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Review on Biodiesel Wastewater Treatment Using Membrane Bioreactor (MBR) Technology

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Abstracts: Reclaiming and reusing water are both economically and environmentally significant concerns for enterprises to consider. The creation of biodiesel fuel is one of the outcomes of the development of technology that provide alternatives to fuel derived from petroleum. Most of the research done on biodiesel thus far has concentrated on its creation and fuel qualities, while environmental control has received very little attention. This review paper discusses biodiesel fuel production from edible oil, non-edible oil, animal fats, and other sources, as well as the transesterification process and the main source of biodiesel wastewater. With a growing population, depleting natural resources, and a plethora of environmental challenges, it is crucial that we find ways to reuse, recycle, and reproduce. This paper provides a summary of research done on the topic of recycling biodiesel wastewater. The pretreatment of wastewater from biodiesel production leads to an improvement in the efficiency of biological processes. By combining the biological process with membrane filtration, MBR technology is developed, which has many advantages over traditional approaches. This review has also examined the relevance, potential, and feasibility of using MBR Technology to treat biodiesel wastewater released from a biodiesel production process. Reusing wastewater after it has been treated with MBR Technology is a further feasible option.

Keywords: Membrane Bioreactor, Biodiesel, Biodiesel Wastewater Treatment, Membrane, Integrated Approach.

1. INTRODUCTION

The world's population is growing, which raises the energy demand every two years. This need for energy drives people to look for alternative energy sources like solar power or the creation of biofuels from waste materials. The project's motive looks to create an effective method of producing biofuels as an alternative fuel. A substitute fuel must meet a number of requirements, including sustainability, biodegradability, non-toxicity, low sulphur content, low carbon monoxide generation, and an emission profile devoid of aromatics. The majority of these requirements are satisfied by biodiesel or fatty acid methyl esters (FAME), which also benefits the environment by reusing waste fat and oil [1]. When the globe began to discover alongside developing non-conventional energy sources, motivated by the diminishing of non-renewable fuel sources, the concept of utilizing biodiesel fuel emerged. The high reliance on fossil fuels has resulted in supply and pricing unpredictability. Water, sun, wind, and biofuels are a few alternatives that can take the place of fossil fuels. The growing awareness about the environmental effects of emissions caused by the burning of traditional fossil fuels plus falling in production of domestic oil are further factors contributing to the rise in demand for biodiesel [2]. When triglycerides obtained through vegetable oils/fats are trans-esterified with alcohols (methanol, ethanol) in the presence of a homogeneous base catalyst such as NaOH, or KOH. Then fatty acid alkyl esters (that is, biodiesel) plus glycerol are formed. In order to eliminate impurities including suspended particles and catalysts, water is typically added to the biodiesel during the last stage of manufacturing. Depending on how many contaminants are present in the methyl ester, this washing phase is repeated 2–5 times [3-5].

For every 100 liters of biodiesel produced, this method produces 20 to 120 liters of effluent. Because seeds are the main raw source used in the production of biodiesel, the wastewater has an intense yellow-creamy color, is highly turbid, also with an unpleasant smell, has a rich organic load, and increased levels of suspended plus dissolved solids, sodium, nitrogen, phosphorus, and potassium. It also has increased levels of chemical oxygen demand (COD) also biological oxygen demand (BOD) [2]. The biodiesel production procedures had a major effect on the properties of the biodiesel wastewater, and it was also discovered that the pH of the wastewater is impacted by the cycle of the biodiesel plant. shown the presence of soap and glycerol in biodiesel wastewater and suggested that this extremely stable colloid wastewater be properly controlled and treated. Due to its high COD, suspended solids, low nitrogen and phosphorus concentration, and high suspended solids, this wastewater has some challenges in naturally decomposing. This wastewater's discharge into the public sewer system might cause clogged drains and microbial activity to be disrupted [4]. The wastewater treatment issue has to be successfully resolved, owing to the significant volume of biodiesel wastewater produced during the process of biodiesel production. The wastewater from the cement industries was incinerated as a different option in the past. No more investigation was announced, though. The cost of using the incineration method is said to be less than the cost of paying a water treatment establishment, but it is compared to all other treatment methods for industrial wastewater, it is still pricey, according to previous studies, which have typically concentrated on the production of biodiesel, do not take environmental management, treatment, and mitigation into account [4,5].

Coagulation, electrocoagulation, biological process adsorption, and microbial fuel cell systems are some of the treatment options that have been documented. Coagulant is introduced during the coagulation process to quickly separate the solution's small particle composition. Destabilized, these particles flocculate to form bigger, settleable flocs. The process of flocculation is in charge of eliminating impurities including metals and hazardous waste as well as lowering COD, BOD, SS, turbidity, and color. The coagulation process involves two stages of mixing: fast and gradual mixing. Slow mixing increases the size of the flocs whereas quick mixing aids in the uniform dispersion of coagulants in a water-based solution [2,4]. The kind of coagulant employed or the amount of pre-hydrolyzed metal salt used, the dosage of the coagulant, the pH, the mixing rate, and the settling time are all variables that might impact how well the coagulation process works. Alum, polyamine, poly-aluminum chlorides, ferric chloride, and titanium chloride are just a few of the many coagulants that are employed [2,6,7]. Coagulation and flocculation are effective techniques for pre-treating wastewater from biodiesel production, according to research. Additional treatment is required to get better % removal for COD, suspended particles, color, and oil & grease [3].

The literature for biodiesel wastewater also highlights AOP like heterogeneous photocatalysis, photo-Fenton, and ozonation, however, some authors point out the need for more studies to develop or improve the management of the amount of residue that is anticipated to be created in the future. The catalytic decomposition of hydrogen peroxide in the context of ferrous ions in a water-based solution is the foundation of the Fenton process, which results in the generation of hydroxyl radicals (OH^-) [8, 9]. Based on the study of various methods to treat biodiesel wastewater, the conclusion is that it is essential to advance technology and improve water management in order to meet the growing demand for water while using less energy. A high-quality, well-treated effluent serves two purposes: it increases the likelihood that the water resource may be reused and releases just the legal amount of toxins into water bodies. Wastewater may be recovered as much as possible, which can be done with the use of alternative technologies such as MBR. Because MBR facilities combine biological processes with membrane filtration, which has several advantages over more conventional techniques, they are frequently employed for treating municipal and industrial wastewater. The membrane module and biological reactor tank that make up the MBR are combined [10]. The entire treatment process can be summarized in the Figure 1.

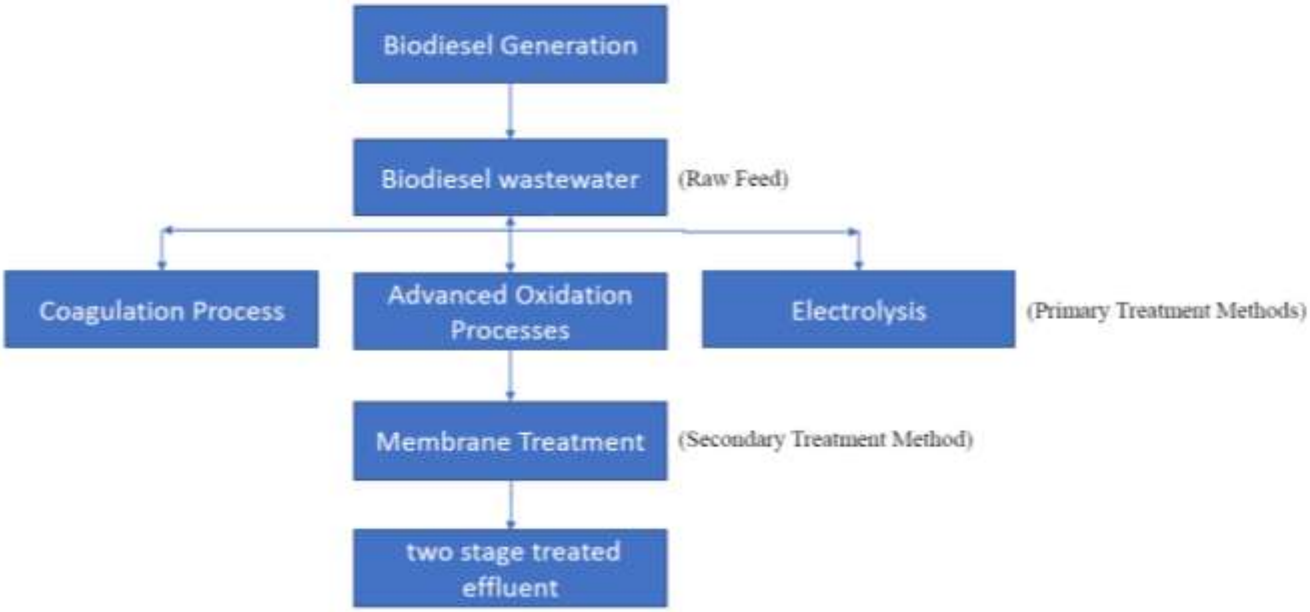

Figure.1. Treatment Process Approach2. MATERIEL AND METHODS

2. BIODIESEL PREPARATION AND WASTEWATER GENERATION

2.1. Biodiesel feedstock

Vegetable oil has historically been the primary source of biodiesel. The sorts of vegetable oils that are offered depend on the country's climate and soil characteristics [11]. Around 90% of the biodiesel produced in Thailand utilizes palm-based oil as a primary ingredient [6]. Soybean oil is the biodiesel feedstock that is most frequently utilized in the United States [12]. Edible oils, non-edible oils, and recyclable sources and wastes can all be used as biodiesel feedstock. Triglycerides and/or free fatty acids (FFAs) are crucial parts of the feedstock used to make biodiesel [13]. All sources of fatty acids are suitable for use in the creation of biodiesel. Based on their FFAs, biodiesel feedstock may be categorized [14].

Palm oil is a widely used feedstock for biodiesel in Malaysia [11]. Due to its accessibility, numerous uses, and ease of discovery, palm oil has a large share in the biodiesel manufacturing sector [13]. It is regarded as one of the sources with a high oil output. Jatropha curcas Linnaeus seed oil is one of the more promising non-edible sources for the feedstock of biodiesel. One option to lower the cost of manufacturing is to use jatropha oil as the main feedstock for making biodiesel [15] Because of India's huge reliance on petroleum imports and the prevalence of this non-edible source there, scientists have been looking into whether jatropha oil may be used to make biodiesel that has qualities that are comparable to or even identical to those of diesel oil [16]. Moreover, it is simple to locate and grow, especially on rocky, sandy, and salty soils [17]. The seeds of the Jatropha curcas plant, which have a 25–30% oil content, are the main source of oil in the plant.

The use of inexpensive feed, such as used/waste cooking oil (WCO), is one of the creative ways for attaining low-cost biodiesel manufacturing [18]. Using WCO has several advantages since it can stop the substance from being released into the drainage system [19]. The total cost of producing biodiesel frequently includes the cost of feed, the cost of transforming the unprocessed material, the price of oil extraction and cleaning, the expense of transesterification, the electricity price, the cost of transportation, and the cost of working capital [20,21]. The price of raw materials and operational costs are the two main elements that have the greatest impact on production costs [11]. The cost of the feedstock made up roughly 80% of the cost of producing biodiesel. Their investigation into the manufacture of palm oil-based biodiesel under various reaction process circumstances revealed that the price of palm oil (80%), the price of methanol (15%), and the cost of energy (5%), were the three main contributors to the cost of palm oil-based biodiesel production [22]. Edible oil, non-edible oil, animal fats, and other sources are the four primary forms of feedstock for biodiesel synthesis, as summarized in Figure 2.

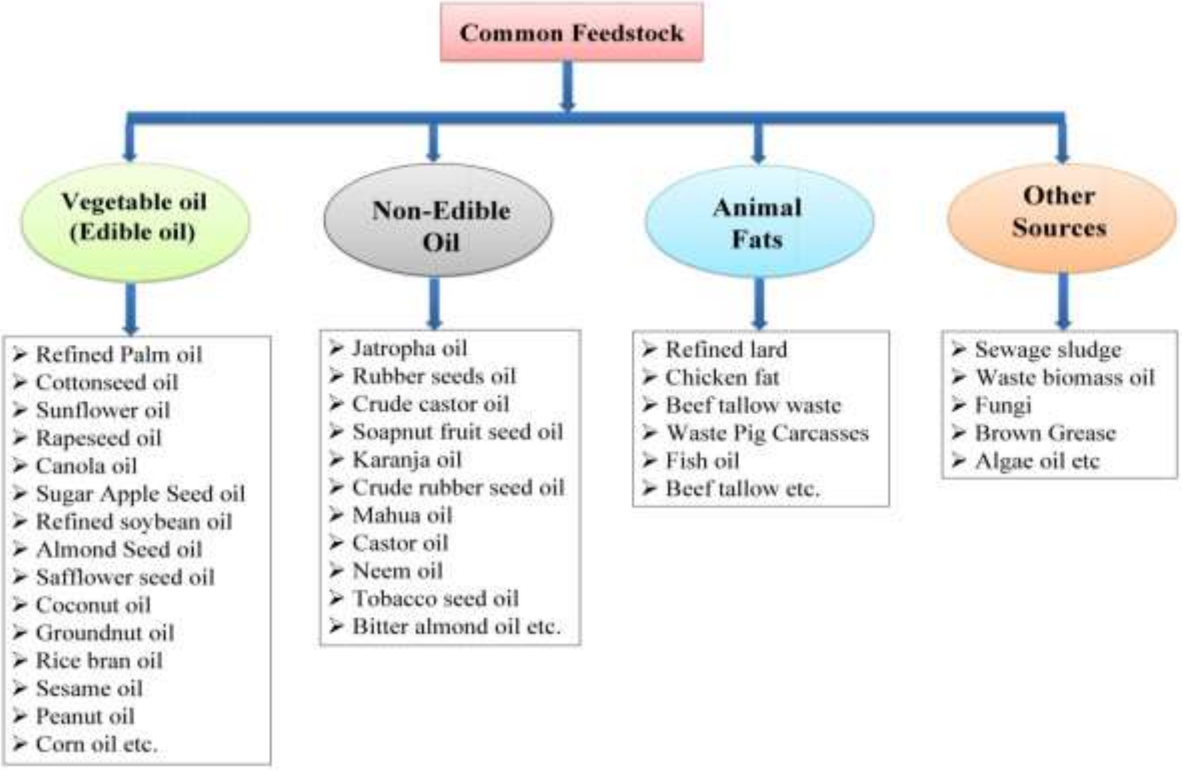

Figure 2. Common feedstocks for biodiesel production [76]

2.2. Biodiesel production

Direct usage and raw oil mixing, micro-emulsions, transesterification, and pyrolysis are the four main methods for making biodiesel [23]. Nevertheless, transesterification is presently the most often employed reaction [11,13,24]. The direct usage approach involves blending or adding diesel fuel to unrefined vegetable oil to make it viscous. The usage of diesel was found to be successful at ratios between 1:10 and 2:10. The literature states that mixtures of oil are not advised for both direct as well as indirect engines. The high viscosity, acid content, FFA content, and gum formation in this condition are all causing problems. According to what was said, the micro-emulsion method was created and is employed to address the issue of the highly viscid vegetable oil. By combining vegetable oil with the appropriate solvents, a micro-emulsion is created. Methanol, ethanol, and 1-butanol are examples of solvents that have been applied and investigated in the past. This method's drawbacks include the potential for significant carbon build-up and insufficient combustion [24]. Biodiesel has been made by pyrolysis, micro-emulsification, and transesterification. All of the solutions have benefits and cons, but they can be done. For large-scale biodiesel production, direct blending, pyrolysis, and micro-emulsification aren't enough. Trans-esterification increases production, simplifies separation, and lowers mass transfer resistance. Trans-esterification is excellent since it uses fatty acid and methanol catalysis. Reversible processes make up transesterification. Triglycerides are converted to diglycerides, diglycerides to monoglycerides, and glycerides to glycerol to make ester molecules. Figure 3 shows the transesterification reaction.

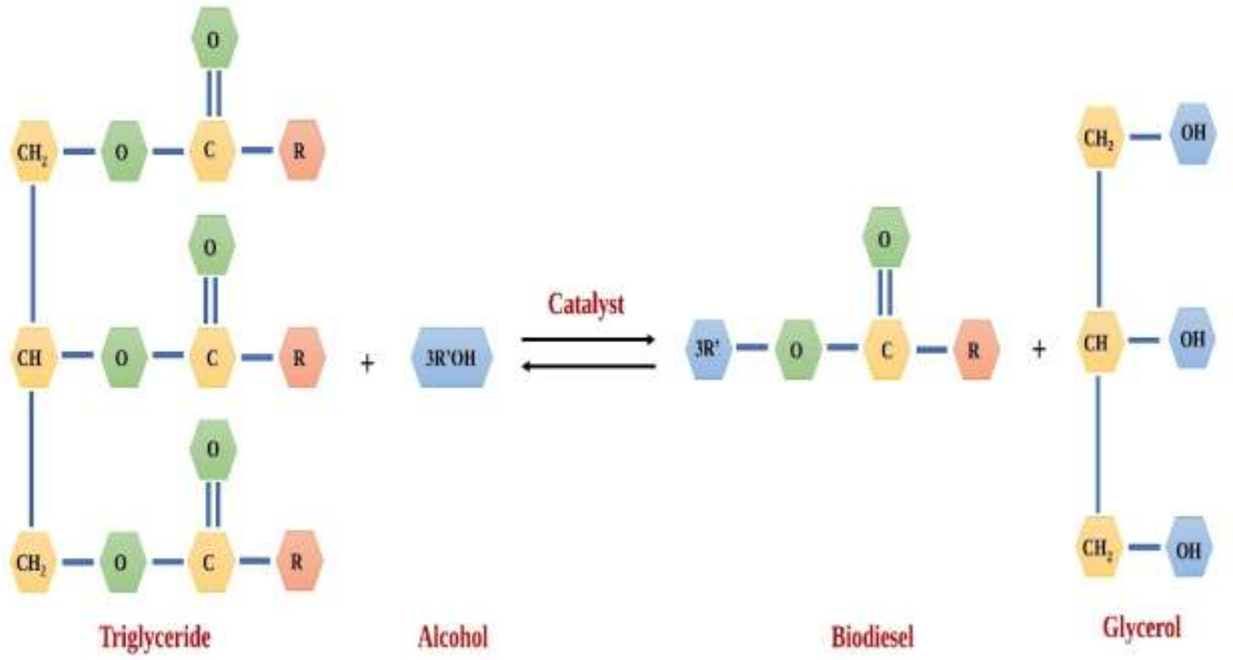

Figure 3. Transesterification reaction [77]

Oils are heated with or without a catalyst during pyrolysis, which changes one organic material into another [25]. As previously noted, biodiesel fuel made by the pyrolysis process, sometimes referred to as bio-oil, is suited for diesel engines, but, because oxygen is removed throughout the process, low-value components are created. Oxygen removal is done to improve the fuel generated so that it will be desirable and acceptable from an economic standpoint. The uses of biodiesel produced by this method are periodically constrained by undesired features such as low calorific value, partial volatility, and instability [26]. This approach has a number of advantages, such as lower processing costs, simplicity, less waste, and less pollution, although it does need expensive equipment [27]. The pyrolysis technique was recommended as a viable option for WCO processing [28]. Since it may lower the viscosity of the oil, transesterification is thought to be the process that produces biodiesel most favorably [24]. Glycerol and methyl esters are produced during the transesterification process when oil feedstock reacts with alcohol while coming in contact with a catalyst. The extraction of biodiesel and glycerin is followed by the recovery of the alcohol in the process. Although the created methyl ester is transported for purification, sometimes referred to as the washing stage, recycled alcohol returns to the beginning procedure. It will next undergo drying in order to generate perfected biodiesel.

Biodiesel production uses alkali catalysts for a single-step transesterification reaction. The FFA and water level may require an acid-catalyzed alcoholysis, or esterification, preceding transesterification. Researchers employ numerous feedstocks to make biodiesel, each with its own manufacturing method [41, 42]. Due to its inexpensive cost, methanol is most often utilized as fuel. Ethanol, propanol, butanol, and amyl-alcohol are among the many alcohols appropriate for transesterification [43]. Transesterification can be done at 60-70℃ atmospheric pressure and higher temperatures [44]. Transesterification yields glycerol. By equilibrium, most transesterification will have occurred, according to research. Settling the mixture completes the reaction. The upper methyl ester layer continues processing while one layer is cleansed. A rectifying column purifies and regenerates the excess methanol after distillation. Figure 4, shows single and double-step biodiesel production schematics.

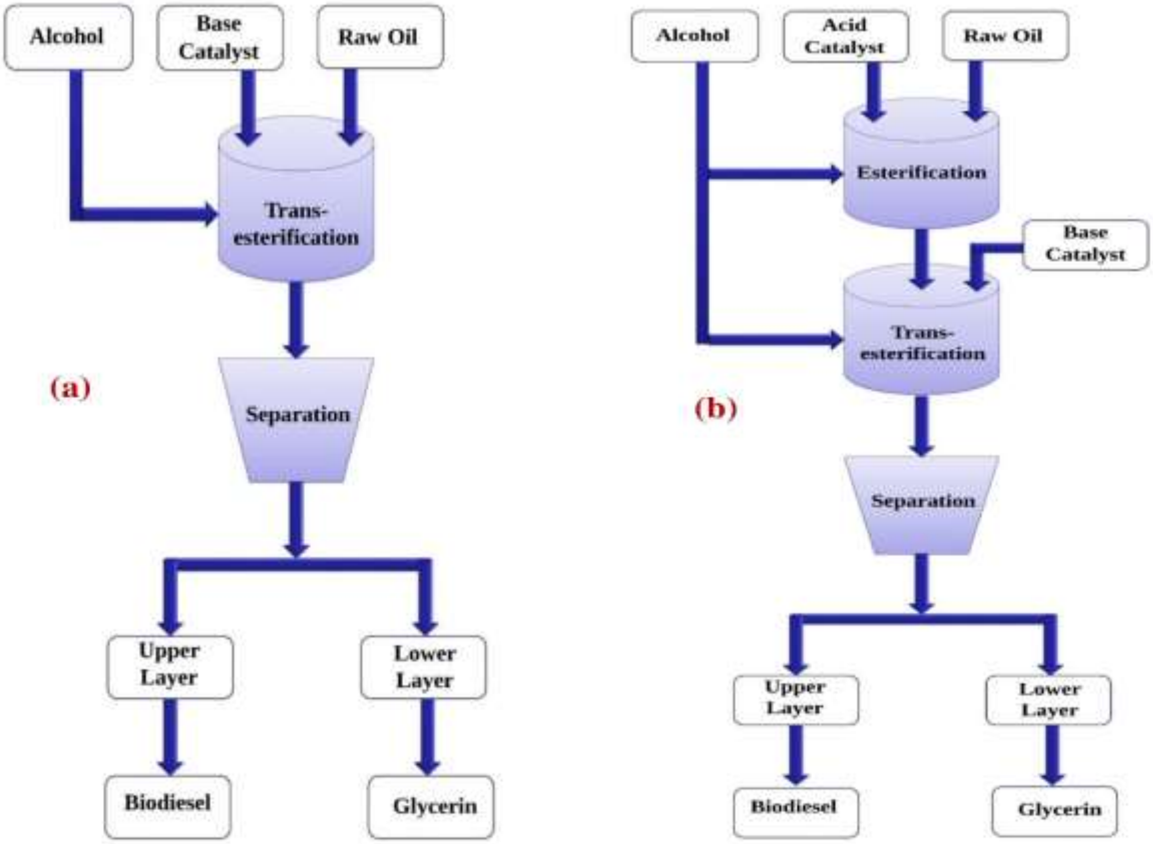

Figure 4. Single (a) and double (b) step biodiesel production [77]

The nature of the catalyst, the mole ratio of alcohol to vegetable oil, the composition of water and FFAs, temperature, and reaction time are all variables that might impact the transesterification yield [11]. Catalysts come in three different varieties: alkalis, acids, and enzymes. Given that it produces a large amount of fuel in a short period of time and doesn't harm industrial machinery, it is frequently employed in commercial production [29, 30]. When compared to other catalysts, it is claimed to have a very quick response. [11, 31] The creation of soap can reduce the output of biodiesel and interfere with the interaction of FFA with an alkali catalyst is undesired because it results in the dissociation of glycerin from methyl ester [32]. Moreover, it results in the use of catalysts. Soap generation can be prevented with the use of an enzyme catalyst. This catalyst is pricey and has a prolonged reaction period than acid catalysts. The beginning material and the circumstances of the reaction typically determine the catalyst that is used. Methanol, ethanol, propanol, butanol, and amyl alcohol are among the alcohols that are often utilized, according to textbooks. Methanol is more advantageous since it is less expensive, simpler to get, and capable of reacting swiftly with triglycerides and dissolving the alkali catalyst [31-33]. Biodiesel properties based on the variety of feedstock used are summarized in Table 1. Important characteristics of any fuel are its viscosity and flash point. The change in the biodiesel properties according to the use of raw feed is demonstrated in Figure 5.

Table 1. Biodiesel properties

Feedstock	Yield (%)	Viscosity (mm^2/sec)	Flashpoint (℃)	Cloud point (℃)	References
Palm oil	-	4.91	179	14	[32]
Castor oil	61	10.75	160	-13	[94]
Jatropha oil	76	5.25	166	-6	[94]
Jatropha oil	-	4.82	128	8	[16]
Jatropha oil	98	5.2	162	0	[95]
Sunflower oil	-	4.72	183	4	[95]
Waste cooking oil	87	5.3	196	-	[18]

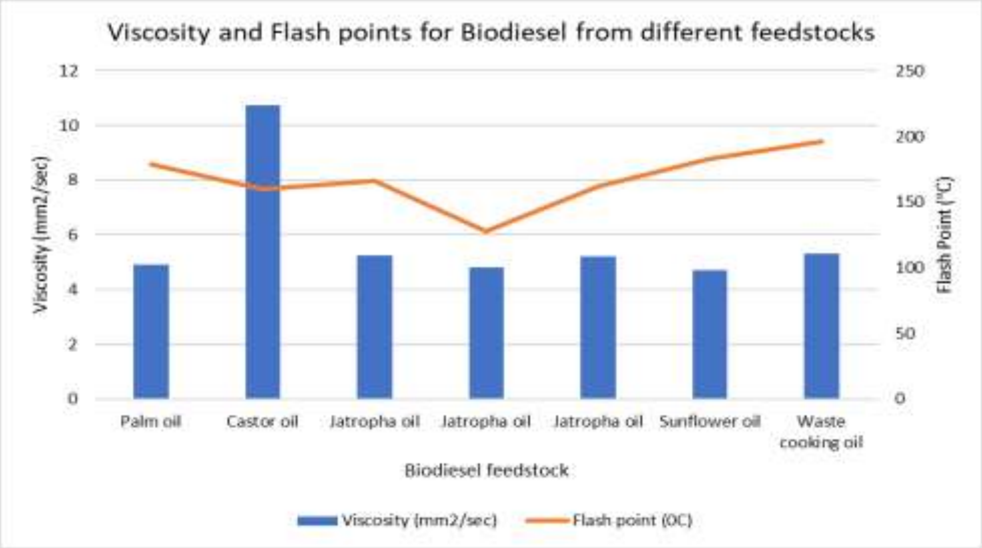

Figure 5. Biodiesel properties according to the use of feedstock

2.3. Wastewater generation

The washing process is primarily responsible for producing biodiesel effluent. It is essential to remove additional pollutants and contaminants during the wash process in order to ensure that only premium biodiesel that satisfies stringent specifications is created [32, 34]. In the washing process, water, soap, catalysts, free glycerin, leftover alcohols, and FFAs are among the unwanted materials eliminated. Non-removed impurities will lower biodiesel quality and have an impact on engine performance [6, 31, 32]. Wet and dry washing are the two methods most frequently used in the washing process. Membrane extraction has recently been studied as an alternate cleaning technique [31].

Glycerin, liquor, salts, and soap are eliminated during the wet washing process using purified hot water or softened solutions. Spraying water mist over the impure product causes the water and impurities to settle and drain away as effluent. Repeating this procedure and getting colorless water shows that all contaminants have been completely removed. The removal of both pollutants with this procedure is advantageous and successful due to the water's potential to dissolve glycerin and methanol. Long separation times and yield loss were listed as two drawbacks of this procedure in the literature [35]. A large portion of the highly contaminated liquid effluent is produced as a result of the decrease in methyl ester yields from fatty acids in rinse water [36]. A key issue for the business and environment is the quantity of biodiesel wastewater produced by the wash process.

Ion exchange resins also known as magnesium silicate powder are utilized for the dry-cleaning procedure [32, 35]. In order to eliminate contaminants, these compounds are utilized in place of water [31, 37]. To increase process efficiency, the filtration step is typically introduced towards the end. The benefits of this treatment are the absence of wastewater production and the reduction of the wash tank's overall surface area coverage [32]. The magnesium silicate used in this procedure may be recycled, but synthesized magnesium silicate has been further used as a composting component and an ingredient in animal feed. [38] Although this method has the benefit of not requiring any water, it is noted that the end products never match the requirements of the international biodiesel standard [37].

The membrane extraction technique was created with the intention of lowering the amount of water used for the washing process. Due to a decrease in the amount of oil in the water released, this approach might have a less detrimental effect on the environment. It is believed that the use of membrane extracting is favorable in that it uses less water, successfully prevents emulsifying agents during the cleaning stage, and reduces the loss of methyl ester throughout the refining process, which is a promising method of purifying biodiesel [37, 39]. Two different membrane types were used in the investigations on membranes: flat polymeric membranes for flat ultrafiltration of polytetrafluoroethylene (PTFE) and flat microfiltration of mixed cellulose acetate (MCA) [35]. The raw biodiesel was fed from the circulation tank to the membrane pores while the methyl ester permeates, which passes through the membrane, was gathered in glassware and the reject fluid was recirculated to the recirculation tank. According to their research, a larger amount of methyl ester was effectively filtered by the ultrafiltration PTFE polymeric membrane than by the MCA polymeric membrane. Tubular Al2O3/TiO2 membranes with typical sizes of 0.2, 0.1, and 0.05 m; 20 kDa were utilized. Membrane technology was also employed. Glycerol was successfully separated throughout the experiment utilizing 10% mass concentration oxidized water, resulting in a final free glycerol content that was less than 0.02% of the maximum value [39].

2.3. MEMBRANES

Water may be removed from wastewater using a variety of methods, including centrifugation, Gravity sedimentation, floatation, coagulation, and membrane filtration. Membrane filtration is extra favorable over other methods due to its low energy need, high recovery efficiency, and lack of toxicity. Traditional wastewater recovery techniques require the use of an energy-intensive evaporator, whereas membrane technology uses significantly less energy and has reduced operational costs.

3.1. Membrane-based separation

The technique of membrane separation may now be used to treat wastewater. Water with a high concentration of pollutants is created during the synthesis of biodiesel and was formerly released into the environment or into water sources, such as rivers, but this practice is now prohibited in many places. Also, it is urged to reuse and save water, especially in locations with a limited supply. The membranes can also be used for biological wastewater recovery. Biological treatment is frequently used to reduce chemical and biochemical oxygen demand in waste streams. First, bio sludge is recovered water for reuse in the utility, particularly in water-scarce locations, by passing it through a reverse osmosis membrane after being concentrated using an ultrafiltration membrane [40].

A high surface area to volume ratio, good control over constituent blending across two stages, and a high surface area to volume ratio are all desirable. The membrane is an engineered device that separates materials based on their size, shape, or characteristics by serving as a barrier to movement in liquid-liquid and liquid-solid processes [41]. The wastewater, dairy, carbohydrates, foodstuff, starches, liquor, enzyme, and sugar sectors all employ membrane filtering extensively [42, 43]. Pressure-driven operations are the most typical membrane treatment method [44, 45] The division of membrane processes is under pressure. The pore size and filtering method of polymeric membranes can be used to classify them. This categorizes membranes into reverse osmosis (RO), microfiltration (MF), ultrafiltration (UF), and nanofiltration (NF). The pore sizes for MF vary across 1 and 0.1 micrometers. It’s used to separate microorganisms, colloids, macromolecules, and other types of particles. UF membranes have pores with diameters that fall between 0.1-0.01 m to filter out viruses, nanoparticles, and solutes with high molecular weights as permeate side, enabling water or solutes with low molecular weights to flow through the pores.

The employment of ultrhaveltration separation technology in the food, medicinal, biotechnology, papermaking, and even dairy industries has gained more attention [44]. The most popular polymeric ultrafiltration membrane substances, such as polyvinylidene fluoride (PVDF), polyethylene (PE), polysulfone (PSF), polyether sulfone (PES), and polypropylene (PP), are frequently been used as the structural constituents of the membrane [44–47]. Fouling is one of the main issues with membranes, which decreases permeation flow, removal percentage, and membrane life when filters are operating. biological fouling, biofouling, colloid fouling, and chemical fouling can all be classified as fouling via the membrane processes [48, 49]. Because fouling leads to membrane pore obstruction, creating anti-fouling membranes are a popular solution to this issue. Antifouling membranes can therefore aid in lowering the need for energy. These membranes are distinguished by their hydrophilicity, smoothness of surface, and biocidal characteristics that prevent the build-up of foulants on the surface of the membrane [50–52]. AlFannakh et al. (2020) describe in detail pressure-driven membrane processes, summarized in Table 2.

Table 2. Pressure-driven membrane processes

Process	Pore size (μm)	Transmembrane Pressure (bar)
Micro Filtration (MF)	10^-1 - 10	Upto 1
Ultra Filtration (UF)	10^-2 - 10^-1	1 - 10
Nano Filtration (NF)	10^-3 - 10^-2	20 - 40
Reverse Osmosis (RO)	10^-4 - 10^-3	30 - 60

3.2 Characterization Of Membranes

3.2.1 Scanning Electron Microscopy

After covering the sample surface with gold, a scanning electron microscope (JEOL 5410) was utilized to examine this produced mix membrane's morphology. The material was examined using a 20 kV voltage and a 6000x magnification [53].

3.2.2 Mechanical Property Estimation

The tensile strength of the produced samples was investigated using the H5KS TINIUS OLSEN mechanical system. The materials were evaluated under uniaxial stress at a rate of 0.5 mm/min at ambient temperature (25℃) [53].

3.2.3 Porosity Estimations

One of the approved standard techniques for determining a membrane's porosity and air permeability is the use of densitometers. A sample area of roughly 25 cm^2 was examined using the Gurley Standard Densometer. By comparing the weights of the same sample when it was wet and when it had dried to a stable value, the porosity could be calculated. Porosity was calculated using the following equation [53]:

$$\varepsilon = Mw - Md/AL\rho \qquad (1)$$

where ρ is the density of pure water (g/cm^3), L is the thickness (cm), and A is the effective area (cm^2).

3.2.4 Pore Size Distribution Measurement

The model's Brunauer-Emmett-Teller (BET) apparatus was utilized to determine the created blend membranes' pore size distribution (ChemBET-3000, Quanta chrome). The BET equipment (glass column) was used to dry and degas a lengthy band of specimens with weights that are known for three hours at 80℃. A BET single point was used to calculate the average area [54, 55]. A detailed study of characterization done by various authors for membranes' use in biodiesel wastewater treatment is summarized in Table 3.

Table 3. Characterization of membranes' use in biodiesel wastewater treatment

Membrane Process	Geometry	Material type	Material	Selective layer	Pore diameter	MWCO	Area (m^2)	References
Nano Filtration (NF)	Flat sheet	Polymeric	Polyester Polysulfone	Polyamide thin film	0.9 nm	200 Da	0.00146	[56]
Nano Filtration (NF)	Tubular	Ceramic	Al_2O_3	TiO_2	1.8 nm	450 Da	0.00879	[56]
Ultra Filtration (UF)	Hollow fiber	Polymeric	Polyether sulfone	External	NA	50 kDa	0.027	[97]
Micro Filtration (MF)	Hollow fiber	Polymeric	Polyimide	External	0.4 μm	NA	0.027	[97]

3.2.5 Performance Of Membranes

For each membrane, the permeate flow and separation % were computed from equations (2) and (3):

$$J(W) = Q/\Delta t * A \qquad (2)$$

where A is the effective membrane surface (m^2), Q is the filtrate volume (l), and t is the permeation time (h).

$$S = (1 - Op/Of) \times 100 \qquad (3)$$

where Of and Op are the solute concentrations for feed and permeate, accordingly [55].

3.3 Membrane Test Unit

The literature-recommended dead-end filtration cell was used for the NF99HF plain membrane permeation experiments. The grade steel 316 L test cell (DEC-Sterlitech HP4750, USA) has an internal diameter of 5.1 cm and a height of 19.9 cm, and it can store a maximum initial charge of 250 mL. The membrane was supported on a compacted porous stainless-steel disc on top of Whatman filter paper, with a Viton O-ring acting as a sealer to prevent leaks. The effective area A of the membrane, which had a 4.9 cm diameter, was 1.03 m^2. The feed solution required to be agitated with a magnetic bar spinning at 500 rpm across the membrane surface to reduce the fouling phenomenon. A nitrogen tank (N) connected to the top of the cell and equipped with a multiple manifold (M) for pressure regulator and a pressure relief mechanism (R) supplied transmembrane pressure (delta p). The magnetic stirrer's temperature controller (1C) maintained a consistent temperature during the experiment (TC, PH) [56]. The permeation assembly for the tubular ceramic membrane comprises a multistage water pump and a single tubular cross-flow penetration module (CFC) (W) GRUNDFOS CRN2-150 APG-BUBE (1.5 Kw), a 15 L thermostat (CH), a stainless-steel wastewater drum with a 20 lit. capacity and an aluminum coil (0.16 m^2) attached to the chiller (CH). 50–500 mL graduated cylinder glasses were used to collect permeate, and a hand chronometer was used to record the collection's start and end times. Two 020 bar stainless steel pressure valves with glycerin baths (M) were mounted at the entry and exit of the module to measure the system pressure. The readings were assisted by a mercury column thermostat in the wastewater storage tank and a thermocouple in the module intake (T) was used to monitor the temperature (TK) [56].

4. BIODIESEL WASTEWATER CHARACTERIZATION

Researchers are interested in the significant volume of wastewater produced by the widely utilized wet wash procedure. According to prior research, the washing procedure is often depending mostly on methyl ester contamination level, two to five times, resulting in 20 to 120 liters of effluent per every 100 liters of biodiesel generated [29]. More than 20 L of wastewater was created per 100 liters of biodiesel production, according to other literature [57]. The discharge of wastewater from this rapidly expanding sector is said to raise environmental problems. It is frequently discovered to have large quantities of COD, SS, oil as well as grease (O&G), and a spectrum of pH values depending on the process being used. Wastewater due to the production of biodiesel is a dense, opaque white liquid [58]. This effluent is difficult to naturally decompose because of its higher pH values, substantial amounts of oil produced using hexane, and little nitrogen or phosphorus content, which are unfavorable for the development of microbes [29]. According to research by and other related studies, residual leftover oil is the predominant constituent of biodiesel effluent [6, 57]. As a result, dumping biodiesel wastewater into a public drainage system runs the risk of clogging the drainage because of the substantial oil content, interfering with biological activity during sewage treatment. The authors' investigations revealed the presence of water, glycerin, FFAs, accelerators, methanol, soaps, and fraction amounts of methyl ester in biodiesel effluent. The elevated concentrations of COD and O&G are a result of these pollutants [29, 34]

A small-scale, industrial biodiesel manufacturing facility that uses an alkali-catalyzed transesterification method provided the test effluent for the experiment. Crude palm oil and used frying oil are the feedstock used by this facility. The methyl ester washing steps were the primary source of the effluent. Due to contamination from oil feedstock, soap, methanol, and glycerol, the effluent from the biodiesel production process had significant concentrations of COD and O&G. The total residual O&G measured 6020 mg/L, whereas the quantity of methanol was determined to be 10667 mg/L. The wastewater's pH was higher than predicted at 8.9 because the transesterification process utilized alkali catalysts [59]. In a study, biodiesel made using soy oil using an alkaline transesterification process was rinsed three times with a water and phosphoric acid solution (3-liter wastewater to 1-liter biodiesel). The biodiesel wastewater had a substantial amount of organic matter and chemical oxygen demand, and it was a dense milky liquid (COD). Wastewater from biodiesel production is An translucent liquid with significant TOC, COD, O&G, conductivity, as well as oxygen concentration concentrations is reported [60-63]. The acidic wash employed in this work can be utilized to explain the disparity in Comparison of the study's pH but also electric conductivity with those found in the literature. Environmentalists are worried about biodiesel wastewater since it is also claimed to have a highly hazardous discharge [8].

Newer investigations use a range of produced organic and inorganic chemical concentrations to mimic actual wastewater. The effectiveness of treatment is determined by the fluctuation of COD over the course of 110 days for a big plant, according to a thorough and in-depth investigation of actual biodiesel wastewater. In order to prepare wastewater for this investigation, COD levels between 10,000 to 44,000 ppm were established based on wastewater COD values available in the literature [2, 64, 65]. Owing to the complex chemical balances involved, species solubility, the variety of phases (liquid and solid; emulsion and micro-emulsions), temperatures, ionic strength, and salt utilized, a shift in pH values is noticed, and this behavior is difficult to explain. By adjusting the related concentrations of Oleic acid, NaOH, HCl, plus NaCl, several writers have attempted to simulate the behavior of a system that is comparable to this one. For instance, it was discovered that lowering the pH from 6.5 to 5.0 causes a shift in an emulsion's typical droplet size of 102M oleic acid and 101M of NaCl, obtaining values from 0.45 to 0.40 m [66].

The factors were identified in the study's feed concentration of their sample, which was treated with NF membranes using comparable but actual wastewater: pH = 6.98, TS = 2608 ppm, EC = 1086, COD = 19,880 ppm [66]. In other studies, the COD was estimated to be between $19,000 and $37,000, while the pH ranged from 4.3 to 6.6. These studies employed coagulation, electrocoagulation, and biological treatments to dispose of wastewater [3, 4]. Based on the detailed evaluation of the important characteristics such as pH, COD, SS, and O&G of biodiesel wastewater is summarized in figures 4, 5, 6 and 7 respectively. Also, the BOD characteristics of biodiesel wastewater are summarized in Table 4.

Table 4. Characterization of membranes' use in biodiesel wastewater treatment

BOD (mg/lit.)	References
105000	[48]
168000	[58]
30000	[6]
168000	[34]
15260	[11]
1600	[2]

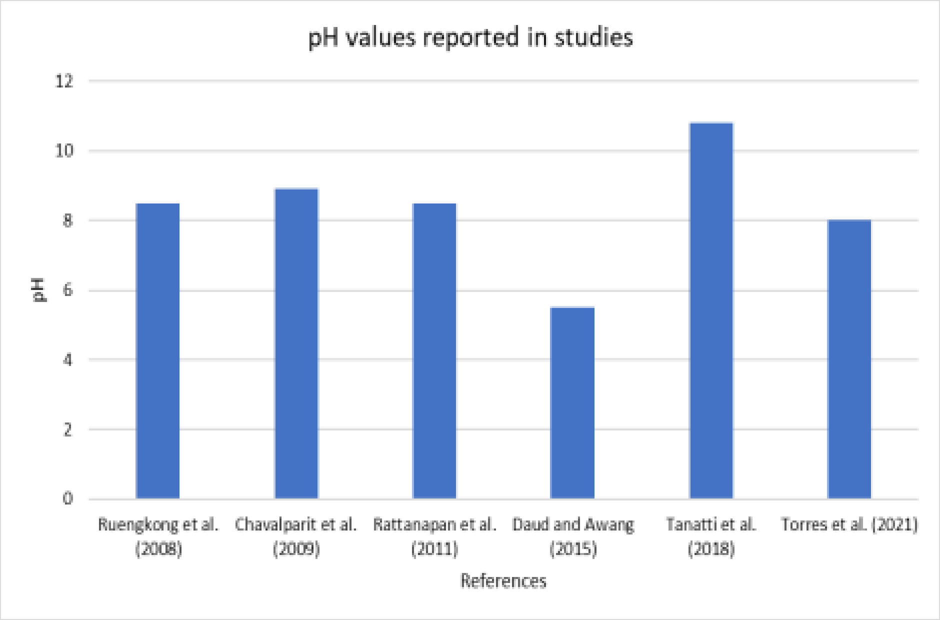

Figure 4. pH characteristics of biodiesel wastewater

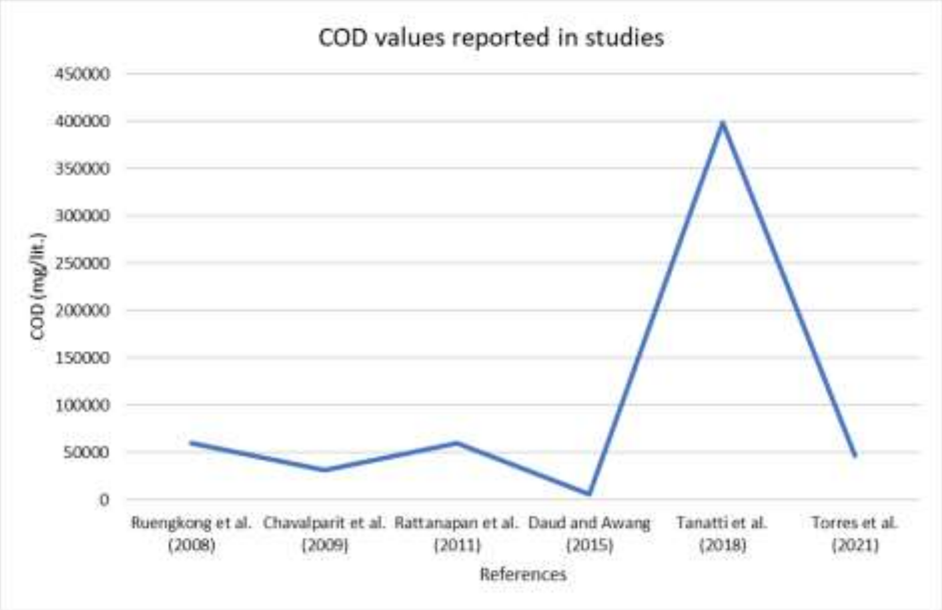

Figure 5. COD characteristics of biodiesel wastewater

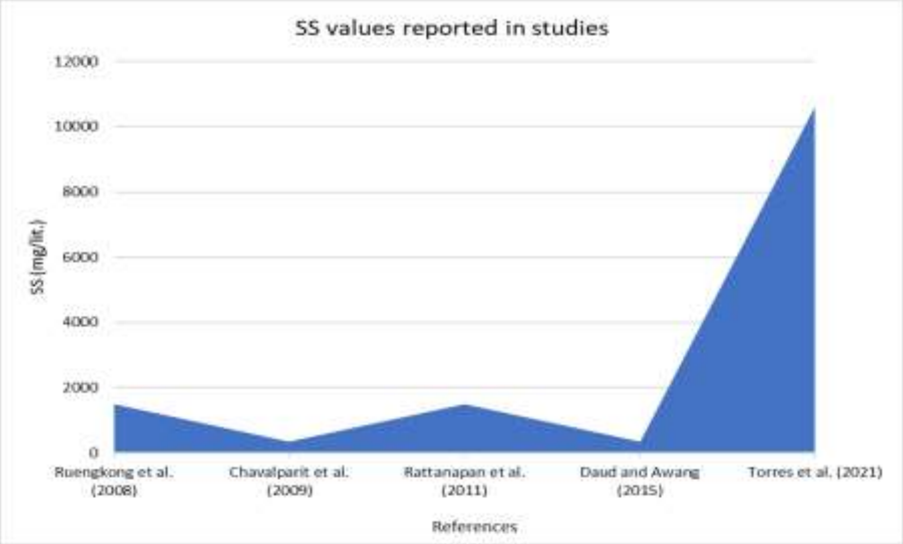

Figure 6. SS characteristics of biodiesel wastewater

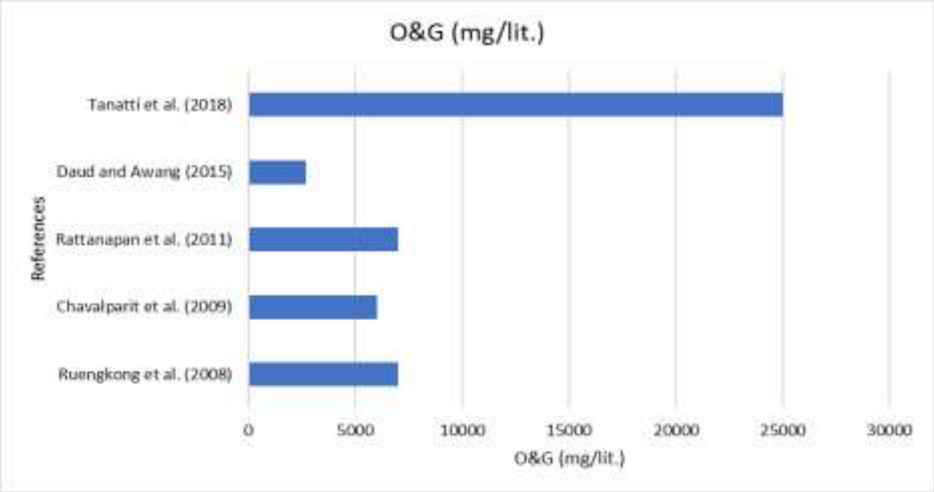

Figure 7. O&G characteristics of biodiesel wastewater

5. PERMEATION

In a detailed study conducted on NF membranes' ability to treat biodiesel wastewater, the following factors were compared with similar works [56].

5.1. Water permeability

There are two records for the NF99HF membrane's hydraulic permeability in the literature: a number that is close to 13.7 L*m^2*h/bar and a greater value that is recorded at 35℃ [67, 68]. A narrow seemingly counterintuitive behavior of permeation (or permeance) found is in line with the hypothesis that increasing pore width should result in a lower permeation flow and, consequently, a higher permeability, according to the Hagen-Poiseuille convective flow model. This suggests that the bigger mean pore width of the Inopor Nano membrane results in greater permeability (rp). Based on the Hagen-Poiseuille equation, the functional membrane thickness (m), the dynamic viscosity of water at 30℃ (Pa*s), and the membrane porosity Am may be used to represent hydraulic permeability. Nevertheless, there are additional characteristics like contact angle, surface quality, selected layer thickness, and membrane penetration to take into account for comparison. It is fascinating to read about the porosity and membrane thickness in relation to these characteristics [69].

6. LOAD EFFECT, SELECTIVITY OF COD

Another topic investigated was the influence of compound wastewater concentration on retention. The wastewater feed combinations were used as the experimental medium in the Inopor Nano membrane. Given the COD readings in permeate weren't lower than 1000 ppm, one unanswered question in this investigation is: What constituents are responsible for the ultimate COD? How does feed concentration affect permeate flow, COD, and EC retentions? Does selective retention exist? The organic load was significantly reduced with both filtering systems, but it was not enough to meet the legal requirements, thus a number of tests were conducted to investigate this problem. Two aqueous solutions were created for COD levels of 1900 ppm, salt (200 mu*S*cm), glycerol, and methanol mentioned in one, and methanol and salts alone in the other [56].

7. ANALYSIS OF SERIES RESISTANCE

The computation of resistances is a rapid technique to learn about fouling phenomena, but it has to be supplemented with experimental knowledge. For instance, recovering the NF99HF membrane required just a straightforward wash process with distilled water to remove both the reversible and irreversible resistance components, however recovering the irreversible resistance component from the ceramic membrane required an extreme cleaning cycle. The membranes examined in this article have been examined by a number of researchers. The research was published as a report that used a cross-flow filtration module to evaluate the NF99HF for NF of weak standard solutions of aniline (100–200 ppm) [73]. The authors created a novel experimental investigation using a 19-channel cylindrical membrane with qualities similar to the commercial ceramic membrane (0.9 nm pore diameter and 450 Da of MWCO). In a pilot facility with 5.1 m^2 available, these authors examined textile washing machine wastewater. Even if the COD load is not stated, the findings show a COD drop of 45–80% with 10-80% for salt content, as well as real fluxes of 100–200 $L*h/m^2$ at 15 bars with v = 2.5-4.5 m/s. [74, 75].

8. MBR CONFIGURATION

Microfiltration (MF) and ultrafiltration (UF) routinely work in membrane bioreactors (MBRs), as are reverse osmosis (RO) and nanofiltration (NF) for advanced cleaning [78]. MBR technology offers benefits such as high-quality effluent, shorter hydraulic retention time (HRT), longer solid retention time (SRT), less slurry formation, the ability to remove organic and inorganic impurities, and resistance to high organic loading [79]. However, it has some drawbacks, including decreased membrane penetrability and higher operating and repairing prices, requiring membrane substitution [80]. The two types of fouling that can occur are reversible and irreversible fouling. Fouling that can be eliminated by relaxation or backwashing is referred to as reversible fouling, whereas irreversible fouling is defined as fouling that is strongly adhered to and requires chemical treatment [5]. Solid retention time (SRT) is regarded as the important operational parameter that influences the properties of activated slurry in terms of microbial diversification, biodegradation kinetics, biopolymer concentration, floc generation, and size separation, which in turn influences effluent quality and membrane fouling propensity. MBR operation at short SRT (e.g., 10 days), for example, restricts the growth of useful microorganisms such as low-growing nitrifiers [81].

The bioreactor initiates biological mechanisms, and the membrane facilitates the removal of suspended particles and contaminants by filtration. Because MBR is one of the most trustworthy methods, the membrane's cost is fairly high. In addition to this, periodic maintenance of membranes and modes of operation is a predictive measure of sustainable membrane technology. Amongst the other types of MBR, submerged membrane bioreactor resulted in a low cost of operation. Membrane act as a barrier between solids and fluid whereas effluent particles are converted into microbial aggregates in an activated sludge process [82].

A side-stream or external membrane bioreactor and an immersed membrane bioreactor (iMBR) or submerged membrane bioreactor (sMBR) are type of MBR. In a side stream MBR system, the feed wastewater comes into direct touch with the biomass. A membrane-based recirculation loop is used to pump both wastewater and biomass. While the water outflow is discharged, the concentrated slurry is reprocessed back to the reactor. A separation of the membrane and the bioreactor is intended to make membrane improvement easier, but it will increase operational costs due to the construction of a recirculation loop [79]. The immersed MBR system has lower operational costs than the side-stream MBR system since there is no recirculation loop and a biological treatment takes place around the membrane in immersed MBR. To maintain sludge age, both MBRs must pump back surplus sludge. Membrane transportation can take effect under pressure or under a vacuum. Pressure-driven filtration is utilized in side-stream MBR while vacuum-driven filtration is used in immersed MBR, which functions in dead-end mode, according to Radenkovic et al. Both systems get air bubbles for aeration and scouring, with the submerged system receiving more air bubbles to decrease membrane fouling in the cross-flow action across the membrane surface [80,82].

MBRs divided into two categories: aerobic and anaerobic. Aerobic MBRs use oxygen as a growth medium for the bacteria, whereas anaerobic MBRs do not. Anaerobic MBRs are less effective at eliminating COD and require a considerable start-up period. Anaerobic treatment is typically used to treat high-strength wastewater at a less temperature that is conducive to microbial development. When compared to aerobic at low flux, it's difficult to maintain a low temperature for the waste feed and it creates a lot of fouling. Organic matter was destroyed within the biofilm under aerobic conditions, with nearly all of the oxygen provided being utilized for biodegradation. It's challenging to maintain the ideal biofilm thickness for adequate oxidation; alternatively, too much biofilm can obstruct liquid flow [79]. EMBR is commonly utilized in conditions where significant quantities of inorganic compounds, such as excessive salt concentration or a high pH, could hinder biodegradation treatment. Only specific organic pollutants (phenol, hydrogen sulphone, and a few inorganic) that can be decomposed in a separate bioreactor are removed by EMBR [83].

9. IMPORTANT CHALLENGES

9.1 Water Scarcity

Water scarcity poses a complex barrier to our community's long-term growth. Climate change, rising population, industrialization, urbanization, and pollution are all contributing to this problem.

9.2. Membrane Fouling

Membrane fouling by reason of the deposition of slurry flocs (dirt) on the membrane surface, which decreases membrane penetrability, maximizes purification frequency and reduction in membrane life. Up until now, there has been a lot of exploration on membrane fouling, factors that influence membrane fouling, and mechanisms, as well as a lot of mitigation approaches [84, 85].

9.3. Membrane Cost

Another commercialization barrier is the cost of membrane bioreactor operations, which varies depending on the variety of membranes and applications. Membrane bioreactors have the capability to lower production expenditure by reducing the number of unit performances for products that demand higher purity, such as lactic acid. Membrane module costs, energy requirements, aeration schemes, and other equipment should all be taken into account. Cost expenditure (CAPEX), operating expenditure (OPEX), substances, and serviceability are the three primary aspects of the cost. Costs for the fermenter, membrane modules, and other bioprocess apparatus are included in capital expenditures. The total electrical and thermal power used is included in the operating costs. To fully commercialize, all cost components should be kept to a minimum [86].

9.4. Salinity Build-Up

Wastewaters with high salt concentrations pose a barrier to biological treatment, increasing the physicochemical and microbiological interaction dynamics. Increased salt concentrations have also been discovered to be major performance-restricting variables in anaerobic systems due to their adverse effects on biomass. When fed with slowly biodegradable wastewaters consisting of proteins or lipids, anaerobic MBRs can achieve better performance efficiencies at maximal SRTs and balanced surrounding conditions. Raised salt concentrations in aerobic MBRs can have a detrimental influence on system performance by inhibiting microbial activity and growth. In both anaerobic and aerobic settings, increased salt causes cell plasmolysis, which reduces metabolic activity [87]. Controlling process limitations and managing biohydrogen logistics, recovery of dissolved methane, extraction and purification of volatile fatty acid (VFA) and the need to reduce costs while using the Anaerobic Membrane Bioreactor are all challenges that must be overcome to improve energy recovery from the Anaerobic Membrane Bioreactor [88].

9.5. Temperature

The essential heat is provided by heating the reactors in the anaerobic biological process of sewerage, which operates at mesophyll or thermophilic temperatures. The operational viability of anaerobic reactors, however, remains a difficulty due to the lower energy content of treated wastewater needed to maintain the necessary temperature, posing a barrier to long-term operation. As a result, operating anaerobic systems at room temperature could be one solution to this problem in cold climates [8]. investigated an MBR for household wastewater management at psychrophilic temperatures and found that COD removal was excellent at temperatures ranging from 15℃ to 6℃, although replacement was hampered at 3℃ [89]. discovered that at less temperatures, organics convert poorly to biogas. Low temperatures hinder anaerobic microbes' activities, including hydrolysis and methanogenesis, to some extent, according to an earlier study [90,91].

10. ENVIRONMENTAL SUSTAINABILITY OF MEMBRANE BIOREACTOR SYSTEM

Membrane bioreactors are an effective method for eliminating organic debris, Nitrogen compounds, and fouling from household and hospital wastewater. Membrane bioreactors may be the right answer for avoiding environmental infractions and minimizing our carbon footprint [80]. Sludge formation that is excessive is considered a polluted source, and additional expenditures are included [86]. The use of a coagulant can assist in reducing the cake layer and the degree of fouling. The residual organic load and salt concentration are key issues for the activated sludge process because they raise capital investment. Additional costs are incurred when Fento's procedure and the aerobic process are combined. A 0.9 Euro per cubic meter ozone-enhanced biofilter granular reactor costs. The cost of bio-filter granular reactors is approximately 0.9 Euro per meter cube of tannery sewerage. The price was lowered to 0.36 Euro per cubic meter because of the many processes of anoxic MBR. Membrane bioreactors were identified as a viable technique for tannery wastewater treatment due to the reduction in treatment costs. This technique ensures a consistent treatment while minimizing costs [92,93].

CONCLUSIONS

The increasing demand for biodiesel has brought to light a new challenge brought about by the scarcity of fossil fuels. The operation generates a sizable amount of effluent. Effluents that contain soap, glycol, methanol, oil, and grease cannot be treated in a single step. The creamy appearance and pungent smell of this wastewater indicate a pressing need for treatment. Several methods, each with its own set of pros and cons, are being explored, evaluated, and implemented for treating and pre-processing aqueous waste from biodiesel production. Several studies show that nanofiltration is a sufficient but imperfect approach for eliminating COD from biodiesel effluent. The final content of the pollutants shows a considerable drop after effluents have been treated. Recent papers in the relevant literature, as well as the calculated findings for COD and EC stability, were found to be equivalent. Although the conductivity was lowered by 87% and 89%, respectively, the highest COD holding with the polymer membranes was 80% and the maximum retention with the ceramic membrane was 85%. Minimal fouling processes are responsible for the ceramic membrane's modest permeate flow reduction. Acceptable discharge waste values can be attained by combining membrane NF-treated wastewater with other technologies. Impurities in biofuels can be removed using ultrafiltration and microfiltration, yeast and fermentation inhibitors can be removed using pervaporation, and nanofiltration can desalt and purify the fuel. Low molecular weight components and recycled water are separated via reverse/forward filtering. Industrializing membrane-based renewables will result in larger economic and energy benefits, but their development is contingent on the efficiency of the membrane process and the quality of the membranes. Improving the present membranes' flow and selectivity is essential for getting the price and energy down. The longevity, mechanical strength, thermal and chemical stability, and fouling impacts of the membrane systems need more study.

ABBREVIATIONS

FAME = Fatty Acid Methyl Esters; COD = Chemical Oxygen Demand; BOD = Biological Oxygen Demand; BOD5 = Biological Oxygen Demand test over 5 days; SS = Suspended Solids; AOP = Advanced Oxidation Process; MBR = Membrane Biological Reactor; FFA = Free Fatty Acid; WCO = Waste Cooking Oil; PTFE = Polytetrafluoroethylene; MCA = Mixed Cellulose Acetate; RO = Reverse Osmosis; MF = Microfiltration; UF = Ultrafiltration; NF = Nanofiltration; PVDF = Polyvinylidene Fluoride ; PE = Polyethylene; PSF = Polysulfone ; PES = Polyether Sulfone ; PP = Polypropylene; BET = Brunauer-Emmett-Teller; O&G = Oil and Grease; MWCO = Molecular Weight Cut Off; TOC = Total Organic Carbon; EC = Electric Conductivity

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