

Effective biosorption of Allura red dye from aqueous solutions by the dried-lichen (*Pseudoevernia furfuracea*) biomass

Zeynep Mine Şenol

Department of Food Technology, Zara Vocational School, Cumhuriyet University, Sivas, Turkey

ABSTRACT

Allura red (AR), which is known as the monoazo class of synthetic food colourant(E129) has been widely used in food industries. Due to the potential toxicity of azo dyes and pathogenicity, the removal of AR from industrial wastewaters is very important environmentally. So, this article aims to investigate the biosorption process of AR by lichen (Pseudoevernia furfuracea) from aqueous solutions. Batch biosorption conditions of AR food dye onto lichen biosorbent as initial AR concentration, solution pH, contact time, temperature and recovery were investigated. From the results, it has been observed that the highest removal efficiency is approximately 87% at a contact time of 5 hours, initial AR food dye concentration of 1000 mgL⁻¹ and agitation speed of 150 rpm at natural pH 8.0. The maximum ARbiosorption capacity from the Langmuir model was found as 0.280 mol kg⁻¹ at 25°C. Biosorption kinetics were analysed by using intra-particle diffusion and pseudo-secondorder models. Biosorption thermodynamics has shown that ARbiosorption onto lichen biosorbent is endothermic, possible and spontaneous. The lichen (Pseudoevernia furfuracea) can become an alternative biosorbent for the removal of AR from the environment and wastewater.

ARTICLE HISTORY

Received 6 May 2020 Accepted 14 June 2020

KEYWORDS

Lichen; *Pseudoevernia furfuracea*; biosorption; food dye; Allura red



1. Introduction

Allura Red (AR) is an azo dye used to synthetic dye in the food industry.AR food dye could be found in many foodstuffs, for example, ice cream, soft drinks, candies and bakery products [1]. The azo dyes are dangerous for human health and the environment due to their mutagenic and carcinogenic properties [2]. Therefore, removal of the azo dye from industrial wastewaters is very important environmentally. Various methods have been used for removal of dye industry wastewater such as membrane filtration, coagulation, ozonation, oxidation, precipitation, filtration and biosorption [3,4]. Among these methods, biosorption is an alternative eco-friendly method for the removal of azo dyes from aqueous effluents and industrial wastewater [5]. Biosorption has the advantages of simplicity of design, high selectivity, low cost and easy usability [6,7]. The use of biosorbent is very important for reasons that are environmentally friendly, effective, inexpensive biosorbent and non-hazardous materials. Many biological materials (biosorbents) have been used to remove dyes from aqueous solutions, such as fungus [8], algae [9], bacteria [10] and lichen [11]. Lichens are organisms composed of a fungus and an alga in a symbiotic relationship. Lichens are among the widely used indicators of environmentally due to their retaining a variety of pollutants and higher capacity for accumulation [12]. Lichens have been widely used as environmental pollution biomonitoring due to their capability to strongly bind and accumulate many inorganic and organic compounds [13]. However, there is limited information about the usage of lichens in wastewater treatment technologies in the literature. Recent studies focus on the usage of lichens for removal of heavy metal from wastewater [3,14]. Also some new ones reported textile dye biosorption by lichens efficiently [11,15,16]. However, there is not enough study about the removal of food colourants.

The main objective of this study is to investigate the potential use of this lichen *Pseudoevernia furfuracea*, as a natural, economic and eco-friendly biosorbent for removal of Allura red food dye. The dried-lichen biomass without making any chemical treatment was selected as a low-cost adsorbent due to its eco-friendly, cost-effective, wide availability, safe, renewable and easy collection. To my present knowledge, this is the first report about the utilisation of lichen called *Pseudoevernia furfuracea* obtained from Bilecik province, Turkey for the biosorption of food colourants from aqueous solutions. This study shows the food colourant biosorption properties of a novel lichen species including isotherm, kinetic and thermodynamic studies. Also the characterisation analyses of this novel lichen biosorbent were carried out in the current study.

2. Material and methods

2.1. Dye

Allura Red (AR) was purchased from Merck (Germany). The chemical structure of the Allura red food dye is shown in Figure 1.

The stock solution was prepared by dissolving 200 mg of AR food dye in 100 mL double-distilled water. The AR solutions of different concentration ($25-2000 \text{ mgL}^{-1}$) were prepared by successive dilutions the stock solutions with a suitable volume of double-distilled water.



Figure 1. Structure of Allura red food dye.

2.2. The collection and preparation of Lichen Biosorbent

In the current study, *Pseudoevernia furfuracea* was used for the removal of AR dye. The lichen samples were collected from Bilecik province (N 400 11.5262', E 0290 57.962') and, dried lichen was used as biosorbent. The lichen samples were cleaned from foreign matters such as soil or bark under the binocular microscope (Primo Star Zeiss) and washed with double-distilled water. The lichen samples were dried at 70 °C throughout one night, then powdered to prepare biosorbent.

2.3. Reagents and equipment

Double-distilled water was used in all experiments. All chemicals were supplied from Merck (Germany). All experiments were studied in duplicate. The AR concentrations were detected using a UV-Visible spectrophotometer (Shimadzu, 160 A model, Kyoto, Japan). Lichen biosorbent was characterised by FT-IR and SEM analysis. FT-IR spectra of lichen biosorbent were recorded in a Perkin Elmer 400 spectro-photometer. SEM images were obtained with a Leo 440 Computer Controlled Digital System.

2.4. Batch biosorption experiments

The biosorption of AR on lichen was investigated by using the batch method. For the biosorption experiments, the biosorbent-solution systems were equilibrated with 100 mg of lichen biosorbent and 1000 mg L⁻¹AR dye concentration at natural pH 8.0, for 24 h at 25° C in 10 mL polypropylene tubes containing 10 mL of AR dye solution was kept in a thermostatic water bath with constant agitation speed (150 rpm). The pH was adjusted with dilute HCl and NaOH solutions (each one, 0.2 and/or 2.0 mol L⁻¹). The concentrations of AR in solution were

spectrophotometrically determined by measuring the absorbance of the solutions at 506 nm [17]. Biosorption% and Q (mol kg^{-1}) were calculated with Equations 1 and 2.

$$Biosorption\% = \left[\frac{C_i - C_f}{C_i}\right] x100$$
 Eq.1

$$Q = \left[\frac{C_i - C_f}{m}\right] x V$$
 Eq.2

where C_i is the initial concentration (mg L⁻¹), m refers to the adsorbent mass (g), C_f is equilibrium concentration (mg L⁻¹), and V is the solution volume (L).

All of the experiments were performed in duplicate. The standard error of data is calculated according to Equation 3.

$$SE = \sqrt{\sigma^2}$$
 Eq.3

where σ represents the square root of the estimated error variance of the quantity.

2.5. Desorption procedure

In this study, HCl, NaOH, HNO₃ and Ethyl alcohol solutions (each one, 0.1 mol L^{-1}) were used for desorption of the AR dye from the surface of the lichen biosorbent. In order to determine the recovery rates of the lichen biomass with desorption in Figure 10, the experiments were repeated three times with the same biosorbent for the biosorption/ desorption cycle. At the end of each experiment, the solutions were centrifuged at 5000 rpm for 10 min to ensure liquid-solid separation and the amounts of AR dye in the equilibrium solution were detected at 506 nm utilising UV-Visible spectrophotometer. Desorption % is calculated with Equation 4.

Desorption% =
$$\frac{Q_{des}}{Q_{ads}} x100$$
 Eq.4

In this equation; Q_{des} ; the desorbed amount of AR (mol kg⁻¹), Q_{ads} ; biosorbed amount of AR (mol kg⁻¹).

3. Results and discussion

3.1. FT-IR analysis

The FT-IR analyses give important information about the functional groups of lichen biosorbent in the biosorption process. In the current paper, the FT-IR analyses of lichen samples after and before biosorption was done (Figure 2).

FT-IR study clearly seems that lichen has characteristic peaks. The strong bands at 3418–3260 cm⁻¹ were due to bound amine (–NH) or hydroxyl (–OH) groups. The peaks at 2926 cm⁻¹ were attributed to the stretching vibration of = C–H [18,20]. The peaks of the carboxyl group (–C = O) groups were observed at 1651–1614 cm⁻¹. The peak at 1038 cm⁻¹ was attributed to –C = O stretching of alcohols and carboxylic acids. The peak at 1014 cm⁻¹ was attributed to the stretching vibration of -CN. After biosorption of Allura red, lichen surface functional groups shifted to 3430–3255, 2937, 1622, 1315 and 831 cm⁻¹ which corresponds to



Figure 2. FT-IR spectra of lichen biosorbent before and after biosorption of AR.

the functional groups such as – OH, = C–H, –C = O, –CHO and – CN, respectively [21]. These results indicated that the hydroxyl (–OH) and carboxyl (–COOH) groups of the biosorbent to be mainly related in the biosorption of AR onto lichen biosorbent. The FT-IR analysis results indicated the formation of hydrophobic interactions, H-bonding and surface complexation between AR with the functional groups of the lichen surface.

3.2. SEM analysis

In order to define the surface morphology of the lichen biosorbent, before and after biosorption was taken from the samples. SEM images of lichen biosorbent before and after AR biosorption are shown in Figure 3. SEM images of lichen biosorbent showed particles that are a huge porous with irregular shapes, rough and edges. The surface of the lichen after AR biosorption was generally smooth and rounded indicating that the particles deposited were AR. This view might be because of the H-bonding and hydrophobic interactions between AR with the active sites on the surface of the lichen biosorbent.

3.3. Effect of point of zero charge and initial pH

Surface charge of lichen biosorbent is an important factor for affecting AR biosorption. The solution pH at which the surface charge of the lichen biosorbent becomes zero is defined as the point of zero charge (PZC). In order to determine the PZC values of the lichen biosorbent, 100 mg biosorbent was incubated for 24 hours in pH solutions ranging from 1.0- to 12.0. The pH was adjusted with dilute HCl and NaOH solutions (each one, 0.1 and/or 1.0 mol L⁻¹). And then, the equilibrium pH values were measured. A point of zero charge (PZC) was used to characterise the electronegativity of the biosorbent. The PZC value of the lichen biosorbent was found to be 4.53 (Figure 4).

As shown in Figure 4, the PZC value of lichen was determined as 4.53. The surface charge of lichen was positive at pH <4.53 and, negative at pH >4.53. In biosorption



Figure 3. SEM photographs of Lichen (a) and AR biosorbed Lichen (b).



Figure 4. PZC plots of the lichen biosorbent in range of pH 2.0–9.0.



Figure 5. Effect of pH on the biosorption of AR onto lichen biosorbent.

biosorbent, resulting in less biosorption of anionic dye ($pH_{AR}>pH_{PZC}$). This results from H-bonds and hydrophobic interactions between the negatively charged lichen biosorbent surface and anionic AR dye [22].

3.4. Effect of biosorbent dosage on biosorption

Biosorbent-solution ratio is an important factor for determining the biosorption capacity of biosorbent. To investigate the influence of initial biosorbent dosage on AR dye was examined by varying dosages between 0.1-20 g L⁻¹. The obtained results presented in Figure 6. The biosorption of AR dye by the lichen biosorbent was increased with the number of active centres in the biosorbent. So, when the biosorbent dosage was



Figure 6. The effect of biosorbent dosage on the biosorption of AR onto lichen biosorbent.

8 👄 Z. M. ŞENOL

increased, the AR dye biosorption yield increased. As seen from Figure 6., the maximum biosorption was found to be 87% in the biosorbent dosage of 20 g L⁻¹. The biosorption was found to be 80% with the biosorbent dosage of 10 g L⁻¹. It was thought that biosorption may be increased with the increase in the amount of biosorbent by the effect of – OH, -SO₃Na, -N = N- [23] groups in AR food colouring dye structure and phenolic groups in the structure of a lichen.

3.5. Biosorption isotherm models on biosorption

Biosorption isotherms describe the biosorption behaviour of AR dye by lichen. Three equilibrium models,

Langmuir, Freundlich and Dubinin Radushkevich (D-R) isotherm models were used to specify the biosorption process, surface properties and biosorption mechanism of the lichen biosorbent [24,26]. The Langmuir, Freundlich and D-R isotherm equations are expressed by the following (Equations. 5, 6 and 7, respectively)

$$Q = \frac{X_L K_L C_e}{1 + K_L C_e}$$
 Eq.5

$$Q = K_F C_e^{\beta}$$
 Eq.6

$$Q_e = Q_{DR} e^{-K_{DR}\epsilon^2} Eq.7$$

where Q is the amount of biosorbed dye (mol kg⁻¹), X_L is the maximum biosorption capacity, K_L is the parameter for Langmuir isotherm and C_e is the equilibrium concentration (mol L⁻¹) and K_F: Freundlich constant, β : adsorbent surface heterogeneity. X_{DR} is a measure of biosorption capacity, K_{DR} is the activity coefficient (mol² KJ²) and ϵ stands for constant related to the biosorption energy and the Polanyi potential. R (8.314 Jmol⁻¹ K⁻¹) and T are the ideal gas constant and the absolute temperature (K).The Polanyi potential (ϵ) is expressed by the following Equation 8:

$$\varepsilon = RTln\left(1 + \frac{1}{C_e}\right)$$
 Eq.8

The biosorption energy (E, kJ mol^{-1}) is expressed by the following Equation 9:

$$E_{DR} = (2K_{DR})^{-0.5}$$
 Eq.9

If the biosorption energy is $8 < E < 16 \text{ kJ mol}^{-1}$, the biosorption is physically controlled and $E < 8 \text{ kJmol}^{-1}$ indicates that the biosorption proceeds physically [27].

Standard deviation 0.0947 mol kg⁻¹, standard error 0.0244 mol kg⁻¹

The biosorption isotherms of the lichen biosorbent for AR dye are shown in Figure 7. The biosorption isotherm constants for AR biosorption are presented in Table 1. It was observed that, when the nonlinear regression coefficient (R^2) which, obtained Langmuir and Freundlich isotherms were compared, the Langmuir isotherm model ($R^2 = 0.977$) was suitable for defining the biosorption of AR by lichen. This situation indicated that all the AR biosorbat molecules were in contact with the active sites on the surface of the lichen biosorbent and the biosorption process was monolayer character. The monolayer



Figure 7. The experimentally obtained biosorption isotherms of ARonto lichen biosorbent.

lsotherm	Parameter	Value	R ²
Langmuir	X _L (mol kg ⁻¹)	0.280	0.977
	K_L (L mol ⁻¹)	6127	
Freundlich	X _F	3.18	0.900
	β	0.381	
D-R	X_{DR} (mol kg ⁻¹)	14.5	0.998
	K _{DR} x10 ⁹ /mol ² KJ ⁻²	9.48	
	E _{DR} /kJ mol ⁻¹	7.26	

 Table 1. Langmuir, Freundlich and Dubinin-Radushkevich isotherm model parameters.

biosorption capacity of the lichen biosorbent for AR dye was 0.280 mol kg⁻¹. The K_L value was found in 6124 L mol⁻¹. The K_F 3.18 which is a measure of the biosorption capacity, and β surface heterogeneity was also found as 0.381, were obtained from the Freundlich model. Since the β value was between 0 and 1, the biosorption of AR onto lichen biosorbent was favourable. The E_{DR} (kJ mol⁻¹) value gives information about biosorption mechanism, physical or chemical [27]. From the D-R model, the biosorption energy was calculated to be 7.26 kJ mol⁻¹. This result suggests that the biosorption process of AR dye onto the lichen biosorbent may be carried out by a mechanism being physical in nature because the biosorption energy lies within E_{DR}<8 kJ mol⁻¹.

3.6. Biosorption kinetics

To investigate the biosorption kinetic, three different kinetic models, pseudo-first-order kinetic (PFO), pseudo-second-order kinetic (PSO) and, Intra-particle diffusion (IPD) models were used to fit the experimental data and the related parameters were derived Equations 10,11 and 12, respectively [28,31].

$$Q_t = Q_e \big[1 - e^{-k_1 t} \big] \hspace{1cm} Eq.10$$

$$Q_t = \frac{t}{\left[\frac{1}{k_2 Q_e^2}\right] + \left[\frac{t}{Q_e}\right]} \tag{Eq.11}$$

$$Q_t = k_i t^{0.5}$$
 Eq.12

where Q_t (mol kg⁻¹) is the lead biosorbed amount at time t (min), Q_e (mol kg⁻¹) is the biosorbed amount at equilibrium, k_1 , k_2 and k_i is the rate constant of the PFO (min⁻¹), the PSO model (mol⁻¹ kg min⁻¹) and the intra-IPD (mol⁻¹ kg min^{-0.5}) model, respectively.

Figure 8 shows the compatibility to the PFO, the PSO and the IPD models and the parameters derived from these models are summarised in Table 2. As seen from Figure 8, biosorption was increased rapidly up to 300 min, and then it was seen to equilibrium. Removal of AR by using lichen biosorbent was fast and the equilibrium was achieved in

 Table 2. Pseudo-first-order, pseudo-second-order and intra-particle diffusion kinetic model parameters.

Kinetic model	Parameter	Value	R ²
Pseudo-first order	Q_t /mol kg ⁻¹	0.229 0).963
	$Q_e/mol kg^{-1}$	0.257	
	$k_1 \times 10^3 / dk^{-1}$	8.29	
	$Hx10^{3}/mol kg^{-1} min^{-1}$	1.83	
Pseudo-second order	Q_t /mol kg ⁻¹	0.229 0).973
	Q _e /mol kg ⁻¹	0.221	
	$k_2 \times 10^3 / mol^{-1} kg min^{-1}$	39.4	
	$Hx10^{3}/mol kg^{-1} min^{-1}$	2.60	
Intra-particle diffusion	k _i x10 ³ /mol kg ⁻¹ min ^{-0.5}	12.6 0).959

Standard deviation 0.0766 mol kg^{-1} , standard error 0.0181 mol kg^{-1}



Figure 8. Compatibility of AR biosorption kinetics to pseudo-first-order, pseudo-second-order and intra-particle diffusion models.

300 min. Because of the correlation coefficient (R²) values, the value of PSO kinetic model was higher than PFO kinetic model. Moreover, theoretically calculated $Q_t(0.229 \text{ mol kg}^{-1})$ values from the PSO model agree very well with the experimental $Q_e(0.221 \text{ mol kg}^{-1})$ values. The results indicated thatAR biosorption onto lichen fitted best with PSO kinetic model. The first linear part of the IPD is due to the bonding of the AR dye to the active centres on the lichen surface and surface biosorption. The second linear part; is due to dye diffusion to the active areas in the pores of the lichen. In this case, lichen dye biosorption is explained by the PSO and the IPD models.

3.7. Biosorption thermodynamics

Thermodynamic parameters of biosorption are very important to explain the effect of the temperature on the AR biosorption on lichen. Thermodynamic parameters (enthalpy, ΔH^0 and entropy, ΔS^0) were obtained from lnK_D against 1/T the graph [32,33]. The free Gibbs energy (ΔG^0) is calculated from Eq. 16. ΔH^0 , ΔS^0 and ΔG^0 were calculated using the following equations;

$$K_D = \frac{Q}{C_e}$$
 Eq.13

$$\Delta G = -RTlnK_D \qquad \qquad \text{Eq.14}$$

$$InK_D = \frac{\Delta S^0}{R} - \frac{\Delta H^0}{RT}$$
 Eq.15

$$\Delta G^0 = \Delta H^0 - T \Delta S^0$$
 Eq.16

In Figure 9(In K_D-1/T) the values of ΔH^0 and ΔS^0 were calculated from the slope of the graph and the cut-off, respectively. Biosorption enthalpy was found negative. ΔH^0 was



Figure 9. The effect of temperature on the biosorption of AR by lichen biosorbent.

calculated as 29.4 kJ mol⁻¹showed that the biosorption process was endothermic. ΔS^0 was calculated as 150 Jmol⁻¹ K⁻¹. The free energy value was found as -15.3 kJ mol⁻¹ at 25°C. The negative free energy value indicated that spontaneous adsorption was possible.

3.8. Desorption efficiency

Desorption studies are very important to make the biosorption process more economical. Reusability investigated the desorption ability for AR biosorption onto lichen. The lichen biosorbent was regenerated using HNO₃, NaOH, HCl and ethyl alcohol. Desorption results are given in Figure 10. The maximum recovery percentage for AR onto lichen biosorbent was achieved with HCl (76%). The minimum recovery percentage for AR onto lichen biosorbent was achieved with Ethyl alcohol (2.0%).

According to the results of this study, the usage of HCl solution ensured the best desorption efficiency. So, HCl was selected as a suitable desorption solvent for the regeneration of the lichen biosorbent. The lichen biosorbent can be recycled at least eight times without the expense of adsorption capacities. After the end of eight cycles, the biosorption capacity was around 72% with a decrease of 5.0% in the recovery rate. This reasonable and acceptable reusability performance indicates that the lichen biosorbent can be used in practical applications for the removal of AR dye.

3.9. Comparison of the maximum sorption capacity of AR on the lichen biosorbent with those of other biosorbents

A comparison has been made between the performances of the lichen biosorbent and other sorbents reported in the literature for sorption of AR dye. The maximum sorption capacity obtained from the Langmuir model was used for comparison with those of different sorbents with and without modification. As can be seen in Table 3, the maximum sorption capacity of the lichen biosorbent was either higher or comparable than that of



Figure 10. The recovery rates of various solvents for desorption of AR.

Sorbent	рН	Temperature ^o C	Max. sorption capacity, mol kg ⁻¹	References
a-picoline ionic liquid-β-cyclodextrin-cross-linked polymer	3.0	25	0.0143	[34]
Activated pine wood	2.0	25	0.0143	[35]
Mn- and Cu- @ ZnS nanoparticles loaded on activated carbon	2.5	25	0.101	[36]
Silica/AgNPs-glucose	4.0	30	0.136	[37]
Activated carbon	7.0	25	0.146	[38]
Poly(ionic liquid)immobilised magnetic nanoparticles	-	30	0.213	[39]
Spirulina platensis	-	25	0.944	[40]
Chitosan/Polyurethane Foam	-	25	0.217	[41]
Natural sawdust	2.0	25	0.051	[42]
Hexadecylpyridinium bromidetreated sawdust	2.0	25	0.151	[42]
Pseudoevernia furfuracea biomass	8.0	25	0.280	This study

Table 3. Comparison of the maximum sorption capacity of AR dye on the lichen biosorbent with those of other sorbents.

the other sorbent materials. According to the obtained results, reasonable sorption capacity and a performance that can compete with other sorbents in this work indicated that the lichen biosorbent was simple, easy to use and cost-effective biosorbent for removal of toxic AR food dye at ppm levels from aqueous solutions.

4. Conclusions

In this research, lichen biosorbent can be used as an effective, alternate biosorbent for AR food dye removal. Optimal working parameters were found as pH of the solution: 8.0, biosorbent dosage: 100 mg, temperature: 25° C and contact time: 5 hours. Adsorption isotherm models showed that AR biosorption onto lichen was more appropriate for the Langmuir model. The adsorption free energy was found as E_{DR} (7.26 kJmol⁻¹), which indicated that AR onto lichen biosorbent composite was physically performed. The maximum biosorption capacity was 0.280 mol kg⁻¹ from the Langmuir isotherm model. The biosorption kinetic was also explained with PSO and intra-IPD models. The biosorption thermodynamics showed that AR biosorption was possible, spontaneous and endothermic. The results found in this study showed that the dried-lichen biosorbent in the removal of AR food dye pollutants from the wastewater.

Acknowledgments

The present study was partly supported by the Cumhuriyet University Scientific Research Projects Commission. The author strongly declares that no scientific and/or financial conflicts of interest exist with other people or institutions.

Disclosure statement

No potential conflict of interest was reported by the author(s).

ORCID

Zeynep Mine Şenol ib http://orcid.org/0000-0002-5250-1267

References

- [1] K. Rovina, S. Siddiquee and S.M. Shaarani, Front. Microbiol. 7, 798. (2016).
- [2] K.T. Chung, J. Environ. Sci. Health. C. **34**, 233 (2016). doi:10.1080/10590501.2016.1236602.
- [3] Z.M. Şenol, Ü.D. Gül and S. Şimşek, Environ. Sci. Pollut. R. 26, 27002 (2019). doi:10.1007/ s11356-019-05848-x.
- [4] N.A. Salvi and S. Chattopadhyay, Appl. Water. Sci. 7, 3041 (2017). doi:10.1007/s13201-016-0417-0.
- [5] C. Patra, T. Shahnaz, S. Subbiah and S. Narayanasamy, Environ. Sci. Pollut. Res. 1, 16 (2020).
- [6] T. Shahnaz, S.M.M. Fazil, V.C. Padmanaban and S. Narayanasamy, Int. J. Biol. Macromol. 151, 322 (2020). doi:10.1016/j.ijbiomac.2020.02.181.
- [7] S. Kumar, S. Narayanasamy and R.P. Venkatesh, Sep. Sci. Technol. 17, 2783 (2019). doi:10.1080/01496395.2018.1560333.
- [8] T.R. Waghmode, M.B. Kurade and S.P. Govindwar, Int. Biodeter. Biodegr. 65, 479 (2011). doi:10.1016/j.ibiod.2011.01.010.
- [9] Z. Aksu and S. Tezer, Process. Biochem. 40, 1347 (2005). doi:10.1016/j.procbio.2004.06.007.
- [10] A.A. Kadam, J.D. Kamatkar, R.V. Khandare, J.P. Jadhav and S.P. Govindwar, Environ. Sci. Pollut.
 R. 20, 1009 (2013). doi:10.1007/s11356-012-0929-6.
- [11] G. Bayazıt, Ü.D. Gül and D. Ünal, Int. J. Environ. Stud. 76, 608 (2019). doi:10.1080/ 00207233.2018.1502959.
- [12] T.H. Nash and V. Wirth, Bibl. Lichenol. 30, 1 (1988).
- [13] F. Ekmekyapar, A. Aslan, Y.K. Bayhan and A. Cakici, Int. J. Environ. Res. 6, 417 (2012).
- [14] Ü.D. Gül, Z.M. Şenol, N. Gürsoy and S. Şimşek, J. Environ. Radioact. 205, 93 (2019). doi:10.1016/ j.jenvrad.2019.05.008.
- [15] T. Tay and R. Keçili, Biol. Divers. Conserv. 9, 189 (2016).
- [16] H. Koyuncu and A.R. Kul, Appl. Water Sci. 7, 72 (2017). doi:10.1007/s13201-020-1156-9.
- [17] M. Soylak, Y.E. Unsal and M. Tuzen, Food Chem. Toxicol. 49, 1183 (2011). doi:10.1016/j. fct.2011.02.013.
- [18] F. Ekmekyapar, A. Aslan, Y.K. Bayhan and A. Cakici, J. Hazard. Mater. 137, 293 (2006). doi:10.1016/j.jhazmat.2006.02.003.
- [19] O.D. Uluozlu, A. Sarı, M. Tuzen and M. Soylak, Bioresour. Technol. 99, 2972 (2008). doi:10.1016/ j.biortech.2007.06.052.
- [20] Z. Kılıç, O. Atakol, S. Aras, D. Cansaran-Duman and E. Emregul, J. Air Waste Manag. Assoc. 64, 1112 (2014). doi:10.1080/10962247.2014.926299.
- [21] N.K. Mondal and M. Kundu, Water Conserv. Sci. Eng. 1, 143 (2016). doi:10.1007/s41101-016-0009-8.
- [22] A.F. Streit, L.N. Côrtes, S.P. Druzian, M. Godinho, G.C. Collazzo, D. Perondi, G.L. Dotto, Sci. Total Environ. 660, 287 (2019).
- [23] M. Wawrzkiewicz and Z. Hubicki, Environ. Technol. 30, 1059 (2009). doi:10.1080/ 09593330903055650.
- [24] J. Langmuir, J. Am. Chem. Soc. 40, 1361 (1918). doi:10.1021/ja02242a004.
- [25] H. Freundlich and W. Heller, J. Am. Chem. Soc. 61, 2228 (1939). doi:10.1021/ja01877a071.
- [26] K.Y. Foo and B.H. Hameed, Chem. Eng. J. 156, 2 (2010). doi:10.1016/j.cej.2009.09.013.
- [27] K. Vijayaraghavan, T.V.N. Padmesh, K. Palanivelu and M. Velan, J. Hazard. Mater. 133, 304 (2006). doi:10.1016/j.jhazmat.2005.10.016.
- [28] S. Lagergren, Handlingar **24**, 1 (1898).
- [29] Y.S. Ho and G. McKay, Process. Biochem. 34, 451 (1999). doi:10.1016/S0032-9592(98)00112-5.
- [30] Y.S. Ho and C.C. Wang, Process. Biochem. 39, 761 (2004). doi:10.1016/S0032-9592(03)00184-5.
- [31] F.C. Wu, R.L. Tseng and R.S. Juang, Chem. Eng. J. 153, 1 (2009). doi:10.1016/j.cej.2009.04.042.

- [32] S. Şimşek, Z.M. Şenol and H.I. Ulusoy, J. Hazard. Mater. 338, 437 (2017). doi:10.1016/j. jhazmat.2017.05.059.
- [33] V.K. Gupta, A. Rastogi, V.K. Saini and J. Neeraj, J. Colloid. Interface. Sci. 296, 59 (2006). doi:10.1016/j.jcis.2005.08.033.
- [34] X. Qin and X. Zhu, Anal. Lett. 49, 189 (2016). doi:10.1080/00032719.2015.1065880.
- [35] M.A. Al-Ghouti, A.A. Issa, B.S. Al-Saqarat, A.Y. Al-Reyahi and Y.S. Al-Degs, Desalin. Water. Treat. 57, 27651. (2016).
- [36] A. Ostovan, H. Asadollahzadeh and M. Ghaedi, Ultrason. Sonochem. 43, 52 (2018). doi:10.1016/j.ultsonch.2018.01.002.
- [37] M.A. Salem, R.G. Elsharkawy, M.I. Ayad and M.Y. Elgendy, J. Sol-Gel Sci. Techn. 91, 523 (2019). doi:10.1007/s10971-019-05055-7.
- [38] S.A. Alkahtani, S.S. Abu-Alrub and A.M. Mahmoud, Int. J. Food Allied Sci. 3, 10 (2017). doi:10.21620/ijfaas.2017110-26.
- [39] A.A.A. Bakheet and X.S. Zhu, J. Mol. Liq. 242, 900 (2017). doi:10.1016/j.molliq.2017.07.097.
- [40] G.L. Dotto, M.L.G. Vieira, V.M. Esquerdo and L.A.A. Pinto, Braz. J. Chem. Eng. 30, 13 (2013). doi:10.1590/S0104-66322013000100003.
- [41] R. da Rosa Schio, B. Cruz da Rosa, N.P.G. Salau, E.S. Mallmann and G.L. Dotto, Chem. Eng. Technol. 42, 2434 (2019). doi:10.1002/ceat.201800749.
- [42] T.K. Saha, R.K. Bishwas, S. Karmaker and Z. Islam, ACS Omega. 5, 13374 (2020).