https://doi.org/10.33261/jaaru.2021.28.2.001 *Association of Arab Universities Journal of Engineering Sciences (2021)28(2):01–07*

Association of Arab Universities Journal of Engineering Sciences
مجلة اتحاد الجامعات العربية للدراسات والبحوث الهندسية

Removal of Congo Red from Aqueous Solution by Circulating Fluidized Bed(CFB)

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Published online: 30 June 2021.

Abstract— In this study circulating fluidized bed was adopted to remove of Congo Red from wastewater using Eichhornia crassipes as a adsorbent. Solution flow rate(6,12 and 18)l/hr, bed height(2,4 and 6) cm and Congo Red initaial concentration (10,25 and 50)mg/l were examined in experiments to show their effects on breakthrough curves and time required to reach the adsorbent to fully saturated curve. The mass transfer coefficient "K_L"decreased with decreasing the liquid flow rate. The minimum fluidization velocities of bed found equal to 1.6, 2, 2.5 mm/s for heights of 2, 4,6 cm respectively. The increasing of the bed height will increase the contact time of the solute in the bed, and these improve the solute removal efficiency. the increasing in flow rate and initial concentration will increase the mass transfer rate.

Keywords— Congo Red dye, Eichhornia crassipes. Circulating Fluidized Bed, Minimum fluidization velocity, Mass transfer

1. Introduction

Fresh water is vital to human life and economic well-being, and societies extract vast quantities of water from rivers, lakes, wetlands, and underground aquifers but most of these freshwater sources are polluted by different chemicals discharged from industries. need for fresh water has long caused to overlook equally vital benefits of water that remains in streams to sustain healthy freshwater habitats [1][33]. Water is being polluted largely due to increased industrialization. Dyestuff manufacturing industries and many other dye and pigment-using industries such as rubber, paper, textile, plastics, and cosmetics generate highly colored and toxic effluent. During dyeing processes 50% of dye is lost. The release of these dyes containing waste water into rivers, seas has adverse effects on the people who use such contaminated water for living purposes like washing, bathing and drinking. Even a small quantity of dye (1.0 mg/L) in water is detectable and objectionable. Besides this dyes can influence aquatic plants by reducing transmission of sunlight into water.

Dyes are mutagenic, dangerous and cause harmful effects to humans[21], [22], [31]. In up-to-date data, more than 100,000 commercially available dyes with the rate of 7×10^5 tons per year are produced and usually two percent of the products are discharged into water systems as waste [6]. Most of the dyes are harmful when brought in contact with living tissues for a long time. The discharge of these dyes to the river stream without proper treatment causes irremediable damage to the crops and living beings, both aquatic and terrestrial [30][7]. Consequently, removal of dyes from such wastewaters is an important environmental problem and complete dye removal is needed because dyes are visible even at low concentration [26]. Anthraquinone-based dyes are the most resistant to degradation due to their fused aromatic structures, which remain coloured for a longer period of time [10].

Different procedures were developed for removal of organic dyes from wastewater. These procedures include electrocoagulation[18], flotation[3], chemical oxidation[10], filtration[12], ion exchange[16], ozonation[16], membrane separation and microbial degradation[16]. Adsorption is found a good way to treat industrial waste effluents, it has significant advantages in comparison with conventional methods, especially from economical and environmental viewpoints [13][1][8]. Natural materials or the wastes, by-products of industries or synthetically prepared materials[5], which are cheap and can be used as such or after some minor treatment as adsorbents, are generally called low-cost adsorbents [19][35][2].So many researchers search for cost effective and efficient alternative materials such as tree fern[38], teak tree bark powder[26], orange peel[10], banana pith[36], date pits[20] and tea waste [29][30]. The most effective and optimized utilization of a biomass demands a detailed understanding of the binding mechanism [37]. Eichhornia crassipes is water hyacinth, found in large amounts around the fields of irrigations and in the fresh water bodies through the year in tropical and subtropical countries including Iraq [39]. The potential of using E. crassipes as alive or a dead biomass to remove dyes from solutions was recently investigated. The results showed that it is a promising cheap biosorbent source for dyes [40][34][16]. In recent years the circulated fluidized bed reactor has received extensive consideration and varied usage in the treatment of wastewater owning to its many benefits such as high contact between gas, liquid, and solid phase producing high mass transfer and high reaction rate, it could reduce the operational problems occur in packed-bed like bed clogging and high pressure drop, it is simple, highly efficient, economical operation compared to other reactor configurations and stable [6][15].This paper aims to study the process of separation and purification wastewater through the removal of Congo red using Water hyacinth (Eichhornia Crassipes)by circulated fluidized bed.

2. Materials preparation

2.1 *Adsorbate*

A Congo Red dye obtained from Sigma Aldrich with 99.99% purity was dissolved in distilled water to prepare (10, 25 and 50)mg/L concentration of CR used in the experiment work. 0.1 M of hydrochloric acid or sodium hydroxide base was added to regulate the solution pH to the desired value. Congo red dye is the sodium salt of 3,3′{[1,1’-Biphenyl]-4,4′-diylbis(azo)}bis(4-amino-1-naphthalene sulfonic acid disodium salt) has a molecular weight of 696.66 g/mol with a formula ($C_{32}H_{22}N_6Na_2O_6S_2$), and their chemical composition structure was shown in **Figure 1**.

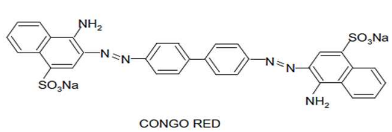

Figure 1: Chemical formula of Congo Red

2.2 *Biomass preparation*

Water hyacinth (Eichhornia crassipes) was prepared as follow:

1. Collected from Tiger River in Aldora district of Baghdad.
2. Washed several times with distilled water in order to make it clean and the washed water hyacinth were left under the sunlight for five days to dry,then oven dried at 105°C for 2 hours to ensure that the sample dried completely.
3. Water hyacinth were separated into stems , leaves and roots.
4. The samples grinded by electric grinder and seived to different particle size as show in **Figure 2.**

Figure 2: Water hyacinth (Eichhornia crassipes)

2.3 Circulating fluidized bed *(*CFB)

G-L-S fluidization is an operation involving a bed of suspended particles in gas and liquid media. This occurs due to a net drag force of gas and liquid in the flow direction on the particles.The solid particles are circulated between the riser and the downer. **Figure 3** show the schematic diagram for CFB column used in removal of CR from wastewater and **Table 1** represents the major parameters examined.

Table 1: Parameters range tested in CFB

Parameters	Range	Purpose
Liquid flow rate (L/hr)	6, 12 and 18	To determine optimum flow rate and to find the effect of flow rate on breakthrough curves .
Bed height, cm	2, 4 and 6	To study the effect of EC bed height on Breakthrough curves.
Initial concentration, mg/L	10,25 and 50	To study the effect of CR initial concentration on breakthrough curves
The condition of this work were pH 6, room temperature, flow rate of air 250 cm^3/min, particle size 650 µm and 6 hr .		

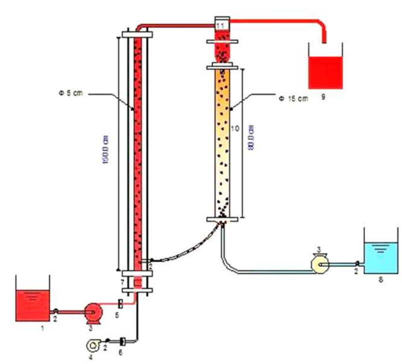

Figure 3: GLSCFB schematic diagram (1: Feeding tank, 2: Valve, 3: Pump, 4: Air compressor, 5: Flow meter of liquid, 6: Gas flow meter, 7: Distributer, 8: Distilled water for the downer 9: Treated wastewater, 10: Downer column, 11: Solid-liquid separator, 12: Riser column.)

3. Results and Discussion

3.1 Minimum fluidization velocity (Ulmf)

The minimum fluidization velocity (Ulmf) was determined experimentally by measuring the pressure drop through the bed of adsorbent particles. The values of minimum fluidized velocity are 0.0016, 0.002 and 0.0025 m/s for the different bed height 2, 4 and 6 cm respectively. Ulmf in this study has been obtained as displayed in **Figure 4**, it is noticed that initial static bed height Hs affect Umf, when Hs increase, pressure drop increase then Umf increase. This owing to the fact that bed fluidization achieved when equaling the upward drag and inertial forces applied by the fluids on the particles with the bed buoyant weight, the initial static bed height (Hs) only effect on the Umf as expected[6].

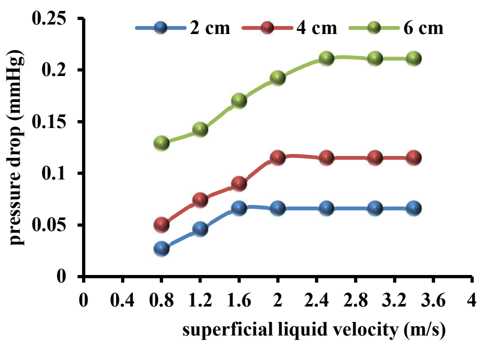

Figure 4: Pressure drop Variant with U_L for different H_s values

3.2 Bed expansion

It is important to be able to establish the relationship between the superficial liquid velocity (U) and the bed voidage (ε)[15]. The bed voidage of fluidized EC biomass was found experimentally using Equation (1) and compared with the theoretical value that calculated using Equation (2). Experimental and theoretical voidage (ε) values are shown in **Table 2**

$$\varepsilon = \frac{V_\varepsilon}{V_b} = \frac{V_b - V_p}{V_b} = 1 - \frac{V_p}{V_b} = 1 - \frac{m_p}{\rho_p . V_b} = 1 - \frac{m_p}{\rho_p . A . H} \quad (1)$$

$$\frac{U}{Ui} = \varepsilon^n \quad (2)$$

Table 2: Values of n, theoretical and experimental voidage for particle size ranges.

Particle size (mm)	Uri R (m/s)	index (n)	ε Eq.(2)	ε Eq.(1)
0.7	0.022	3.87	0.71	0.93

3.3 Breakthrough Curves

By plotting Ce/Ci versus time for CR, the breakthrough curves for contaminant was obtained.

3.3.1 Effect of liquid flow rate:

The fluid flow rate is a major parameter in the design of biosorption column especially fluidized bed reactor due to its effect on the contact time between the particles and dye solution[15]. When the flow rate increases the breakthrough curves become steeper. At a high flow rate, the adsorbate solution leaves the column before the complete equilibrium occurs due to the reduction in the contact time as shown clearly in figure below **Figure5**. The increasing of flow rate will cause a reduction in the thickness of the surface film which considered the resistance for the mass transfer. As a result of that increasing in flow rate will increases the mass transfer rate[12]. These results agree with that obtained by [15], [34] .

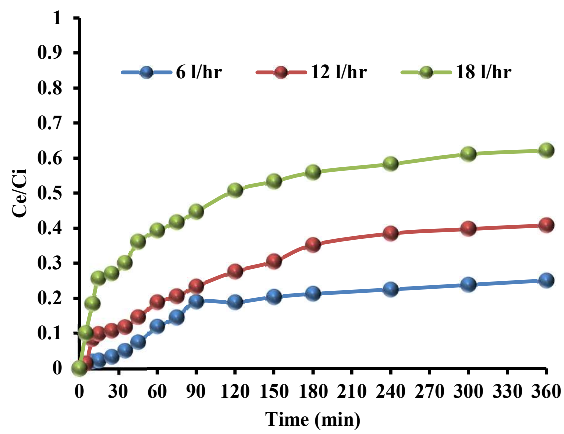

Figure 5: Effect of flow rate on removal of CR at initial conditions (8 cm), pH 6, 10 mg/L, and air flow rate 250 cc/min.

3.3.2 Effect of bed height:

It is obvious that when the bed height increases, the time to reach breakpoint increase. The increasing of bed height will provide an extra surface area for the adsorption process. This shows that at low bed height the effluent adsorbate concentration ratio increases more rapidly thanfor a higher bed height. Furthermore, in a low bed height, the bed is saturated in less time. A lower bed height corresponds to lesser amount of adsorbent and weak capacity for the bed to adsorb dye from solution. At constant flowrate, the increasing of the bed height will increase the contact time of the solute in the bed, and these improve the solute removal efficiency[17]. These results agree with that obtained by [16], [27].

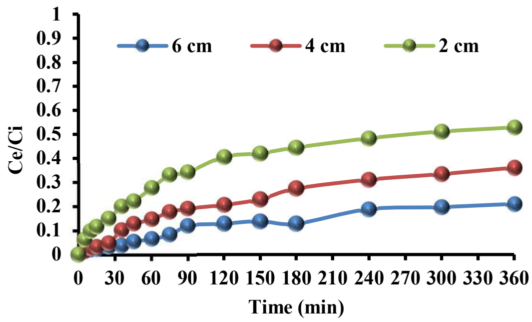

Figure 6: Effect of bed height at initial conditions pH 6, 10 mg/l, 6 L/hr, and air flow rate 250 cc/min on removal of CR.

3.3.3 Effect of initial concentration:

The effect of different initial CR dye concentrations (10, 25 and 50 ppm) on the removal efficiency were examined at constant water, air flow rate, bed height and the results were plotted in **Figure7** in terms of C/Co versus time. From this figure, it is obvious that saturation time decreases with increasing the initial concentration and there is inverse relation between the breakpoint and initial concentration. Low initial solute concentration make the saturation time of diffusion rate longer. As the CR influent concentration increases, the adsorption capacity also increase. This was attributed to a high concentration difference (driving force for adsorption) between liquid and solid phase also, this will increase solute mass transfer rate to attach adsorbent active free sites. If the initial CR concentration is high, faster bed saturation occurred and the slope of the breakthrough curve was high.

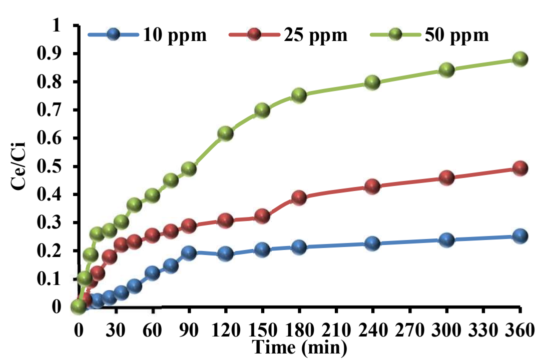

Figure 7: Effect of initial concentrations on the removal of CR at initial conditions pH 6,(Hs = 4 cm), 6 L/hr and 250 cc/min.

a) Mass transfer

Several authors proposed generalized correlation to predict the mass transfer coefficient (K_L) in continuous systems. Fluidized bed reactor offers high available surface area, since there is no contact between particles, and contact of the entire surface with the wastewater. Segregation of the adsorbent particles occurs in fluidized bed adsorber, so that nearly its fully capacity can be realized with this form of operation. Liquid solid mass transfer coefficient is one of the most important parameters for the design, scale-up and performance characterization of columns[27]. The three-phase circulating fluidized bed can increase the heat and mass transfer coefficients at a higher range of superficial liquid velocity, and minimizing the dead zone in the reacting or contacting system by means of the circulating fluidization mode[15]. Various studies have been done to determine the mass transfer in a bed and many correlations have been suggested to predict solid-liquid mass transfer coefficients for different systems, can use equation below to predicted of K_L values[16].

$$D_m = 2.74 * 10^{-9}\,(Mwt)^{-\frac{1}{3}} \tag{3}$$

$$Sh_z = 2.27\, Sc^{0.33}\, Re_{zl}^{0.494}\, Re_{zg}^{0.178}\, Ga^{-0.28} \tag{4}$$

$$K_l = \frac{Sh_z \,.\, D_m}{d_p} \tag{5}$$

When the liquid flow rate increased from 6 to 18 L/hr, K_L values were increased from 3.63E-06 to 4.19E-06 for CR dye. The reason behind this action is that the change in flow rate will affect the film diffusion, but not the intra-particle diffusion. The higher the flow rate the smaller the film resistance to mass transfer and larger K_L values were tabulated in **Table 3**.

Table 3: Mass transfer coefficient K_L values of CR at different operating conditions.

contaminant	Ul (m/sec)	Dp (m)	Re_p	U_g (m/s)	k_L (m/s)
CR	0.0024	0.00065	1.95	0.003	3.63E-06
	0.0024	0.00065	1.95	0.0022	3.63E-06
	0.0024	0.00065	1.95	0.0022	3.63E-06
	0.0028	0.00065	2.275	0.0022	3.92E-06
	0.0028	0.00065	2.275	0.0022	3.92E-06
	0.0028	0.00065	2.275	0.0022	3.92E-06
	0.0032	0.00065	2.6	0.0022	4.19E-06
	0.0032	0.00065	2.6	0.0022	4.19E-06
	0.0032	0.00065	2.6	0.0022	4.19E-06

5- Conclusions

Eichhornia crassipes was found to be feasible media for use as a bed column for the removal of CR from contaminated water.A fluidized -bed column was used to investigate the sorption of CR studied and it was depended on feed flow rate, initial CR concentration and bed height of adsorbent. The minimum fluidization velocity of bed were found equal to 1.6, 2, 2.5 mm/s respectively for bed height of 2, 4, 6 cm respectively. The variation of liquid superficial velocity from 0.0024 to 0.0032 m/s resulted an obvious depletion in sorption removal efficiency. When the flow rate increases the breakthrough curves become steeper. The variation of bed height from 2 to 6 cm for CR resulted in an obvious increase in biosorption capacity. Increasing initial contaminant concentration cause an increase in biosorption capacity. The mass transfer coefficient "KL" increased with increasing the liquid flow rate.

Nomenclature

Abbreviation	Description
CR	Congo Red
EC	Eichhornia crassipes
dp	Particle diameter
GLSFB	Gas-Liquis-Solid Fluidized Bed
Hs	Bed height
Pd	Pressure drop
PIS	Pistachio shell
D_m	Diffusivity coefficient
Sh_z	Modified Sherwood number, K_L. ŋZ/D_m
Re_{zg}	Modified Reynolds number for gas, Ug ρ_g/a μ_g
TEC	Tetracyclin
Ug	Gas velocity
UL	Liquid velocity
Ulmf	Minimum fluidization velocity
U_i	The settling velocity of a particle
ε	Bed voidage
V_b	volume of wastewater solution
V_p	Volume of particle
V_ε	Void volume
m_p	Mass of particles
ρ_p	Real density of particles
H	Bed height

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إزالة الكونغو الأحمر (CR) من محلول مائي عن طريق تعميم السرير المميعة (CFB)

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نشر في: 30 حزيران 2021

الخلاصة – في هذه الدراسة ، تم تبني طبقة مميعة متداولة لإزالة الكونغو الأحمر من مياه الصرف الصحي باستخدام crassipes Eichhornia كمواد ماصة. معدل تدفق محلول Congo Red ، ارتفاع السرير وتركيز الكونغو الأحمر هي العوامل التي تم فحصها في التجارب لإظهار تأثيرها على منحنى الاختراق والوقت المطلوبة للوصول إلى الممتزات إلى منحنى مشبع تماما. انخفض معامل نقل الكتلة "KL" مع انخفاض معدل التدفق السائل. تم العثور على الحد الأدنى من سرعة التميع في السرير مساوياً 1.6 ، 2 ، 2.5 مم / ثانية على التوالي لارتفاع السرير 2 ، 4،6 سم على التوالي. تم استخدام ظروف التشغيل المختلفة. ارتفاع السرير الثابت (2 ، 4 و 6 سم) من الكتلة الحيوية لأنابيب Eichhornia لكونغو ريد ؛ معدل التدفق السائل (6 ، 12 18 a لتر / ساعة) ؛ وتركيزات الملوثات الأولية (10 و 25 و 50 ملغم / لتر). ستؤدي زيادة ارتفاع السرير إلى زيادة وقت التلامس في المذاب في السرير، مما يؤدي إلى تحسين كفاءة إزالة المذاب. زيادة معدل التدفق والتركيز الأولي يزيد من معدل نقل الكتلة. الكلمات الرئيسية – تكتب 3-5 كلمات رئيسية وتكون منفصلة باستخدام "، ".

الكلمات الرئيسية – الكونغو الأحمر ، السرير المميع الدائري ، الحد الأدنى لسرعة التميع ، ارتفاع السرير ، زهرة النيل.