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Biodegradation and fate of linear alkylbenzene sulfonate in integrated fixed-film activated sludge using synthetic media

Hadi Eslami[a], Mohammad Reza Samaei[b], Ebrahim Shahsavani[c], Ali Asghar Ebrahimi[a,*]

[a]*Environmental Science and Technology Research Center, Department of Environmental Health Engineering, Shahid Sadoughi University of Medical Sciences, Yazd, Iran, Tel. +98 35 38209100-14; Fax: +98 35 38209111; emails: ebrahimi20007@gmail.com, ebrahimi20007@ssu.ac.ir (A.A. Ebrahimi), hadieslami1986@yahoo.com (H. Eslami)*
[b]*Department of Environmental Health Engineering, School of Health, Shiraz University of Medical Sciences, Shiraz, Iran, email: mrsamaei@sums.ac.ir*
[c]*Research Center for Social Determinats of Health, Jahrom University of Medical Sciences, Jahrom, Iran, email: eshahsavani@yahoo.com*

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ABSTRACT

Linear alkylbenzene sulphonate (LAS) is widely used for household and industrial purposes while influencing negatively on the environment. Present paper aimed to study LAS biodegradation among different loading rates and fate of LAS in integrated fixed-film activated sludge (IFAS) using synthetic media. A synthetic wastewater among three LAS loading rates with LAS concentrations of 5, 12 and 20 mg/L was investigated within an operative period of 111 d. In doing so, a kinetic model was developed to explain the biodegradation rate of LAS. Finally, the obtained data were analyzed by analysis of variance statistical test. The mean removal efficiency of LAS among three LAS loading rates were 92.32% ± 2.81%, 95.55% ± 2.74% and 96.22% ± 2.74%, respectively. Nevertheless, in terms of total removal efficiency of LAS, the contributions of LAS biosorption in sludge among the three LAS loading rates were 21.3%, 34.2% and 48.5%. The mean removal efficiency of chemical oxygen demand (COD) in among three LAS loading rates were 92.17% ± 4.32%, 91.53% ± 3.34% and 90.91% ± 2.98%, respectively. Moreover, the higher LAS loading rate, the higher removal efficiency of LAS ($p \leq 0.001$) and the lower COD removal efficiency ($p \leq 0.001$). The results of Michaelis–Menten model for biodegradation kinetics showed that the LAS biodegradation follows the first-order reaction kinetics ($R^2 = 0.9949$). In addition, biodegradation kinetic and removal efficiency of LAS showed that following the increased concentration of LAS among different loading rates, the LAS biodegradation rate was increased. Therefore, IFAS system is argued to be applicable for wastewater treatment in low and high concentrations of LAS up to 20 mg/L.

Keywords: Biodegradation; Linear alkylbenzene sulphonate; Integrated fixed-film activated sludge; Synthetic media; Kinetic model

1. Introduction

Surfactants compose a diverse group of chemical compounds that are widely used for domestic and industrial cleaning, emulsification as well as wetting agents [1]. Surfactants can be classified into such groups as anionic, cationic, non-ionic, and amphoteric [2–4]. Anionic surfactants are used as additives in foodstuffs, cosmetics, pharmaceutical products, cleaning products, detergents and foams in any form [5]. Among anionic surfactants, linear alkylbenzene sulphonate (LAS) is widely used for domestic and industrial purposes due to its inexpensiveness and high cleansing properties [6–8]. LAS molecule contains a linear alkyl chain with 10–14 carbon atoms that are bonded to a sulfonated aromatic ring [9–13] (Fig. 1).

* Corresponding author.

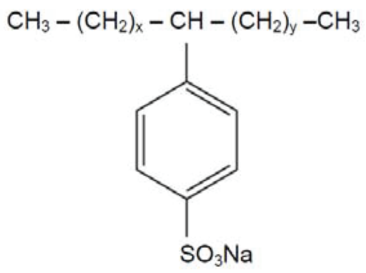

Fig. 1. A structure of LAS, where *x* and *y* are related to the number of CH_2.

LAS has been extensively used for over 40 years with an estimated global consumption of 18.2 million tons/year [14,15]. LAS is a cleaning agent widely used in clothes washing with a concentration up to 25% and 30% in consumable and commercial products, respectively. However, over 80% of LAS usage is seen in household detergents as a component of liquid cleaning products. After being used, LAS is discarded into aquatic ecosystems as it is discharged from both treated and untreated wastewater [16,17]. An average LAS concentration of 1–18 mg/L can be found in municipal wastewater [14]. Attention has been paid to LAS since it influences aquatic/terrestrial ecosystems and is used in large quantities of consumable products [18]. Entry of LAS in aquatic ecosystems may lead to foaming in water resources and consequently result in spread of contaminants [19]. LAS can be harmful for living organisms due to surface-active abilities which may disrupt membranes and lead to denaturation of proteins [5]. Researchers report that 0.02–1.0 mg/L LAS can cause adverse effects on aquatic organisms, especially fish gills and blue mussel larva [14]. Negative effects of LAS on the aquatic ecosystems and water pollution by such surfactants caused researchers to consider their treatment and removal from aquatic environments [20].

LAS degradation and treatment are studied in diverse processes such as chemical coagulation and electrocoagulation, photo- and chemical oxidation, photocatalytic and sonochemical degradation, adsorption and biological and biodegradation [21–27]. Among these LAS treatment methods, biological processes are more widely implemented due to high performance, low cost and environmentally friendly [20,28]. Accordingly, among the biological processes, the integrated fixed-film activated sludge (IFAS) brings up the advantages of both the activated sludge and attached growth bioreactors. This system has higher resistance compared with organic and hydraulic load shock and also higher flexibility and purification power compared with the conventional activated sludge (CAS) process [29].

Due to appropriate properties of the IFAS process for contaminant removal, the present study looked for investigation of LAS biodegradation among different loading rates and fate of LAS in IFAS using synthetic media.

2. Materials and methods

2.1. Pilot plant setup

In present work, IFAS reactor with the sedimentation basin was built as the pilot scale. Fig. 2 shows schematic of

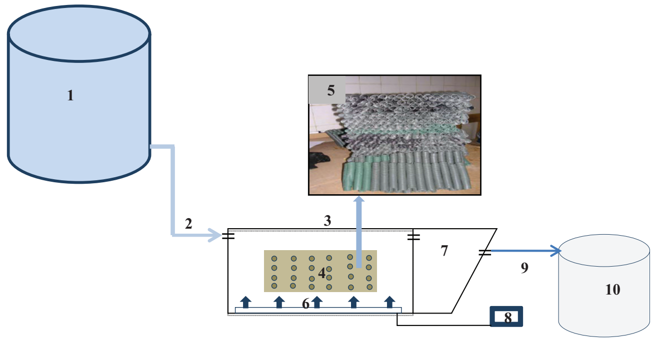

Fig. 2. A schematic of the IFAS system (1. feed tank, 2. inlet of IFAS reactor, 3. aerated reactor, 4. synthetic media, 5. real synthetic media used in IFAS reactor, 6. air inlet, 7. settling area, 8. air pump, 9. effluent of IFAS reactor, 10. effluent storage tank).

Table 1
Design parameters of IFAS reactor and sedimentation basin in pilot scale

Specifications	IFAS reactor	Sedimentation basin
Length (cm)	40	30
Width (cm)	30	30 in surface 10 in bottom
Height (cm)	25	25
Freeboard (cm)	5	5
Volume (L)	27.5	18
Hydraulic retention Time (h)	6	4
Input flow rate (L/d)	110	110

this pilot. In Table 1, the design parameter specifications of the IFAS system are presented. The media used in IFAS was a tube type produced by the waste of electrical mounting sleeves. Specific surface of this media was 154 m^3/m^2 that occupied 25% of the reactor volume.

2.2. Experimental method

The reactor was fed by a synthetic wastewater having similar characteristics to domestic wastewater. The corresponding chemical formula is given in Table 2. Initially, the reactor was launched with a loading rate of 250 mg/L chemical oxygen demand (COD). Then, inlet COD loading rate changed respectively to 300 and 500 mg/L. After a period of 4 weeks, the COD removal efficiency in the reactor was fixed. Then, LAS was added to the inlet wastewater with three loading rates and concentrations of 5, 12 and 20 mg/L for 9 weeks. The total operation time in this study was 111 d. Hydraulic retention times (HRT) in IFAS rector and sedimentation basin were 6 and 4 h, respectively, while, sludge retention time in IFAS system was 10 d. Finally, the removal efficiency was determined by measuring the concentration of LAS in inlet and outlet of the system. In addition, LAS biosorption was measured by mass balance between influent, effluent LAS concentration and measuring of LAS in sludge in loading rates.

The air required for the pilot was supplied by a compressor of 150 m^3 and the system was operated under the conditions of dissolved oxygen in a range of 2–3 mg/L. The temperature was regulated by an aquarium heater that was implanted vertically inside the reactor. Furthermore, 2 samples from inlets and 6 samples from outlets were taken for each parameter in each loading. In this study, the COD and LAS parameters were measured while additional parameters such as dissolved oxygen (DO), temperature (T), air intake and pH were controlled. Finally, 48 samples were taken and measured.

The measurements of LAS was carried out by methylene blue active substances method using spectrophotometer at a wavelength of 652 nm and COD was measured by dichromate method (closed reflux, 5220 B, colorimetric method, Spectrophotometer Milton Roy Company 2OD) according to standard methods for the examination of water and wastewater [30]. Moreover, other parameters such as DO, pH and temperature were measured by YSI portable device.

2.3. Statistical analysis

The results were shown as the mean ± standard deviation (SD) with three replicates, and then the resulted data were imported to the software packages MS Excel 2010 and SPSS V.18.0 and analyzed by one-way analysis of variance (ANOVA) test. In addition, the value of $p < 0.05$ was considered as the significance level.

3. Results and discussion

3.1. LAS and COD removal

The results of this study showed that at LAS loading rates of 5, 12 and 20 mg/L, the mean concentrations of LAS in the effluent were 0.181 ± 0.069, 0.417 ± 0.214 and 0.703 ± 0.508, respectively (Table 3), and the mean removal efficiency of LAS in three LAS loading were 92.32 ± 2.81%, 95.55 ± 2.74% and 96.22 ± 2.74%, respectively (Fig. 3(a)). Plenty of studies on biodegradation of LAS show that the surfactant is well biodegradable under a wide variety of aerobic conditions [7].

Table 2
Synthetic wastewater formulation with the amount of each chemical substance

Synthetic wastewater formulation	COD 250 mg/L	COD 300 mg/L	COD 500 mg/L
	Chemical substance g/100 mL	Chemical substance g/100 mL	Chemical substance g/100 mL
$C_6H_{12}O_6$	30	36	60
CH_3COONH_4	18	18	18
$NaHCO_3$	5	5	5
$FeSO_4.7H_2O$	0.05	0.05	0.05
$NiSO_4.7H_2O$	0.05	0.05	0.05
$FeCl_3.6H_2O$	0.005	0.005	0.005
$Na_2HPO_4.7H_2O$	3.3	3.3	3.3
K_2HPO_4	2	2	2
KH_2PO_4	0.8	0.8	0.8
$MgSO_4.7H_2O$	0.05	0.05	0.05
$CoCl_2$	0.004	0.004	0.004
Na_2SO_4	0.001	0.001	0.001
$CaCl_2$	0.1	0.1	0.1

Table 3
Mean and standard deviation of studied parameters in different loading of input LAS

Parameters	Input of LAS concentration						p value[a]
	5 mg/L		12 mg/L		20 mg/L		
	Mean	SD	Mean	SD	Mean	SD	
LAS_{out} (mg/L)	0.281	0.069	0.517	0.214	0.703	0.508	≤0.001
COD_{in} (mg/L)	512.38	51.31	529.22	49.73	551.03	28.21	≤0.001
COD_{out} (mg/L)	38.63	4.12	44.97	13.83	49.65	18.09	≤0.001
pH	8.05	0.307	7.74	0.158	7.21	0.196	≤0.001
Temperature (°C)	21.17	0.310	20.85	0.397	20.80	30.82	0.075
DO (mg/L)	2.39	0.471	2.46	0.291	2.49	0.280	0.917
Air intake (mg/L)	7.01	1.45	11.50	1.32	19.10	0.964	≤0.001
Operation time (day)	18		19		23		

[a]One-way ANOVA.

In a study by Khamutian et al. [31], the results showed that the LAS removal efficiencies in CAS within winter and summer were 90.8% and 96.5%, respectively. Mollaei et al. [20] studied moving bed biofilm reactor for removing LAS using synthetic media. Their results showed that in optimum condition (HRT of about 72 h and LAS concentration of 200 mg/L), removal efficiency was 99.2% [20]. LAS biodegradation studies indicate that under aerobic conditions, LAS co-metabolism generates shorter chain homologues and then will be mineralized to CO_2 and H_2O by several microorganisms [14,32,33]. Microorganisms can both utilize surfactants as substrates and carbon resource and also co-metabolize them through the initial reactions involved in catabolic pathways [14]. In addition to the biodegradation by microorganisms, there are additional mechanisms such as biosorption and precipitation by suspended solids in LAS biological treatment process [5,14,17]. In this research, the contributions of LAS biosorption in sludge in the three LAS loading rates of 5, 12 and 20 mg/L were 21.3%, 34.2% and 48.5%, respectively, from total removal efficiency of LAS. Further LAS mass balance is presented in Table 4.

In addition, in this study, as shown in Fig. 3(a), by increasing LAS loading, the removal efficiency of LAS was increased as well. The ANOVA test was showed that such a relationship is statistically significant ($p \leq 0.001$). The studies conducted by other researchers show that with increased concentrations of LAS in influent, biodegradation of LAS is increased, as well [11,34]. As a result, with increased LAS concentration among different loading rates, LAS degrading microorganisms can be used from LAS as an alternative additional carbon resource and adopted in the IFAS reactor. With domination of these microorganisms, LAS biodegradation and removal efficiency were increased [34].

Furthermore, by increasing inlet LAS loading, COD in inlet and outlet was increased and the ANOVA test showed this relationship is statistically significant ($p \leq 0.001$). Calculations showed that, a COD of 2.3 ± 0.3 is created per mg/L of inlet LAS (Table 4).

In this study, the mean removal efficiency of COD among the three LAS loading rates were 92.17% ± 4.32%, 91.53% ± 3.34% and 90.91% ± 2.98%, respectively; while, with increased LAS concentration in influent, COD removal efficiency was decreased (Fig. 3(b)). According to the ANOVA test result, this relationship was statistically significant ($p \leq 0.001$). The high efficiency of COD in this study indicates that IFAS system is an integrated system with attached and suspended growth and has higher efficiency and in fact, a consortium of the microorganisms with attached and suspended growth is able to enhance efficiency of organic materials removal [34,35]. Nevertheless, with increased LAS in high concentration, biodegradation of organic materials and COD removal efficiency were decreased due to adverse effects of LAS on microorganisms population in IFAS reactor [5]. However, LAS degrading microorganisms with increased LAS concentration were dominated and can be used as an alternative carbon resource and thereby have more growth [34].

3.2. pH and DO changes

In Fig. 4, the pH and DO changes during the operation time are shown. As it can be seen, with increased LAS concentration in influent, pH was decreased. It can be argued

Table 4
Mass balance of LAS in IFAS reactor to analyze the fate of LAS

	Mass of LAS (5 mg/L)		Mass of LAS (12 mg/L)		Mass of LAS (20 mg/L)	
	mg	%	mg	%	mg	%
Mass added	137.5	100	330	100	550	100
Mass in Effluent	7.72	5.61	14.21	4.30	19.33	3.51
Mass bio-sorbet	29.28	21.30	112.86	34.2	266.75	48.50
Mass biodegraded	100.49	73.08	202.93	61.49	263.92	47.98

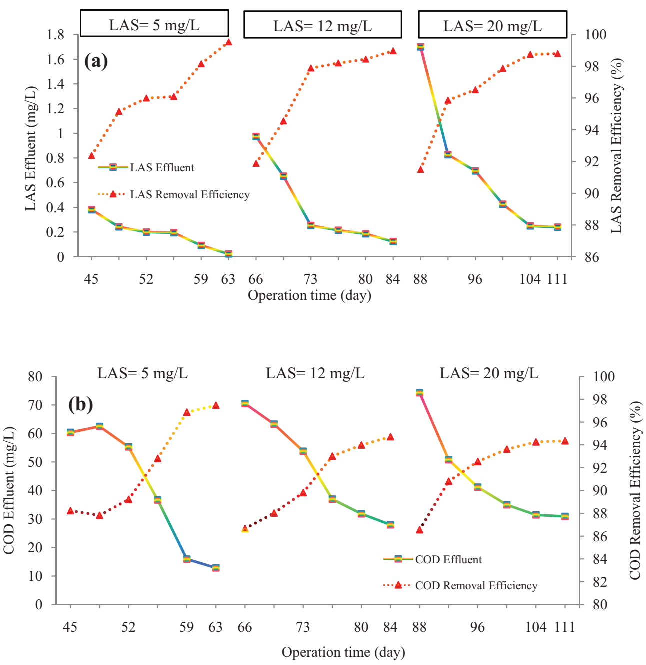

Fig. 3. Effluent and removal efficiency of LAS (a) and COD (b) in different LAS loading rates.

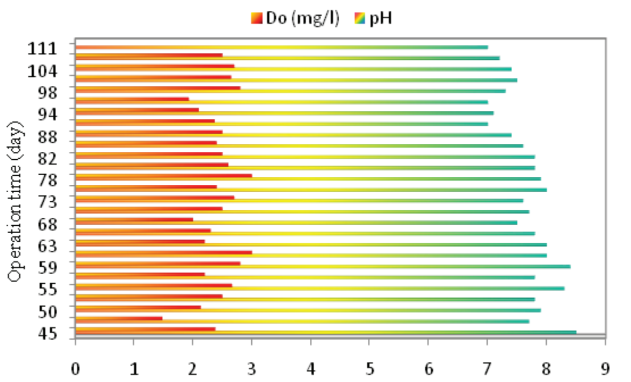

Fig. 4. pH and DO changes in different LAS loading rates and operation time.

that due to the acidic nature of LAS, the pH was decreased along with increased LAS concentration [36,37].

3.3. Air intake and LAS loading relationship

The relationship between air intake and LAS loading rates is shown in Fig. 5. In addition, the results showed that with increased LAS concentration in the influent, air intake rate was increased. Such a relationship was measured by linear regression model. As it can be seen, determination coefficient (R^2) was 0.9784 which indicates a reasonable relationship between air intake and LAS loading rates. According to the calculations, the amount of air intake at a loading rate of 5 mg/L inlet LAS was 1.402 mg/L, while in that of 12 and 20 mg/L inlet LAS, it was 0.95 mg/L. In this study, with an increase of 1 mg/L for LAS, the concentration of COD had an enhancement of 2.3 mg/L.

3.4. Biodegradation kinetics

The biodegradation kinetics was estimated using the Michaelis–Menten (MM) model [11].

$$r_{\text{bio}} = K_{\text{bio}} \frac{C}{K_0 + C} X \quad (1)$$

$$r_{\text{bio}}/X = \frac{S_0 - S}{\theta X} \quad (2)$$

where r_{bio} is the rate of LAS biodegradation (µg/L h), k_{bio} is a biodegradation rate constant (µg/gVSS h), X is the concentration of volatile suspended solids (g/L) and K_C is the LAS concentration at which half of the maximum biodegradation rate is reached (µg/L). Furthermore, r_{bio}/X is the specific biodegradation rate (µg/gVSS h), S_0 is the concentration of LAS in influent, S is the concentration of LAS in an effluent and θ is the hydraulic retention time (h). MM model showed that with increased LAS concentration, the biodegradation rate of LAS increases and LAS biodegradation follows the first-order reaction kinetics and the correlation coefficient R^2 in the model was 0.9949 to predict and the biodegradation rate of LAS in IFAS (Fig. 6). In addition, biodegradation kinetics and removal efficiency of LAS showed that by increasing the concentration of LAS in different loading rates, the LAS biodegradation rate was increased. K_c and k_{bio} in this model were 5,881 µg/L and 6241 µg/gVSS h, respectively [11].

4. Conclusions

The mean removal efficiency of LAS in the loading rates of 5, 12 and 20 mg/L LAS within an operational period of 111 d were 91.32% ± 2.81%, 95.55% ± 2.74% and 96.22% ± 2.74%, respectively, while in terms of total removal efficiency of LAS, the contributions of LAS biosorption in sludge among the three LAS loading rates were 21.3%, 34.2% and 48.5%, respectively. In addition, by increasing LAS loading, the removal efficiency of LAS was increased. The ANOVA test results showed this relationship statistically significant ($p \le 0.001$). The mean removal efficiency of COD among the three LAS loading rates were 92.32% ± 2.81%, 95.55% ± 2.74% and 96.22% ± 2.74%, respectively, while with increased LAS concentration in influent, COD removal efficiency was decreased. According to the ANOVA test, this relationship was statistically significant ($p \le 0.001$). Biodegradation kinetics of LAS follow the first-order reaction kinetics and the correlation coefficient R^2 in the model was 0.9949 to predict and the biodegradation rate of LAS in IFAS. Therefore, the IFAS system is argued to be applicable for wastewater treatment in low and high concentrations of LAS up to 20 mg/L.

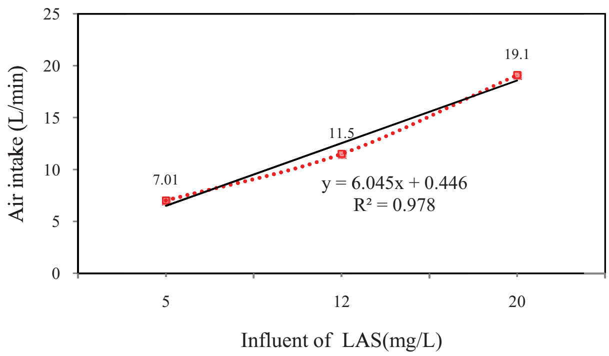

Fig. 5. The relationship between air intake and LAS concentration in different loading rates.

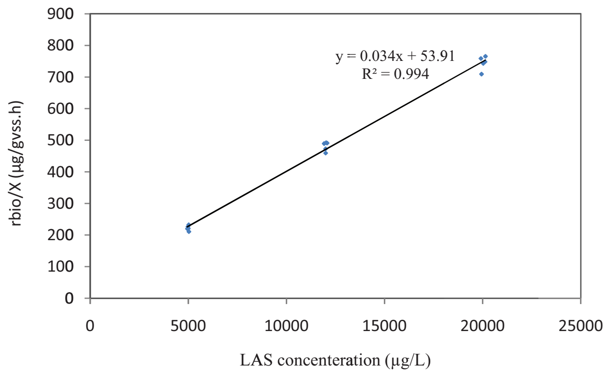

Fig. 6. Specific biodegradation rate of LAS (µg/gVSS h) in IFAS at different concentration of LAS at base on MM model.

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