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Equilibrium kinetic and thermodynamic studies of Cr (VI) ions removal by *Pleurotus mutilus* biomass in torus reactor

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ABSTRACT/RESUME

Abstract: *The aim of this research was the kinetic equilibrium and thermodynamic studies of hexavalent chromium removal from aqueous solution by Pleurotus mutilus biomass, in torus reactor. Experiments were carried out in laboratory scale. The effect of sorbent amount, pH and initial concentration of hexavalent chromium on biomass removal capacity were determined after 3 hours of treatment. The optimum conditions were found to be a chromium initial concentration of 100 mg/L, pH 5.4 and biosorbent dose 2 g/L; in these conditions the biosorption capacity reaches 29.46 mg/g. The modeling study showed that the pseudo-first-order model fitted well the adsorption kinetics. The kinetics of diffusion has been analysed, it confirms that the external mass transfer controls the biosorption process. The adsorption equilibrium data could be described by Langmuir, Temkin and Dubinin-Radushkevich isotherms models. Desorption can recover biomass and concentrated metal in a reduced volume. The recovered metal can be reused or treated by physical or chemical methods. The thermodynamic study indicates that biosorption was endothermic and spontaneous in nature. This biomass was destined to incineration, valorized as a potential biosorbent for heavy metals with no treatment indeed. The torus reactor has demonstrated interesting results for its first application in biosorption process.*

I. Introduction

Heavy metals are largely used in industrial manufacturing processes and surface treatement. They represent a big problem for diposal in wastewater. Among these heavy metals, mention is made of hexavalent chromium Cr(VI) which is widely used in metal finishing, paint, paper industries,electrical and electronics equipements and catalysis of chemical manufacturing[1].

Much of the Cr (VI) released in wastewater comes from the leather industry, the high concentration of Cr (VI) found in sludge is due to the presence of chromium salts in the tanning process, it can reach 2075 mg / L in tanning effluents [2]. It is known that chromium exists mainly in two oxidation state: Cr(III) and Cr(VI) [3], [4] this last is more toxic, affecting public health and causing environmental pollution [5]. Human activities are the principle cause of elevated levels of Cr(VI) in the aquatic environment, groundwater and spring waters [3], [4]. Chromium is carcinogen, it is harmful even at low concentrations, it has been considered by the International Agency for Research on Cancer as a group I human carcinogen [6]. The World Health Organization (WHO) has been authorized a chromium level in surface and drinking waters respectively of about 0.1 and 0.05 mg/L [4]. A larger quantities of chromium discharged in industrial wastewater, as a concentrations varied

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between 5 and 220 mg/L, which contaminates soil and water [5], [7].

Therefore, the treatment of chromium before its discharge into the wastewater becomes necessary to have the permitted level of chromium concentration. Hence, numerous methods have been used such as chemical and electrochemical precipitation, reduction, ion exchange, evaporation, reverse osmosis and membrane separation. These methods are very expensive and ineffective when metal concentration is less than 100 mg/L [8], [9]. However, the choice of the right method is based on cost. Therefore, microbial biomass and different lignocellulosic materials are selected and studied among several types of adsorbents. These natural biosorbents, known by their abundance in nature and their low prices compared to other [10].

The biosorption process is fundamentally based on solid liquid contact. As a result, different reactors and bioreactors were developed by authors in order to increase heavy metals uptake, Such as, stirred-tank and continuous stirred tank, fixed packed-bed, fluidized-bed and multiple-bed, airlifts and bubble column [10]. Batch stirred reactors and columns are largely used in Cr (VI) removal. The column used in the biosorption experiments has a limitation of mass transfer caused by the preferential paths of the effluent through the compacted biomass bed. The torus reactor was used in biosorption of Cr (VI) dead biomass of *Pleurotus mutilus* by Alouache et al [11]. It was first proposed in polymerization of olefin. It has potential applications in biochemical reaction and viscous liquids process. According to previous works [12]–[14], the torus reactor has several advantages, such as heterogeneous solution can be avoided under a high Reynolds number, by keeping all particles in suspension. This reactor is known for its easy scale up and geometry design, which prevents vortex formation and dead volume, encountered in stirred batch reactors.
The aim of the present work is the study of hexavalent chromium removal from aqueous solutions by *Pleurotus mutilus* biomass in torus reactor. For this purpose, the effect of the sorbent dose, initial pH, initial chromium concentration were studied. Thermodynamic, Kinetic models and equilibrium isotherm were investigated in order to identify the adsorption feasibility and capacity, and rate-controlling step of chromium biosorption on *Pleurotus mutilus*.

II. Materials and methods

II.1. Biomass preparation and characterization

The fungal waste (*Pleurotus mutilus* biomass), was obtained after *Pleuromutiline* extraction. It was provided by the SAIDAL antibiotic complex of Medea (Algeria). Generally, this waste was destined for incineration. However it can be valorized for wastewater treatment. It was thoroughly washed and dried for 24 h at 50°C. The dried residue was manually milled in a mortar, sieved and stored before adsorption experiments. The chemical and physical properties of this biosorbent are presented in Table 1 according to Khitous et al [11], [15].

Table 1. *The physical and chemical characteristics of Pleurotus mutilus biomass*

Particle size	d_p	ρ_{app}	S_p	Zeta
(µm)	(µm)	(g/cm^3)	(m^2/g)	(V)
160-200	180	0,43	0,13	-0,062

II.2. Hexavalent Chromium solutions preparation and analysis

The stock solution (1000 mg/L) of Cr (VI), used for all experiments was prepared by adding the required amount of ($K_2Cr_2O_7$) in distilled water**.** Hexavalent chromium solutions were prepared by successive dilution in distilled water for experimental tests. The pH of solutions was adjusted using NaOH and/or HCl solutions (0.1 mol/L). Sampling was done at a specific time followed by filtration in order to remove biomass from solution.
The residual concentration of Cr (VI) was measured by titration with 1,5diphenylcarbazide (DPC) under acidic conditions, and analyzed at 540 nm with UV-visible spectrophotometer (OPTIZEN 1412 V) [4], [7], [8], [16]. Therefore, the Cr(VI) uptake on biomass was determined by using the following equation:

$$q_e = \frac{(C_0 - C_e)V}{m} \qquad \textbf{(1)}$$

Where, q_e is the chromium uptake at equilibrium (mg/g), C_e and C_0 are the equilibrium and the initial Cr(VI) concentrations (mg/L) respectively in the aqueous solution; V is the volume of the Cr(VI) solution (L), m is the amount of biosorbent (g), the percentage of Cr (VI) removal was determined using equation(2) as follows:

$$Removal\% = \frac{C_0 - C_e}{C_0} \times 100 \qquad \textbf{(2)}$$

II.3. Biosorption experiments in torus reactor

The torus reactor is presented in Figure 1. It was made from transparent pipe with an internal diameter D_t, a circumference length L_t, and circumference diameter R_t with total working volume V_R. A marine screw impeller is used for mixing, the stirring is driven by a variable speed motor (IKA-WERK RW20) providing a speed of 800 rpm, sufficient to have homogeneous solution at which the torus biosorption experiments were investigated.

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The torus reactor characteristics are given in Table 2. This reactor is similar to those used by authors [14,17–19].

The biomass was introduced into the reactor until the desired concentration was reached.

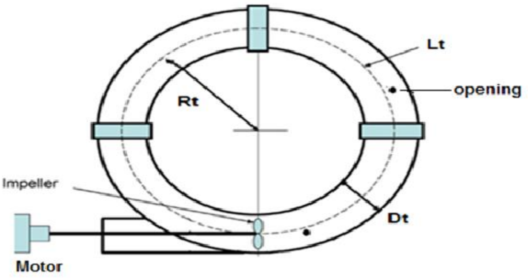

Figure 1. *Horizontal view of the torus reactor*

The adequate amount of stock solution was added and the total volume of 2.9L of the reactor was completed by distilled water. Samples were taken from a special opening in the top of the reactor for different time intervals, then filtered and analyzed for remaining Cr (VI) concentrations.

Table 2. *Characteristics of the reactor*

V_R	D_t	L_t	R_t
(l)	(mm)	(mm)	(mm)
2.9	50	1400	250

The experiments were investigated at room temperature (20°C). The influence of experimental parameters such as sorbent dose, initial pH, and initial chromium concentration were studied by varying one parameter while keeping others constant. All experiments were performed in duplicates and only, the mean values were used.

III. Results and discussion

III.1. Effect of biosorbent dose

The effect of biomass dose on hexavalent chromium removal from aqueous solution was investigated at various sorbent amounts ranging from 1 to 6 g/L at pH value around 5.4 and 100 mg/L fixed as initial Cr (VI) concentration. As illustrated in Figure 2 the percentage removal increases from 31.53 to 77.43% with increasing the sorbent dose from 1 to 6g/L. This can be due to the increase in surface area and number of active sites offered by the increased biosorbent dose [20], [21]. Adsorption capacity was decreased from 36.53 to 12.91mg/g as *Pleurotus mutilus* dose increased from 1 to 6g/L. The high chromium uptake was obtained at low biosorbent dose, it could be attributed to the availability of free active sites for metal uptake [21]. The decreased biosorption capacity at higher biosorbent dose is probably due to the increase of the vacant sorption sites with more sorbent present in solution [4]. Also it appears that the maximum chromium uptake corresponds to the biosorbent dose of 2g/L. Therefore, it has been selected for biosorption experiments.

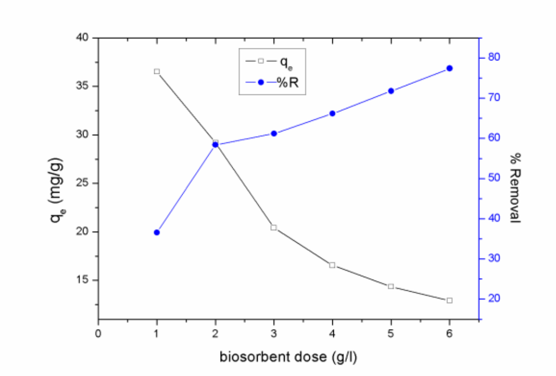

Figure 2. *Effect sorbent dose of Pleurotus mutilus biomass on Cr (VI) uptake*

III.2. Effect of chromium initial concentration

The adsorption experiments were performed by varying Cr (VI) concentration and keeping other parameters constant such as pH 5.4, a sorbent dose of 2 g/L, and agitation speed of 800 rpm. The particle size PS ranging from 160 µm to 200 µm. When the initial metal concentration increases from 25 to 150 mg/L respectively the Cr (VI) uptake increases from 9.52 to 31mg/g this results was presented by Alouache et al [11]. This increase can be explained by the limited number of active sites that become saturated by exceeding a certain concentration of Cr (VI) [1]. Also the increase in Cr (VI) concentration gradient provides a driving force for overcoming the mass transfer resistances between the metal ions of the aqueous solution and the active sites of the biosorbent [8], [20]. As the initial Cr (VI) increases, over 100 mg/L, a decline in sorption uptake is revealed. This phenomena can be due to the binding sites saturation caused by the high number of chromium ions in solution [21]. Hence, increasing in sorption capacity became quite slower [22].

Finally, a dynamic equilibrium form has been established between the chromium ions in solution and those of the adsorbent surface, where all the active sites of the adsorbent get occupied [7,8]. Therefore the Cr (VI) removal was highly dependent on the initial Cr (VI) concentration. Similar trend of metal biosorption was reported by Rangabhashiyam et al [8] using activated Sterculia guttata shell. Consequently, a concentration of 100 mg/L was selected as the optimum initial

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hexavalent chromium concentration for biosorption experiments according to Alouache et al [11].

III.3. Effect of initial pH on metal ion biosorption

The pH value is one of the important parameters affecting the biosorption capacity for metal ions removing in aqueous solutions. Experiments were carried out in a pH range from 3 to 10 while keeping Cr (VI) concentration equal to 100 mg/L, a sorbent dose of 2 g/L, agitation speed of 800 rpm, and the particle size PS ranging from 160 µm to 200 µm. Alouache et al [11] were found a maximum chromium uptake around pH 5.4. Also, this result can be explained by the point of zero charge (pH_{pzc}) of the *Pleurotus mutilus* biomass, at which the electric charge on biosorbent surface is zero as reported by Chergui et al [23]. They found that the overall charge of biomass was positive in acidic pH values.

The decrease in sorption could be due to the decrease of the electrostatic attraction, and the biosorption competitiveness between the chromium species in solution ($HCrO_4^-$) (CrO_4^{-2}) and (OH^-) ions on active sites of the sorbent. From pH great than 8, more hydroxyl ions presents in solution, then the surface being charged negatively, and repulsive towards chromium ions. Causing a low biosorption capacity for Cr(VI) [24], [25]. Hence this value was taken as the optimum for further adsorption experiments. Similar trends were observed for the sorption of Cr(VI) by MgAl-NO_3 hydrotalcite in fixed-bed column by Khitous et al [4]. They have obtained a high sorption capacity at pH solution of 5 and 6 with maximum uptake of (48mg/g), which suggests an optimum pH for efficient uptake of Cr(VI) ions. The same phenomena was observed by Kahu et al [26] in the detoxification study of Cr(VI) using sulfate-crosslinked chitosan. They have obtained a maximum uptake at pH 6. Similarly, an optimum pH value of 5 was obtained for the biosorption of Cr(VI) onto *Spirulina sp* by Rezaei [9]. Also, a maximum removal of chromium (82.3%) by dead biomass of Trichoderma sp. BSCR02 was observed at pH value of 5 by Rose et al [27].

III.4. Kinetic studies

Kinetic studies were investigated in torus reactor. Experiments were conducted at 20°C, and biomass dose of 2g/L and initial Cr (VI) concentration of 100 mg/L, initial pH 5.4 and agitation speed 800rpm. The remaining Cr (VI) concentration was determined as function of time.

Biosorption chromium analyses were carried out by fitting the kinetics data to pseudo-first-order and pseudo-second-order models, intra particle diffusion models (Weber and Morris), and (Urano and Tachikawa). Boyd model and external mass transfer were used in order to identify the rate controlling step in Cr (VI) biosorption onto *Pleurotus mutilus* biomass. These models are largely used to describe heavy metals biosorption, by including all steps of adsorption [8], [25], [28].

III.4.1. Pseudo-first order

The biosorption kinetic of Cr(VI) was modeled using a pseudo-first-order model proposed by lagergren [29], [30] and expressed in the linearized form as follows:

$$log_{10}(q_e - q_t) = log_{10}(q_e) - \frac{K_1}{2.3} t \qquad \textbf{(3)}$$

Where, q_t and q_e are the uptake capacity of Cr (VI) at time (t) and equilibrium, respectively (mg/g). k_1 is the first-order rate constant (min^{-1}).

3.4.2. Pseudo-second order

The biosorption kinetic was also described by a pseudo-second–order reaction [29], [30]. The linearized form was presented as follows:

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{t}{q_e} \qquad \textbf{(4)}$$

Where k_2 is the rate constant of the pseudo-second–order model (g/(mg.min)).

Information on Cr (VI) uptake is necessary to select the operating conditions for removal processes. The obtained kinetics data were plotted by applying different models, as shown in Figure 3.

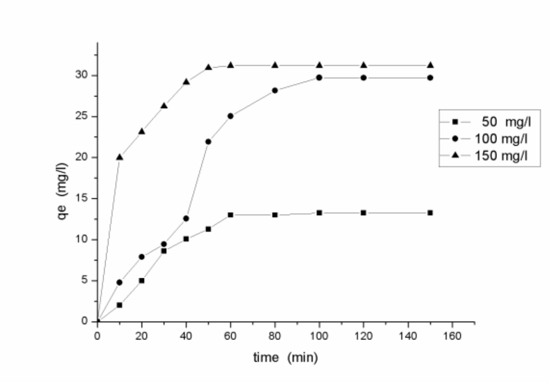

Figure 3. *q_t versus time for Cr(VI) biosorption onto Pleurotus mutilus biomass*

The increase of interaction time increases the removal of Cr (VI). The adsorption rate of Cr (VI) appears to be rapid for the initial contact period of 60 min, which can be explained by the availability of more active sites on the biomass external surface in the process beginning. With gradual decrease in these active sites, the adsorption process becomes slower in the later stages and took more time to adsorb Cr(VI) ions [16]. Consequently, the adsorption has a slower rate and become almost constant around 150 min. With increasing the contact time, the active sites became more occupied, which indicate that the adsorption equilibrium of Cr (VI) was reached. Similar results were observed by authors [4], [7], [16]. For the pseudo-first–order reaction, q_e and k_1 were

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determined from the intercept and slope of the plot of log (q_e-q_t) versus time t as illustrated in Figure 4.

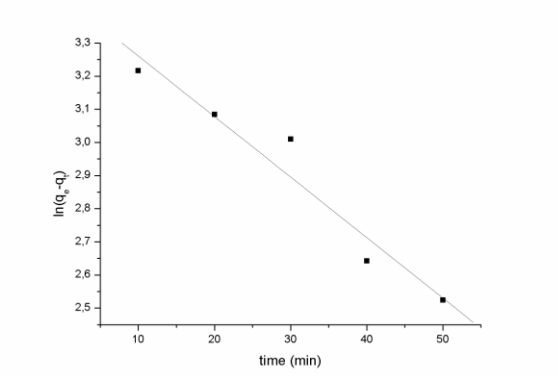

Figure 4. *The plot of the pseudo-first-order biosorption kinetics of Chromium onto Pleurotus mutilus biomass*

The biosorption kinetics was also described by a pseudo-second–order reaction, where q_e and k_2 can be determined from the slope and intercept of the linear plot of (t/q_t) versus time t not shown here. The pseudo-first-order and pseudo-second-order parameters are presented in Table 3.

Table 3. *kinetics isotherm parameters for Cr (VI) biosorption by Pleurotus mutilis biomass*

Kinetics model	Parameter	Value
Pseudo-first-order	K_1(min^{-1})	0.018
	q_e (mg/g)	30.96
	R^2	0.943
Pseudo-second-order	K_2(g.mg^{-1}min^{-1})	8.4110^{-4}
	q_e (mg/g)	26.31
	R^2	0.903

The obtained determination coefficient value for the pseudo-second-order model was low, and the calculated value $q_{e,cal}$ (26.31 mg/g) was not close to the experimental value $q_{e,exp}$ (29.46 mg/g). This observation suggested that the Cr (VI) adsorption data diverged from the pseudo-second-order model to the pseudo-first-order model. It appears clearly that the calculated value 30.96 mg/g for the pseudo-first -order model converge to experimental value $q_{e,exp}$ (29.46 mg/g), with a high value of determination coefficient (R^2), compared to the pseudo- second -order model. Consequently our experimental data were well fitted with pseudo-first order model kinetic. Similar kinetic results have been obtained for the biosorption of Cr(VI) and Cu(II) by waste tea fungal biomass by Razmovski et al [31]. Also, Dakiky et al [29] were observed that Cr (VI) adsorption on various lignocellulosic adsorbents fitted well with a pseudo-first order mechanism.

III.4.3. Intra particle mass transfer (Weber and Morris model).

The diffusion mechanism of biosorption could not identified by the pseudo-first-order and pseudo-second-order kinetic models [32]. The overall rate of biosorption will be controlled by the slowest step, which would be either external mass transfer or intra-particle diffusion [25].

Knowing the steps involved during the adsorption, became necessary to predict the limiting step which is an important parameter in the biosorption process. Experimental kinetics data were analyzed by applying the intra-particle diffusion model, proposed by Weber et al [33] as follows:

$$q_t = K_i \,.\, t^{0.5} \qquad \textbf{(5)}$$

Where k_i is the intra-particle diffusion rate constant (mg/(g.min$^{0.5}$), $t^{0.5}$ is square root of time (min$^{0.5}$). K_i can be obtained from the slope of the plot of (q_t) versus ($t^{0.5}$). The slope of each linear part represents the limiting diffusion mechanism; the two steps show that the biosorption process takes place by surface sorption and intra particle diffusion. The initial linear step of the plot indicates a boundary layer effect, while the second linear step is due to the intra particle or pore diffusion [25], [34]. By taking into account only the initial period according to Weber and Morris model, the diffusion coefficient in the solid phase D_w will be calculated, using Eq.(6):

$$K_{i1} = \left(\frac{12\,q_e}{d_p}\right)\left(\frac{D_W}{\pi}\right)^{0.5} \qquad \textbf{(6)}$$

As shown in Figure 5, it is clear that the biosorption process follow two phases: surface diffusion with slope (k_{i1}=1.814 mg/(g.min$^{0.5}$), followed by intra particle diffusion with slope (k_{i2}=2.000 mg/(g.min$^{0.5}$) The diffusion coefficient value was (D_w =4.4610^{-14} m^2/s). Similar results were found by Rangabhashiyam et al [8]. On the other hand, the intra particle diffusion parameter k_{i2} (mg/(g.min$^{0.5}$) is equal to the slope of the second linear portion of the curve; the intercept of the plot represents the boundary layer effect. when interception is greater, the contribution of surface sorption is important [34]. Regarding all these results, it is important to estimate which is the limiting step (pore or surface diffusion) in the sorption process.

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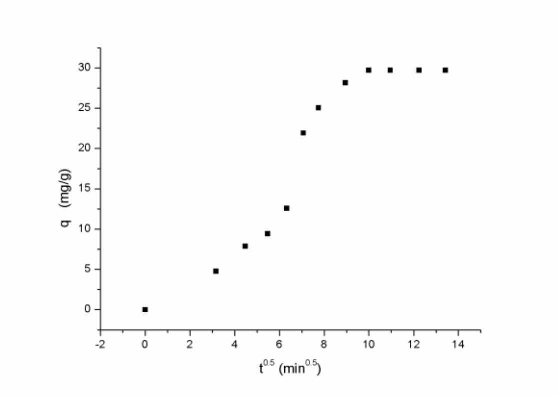

Figure 5. *Weber and Morris plot for Cr(VI) biosorption onto Pleurotus mutilus*

III.4.4. Intra particle mass transfer (Urano and Tachikawa model)

Urano et al [35] have proposed another type of intra particle diffusion model. The rate of adsorption was considered as independent with the agitation speed, and external diffusion negligible compared to the low adsorption rate[36]. The kinetics of biosorption was modeled by Eq.(7):

$$\ln\left[1-\left({q_t}/{q_e}\right)^2\right] = -\frac{4\pi^2 D_i t}{d_p^2} \qquad \textbf{(7)}$$

Where d_p is the mean particle diameter (m), D_i is the average diffusion coefficient in solid (m^2/s).

D_i was computed by plotting $-\ln\left[1-\left({q_t}/{q_e}\right)^2\right]$ versus time t according to Urano and Tachikawa model [35]. As shown in Figure 6, the slope of the plot gives the D_i value equal to $3.24.10^{-12} m^2/s$. The comparison of D_w (Weber and Morris model), and D_i (Urano and Tachikawa model) coefficients values with those of molecules in water (10^{-9} m^2/s) indicates a poor intra particle diffusion. Consequently, the external mass transfer of Cr(VI) through the particle-liquid film was assumed to be the controlling step in the overall sorption process.

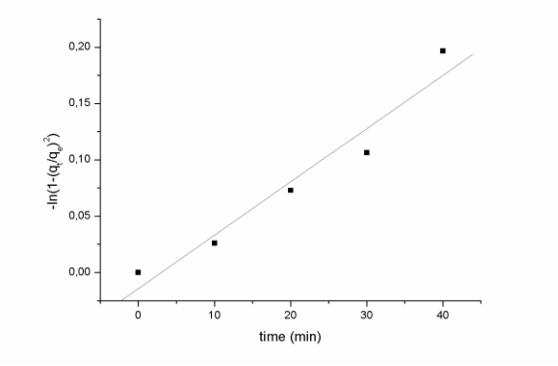

Figure 6. *Urano and Tachikawa plot for Cr (VI) biosorption onto Pleurotus mutilus*

III.4.5. External mass transfer model

The rate constant of external mass transfer of Cr(VI) was calculated using the model proposed in litterature [33,36]. Different steps control the biosorption kinetics including diffusion processes [33,35]. Three steps can be applied to chromium removal; (i) transfer of chromium ion from the boundary film to the particle's surface, and (ii) transfer of the Chromium ion from the surface to the intra particle actives sites, finally (iii) the uptake of chromium ion on the active sites, via sorption and complexation.

In agitated solid/Liquid reactor, mixing in the liquid is very rapid, and then the concentrations of heavy metal and sorbent are nearly uniform. The Fick's law is applied in this model, to characterize chromium concentration evolution in solution C (mg/L). As a function of concentration difference of Cr (VI) ion between solution, and surface particle C_S (mg/L) according to Eq. (8) as follows:

$$\frac{dc}{dt} = -k_s.S.(C - C_s) \qquad \textbf{(8)}$$

Where k_s is the mass transfer coefficient (m/s) and S the specific surface area (m^{-1}). By making some assumptions such as; initial surface concentration (C_S) negligible, and (C_0) the initial concentration in solution, the previous equation can be simplified to:

$$\left[\frac{d\left({C}/{C_0}\right)}{dt}\right]_{t\to 0} = -k_s.S \qquad \textbf{(9)}$$

The initial rate of sorption, $-K_s.S$ (s^{-1}) can be obtained by polynomial linearization of C/C_0 curves, and subsequent derivation at $t = 0$. The specific surface area S, supposed spherical was calculated as the external surface, as follows:

$$S = \frac{6.w}{d_p.\rho_{app}} \qquad \textbf{(10)}$$

Where w is the mass concentration of the sorbent in solution (kg/m^3), d_p the average particle diameter (m) and ρ_{app} the sorbent apparent volume mass (kg/m^3). According to the slope value at C=0 obtained by plotting C/C_0 versus time t, as illustrated in Figure 7 the external mass transfer coefficients was determined as $K_s = 5.16 \times 10^{-5}$ m/s. The kinetic of chromium sorption onto *Pleurotus mutilus* was governed by an external diffusion process; it was confirmed by the low value of Ks.

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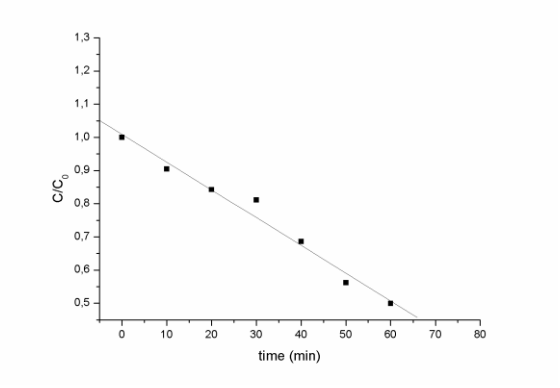

Figure 7. *External mass transfer plot for Cr(VI) biosorption onto Pleurotus mutilus*

III.4.6. Boyd model

Boyd's model was applied to identifies whether the adsorption mechanism is a film or an intra particle diffusion due to the dual dependence of adsorption rate [25], [37]. The experimental data were analyzed by this model in order to identify the predominant phenomena in the biosorption process, the kinetic expression of this model was presented by Aravindhan et al [38] in the following equation:

$$B_t = -0.4977 - ln\left(1 - \frac{q_t}{q_e}\right) \qquad (11)$$

Where B_t is the liquid film diffusion constant, in order to distinguish whether the external or intraparticle transport controls the rate of biosorption, the linearity of the plotting of B_t versus time t has been exploited; if the curve is linear without passing through the origin, the adsorption is controlled by external diffusion, if it passes by the origin, the intra particle transport controls the adsorption process [25], [28], [34], [38]. The effective diffusion coefficient (D_j) can be calculated using Eq.(12) as follows:

$$B_t = \left(\frac{\pi^2}{r_p^2} D_j\right) \times t \qquad (12)$$

Where r_p represents the radius of the assumed spherical particle (m), according to Figure 8, the relation between B_t and time (t) is linear ($B=0.043 min^{-1}$; $R^2=0.91$), but does not pass through origin, which indicate that the surface diffusion is the rate-limiting step. The obtained diffusion coefficient value was equal to $2.35.10^{-12}$ m^2/s, it is lower than that of ions diffusion coefficient in the liquid. Similar results were obtained by Hachi et al [28] for methylene blue adsorption on *Pleurotus mutilis* biomass. The low values of K_{i2} of (Weber and Morris model) and D_j coefficients of (Boyd model), suggest that the intra particle diffusion is negligible in comparison to the external mass transfer phenomena, and adsorption will be occurred on biomass surface [38].

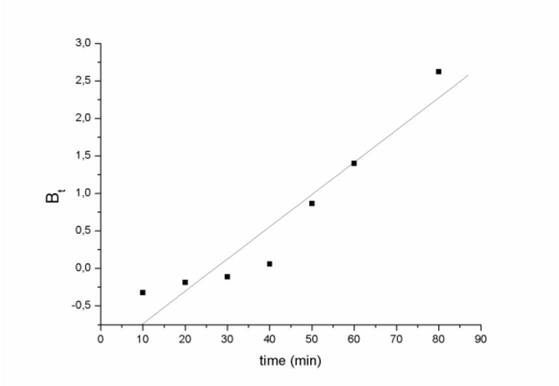

Figure 8. *Boyd plot for Cr (VI) biosorption onto Pleurotus mutilus*

III.5. Adsorption isotherms

The equilibrium adsorption isotherms are fundamental to identify the mechanisms of Cr (VI) uptake and the interaction between adsorbent and adsorbate. Equilibrium adsorption studies were carried out, by monitoring the Cr (VI) uptake capacity q_e as function of the residual Cr (VI) concentration C_e in aqueous solution. The experimental data at equilibrium were applied to various isotherms models such as: Langmuir, Freundlich, Temkin and Dubinin-Radushkevich.

III.5.1. Langmuir model

The removal of metal ions in this model, occurs on homogeneous surface through monolayer adsorption without interaction between the adsorbate present simultaneously on the adjacent sites [39]. This model doesn't give any information about the mechanism. It gives only the sorbent adsorption capacity [27, 39].this model can be presented by equation as follow:

$$q_e = \frac{q_m b C_e}{1 + b C_e} \qquad (13)$$

Where q_m is the maximum adsorption capacity (mg/g), and b is the equilibrium constant (L/mg), which represents the adsorption energy [20], b and q_m can be determined using Eq. (14) the plot of C_e/q_e versus C_e.

$$\frac{C_e}{q_e} = \frac{1}{q_m b} + \frac{C_e}{q_m} \qquad (14)$$

The distribution factor R_L defined in Eq. (15) can describe the characteristics and type of Langmuir isotherm.

$$R_L = \frac{1}{1 + b C_0} \qquad (15)$$

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Where, C_0 is the initial adsorbate concentration. A further analysis of Langmuir isotherm can be made according to R_L value. A favorable adsorption process is dominant for R_L varying between 0 and 1. While an unfavorable adsorption is for $R_L > 1$, when $R_L=1$ adsorption is linear and if $R_L = 0$, it is irreversible [7], [9], [40].

According to results obtained by Alouache et al [11] the plot of C_e/q_e vs C_e, gives a straight line of slope $1/q_m$ and intercepts $1/b.q_m$. The Langmuir parameters are summarized in Table 4. The high value of R^2, indicate a best fit of this model to the adsorption of Cr (VI) by *Pleurotus mutilus* biomass. The R_L was found between 0–1, which confirms the favorable adsorption process for hexavalent chromium removal [1], [22].

Table 4. *Isotherm model parameters for Cr (VI) Biosorption onto Pleurotus mutilus*

Isotherm	parameter	Value
Langmuir	q_m (mg/g)	23.25
	b(L/mg)	1.13
	R^2	0.916
	R_L	0.0087
Freundlich	n	1.32
	K_f	1.29
	R^2	0.888
Temkin	K_T(L/g)	381.4
	B(J/mol)	22766.3
	R^2	0.972
Dubinin-Radushkevich	q_{max}(mg/g)	32.29
	β (mol^2/J^2)	3.10^{-5}
	E (KJ/mol)	0.129
	R^2	0.978

II.5.2. Freundlich model

The Freundlich isotherm is applied under the assumption of the heterogeneous adsorption surface and active sites involving different energies, this model expressed by Eq. (16):

$$q_e = K_F C_e^{1/n} \qquad \textbf{(16)}$$

Where k_F is the Freundlich constant related to the uptake capacity, the Freundlich exponent 1/n, represent the intensity of adsorption [41], which is lower for more heterogeneous surfaces. K_F and n can be obtained from the linear forms of Eq. (16) as follows:

$$lnq_e = lnK_F + \frac{1}{n} lnC_e \qquad \textbf{(17)}$$

The Freundlich constants values were determined using the straight line of the plot of (lnq_e) versus (lnC_e), and presented in Table 4 according to Alouache et al [11], it can be seen clearly that the value of determination coefficient R^2 for Freundlich model is lower than that of Langmuir model.

III.5.3. Temkin model

The linear decrease in biosorption heat with increasing biosorbent coverage is the basis of the Temkin isothermal hypothesis [42]. This model is presented by Eq. (18) as follows:

$$q_e = (RT/B) ln(K_T C_e) \qquad \textbf{(18)}$$

K_T and B values were obtained by the linearized form of Eq. (18).

$$q_e = (RT/B) lnK_T + (RT/B) lnC_e \qquad \textbf{(19)}$$

Where K_T is the Temkin isotherm equilibrium binding constant (L/g), R is the universal constant of gas (8.314J/(mol.K), B is a constant related to the biosorpton heat (J/mol), T is the temperature (K). The plot of q_e versus $ln(C_e)$ gives a straight line of slope (RT/B) and intercepts ((RT/B).lnK_T). Temkin parameters are summarized in Table 4. These results confirm that increasing biosorbent coverage leads to a decrease in biosorption heat.

III.5.4. Dubinin-Radushkevich model

The Dubinin-Radushkevich isotherm is analogical to Langmuir type and more general since it does not assume a constant adsorption potential or homogeneous surface. This isotherm was developed by taking into account the effect of the porous structure of the biosorbent [42], [43]. However, it can be used to identify the physical or chemical nature of biosorption. This model is commonly presented by Eq. (20) as follows:

$$q_e = q_m exp^{-\beta\varepsilon^2} \qquad \textbf{(20)}$$

The linearized form of Dubinin-Radushkevich isotherm was presented in Eq. (21) as follows:

$$lnq_e = lnq_{max} - \beta\varepsilon^2 \qquad \textbf{(21)}$$

Where β is the activity coefficient (mol^2/J^2) related to the mean biosorption free energy E, and ε is the Polanyi potential (J/mol) which can be determined using Eq. (22):

$$\varepsilon = RT ln\left(1 + \frac{1}{C_e}\right) \qquad \textbf{(22)}$$

The Polanyi sorption approach is based on the hypothesis of the existence of a fixed volume of sorption space near the surface of the adsorbent with a sorption potential existing on these spaces. The sorption space near the solid surface is characterized by equipotential surfaces series which have the same sorption potential. This sorption potential is not dependent on temperature. It varies with both sorbent and sorbate nature [44].
The plot of lnq_e versus ε^2 gives a straight line, with a slope equal to $(-\beta)$. The average free sorption energy per sorbate molecule necessary for transferring quantity of one mole of metal ion from infinity in the solution to the biomass surface [44].

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Thus the mean energy (E) can be determined using Eq. (23):

$$E = \frac{1}{\sqrt{2\times\beta}} \qquad \textbf{(23)}$$

The obtained energy value E is significant to predict the type of adsorption processes [42], [45]. For physisorption process, the value of mean adsorption energy is less then 8 kJ/mol. While, for ion exchange is between 8 and 16 kJ/mol. Finally, for chemisorption process the value is ranging between 16 and 400 kJ/mol. The Dubinin-Radushkevich parameters are presented in Table 4. The value of adsorption energy is equal to 0.129 kJ/mol. It corresponds to a physical biosorption process in nature. The adsorption capacity using the Dubinin–Radushkevich isotherm model was obtained q_{max}(32.29 mg/g), it converge to experimental value $q_{e,exp}$ (29.46 mg/g). The determination coefficient value (R^2) was found higher than the other isotherm models which confirm the physical biosorption of Cr (VI) onto *Pleurotus mutilus* biomass. Similar results were obtained by Rangabhashiyam et al [8].

Table 5. *Comparison of adsorption capacities of the adsorbents for the removal of Cr(VI)*

Biosorbent	q_e (mg/g)	references
Sterculia guttata shell	90.90	[8]
Bacillus cereus	30.93	[22]
Scallop shell	34.48	[1]
Spirulina sp	90.91	[9]
activated carbon	7.80	[7]
Pleurotus mutilis	29.46	Our study

The adsorption capacity of *Pleurotus mutilis* biomass has been compared with similar biosorbents reported in literature and presented in Table 5. The experimental data of the present study are similar to those obtained by authors in some cases. The adsorption capacity varies and depends on adsorbent characteristics.

III.6. Desorption study

The recycling of the adsorbent through desorption process is a most important aspect from an economic point of view. Furthermore, the process helps to regenerate the adsorbents so that it can be subjected for the successive cycles in adsorption process. It allows to decrease the processing cost and to open the possibility of recovering the metal extracted from the liquid phase [46]. For our experiments, the regeneration of sorbent material was carried out in order to reuse the biomass and also recover the metal.

After adsorption, desorption experiments were performed in batch by introducing of a known amount of metal loaded biomass in a beaker of 50 mL and kept in agitation for 1h. The experiments were carried out with NaOH solution (1.0 mol/L) used as eluting agent. Then, the biomass was collected by filtration and dried in an oven for 5 h at 105°C. It was regenerated without a load to the environment in terms of disposal [47]. The filtrate obtained contains chromium concentrated solution. This operation allows to reducing the volume of the pollution, after that it can be reused or treated by physical and/or chemical methods.

III.7. Adsorption thermodynamic

It is well known that, temperature influences the sorption process rate, through two main effects over the adsorbents [46].Temperature can affect the diffusion rate of the metal ions by decreasing solution viscosity and increase in the number of the adsorption sites [48]. The effect of temperature on Cr(VI) adsorption onto *Pleurotus mutilus* was investigated at temperatures of 293 to 323 K at pH 5.4 with initial Cr(VI) concentration of 100 mg/l, and sorbent dose 2 g/l. An increase in temperature increases the chromium uptake of Cr(VI) by *Pleurotus mutilus*.

The thermodynamic constants, free energy ΔG°enthalpy ΔH° and entropy ΔS° were calculated to confirm the nature of adsorption and to evaluate the thermodynamic feasibility of the process.

The values of ΔH° and ΔS° were calculated from the intercept and slope of the plot of lnK_L versus 1/*T* according to Eq (24). The free energy ΔG° is calculated using Eq (25):

$$lnK_L = -\Delta H^\circ/_{RT} + \Delta S^\circ/_{R} \qquad \textbf{(24)}$$

$$\Delta G^\circ = \Delta H^\circ - T\Delta S^\circ \qquad \textbf{(25)}$$

where *R* is the universal gas constant (8.314 J / mol K), *T* is the temperature in Kelvin and $K_L$is the equilibrium constant, calculated as the surface and solution metal distribution ratio ($K_L = q_e / C_e$) [46], [49].

The values of ΔG°, ΔH° and ΔS° are given in Table 6. The positive values of ΔH° show that the adsorption process was endothermic and that an increase in temperature favored the adsorption [46], [49].The reason why adsorption capacity increased with increasing temperature can be explained by the value of ΔH° [50].

The negative values of ΔG° indicate that Cr (VI) adsorption process is spontaneous. The positive

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value of ΔS° denote the increased randomness during the sorption process of Cr (VI) on the surface of *Pleurotus mutiuis biomass*. The increase in sorption with temperature may be attributed to either increase in the number of active surface sites available for sorption or due to the decrease in the boundary layer thickness surrounding the sorbent, so that the mass transfer resistance of sorbate in the boundary layer decreased [46]

Table 6. *Thermodynamic parameters for hexavalent chromium biosorption*

T(k)	ΔG°(KJ.mol^{-1})	ΔH°(KJ.mol^{-1})	ΔS°(J.mol^{-1}K^{-1})
308	-0.58	24.40	81.10
313	-0.98		
318	-1.39		

IV. Conclusion

In this research study, it has been showed that the *Pleurotus mutilus* biomass have a promising application in the removal of Cr (VI) from aqueous solutions. The maximum sorption capacity was found to be 29.46 mg/g under optimal conditions (pH 5.4 and initial concentration of 100 mg/L and biosorbent dose of 2 g/L). The modeling study verifies that the adsorption kinetics follow the pseudo-first-order. The obtained values of D_w , D_i and D_j diffusion coefficients according to Weber-Morris, Urano-Tachikawa and Boyd models respectively are low compared to the value of ion diffusion in liquid (D). These results demonstrate that the external mass transfer is the predominant mechanism that control biosorption of Cr (VI) onto *Pleurotus mutilus* biomass. In addition the low value of the external mass transfer coefficients K_s further confirms the interpretation above.

The study of adsorption isotherm showed that the equilibrium data could be fitted by Langmuir, Temkin and Dubinin-Radushkevich isotherms equations confirming the physical sorption of Cr(VI). The thermodynamic analysis indicates that biosorption was endothermic and spontaneous in nature.

This first application of the torus reactor in biosorption phenomena shows interesting results. This type of reactor has promising applications in wastewater treatment, as it has many advantages in the effluents flow and the good mixing of heterogeneous solutions even at high concentrations, so it can be suggested in continuous mode to facilitate its industrial application.

The research results indicate that the *Pleurotus mutilus* biomass could serve as an inexpensive source for chromium recovery from contaminated water due to its low cost (waste material first destined to incineration valorized with no treatment indeed), it could be an excellent materiel for water treatment application. It is anticipated that this study could lead to further investigation in the removal of other heavy metals using *Pleurotus mutilus* or other biosorbent in a torus reactor.

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