MODELING AND SIMULATION OF FIXED BED BIOSORPTION SYSTEM FOR CADMIUM AND LEAD IONS FROM WASTEWATER

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Abstract

It is the application of simulated data obtained from experimental results that can be used for further scale up of continuous fixed bed system for biosorption of heavy metals from wastewater not batch system that will make use of bioreactor more attractive to the industries. Modeling and simulation of microbial biosorption of cadmium and lead ions from synthesis wastewater in a continuous fixed bed system were achieved using plexiglass tube with height 30.0 cm and column internal diameter 1.0 cm . The simulated and validation results show that sum of the squares of error values of $4.6 \text{ X } 10^{-8}$, $8.84 \text{ X } 10^{-7}$, $1.365 \text{ X } 10^{-2}$ and $8.4\,95 \text{ X } 10^{-5}$ for Adams-Bohart model indicating lowest fitness to $2.6 \text{ X } 10^{-8}$, $8.6 \text{ X } 10^{-7}$, $9.9 \text{ X } 10^{-3}$ and $7.6 \text{ X } 10^{-5}$ recorded for Thomas model and Yoon-Nelson model for all biosorbates biosorption carried out. Cadmium ions biosorption onto Saccharomyces cerevisiae gives the best acceptable data with values of coefficient of determination ranged from $0.9688 - 0.9689$ and sum of the squares of error which ranged from $(2.6 - 4.6) \text{ X } 10^{-8}$ for the three models validated. Moreover, lowest acceptable data were observed with lead ions biosorption onto Saccharomyces cerevisiae at values of coefficient of determination ranged from $0.4204 - 0.4427$ and sum of the squares of error gives the range from $9.9 \text{ X } 10^{-3} - 1.365 \text{ X } 10^{-2}$ for the three models validated.. The three models are suitable in designing a fixed bed system for biosorption of cadmium and lead ions onto immobilised nonliving Bacillus circulans and Saccharomyces cerevisiae biosorbents

Keywords: Biosorption, Breakthrough curve, Statistical analysis, Models validation

Introduction

The removal of heavy metals from wastewater by physical, chemical and biological techniques has been studied or reviewed extensively by many researchers (Wang and Chen, 2009; Fomina, and Gadd, 2014; Srivastava et al., 2015; Luka *et al.*, 2021a; Luka *et al.*, 2021b). The physical and chemical methods are ineffective in terms of process cost, energy and chemical products consumption, generation of toxic sludge and disposal problems (Das *et al.*, 2008; Wang and Chen, 2009). Nowadays, applying biological technique also known as biotechnology in controlling and removing metal pollution has drawn attention of many investigators and gradually becomes topic of interest in the field of metal pollution control because of its potential application (Fu and wang, 2011; Wu et al., 2012; Fomina, and Gadd, 2014). Batch process systems are not attractive for industrial application and treatment of large volume of wastewater to Continuous process system such as fixed bed contactor (Luka *et al*, 2021a).

The performance of a fixed bed system in biosorption is generally viable only with small laboratory columns. The data obtained in the laboratory experiments can be utilised for estimating and evaluating the performance of industrial or practical size contactor by applying suitable mathematical models developed by many investigators for such purposes. Various models have been applied to estimate the breakthrough performance and also to calculate the column kinetic constants and adsorption capacity of the fixed-bed system (Cruz-Olivares *et al.*, 2013; Rout *et al.*, 2014). Among the several models developed by researchers some of the models that can be used for modeling and simulation of fixed bed system for biosorption of heavy metals from wastewater includes: Adams-Bohart model, Thomas model and Yoon-Nelson model (Yunnen *et al.*, 2017; Benjreid *et al.*, 2018). Simulation of biosorption processes will help in further scale up of economically effective and eco- friendly fixed bed system for bioremediation of wastewater for intended use.

The aim of this research is to simulate and validate models of continuous biosorption process for a fixed bed system using immobilised *Bacillus circulans* and *Saccharomyces Cerevisiae* as biosorbents to remediate cadmium and lead from wastewater.

Materials and Methods

Preparation of Solution and Serial Dilution

Stock solutions optimum concentration ($100\ mg/l$) of Cadmium and $40\ mg/l$ Lead ions were prepared from Cadmium nitrate ($Cd(NO_3)_2.4H_2O$), and Lead nitrate ($Pb(NO_3)_2$), respectively. The Cadmium ions solution optimum operating pH of 7.0 and 6.0 for *Bacillus Circulans* and *Saccharomyces Cerevisiae* respectively as well as that of Lead ions solution operating pH of 3.0 for both *Bacillus Circulans* and *Saccharomyces Cerevisiae* in the continuous fixed bed process system were achieved using $0.1\ M\ NaOH$or HCl (Luka *et al.*, 2021a; Luka *et al.*, 2021b). Nitrate salts were used as the counter ion for metal ions where necessary because of its low tendency to form complexes (Amirnia, 2015).

Immobilisation of Biosorbent

Entrapment method that involved drop wise addition of biomass suspended in polymer matrix of $2\ \%\ (w/v)$ sodium alginate solution and $5\ \%\ (w/v)$ biomass was adapted for the production of the bead (Luka *et al.* 2021a). After drying, each strain of immobilised biomass bead were kept inside tight bottle for future use as biosorbent.

The Fixed Bed Process Systems

The continuous fixed bed experimental process system set up was adapted from Luka *et al.* 2021a. The bed was made from a plexiglass tube with inner diameter (d) $1.0\ cm$ and height (H) 30.0 cm . A layer of glass wool $1\ cm$ was installed at the bottom and top of the bed. An immobilised biomass was served as biosorbent packed with height $9\ cm$ for both *Bacillus circulans* and *Saccharomyces cerevisiae*, respectively. The flow rate (Q) was set constant at $10\ ml/min$. Each metal solution was pumped into the column in an up-flow mode. The experiments were conducted at room temperature of 32 ℃. The concentrations of Cadmium and Lead ions after biosorption were evaluated using standard method for examination of water and wastewater (Luka *et al.*, 2021a). The concentration of the effluent stream of Cadmium and Lead ions were detected each using VGP210 - Atomic absorption spectroscopy. Figure 1, shows the schematic presentation of the fixed bed contactor process system.

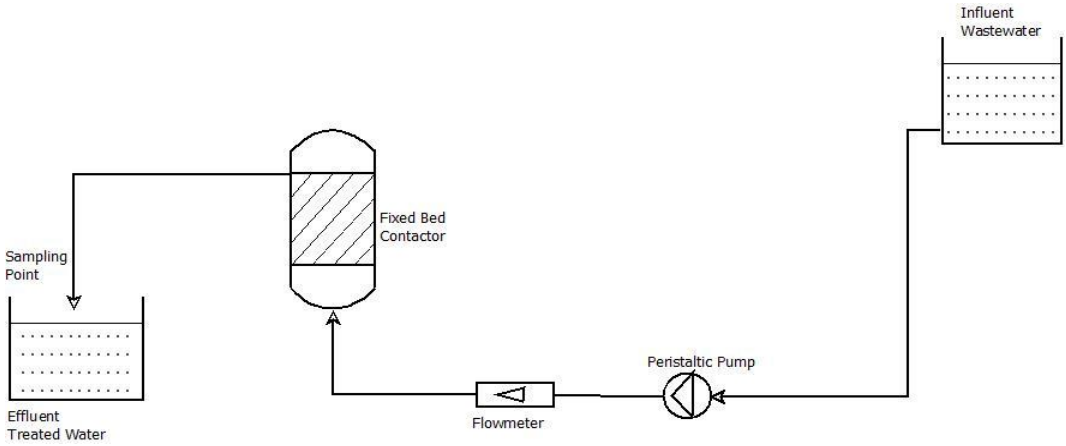

Figure 1. Schematic Presentation of the Fixed Bed Process System (Luka *et al.*, 2021a)

Model Selection

The biosorption experimental data were all generated at room temperature (32 ℃) for continuous fixed bed process systems. Some selected existing biosorption breakthrough curves models of Adams-Bohart, Thomas, and Yoon-Nelson models were used to evaluate the Performance of *Bacillus circulans* and *Saccharomyces cerevisiae* biosorbents with the heavy metals under investigation (Cruz-Olivares *et al.*, 2013; Yunnen *et al.*, 2017; Benjreid *et al.*, 2018). The modeling and simulation of biosorption processes were carried out using R2009b - MAT LAB Software and the model's parameters were obtained from the plots of each model equation.

Statistical Analysis, Simulation and Validation of Models of Fixed Bed Contactor

The R2009b - MAT LAB was used for writing codes and simulation, while experimental and simulated data were validation using a plot of $\frac{C_t}{C_o}$ versus t, R^2 and statistical or error analysis known as Sum of the Squares of Error (SSE).

The Sum of the Squares of Error (SSE) was obtained using equation (1) (Rout *et al.*, 2014; Amirnia, 2015):

$$SSE = \frac{\Sigma\left[\left(\frac{C_t}{C_o}\right)_c - \left(\frac{C_t}{C_o}\right)_e\right]^2}{N} \quad (1)$$

Where, $\left(\frac{C_t}{C_o}\right)_c$ = ratio of effluent and influent biosorbate concentration gotten from the model calculation; $\left(\frac{C_t}{C_o}\right)_e$ = ratio of effluent and influent biosorbate concentration gotten from the experiment; N = total number of experimental point.

Results and Discussion

Continuous fixed bed breakthrough curves models

The breakthrough curves models testing for cadmium ions biosorption onto *Saccharomyces cerevisiae* ($Cd\ Y$) and Bacillus c*irculans* ($Cd\ B$) for Adams-Bohart model (AB), Thomas model (TH) and Yoon-Nelson model (YN) were carried out and the results are given in Figures 2 - 4, respectively.

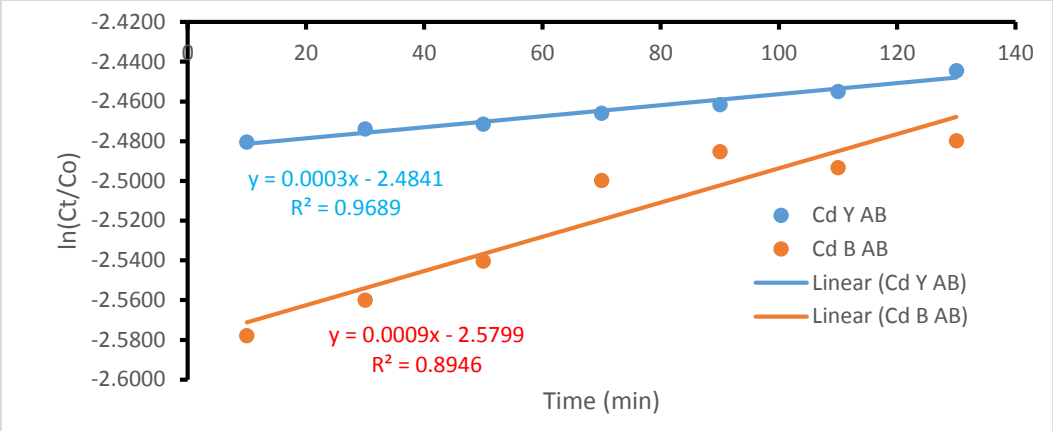

Figure 2: Adams-Bohart Model for Cadmium Biosorption onto *Bacillus Circulans* and *Saccharomyces Cerevisiae* [Q=10 ml/min, H=9 cm, d=1.0 cm]

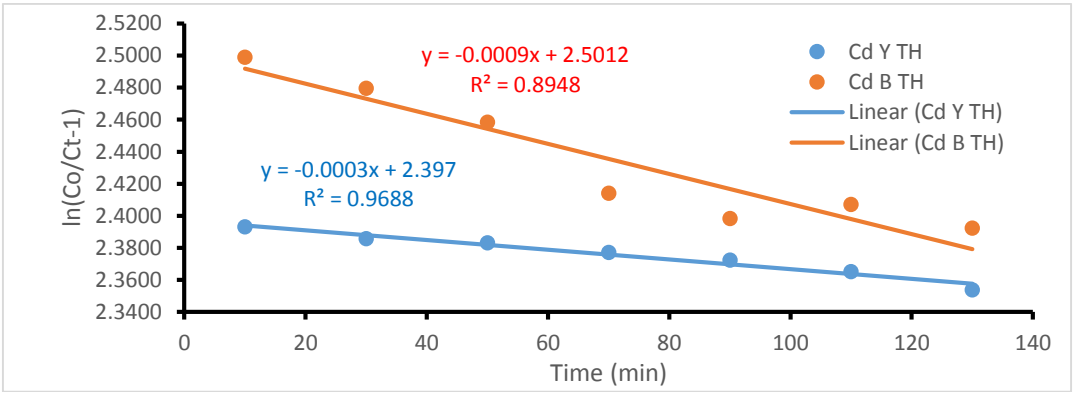

Figure 3: Thomas Model for Cadmium Biosorption onto *Bacillus Circulans* and *Saccharomyces Cerevisiae* [Q=10 ml/min, H=9 cm, d=1.0 cm]

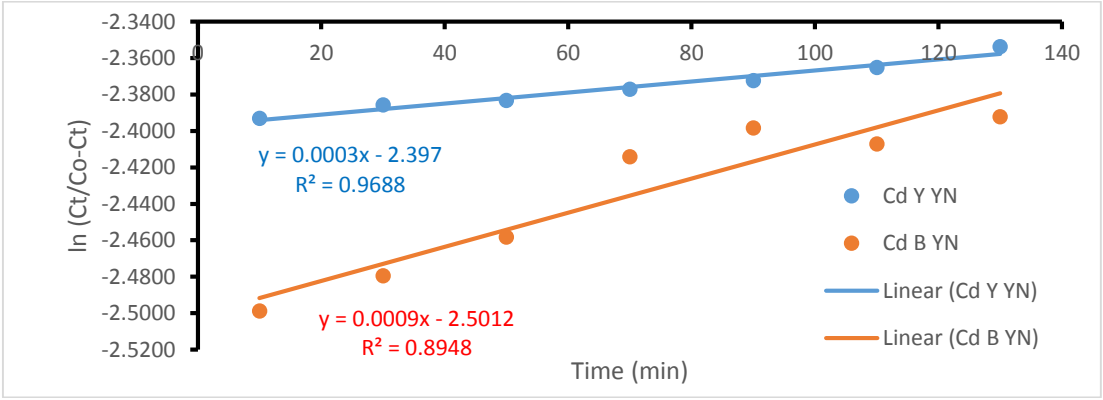

Figure 4: Yoon-Nelson Model for Cadmium Biosorption onto *Bacillus Circulans* and *Saccharomyces Cerevisiae* [Q=10 ml/min, H=9 cm, d=1.0 cm]

The value of R^2 ranged from $0.9688 - 0.9689$ for cadmium ions biosorption onto *Saccharomyces cerevisiae* ($Cd\ Y$) while cadmium ions biosorption onto Bacillus *circulans* ($Cd\ B$) R^2 values ranged from $0.8946 - 0.8948$ which are close to unity an indicative of fitness of the three models tested. From the value of R^2, it was observed that cadmium ions biosorption onto *Saccharomyces cerevisiae* ($Cd\ Y$) give a better fitness to cadmium ions biosorption onto Bacillus *circulans* ($Cd\ B$) for the three breakthrough curves models tested.

Figures 5 – 7 illustrate results of lead ions biosorption onto *Saccharomyces cerevisiae* ($Pb\ Y$) and Bacillus *circulans* ($Pb\ B$) for the Adams-Bohart, Thomas and Yoon-Nelson Models, respectively.

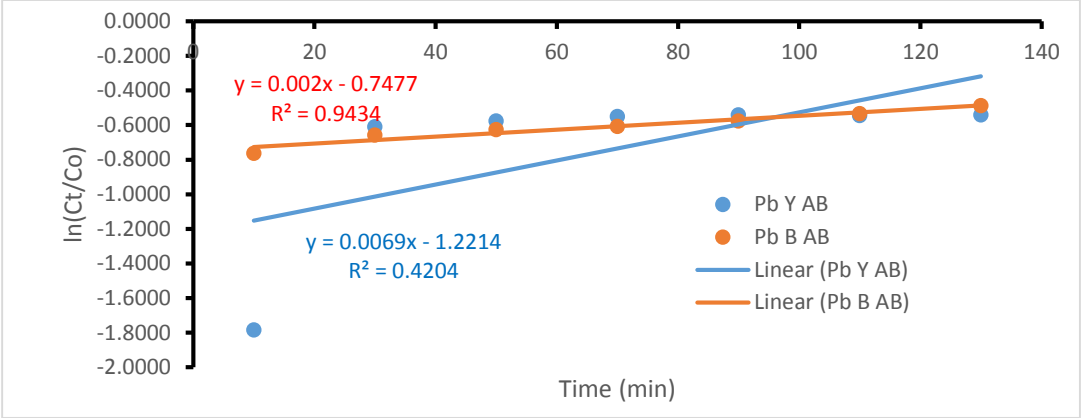

Figure 5: Adams-Bohart Model for Lead ion Biosorption onto *Bacillus Circulans* and *Saccharomyces Cerevisiae*
[Q=10 ml/min, H=9 cm, d=1.0 cm]

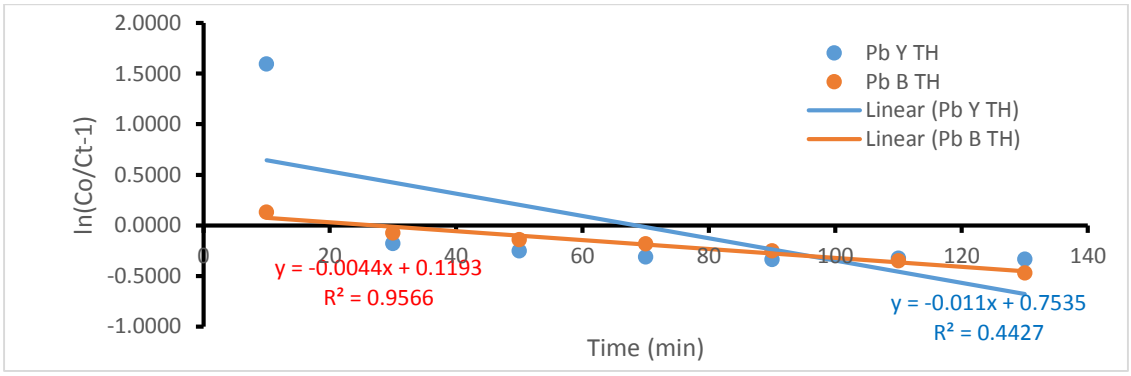

Figure 6: Thomas Model for Lead ion Biosorption onto *Bacillus Circulans* and *Saccharomyces Cerevisiae*
[Q=10 ml/min, H=9 cm, d=1.0 cm]

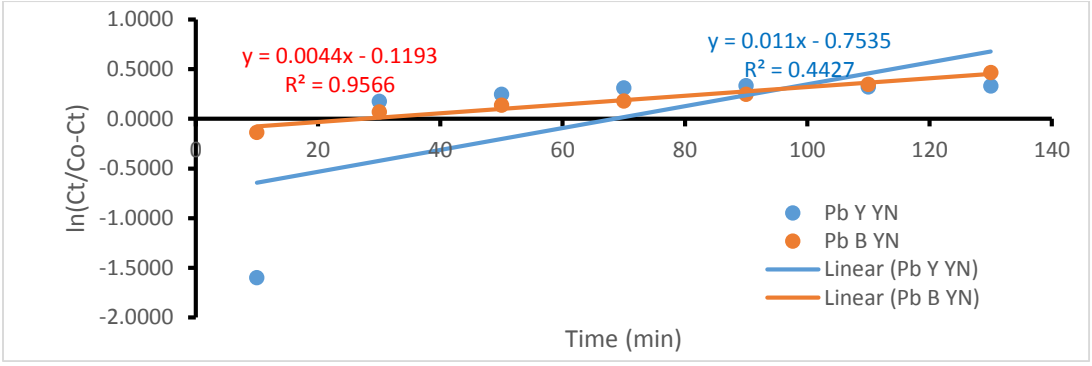

Figure 7: Yoon-Nelson Model for Lead ion Biosorption onto *Bacillus Circulans* and *Saccharomyces Cerevisiae*
[Q=10 ml/min, H=9 cm, d=1.0 cm]

It was noted that values of R^2 ranged from $0.4204 - 0.4427$ for lead ions biosorption onto *Saccharomyces cerevisiae* ($Pb\ Y$) whereas lead ions biosorption onto Bacillus *circulans* ($Pb\ B$) R^2 values ranged from $0.9434 - 0.9566$. The values of R^2 show that lead ions biosorption onto Bacillus *circulans* ($Pb\ B$) give better fitness to lead ions biosorption onto *Saccharomyces cerevisiae* ($Pb\ Y$) for the three breakthrough curves models tested.

Statistical Analysis, Simulated Data Validation and Models Kinetic Parameters of Fixed Bed Contactor

The breakthrough curves for Adams-Bohart, Thomas and Yoon-Nelson Models validation for continuous Fixed bed contactor are illustrated in Figures 8 - 11.

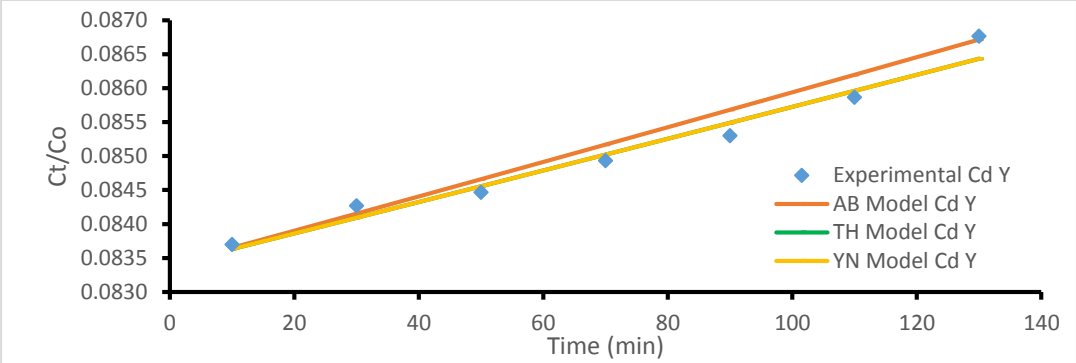

Figure 8: Models Validation for Cadmium Biosorption onto Saccharomyces *Cerevisiae*
[Q=10 ml/min, H=9 cm, d=1.0 cm]

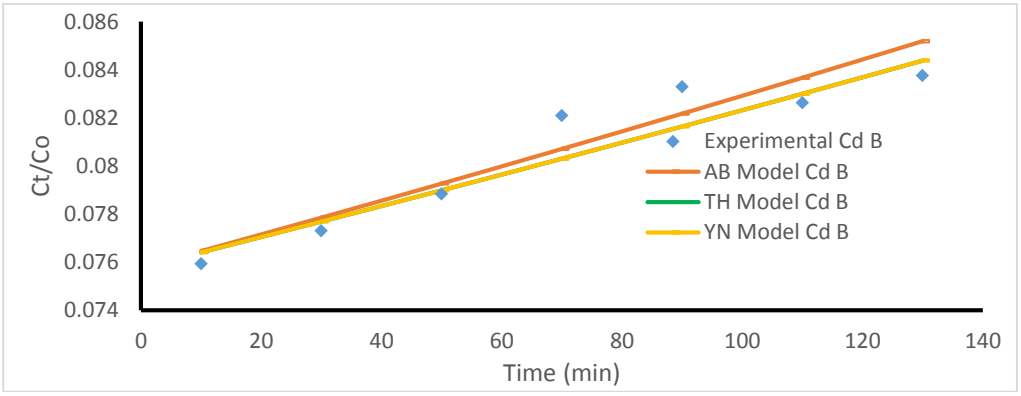

Figure 9: Models Validation for Cadmium Biosorption onto *Bacillus Circulans*
[Q=10 ml/min, H=9 cm, d=1.0 cm]

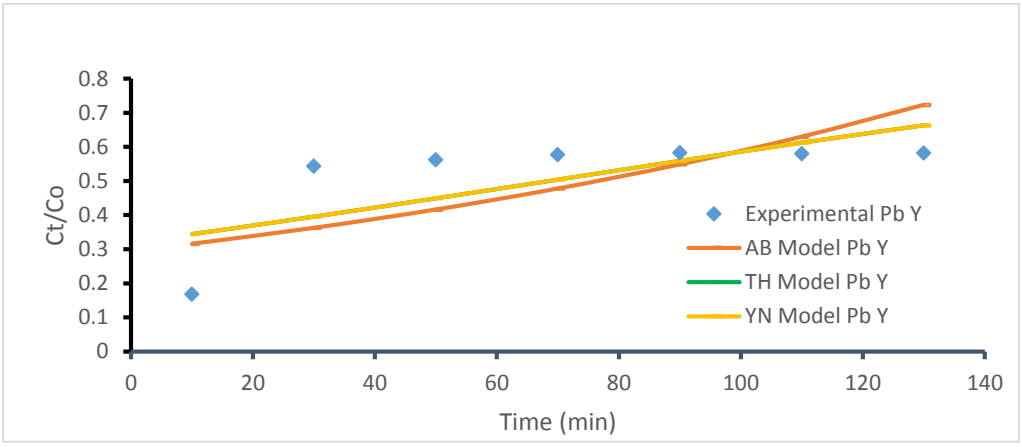

Figure 10: Models Validation for Lead Biosorption onto Saccharomyces *Cerevisiae*
[Q=10 ml/min, H=9 cm, d=1.0 cm]

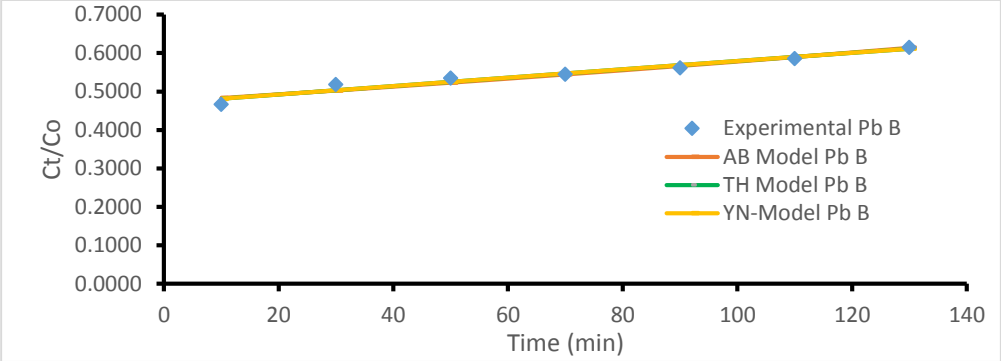

Figure 11: Models Validation for Lead Biosorption onto *Bacillus Circulans*
[Q=10 ml/min, H=9 cm, d=1.0 cm]

Kinetic parameters, R^2 and Sum of the Squares of Error (SSE) generated from simulation as well as experimental data are presented in Tables 1 – 3, in chronological order.

Table 1: Adams-Bohart Model and Validation Parameters for cadmium and lead onto *Bacillus Circulans* and *Saccharomyces Cerevisiae*

Sample	K_{AB} (ml/mg min)	N_o (mg/l)	R^2	SSE
$Cd\ Y$	$3.0\ X\ 10^{-6}$	1,171,427.96	0.9689	$4.6\ X\ 10^{-8}$
$Cd\ B$	$9.0\ X\ 10^{-6}$	405,531.61	0.8946	$8.84\ X\ 10^{-7}$
$Pb\ Y$	$1.725\ X\ 10^{-4}$	10,016.90	0.4204	$1.365\ X\ 10^{-2}$
$Pb\ B$	$5.0\ X\ 10^{-5}$	21,155.42	0.9434	$8.4\ 95\ X\ 10^{-5}$

Table 2: Thomas Model and Validation Parameters for cadmium and lead onto *Bacillus Circulans* and *Saccharomyces Cerevisiae*

Sample	K_{TH} (ml/mg min)	q_o (mg/g)	R^2	SSE
$Cd\ Y$	$3.0\ X\ 10^{-6}$	3,995,000.00	0.9688	$2.6\ X\ 10^{-8}$
$Cd\ B$	$9.0\ X\ 10^{-6}$	1,323,380.00	0.8948	$8.6\ X\ 10^{-7}$
$Pb\ Y$	$2.75\ X\ 10^{-4}$	13,700.00	0.4427	$9.9\ X\ 10^{-3}$
$Pb\ B$	$1.1\ X\ 10^{-4}$	5,164.50	0.9566	$7.6\ X\ 10^{-5}$

Table 3: Yoon-Nelson Model and Validation Parameters for cadmium and lead onto *Bacillus Circulans* and *Saccharomyces Cerevisiae*

Sample	K_{YN} (min^{-1})	τ (min)	q_{YN} (mg/g)	R^2	SSE
$Cd\ Y$	$3.0\ X\ 10^{-4}$	7990.00	3,995,000.00	0.9688	$2.6\ X\ 10^{-8}$
$Cd\ B$	$9.0\ X\ 10^{-4}$	2779.10	1,323,380.00	0.8948	$8.6\ X\ 10^{-7}$
$Pb\ Y$	$1.1\ X\ 10^{-2}$	68.50	13,700.00	0.4427	$9.9\ X\ 10^{-3}$
$Pb\ B$	$4.4\ X\ 10^{-3}$	27.11	5,164.50	0.9566	$7.6\ X\ 10^{-5}$

Table 1 gives kinetic parameters of Adams-Bohart model for cadmium and lead ions onto *Bacillus circulans* and *Saccharomyces cerevisiae* biosorbents. The Adams-Bohart model kinetic or rate constant (K_{AB}) and maximum volumetric biosorption capacity of bed (N_o) values were recorded as $3.0\ X\ 10^{-6}\ ml/mg\ min$ and $1{,}171{,}427.96\ mg/l$, respectively for cadmium ions biosorption onto *Saccharomyces cerevisiae* ($Cd\ Y$). The values of K_{AB} and N_o for cadmium ions biosorption onto Bacillus *circulans* ($Cd\ B$) were noted as $9.0\ X\ 10^{-6}\ ml/mg\ min$ and $405{,}531.61\ mg/$

l, respectively. Kinetic parameters of Adams-Bohart Model for lead ions onto *Saccharomyces cerevisiae* $(Pb\ Y)$ recorded K_{AB} and N_o values as $1.725\ X\ 10^{-4}\ ml/mg\ min$ and $10,016.90\ mg/l$, respectively. Lead ions biosorption onto Bacillus c*irculans* $(Pb\ B)$ measured K_{AB} and N_o values as $5.0\ X\ 10^{-5}\ ml/mg\ min$ and $21,155.42\ mg/l$, respectively. The higher the value of N_o the better the performance of the fixed bed contactor whereas lower value of K_{AB} is an indicative of better performance of the fixed bed contactor. The Adams-Bohart model kinetic rate constant shows an inverse relation with maximum volumetric biosorption capacity of bed (Yunnen *et al.*, 2017; Benjreid *et al.*, 2018). This implies that concerning the two heavy metals ions investigated cadmium ions biosorption shown a perfect fit for Adams-Bohart model using either *Bacillus circulans* or *Saccharomyces cerevisiae* as biosorbent compared to that of lead. To be precise, the overall fitness was demonstrated by cadmium ions biosorption onto *Saccharomyces cerevisiae* $(Cd\ Y)$ whereas Lead ions biosorption onto *Saccharomyces cerevisiae* $(Pb\ Y)$ shown the lowest fitness as presented in Table 1.

Kinetic parameters of Thomas Model for each heavy metal of cadmium and lead ions onto *Bacillus circulans* and *Saccharomyces cerevisiae* are presented in Table 2. The Thomas Model rate constant (K_{TH}) and maximum solid phase concentration of metals ion (q_o) values were noted as $3.0\ X\ 10^{-6}\ ml/mg\ min$ and $3,995,000.00\ mg/g$, respectively for cadmium ions biosorption onto *Saccharomyces cerevisiae* $(Cd\ Y)$. Cadmium ions biosorption onto *Bacillus circulans* $(Cd\ B)$ recorded K_{TH} and q_o values as $9.0\ X\ 10^{-6}\ ml/mg\ min$ and $1,323,380.00\ mg/g$, respectively. The values of K_{TH} and q_o for lead ions biosorption onto *Saccharomyces cerevisiae* $(Pb\ Y)$ were measured as $2.75\ X\ 10^{-4}\ ml/mg\ min$ and $13,700.00\ mg/g$, respectively. The values of K_{TH} and q_o for lead ions biosorption onto Bacillus *circulans* $(Pb\ B)$ were noted as $1.1\ X\ 10^{-4}\ ml/mg\ min$ and $5,164.50\ mg/l$, respectively. Thomas Model shown that the higher the value of q_o the better the performance of the column contactor whereas lower value of K_{TH} is an indicative of better performance of the column contactor (Benjreid *et al.*, 2018). Results in Table 2 shown that cadmium ions biosorption onto *Saccharomyces cerevisiae* $(Cd\ Y)$ gave better fitness of Thomas model to other five different contacting processes investigated whereas lowest fitness was illustrated by Lead ions biosorption onto Bacillus *circulans* $(Pb\ B)$.

Table 3 shows the results of Yoon-Nelson Model kinetic parameters for cadmium and lead ions onto *Bacillus circulans* and *Saccharomyces cerevisiae*. The Yoon-Nelson model total quantity of metal ions removal capacity of the column (q_{YN}) gives the same results for each metal ions biosorption onto biosorbent under investigation with Thomas model maximum solid phase concentration of metals ion (q_o) as shown in Table 2. Yoon-Nelson model rate constant (K_{YN}) and the time required for 50 % biosorbate breakthrough (τ) of cadmium ions biosorption onto *Saccharomyces cerevisiae* $(Cd\ Y)$ values were recorded as $3.0\ X\ 10^{-4}\ min^{-1}$ and 7990.00 min, respectively. Regarding, cadmium ions biosorption onto *Bacillus circulans* $(Cd\ B)$ values recorded for K_{YN} and τ were $9.0\ X\ 10^{-4}\ min^{-1}$ and $2779.10\ min$, respectively. The results for lead ions biosorption onto *Saccharomyces Cerevisiae* $(Pb\ Y)$ were measured as $1.1\ X\ 10^{-2}\ min^{-1}$ and $68.50\ min$ for K_{YN} and τ, respectively. The values of K_{YN} and τ for lead ions biosorption onto *Bacillus circulans* $(Pb\ B)$ were recorded as $4.4\ X\ 10^{-2}\ min^{-1}$ and $27.11\ min$, respectively. The Yoon-Nelson model illustrated that higher value of q_{YN} were obtained at higher τ and lower K_{YN} simultaneously. In addition, it was observed that K_{YN} is inversely proportional to τ. Hence, higher value of τ, q_{YN} and R^2 is an indicative of better performance of the column contactor. From Table 3, it is clearly illustrated that cadmium ions biosorption onto *Saccharomyces cerevisiae* $(Cd\ Y)$ gave better fitness of Yoon-Nelson model to others contacting processes investigated.

Figures 8 – 11 and Tables 1 - 3 clearly illustrated that cadmium ions biosorption onto *Saccharomyces cerevisiae* $(Cd\ Y)$ gives the best acceptable data with values of R^2 ranged from $0.9688 - 0.9689$ and SSE which ranged from $(2.6 - 4.6)\ X\ 10^{-8}$ for the three models validated. Lowest acceptable data were observed with lead ions biosorption onto *Saccharomyces cerevisiae* $(Pb\ Y)$ at values of R^2 ranged from $0.4204 - 0.4427$ and SSE gives the range from $9.9\ X\ 10^{-3} - 1.365\ X\ 10^{-2}$ for the three models validated (Rout *et al.*, 2014; Tejada-Tovar *et al.*, 2018). The results show that SSE values of $4.6\ X\ 10^{-8}$, $8.84\ X\ 10^{-7}$, $1.365\ X\ 10^{-2}$ and $8.4\ 95\ X\ 10^{-5}$ for Adams-Bohart model indicating lowest fitness to $2.6\ X\ 10^{-8}$, $8.6\ X\ 10^{-7}$, $7.46\ X\ 10^{-7}$, $7.83\ X\ 10^{-5}$, $9.9\ X\ 10^{-3}$ and $7.6\ X\ 10^{-5}$ recorded for Thomas model and Yoon-Nelson model for the six different heavy metal sorption onto adsorbent, respectively (Rout *et al.*, 2014; Tejada-Tovar *et al.*, 2018).

Conclusions

The following conclusions can be drawn from the results of this study:

1. The Adams-Bohart, Thomas and Yoon-Nelson models were used for modeling the performance of cadmium and lead ions in the fixed bed system with success.
2. The simulated and validated model data could be used for further scale up purposes of continuous fixed bed contactor for biosorption of cadmium and lead ions onto *Bacillus circulans* and *Saccharomyces cerevisiae*.
3. The better fitness was observed for biosorption of cadmium ions onto *Saccharomyces cerevisiae* in the fixed bed system to other three different contacting processes.
4. The R2009b - MAT LAB software as well as Microsoft Excel- 2016 software were successfully used as tools for modeling and simulation of the fixed bed system, respectively.

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