) membrane. Tomaszewska et al. [28] adopted an integrated UF/RO system to treat oily bilge water. The influence of transmembrane pressure on the permeate flux, degree of the retention of organic (including oil), and inorganic compounds was investigated. The results demonstrated that a high effectiveness of purification can be achieved. The first stage of bilge water treatment with UF process had the oil content below 10 mg/L and no SS whereas almost all turbidity was removed. The second stage of the treatment with RO process resulted in removing 70% TOC, 90% cations, P_2O_5 and sulfate anions. Karakulski et al. [29] applied UF to in-depth treatment of oily bilge water. After preliminary treatment of bilge water collected at the port by coagulation/flotation process, the effluent with oil content of 7–13 mg/L was obtained. Then the effluent was performed with FP100 membranes and the turbidity of obtained UF permeate was varied in the range of 0.08–0.26 NTU and the oil content was at a level of 0.9–1.1 mg/L. The membrane separation can treat dispersed oil well, and achieve a better cost-effectiveness ratio on the basis of the same input.

3.2. Chemical Treatment

As one of the most widely used methods in the treatment of marine oily wastewater, chemical treatment uses chemical reactions to deal with oil, suspended solids (SS), chemical oxygen demand (COD), and so on in oily sewage. Recently, several chemical technologies have been applied to treat marine oily wastewater, such as adsorption and electrochemical oxidation. A summary of the treatment efficiencies of chemical treatment obtained in some works aimed at removing pollutants from different sources of marine oily wastewater is presented in Table 2.

3.2.1. Adsorption Method

The Adsorption method is a method that uses solid adsorption materials with good porosity and high specific surface area as a sorbent to adsorb small- and medium-sized oil molecules on the surface, so as to achieve oil/water separation. When the adsorbent is used to treat oily sewage, it can not only adsorb indecomposable hydrocarbon organics and reduce COD, but also decolorize and deodorize the sewage. As it can treat the sewage to a reusable level, the adsorption method has been widely used in ship oily sewage treatment. Traditional adsorbents, such as zeolite and activated carbon, have a disadvantage of limited adsorption capacity, long adsorption time, and high cost. In recent years, scientists have developed various novel adsorbents with different materials to treat marine oily wastewater, such as foam [30], biomass [31–33], metal organic framework [34,35], chitosan [36–38], cotton [39], sponge [40–42], magnetite nanoparticles [43–45], and so on.

Abdelwahab et al. [32] evaluated a modified sugarcane bagasse (B) hydrophobic sorbent for diesel oil removal from artificial seawater. Their results showed that additional hydrophobicity was achieved by coating the esterified bagasse (EB) with a hydrophobic polymer (PAN), as a peak adsorption capacity of 8.4 g/g for diesel oil and reused for more than six cycles were reported at a pH of 5.5, sorbent dose of 2 g/L, initial oil concentration of 20 g/L, and contact time of 40 min. They concluded that the advantages of hydrophobicity were clearly addressed by the reduction of water adsorption capacity (WAC) and increased oil adsorption capacity (OAC). Generally, the results are promising and the prepared sorbents can positively add to solve environmental problems. Vlaev et al. [33] attempted to establish the possibilities to use rice husk ash adsorbents for cleaning spilled oil, oil products, or bilge water. Black rice husk ash (BRHA) and white rice husks ash (WRHA) were obtained via pyrolysis of raw rice husks in a pilot plant fluidized-bed reactor under different conditions. The results showed that BRHA have higher adsorption capacity than WRHA. At a given temperature of 298K, BRHA sorbed 6.22g/g of crude oil, and 5.02g/g of diesel fuel. The results obtained showed that the material studied has a high adsorption capacity and low cost and may be used successfully as an effective adsorbent to cleanup of bilge water and spills of oil and oil products in water basins.

Rahmani et al. [35] utilized the prepared nanocomposites as new oil cleanup materials for the removal of oil spills. MIL-101 and MIL-101@nanoporous graphene (NPG), with different nanoporous graphene contents (30, 60, and 90 wt%) were herein synthesized via a solvothermal method. The high oil adsorption capacity (14 g/g) was achieved for the synthesized adsorbents with the high surface area of MIL-101 (4293 m^2/g) and MIL-101@NPG 60wt% (4642 m^2/g), and the high pore volume of MIL-101 (2.42 cm^3/g) and MIL-101@NPG 60 wt% (2.62 cm^3/g). The results showed that the crude oil adsorption capacity was enhanced by adding NPG to the virgin MIL-101, which is higher than previous scientific reports. Furthermore, the synthesized adsorbents could be further reused for 10 times with no significant adsorption loss. Digital photographs of the removal of crude oil from water surface by synthesized nanocomposite are shown in Figure 5.

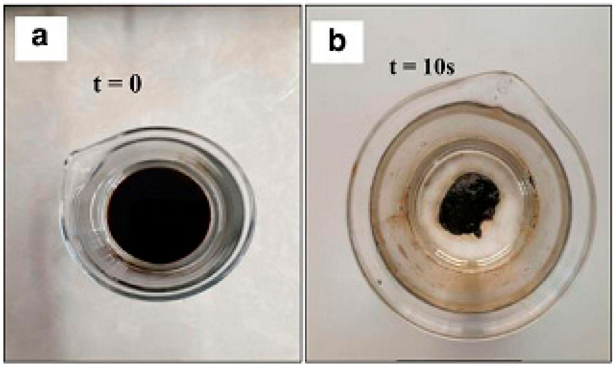

Figure 5. Digital photograph images of removal of crude oil from water surface by synthesized nanocomposite (**a**,**b**) [35].

Doshi et al. [37,38] studied the effectiveness of N, O-carboxymethyl chitosan (NO-CS) and sodium form of amphiphilic chitosan as adsorbents, respectively, in the oil spill response for the removal and recovery of floating oil from the sea water (Figure 6). Water-soluble NO-CS was synthesized by carboxymethylation of chitosan in a hydro-alcoholic medium at 50 °C by chloroacetic acid. Sodium salt of oleoyl carboxymethyl chitosan (NaO-CMCS) was synthesized by introducing hydrophobic groups on amino groups of hydrophilic sodium salt of carboxymethyl chitosan (Na-CMCS). The effectiveness of NO-CS was studied by considering the dosage, salinity, and pH on the marine diesel, diesel, and marine-2T oil. Their results showed that the destabilization of marine diesel was the most effective among the studied three oils, which showed excellent adsorption at sea water alkalinity and salinity. They also found that hydrophobically-modified NaO-CMCS could sustain the oil-in-water (o/w) emulsion stability for more than six weeks with droplet sizes ≤ 30 μm. The recovery of oil was 75–85% and 19–49% from deionized water and sea water, respectively.

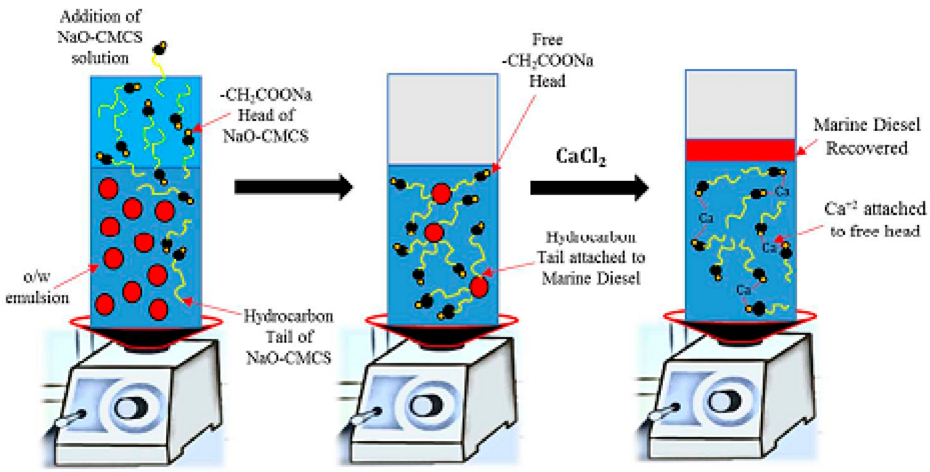

Figure 6. Schematic of removal of floating oil with NaO-CMCS [37].

Gupta et al. [39] prepared a surface-tailored wettability modified superhydrophobic/superoleophilic Janus membrane for separation of offshore petroleum wastewater. They impregnated a nano-sized polytetrafluoro-ethylene (PTFE) dispersion onto a cotton substrate using the Meyer rod coating technique. The schematic of the fabrication is shown in Figure 7. The membrane exhibited excellent separation efficiency up to 98% and retained its intrinsic properties for at least

30 recurrences. The application of filtration separation method in shore-based oil wastewater treatment has a broad prospect.

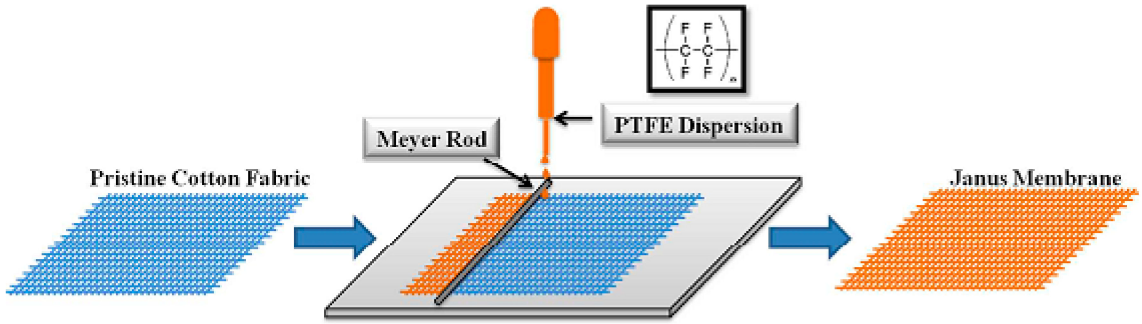

Figure 7. Schematic of the fabrication of Janus membrane [39].

Wang et al. [41] investigated the adsorption behaviors of sponges for the separation of oil–water mixtures from marine spills. A superhydrophobic and superoleophilic polyurethane (PU) sponge was synthesized via an environmentally friendly surface grafting of a polymer molecular brush (Figure 8). Their results showed that the grafted PU sponge possessed high absorption capacity (23 times its own weight), high oil retention (93%), high mechanical strength, and good recyclability (more than 400 times), because of the expansion in oil and collapse in water of the polymer molecular brushes.

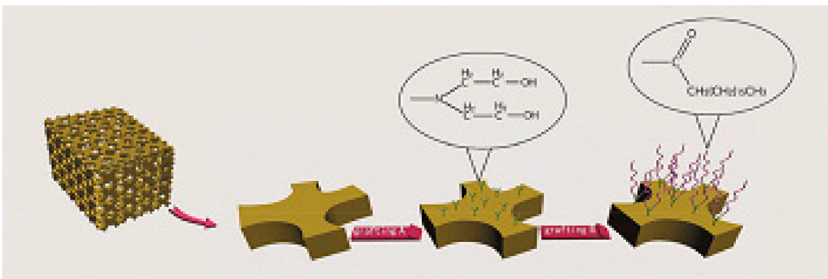

Figure 8. Illustration for the fabrication of a high oil wettability PU sponge [41].

Turco et al. [42] proposed an innovative approach for the fabrication of porous polydimethylsiloxane (PDMS) starting from an inverse water-in-silicone procedure able to selectively collect oil from water in few seconds (Figure 9). Their results showed that the oil mass absorption was in the range of 3.9–32.3 g/g depending on viscosity, density and surface tension of the organics. This innovative fast and simple approach can be successful in case of emergency, as oil spill accidents, permitting in situ fabrication of porous absorbents. The porous material evidenced a higher volume sorption capacity for oil sorption from water, excellent mechanical, reusability properties, and low cost.

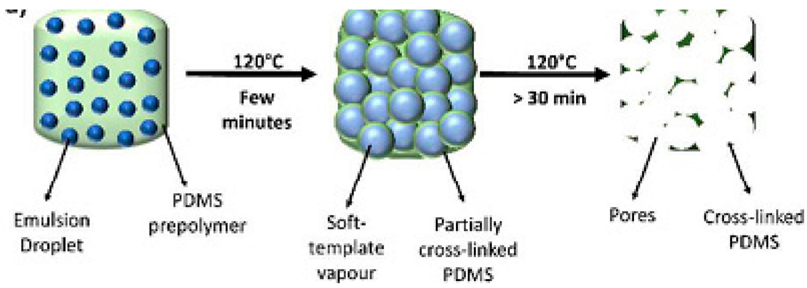

Figure 9. Schematic illustration of the preparation of the PDMS sponge [42].

Magnetic separation uses magnetic materials as carriers, taking advantage of magnetization effects and leaves magnetic materials and adsorbed oil in the magnetic field, so as to achieve the purpose of oil/water separation. Mirshahghassemi et al. [43,44] synthesized polyvinylpyrrolidone (PVP)-coated magnetite nanoparticles by a hydrothermal method, which could effectively remove MC252 oil from the oil-water mixture prepared by fresh water and sea water with a removal rate of nearly 100%, respectively. Furlan et al. [45] manufactured high-performance magnetic polydimethylsiloxane (PDMS) nanocomposite sponge materials for the treatment of oily wastewater from vessels. Their results showed the sorption capacity measured by the weight gain for cleaning agents (440%), emulsified oil droplets (580%), and oils of varied viscosities and densities (415–760%). They also showed that the magnetic sponges are resistant to corrosive chemicals, exhibit high adsorption capacity for oil and detergents, and selective adsorption for oil in oil–water emulsion. The sponges can be repeatedly used without any signs of deterioration. They are magnetically controllable and remain in position when magnetized, allowing for separation from the motion of the ship. The magnetic separation method is a simple and feasible separation technology with high treatment efficiency, but it also has its technical difficulties and limitations. For example, it is necessary to select the polymagnetic medium with high magnetic saturation. Conventional magnetic materials require special conditions, such as organic solvents and high temperatures, which make large-scale production difficult, costly, and potentially toxic.

3.2.2. Electrochemical Oxidation Method

Chemical oxidation is one of the effective methods to remove COD and petroleum hydrocarbons from oily wastewater. It utilizes redox reactions in degrading organic matters into easily degradable materials directly or indirectly. As one of chemical oxidation technologies, micro-electrolysis has the advantages of having no additional chemical operation, has easy to automatic system control, low space requirements, and efficient treatment in a short time. The traditional coagulation method is to add coagulant to the system and take advantage of the adsorption and encapsulation of flocs formed by medicament to realize the treatment of oily wastewater. In order to overcome the disadvantages of traditional coagulation method, researchers used micro electrolysis to achieve the coagulation effect, also known as electro-coagulation/flocculation [46]. The electro-coagulation method uses aluminum, iron, and other metals as anodes. Under the action of direct current, the anodes are electrolyzed to produce aluminum and iron ions. Then various hydroxyl complexes and hydroxides are formed through a series of hydrolysis, polymerization, and ferrous oxidation processes, so that oil droplets and suspended substances in the waste water condense, precipitate, and separate.

Aswathy et al. [47] used aluminum electrodes to check the treatability of synthetic bilge water with a batch electrocoagulation study. The studies were conducted to investigate the effect of various operational parameters on the treatment efficiency. At a pH of 7, applied voltage of 10 V, spacing of 1 cm, and effective electrode area of 45 cm^2, a maximum soluble COD removal efficiency of 85% was obtained after an electrolysis time of 120 min. Among different electrode combinations of aluminum and iron, Al-Al combination showed maximum removal efficiency. A case study on real bilge water was conducted and a maximum removal of 89.84% was obtained at optimum conditions using aluminum electrodes. Rincon et al. [48] presented electrocoagulation to treat bilge water, with a focus on oily emulsions and hazardous metals (copper, nickel, and zinc) removal efficiency. Experiments were run with a continuous flow reactor, manufactured by Ecolotron, Inc., and a synthetic emulsion as artificial bilge water. The synthetic emulsion contained 5000 mg/L of oil and grease, 5 mg/L of copper, 1.5 mg/L of nickel, and 2.5 mg/L of zinc. The experimental results demonstrate that the best treatment and cost efficiency was obtained when using a combination of carbon steel and aluminum electrodes, at a detention time less than one minute, a flow rate of 1 L/min, and 0.6 A/cm^2 of current density. The final effluent concentration of oil and grease was always less than 10 mg/L. 99% of zinc removal efficiency and 70% of copper and nickel removal efficiency were achieved. Ulucan et al. [49,50] investigated the treatability of bilge water by electrocoagulation/flotation process with aluminum electrodes. The experiments were carried out in accordance with statistical runs assigned by response surface methodology. Under 9.87 mA/cm^2 of current density in approximately 13 minutes and in approximately 29 °C of inlet temperature, optimum COD and oil were removed by 90.3% and 81.7%, respectively. Bilgili et al. [51,52] investigated the capability of electrocoagulation with Al anode in the treatment of oily wastewater from a port reception facility in Turkey (Figure 10). Their results showed that when the current density is 50 A/cm^2 and electrolysis time is 20 min, SS, oil, COD removal rate are 65%, 75%, and 60%, respectively in batch operations. In continuous operation, the removal rates of SS, oil and COD were 80%, 90%, and 77%, respectively.

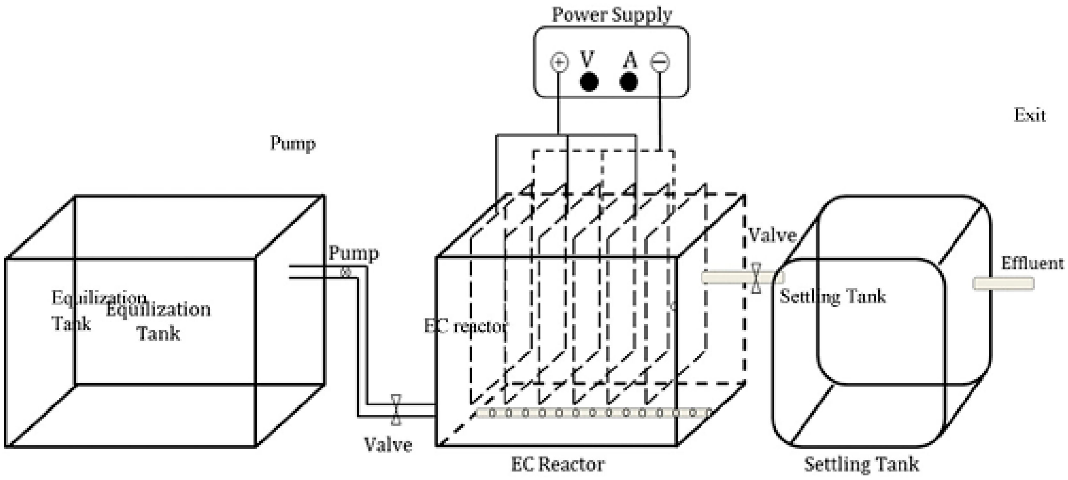

Figure 10. Schematic view of the pilot electrocoagulation plant [52].

Korbahti et al. [53,54] studied the treatment of bilge water using electrochemical oxidation with platinum iridium electrodes in a batch electrochemical reactor (Figure 11). The results showed that the optimized condition is 32 °C reaction temperature and 12.8 mA/cm^2 current density. Additonally, 99.2% COD removal, 93.2% oil removal, and 91.1% turbidity removal, and an average energy consumption of 33.25 kWh/kg COD removal were obtained. The method realized the application of low-energy portable shipboard wastewater treatment system for bilge water.

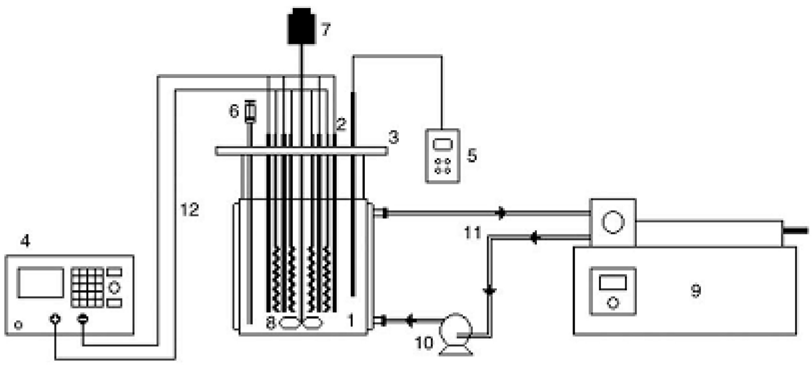

Figure 11. Batch electrochemical reactor system (1: stirred batch electrochemical reactor, 2: Pt/Ir anodes and Fe cathodes, 3: Polyamide®reactor lid, 4: programmable DC power source, 5: digital thermometer, 6: sampling cell, 7: driving motor, 8: glass mixer, 9: heating/cooling tank, 10: heating/cooling pump, 11: heating/cooling feed, 12: power supply connections) [53].

Carlesi et al. [55] investigated the treatment of bilge water by using an electrochemical reactor, with parallel plate electrodes, to achieve separation and flotation of oily traces, electro-reduction of contained metals and electro-oxidation of dissolved organic matter. Their results showed that color, turbidity, COD, and Pb-Zn concentration were reduced by 80%, 70%, 50%, and 40%, respectively in 2 h.

Table 2. Removal efficiencies of pollutants from marine oily wastewater by chemical treatment.

Treatment Method		Source of Oily Wastewater	Experiment Result	Ref
Adsorption method	Polyacrylonitrile-coated esterified bagasse (PAN-EB) sorbent	Synthetic oil-seawater mixtures with diesel oil	8.4 g/g diesel oil; reused > 6 cycles	[32]
	Black rice husk ash (BRHA) adsorbent	Synthetic oily bilge water with crude oil and diesel oil	6.22 g/g crude oil; 5.02 g/g diesel oil	[33]
	MIL-101@NPG	Simulate offshore oil spill	14g/g crude oil	[35]
	Sodium salt of oleoyl carboxymethyl chitosan (NaO-CMCS) adsorbent	Synthetic oil-in-water emulsion with marine diesel	75–85% oil in deionized water; 19–49% oil in seawater	[37,38]
	Janus membrane (cotton substrate)	Synthetic oil-seawater mixtures with diesel oil	98% separation efficiency; 30 recurrences	[39]
	Polyurethane (PU) sponge adsorbent	Synthetic oil-water emulsion with crude oil	93% oil removal; 40 times recyclability	[41]
	PVP-coated magnetite nanoparticles	Synthetic oil-seawater mixtures with crude oil	nearly 100% oil removal	[43,44]
	magnetic PDMS nanocomposite sponge	Oily wastewater from vessels	440% cleaning agent, 580% emulsified oil droplets, 415-760% oils of varied viscosities and densities	[45]
Electrochemical oxidation method	Al-Al electrode, pH = 7, applied voltage 10 V, electrolysis 120 min	Synthetic oily bilge water and real oily bilge water	85% COD for synthetic bilge water; 89.94% COD for real bilge water	[47]
	Carbon steel and Al electrodes, flow rate 1 L/min, current density 0.6 A/cm^2	Synthetic bilge water with 5000 mg/L oil, 5 mg/L copper, 1.5 mg/L nickel, 2.5 mg/L zinc	<10 mg/L oil, 99% zinc, 70% copper, and nickel removal	[48]
	6.95 of pH, 9.87 mA/cm^2 of current density, 29 °C inlet temperature	Bilge water from Haydarpaşa Port Waste Acceptance Plant in Turkey with 1033mg/L of COD and 338 mg/L of oil	90.3% COD and 81.7% oil	[49,50]
	Al electrode with current density of 50 A/cm^2, electrolysis 20 min	Oily wastewater from a port reception facility in Turkey	65% SS, 75% oil and 60% COD in batch reaction; 80% SS, 90% oil and 77% COD in continuous reaction	[51,52]
	Pt/Ir electrodes, reaction temperature 32 °C, current density 12.8 mA/cm^2	Synthetic bilge water with 50%/50% seawater/freshwater, 3080 mg/L COD and 2000 mg/L oil	99.2% COD, 93.2% oil, and 91.1% turbidity	[53,54]
	parallel plate electrodes (stainless steel and oxidized titanium), electrolysis 2h	Bilge water from Valparaiso and San Antonio ports in Chile	80% color scale; 70% NTU; 50% COD; 40% Pb-Zn	[55]

3.3. Biological Treatment

Biological methods have been widely applied for oily wastewater treatment. The performance of several biological technologies has been investigated and has yielded some impressive results recently. These biological technologies include the microbial metabolism method, activated sludge method, and biofilm method. Table 3 provides some of the performance data obtained from the application of biological treatment for marine oily wastewater.

3.3.1. Microbial Metabolism Method

The microbial metabolism method is the use of microbial degradation of crude oil, to convert organic matter, including oil, as nutrients into the microbe organic ingredients or to proliferate into new microorganisms, thereby to remove organic matter in wastewater [56]. Chanthamalee et al. [57] examined application of polyurethane foam (PUF)-immobilized *Gordonia* sp. JC11, a known lubricant-degrading bacterial inoculum, for the treatment of bilge water. The experiments showed that the PUF-immobilized bacteria were more efficient at removing oil than indigenous microorganisms and were able to remove approximately 40–50% of boat lubricant (1000 mg/L). Crisafi et al. [58] investigated the effect of three treatments in oily-seawater after a real oil-spill occurred in the Gulf of Taranto (Italy). Biostimulation with inorganic nutrients allowed the biodegradation of the 73 ± 2.4% of hydrocarbons, bioaugmentation with a selected hydrocarbonoclastic consortium degraded 79 ± 3.2%, while the addition of nutrients and a washing agent has allowed the degradation of the 69 ± 2.6%. Ma et al. [59] using crude oil as a carbon source, isolated eight strains with high petroleum-degrading activities from an oil terminal's surface water and mud, and selected two strong petroleum-degrading yeasts through enrichment culture. After five days of mixed culture, the petroleum degradation rate reached 51%.

3.3.2. Activated Sludge Method

Activated sludge is a kind of aerobic biological treatment method that uses suspended microbial floc to treat wastewater. A typical complete cycle of a sequencing batch reactor (SBR) consists of five stages, composed of water inlet, reaction, sedimentation, water outlet, and idle. SBR is widely used in marine sewage treatment due to the characteristics of quick response, high efficiency, strong resistance, and low operation cost [60–62]. Recently, Uma et al. [63] studied synthetic oily bilge water treatment and the subsequent production of biopolymer by using a SBR with the isolated novel soil bacteria from the hydrocarbon contaminated site near Karaikal port, India. The experimental results showed that a decrease in substrate concentration to 5000 mg/L of soluble COD showed maximum organic removal (81%) and maximum polyhydroxyalkanoates (PHA) yields of its cell dry mass (81%). The PHA yield was maximum at SRT of 5 d, pH = 7 and cycle time (CT) of 24 h.

3.3.3. Biofilm Method

The biofilm method is a kind of sewage biological treatment technology in parallel with the activated sludge method. Biofilm is formed by growing and breeding microorganisms on filter material or carrier. The sewage contacts the biofilm and organic pollutants in wastewater are taken in as nutrients by the microorganisms on the biofilm, so as to realize the purification of sewage. The main form is a biological filter. In recent years, novel membrane reactors, including the microporous membrane bioreactor (MBR), moving bed biofilm reactor (MBBR), sequential batch biofilm reactor (SBBR), and upflow anaerobic sludge bed-anaerobic biological filter (UASB-BF) [64,65]. Among them, MBR is a combination of membrane components and traditional biological treatment units, which have the characteristics of good separation effect and impact resistance, and are also a hot research field recently used by scholars to treat marine oily wastewater. Sun et al. [66,67] investigated biofilm-MBR technology to treat shipboard wastewater containing grey/black water and oily bilge water. Both dead-end side-stream and recycle side-stream configurations of a biofilm-MBR concept with flat sheet ceramic membranes have been investigated (Figures 12 and 13). A good membrane permeate quality was achieved in each process configuration, with <5 mg/L oil concentration.

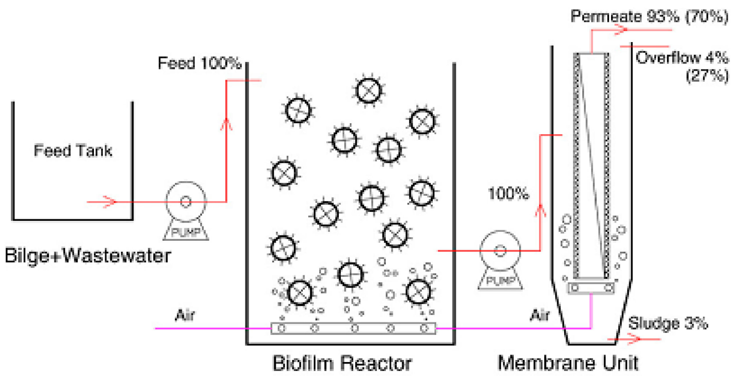

Figure 12. Dead-end side-stream configuration of bio film-MBR, membrane filtration unit recovery: 93% or 70% [67].

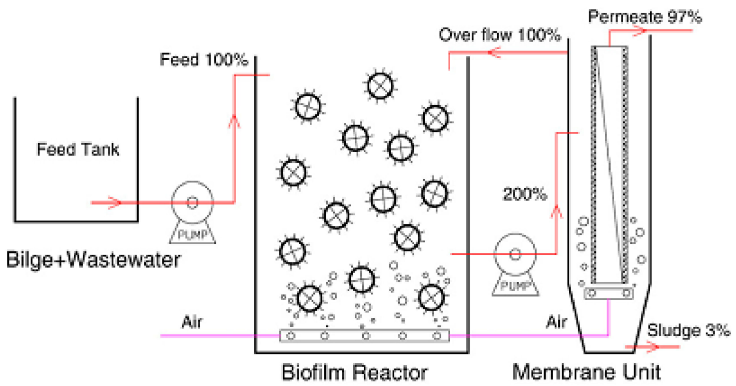

Figure 13. Recycle side-stream configuration of biofilm-MBR [67].

Wei et al. [68] established an anoxic/oxic-MBR (A/O-MBR) system for treating harbor oily wastewater (Figure 14). It showed good removal performance for COD, oil content, SS, and other pollutants. Then, they optimized MBR via improving the aeration rate and reducing the ratio of A_r/A_d.

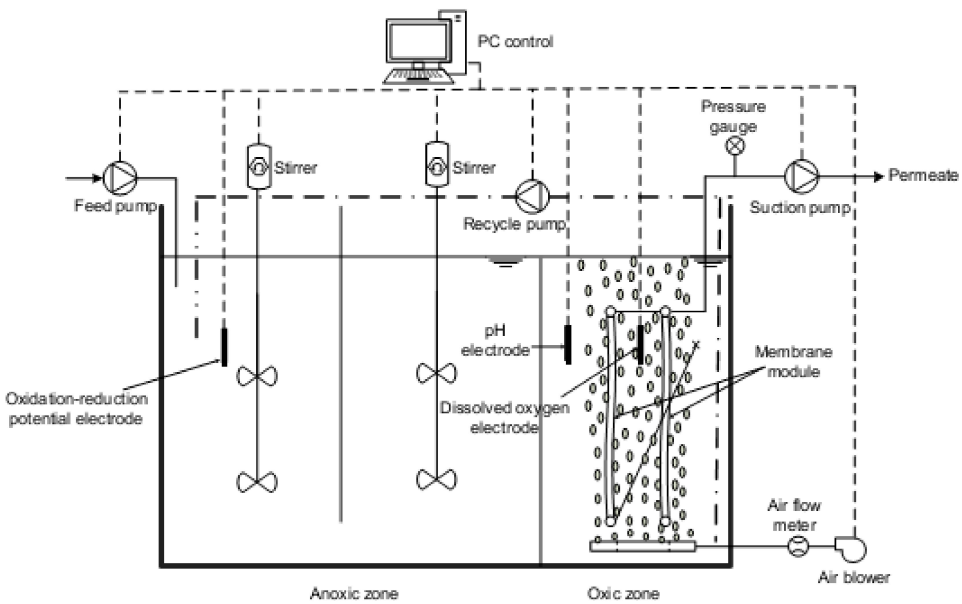

Figure 14. Schematic diagram of the pilot-scale A/O-MBR [68].

Vyrides et al. [69] examined the biodegradation of bilge water using first anaerobic granular sludge followed by inoculation of aerobic microbial consortium (consisting of five strains) in MBBR. After treating real bilge water for 165 days with 36 h HRT, the MBBR with a filling fraction of 40% resulted in the highest COD decrease (60%). Emadian et al. [70] used a hybrid up-flow anaerobic sludge blanket (HUASB) bio-reactor to treat dilute bilge water. Their results showed that the COD removal rate reached 75% with 8 h HRT and a 0.6 g COD/d organic load rate (OLR). In addition, the bio-reactor showed a good performance in removing oil from the waste stream, which was significantly lower than the standard value of bilge water discharge by International Maritime Organization (IMO). Compared with microbial metabolism method and activated sludge method, the biofilm method has the advantages of simple operation and good separation effect, which has attracted wide attention from scholars at home and abroad.

Table 3. Removal efficiencies of pollutants from marine oily wastewater by biological treatment.

Treatment Method		Source of Oily Wastewater	Experiment Result	Ref
Microbial metabolism method	PUF-immobilized bacteria	Bilge water from fishing vessels in the Sing Amnuai fishing port, Thailand	40–50% of boat lubricant	[57]
	Biostimulation, bioaugmentation and addition of nutrients	real oil-spill wastewater from Gulf of Taranto (Italy)	73 ± 2.4% of hydrocarbons through biostimulation, 79 ± 3.2% through bioaugmentation, 69 ± 2.6% while the addition of nutrients and a washing agent	[58]
	petroleum-degrading yeasts	Synthetic oil-spill wastewater	51% oil degradation rate	[59]
Activated sludge method	SBR, SRT of 5 d, pH = 7 and CT of 24 h	synthetic oily bilge water	81% COD	[63]
Biofilm method	biofilm-MBR	shipboard wastewater containing grey/black water and oily bilge water	<5 mg/L oil concentration	[66,67]
	A/O-MBR	harbor oily wastewater	About 90 % COD	[68]
	MBBR with anaerobic granular sludge followed by inoculation of aerobic microbial consortium	bilge water from treatment company at Zygi, Cyprus	60% COD	[69]
	HUASB bio-reactor with 8 h HRT and 0.6g COD/d OLR	dilute bilge water from Amirabad port, Iran	75% COD	[70]

3.4. Combined Treatment

Due to the various forms of marine oily wastewater, the use of a single technology is often unable to take into account the removal of floating oil, dispersed oil, emulsified oil, and dissolved oil at the same time. Furthermore, marine oily wastewater composition is complex, not only containing oil pollution, but also involves SS, COD, hazardous metals, and so on. Therefore, it is necessary to combine a variety of technologies to achieve a higher treatment effect. Currently, it has been reported that integrated processes applied in the treatment of marine oily wastewater include electrocoagulation/nanofiltration (EC/NF) process, photocatalysis and air-stripping integrated methods, photo-electro-Fenton hybrid systems, photocatalytic membrane separation, and coagulation-adsorption methods. Table 4 shows the performance data recorded by authors who have worked on the application of hybridized methods and systems to remove oil from different sources of marine oily wastewater.

Akarsu et al. [71] designed a compact wastewater treatment system for bilge water by electrocoagulation and nanofiltration integrated process (Figure 15). The influence of voltage, time, and initial pH on the removal efficiencies of COD was analyzed with aluminum electrodes as both the anode and cathode. Moreover, a commercial NF270 flat-sheet membrane was also used for further purification. The Box–Behnken design combined with a response surface methodology was used to study the response pattern and determine the optimum conditions for maximum COD removal and minimum metal ion contents of bilge water. The total reduction of COD at the level of 74% results in COD content below 150 mg COD/L in the EC/NF integrated system.

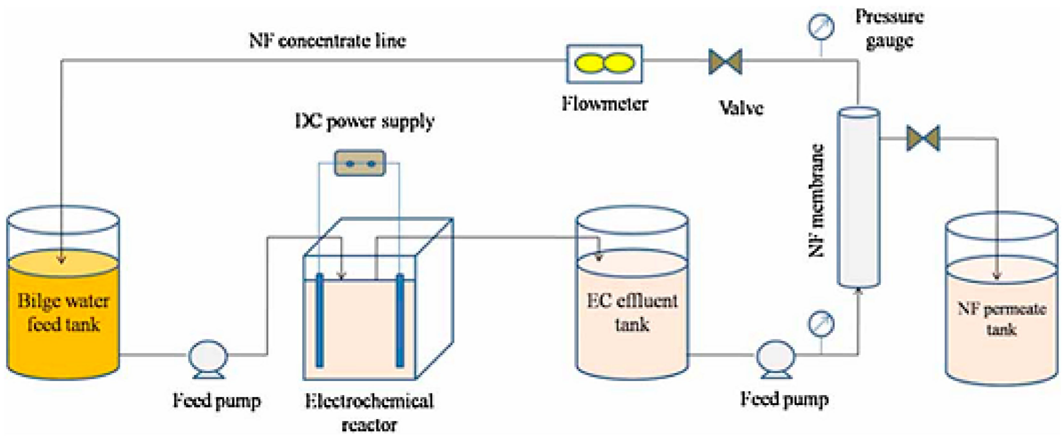

Figure 15. A schematic plot of the EC/NF integrated system of the case study [71].

Photocatalytic oxidation generally refers to the decomposition of oil substances using ultraviolet lamps, which has the advantages of high treatment efficiency and no pollution. However, the high energy consumption restricts its application. Therefore, photocatalytic oxidation is often used in combination with other methods in the oxidation decomposition reaction of marine oily sewage. Cazoir et al. [72] combined photocatalysis and air-stripping in a cylindrical 1 L batch liquid phase reactor for hydrocarbon removal from a real oily bilge water (Figure 16). Their results showed that the hydrocarbon oil index could be reduced to its maximum permissible value of 15 ppm in only 8.5 h.

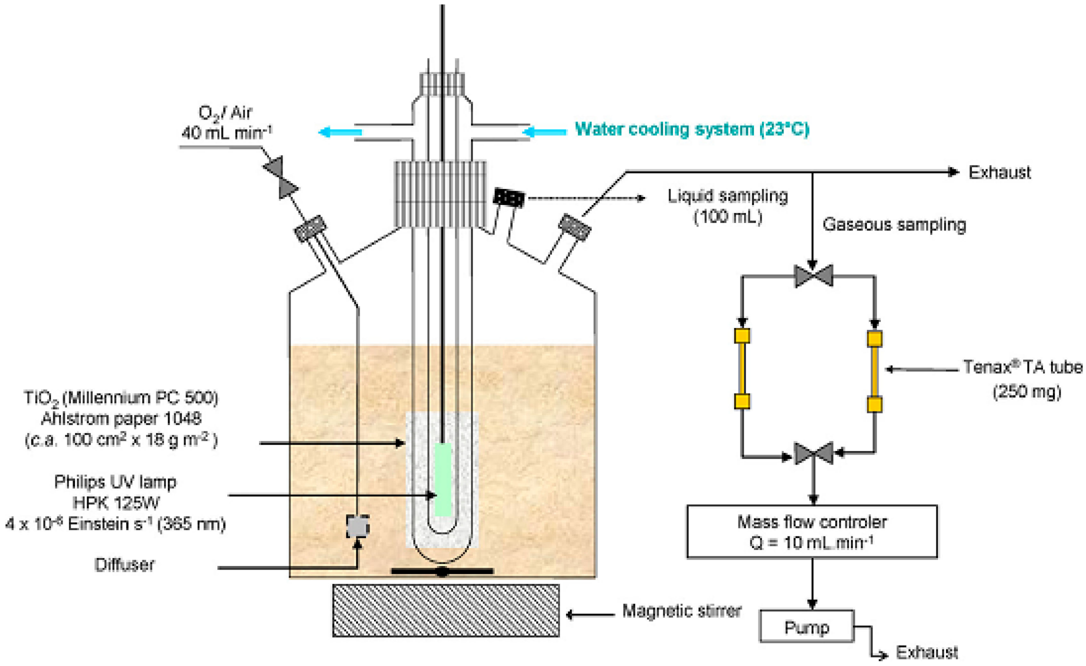

Figure 16. Schematic of the 1 L batch photo reactor [72].

Eskandarloo et al. [73] designed a photo-electrochemical system for in situ generation of H_2O_2 and de-emulsification of bilge water (Figure 17). By coupling photocatalytic oxidation at the Pt/TiO_2 anode and electro-Fenton oxidation at the Fe_2O_3 loaded graphite cathode in an undivided cell, it was found that the hydroxyl radical (·OH) formed can quickly break the emulsification of oily bilge sewage. Their results showed that a COD removal efficiency of over 70% on oily bilge water by 240 min visible-light irradiation. After 10 consecutive experiments, the removal efficiency of COD and the generation of H_2O_2 showed no significant changes, and the catalytic activity could be maintained for a long time.

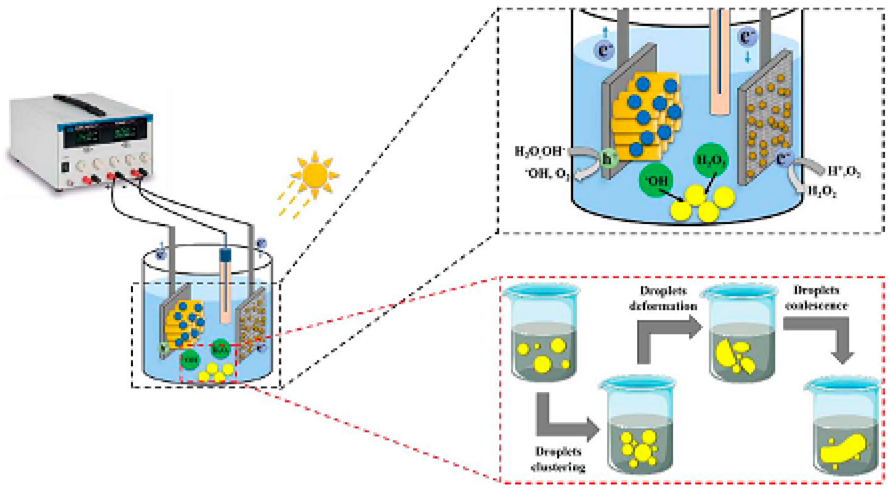

Figure 17. Schematic illustrations of proposed mechanisms and transient behavior of oil droplets during photo-electrochemical treatment [73].

Moslehyani et al. [74–76] studied the hybrid system consisting of a photocatalytic reactor and ultrafiltration membrane for the degradation and separation of oily bilge wastewater (Figure 18). In the PR-UF system, UV irradiation was made on a TiO_2 photocatalyst. The catalyst particles were suspended in the oily water in PR and ultrafiltered by UF hollow fibers. Their results showed that more than 90% of bilge wastewater is degraded by the photocatalytic reaction while the degradation rate of bilge wastewater increased to 99% by combining with an ultrafiltration nanocomposite membrane.

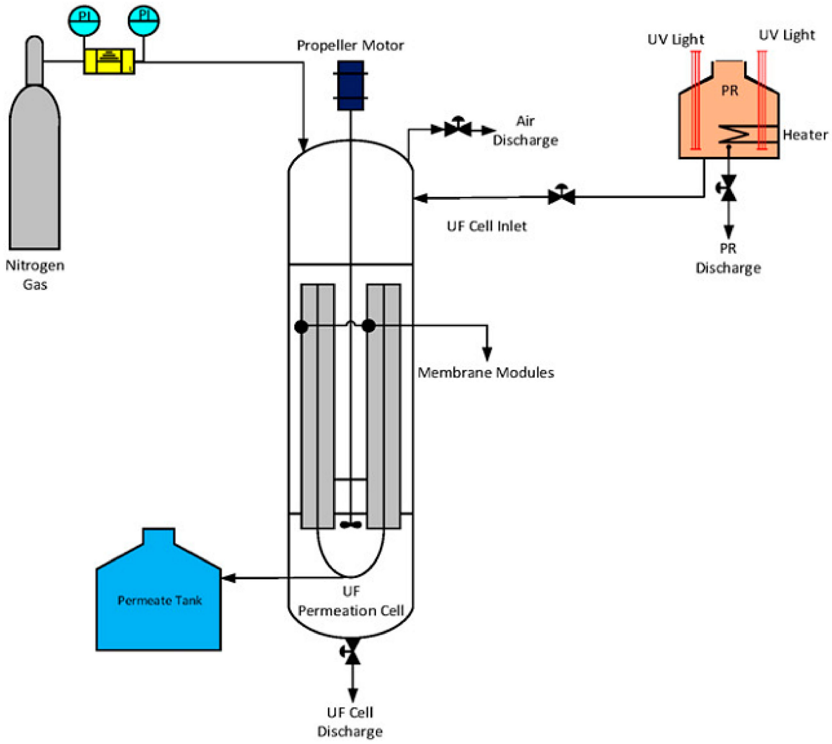

Figure 18. Schematic diagram of the PR-UF system [74].

Mancini et al. [77] verified the feasibility of treating an emulsified oily and salty wastewater (slop) through a chemical coagulation pretreatment followed by a granular activated carbon (GAC) filtration including the on-line bio-regeneration of the exhaust carbons (Figure 19). Their results showed that aluminum sulfate ($Al_2(SO_4)_3$) operated well (30% of COD removal) when applied at the optimal dose (90 mg/L) in the coagulation pre-treatment. The results from the column filtration tests indicated the feasibility of using the selected GAC to achieve the respect of the discharge limits in the slops treatment with a carbon usage rate in the range 0.1–0.3 kg/m^3 of treated effluent. Bioregeneration of the spent GAC was performed through the use of the marine hydrocarbonoclastic bacterium *Alcanivorax borkumensis* SK2. The preliminary cost evaluation indicates that one cycle of biological regeneration of the spent AC could achieve approximately 90% of the initial sorption capacity, which reduces the operation costs and the need for waste disposal.

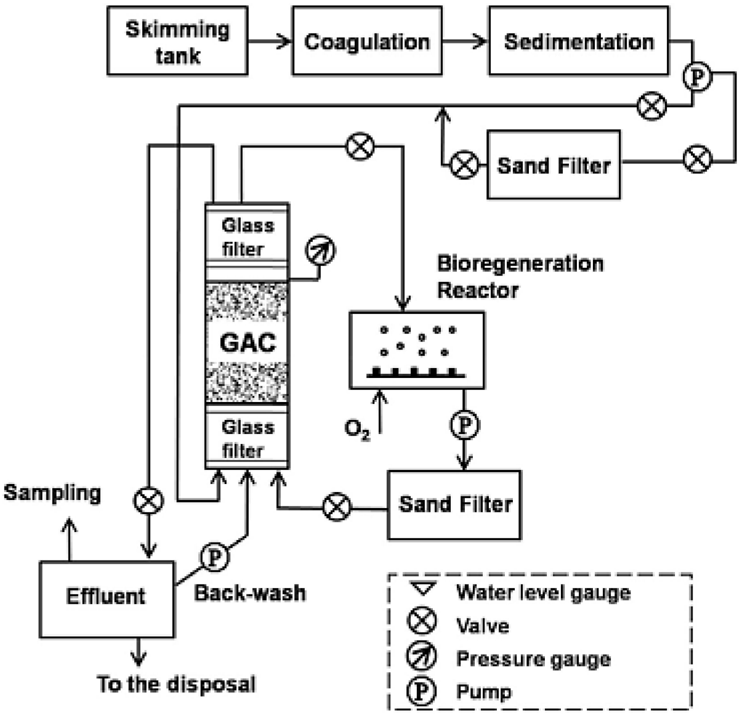

Figure 19. Schematic representation of the plant for treatment of slops as proposed [77].

While many hybrid systems have achieved success, some scholars have attempted to combine the data simulation technology with the experimental technology in treatment of marine oily wastewater, realizing interdisciplinary cross-association. Jing et al. [78,79] used an artificial neural network model (ANN) to predict the degradation process of polycyclic aromatic hydrocarbons (PAHs) in marine oily wastewater under UV irradiation. The photodegradation kinetics of naphthalene in natural seawater was studied by a full factorial design of experiments (DOE). The results showed that the method can remove all naphthalene pollutants, and the neural network model is consistent with the experimental results. Piao et al. [80] studied the effects of geometric features on oil-recovery rate based on a two-dimensional numerical simulation and proposed a new design of oil-water separation system. The working mechanism of the separator is to utilize the density difference between oil and water. They realized that the effective separation of layered oil could be strengthened by adding momentum to oil-water mixture flow through a "U-shaped" passage. They also used a basic experimental test with a three-dimensional oil-water separator model to optimize the conditions to maximize the oil recovery.

Table 4. Application progress of the combined method in the treatment of oily wastewater from ships.

Combined Method	Experiment Condition	Source of Oily Wastewater	Experiment Result	Ref
Electrocoagulation/nanofiltration (EC/NF) process	Aluminum electrode, EC/NF integrated system, commercial NF270 flat-sheet membrane, pH = 8, t = 90 min, 10 V of potential	Bilge water from Mersin Waste Receiving Facilities in Turkey with 423 mg/L of COD	52% COD after EC, 74% COD and <150 mg COD/L after EC/NF integrated process	[71]
Photocatalysis and air-stripping	Photo catalysis and air-stripping in a cylindrical 1 L batch liquid phase reactor for 8.5 h	Bilge water from a French military ship	<15ppm oil	[72]
Photo-electrochemical system	Photocatalytic oxidation on Pt/TiO_2 anode under visible light irradiation, electro-Fenton oxidation on Fe_2O_3 supported graphite cathode	Simulated bilge water with diesel fuel marine, lubricating oil, detergents and surfactants	70% COD, continuous operation for 10 times	[73]
Photocatalytic reactor-ultrafiltration (PR-UF)	Photolysis with UV light for 6 h, ultrafiltration with PVDF/HNT membrane for 1 h	Bilge water from Malaysian oil tanker with 400 ml/L of hydrocarbon concentration	>80% TOC and >90% hydrocarbons by photolysis; >99% hydrocarbons by ultrafiltration	[74–76]
Coagulation-filtration-bioregeneration	90 mg/L ($Al_2(SO_4)_3$, GAC column filtration with 0.1–0.3 kg/m^3 carbon usage rate, biological regeneration process with marine hydrocarbonoclastic bacterium *Alcanivorax borkumensis* SK2.	Emulsified oily bilge water from the barges transiting in the industrial port of Augusta (Sicily)	30% COD by coagulation; reached emission limit by filtration; 90% of the initial sorption capacity by regeneration process	[77]
ANN-UV irradiation	UV light, ANN with DOE to predict the degradation process of PAHs	Synthetic marine oily wastewater	All naphthalene pollutants were removed, and ANN is consistent with the experimental results.	[78,79]
Numerical simulation and Gravity separation	Used a two-dimensional numerical simulation and proposed a new design of oil-water separation system with a "U-shaped" passage	Synthetic marine oily wastewater	>95% oil-recovery rate	[80]

4. Discussion

There is no one-size-fits-all method to remove all pollutants from marine oily wastewater due to their diverse forms, complex composition, and variable content. Application scope and feature analysis of different technologies are shown in Table 5. Although the marine oily wastewater treatment technology has been widely concerned and studied by researchers at home and abroad, and some treatment technologies have made great progress, most of them are still at the lab scale, and there are still many problems to be solved in order to realize large-scale application in practical treatment. (1) As far as we know, filtration of oils selectively and effectively from water have been achieved by developing functional materials with special wettability. However, traditional superoleophobic surface-coated materials suffer from mechanical damage and loss of their superoleophobicity in high-salinity environments. Additionally, they also have limitations for complicated fabrication procedures that require high cost, time consuming, and chemical preparation processes. Therefore, it is essential to fabricate new material that can be bent and physically manipulated without diminishing the oil/water-wetting properties as well as offer excellent antifouling properties and environmental adaptability. Meanwhile, it would be a good choice to introduce endless and cheap biomass resources into the preparation of filtration materials [81]. On the other hand, the research on the mechanisms behind the superoleophobicity, antifouling, and self-cleaning properties of materials should be strengthened.

(2) Currently, some applications in marine oily wastewater have reached a significant development by membrane separation with UF or RO. Nevertheless, several challenges still need to be addressed in order to overcome the disadvantages in terms of separation performance, antifouling property, and long-time stability. For example, according to the European emission standards, the discharge of marine oil sewage must below 5 ppm. That means the current ship oil/water separator must be equipped with third-stage membrane separation to deal with emulsified oil and dissolved oil. Therefore, it is of great significance to develop novel membranes with high-performance pores and high separation efficiency. Meanwhile, membranes are susceptible to fouling at extreme pH levels, and high saline or surfactant concentrations. They are not beneficial for high-efficiency oil/water separation. How to improve the antifouling property of membranes and achieving long-term cyclic stability will be dominant requirements in future.

(3) The adsorption method has shown good results in the treatment of emulsified oil and dissolved oil wastewater generated by ship operational discharge and offshore oil spills. However, the synthesis and preparation of materials are time-consuming, complex, and expensive, which limits their application in emergency situations. Porous materials with higher adsorption capacity, excellent mechanical properties, and advantages of reuse will stand out in future offshore oil spill sewage treatments. New adsorbents with low cost, high selectivity, high adsorption capacity, high reuse rate, and simple preparation process will be the research hotspots in this field.

(4) In addition, electrochemical treatment of oily sewage from ships is regarded as an efficient and feasible method. However, the demand for electricity leads to high operation cost, and a large number of electrode metals will be released, which will cause another environmental problem. Undoubtedly, the future trend should be to strengthen the research and development of new electrode materials to ensure lower energy consumption and less pollutant precipitation.

(5) Biological treatment has the advantages of low investment and low operating cost, but the cultivation process involving microbial strains is time-consuming and sometimes needs to be supplemented by expensive enzyme action, which has limitations in the treatment of oily sewage from ships. How to cooperate with other treatment technologies and give full play to the advantages of low operating cost of biological treatment still needs to be further explored.

(6) Most ship oil and sewage treatment studies are focused on the separation of oil and water, and there are limited studies on the simultaneous treatment of other pollutants in oily water. For example, the presence of hazardous metal ions and harmful chemicals in oily sewage can have more

harmful effects, and how to simultaneously treat these pollutants and reduce their toxicity also requires future attention.

(7) Energy consumption is an important aspect that needs to be considered in the oil-bearing sewage treatment technology of ships. At present, renewable energy technology has been applied in the field of sewage treatment, which can help save energy and reduce consumption, reduce treatment cost, additional pollutant discharge, and environmental pressure. Therefore, the best way to reduce energy consumption is to combine oily wastewater treatment with renewable energy or clean energy in the future.

(8) The traditional single oil-bearing sewage treatment technology for ships is often inefficient and ineffective. The combination of multiple processes can effectively treat various pollution components in oil-bearing sewage and achieve a very high oil-removing effect. In recent years, scholars at home and abroad have made extensive attempts to degrade ship oily wastewater by joint treatment. Most of the reported treatment methods are expensive, cannot be implemented on a commercial scale, and cause environmental pollution. Therefore, joint treatment technologies with synergistic effects will be one of the future development directions.

Table 5. Comparison of commonly used methods of marine oily wastewater treatment.

Treatment Method		**Application Scope**	**Characteristic**
Physical treatment	Gravity separation method	Floating oil and dispersed oil	Low cost, simple device, easy to operate, but large area, limited separation capacity and poor treatment effect on emulsified oil.
	Coagulation separation method	Dispersed oil and emulsified oil	Low cost, small equipment, easy to operate, poor treatment effect with surfactant.
	Filtration separation method	Dispersed oil, emulsified oil, dissolved oil and hazardous metal ions	Low cost on equipment, but the filter need to be rinsed repeatedly
Chemical treatment	Adsorption method	Emulsified oil and dissolved oil, detergents	High cost on equipment, small area, adsorbents regeneration difficult.
	Electrochemical oxidation method	Emulsified oil, hazardous metal ions	High cost, large power consumption, complex device, but low space requirements and efficient treatment in a short time.
Biological treatment	Microbial metabolism method	Dissolved oil	Low cost, no additional chemical operation, but time-consuming, low efficiency and difficult to handle on a large scale.
	Activated sludge method	Emulsified oil and dissolved oil	Low cost, low treatment capacity, easy to automatic system control, but large area, strong resistance.
	Biofilm method	Emulsified oil and dissolved oil	Low cost, simple operation and good separation effect, good separation effect and impact resistance.

5. Conclusions

Marine oil pollution has brought significant negative impacts to the ecological environment, human security, and economic development, which has attracted great attention of the international community. With increasingly strict environmental protection laws and standards, the existing oil-bearing sewage treatment technology of ships can no longer meet the demand. In the future, the development trend of oil-bearing sewage treatment technology of ships will definitely move forward towards efficiency, environmental protection, economy, and energy conservation, and the concept of sustainable development will also become more prominent.

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References

1. Jernelov, A. The threats from oil spills: Now, then, and in the future. *Ambio* **2010**, *39*, 353–366. [CrossRef] [PubMed]
2. Burgherr, P. In-depth analysis of accidental oil spills from tankers in the context of global spill trends from all sources. *J. Hazard. Mater.* **2007**, *140*, 245–256. [CrossRef] [PubMed]
3. Soto, L.A.; Botello, A.V.; Licea-Duran, S.; Lizarraga-Partida, M.L.; Yanez-Arancibia, A. The environmental legacy of the Ixtoc-I oil spill in Campeche Sound, southwestern Gulf of Mexico. *Front. Mar. Sci.* **2014**, *1*. [CrossRef]
4. Buskey, E.J.; White, H.K.; Esbaugh, A.J. Impact of oil spills on marine life in the Gulf of Mexico: Effect on plankton, nekton, and deep-sea benthos. *Oceanography* **2016**, *29*, 174–181. [CrossRef]
5. Beyer, J.; Trannum, H.C.; Bakke, T.; Hodson, P.V.; Collier, T.K. Environmental effects of the Deepwater Horizon oil spill: A review. *Mar. Pollut. Bull.* **2016**, *110*, 28–51. [CrossRef]
6. Cho, D.O. The effects of the M/V Sea Prince accident on maritime safety management in Korea. *Mar. Policy* **2007**, *31*, 730–735. [CrossRef]
7. Ung, S.T. Human error assessment of oil tanker grounding. *Saf. Sci.* **2018**, *104*, 16–28. [CrossRef]
8. Zafirakou, A.; Themeli, S.; Tsami, E.; Aretoulis, G. Multi-criteria analysis of different approaches to protect the marine and coastal environment from oil spills. *J. Mar. Sci. Eng.* **2018**, *6*, 125. [CrossRef]
9. Wang, F.J.; Lei, S.; Xue, M.S.; Ou, J.F.; Li, W. In situ separation and collection of oil from water surface via a novel superoleophilic and superhydrophobic oil containment boom. *Langmuir* **2014**, *30*, 1281–1289. [CrossRef] [PubMed]
10. Bullock, R.J.; Perkins, R.A.; Aggarwal, S. In-situ burning with chemical herders for Arctic oil spill response: Meta-analysis and review. *Sci. Total Environ.* **2019**, *675*, 705–716. [CrossRef] [PubMed]
11. Prince, R.C. Oil spill dispersants: Boon or bane? *Environ. Sci. Technol.* **2015**, *49*, 6376–6384. [CrossRef] [PubMed]
12. Socolofsky, S.A.; Gros, J.; North, E.; Boufadel, M.C.; Parkerton, T.F.; Adams, E.E. The treatment of biodegradation in models of sub-surface oil spills: A review and sensitivity study. *Mar. Pollut. Bull.* **2019**, *143*, 204–219. [CrossRef]
13. Zhang, T.; Li, Z.D.; Lu, Y.F.; Liu, Y.; Yang, D.Y.; Li, Q.R.; Qiu, F.X. Recent progress and future prospects of oil-absorbing materials. *Chin. J. Chem. Eng.* **2019**, *27*, 1282–1295. [CrossRef]
14. Wilewska-Bien, M.; Anderberg, S. Reception of sewage in the Baltic Sea-The port's role in the sustainable management of ship wastes. *Mar. Policy* **2018**, *93*, 207–213. [CrossRef]
15. Lagring, R.; Degraer, S.; de Montpellier, G.; Jacques, T.; Van Roy, W.; Schallier, R. Twenty years of Belgian North Sea aerial surveillance: A quantitative analysis of results confirms effectiveness of international oil pollution legislation. *Mar. Pollut. Bull.* **2012**, *64*, 644–652. [CrossRef]

16. Ministry of Ecology and Environment. *General Administration of Quality Supervision, Inspection and Quarantine of the People's Republic of China, Discharge Standard for Water Pollutants from Ships: GB 3552-2018*; Environmental Science Press: Beijing, China, 2018.
17. Mysore, D.; Viraraghavan, T.; Jin, Y.C. Oil/water separation technology-A review. *J. Residuals Sci. Tech.* **2006**, *3*, 5–14.
18. Le, T.V.; Imai, T.; Higuchi, T.; Yamamoto, K.; Sekine, M.; Doi, R.; Vo, H.T.; Wei, J. Performance of tiny microbubbles enhanced with "normal cyclone bubbles" in separation of fine oil-in-water emulsions. *Chem. Eng. Sci.* **2013**, *94*, 1–6. [CrossRef]
19. Kratochvil, M.J.; Manna, U.; Lynn, D.M. Superhydrophobic polymer multilayers for the filtration-and absorption-based separation of oil/water mixtures. *J. Polym. Sci. Pol. Chem.* **2017**, *55*, 3127–3136. [CrossRef]
20. Zhu, Y.Z.; Wang, J.L.; Zhang, F.; Gao, S.J.; Wang, A.Q.; Fang, W.X.; Jin, J. Zwitterionic nanohydrogel grafted PVDF membranes with comprehensive antifouling property and superior cycle stability for oil-in-Water emulsion separation. *Adv. Funct. Mater.* **2018**, *28*. [CrossRef]
21. Liu, Y.A.; Su, Y.L.; Guan, J.Y.; Cao, J.L.; Zhang, R.N.; He, M.R.; Jiang, Z.Y. Asymmetric aerogel membranes with ultrafast water permeation for the separation of oil-in-water emulsion. *ACS Appl. Mater. Interfaces* **2018**, *10*, 26546–26554. [CrossRef]
22. Rana, M.; Chen, J.T.; Yang, S.D.; Ma, P.C. Biomimetic superoleophobicity of cotton fabrics for efficient oil-water separation. *Adv. Mater. Interfaces* **2016**, *3*. [CrossRef]
23. Padaki, M.; Murali, R.S.; Abdullah, M.S.; Misdan, N.; Moslehyani, A.; Kassim, M.A.; Hilal, N.; Ismail, A.F. Membrane technology enhancement in oil-water separation. A review. *Desalination* **2015**, *357*, 197–207. [CrossRef]
24. Li, Y.Q.; Zhang, H.; Fan, M.Z.; Zhuang, J.D.; Chen, L.H. A robust salt-tolerant superoleophobic aerogel inspired by seaweed for efficient oil-water separation in marine environments. *Phys. Chem. Chem. Phys.* **2016**, *18*, 25394–25400. [CrossRef] [PubMed]
25. Li, Y.Q.; Zhang, H.; Fan, M.Z.; Zheng, P.T.; Zhuang, J.D.; Chen, L.H. A robust salt-tolerant superoleophobic alginate/graphene oxide aerogel for efficient oil/water separation in marine environments. *Sci. Rep.* **2017**. [CrossRef]
26. Zhang, H.; Li, Y.Q.; Shi, R.H.; Chen, L.H.; Fan, M.Z. A robust salt-tolerant superoleophobic chitosan/nanofibrillated cellulose aerogel for highly efficient oil/water separation. *Carbohyd. Polym.* **2018**, *200*, 611–615. [CrossRef]
27. Cortese, B.; Caschera, D.; Federici, F.; Ingo, G.M.; Gigli, G. Superhydrophobic fabrics for oil/water separation through a diamond like carbon (DLC) coating. *J. Mater. Chem. A* **2014**, *2*, 6781–6789. [CrossRef]
28. Tomaszewska, M.; Orecki, A.; Karakulski, K. Treatment of bilge water using a combination of ultrafiltration and reverse osmosis. *Desalination* **2005**, *185*, 203–212. [CrossRef]
29. Karakulski, K.; Gryta, M. The application of ultrafiltration for treatment of ships generated oily wastewater. *Chem. Pap.* **2017**, *71*, 1165–1173. [CrossRef]
30. Hoang, A.T.; Bui, X.L.; Pham, X.D. A novel investigation of oil and heavy metal adsorption capacity from as-fabricated adsorbent based on agricultural by-product and porous polymer. *Energy Sources A* **2018**, *40*, 929–939. [CrossRef]
31. Nguyen, H.N.; Pignatello, J.J. Laboratory tests of biochars as absorbents for use in recovery or containment of marine crude oil spills. *Environ. Eng. Sci.* **2013**, *30*, 374–380. [CrossRef]
32. Abdelwahab, N.A.; Shukry, N.; El-Kalyoubi, S.F. Preparation and characterization of polymer coated partially esterified sugarcane bagasse for separation of oil from seawater. *Environ. Technol.* **2017**, *38*, 1905–1914. [CrossRef] [PubMed]
33. Vlaev, L.; Petkov, P.; Dimitrov, A.; Genieva, S. Cleanup of water polluted with crude oil or diesel fuel using rice husks ash. *J. Taiwan Inst. Chem. Eng.* **2011**, *42*, 957–964. [CrossRef]
34. Mukherjee, S.; Kansara, A.M.; Saha, D.; Gonnade, R.; Mullangi, D.; Manna, B.; Desai, A.V.; Thorat, S.H.; Singh, P.; Mukherjee, A.; et al. An ultrahydrophobic fluorous metal-organic framework derived recyclable composite as a promising platform to tackle marine oil spills. *Chem. Eur. J.* **2016**, *22*, 10937–10943. [CrossRef] [PubMed]
35. Rahmani, Z.; Shafiei-Alavijeh, M.; Kazemi, A.; Rashidi, A.M. Synthesis of MIL-101@nanoporous graphene composites as hydrophobic adsorbents for oil removal. *J. Taiwan Inst. Chem. Eng.* **2018**, *91*, 597–608. [CrossRef]

36. Eldin, M.S.M.; Ammar, Y.A.; Tamer, T.M.; Omer, A.M.; Ali, A.A. Development of low-cost chitosan derivatives based on marine waste sources as oil adsorptive materials: I. Preparation and characterization. *Desalin. Water Treat.* **2017**, *72*, 41–51. [CrossRef]
37. Doshi, B.; Repo, E.; Heiskanen, J.P.; Sirvio, J.A.; Sillanpaa, M. Effectiveness of N, O-carboxymethyl chitosan on destabilization of Marine Diesel, Diesel and Marine-2T oil for oil spill treatment. *Carbohyd. Polym.* **2017**, *167*, 326–336. [CrossRef] [PubMed]
38. Doshi, B.; Repo, E.; Heiskanen, J.P.; Sirvio, J.A.; Sillanpaa, M. Sodium salt of oleoyl carboxymethyl chitosan: A sustainable adsorbent in the oil spill treatment. *J. Clean. Prod.* **2018**, *170*, 339–350. [CrossRef]
39. Gupta, P.; Kandasubramanian, B. Directional fluid gating by Janus membranes with heterogeneous wetting properties for selective oil-water separation. *ACS Appl. Mater. Interfaces* **2017**, *9*, 19102–19113. [CrossRef]
40. Shiu, R.F.; Lee, C.L.; Hsieh, P.Y.; Chen, C.S.; Kang, Y.Y.; Chin, W.C.; Tai, N.H. Superhydrophobic graphene-based sponge as a novel sorbent for crude oil removal under various environmental conditions. *Chemosphere* **2018**, *207*, 110–117. [CrossRef]
41. Wang, G.; Zeng, Z.X.; Wu, X.D.; Ren, T.H.; Han, J.; Xue, Q.J. Three-dimensional structured sponge with high oil wettability for the clean-up of oil contaminations and separation of oil-water mixtures. *Polym. Chem.* **2014**, *5*, 5942–5948. [CrossRef]
42. Turco, A.; Primiceri, E.; Frigione, M.; Maruccio, G.; Malitesta, C. An innovative, fast and facile soft-template approach for the fabrication of porous PDMS for oil-water separation. *J. Mater. Chem. A* **2017**, *5*, 23785–23793. [CrossRef]
43. Mirshahghassemi, S.; Lead, J.R. Oil recovery from water under environmentally relevant conditions using magnetic nanoparticles. *Environ. Sci. Technol.* **2015**, *49*, 11729–11736. [CrossRef] [PubMed]
44. Mirshahghassemi, S.; Cai, B.; Lead, J.R. Evaluation of polymer-coated magnetic nanoparticles for oil separation under environmentally relevant conditions: Effect of ionic strength and natural organic macromolecules. *Environ. Sci. Nano* **2016**, *3*, 780–787. [CrossRef]
45. Furlan, P.Y.; Ackerman, B.M.; Melcer, M.E.; Perez, S.E. Reusable magnetic nanocomposite sponges for removing oil from water discharges. *J. Ship Prod. Des.* **2017**, *33*, 227–236. [CrossRef]
46. Cerqueira, A.A.; Souza, P.S.A.; Marques, M.R.C. Effects of direct and alternating current on the treatment of oily water in an electroflocculation process. *Braz. J. Chem. Eng.* **2014**, *31*, 693–701. [CrossRef]
47. Aswathy, P.; Gandhimathi, R.; Ramesh, S.T.; Nidheesh, P.V. Removal of organics from bilge water by batch electrocoagulation process. *Sep. Purif. Technol.* **2016**, *159*, 108–115. [CrossRef]
48. Rincon, G.J.; La Motta, E.J. Simultaneous removal of oil and grease, and heavy metals from artificial bilge water using electro-coagulation/flotation. *J. Environ. Manag.* **2014**, *144*, 42–50. [CrossRef]
49. Ulucan, K.; Kabuk, H.A.; Ilhan, F.; Kurt, U. Electrocoagulation process application in bilge water treatment using response surface methodology. *Int. J. Electrochem. Sci.* **2014**, *9*, 2316–2326.
50. Ulucan, K.; Kurt, U. Comparative study of electrochemical wastewater treatment processes for bilge water as oily wastewater: A kinetic approach. *J. Electroanal. Chem.* **2015**, *747*, 104–111. [CrossRef]
51. Sekman, E.; Top, S.; Uslu, E.; Varank, G.; Bilgili, M.S. Treatment of oily wastewater from port waste reception facilities by electrocoagulation. *Int. J. Environ. Res.* **2011**, *5*, 1079–1086.
52. Bilgili, M.S.; Ince, M.; Tari, G.T.; Adar, E.; Balahorli, V.; Yildiz, S. Batch and continuous treatability of oily wastewaters from port waste reception facilities: A pilot scale study. *J. Electroanal. Chem.* **2016**, *760*, 119–126. [CrossRef]
53. Korbahti, B.K.; Artut, K. Electrochemical oil/water demulsification and purification of bilge water using Pt/Ir electrodes. *Desalination* **2010**, *258*, 219–228. [CrossRef]
54. Korbahti, B.K.; Artut, K. Bilge water treatment in an upflow electrochemical reactor using Pt anode. *Sep. Sci. Technol.* **2013**, *48*, 2204–2216. [CrossRef]
55. Carlesi, C.; Ramírez, N.G.; Carvajal, D.; Hernandez, M.C.; Fino, D. Electrochemical treatment of bilge wastewater. *Desalin. Water Treat.* **2015**, *54*, 1556–1562. [CrossRef]
56. Xue, J.L.; Yu, Y.; Bai, Y.; Wang, L.P.; Wu, Y.N. Marine oil-degrading microorganisms and biodegradation process of petroleum hydrocarbon in marine environments: A review. *Curr. Microbiol.* **2015**, *71*, 220–228. [CrossRef] [PubMed]
57. Chanthamalee, J.; Wongchitphimon, T.; Luepromchai, E. Treatment of oily bilge water from small fishing vessels by PUF-immobilized *Gordonia* sp. JC11. *Water Air Soil Poll.* **2013**, *224*, 1601. [CrossRef]

58. Crisafi, F.; Genovese, M.; Smedile, F.; Russo, D.; Catalfamo, M.; Yakimov, M.; Giuliano, L.; Denaro, R. Bioremediation technologies for polluted seawater sampled after an oil-spill in Taranto Gulf (Italy): A comparison of biostimulation, bioaugmentation and use of a washing agent in microcosm studies. *Mar. Pollut. Bull.* **2016**, *106*, 119–126. [CrossRef]
59. Ma, C.; Liu, J.; Zhou, T. Study on characteristics of marine petroleum-degrading strains and their bioremediation utilization of carbon source spectrum. *J. Bionanosci.* **2015**, *9*, 127–134. [CrossRef]
60. Dokianakis, S.N.; Fountoulakis, M.S.; Kornaros, M.; Lyberatos, G. Biological treatment of sewage from ships in a sequencing batch reactor. *Fresen. Environ. Bull.* **2007**, *16*, 1608–1615.
61. Zhang, W.J.; Wei, Y.; Jin, Y. Full-scale processing by anaerobic baffle reactor, sequencing batch reactor, and sand filter for treating high-salinity wastewater from offshore oil rigs. *Processes* **2018**, *6*, 256. [CrossRef]
62. Choi, Y.I.; Ji, H.J.; Shin, D.Y.; Mansoor, S.; Kwan, M.J.; Lee, S.C.; Jung, J.H.; Jung, B.G.; Sung, N.C.; Wang, J.P. An evaluation of the water quality characteristics of shipboard sewage disposal and usability based on water quality enhancement. *Appl. Sci.* **2019**, *9*, 418. [CrossRef]
63. Uma, V.; Gandhimathi, R. Organic removal and synthesis of biopolymer from synthetic oily bilge water using the novel mixed bacterial consortium. *Bioresource Technol.* **2018**, *273*, 169–176. [CrossRef] [PubMed]
64. Le-Clech, P.; Chen, V.; Fane, T.A.G. Fouling in membrane bioreactors used in wastewater treatment. *J. Membr. Sci.* **2006**, *284*, 17–53. [CrossRef]
65. Meng, F.G.; Chae, S.R.; Drews, A.; Kraume, M.; Shin, H.S.; Yang, F.L. Recent advances in membrane bioreactors (MBRs): Membrane fouling and membrane material. *Water Res.* **2009**, *43*, 1489–1512. [CrossRef]
66. Sun, C.; Leiknes, T.O.; Weitzenbock, J. The effect of bilge water on a biofilm-MBR process in an integrated shipboard wastewater treatment system. *Desalination* **2009**, *236*, 56–64. [CrossRef]
67. Sun, C.; Leiknes, T.O.; Weitzenbock, J. Development of a biofilm-MBR for shipboard wastewater treatment: The effect of process configuration. *Desalination* **2010**, *250*, 745–750. [CrossRef]
68. Wei, Y.J.; Li, G.Y. Membrane fouling behavior and microbial community succession in a submerged membrane bioreactor treating harbor oily wastewater. *J. Zhejiang Univ. Sci. A Appl. Phys. Eng.* **2016**, *17*, 745–757. [CrossRef]
69. Vyrides, I.; Drakou, E.M.; Ioannou, S.; Michael, F.; Gatidou, G.; Stasinakis, A.S. Biodegradation of bilge water: Batch test under anaerobic and aerobic conditions and performance of three pilot aerobic Moving Bed Biofilm Reactors (MBBRs) at different filling fractions. *J. Environ. Manag.* **2018**, *217*, 356–362. [CrossRef]
70. Emadian, S.M.; Hosseini, M.; Rahimnejad, M.; Shahavi, M.H.; Khoshandam, B. Treatment of a low-strength bilge water of Caspian Sea ships by HUASB technique. *Ecol. Eng.* **2015**, *82*, 272–275. [CrossRef]
71. Akarsu, C.; Ozay, Y.; Dizge, N.; Gulsen, H.E.; Ates, H.; Gozmen, B.; Turabik, M. Electrocoagulation and nanofiltration integrated process application in purification of bilge water using response surface methodology. *Water Sci. Technol.* **2016**, *74*, 564–579. [CrossRef]
72. Cazoir, D.; Fine, L.; Ferronato, C.; Chovelon, J.M. Hydrocarbon removal from bilgewater by a combination of air-stripping and photocatalysis. *J. Hazard. Mater.* **2012**, *235*, 159–168. [CrossRef] [PubMed]
73. Eskandarloo, H.; Selig, M.J.; Abbaspourrad, A. In situ H_2O_2 generation for de-emulsification of fine stable bilge water emulsions. *Chem. Eng. J.* **2018**, *335*, 434–442. [CrossRef]
74. Moslehyani, A.; Mobaraki, M.; Matsuura, T.; Ismail, A.F.; Othman, M.H.D.; Chowdhury, M.N.K. Novel green hybrid processes for oily water photooxidation and purification from merchant ship. *Desalination* **2016**, *391*, 98–104. [CrossRef]
75. Moslehyani, A.; Ismail, A.F.; Othman, M.H.D.; Isloor, A.M. Novel hybrid photocatalytic reactor-UF nanocomposite membrane system for bilge water degradation and separation. *RSC Adv.* **2015**, *5*, 45331–45340. [CrossRef]
76. Moslehyani, A.; Mobaraki, M.; Isloor, A.M.; Ismail, A.F.; Othman, M.H.D. Photoreactor-ultrafiltration hybrid system for oily bilge water photooxidation and separation from oil tanker. *React. Funct. Polym.* **2016**, *101*, 28–38. [CrossRef]
77. Mancini, G.; Panzica, M.; Fino, D.; Cappello, S.; Yakimov, M.M.; Luciano, A. Feasibility of treating emulsified oily and salty wastewaters through coagulation and bio-regenerated GAC filtration. *J. Environ. Manag.* **2016**, *203*, 817–824. [CrossRef]
78. Jing, L.; Chen, B.; Zhang, B.Y.; Zheng, J.S.; Liu, B. Naphthalene degradation in seawater by UV irradiation: The effects of fluence rate, salinity, temperature and initial concentration. *Mar. Pollut. Bull.* **2014**, *81*, 149–156. [CrossRef]

79. Jing, L.; Chen, B.; Zhang, B.Y.; Li, P. Process simulation and dynamic control for marine oily wastewater treatment using UV irradiation. *Water Res.* **2015**, *81*, 101–112. [CrossRef]
80. Piao, L.F.; Kim, N.; Park, H. Effects of geometrical parameters of an oil-water separator on the oil-recovery rate. *J. Mech. Sci. Technol.* **2017**, *31*, 2829–2837. [CrossRef]
81. Doshi, B.; Sillanpaa, M.; Kalliola, S. A review of bio-based materials for oil spill treatment. *Water Res.* **2018**, *135*, 262–277. [CrossRef]