Recent Advances in Physicochemical and Biological Techniques for the Management of Discharges Loaded with Surfactants

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The spectacular evolution of the urban and industrial sector today poses real environmental challenges of water pollution that requires immediate attention. Surfactants are emerging contaminants that pose a significant problem in wastewater treatment, and their presence causes difficulty in traditional treatment processes. In this context, the present work critically reviews the impacts of surfactants and their toxicity on the environment and human health while presenting the various techniques used in wastewater treatment plants to reduce their effects. Surfactants are removed from wastewater using different techniques, including physical, chemical, biological, and membrane treatment. The choice of the most appropriate technique for wastewater treatment is based on many criteria, such as effluent quality, standards to be respected, investment and operating costs, and environmental footprint. Adsorption and coagulation-flocculation are the most suitable techniques for removing detergents from wastewater due to their effectiveness, ease of use, environmental friendliness, and cost-effectiveness.

Keywords
pollution, wastewater, surfactant, impacts, treatment

Introduction

Industrial wastewater rich in toxic materials poses serious problems for the environment. A large number of pollutants, such as heavy metals, dyes, surfactants, pharmaceuticals, and pesticides are used in industrial and domestic products today[1,2]. After intensive use of these products, a large number of pollutants end up at different levels of the water cycle, especially in urban areas[3]. Surfactants are one of the most encountered families of pollutants in wastewater[4,5].

Detergents are compounds widely used in domestic and industrial applications for different purposes, such as foaming, emulsion stabilization, paints, mineral separations, pharmaceutical formulations, fire-fighting applications, and herbicide and insecticide formulations[6]. These surfactants are necessary and irreplaceable compounds in the field of hygiene and cosmetology due to their amphiphilic chemical structures characterized by two parts of different polarity, one lipophilic is apolar, the other hydrophilic is polar[7]. In addition, surfactants allow immiscible substances, such as water and oil, to mix to form and stabilize assemblies[4,8]. Micelles begin to form as the surface tension decreases with increasing surfactant concentration in the aqueous medium. Indeed, a suitable surfactant can reduce the interfacial tension between polar and apolar liquids from 40 mN m^{-1} to 1 mN m^{-1} (between water and n-hexadecane)[9]. Surfactants are classified into four main classes based on the charge of the hydrophilic part: anionic, cationic, nonionic, and amphoteric[7,10].

Detergent use is widespread and has been growing for several years, from 15.93 million tons used in 2014 to 24.19 million tons projected in 2022[11]. Many industries use detergents, including the textile, pharmaceutical, cosmetic, mining, oil recovery, paper industries, etc.[12] Aditionally, fluorinated surfactants constitute an important class of fluorinated compounds used in plastics and pharmacology, and in the anti-icing treatment of aircraft[13]. Moreover, the detergents used in developing countries are often difficult to biodegrade, since the population often turns to cheap detergents (non-biodegradable products authorized in the developing countries' market).

After use, surfactants are ultimately released into the aquatic ecosystem due to wastewater treatment plant discharges into rivers, oceans, lakes, and estuaries or through direct discharge of raw sewage[14]. Its presence in surface waters is primarily

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linked to anthropogenic activities like runoff and storm water discharges from urban and industrial areas, and direct effluent discharge[15]. As a result, significant amounts of surfactants are found in the environment (rivers, lakes, soils, and sediments). Domestic wastewater can contain up to 10 mg L^{-1} of surfactants, while wastewater from industries that manufacture surfactants can contain up to 300 mg L^{-1} [16,17]. They are often highly persistent, water-soluble, and poorly biodegradable contaminants[18]. In addition, they can pass through wastewater treatment processes and reach drinking water resources, posing a serious health risk to humans, animals, and aquatic life[19,20].

Non-biodegradable and more persistent surfactants, such as perfluorinated surfactants, are dangerous and harmful compounds causing the destabilization of aqueous flora and fauna[20]. They are detrimental to humans, aquatic life, and vegetation. The existence of surfactants in water above a specific concentration reduces water quality, induces unpleasant taste and odor, and causes short- and long-term changes in the ecosystem[10].

Many studies have reported various surfactants' toxicity and environmental effects, mainly on different aquatic organisms[21]. Acute toxicity tests show that LC50 values for some detergents vary considerably between species for nonionic surfactants, cationic surfactants, and anionic surfactants[21]. According to the authors, many of the surfactants tested, anionic and nonionic, were found to be toxic enough to be classified as very toxic and harmful, according to the European Union Directive 67/548/EEC.

Disposal of detergents by different techniques becomes a necessity for environmental protection[22]. Various physical, chemical, biological, and membrane treatment techniques are used to remove surfactants from wastewater[2,10,23]. Wastewater treatment is chosen based on various factors, including effluent quality, ecological consequences, treatment cost, and secondary waste generation.

This study aims to assess the toxicity of surfactants in the environment, their behavior in different ecological systems, their impacts on wastewater treatment plants, and the various treatment processes used to remove surfactants.

Environmental impacts of effluents containing detergents, and their toxicity

Surfactants are dangerous and harmful compounds that cause destabilization of the aqueous flora and fauna, making them harmful to humans, aquatic life, and vegetation[1,12,24].

Table 1 – *Hazards associated with surfactants*

Risks	References
Disruption of self-remediation processes in aqueous systems, toxicity to aquatic organisms.	29
Anionic surfactants, which are the most commonly used, are more toxic to fish than are nonionic surfactants.	30
Eutrophication of water systems.	31
Persist by becoming a source of ecotoxicity in the environment.	32
Acute toxic effects on the skin, eyes, mucous membranes, upper respiratory tract, and stomach for humans.	33
Affect different stages of the respiratory process.	34
Destroy the function and structure of bacterial membranes of microorganisms.	10
Toxic effect on marine organisms.	35
Inhibit photosynthesis and the growth of cyanobacteria.	36
Cationic surfactants are not readily biodegradable in seawater.	37

Surfactants act in synergy with other toxic compounds in the water, which can change the physicochemical characteristics while producing other toxic products[25]. Furthermore, perfluorinated surfactants repel water, grease, and dust, which justifies their use as anti-adhesive, waterproofing, and protective agents[26]. They persist and accumulate in living organisms, causing developmental and reproductive problems and metabolic disorders. Indeed, some detergents have been banned by the European Directive 2006/122/EC after being detected in human blood. However, special authorizations have been granted to specific industries where certain detergents cannot be replaced[26]. Borghi *et al.*[27] noted that the toxicity of surfactants increases with increasing hydrophobicity. Furthermore, increasing the length of alkyl groups increases the hydrophobicity of surfactants, leading to the increase in toxicity of the molecule, while increasing the ethylene oxide (EO) in a molecule decreases the hydrophobicity and toxicity of surfactants[28]. The hazards associated with surfactants are summarized in Table 1.

Voloshina *et al.*[38] have shown that surfactants exhibit marked biological activity. Anionic detergents can combine with bioactive macromolecules such as peptides, enzymes, and DNA[39]. Binding to proteins and peptides can alter a molecule's polypeptide chain folding and surface charge, changing biological function.

The primary target site for cationic surfactants is the bacteria's cytoplasmic (inner) membrane. Quaternary ammonium compounds (QACs) bind to

Table 2 – *Toxicity of different types of surfactants to various aquatic and terrestrial species*

Species		Surfactants	End point	Concentration	References
Aquatic	*Vibrio fischeri* (Bacteria)	QAC	EC_{50} (Luminescence 30 min)	0.5 mg L^{-1}	41
	Daphnia magna (Invertebrate)	AEO	EC_{50}	0.36 – 50.5 mg L^{-1}	15
	Dunaliella sp. (Bacteria)	LAS	EC_{50} (24 h)	3.5 mg L^{-1}	42
	Pseudokirchneriella subcapitata (Algae)	PFOS	EC_{50}	146 µM	15
	Gammbusia affinis (Fish)	SDS	EC_{50} (Immobilization 48 h)	40.15 mg L^{-1}	42
Terrestrial	*Brassica rapa* (Terrestrial)	LAS	EC_{50} (14 days)	137.7 mg kg^{-1}	43
	Isotoma viridis (Soil fauna)	LAS	LC_{50} (Mortality)	661 mg kg^{-1}	43
	Folsomia candid (Invertebrate)	NP	EC_{50} (21 days)	5–133 mg kg^{-1}	15

the cell's internal membranes and disrupt them via their long alkyl chain.

In addition, nonionic surfactants exert antimicrobial activity by binding to various proteins and phospholipid membranes[40]. This binding increases the permeability of membranes and vesicles, causing low molecular weight compounds to leak, which can lead to cell death or damage by losing ions or amino acids.

To date, increased attention has been paid to the ecological risk and potential toxic effect of environmental surfactant residues. Table 2 presents the toxicity tests of certain surfactants for various aquatic and terrestrial organisms.

Excessive use of any surfactant and its release into the environment, particularly aquatic organisms, can have serious ecosystem impacts[12]. The amounts of anionic, nonionic, and cationic surfactants released into wastewater and aquatic environments must be monitored and regulated. Cationic surfactants are recognized as presenting the greatest hazard, and their limits in wastewater should be the lowest[42].

Impacts of surfactants on the operation of wastewater treatment plants

Most detergents persist in the wastewater to be treated before being broken down throughout the various stages of the treatment plant, which disrupts its operation. The foam generation is one of the problems detected in the coagulation-flocculation process of the pretreatment station in Kenitra (Morocco)[44]. The leachate samples taken at the level of the coagulation-flocculation basin, presented in Fig. 1, show the presence of foams generated by the

Fig. 1 – *Foam generated by the detergents at the WWTPs of the city of Kenitra*

Table 3 – *Impacts of surfactants on WWTPs*

Title	Impact on WWTPs	References
Surfactants in the aquatic and terrestrial environment: occurrence, behavior, and treatment processes	Surfactants can cause foaming and reduce reoxygenation and oxygen levels, resulting in deterioration of water quality and toxic effects on water-dwelling organisms.	15
Surfactants and personal care products removal in pilot-scale horizontal and vertical flow constructed wetlands while treating greywater	With excessive operation and maintenance costs, conventional systems require an uninterrupted power supply.	45
Scale-down studies of membrane bioreactor degrading anionic surfactants wastewater: Isolation of new anionic-surfactant degrading bacteria	Surfactants can affect the biological treatment process of wastewater and cause problems in aeration and wastewater treatment plants due to their high foaming power, low oxygenation potential, and the death of waterborne organisms.	46
Treatment of high strength aqueous wastes in a thermophilic aerobic membrane reactor (TAMR): performance and resilience	Toxic effects and foam formation at high concentrations.	47
Removal of nonionic and anionic surfactants from real laundry wastewater using a full-scale treatment system	For biological treatments: long reaction time, foaming, and death of the biomass at high surfactant concentration, and high sewage sludge production must be disposed of when the surfactant concentration is very high.	48
	For physical-chemical treatments: high operating costs due to the need to regenerate/dispose of exhausted adsorbent materials, high oxidant costs, and high sludge production to be disposed of (both types of treatments).	
Effects of detergents on natural ecosystems and wastewater treatment processes: a review	Reduce the efficiency of wastewater treatment plants.	19
	Interfere with water coagulation. High resistance to biological degradation.	
Impact of various surfactant classes on the microorganism community used for WWTP biodegradation treatment	The massive presence of surfactants in domestic and industrial wastewater could affect wastewater treatment plants by inhibiting the activated sludge in the pollutant biodegradation treatment step.	5

detergents present in the leachate in contact with the ferric chloride ($FeCl_3$), which was used as a coagulant. Indeed, the addition of antifoam was used to avoid the formation of foam, which allowed good agglomeration of the pollutants during treatment with $FeCl_3$.

Thus, several studies, shown in Table 3, have examined the impacts of detergent-rich wastewater on different types of wastewater treatment plants and their operation.

Treatment methods

Physical-chemical treatment

For the control of wastewater pollution by detergents, several physical-chemical treatment methods are used:

Treatment of detergents by coagulation-flocculation

The coagulation-flocculation process is one of the first steps in wastewater treatment, and is considered one of the most important and widely used treatment processes as a common large- and small-scale method in urban and industrial wastewater treatment, due to its simplicity and efficiency[49,50].

In the coagulation/flocculation process, many factors can influence the efficiency of the process, such as the type and dosage of coagulant/flocculant, pH, mixing speed and time, temperature, and retention time, etc.[51,52]. Optimizing these factors can significantly increase the efficiency of the process[53].

Coagulation-flocculation is now commonly used for the first segment of surfactant effluent treatment due to its high efficiency, low cost, and simple operation[54,55].

Several studies have focused on optimizing the operating parameters of coagulation-flocculation: the type of coagulant and flocculant used, dose, pH, agitation speeds, and settling time. Zhu *et al.*[56] conducted a comparative study of different coagulants of Al^{3+}, Fe^{3+}, or Ca^{2+} with ecological bamboo pulp cellulose-g-polyacrylamide (BPC-g-PAM) as a flocculant; the results suggested that the combination of Fe^{3+} + BPC-g-PAM showed the best coagulation-flocculation performance to the surfactant effluent, due to its minimum dosage of coagulant, BPC-g-PAM, and fast processing time.

To treat laundry wastewater containing surfactant (linear alkylbenzene sulfonate/LAS), Maryani *et al.*[57] used PAC as a coagulant for coagulation-flocculation treatment. The results showed that the treatment process was highly effective for 60 minutes with the addition of 100 grams of PAC, which decreased LAS concentration from 2.02 mg L^{-1} to 0.02 mg L^{-1}.

On the other hand, another study investigated the use of Alyssum mucilage as a new natural coagulant for the treatment of detergent-rich wastewater; under the optimal conditions of the coagulation-flocculation process, the maximum COD and surfactant removal efficiencies were determined to be 84.63 % and 99 % respectively[58].

Mohan[59] demonstrated a laundry rinse water treatment system to remove surfactants from laundry waste using a natural coagulant (Nirmali seeds). In this experiment, 96.3 % of the anionic surfactant linear alkyl benzene sulfonate (LAS) was removed from the overall treatment system when Nirmali seeds were used as coagulant. This showed that the treated water was safe if released into the environment.

Bakraouy *et al.*[44] tested a ferric chloride-rich discharge as a coagulant compared to its combined effect with poly aluminum chloride (PAC), as well as with PAC alone, on the treatment of leachate from the detergent-rich landfill of the city of Kenitra, Morocco. Applying the new coagulant ($FeCl_3$-rich industrial discharge) alone resulted in a 97 % removal of detergents. The study of the mixture of the coagulant rich in ferric chloride with the PAC allowed the elimination of 91 % of the detergents, while the PAC alone allowed 95 % removal of detergents.

The coagulation/flocculation process remains an available treatment for removing detergents from industrial wastewater. However, using chemical coagulants and flocculants can lead to residuals and metal compounds in the generated sludge, and is difficult to remove after treatment, which produces secondary pollution to the receiving environment[64]. For these reasons, studies are pushed towards using coagulants and flocculants of natural origin that are readily biodegradable, available from reproducible natural resources, and do not produce secondary pollution, as a biological process that can effectively degrade the generated sludge.

Detergent treatment by flotation

Flotation is one of the most considered and used separation processes, combining three phases: gas phase, liquid phase, and solid phase[65]. Generally, flotation is a technique based on separating solids from a liquid body using air bubbles (artificial flotation) or without air injection (natural flotation). In principle, it is a solid-liquid separation process using bubbles generated by O_2 soluble in the liquid[66]. However, air remains the most frequently used and functional gas in practice because it is cheaper and easily affordable, and different flotation processes are used depending on their applications. This technique has been used in many fields, such as wastewater treatment, mineral treatment, and drinking water treatment. The different types of flotation processes are distinguished according to the method of bubble production: electrolytic flotation, dispersed air flotation, dissolved air flotation, and natural flotation to be used[66,67]. After the introduction of air bubbles that act as a transport medium for the particles in the liquid medium to be treated, the particles attach to the air bubbles to move to the surface of the aqueous solution[68,69].

Flotation is an effective, readily employed, and economical technology for treating wastewater con-

Table 4 – *Removal of COD and surfactants from wastewater by a coagulation-flocculation process*

Origin of wastewater	Surfactant (mg L^{-1})	COD (mg L^{-1})	Coagulant/flocculant	Coagulant/ flocculant concentration (mg L^{-1})	Surfactant removal (%)	COD removal (%)	References
Laundry wastewater	19.68	280.00	$ZnCl_2$+Mineral ash / Praestol -650	29.54 + 1936.35 / 196.38	74.00	68.00	60
Lixiviat	77.58	4416.00	PAC	1710.00	95.00	96.00	44
Industrial wastewater	80.33	48241.00	Cactus pads (genus opuntia) + 30 % iron chloride 3 ($Fecl_3$)	1480.00	78.00	90.00	61
Car wash wastewater	81.00	543.00	TanFloc (naturel coagulant)	220.00	64.20	61.00	62
Oily saline wastewater	55.00	1203.00	Lallemanta mucilage	10.00	20.60	87.60	63
Laundry wastewater	2.02	–	PAC	1000.00	99.00	–	57
Oily saline wastewater	55.00	1202.50	Mucilage d'Alyssum	40.50	99.00	85.00	58

taining detergents, because detergents consist of a polar ionic head and a nonpolar hydrocarbon chain. The attachment of the polar head group to the target ion leads to the exposure of the nonpolar hydrophobic section of the surfactant in the solution, which facilitates contact when air bubbles are introduced into the flotation cell[70].

Detergents can adsorb to the solid/liquid interface used as a collector to alter the surface wettability of mineral particles[71].

The results of a study evaluating the separation efficiency of the oil fraction from soap and detergent industry effluents, using the dissolved air flotation (DAF) process with a biosurfactant (*Pseudomonas cepacia*) as an alternative collector, showed that the DAF-biosurfactant system increased the process oil separation efficiency from 69.5 % using only microbubbles to 89.8 % using the isolated biosurfactant[72].

Dyagelev *et al.*[73] used laboratory flotation equipment (LFM-001) to treat wastewater with high petroleum and surfactant content. After water-air mixing, the oil particles formed flakes that floated at the interface in the presence of a coagulant.

Another study by Kastali *et al.*[22] illustrates the removal of detergents from vegetable oil-laden wastewater by natural flotation. The results showed that natural flotation could remove 80 % of the detergents. This offers the value of natural flotation for the discharge of surfactant-laden vegetable oils.

Detergent treatment by adsorption

The adsorption process remains the most studied and applied method in industries for removing detergent-related pollution. It can occur at an interface between two-phase interfaces such as liquid-solid, liquid-liquid, or gas-liquid[74].

Adsorption processes are widely used for wastewater treatment and industrial effluents[75,76]. Piccin *et al.* and Syafiuddin *et al.*[77,78] have shown that the economic and technical feasibility of adsorption processes depends on several factors, including the type of adsorbent, the physicochemical characteristics of the effluent to be treated, and the pollutants to be removed, operating conditions, process configuration, discharge standards, and regeneration of the adsorbent material.

In general, adsorbents must have relatively large surface areas and mechanical stability, and must be recyclable[79]. In addition, several factors can influence the effectiveness of the adsorbent, temperature, pH, agitation time, filtration rate in the dynamic study, and pollutant load[80].

To this end, activated carbon (AC) is the universal adsorbent for the liquid phase, and remains the main commercial product for removing water pollution, especially detergents[77]. It is an effective adsorbent in wastewater treatment due to its ability to remove a wide variety of pollutants: pesticides, heavy metals, surfactants, color, odor, and other unwanted organic and inorganic pollutants in wastewater treatment.

Activated carbons can be produced from natural or synthetic materials. As agricultural residues, olive grains and biomass are the most favorable due to their abundance and low cost[81].

Activated carbons have a high capacity to adsorb surfactants due to their large specific surface areas and hydrophobic nature. The combination of high pH-independent power with a solid carbon-surfactant interaction means that activated carbons are among the most effective adsorbents for removing surfactants from wastewater[82].

The effect of particle size and pore size on the adsorption of the anionic surfactant sodium dodecyl sulfate (SDS) has been studied extensively. Binders and Franco[83] used four activated carbons with different particle and pore sizes. The study showed that activated carbon with small pore sizes of 0.56 to 0.77 nm adsorbed more SDS surfactant than other activated carbons.

Valizadeh *et al.*[84] conducted a study to remove anionic surfactant sodium dodecylbenzene sulfonate (SDBS) using adsorption on activated carbon prepared from pine cones and activated by potassium carbonate. The results showed an adsorption capacity of 97.6 mg g^{-1} in adsorption by a mass of 303 mg g^{-1} of granular activated carbon (GAC) in a fixed bed column with a maximum exhaustion time of about 10 days at an initial SDBS concentration of 50 mg L^{-1}, bed height of 2 cm, and flow rate of 120 mL h^{-1}. Indeed, the removal of SDBS in the presence of an increasing dose of adsorbent from 0.1 to 0.5 g L^{-1} increased from 69.3 % to 96 %. This was due to the extension of ion exchange sites on the surface of (GAC), leading to more significant binding of SDBS to the adsorbent surface.

The use of microfiltration as a secondary membrane on the surface or in the pores of the membrane during powdered activated carbon treatment to remove anionic surfactants linear alkyl benzene sulfonate (LABS) and cationic cetyltrimethylammonium bromide (CTAB) increased removal efficiency[10].

According to a study by Siyal *et al.*[85], charcoal fly ash showed 96 % removal of the anionic surfactant sodium dodecyl sulfate (SDS) with a reported adsorption capacity of 0.96 mg g^{-1} with a dose of 100 g L^{-1} of fly ash.

To remove detergents from laundry wastewater by adsorption on different materials, Siswoyo *et al.*[86] investigated the ability of aluminum sulfate-rich drinking water treatment plant sludge as an adsorbent combined with phytoremediation system to remove chemical oxygen demand (COD) and surfactant in laundry wastewater. The results showed that the combined phytoremediation adsorption system could be considered a suitable environmental technology since it removed 77.5 % and 99.9 % of chemical oxygen demand (COD) and surfactant.

In another research investigating the use of waste material as an environmentally friendly adsorbent, Azad[87] prepared an adsorbent material of recycled egg cartons coated with candle soot in a mixture with acetone to remove the detergent from wastewater. The results showed that the carbon-coated recycled egg carton had a good adsorption capacity for the detergent.

To test the removal of four linear alkylbenzene sulfonates (LAS) LAS C10, LAS C11, LAS C12, and LAS C13, from wastewater samples, Orta *et al.*[79] used two high-swelling micas (Na-Mica4 and C18-Mica-4) as adsorbent material. Adsorption tests showed that equilibrium for C18-Mica-4 was reached within 30 min with removal rates up to 98 %, while Na-mica-4 required more time to reach low removal rates compared to the results obtained with C18-Mica-4. Table 5 shows the removal of different types of surfactants from wastewater using different adsorption materials.

The adsorption treatment of detergent-loaded wastewater remains a very useful and effective technique for removing several surfactants, even at low concentrations. Indeed, activated carbon is very impressive for the adsorption of surfactants. However, its use is not very limited due to economic considerations. Therefore, it is crucial to develop highly effective adsorbents that are low-cost, environmentally friendly, and readily available to absorb surfactants from wastewater. Using low-cost adsorbents has some advantages, such as high adsorption capacity, easy separation, abundance, low cost, and renewability.

Treatment of detergents by membrane filtration

Membrane filtration systems are widely used in different industrial fields, such as water desalination, wastewater treatment, biotechnology, etc.[89] The membrane filtration process is a physical separation technique in the liquid phase using a permeable and selective membrane, described as permselective. Depending on its intrinsic characteristics and mode of operation, the membrane blocks the passage of some contaminants while allowing other pollutants to pass[90]. During the filtration process, dissolved and particulate compounds in the water are separated by the membrane[91,92].

Membrane filtration is an increasingly important technique for treating wastewater from various sources (municipal wastewater, plant effluent wastewater). In addition, membrane filtration removes dissolved particles, such as detergents, heavy metals, oils, and greases. Moreover, the treatment of wastewater could result in water that meets the standards for irrigation, thus reducing the demand for freshwater. This shows that membrane filtration must be adapted to each specific process[93].

Several studies have been conducted on membrane filtration processes for detergent removal from wastewater[94].

Linclau *et al.*[95] used nanofiltration combined with a membrane bioreactor (MBR) to treat and recycle wastewater from a detergent production site using tubular membranes. This resulted in a permeating quality with COD <50 mg L^{-1} and anionic surfactant concentration <0.5 mg L^{-1}, producing water that met the standards of water used for cooling. On the other hand, Hube *et al.*[96] carried out treatment of laundry wastewater that contained 10.06 mg L^{-1} of anionic surfactant by ultrafiltration (UF) and reversed osmosis (RO). After ultrafiltration, the anionic surfactant was insufficiently reduced (still 7.20 mg L^{-1} to be removed), so the reverse osmosis step was necessary. The ultrafiltration step, howev-

Table 5 – *Removal of surfactants from wastewater by adsorption process*

Types of wastewater	Adsorption material	Types of surfactants	Percentage of removal	References
River water (with different concentrations of SDBS added in the laboratory)	Activated carbon prepared from pine cones and activated by potassium carbonate	Anionic surfactant sodium dodecylbenzene sulfonate (SDBS)	69 to 96 %	84
Aqueous solution	Charcoal fly ash	Anionic surfactant sodium dodecyl sulfate (SDS)	96 %	85
Laundry wastewater	Sludge from wastewater treatment plants		99 %	86
Wastewater treatment plant located in Sevilla (southern Spain)	High-swelling mica (C18-Mica-4)	Linear alkylbenzene sulfonates (LAS)	98 %	79
Aqueous laboratory solution	Chitosan films	Anionic surfactant sodium dodecylbenzene sulfonate (SDBS)	–	88

er, guaranteed good performance and duration of the reverse osmosis membrane. The total anionic surfactant's overall removal efficiency (ultrafiltration + reverse osmosis) was 99.2 %, with a remaining concentration of 0.91 mg L^{-1}. In addition, the recovery of water and detergent from laundry wastewater by Barambu *et al.*[97] using a tiltable filtration panel system for the treatment of laundry wastewater by filtration allowed the reuse of water with detergent recovery. The microfiltration membrane was chosen because of its low intrinsic strength, allowing the filtration system to operate under intense pressure. In this system, the combination of aeration rate and inclination angle improved permeability up to 83 %, with a recovery of 32 % of detergent.

There is interest in treating detergent-rich wastewater with ultrafiltration (UF). This process is often required to remove pollutants from secondary solid waste caused by other upstream treatments[98]. Recently, attention has been given to the potential of UF to recover key chemicals from wastewater streams, including nutrient recovery for algal bioreactor growth[99], brackish water from detergent-rich textile wastewater[100], and fluoride[101].

However, ultrafiltration often accepts low molecular weight compounds, such as phenol, a particularly harmful wastewater constituent from the olive oil industry[102]. Adding surfactants above the critical micellar concentration (CMC) solubilizes pollutants in larger micelles and removes them[102]. This technology has recently received significant attention in the literature due to the improved performance demonstrated, and focuses primarily on improving the performance of the technology[101,103].

Treatment of detergents by advanced oxidation processes

Advanced oxidation processes (AOPs) are considered a highly competitive and very reasonable technology for treating domestic and industrial wastewater, especially for removing organic compounds classified as bio-recalcitrant, detergents, and the demobilization of pathogens not reachable by traditional methods[104,105].

The study conducted by Tri Wahyuni[106] showed good removal of detergents from a laundry wastewater using photodegradation under $UV/TiO_2/H_2O_2$ (photo-Fenton type) and $UV/Fe^{2+}/H_2O_2$ (photo-Fenton). The efficiency of surfactant photodegradation was found to be controlled by TiO_2 dose, pH, H_2O_2 concentration, and treatment time for the $UV/TiO_2/H_2O_2$ system, and that of Fe(II) and H_2O_2 concentrations, pH and UV exposure time for the $UV/Fe(II)/H_2O_2$ (photo-Fenton) process.

To remove an anionic surfactant, sodium dodecyl sulfate (SDS), Mirbahoush *et al.*[107] used a coupled coagulation-Fenton treatment process. In the pretreatment step, coagulation using flaxseed mucilage (FSM) at the optimal dose of 100 mg L^{-1} resulted in 80.8 % surfactant removal. In the post-treatment step, heterogeneous photo-Fenton oxidation using $MnFe_2O_4$ nanocatalyst was applied to remove the remaining SDS, and complete removal of surfactant was achieved in a short reaction time and at low doses of $MnFe_2O_4$ nanocatalyst and H_2O_2. The combination of the two processes showed excellent performance for the treatment of real wastewater samples from a car wash.

Another study examined the removal of the anionic surfactant sodium dodecyl sulfate (SDS) by the $O_3/UV/H_2O_2$ process using a central composite design based response surface methodology that helps to evaluate the process parameters, especially the initial pH, H_2O_2 concentration, ozone dosage, and reaction time on SDS removal. The study showed that the $O_3/UV/H_2O_2$ process achieved 96 % removal of the anionic surfactant sodium dodecyl sulfate (SDS)[108].

Sugiharto[109] tested the removal of an anionic surfactant sodium dodecylbenzenesulfonate (DBS) from laundry wastewater by photodegradation with TiO_2 as a photocatalyst for DBS removal under different conditions using TiO_2 without UV light, and both UV light and TiO_2. The results showed that 16 % of DBS was removed using TiO_2 without UV light treatment. However, when steaming TiO_2 with both UV light treatments, the removal of DBS reached 98 %, indicating significant photodegradation.

Also, Mondal *et al.*[110] performed a comparative study of UV and UV-H_2O_2 advanced oxidation process (AOP) in a batch reactor emitting monochromatic light centered at 253.7 nm to test the degradation of anionic surfactant sodium dodecyl sulfate (SDS) in municipal wastewater. The results showed that only 45 % of SDS had degraded under direct UV; however, almost complete degradation of SDS was observed for UV-H_2O_2.

Biological treatment

Biological wastewater treatment aims to degrade or adsorb dissolved, colloidal, particulate, and settleable materials into biological flocs or biofilms through a series of important processes that have in common the use of microorganisms on water-soluble components. These processes take advantage of the ability of microorganisms to assimilate organic matter and nutrients (nitrogen and phosphorus) dissolved in the wastewater for their growth. When they reproduce, they aggregate and form macroscopic flocs[111].

The design of biological processes is based on creating and exploiting ecological niches that select the most suitable microorganisms to reproduce in such environmental conditions[112].

In most cases, organic matter provides the carbon energy that microorganisms need for growth. However, it is also necessary to rely on the presence of nutrients contained in the essential elements for growth, particularly nitrogen and phosphorus. Oxygen is not always crucial because microorganisms can degrade organic matter under anaerobic conditions. Wastewaters with a high organic matter content favor technology using bacteria under anaerobic conditions.

Biological treatment is particularly common for surfactant treatment in domestic wastewater due to its cost-effectiveness and environmentally friendly effects, while physical and chemical treatment methods are expensive[113]. Surfactant biodegradation occurs when microorganisms use the surfactant either as a source of energy or nutrients, or by co-metabolizing the surfactant in catabolic pathways[113].

To remove anionic surfactants, sodium lauryl sulfate (SLS) and sodium lauryl polyoxyethylene ether sulfate (SLES) in the influent accumulated in the supernatant of an anaerobic membrane bioreactor (AnMBR) designed to hydrolyze organic matter and convert organic nitrogen. To address this problem, micro-aeration, which had rarely been reported to improve surfactant biodegradation in the anaerobic process, was introduced and proved an effective approach that decreased surfactant concentrations in the supernatant from 9000 mg L^{-1} to 2000 mg L^{-1}. After the introduction of micro-aeration, the emerging genera *Aquamicrobium, Flaviflexus*, *Pseudomonas*, and *Thiopseudomonas* in the microbial community could be responsible for the efficient biodegradation of surfactants[114].

In addition, Zhu *et al.*[115] conducted a study on two typical industrial and domestic wastewater treatment plants with different treatment technologies, in particular, the anaerobic-oxic (A/O) treatment process and the cyclic activated sludge technology (CAST) process. The objective of these processes was to study the treatment efficiency of two types of surfactants, linear alkylbenzene sulfonates (LAS) and benzalkonium chlorides (BAC). The biological treatment unit in the A/O treatment process and the cyclic activated sludge system in the CAST treatment process were the main surfactant removal units, with removal efficiencies greater than 83 %. These results showed that surfactants could be strongly degraded under aerobic conditions. However, seasonal changes had no significant influence on the removal efficiency of surfactants.

A new approach was performed by Hena *et al.*[116] to remove surfactants from municipal wastewater by culturing selected microalgae. Out of 76 strains, only 11 were finally able to grow in wastewater containing the detergents, suggesting that wastewater detergents are highly toxic for aquatic organisms, especially microalgae.

In addition, Serejo *et al.*[117] investigated surfactant removal efficiency in three high-rate algal ponds for primary treatment of domestic wastewater. Semi-continuous feeding in the high rate algal ponds operated during daylight was found to be more advantageous than the normal continuous operation regarding both microalgae biomass productivity and surfactant removal efficiency, which reached 97 % removal rate.

Khosravi *et al.*[118] conducted a sequencing batch reactor (SBR) study as a method for purification of textile wastewater loaded with anionic surfactant sodium dodecyl sulfate (SDS) with a concentration of 20 mg L^{-1}. The experimental results of the biological treatment by SBR in two anaerobic-aerobic phases showed an excellent degradation of sodium dodecyl sulfate (SDS) (0.4 mg L^{-1}) with a removal rate of up to 98 %. Also, Kamińska and Marszałek[119] treated greywater using a laboratory SBR reactor with a capacity of 3 L, operating in a 24-h cycle. The treatment gave very good results, with complete removal of nonionic surfactants, and 97 % removal of anionic surfactants from the greywater. Kruszelnicka *et al.*[29] tested the removal efficiency of surfactants in domestic wastewater from an SBR-type treatment plant in Poland (type SBR-K-6). The results showed that anionic surfactants were removed up to 88 %; on the other hand, lower removal efficiency was obtained in the case of nonionic surfactants, which reached 56 %.

Ran *et al.*[120] conducted a study on the evaluation of detergent removal at a pilot-activated sludge treatment plant in a membrane bioreactor with linear alkyl benzene sulfonate (LAS) loaded wastewater. During treatment, the concentration of added LAS was gradually increased from 25 mg L^{-1} to 200 mg L^{-1}. The results showed that the removal rate of LAS was relatively stable and reached 80 %. In addition, when the LAS concentration increased by more than 175 mg L^{-1}, the LAS removal rate decreased significantly.

In their study, Bering *et al.*[121] targeted the treatment of laundry wastewater mainly containing surfactants and impurities from washed fabrics, using a two-stage moving bed bioreactor operating under aerobic conditions. The treatment results showed good surfactant removal efficiency, which was 79–99 % for anionic, 88–99 % for nonionic, and 85–96 % for the sum of anionic and nonionic surfactants. Table 6 shows the removal of different types of surfactants from wastewater by different biological treatments.

Table 6 – *Removal of surfactants from wastewater by biological treatment*

Types of wastewater	Biological treatment	Bacterial cultures	Types of surfactants	Percentage of removal	Processing times	References
Synthetic wastewater	Anaerobic membrane bioreactor (AnMBR)	*Aquamicrobium*, *Flaviflexus*, *Pseudomonas* and *Thiopseudomonas*	Anionic surfactants SLES (sodium lauryl polyoxyethylene ether sulfate) and SLS (sodium lauryl sulfate)	77 %	_	114
Wastewater of Lepenica River (Kragujevac, Serbia)	Biodegradation	*Aspergillus niger*	Anionic surfactants	30 %	16 days	122
Industrial wastewater (textile industry)	Biodegradation	*Aspergillus versicolor*	Cationic surfactant dodecyl trimethylammonium bromide (DTAB)	100 %	24 hours	113
Textile wastewater	SBR	Anaerobic and aerobic bacteria	Anionic surfactant sodium dodecyl sulfate (SDS)	98 %	(8 h anaerobic: 13 h aerobic)	118
Laundry wastewater	Moving bed bio reactor (MBBR)	Not selected	Anionic and nonionic surfactants	85–96 %	_	121
Industrial and domestic wastewater	A/O and CAST treatment processes	Not selected	LAS (linear alkylbenzene sulfonates) BAC (benzalkonium chlorides)	83 %>	_	115

Biological treatment of detergent-laden wastewater remains the best option due to low operating costs and high detergent removal rates. Several parameters influence the biological treatment and biodegradation progress, especially of detergent-laden wastewater, the types, and concentrations of microorganisms, degradation reaction conditions, aeration, surfactant types and availability, surfactant concentration, presence of toxicants, and residence time[123].

New advanced treatments with carbon nanotubes (CNT)

A potential strategy for future surfactant removal could be to promote new techniques that are more suitable for surfactant processing.

Carbon nanotubes (CNTs) have been researched for various applications. In the context of detergent-rich wastewater treatment, CNTs could be used as a nanomaterial for adsorption-based treatment processes as an alternative to traditional materials, such as activated carbon (AC), zeolite, and kaolin[124,125]. The results demonstrated that CNTs differed from other carbonaceous materials in their adsorption characteristics. For example, perfluorooctane sulfonate (PFOS) had a maximum sorption capacity of 656 mg g^{-1} on CNTs compared to 196.2 mg g^{-1} for granular activated carbon[126]. Indeed, CNT should be a perfect sorbent to remove surfactants.

This technology has received much attention for its use in detergent-rich wastewater treatment due to its large adsorption surface area and ability to adsorb a range of difficult-to-remove pollutants, and is of interest to the detergent-rich wastewater treatment industry.

Conclusion

This review has clarified, in general, the impacts of detergents on the environment, health, and operation of wastewater treatment plants. The methods of elimination by different physicochemical techniques (coagulation-flocculation, adsorption, membrane treatment) or biological, and sustainable and environmentally friendly treatment methods have been reviewed. More research is needed in the field, including the possibility of merging one or more techniques to obtain better results. However, efforts must be made to mitigate the environmental impacts of detergents, and move more towards developing and using green surfactants that are readily biodegradable and do not present environmental and health risks. This review has revealed a dynamic and challenging area that could benefit from greater attention from the research community to control detergent-laden wastewater pollution.

Nomenclature

EO	–	ethylene oxide
WWTPs	–	wastewater treatment plants
QACs	–	quaternary ammonium compounds
BPC-g-PAM	–	bamboo pulp cellulose-g-polyacrylamide
LAS	–	linear alkylbenzene sulfonate
PAC	–	poly aluminum chloride
DAF	–	dissolved air flotation
AC	–	activated carbon
SDS	–	sodium dodecyl sulfate
SDBS	–	sodium dodecylbenzene sulfonate
GAC	–	granular activated carbon
LABS	–	linear alkylbenzene sulfonate
CTAB	–	cetyltrimethylammonium bromide
COD	–	chemical oxygen demand
MBR	–	membrane bioreactor
UF	–	ultrafiltration
RO	–	reverse osmosis
FSM	–	flaxseed mucilage
DBS	–	dodecylbenzenesulfonate
AOP	–	advanced oxidation process
SLS	–	sodium lauryl sulfate
SLES	–	sodium lauryl polyoxyethylene ether sulfate
AnMBR	–	anaerobic membrane bioreactor
CAST	–	cyclic activated sludge technology
LAS	–	linear alkylbenzene sulfonates
BAC	–	benzalkonium chlorides
SBR	–	sequencing batch reactor
CNT	–	carbon nanotubes
EC50	–	concentration that induces 50 % of the substance maximum effect
LC50	–	concentration required to kill half the members of a tested population
PFOS	–	perfluorooctane sulfonate
AEO	–	alcohol ethoxylate
NP	–	nonylphenol

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