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Membrane techniques for removal of detergents and petroleum products from carwash effluents: a review

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Abstract

One of the most significant urban services is the carwash, which generates large amounts of wastewater containing a variety of pollutants, including sand, gravel, suspended solids, surfactants, oil products, diesel cleaners, etc., that may cause environmental pollution when transferred to the sewage system without any treatment. The effective treatment is crucial to prevent environmental pollution as well as to recycle the water source. Contaminants are removed from carwash effluent using a variety of treatment technologies. This review focuses on identifying and comparing efficiency of using advanced commercial and modified membrane filtration techniques, meeting discharge standard regulations, to treat carwash impurities, especially detergents/surfactants (anionic surfactant) and petroleum products (oil/grease). The results of this review indicate that ultrafiltration membrane (UF) is the most common membrane filtration technology for carwash wastewater treatment. Additionally, the adoption of traditional pre-treatment processes may be advantageous before utilization of membrane process for treating carwash wastewater; although conventional treatment processes can produce a high quality of effluent, they are less effective than membrane systems.

Keywords:

anionic surfactant
carwash
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Key findings

- Pollutant removal efficiency of surfactants, oil products and TSS from carwash wastewater depends on the pre-treatment method and the type of membranes used.
- The main method for obtaining new modified membrane structures is the combination of various polymers with other materials to improve the membrane performance.

1. Introduction

One of the activities that consumes huge amount of water is washing cars. The amount of water consumed depends on both the geographical area and the carwash type, and 150–600 L of water are often used to wash a car [1]. The discharge of the car wash station without treatment resulted in different harmful impacts on the environment because it includes various types of impurities like oils, hydrocarbons, fats, petroleum fractions, road surface pollutants, small exhaust particles and water-soluble cleansing agent [2]. The composition of carwash wastewater varies greatly depending on the washing technique, type and size of the car that is being washed [3, 4]. The proper treatment and purification of carwash wastewater is considered as a new source of water. In international practices, great attention is paid to the quality of carwash wastewater and the selection of the optimal methods for cleaning it. Recently, membrane separation processes have been effectively used to remove carwash contaminants. The manufacturing materials for the membranes are either organic (polymers) or inorganic (ceramic, steel, aluminum, and glass). The membrane processes are able to carry out an effective treatment to decrease the oil content, detergent and other pollutants in carwash wastewater to an acceptable level [5–10]. By conducting a literature survey, it no information was found about the use of modified membranes technologies for treating the contaminants present in carwash wastewater, which is one of the relevant areas of research. In this regard, the purpose of this literature review was to evaluate the efficiency of commercial and modified membrane filtration techniques and hybrid membrane methods for removing surfactants, oily products and other contaminants of carwash units with the possibility of ensuring their compliance with the regulations for reuse or discharge into the centralized sewer network.

2. Carwash units' specifications

The carwash wastewater (CWW) composition depends on the number of washed cars, the season, the type of carwash, and the parameters like detergent types and concentrations, and the amount of water used. The contaminants of carwash wastewater originate from detergents used in the washing process, oils and greases of the engine, organic materials, metals, residual petroleum, and particles, like carbon, dust, and salt [11, 12]. The International Car Wash Association listed the main parameters of interest to carwash operators, which include: total dissolved solids (TDS), total suspended solids (TSS), biochemical oxygen demand (BOD), chemical oxygen demand (COD), oil and grease, detergent, zinc, lead, and trace quantities of other metals [13]. Of these constituents, the carwash operator uses the detergent only to wash off the other parameters from the car surfaces. The parameters characteristic of carwash wastewater mentioned in the literature are listed in Table 1.

The concentrations of COD, oil and grease, and turbidity have a large range, as indicated in Table 1, although some parameters, such as pH, have fewer variation in the studies. According to Table 1, several studies revealed the pH of CWW were between 6.3–8.7, with the higher value recorded by [1]. Also, the electrical conductivity (EC) values were within 138.8–1570 μS/cm. Other CWW Constituents are present in solids that enter the vehicle from dust, mud, and silts, which also increase the turbidity of the wastewater. Van Bruggen et al. recorded the maximum value of total suspended solids (TSS) within 2458 mg/L [14], while Shete and N.P. Shinkar, recorded the high value of total dissolved solids (TDS) within 7920 mg/L [2], and the maximum value of turbidity recorded by Mirshahghassemi et al. was within 1400 NTU [15].

The chemical oxidation demand (COD) in CWW was ranged between 59–1810 mg/L. The countries such as Brazil recorded 59–85 mg/L of COD [16]; Indonesia 700 mg/L [17]; Malaysia 75.0–738 mg/L [1]; Turkey recorded 314 mg/L [18]; and India 460 mg/L [19]. The higher COD observed in Belgium was 1810 mg/L [14]. In carwash stations, emulsified oil obtained from washing engine and detergents utilized for vehicles washes contribute to increased BOD levels. Based on the literature review, the biochemical oxygen demand (BOD5) values were 70–320 mg/L in Belgium [14] and 27–650 mg/L in South Africa [20]. Lau et al. recorded the BOD5 values in Malaysia (10.5–11.9 mg/L that are lower than the others [1]. The oil and grease which are among the CWW's most significant pollutants were stated from less than 0.1 mg/L [16] up to more than 1750 mg/L [21]. Also, Van Bruggen et al. found high level of anionic surfactant in CWW, about 15.5 mg/L [14]. Many studies in the field of treatment carwash wastewater determined, total dissolved solids, total suspended solids, turbidity, and chemical oxygen demand (COD), but they did not measure detergents and oil products despite their great impact on the environment [22].

The two important parameters in carwash wastewater (detergents, and oil products) are described in the following paragraphs.

2.1. Structure characteristics and properties of the detergents in carwash technologies

In car wash stations, detergents used as cleaning agents contain different types of surfactants. Surfactants are surface-active chemicals that reduced the surface tension and are concentrated at the interfaces between bodies or droplets of water or oil to act as foaming or emulsifying agents. Their chemical structure has a direct bearing on this mode of action [28]. They are long-chain molecules containing a head group that is hydrophilic (soluble in water) and a tail group that is hydrophobic (oil soluble).

Table 1 The characteristics of carwash wastewater.

Parameter	Units	Range	References
pH	–	6.3–8.7	[1, 2, 16, 18, 19, 23, 24]
Conductivity (EC)	μS/cm	138.8–1570	[1, 16, 18, 19, 23]
Turbidity	NTU	7.7–1400	[1, 2, 15–17, 23–25]
Total Suspended solids (TSS)	mg/L	60–2458	[2, 14, 16, 18, 19, 26]
Total Dissolved solids (TDS)	mg/L	120–7920	[1, 2, 16, 23, 25]
Chemical Oxygen Demand (COD)	mg/L	59–1810	[1, 2, 14, 16–19, 23–27]
Biochemical oxygen demand	mg/L	10.5–650	[1, 14, 20]
Oil and grease (O&G)	mg/L	0.1–1750	[16, 17, 21, 24]
Anionic surfactant	mg/L	0.7–51	[24, 26, 27]

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The major classification of surfactants into classes is based on the characteristics of the hydrophilic head group: anionic with a negative charge, cationic with a positive charge, non-ionic without charge, and zwitterionic with negative and positive charges. The hydrophobic tail group may be long chain hydrocarbons, fluorocarbons, siloxane chains, and short polymer chains [29, 30].

Among the numerous surfactant types, anionic surfactants are particularly important because they account for 60% of all soap production worldwide [31]. Anionic surfactants are most commonly used in car washing technologies. Among the most widespread are those which have a lengthy hydrophobic alkyl chain and a hydrophilic end group that is charged with sulfonate (e.g., Linear alkylbenzene sulfonate (LAS), Secondary alkane sulfonate (SAS), a-Olefin sulfonate (AOS), and Methylester sulfonate (MES)) or a sulfate charged hydrophilic end group (e.g., Alkyl sulfate (FAS or AS) and Alkylether sulfate (AES)) (Figure 1), as well as sodium, potassium, or ammonium counterions. Sodium dialkylsulfosuccinate, sodium dodecylsulfonate, and sodium odecylbenzenesulfonate are common surfactants.

Anionic surfactants play a significant part in technical applications owing to their excellent properties like superior water solubility, great cleaning efficiency, and low cost [32]. The methylene blue active substances method (MBAS) is used to determine the anionic surfactant content for water and wastewater. Anionic surfactants are the most notable of the materials exhibiting methylene blue activity, which is usually utilized in formulations of detergent which are strongly responsive to this approach [33]. MBAS with a high concentration of linear alkylbenzene sulfonates (LAS) may coat the oil and grease with MBAS molecules to make these droplets soluble in water. Thus, the effluent did not have lower values [34]. The anionic surfactants contained in carwash wastewater and its treatment methods are listed in Table 2.

2.2. Structure characteristics of the petroleum products in carwash wastewater

Carwash wastewater can carry petroleum hydrocarbon residues, hydraulic fluids, and lubricating oils. Lubricating oils are formed from a basic stock of heavier petroleum hydrocarbons, which are generated from crude oil, together with a variety of additives to enhance their specific qualities. They are mixed using base oils made of petroleum hydrocarbon, polyalphaolefins (PAO) or their mixtures in varying ratios [37]. Oil and grease that originate from different types of petroleum products in car washes may be released from the vehicle surface, tires, or may be leaked out from the braking system, engine parts, and connections [38]. Oil composition is complex, typically containing aliphatic hydrocarbons (73–80%), monoaromatic hydrocarbons (11–15%), diaromatic hydrocarbons (2–5%) and polyaromatic hydrocarbons (4–8%). Also, it consists approximately of 20% of the lubrication additives, which include zinc diaryl, zinc dithiophosphate, molybdenum disulfide, metal soaps and other organometallic composites.

Oil products in carwash wastewater are considered a serious environmental problem because they contain toxic materials (e.g. polyaromatic hydrocarbon, phenol, etc.) affecting the environment [39]. Table 3 shows oil and grease content in carwash wastewater and its treatment methods.

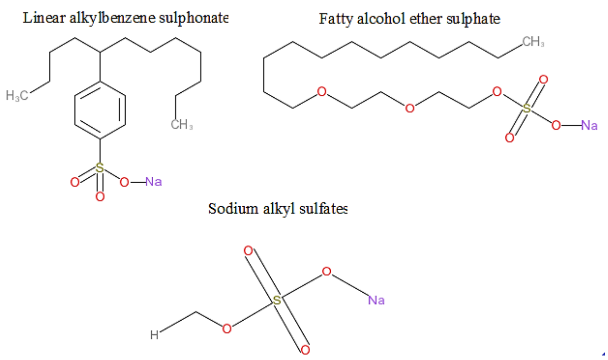

Figure 1 Structural formula of common anionic surfactants.

Table 2 Anionic surfactants in various carwash wastewater and the treatment methods.

Anionic surfactants	Initial concentration (mg/L)	Final concentration (mg/L)	Treatment method	References
Sodium dodecyl sulfate (SDS)	100	9.9–51.7	Membrane separation	[14]
Sodium dodecylbenzene sulfonate (SDBS)	0.7–2.5	Not available	Membrane separation	[26]
Sodium dodecyl benzene sulfonate (SDBS)	0.8	0.1–0.4	Membrane separation	[27]
Linear alkylbenzene sulphonates (LAS)	Not declared	<0.06	Coagulation; membrane separation; granular activated carbon	[35]
Linear alkylbenzene sulphonates (LAS)	2–5	Not available	Coagulation; membrane separation	[24]
Alkylbenzene surfactant (ABS)	3–20	<0.05	Sedimentation; surface degreasing; sand filtration; ozonation; UV irradiation, membrane separation	[21]
Dodecyl benzene sulfonate (DBS)	21	12	Flocculation column; flotation; sand filtration; chlorination	[36]

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Table 3 Oil and grease in various carwash wastewater and treatment techniques.

Oil and grease	Initial concentration (mg/L)	Final concentration (mg/L)	Treatment method	References
Oil	Not declared	0.95	Coagulation; membrane separation; granular activated carbon	[35]
Oil	5–25	Not available	Coagulation; membrane separation	[24]
Oil	36	2–8	Membrane separation	[17]
Fossil oil and grease	500–3000	4–20	Sedimentation; surface degreasing; sand filtration; ozonation; UV irradiation; membrane separation	[21]

3. Legislation standards for carwash wastewater

To conserve natural resources and give a high-quality wash, a new carwash technique must be used, involving water reusing. Some countries have achieved considerable advancements in the reuse of wastewater by establishing laws and regulations, whereas the other ones still lack sufficient planning and restrictions [40]. Environmental legislation and regulations have been published for wastewater disposal into the public sewage system. Though many countries have their own legislations on environmental quality, the carwash sector rarely enforces application of these laws [41]. Greywater standard on carwash wastewater was created in countries like United States, Australia, and Europe. Some countries in Europe restricted consumption of fresh water for carwash, or imposed a reclamation (recycling and reuse) percentage of approximately 70–80% to meet the water quality standard [36].

The standards for the carwash wastewater discharge were included in the Standard of Sewage and Effluent Discharge. Wastewater composition standards (the standards of permissible discharges of substances into water bodies) were established in city of Yekaterinburg, Sverdlovsk region, Russia by the Regulation No. 2329 of 2021. Furthermore, Iraq established Iraqi National Standards set by the Regulation 25 of 1967 for wastewater drained to the water source or into public sewers. Malaysia also uses Standard A and Standard B Regulations 2012 based on Malaysia Sewage and Industrial Effluent Discharge for the effluent discharges of carwash wastewater. These legislations are applied to the carwash wastewater and are focused on suspended solids (SS), oil and grease, and chemical oxygen demand (COD), yet less attention is paid to the surfactant concentration. The legislation standards for wastewater are summarized in Table 4.

4. Membrane techniques

4.1. Commercial membrane techniques

A summary of numerous studies on membrane techniques used for the treatment of wastewater from car washes is provided in Table 5.

The UF membranes were widely used in a variety of industrial applications because of the great efficacy in removing contaminants from wastewater with higher permeation flux than that of the other membrane filtration processes [42–44]. The utilization of UF membranes are the main focus of membrane research in the carwash sector [45, 46]. Different commercial and experimental membranes were applied to the carwash effluents that have a various contaminant like detergents/surfactants, oil/grease and others. Bruggen et al. investigated seven types of commercial UF membranes with carwash effluents. Because of surfactants having a major role in the membrane performance, synthetic detergent solutions present in carwash effluents were employed. Three types of surfactants were utilized: sodium dodecyl sulfate (SDS) as an anionic surfactant, cetrimide as a cationic surfactant, and Triton X-100® as a nonionic surfactant. The results showed that C100F and Ultrafilic UF possessed best performance in term of surfactant rejection due to molecular weight cutoff (MWCO) of these membranes [14]. In another study, Boussu et al. showed that the desirable quality and the intended use (recycling or discharge) of the purified wastewater determine whether to use UF or NF membranes. Four typical membranes were used; two UF membranes and two NF membranes for analyzing COD and anionic, cationic and nonionic surfactants of carwash wastewater. The authors concluded that with the proper membrane type (least membrane fouling) and process format (a hybrid process combining UF and/or a biological treatment with NF), membrane processes can be effective. UF membrane is the best option if the purified wastewater is used for recycling and the surfactants presence does not cause a problem [27]. In a related study, Boussu et al. investigated the application of NF membrane in the carwash sector using two different NF membranes. They revealed that the removal efficiency of NF membrane was 100% in terms of removing COD and surfactants. The membrane with the shortest MWCO, NF270, had the highest retentions, which is consistent with the results. Anionic surfactants (Sodium dodecylbenzene sulfonate (SDBS)) are better retained than other surfactants because the negative charges of surfactant and the membrane surface are electrostatically attracted to one another [26].

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Table 4 The legislation standards for the carwash wastewater.

Parameters	Standard in Yekaterinburg, Sverdlovsk region (Regulation No. 2329 of 2021) Wastewater composition, mg/L			Iraqi National Standards (Regulation No. 25 of 1967) Wastewater composition, mg/L		Malaysia (Industrial Effluents) Regulations 2012 Wastewater composition, mg/L	
	North Canalization Basin	Southern Canalization Basin	Sewerage Basin	Water Source	Public Sewers	Standard A	Standard B
Suspended solids	300	96.80	31.48	–	–	50	100
BOD5	169.40	40.10	30.90	<5	<40	2	40
COD	500.00	176.90	–	–	<100	80	200
Petroleum products	2.21	0.60	1.30	–	10, if water drained/source water is 1:1000; 5, if water drained/source water is 1:500; 3, if water drained/source water is 1:300	1	10
Sulphates	80.10	69.00	90.44	200	<400, if the water drained/source water is 1:1000	0.5	0.5
Chlorides	53.54	72.00	139.0	200	<600, if the water drained/source water is 1:1000	–	–
Ammonium	25.40	3.30	8.54	1	–	–	–
Anionic surfactant	0.80	1.60	2.17	–	–	–	–
Phosphates-P	0.24	0.24	2.45	0.13	0.98	–	–
Phenol	0.008	0.023	–	0.005	0.01–0.05	0,001	1
Chromium (6+)	0.0128	0.01	–	0.05	0.1	0.05	0.05
Chromium (3+)	0.0107	0.01	–	0.05	0.1	0.2	0.1
Iron	0.50	1.032	–	0.3	2	1	5
Zinc	0.05	0.051	–	0.5	2	2	2
Copper	0.0312	0.0221	–	0.05	0.2	–	–
Nickel	0.0099	0.0094	–	0.1	0.2	0.2	1
Aluminium	0.17	0.22	–	0.1	5	10	15
Manganese	0.20	0.10	–	0.1	0.5	0.2	1

In general, the solute rejection of membranes increases with decreasing membrane pore size and porosity. Istirokhatun et al. studied the applicability of four different commercial UF membranes with various molecular weight cut-off through filtration in a cross-flow set up [17]. In their study, PES10 had the highest rejection for all pollutants, which is evident in the fact that this membrane has the smallest MWCO. The UF membranes have within 90–95%, 78–100%, and 100% rejection for COD, oil and grease, and turbidity, respectively [17].

The concentration of COD can indicate contaminants outside of the vehicle such as various composting dust, bird droppings, or fallen fruit. Detergents, too, can contribute to increased levels of COD in effluents and were identified as COD in many studies [1], [18]. Lau et al. examined the treatment of carwash effluents by using three various types of commercial polymeric membranes UF and NF; their characteristics summarized in Table 5. During filtration in a lab-scale cross-flow unit, it was determined that the COD removal depend on the membrane characteristics and the best removal (91.5%) was gained by the NF270 [1]. In a similar study, Uçar [18] filtered carwash wastewater by four UF membranes of varying MWCO and one NF membrane. The COD measurements were made to specify the detergent in this study. The results showed the COD removal efficiency corresponding to 97% for NF membrane, which is more than that for the UF membrane. Furthermore, these studies emphasize the importance of pre-treatment when using membranes for treatment of carwash wastewater.

4.2. Modified membrane techniques

Membranes are generally prepared from ceramic and polymeric materials. Although the cost of manufacturing ceramic membrane is high, it can be minimized by using locally sourced precursors like pore-formers, clay, and plasticizer. Zrelli et al. prepared a ceramic membrane from clay and 22% oasis waste by using semidry-pressing process; the membrane was sintered at 800 °C and molded at 8 bars. This ceramic membrane was used to treat carwash wastewater which was collected from five car washing units located in Tunisia. The characteristics of the used carwash wastewater in this study were total solid (1115 mg/L), oil content (137 mg/L), pH = 7.5, and conductivity (915 μs/cm).

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Table 5 Carwash wastewater treatment techniques with various commercial membranes.

Membrane		Characteristics		Influent characteristics	Effluent characteristics	References
		MWCO (kDa)	Material			
UF	C100F	100	Cellulose	Synthetic solutions: Nonionic surfactant 100 mg/L; Anionic surfactant 100 mg/L; Cationic surfactant 100 mg/L;	Nonionic surfactant 55.9–94.6 mg/L; Anionic surfactant 48.3–90.1 mg/L; Cationic surfactant 5.2–83.3 mg/L	[14]
	P150F	150	Polyethersulfone			
	PES	20	Polyethersulfone			
	PES 10	10	Polyethersulfone			
	HFM 116	100	PVDF			
	HFM 180	250	PVDF			
	Ultrafilic UF	100	Polyacrylonitrile			
UF	UF P150F	150	Polyethersulfone	COD 208–316 mg/L; Nonionic surfactant 26–39 mg/L; anionic surfactant 0–0.8 mg/L; cationic surfactant 4.3–7.9 mg/L	COD 192 mg/L; Nonionic surfactant 23 mg/L; anionic surfactant 0; cationic surfactant 7.3 mg/L	[27]
	Ultrafilic	100	Polyacrylonitrile		COD ND; Nonionic surfactant 24 mg/L; anionic surfactant 0 mg/L; cationic surfactant 3.9 mg/L	
NF	NFPES10	1.2	Polyethersulfone		COD 106 mg/L; Nonionic surfactant 21 mg/L; anionic surfactant 0.4 mg/L; cationic surfactant 0.9 mg/L	
	NF270	0.17	Polyamide		COD 10 mg/L; Nonionic surfactant 1 mg/L; anionic surfactant 0.1 mg/L; cationic surfactant 0.2 mg/L	
NF	NF270	0.17	Polyamide	SS 60–140 mg/L; COD 208–382 mg/L; Nonionic surfactant 32–51 mg/L; Anionic surfactant 0.7–2.5 mg/L; Cationic surfactant 1.7–3.7 mg/L	COD 33–100 mg/L	[26]
	NFPES10	1.2	Polyethersulfone			
UF	PES10	10	Poly (Ether) Sulfone	COD 700 mg/L; O&G 36 mg/L; Turbidity 186.6 NTU	COD 33.3–40 mg/L; Turbidity 0.22–0.25 NTU; O&G 0–2 mg/L	[17]
	PS25	25	Polysulfone		COD 40–70 mg/L; Turbidity 0.24–0.57 NTU; O&G 2–8 mg/L	
	PS50	50	Polysulfone			
	PS100	100	Polysulfone			
UF	GE	1	Composite polyamide	pH 7.3; COD 314 mg /L; EC 729 µS/cm; PO43–P 9.05 mg/L	pH 7.34–7.59 COD 64.5–85.5 mg/L; EC 523–629 µs/cm; PO43–P < 1 mg/L	[18]
	PT	5	Polyethersulfone			
	PW	10	Polyethersulfone			
	MW	50	PAN/Ultrafilic			
NF	NF270	0.2–0.4			pH 7.61; COD 8.1 mg/L; EC 391 µs/cm; PO43–P < 0.05 mg/L	
UF	PVDF100	100	Polyvinylidene difluoride	COD 75.0–738.0 mg/L; Turbidity 34.7–86.0 NTU; TDS 89.2–151.8 mg/L; EC 138.8–260.7 µS/m	COD 56.11–82.41 mg/L; Turbidity 92.37–96.85 NTU; TDS 13.59–16.56 mg/L; EC 16.9–19.6 µS/m	[1]
	PES30	30	Polyethersulfone		COD 54.85–83.89 mg/L; turbidity 97.27–97.34 NTU; TDS 17.61–31.45 mg/L; EC 23.57–35.44 µS/m	
NF	NF270	0.3	Polyamide		COD 70.9–91.49 mg/L; turbidity 94.42–98.75 NTU; TDS 59.99–61.53 mg/L; EC 61.92–63.62 µS/m	

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To reduce the fouling and increase the treatment efficiency, the authors used sedimentation for 1 hour and filtration as a pretreatment of the carwash wastewater samples, and then treated them with a dead-end filtration unit. The authors examined the effect of percentage of oasis waste on open and closed porosity at a molding pressure (8 bars) and sintering temperature (800 °C), as shown in Figure 2. The concepts of open and closed porosity are used to describe different types of porous materials. The volume of fluid that the continuous fluid phase occupies in relation to the entire volume of porous material is referred to as the open porosity. In contrast, closed porosity is the percentage of the total volume in which fluids are present but cannot flow effectively [47]. The membranes had stronger hydrophilic characteristics with increased concentration of oasis waste. When the concentration of oasis waste was between 8–15% for the fabricated ceramic membrane, a sharp increase of the open porosity 26.12–40.05% and a slight variation of the closed porosity were detected, and above this concentration, no effect was observed on the open porosity, while the closed porosity increased. Furthermore, the evolution of permeate flux and oil rejection were compared for oily wastewater with 125 mg/l oil concentration and carwash wastewater for the prepared ceramic membrane, as shown in Figure 3. The authors concluded that the permeate flux decreases with the time increase, and the flux evolution of the carwash wastewater was about 5% lower than that of the oily wastewater. Also, the oil rejection of carwash wastewater was about 93% which complies with the Tunisian standard of wastewater. These outcomes are a result of using detergents composed of surfactants in the carwash units, which interact with the membrane surface to give reduced. These detergents flux reduction and improved the oil rejection [47].

Polymeric modified membranes have been employed for solving issues relating to water treatment or water reuse due to their exceptional efficiency, cost-effectiveness, clean technology, and environmental friendliness by efficiently eliminating oil and grease, detergents, and highly hazardous surfactants from carwash effluents [46, 48].

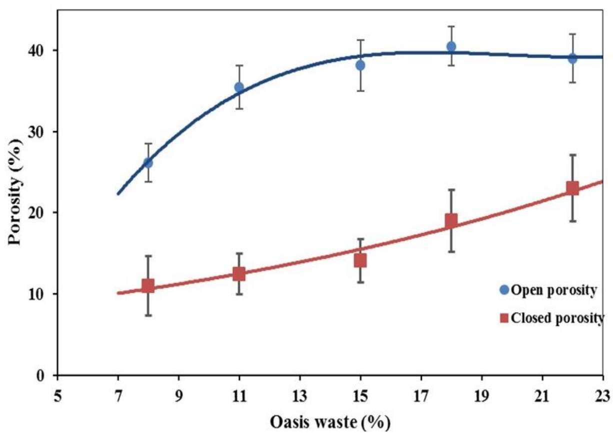

Figure 2 Open and closed porosity evolution with percentage of oasis waste [47].

There are few studies on the use of modified membranes for the treatment of carwash wastewater. Kamelian et al. prepared 10 types of acrylonitrile-butadiene-styrene (ABS) membranes with thickness ranged between 82.4–113.9 μm. The impact of solvent-nonsolvent types, ABS and additive concentrations (polyethylene glycol (PEG)) on the structures and wastewater treatment of carwash was assessed. The 1-methyl-2-pyrrolidone (NMP) and Dimethyl acetamide (DMAc) were utilized as solvents, and water and heptane (C7) were used as antisolvents. The authors clarified through the cross-sectional images that the membranes made by using the NMP/C7 pair (e.g. M2) have a denser structure in comparison with the membranes prepared using the NMP/water pair (e.g. M9) which have a fingerlike structure as shown in Figure 4, because NMP has low solubility in heptane (C7); meanwhile, NMP has a large cross-affinity with water. Also, the membranes prepared by using 20 wt.% ABS have denser and thinner structures in comparison with the membranes prepared by using lower concentration, 17 wt.%, of ABS because the high concentration of ABS increase the casting solution viscosity, cause slow demixing in the coagulation bath, and thus reduce the nuclei growth rate in the structure through phase inversion. The surface SEM images (Figure 5) of M1, M2 and M3 proves the porosity reduction with 0, 6, 10 wt.% PEG addition, respectively. Also, the authors concluded that the membrane structure, which was prepared using NMP/water pair, was negatively impacted by the AN migration to the water coagulation bath through the precipitation process. Furthermore, similar performance of the membranes prepared with NMP and DMAc as solvent results in similar impact on the membrane structure. The authors revealed that the porous membranes fabricated with similar concentrations in the water coagulation bath have lower rejection percentages of COD, TDS, and turbidity, and more stable flux. Additionally, it was shown through the study of the effectiveness of membranes prepared with various ABS concentrations that increasing the ABS concentration raises rejection rates while lowering stable flux.

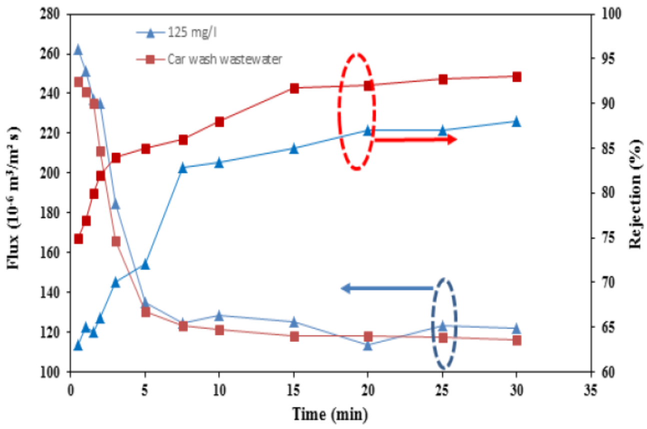

Figure 3 The permeate flux and oil rejection with time [47].

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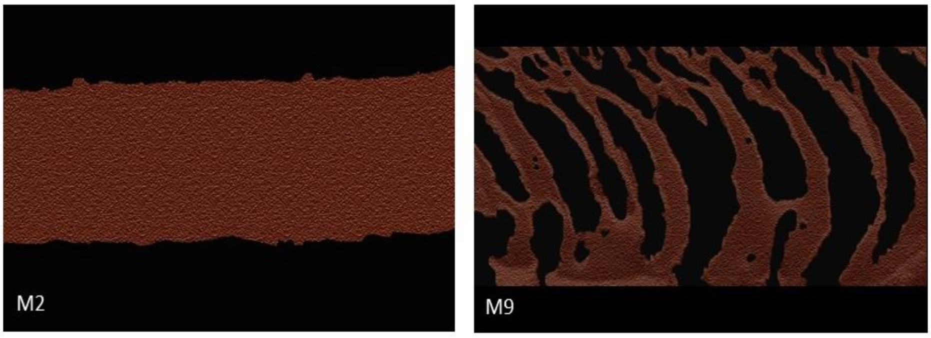

Figure 4 The cross-sectional SEM images of prepared membranes.

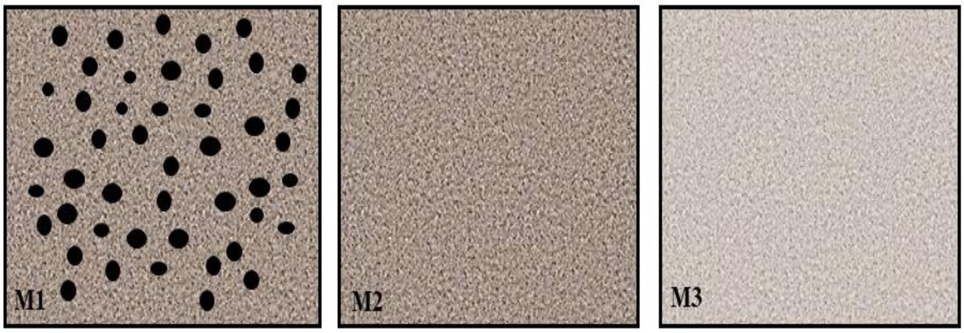

Figure 5 The surface SEM images of some prepared membranes.

Also, increasing the PEG concentration caused an increase in rejection rates and a decrease in stable flux at a constant ABS concentration. All these results are consistent with the cross sectional and surface SEM images of the membranes. Moreover, to study the migration effect from ABS to water, the authors used the Fourier transform infrared (FTIR) spectra, which focused on the characteristic peak of AN. The intensity changes as the coagulation bath changes, which means the AN migration from ABS to water occurred [25].

Kiran et al. fabricated two membranes, Cellulose acetate (CA) and polyethersulfone (PES), by using hydrophilic polymer sulfonated polyether ether ketone (SPEEK) and nanoclay bentonite, and compared their performance with the commercial polyethersulfone (PES) membrane in the removal of carwash effluent. The authors concluded based on the surface SEM images that the membranes CA/SPEEK/bentonite and PES/SPEEK/bentonite have loose porous structures with clear bigger pores, owing to the incompatibility between inorganic bentonite and an organic polymer; thus, demixing the casting dope solution improves the pore formation. Additionally, the bigger surface area and the smaller size of nanoclay bentonite cause it to disperse in both the top and bottom layers of the membrane surface. The membranes' cross-sectional images revealed a thin skin layer and finely interconnected porous structure that were responsible for the improved permeation rate brought on by the addition of both bentonite and SPEEK to the casting dope solution. A higher flux was observed for CA/SPEEK/bentonite membrane (52.3 L/m^2h) compared with that of the commercial PES membrane (41.5 L/m^2h); this might be caused by more solutes being absorbed onto the membrane surface. Also, the higher rejection, 60% COD and 82% turbidity, was gained for the CA/SPEEK/bentonite membrane. In this study, fourier transform infrared (FTIR) spectra (Figure 6) revealed that the wavenumbers 1030–1060 cm^{-1} and 550 cm^{-1} in membranes correspond to alcohols (CH_2OH) and disulfide, respectively, which showed that the pollutant was adsorbed on the surface of the membrane. Yet, the commercial PES membrane displayed a higher adsorption intensity with carwash effluent, which may be related to its hydrophobic properties [23].

5. Hybrid-membrane techniques

Numerous studies were published on using integrated membrane techniques either alone or in combination with conventional methods such us mechanical separation, flotation, chemical coagulation, etc.

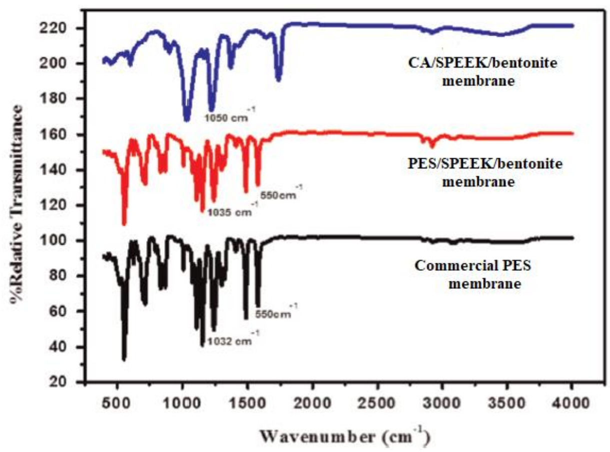

Figure 6 Fourier transform infrared (FTIR) spectra of membranes. Reproduced from ref. [23] © 2015 Elsevier Inc.

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Establishing an effective pre-treatment is crucial to ensuring that the membranes operate consistently and to improving the wastewater quality based on its nature and the discharge standard regulation. In the study of Nagamani et al. the authors used a combined pre-treatment consisting of utilizing a coarse filter (20 µm PP cartridge), an MF ceramic membrane (3 µm kaolin), and ultimately an NF membrane (500 Da Polyamide Membrane) to ensure that the filtrate is of sufficient quality to be recycled and used again. Detergents, oil, grease, and other contaminants may be indicated by the COD concentration. They found that the concentrations of COD post 20 µm cartridge filtration has a minimum change <2%, which means that the particle sizes were less than 20 µm, and the removal efficiency of COD was 99% after filtration by course filter, MF and NF membrane [19]. In another study, Moazzem et al. achieved high removal efficiency of turbidity 99.9%, suspended solids 100%, and COD 96% from carwash wastewater by using two types of membranes, including ceramic UF membrane and reverse osmosis (RO) with pretreatment as coagulation flocculation process and sand filtration [22]. Also, Tan and Tang clarified that it is preferable to utilize pretreatment before UF membrane to prevent more contaminants from adsorbing in pores. It was shown that using chemical coagulation as pretreatment by adding $KMnO_4$ to the coagulant PAC can improve efficiency of the coagulation and help to reduce the blockage of the two types of PS UF membranes with MWCO (6 and 20 kDa) which were used [24]. In a similar study, Tang et al. separated carwash wastewater by using two sorts of PS hollow fiber UF membranes (6 and 20 kDa) with enhanced coagulation and granular activated carbon (GAC), resulting in good removal levels of oil, COD, BOD, and LAS. The GAC tank following the UF membrane can efficiently adsorb LAS, odor, and color. A schematic diagram of the apparatus is presented in Figure 7. AFM analysis was conducted on the membrane (Figure 8), and it was concluded that the LAS existence in carwash wastewater might loosen the gel layer and improve the membrane flux [35].

Moreover, Shete et al. checked the feasibility for obtaining refuse-free water by using sedimentation and induced air flotation as a pretreatment, and then cross-flow UF and reverse osmosis. The removal efficiency values for the total dissolved solids (TDS), total suspended solids (TSS), oil and grease, and COD by UF were 81%, 82%, 74%, and 67%, respectively (the effluent poses can be safely discharged into any local water bodies, including rivers), and by RO these values were 91%, 82%, 90%, and 81%, respectively (it is safe to reuse the effluent for any productive activity, such as car washing or landscape gardening). It was possible to obtain a sufficient quantity of water with high purity from carwash effluent using these procedures [2]. Table 6 provides a summary of several studies on hybrid-membrane techniques that were conducted for the treatment of wastewater from car washes.

In the study by Jiku et al., coagulation-flotation was utilized as a pre-treatment before hollow fiber ultrafiltration membrane for oil removing from car washing wastewater. The oil content in car washing effluents ranged from 5.2 to 13.47 mg/L. According to the their results, the combined processes may remove more than 40% of the oil content, more than doubling the oil removal rate of the traditional coagulation and flotation method [49].

6. Limitations

This review studied the membrane filtration techniques and their effectiveness in eliminating various contaminants from carwash wastewater, including surfactants, oil products, dissolved and suspended particles. The major restriction of membrane filtration for the treatment of carwash is the rapid membrane flux reduction during the operation; therefore, the selection of the suitable membrane or modification of the structure of membrane might be necessary.

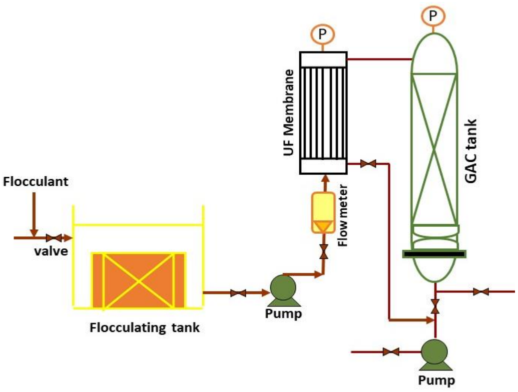

Figure 7 Schematic diagram of system setup.

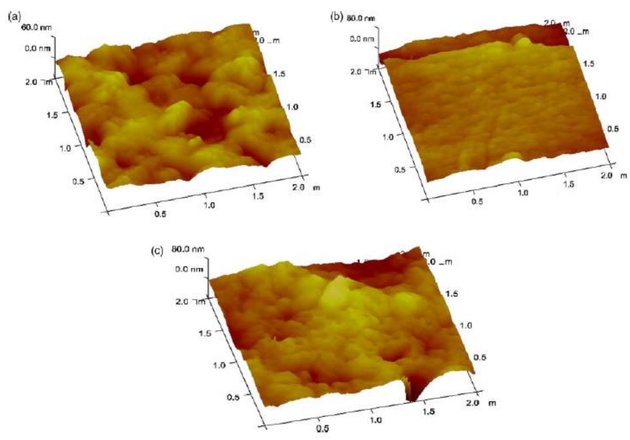

Figure 8 AFM analysis on UF membrane: (a) new UF membrane, (b) membrane contaminated with oil and (c) membrane contaminated with oil and LAS. Reproduced from ref. [35] © IWA Publishing 2007.

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Table 6 Carwash wastewater treatment methods based on Hybrid-Membrane techniques.

Pretreatment	Membrane	Characteristics			Influent characteristics	Effluent characteristics	Ref.
		MWCO (kDa)	Pore size, μm	Material			
Coarse filter PP	Ceramic-MF	–	3	kaolin	pH 6.4; EC 680 μS/cm; COD 460 mg/l; TS 1280 mg /L	After MF+NF pH 7.73; EC 226 μS/cm; COD 4 mg/l	[19]
	NF	0.5	–	Polyamide			
Coagulation-flocculation, sand filtration	Ceramic-UF	–	0.02	Zirconia oxide	pH 4.66–6.42; EC 404–509 μs/cm; Turbidity 522–763 NTU	UF pH 5.32; EC 273–298 μs/cm; Turbidity 1.84–0.86 NTU	[22]
	RO	–	–	–		After UF+RO pH 5.32; EC 7.6–14.6 μs/cm; Turbidity 0.86–0.16 NTU	
Enhanced coagulation	UF	6 20	–	Polysulfone	pH 6.5–8; Turbidity 70 NTU; COD 100–160 mg/L; Oil 5–25 mg/L; LAS 2–5 mg/L	Not declared	[24]
Enhanced coagulation	UF	6 20	–	Polysulfone	Not declared	COD 33.4 mg/L, BOD 4.8 mg/L, Turbidity 0.42 NTU, LAS 0.06 mg/L, oil 0.95 mg/L, SS 1 mg/l DS 115 mg/l	[35]
	Granular activated carbon (GAC)						
Sedimentation & Induced air flotation	UF	–	–	–	pH 8.5; Turbidity 7.7 NTU; TDS 7920 mg/l; TSS 1134 mg/l; COD 288 mg/l; O&G 34.19 mg/l	UF pH 7.26; Turbidity 1.20 NTU; TDS 791.5 mg/l; TSS 124 mg/l; COD 83 mg/l; O&G 3.11 mg/l	[2]
	RO	–	–	–		After UF+RO pH 6.38; Turbidity 0.0 NTU; TDS 140 mg/l; TSS 10 mg/l; COD 12.8 mg/l; O&G 0.31mg/l	
Coagulation - Flotation	Hollow fiber UF	–	–	–	pH 6.9–7.6; Oil and grease 5–13.4 mg/L; Turbidity 362–450 NTU	Oil and grease >2.1–5.4 mg/L	[49]

7. Conclusion and future prospects

According to the reviewed studies into the membrane filtration systems and hybrid membrane technologies, traditional filtration methods such as mechanical separation, coagulation-flocculation process, air flotation, etc., are the most interesting choices for treating carwash wastewater and getting rid of various contaminants that pose a serious threat to the environment. Moreover, these methods cost effective and can produce high-quality filtering. The concentrations of surfactants and oily products were also found to be reduced based on the treatment type and the membrane technology employed. The main approach to obtaining new modified membrane structures is a combination of various polymers and other materials in the manufacturing technology, which allows to improve the removal efficiency and other performance properties of membranes.

This study allows giving some suggestions for the future works in the carwash sector. Firstly, it is necessary to conduct more studies on using the modified membranes to treat wastewater for car washes, to consider the cost and the main characteristics, and then to apply the membranes on field-scale to obtain an efficient wastewater treatment. Also, developing good and cost-efficient membrane filtration techniques and using them in the car washing will reduce the cost of using municipal water through recycling.

• Supplementary materials

No supplementary materials are available.

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• Conflict of interest

The authors declare no conflict of interest.

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