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Chapter 10

Electrocoagulative and Biological Treatment of Laundry Wastewater

Terelle Ramcharan and Ajay Bissessur

Additional information is available at the end of the chapter

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Abstract

The greater demand for potable water, both locally and worldwide, has directed a huge interest amongst researchers to investigate the possibility of recycling and reusing wastewater from laundry run-offs. The advantage of using recycling wastewater from such sources is mainly due to the fact that these bulk volumes of wastewater are considered to be less chemically polluted in comparison to those discarded from industrial effluents and wastewater sources. Almost all laundry detergents contain surfactants, whose main function serves to remove dirt/soil from contaminated items. Thus, an analysis of the surfactant levels before and after a treatment process is important to confirm that the surfactant has in fact carried out its intended purpose. Electrocoagulative treatment of wastewater, a well-researched and well-documented clean-up process that involves the production of aluminium hydroxy species by oxidation of aluminium metal upon the application of a controlled voltage which adsorbs fine particulate matter and pollutants from the wastewater has been investigated as a clean-up application to the treatment of laundry wastewater. The use of a biological treatment process which entails treating the wastewater with aerobic bacterial specie specifically designed to degrade fats, lipids, protein, detergents and hydrocarbons has also been investigated.

Keywords: biological, biospinners, electrocoagulation, laundry wastewater, linear alkylbenzene sulfonates

1. Introduction

The composition of laundry detergents is generally complex due to the numerous factors that have to be taken into consideration to ensure fresh clean garments at the end of the wash process. Sodium dodecylbenzene sulfonate, more commonly known as SDS or linear alkylbenzene sulfonates (LAS),

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is the most abundant anionic surfactant utilised in laundry detergents due to its excellent performance in removing water insoluble substances such as greasy and oily stains. As a commercial commodity, LAS is sold as a sodium salt which contains a mixture of homologues that has between 10 and 14 linear carbon atoms with a phenyl group attached to the linear alkyl chain and the sulfonate anion as shown in **Figure 1** [1–6].

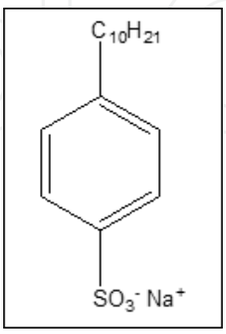


Figure 1. Chemical structure of sodium dodecylbenzene sulfonate (SDS).

The rapid biodegradation of LAS compounds especially under aerobic conditions consumes a large amount of bio-available oxygen that significantly increases the chemical oxygen demand, thus negative impacting on the environment and organisms persisting within that system [4, 5]. Oxidation of LAS by oxygen results in the formation of sulfophenylcarboxylic acid (SPC) that comprises one of the main products of biodegradation [7–9].

2. Quantification of LAS by Ultraviolet-Visible spectrophotometry

Ultraviolet-Visible (UV-Vis) spectrophotometry is one of the commonly used techniques for the quantification of surfactants, whereby the method of determination of anionic surfactants entails the use of a cationic dye that complexes with the anionic surfactant through the mechanism of ion association as shown in **Figure 2** [10, 11].

Valuable structural information by mass spectrometric (MS) detection often allows for the qualitative analysis of surfactants [12]. Analysis of ethoxylated surfactants using soft ionisation techniques such as electrospray ionisation (ESI) and atmospheric pressure chemical ionisation (APCI) determines analytes in a cationized molecular state [13]. The use of mass spectrometry and addition of a volatile reagent like ammonium salt, for example, ammonium acetate that suppresses the formation of alkali salts improves the accuracy of LAS determinations. The determination of non-ionic surfactants is possible via the application of positive or negative ionization modes for ESI and APCI, with the best response obtained using the positive ion mode [13].

Liquid chromatography-mass spectroscopy (LC-MS) is a powerful analytical technique, that is an applied qualitative detection method for non-ionic surfactants as reported by many researchers [12, 14–17]. In addition, a direct application of gas chromatography-mass spectroscopy (GC-MS) is used in the analysis of non-ionic surfactants; however, this method is limited in its

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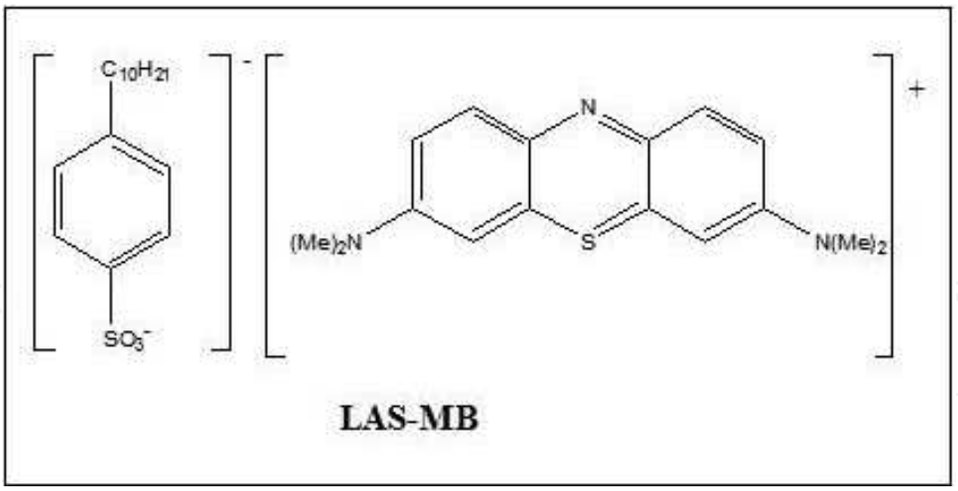


Figure 2. Ion association complex formed between LAS and methylene [10].

application due to the derivatization requirement for long ethoxy chain containing surfactants [17]. The use of solid-phase extraction (SPE) and GC-MS for the direct analysis of APEs is carried out, whereby a graphitized carbon black SPE cartridge and use of methanol/dichloromethane solvent system was implemented [18]. The use of ethyl violet and acridine orange dyes has been reported by researchers for extraction of anionic surfactants [19, 20]. Specifically, toluene and benzene solvents have been used for extraction of LAS complexes, which is deemed less toxic than chloroform, and have therefore been reported as a recommended replacement to the methylene blue method [19, 20]. High-performance liquid chromatography (HPLC) is a commonly applied technique for LAS determination and detection which includes ultraviolet (UV), fluorescence (FL), diode-array detection (DAD) and mass spectroscopy (MS).

Another method for the analysis of LAS by an ion-pair SPE technique and HPLC has been developed [21]. Extraction of LAS using C_8, C_{18} and multiwall carbon-nanotubes was investigated and samples were quantified by reversed-phase HPLC using a C_8 column and UV detection with isocratic elution at a retention time of 15 min using a methanol/water mobile phase containing 5 mM sodium acetate [21]. Quantification of LAS in environmental samples by HPLC-FL has been developed which entails Soxhlet extraction of the sample with gradient elution, retention time of 22 min and application of mobile phases, which include acetonitrile, water, triethylamine and acetic acid [22, 23]. Quantification of LAS in sewage sludge samples using HPLC-FL with a C_8 column with microwave-assisted extraction is used for sample preparation. A comparison of separation of LAS using HPLC-FL and HPLC-DAD showed no significant difference between the two sets of results and that usage of either a FL or DAD detector are applicable [24]. HPLC-MS is considered the most accurate method for determination of LAS as it permits for both a qualitative and quantitative analysis of LAS [3, 5]. GC-MS is less often used for analysis of LAS, as this method would require derivatization of LAS into a volatile compound [25]. Quantification of anionic surfactants and inorganic constituents' viz., phosphates, silicates and zeolite, has been analysed by Inductively coupled plasma-optical emission spectroscopy (ICP-OES) [26]. Specifically, alkylbenzene sulfonates and alkyl sulphates were determined due to their ability to precipitate upon addition of calcium ions [26]. Non-ionic surfactants that are used widely in domestic and

industrial detergents [27] are represented by two major classes, which include alcohol ethoxylates (AE) and alkylphenol ethoxylates (APEOs) [28]. The most common non-ionic surfactants used in detergents are octylphenol ethoxylate (OPEO) and nonylphenol ethoxylate (NPEO) as shown in **Figure 3** [29, 30].

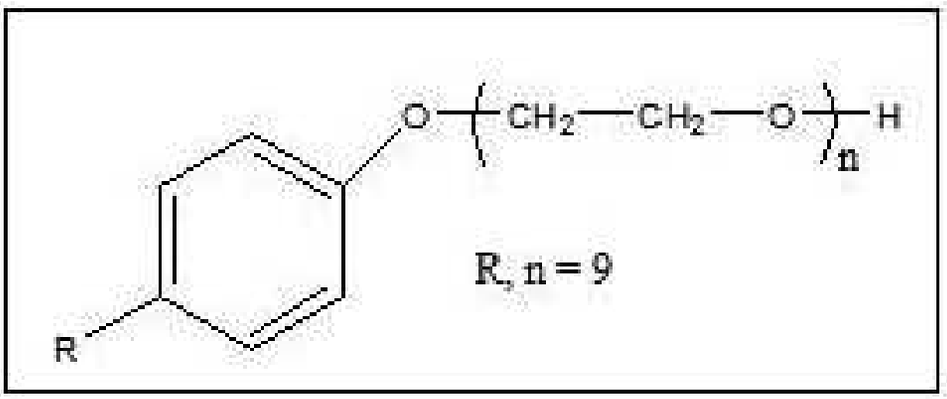


Figure 3. Structure of nonylphenol ethoxylate [31, 32].

The toxicity of the surfactant is dependent on the length of the ethoxy chain. A more toxic behaviour is known to be displayed by APEOs with a shorter ethoxy chain (typically <4) when compared to longer ethoxy chain length APEOs (typically >10) [18]. APEOs can be degraded under both anaerobic and aerobic conditions, thus leading to the biotransformation of APEO into lipophilic metabolites of APEO [33]. The most common degradation products of APEOs include nonylphenol, octylphenol and mono- and diethoxylated compounds of NPEO and OPEO [33, 34], which are deemed toxic and have been found to be persistent in the environment, thus causing endocrine disrupting effects amongst aquatic organisms [34–36]. Other contributing important ingredients found in laundry detergents include builders and anti-foaming agents. A common zeolite-based builder, sodium aluminium silicate, is often used as a builder in laundry detergents to reduce water hardness, while polydimethylsiloxane acts as an anti-foaming reagent.

3. Application of biological and electrocoagulative treatment methods to laundry wastewater

The separation of the solid matrices from the liquid matrices forms the basis for treatment of wastewater, which is most commonly achieved through coagulation-flotation methods [37]. During coagulative processes, an alteration of the surface properties of the individual particles occurs and this permits transformation into larger particles [38]. Inorganic salts of aluminium, iron or calcium are commonly used in coagulation processes [39]. In the coagulation process, small particles may form which decrease the efficiency in removal of pollutants from the wastewater streams and for this reason, flocculent agents are commonly used in conjunction with coagulation agents [40]. The efficiency of coagulation is enhanced by an increase in flocculation through accumulation of particles into larger settleable masses [38]. Polymer-based flocculants are commonly used for this purpose as a result of their large surface area,

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hence enabling the particles to group and settle, thus facilitating easy removal of pollutants from the wastewater.

Biological treatments have been mainly applied to the treatment of industrial effluent wastewater. The advantages associated with biological treatment of wastewater include a decreased amount of toxic and harmful chemicals coupled with an easy to implement green process [41]. Waste from effluents is recycled into an organism-based biomass through biological treatment, and can be easily disposed of naturally into the environment [41]. Major disadvantages associated with biological treatment of wastewater include:

a. large space requirement for the storage of biological waste,

b. longer time periods required for effluent treatment in comparison to chemical treatment,

c. limitation in its application to a wide range of effluents [41].

In the application of biological treatment of wastewater, addition of a specific strain of bacteria to the wastewater is the main thrust of the system that subsequently targets specific oxidation and degradation of pollutants.

Biological wastewater treatment is often seen as an environmental friendly method, as there are generally no added chemicals involved. Some of the major concerns with regard to biological treatment of wastewater include the longer time periods for treatment, a larger surface area required and the addition of specialised bacteria for the specific degradation of pollutants. Chan demonstrated a method for treatment of laundry effluent through a combination of biological and chemical treatment methods [41].

The laundry effluent was treated biologically prior chemical treatment. This treatment method permitted the production of high-quality water that could be used for activities such as flushing and cleaning which reduced the consumption of water by the launderette. The quality of the water was assessed by measuring the following parameters: pH, DO, SS, COD and total surfactant concentration.

Electrocoagulation is often implemented as the primary treatment for wastewater due to its efficient pollutant removal as well as its safe and environmental friendly nature. Electrocoagulation involves the dissolution of sacrificial anodes due to the application of electric current. Aluminium and iron are the most generic anodes used for this purpose.

$$\mathrm{Al(s)} \rightarrow \mathrm{Al^{3+}(aq)} + 3\mathrm{e^-} \tag{1}$$

$$3\mathrm{H_2O}\ (\ell)\ +\ 3\mathrm{e^-} \rightarrow\ \mathrm{H_2(g)} + 3\mathrm{OH^-(aq)} \tag{2}$$

Eqs. (1) and (2) represent the reactions taking place at the anode and cathode, respectively. The resultant metal ion reacts with hydroxide in the wastewater to form various metal hydroxides.

$$\mathrm{Al^{3+}(aq)} + 3\mathrm{H_2O}\ (\ell)\ (\mathrm{aq})\ \rightarrow\ \mathrm{Al(OH)_3} + 3\mathrm{H^+(aq)} \tag{3}$$

$$\mathrm{Al(OH)_3(aq)} + \mathrm{OH^-(aq)}\ \rightarrow\ \mathrm{Al(OH)_4{}^-(aq)} \tag{4}$$

Eqs. (3) and (4) represent the generation of aluminium hydroxy species during electrocoagulation.

Treatment of wastewater by electrocoagulation is known to effectively remove heavy metals, minerals and dyes from wastewater streams, hence making it a good treatment method for laundry wastewater. A high removal efficiency of organic compounds is obtained due to the various mechanisms that occur in the electrocoagulation cell. The pollutants adsorb onto the different aluminium hydroxy species depending on the chemical structure of the pollutant.

The hydrogen gas produced at the cathode induces flotation of the hydroxy species, hence allowing for a quick and efficient removal of pollutants from the wastewater. Aside from the production of aluminium hydroxy species, other mechanisms in the electrocoagulation cell occur which increases the efficient removal of pollutants from the wastewater stream. Reactions at the surface of the cathode also remove carbonate salts, which is abundant in laundry wastewater.

$$HCO^{3-}(aq) + OH^{-}(aq) \rightarrow CO_3^{2-}(aq) + H_2O\ (\ell) \tag{5}$$

$$Ca^{2+}(aq) + CO_3^{2-}(aq) \rightarrow CaCO_3(s) \tag{6}$$

$$Mg^{2+}(aq) + CO_3^{2-}(aq) \rightarrow MgCO_3(s) \tag{7}$$

Eqs. (5)–(7) represent the removal of carbonate from the wastewater as salts of calcium and magnesium. Laundry wastewater is also known to contain chloride salts. Electrolysis generates molecular chlorine, which can lead to the formation of hypochlorous acid and hypochlorite ions as shown in Eqs. (8)–(10). These species contain a relatively high oxidative potential, which allows for further degradation of organic pollutants in the wastewater stream.

$$2Cl^{-}(aq) \rightarrow Cl_2(g) + 2e^{-} \tag{8}$$

$$Cl_2(g) + H_2O \rightarrow HOCl\ (aq) + H^{+}(aq) + Cl^{-}(aq) \tag{9}$$

$$HClO(aq) \rightarrow ClO^{-}(aq) + H^{+}(aq) \tag{10}$$

In research presented by many scientists, electrocoagulation is described as the treatment of laundry effluent [42–44]. Iron and aluminium electrodes are used for electrocoagulation; however, aluminium electrodes had a greater efficiency in removal of pollutants from the laundry wastewater. Some investigations applied an ultrasonic bath during electrocoagulation which had a profound effect on the efficiency of the removal [42].

Over time, the formation of an inhibiting film due to high voltages applied to the electrodes impacts negatively on the efficiency of electrocoagulation. The measured parameters of phosphorous levels, detergent, COD, turbidity and conductivity in the laundry wastewater before and after the process of electrocoagulation are good indicators of the effectiveness of the electrocoagulative process [42–44].

Electrocoagulation using aluminium electrodes, as shown in **Figure 4**, has been applied as a method for treatment of wastewater obtained from a textile industry aimed at the removal of dye substances from wastewater [45, 46]. This method has accounted for a 99% efficiency in removal of the dye substances, measured by determination of the COD before and after treatment [45, 46]. The removal of heavy metals such as nickel, copper, zinc and chromium from synthetic and industrial wastewater by electrocoagulation using aluminium electrodes

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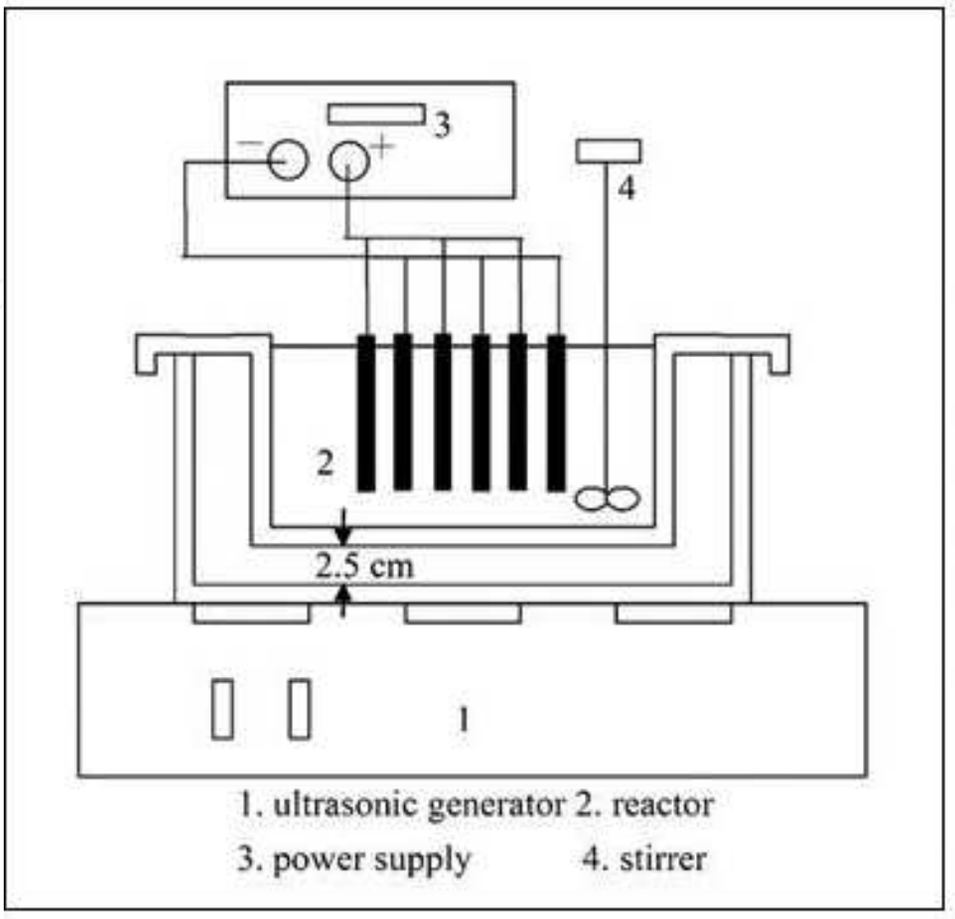


Figure 4. Illustration of an electrocoagulation cell adapted from Wang et al. [42].

has been widely applied. An added advantage of electrocoagulation in addition to removal of heavy metals from wastewater stream also significantly decreased the COD [47]. In a research study by Ramcharan and Bissessur, a comparison of electrocoagulation and biological treatment of Laundry Wastewater (LWW) was reported [48]. The surfactant concentration, chemical oxygen demand and total dissolved solids were the general water guideline parameters used to assess the success of the treatment system. The wastewater was characterised after each wash and rinse cycle discharged from a domestic washing machine and are referred to as first wash cycle wastewater (W1), first rinse cycle wastewater (R1) and second rinse cycle wastewater (R2). The two major parameters, which influenced the above treatment methods, were the period allocated for treatment and the suitability of each treatment method to a variety of wastewater matrixes. The successful treatment of R1 and R2 was obtained using the biological method, while electrocoagulation was successful for W1, R1 and R2 (**Figure 5**). The sample matrix of W1 was not compatible for biological treatment, as the bacterium was not able to cultivate under such harsh conditions. Aeration of W1 proved to decrease the concentration of the surfactant because SDS is susceptible to degradation under oxidative conditions.

Degradation of the bacteria is imminent upon exposure to the strongly basic pH of the first wash laundry wastewater, which increased the organic content thereby increasing the COD in laundry wastewater from the first wash during biological treatment.

The dominance of the electrocoagulative treatment method over the biological method of LWW is further supported by the COD levels attained as shown in **Figure 5**. It is clearly evident that upon treatment of W1, a gradual increase in the COD levels occurs over a prolonged period of time. The highly alkaline nature of the wastewater induces breakdown of bacterial cells, thus implementing an increase in the organic content and thereby consequently

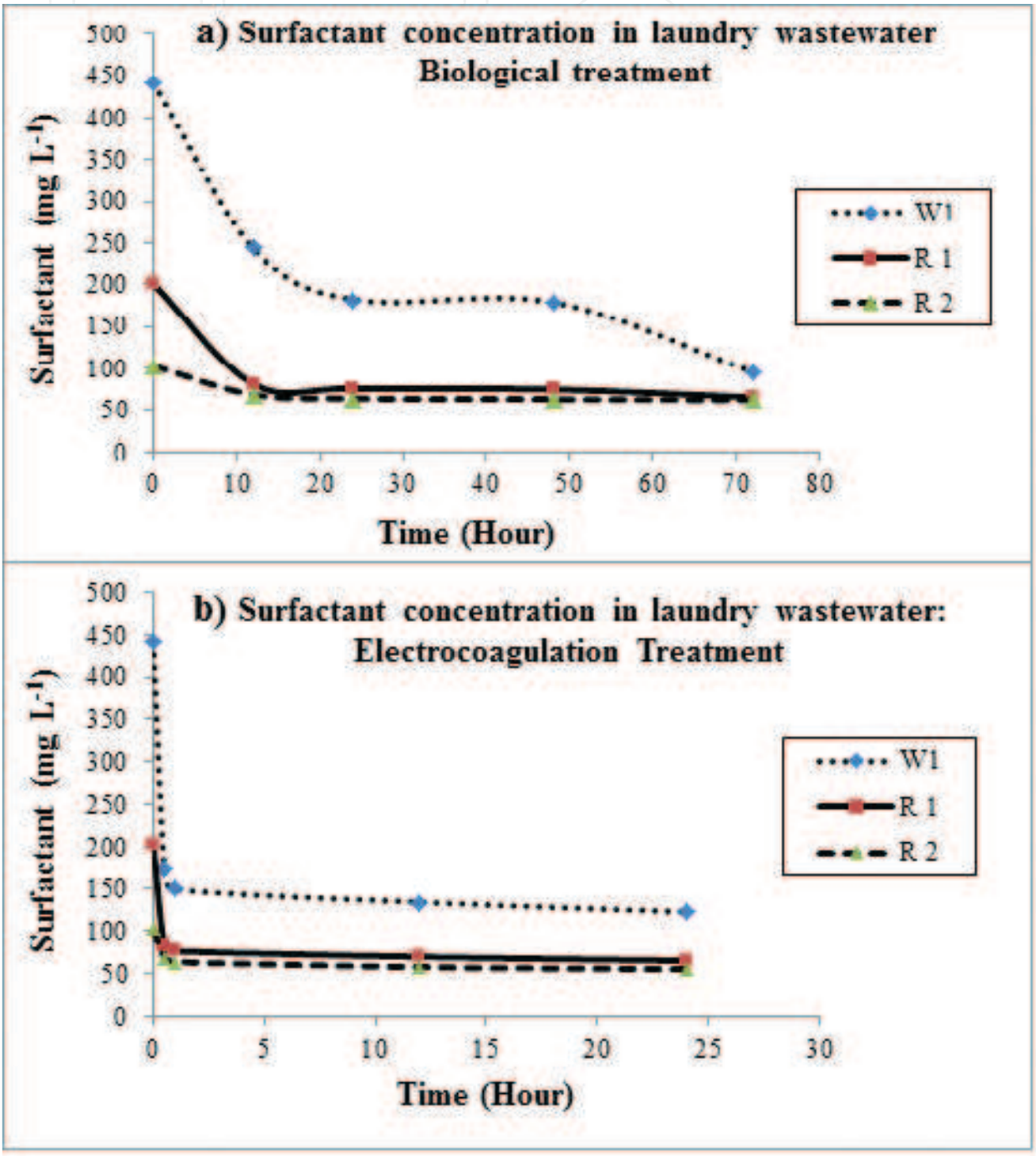


Figure 5. Decrease in surfactant concentration after application of (*a*) *biological treatment* and (b) *electrocoagulative treatments* to laundry wastewater from the first wash (W1), first rinse stage (R1) and second rinse stage (R2). Reproduced from Ref. [48].

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causing an increase in the COD level of W1. However, a marked decrease in COD level occurred during the implementation of the electrocoagulative technique as shown in **Figure 5**. Finally, the persistence of LAS in solution is directly linked to the COD level. The effective removal of LAS by the electrocoagulative treatment caused a marked decrease in the organic content present; thus, a rapid decrease in the COD is observed especially for R2 in the initial onset (within the 5 minutes of implementation) of electrocoagulation as shown in **Figure 6**.

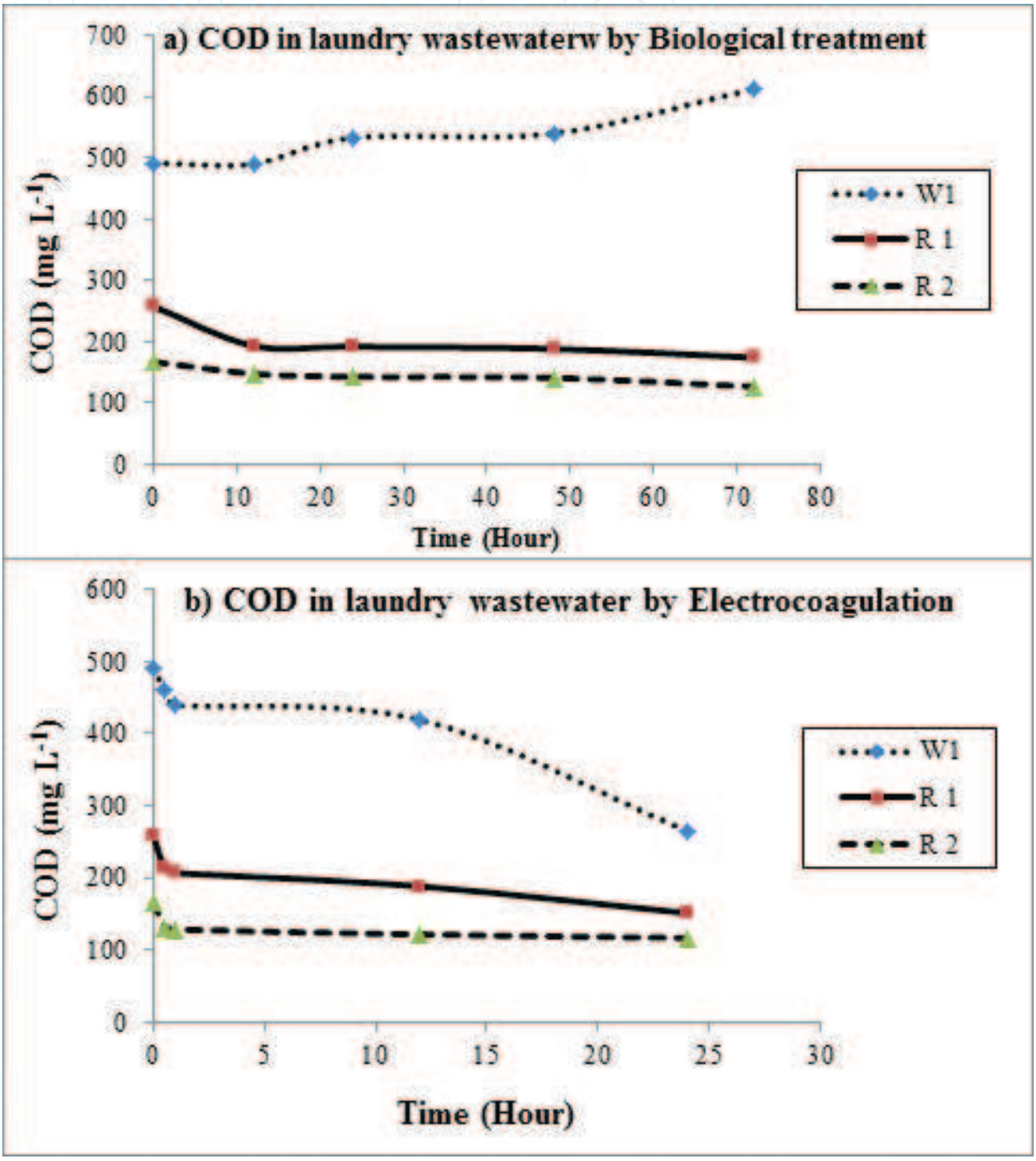


Figure 6. COD Levels of laundry wastewater samples after first wash (W1), first rinse (R1) and second rinse (R2) cycles when subjected to (a) *biological treatment* and (b) *electrocoagulation*. Reproduced from Ref. [48].

TDS levels at the different wash and rinse cycles of LWW showed an increasing trend when treated biologically, while the electrocoagulation method of treatment for LWW showed a decrease in the TDS levels as shown in **Figure 7**. This is chiefly due to the quick polymeric generation of aluminium hydroxide species during electrocoagulation had allowed for adsorption of SDS in LWW whilst promoting effective TDS removal through settlement of the polymeric floc generated.

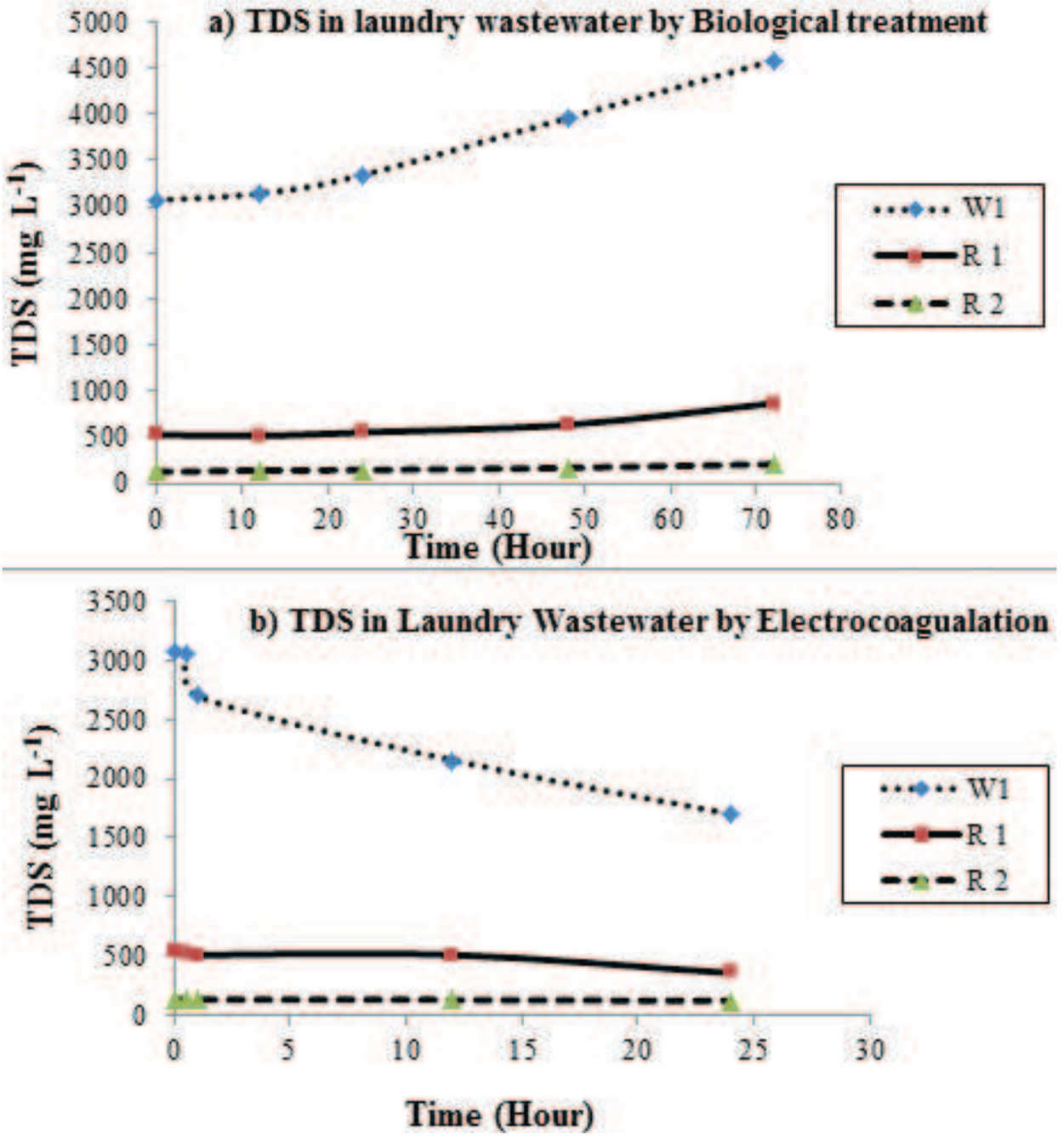


Figure 7. TDS levels in laundry wastewater for first wash (W1), first rinse (R1) and second rinse samples (R2) after (a) *biological treatment* and (b) *electrocoagulation*. Reproduced from Ref. [48].

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Supporting kinetic data is pivotal when implementing pilot wastewater treatment systems. The adsorption kinetics is one of the important parameters used to assess sustainability of the treatment system. A kinetic study on the adsorption capacity of the aluminium hydroxy species was investigated by Ramcharan and Bissessur [48]. The Ho pseudo second-order expression was used to evaluate the adsorption capacity for surfactant removal in laundry wastewater from the first wash, first rinse cycle and second rinse cycle as shown in Eq. (11) below. A second-order reaction was observed from the plot of t/q_t vs. t shown in **Figure 8** with R^2 values >0.99.

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \tag{11}$$

The percentage of efficiency of adsorption (% E) was based on calculations using Eq. (13) below, where C_0 and C corresponds to the initial and specific concentration of the surfactant at time t. The values of the adsorption efficiency at equilibrium (q_e) and rate of adsorption (k_2) was based on calculations using Eqs. (13) and (14), respectively. The rate of adsorption of the surfactants is significantly lower for laundry wastewater discharged from the first wash as compared to laundry wastewater from the first and second rinses as shown in **Table 1**. It can be easily inferred that a reduced amount time is required for the treatment of laundry wastewater disposed after the first and second rinses.

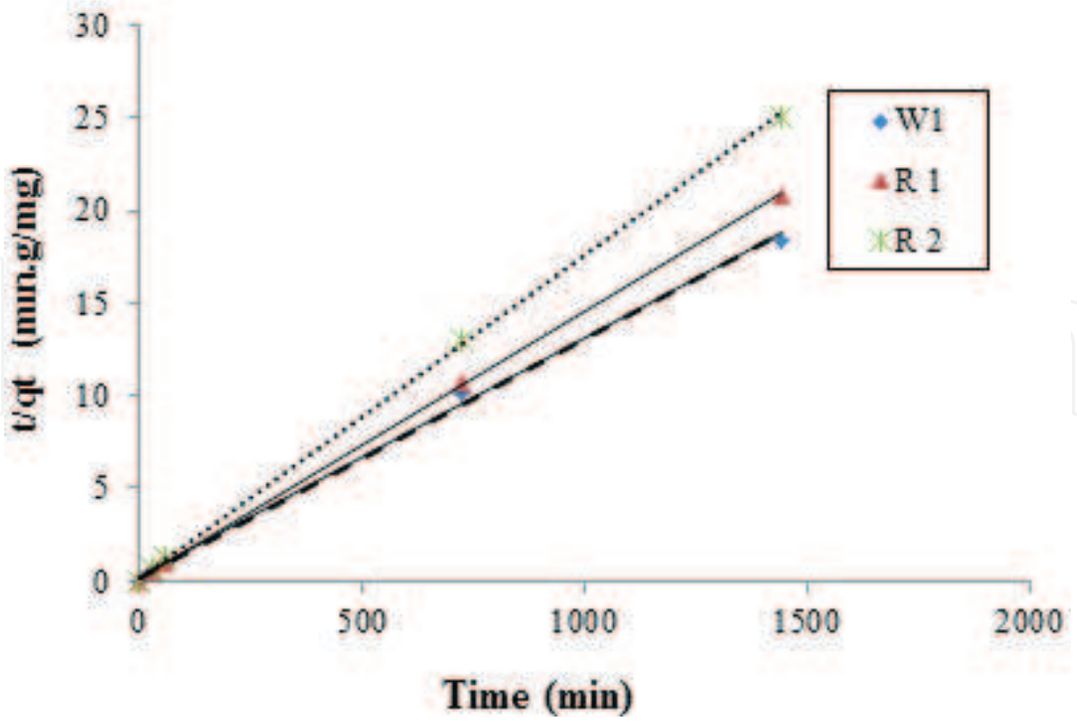


Figure 8. A Plot of t/q_t vs. t showing second-order reaction kinetics for the adsorption capacity of surfactant by aluminium hydroxy species. Reproduced from Ref. [48].

Lagergren parameter	W1	R1	R2
Experimental q_e	77.60	67.27	60.69
Calculated q_e	77.52	68.97	57.47
R^2	0.997	0.999	0.999
k_2	8.53×10^{-4}	2.53×10^{-3}	2.21×10^{-3}

Reproduced from Ref. [48].

Table 1. The Lagergren parameters for adsorption of surfactants by aluminium hydroxy species.

$$\% E = \frac{C_0 - C}{C_0} \times 100 \tag{12}$$

$$q_e = \frac{1}{\text{Slope}} \tag{13}$$

$$k_2 = \frac{\text{Slope}^2}{\text{Intercept}} \tag{14}$$

4. Conclusions

The application of electrocoagulative and biological treatment methods effectively decreased the amount of surfactant concentration in laundry wastewater after all rinsing stages. In comparison, the electrocoagulative technique was found to be a more efficient treatment method of the two due to its ability to reduce the levels of the surfactant, COD and TDS over a considerably shorter period of time and its ability to be applied to a wider range of wastewater samples. A modification to the electrocoagulation treatment process whereby the addition of Biospinners® was carried out and was found to further reduce the levels of the surfactant, COD and TDS within the same applied period of time. Modification due to addition of Biospinners was shown to increase aeration and surface area, and facilitated the removal of an overlaying film of aluminium hydroxy species formed on the electrodes. The adsorption of LAS by aluminium hydroxy species was found to take place at a lower rate for W1, in comparison to R1 and R2 as shown by the kinetics in this study. From this, it is evident that there is a need for isolated treatments of laundry wastewater W1, R1 and R2, thus ensuring a reduced period of treatment and also ensuring the total output cost of the treatment method is kept to a minimum.

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