
*International Research Journal of Pure & Applied Chemistry*

***22(5): 1-11, 2021; Article no.IRJPAC.70089***
***ISSN: 2231-3443, NLM ID: 101647669***

# Removal of Dyes from Wastewater of Artisanal Dyeing Plants by Adsorption in a Fixed Bed Column of Deactivated Lichens

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***Authors' contributions***

*This work was carried out in collaboration among all authors. Author KKD-B performed literature research, wrote the protocol and the first version of the manuscript. Author YZ managed the methodology, analyzes and the final version of the manuscript. Author DDY managed the collection of samples and data. Author EL designed the study. All authors read and approved the final manuscript.*

***Article Information***

DOI: 10.9734/IRJPAC/2021/v22i530405
Editor(s):
(1) Prof. Wolfgang Linert, Vienna University of Technology, Austria.
Reviewers:
(1) Pornpimol Muangthai, Srinakharinwirot University, Thailand.
(2) A. Kistan, Anna University, India.
Complete Peer review History: http://www.sdiarticle4.com/review-history/70089

***Original Research Article***

***Received 20 April 2021***
***Accepted 25 June 2021***
***Published 01 July 2021***

## ABSTRACT

**Aims:** Pollution by wastewaters from various urban activities such as artisanal dyeing plants is a real problem for developing countries. The treatment of wastewater by the adsorption method is carried out by means of less expensive and available adsorbent media. Two techniques of the adsorption method are possible: adsorption in continuous mode (column adsorption) and adsorption in discontinuous mode (batch adsorption). The choice of the continuous adsorption technique is justified by its ability to process large volumes of solutions. In this study, dyes contained in wastewater from artisanal dyeing plants were removed by continuous adsorption in a fixed-bed column of deactivated lichen biomass (Parmotrema dilatatum).

**Study Design:** Random design

**Place and Duration of Study:** Laboratory of Thermodynamics and Environmental Physico-Chemistry (University Nangui Abrogoua, Ivory Coast) between May 2020 and October 2020.

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**Methodology:** Four (4) categories of wastewater were collected in artisanal cotton and leather dyeing plants through two municipalities of the city of Abidjan, economic capital of Ivory Coast. Two (2) wastewaters colored in blue from dyeing of cotton boubous and jeans and two (2) wastewaters colored in red from dyeing of leather jackets and bags. These wastewaters were treated through the fixed bed column of deactivated lichens. The column feed rate was set at 0, 07 $L.min^{-1}$ and the adsorbent bed mass at 100 g.
**Results:** The study showed that, regardless of the nature of the dyed object and regardless of the target dye, the amount of dye adsorbed was better with waters of higher initial concentration. Thus the best amount of adsorbed dye is 44.444 $mg.g^{-1}$ and the best removal rate is 97.9%. These values are obtained with the red wastewater of bags (RWB) treatment which was the most concentrated wastewater.
**Conclusion:** Good efficiency of deactivated lichen bed as adsorbent for the in situ removal of dyes from wastewater by continuous adsorption.

*Keywords: Wastewater; continuous adsorption; dyes; dye plant; artisanal; lichens; fixed bed column.*

## 1. INTRODUCTION

In Ivory Coast, approximately 60% to 85% of industrial activity is concentrated in the city of Abidjan due to the vitality of its road and port infrastructures [1,2]. This industrial activity covers the plastic, food, cosmetics, paper, pharmaceutical, textile and dyeing sectors. In their operation, the industrial units use water for the transformation processes and in the cleaning of installations and tools. Unfortunately, about 10-15% of the wastewater of these industries is discharged without any prior treatment into the receiving environments [3]. The artisanal and modern dye plants are among the main sources of pollution, producing about 85% of wastewaters volume in Ivory Coast [4,5]. These dye plants use a wide range of dyes in their dyeing process. Many of the dyes present in these dye plants wastewaters have harmful effects on health [6,7] and can negatively affect photosynthesis [8,9]. The proliferation of artisanal dyeing plants increases the content of dyes in receiving environments, which can lead to health and environmental problems. To protect its environment and the health of its population, Ivory Coast since its independence has ratified several treaties, conventions and laws aimed at reducing the pollution levels [10]. Despite existence of such a political will, about only 1% of the industries in Abidjan have wastewater treatment stations before discharge [2]. Some of the existing treatment stations are either not functional or poorly managed. The lagoon Ebrié that is the main recipient of these industries wastewater, has been the subject of several studies that have revealed a deterioration in the quality of its water in connection with industrial discharges [1,2,4,11,12]. The data from these studies have prompted some researchers in Ivory Coast to conduct additional studies on the removal of pollutants such as: trace metals [13], humic substances [14] and phosphates [15]. On the other hand, little work has been done in Ivory Coast on the dyes removal of from wastewaters of artisanal dyeing plants.

This study is an approach for the treatment of wastewater from artisanal dyes plants that may contain potentially toxic dyes. The approach uses as adsorbents biomass from lichens (*Parmotrema dilatatum*) previously deactivated and placed in a column to serve as a fixed bed. The choice of the continuous adsorption technique is justified by its ability to treat large volumes of solutions [1]. Lichens are vegetable species that result from a symbiotic association between an alga and a fungus. They can survive extreme dehydration, even for months or years and can also tolerate hard environmental conditions. They grow on rock surfaces, tree trunks, house roofs, monuments and on the soil surface [16,17]. Several studies have highlighted their uses as bioindicators of air quality [18,19-24] and their high capacity to adsorb pollutants such as trace metals [25-31]. In the present study, the lichen species is used to evaluate its ability to serve as an adsorbent bed in continuous adsorption processes of dyes from the wastewater of artisanal dyeing units.

## 2. MATERIALS AND METHODS

### 2.1 Lichen Biomass Collection

The fresh lichens biomass are colleted in the ecologycal reserve of LAMTO (5.02°C W and 6.13°C N). This reserveof 2500 ha, is at174 km from the city of Abidjan (Ivory Coast) between Singrobo and Taabo and extends along the

Bandama River [32]. The average annual temperature is 28.28°C. The value of annual rainfall is 1194 mm with a humidity rate above 58% [33].

The collected specie (*Parmotrema dilatatum*) has a lobed foliaceous thallus between 3-10 cm wide. The foliaceous thallus is the most abundant thallus in the Ivorian vegetation, particularly in the Lamto reserve. Species of the genus *Parmotrema* can survive in humid, arid or semi-arid climates; they possess a very important geographical diversity [34].

The lichens are collected from mature trees with diameters greater than 20 cm with bark that is not cracked, injured or tilted.

## 2.2 Deactivation Process

In the laboratory, the collected lichens are detached from their substrates and then manually cleaned to remove any soil, leaf, dust or insect debris. The thallus is then washed with distilled water. A total of 1500 g of treated fresh lichens were placed at 80°C in an oven (Memmert BM300, Schwabach, Germany) for 48 hours to deactivate them. The deactivated lichens were ground. The grind was sieved to obtain grains with a diameter is d =125µm.

## 2.3 Wastewater Collection

Wastewater samples were collected from artisanal dyeing sites in the city of Abidjan (4°01' W and 5°19' N), specifically in the two neighborhoods of Abobo and Adjame (Fig. 2). These neighborhoods were chosen because of the importance of the artisanal dyeing activities that they record. In these neighborhoods, wastewater from artisanal dyeing plants are discharged directly into the environment and drained into the aquatic environments. The sampling equipment consists of 20L polyethylene drums placed at each site to collect wastewater for 24 hours.

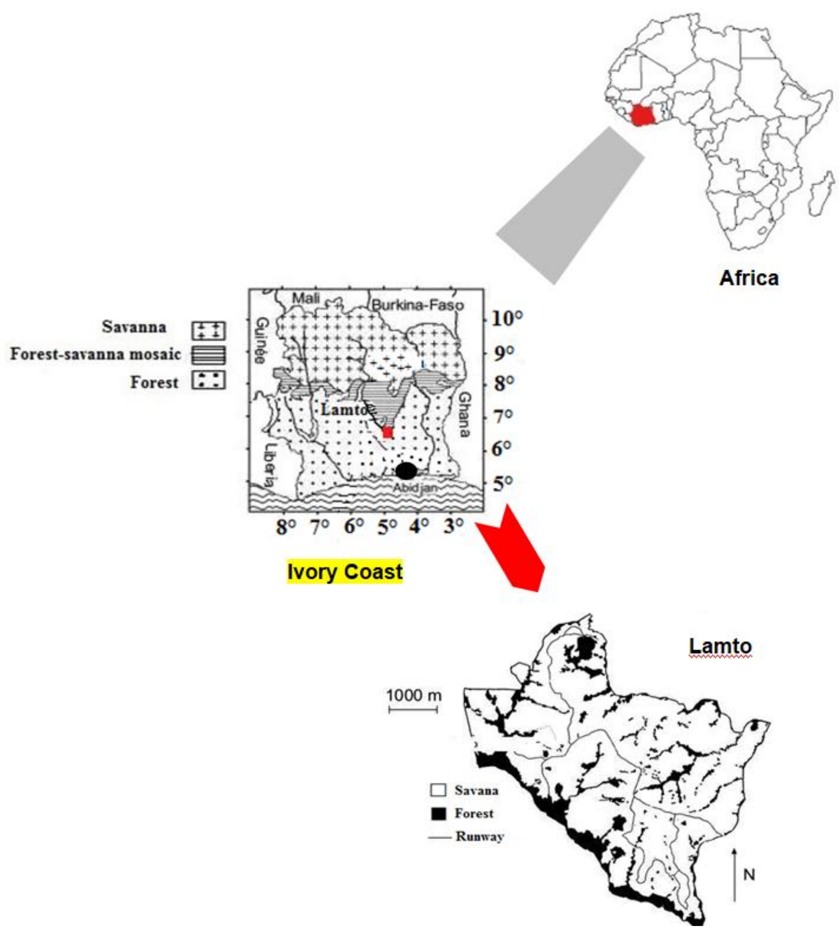


Fig. 1. Location of Lamto reserve [32,33]

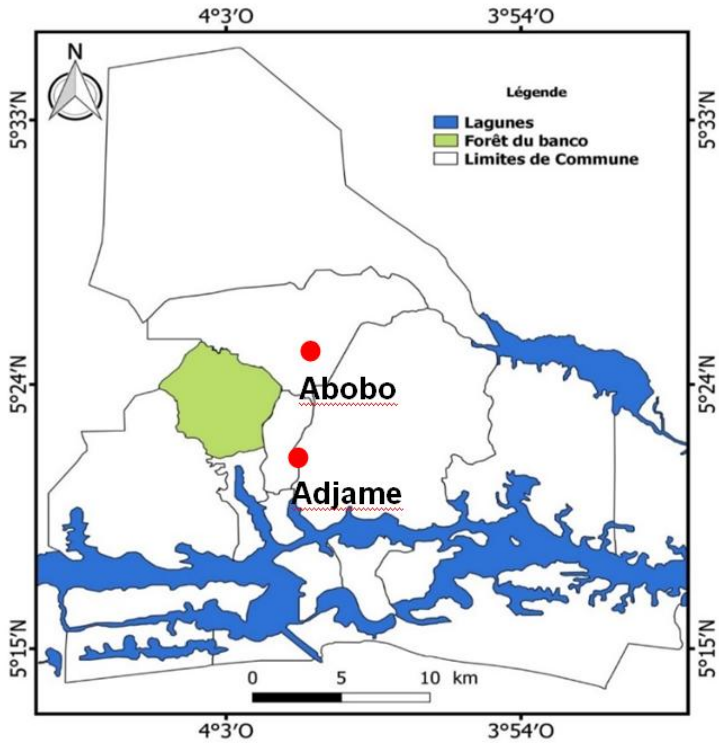


**Fig. 2. Sampling sites location in Abidjan city (Ivory Coast)**

Four (4) wastewater stocks were collected from both study sites. These are the following wastewaters: blue wastewater of cotton basins (BWB), blue wastewater of cotton jeans (BWJ), red wastewater of leather jackets (RWJ), and red wastewater of leather bags (RWB).
The collected wastewaters are sent to the laboratory for dynamic studies.

## 2.4 Dynamic Wastewater Treatment

The two synthetic dyes (blue and red) used locally by the artisans in artisanal dyes plants, were purchased commercially. The dyes are put into solution (Fig. 4) and then their maximum absorption wavelengths ($\lambda_{max}$) were determined by scanning with the UV/Vis spectrophotometer (WFJ-752, Beijing, China).The maximum absorption wavelength of the blue dye is $\lambda_{max}$ = 620 nm and that of the red dye is $\lambda_{max}$ = 510 nm.
The dynamic studies were performed through a glass column with an internal diameter of 8 cm and 22 cm height (Fig. 4). A peristaltic pump was used to pump the wastewater at a desired constant rate, from a 25 L feed recipient. A thin glass wool was used as filter to avoid any loss of adsorbent. The effluent was collected at the column outlet in a glass jar.

For each wastewater stock collected, the pH is adjusted to 7 and then placed in the column feed vessel for treatment. Our previous work [35] allowed us to set the different parameters of the column. Thus, the feed rate Q is 0.07 L min$^{-1}$, the mass m of the deactivated lichen bed is 100 g corresponding to a bed height H of 8 cm. The treated solution, called effluent, is recovered at the exit of the column. The optical density of the effluent is read at 5 min time intervals using an UV/visible spectrophotometer (WFJ-752, Beijing, China). The concentration $C_t$ at time t is obtained using the calibration curve of the target dye. The calibration curve is obtained after dissolving the commercial dye in distilled water.

The amount $q_{ads}$ (mg.g$^{-1}$) of adsorbed dye per gram of lichen is determined using the relationship below [36].

$$q_{ads} = \frac{DC_0 \int_0^{t_s} \left(1 - \frac{C_t}{C_0}\right) dt}{m} \quad (1)$$

With:
$C_0$: Initial concentration (mg.L$^{-1}$) of dye in wastewater

$C_t$: Concentration (mg.L$^{-1}$) at time t of dye in the effluent;
D: Column feed rate (L.min$^{-1}$);
$t_s$: saturation time;
m: Mass (g) of deactivated lichens.
The integral in equation (1) is the area above the breakthrough curve from $\frac{C_t}{C_0}=0$ to $\frac{C_t}{C_0}=1$

The graph $\frac{C_t}{C_0}=g(t)$ is called breakthrough curve of the column.
The removal percentage (R) is given by the following relationship [36]:

$$R=\frac{\int_0^{t_s}\left(1-\frac{C_t}{C_0}\right)dt}{t_s}\times 100 \qquad (2)$$

R and $q_{ads}$ are obtained after integration between t=0 and t=$t_s$ of the polynomial regression of the following function

$$1-\frac{C_t}{C_0}=f(t) \qquad (3)$$

The time corresponding to an effluent outlet with less than 5% of $C_0$ is called breakthrough time tb. It is fixed according to the treatment objective. In this study, it is fixed at 1% of $C_0$ for given dye. At this breakthrough time, the bed is not fully used. The equivalent length of unused bed (LUB) is obtained by equation (4) [37]

$$LUB=H\left(1-\frac{t_b}{t_s}\right) \qquad (4)$$


**Fig. 3. Solutions of the blue dye and the red dye before their use**

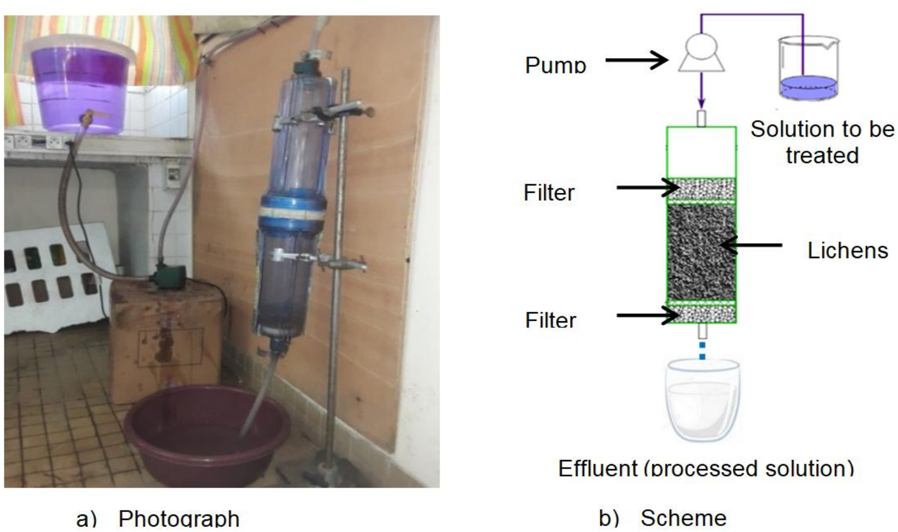


**Fig. 4. Photograph and scheme of the experimental column [34]**

## 2.5 Data Processing

Data are obtained after 3 individual trials. Data homogeneity analyses, polynomial regressions and graphs were performed using STATISTICA 8.0.360

## 3. RESULTS AND DISCUSSION

Fig. 5 shows the breakthrough curve obtained at the end of the treatment of blue wastewater of cotton basins (BWB) that introduced in the column with an initial concentration $C_0$ = 35 mg.L$^{-1}$. The first output of the treated solution at the desired concentration corresponds to a breakthrough time $t_b$ = 7 h 30 min. From $t_b$, the dye gradually saturates the bed of deactivated lichens in the column from top to bottom until complete saturation after the time $t_s$ = 29 h. The column is then in equilibrium and no longer retains dye.

Blue jeans dyeing water (BWJ) is treated from an initial concentration $C_0$ = 24 mg.L$^{-1}$, the evolution of its breakthrough curve is given in Fig. 6.

This figure reveals that during the BWJ treatment, the breakthrough time $t_b$ is 15 h and the saturation time $t_s$ was obtained after 33 h of operation of the column.

The red wastewater from leather jackets (RWJ) was treated from an initial concentration $C_0$ = 34 mg.L$^{-1}$. The evolution of the breakthrough curve is shown in Fig. 7.
The breakthrough time $t_b$ for this RWJ treatment is 8 h and the saturation time $t_s$ was obtained after 27 h.

Finally, the breakthrough curve which result from the treatment of wastewater from a red wastewater of leather bags (RWB) at $C_0$ = 46 mg.L$^{-1}$ is presented in Fig. 8.

The examination of Fig. 8 resulting from the treatment of the wastewater RWB, shows that the breakthrough time is recorded after 5 h and that of the saturation of the sites of the fixed bed of deactivated lichens is observed at $t_s$ = 23 h 30 min.

The volume of the effluent $V_{eff}$ at $t_b$, the unused bed length (LUB) and the temporal parameters derived from the various breakthrough curves of the wastewater from artisanal dyeing plants are recorded in Table 1.

The results recorded in Table 1 show that the breakthrough time is a function of the initial concentration of the dye in the considered wastewater. The higher the initial concentration $C_0$, the lower $t_b$ and $V_{eff}$ are, resulting in rapid saturation of the fixed bed of deactivated lichens. However, the length of unused bed (LUB) at $t_b$ increases with $C_0$. This result can be explained by the availability of active sites in the adsorbent bed. For close initial concentrations (BWB and RWJ), significantly close column parameters are observed. The nature of the dyed objects does not influence the column parameters. Thus, the removal of the blue dye in BWJ, from $C_0$ (23 mg.L$^{-1}$) to 1% $C_0$ is done during 15h and allows to obtain 63 L of purified effluent but by soliciting a great length of the fixed bed of deactivated lichens. This result suggests that for a treatment objective of BWJ up to the 1% $C_0$ threshold, it is necessary to renew the deactivated lichen bed after 15 h of column operation.

The removal percentage reflects the ratio of the mass of dye adsorbed by the bed (between $t_0$ and $t_s$) to the total mass of dye that passed through the column (between $t_0$ and $t_s$). For each wastewater, the amount of dye adsorbed, and the removal rate are recorded in Table 2.

Regardless of the dye color and the nature of the object, residual water dyes are significantly removed through the deactivated lichen fixed bed column. The amount adsorbed at equilibrium and the rate of removal increase as the initial concentration of the dye in the residual water increases. This result on the influence of the initial concentration was observed during our recent work [35] on the adsorption of crystal violet and methyl red dyes in a fixed bed column. Indeed, the results of their work showed that the residence time and the saturation time decreased with the increase of the initial dye concentration. The results of the study are also in agreement with the work of Marzbali et al. [38] on the adsorption of tetracycline on a fixed bed of mesoporous activated carbon. The authors linked this phenomenon to the increased concentration gradient that results in a higher driving force for rapid mass transfer. The best elimination rate is obtained with the treatment of RWB. The high initial concentrations cause a rapid displacement of the mass transfer zone (MTZ) and a denser occupation of the active sites of the adsorbent bed. The effect of gravity could on the one hand explain this rapid movement but also the competition around the active sites of deactivated lichens could lead to a more compact occupation [39, 40].

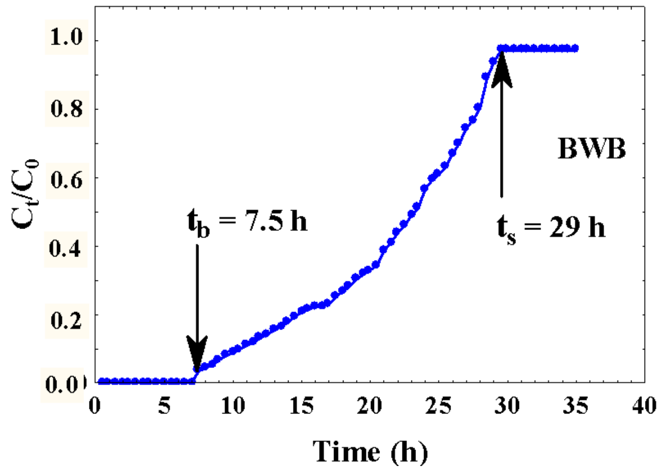


**Fig. 5. Breakthrough curve relating to the treatment of BWB**
*Q = 0.07 L min$^{-1}$; m = 100 g; H = 8 cm; d =125 μm, pH = 7*

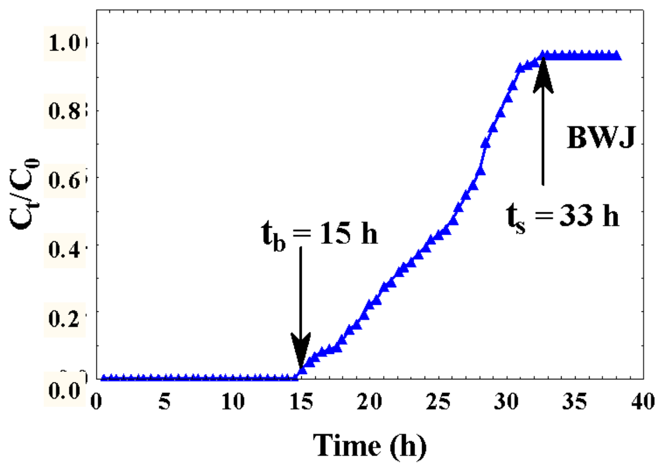


**Fig. 6. Breakthrough curve relating to the treatment of BWJ**
*Q = 0.07 L min$^{-1}$; m = 100 g; H = 8 cm; d =125μm, pH = 7*

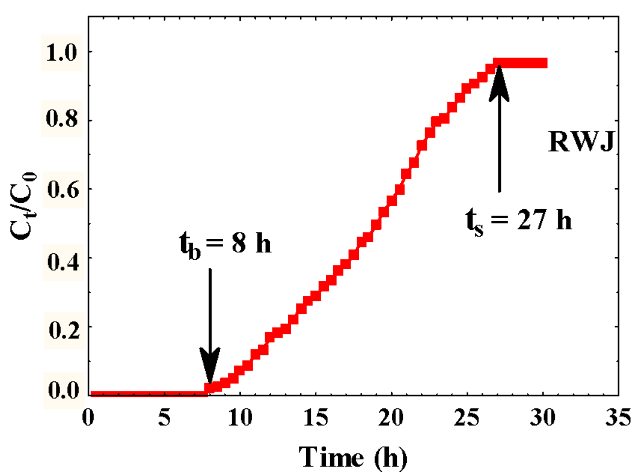


**Fig. 7. Breakthrough curve relating to the treatment of RWJ**
*Q = 0.07 L min$^{-1}$; m = 100 g; H = 8 cm; d =125μm, pH = 7*

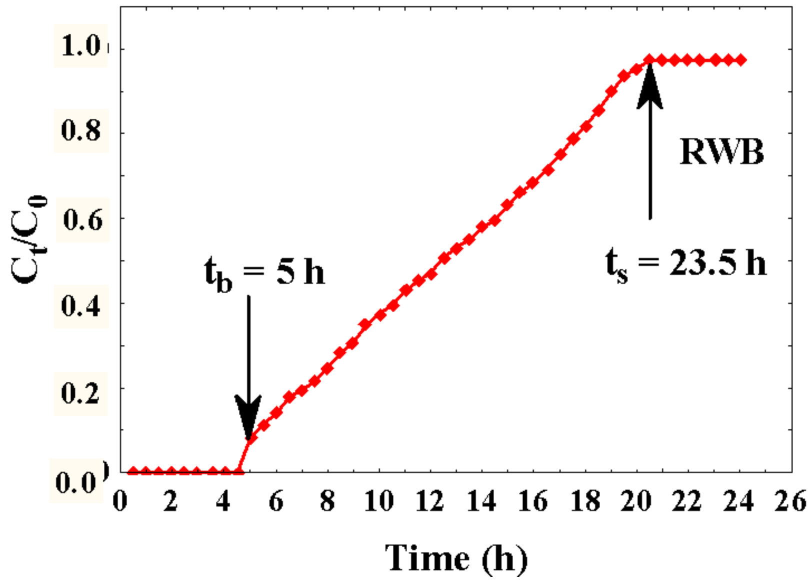


**Fig. 8. Breakthrough curve relating to the treatment of RWB**
*Q = 0.07 L min$^{-1}$; m = 100 g; H = 8 cm; d =125μm, pH = 7*

**Table 1. Some parameters of the column for each treatment**

| | $C_0$ (mg.L$^{-1}$) | $t_b$ (h) | $t_s$ (h) | $V_{eff}$ (L) | LUB (cm) |
|---|---|---|---|---|---|
| BWB | 35 | 7.5 | 29 | 31.5 | 5.93 |
| BWJ | 23 | 15 | 33 | 63 | 4.36 |
| RWJ | 34 | 8 | 27 | 33.6 | 5.63 |
| RWB | 46 | 5 | 23.5 | 21 | 6.30 |

**Table 2. Quantities of adsorbed dye and percentage of removal**

| | $C_0$ (mg.L$^{-1}$) | $q_{ads}$ (mg.g$^{-1}$) | R(%) |
|---|---|---|---|
| BWB | 35 | 41.449 | 0.972 |
| BWJ | 23 | 30.547 | 0.958 |
| RWJ | 34 | 37.467 | 0.972 |
| RWB | 46 | 44.444 | 0.979 |

## 4. CONCLUSION

The present study examined the performance of deactivated lichens as adsorbents for the dynamic removal of dyes from wastewater of artisanal dyeing. The column developed in the laboratory in our previous work had shown good theoretical performance for the removal of dyes in aqueous solution. This real-life study shows that it is possible to effectively remove dyes from wastewater regardless of the nature of the dyed objects. It is possible to eliminate or considerably reduce the concentrations of dyes in wastewater, by sizing fixed bed columns of deactivated lichens, in order to install them at the outlets of artisanal dyeing units while ensuring flow by gravity.

## COMPETING INTERESTS

Authors have declared that no competing interests exist.

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