
Kinetic biosorption study of cadmium onto coconut copra meal as biosorbent

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Abstract: The sorption of cadmium ions on an agricultural by-product, coconut copra meal, was investigated. A batch sorption model, based on the assumption of the pseudo-second-order mechanism, was applied to predict the rate constant of sorption, the equilibrium sorption capacity and the initial sorption rate with the effect of initial cadmium concentration and temperature. Activation energy of sorption was determined based on the pseudo-second-order rate constants. In addition, the experimental results were analysed by pseudo-Langmuir, pseudo-Freundlich and pseudo-Redlich-Peterson isotherms based on pseudo-second order kinetic expression constant. Both pseudo-Langmuir and pseudo-Redlich-Peterson isotherms were found to represent the measured sorption data well. According to the evaluation using the pseudo-Langmuir equation, the monolayer sorption capacity was obtained to be 2.59 mg/g.

Keywords: copra meal; cadmium ions; pseudo-second-order; kinetics.

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1 Introduction

A low-cost sorbent is defined as one which is abundant in nature, or is a by-product or waste material from another industry (Bailey et al., 1999). The use of these low-cost materials could be an alternative to the more costly processes. The potential of biological materials as an alternative to treat low metal-bearing wastewaters has been investigated. Some of the biowastes reported to effectively sequester heavy metals include waste tea, exhausted coffee, exhausted coffee, nut and walnut shells (Orhan and Büyükgüngör, 1993), rice husk (Khalid et al., 1998), etc.

A number of biosorption processes for pollutants have been studied in an attempt to find a suitable explanation of the mechanism and the kinetics. In recent years, biosorption mechanisms involving kinetics-based models have been reported. Numerous kinetic models have described the reaction order of sorption systems based on solution concentration. These include first-order (Saiers et al., 1994) and second-order (McCoy and Liapis, 1991) reversible ones, first-order (Mohan et al., 2002) and second-order (Chu and Hashim, 2003) irreversible ones and pseudo-first-order (O'Shannessy and Winzor, 1996) and pseudo-second-order ones (Zaror, 1997) based on the solution concentration. On the other hand, reaction orders based on the capacity of the adsorbent, such as Lagergren's first-order equation (Lagergren, 1898), Zeldowitsch's model (Zeldowitsch, 1934) and Ho's pseudo-second-order expression (Ho, 1995; Ho and McKay, 1998a, 1999, 2000), have also been presented. In addition, the pseudo-second-order expression has been successfully applied to the sorption of metal ions, dyes, herbicides, oils and organic substances from aqueous solutions (Ho, 2004a; Zheng et al., 2005).

Copra meal produced in large amount has very little application apart from its use as animal feed owing to its high protein and fibre content. There is no information in the literature on the use of coconut copra as biosorbent. In this work, the kinetics of biosorption for the use of copra meal as biosorbent for cadmium ion removal from solution is investigated. In addition, pseudo-isotherms were also presented for the biosorption system.

2 Materials and methods

2.1 Materials

Coconut was obtained from the open market in Benin City. The fruits were dehusked, split and sun dried until the meat (endosperm) reached a moisture content of 5%.

The copra was blandered in a food-processing blender and the oil extracted with hot water. The copra meal was then washed several times with hot water to remove excess oil, sun dried and sieved to obtain a size less than 250 μm . The sieved copra meal was soaked in 0.02 mol/dm³ HCl overnight. The acid solution was filtered off, and the copra meal was washed with distilled water until the pH of the wash became neutral. The copra meal was dried at 373 K for 24 hours and stored in an air-tight container. The stock solution of cadmium(II) (1000 mg/dm³) was prepared in distilled water using cadmium nitrate salt (BDH); all working solutions were prepared by diluting the stock solution with distilled water.

2.2 Methods

Effect of initial concentration

A range of cadmium concentrations (10.5–201 mg/dm³) (adjusted to pH 6) were used, and the flasks agitated for 100 minutes. A 2.0 -g sample of copra meal was added to each 100 ml volume of cadmium solution and agitated at 150 rpm and the temperature was set at 24°C for all experiments. Samples (2.0 ml) were withdrawn at suitable time interval and the filtrate analysed by atomic absorption spectrophotometry (AAS).

Effect of temperature

Two gram of copra meal was added to a 500-ml flask containing 100 ml of 201 mg/dm³ of cadmium (pH 6) stirred at 150 rpm and at temperature 297 K. Samples (2.0 ml) were withdrawn at suitable time intervals and the filtrate analysed by Atomic Absorption Spectrophotometer (AAS). The experiment was repeated at 311, 323 and 333 K.

3 Results and discussion

The sorption of cadmium by copra meal was very rapid, regardless of the metal concentration (Figure 1). It may be further noted that most of the metal uptake by biosorbents occurred during the first few minutes, as in the case of sorption of lead onto tree fern (Ho et al., 2004). As the cadmium concentration is increased, the sorption capacity progressively decreased from 0.330 mg/g in 10.5 mg/dm³ to 2.35 mg/g in 201 mg/dm³ solution. Higher cadmium sorption at higher cadmium solution concentration may be because of the higher metal to sorbent ratio, which increases as the copra meal quantity is decreased. As the solution concentration was increased at a given quantity of sorbent, the cadmium/copra meal ratio increased, which in turn was reflected in an increased cadmium uptake per biosorbent. Fourest and Roux (1992) have explained it to be owing to the net cadmium ion availability around the microenvironment of the sorbent, and the electrostatic interaction between them at non-limiting sorption sites on copra meal.

The sorption may be represented by the pseudo-second-order kinetics (Ho and McKay, 1999, 2000) given by

$$\frac{dq_t}{dt} = k(q_e - q_t)^2 \quad (1)$$

where k is the pseudo-second-order rate constant. For boundary conditions $t = 0$ to $t = t$ and $q_t = q_t$, the integrated form of the equation is

$$\frac{1}{q_e - q_t} = \frac{1}{q_e} + kt \quad (2)$$

which can also be written as

$$q_t = \frac{1}{kq_e^2} + \frac{t}{q_e} \quad (3)$$

or, in the linear form,

$$\frac{t}{q_t} = \frac{1}{h} + \frac{1}{q_e} t \quad (4)$$

where $h = kq_e^2$ can be regarded as the initial sorption rate as t approaches 0. If the pseudo-second-order kinetics is applicable, the plot of t/q_t vs. t gives a linear relationship, which allows computation of q_e , k , and h without having to know any parameter before hand.

Figure 1 Effect of initial concentration on the sorption of cadmium ions onto copra meal

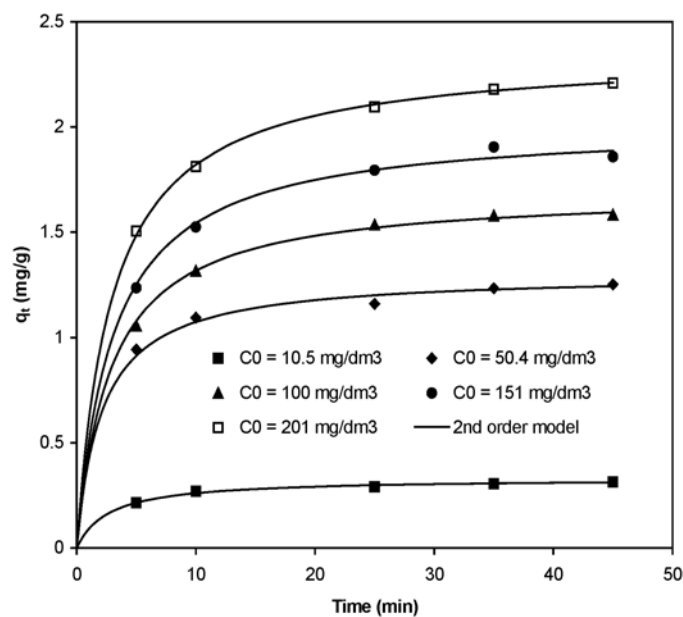


Figure 1 shows a plot of the experimental data points for the sorption of cadmium by copra meal as a function of time. These plots show that, for all initial cadmium concentrations, the amount of cadmium sorbed increases rapidly with time at the beginning and becomes very slow towards the end of the process. The plots also demonstrated that the sorption increased for lower initial cadmium concentrations at any specific time. Furthermore, a large fraction of the total amount of cadmium was removed within a short period of time.

Table 1 lists the rate constant, k , the equilibrium sorption, q_e , the initial sorption rate, h and the coefficients of determination, r^2 , of sorption at various initial concentrations of cadmium; these were calculated from the intercept and slope of the straight line plots of t/q_t vs. t according to equation (4). The initial sorption rate increased with an increase in the initial concentration of cadmium ions in solution. Thus, the initial sorption rate varied from 0.123 mg/g min to 0.805 mg/g min as the initial concentration of cadmium ions was varied from 10.5 mg/dm³ to 201 mg/dm³. The values of rate constants decreased from 1.12 g/mg min to 0.145 g/mg min with an increase in the initial concentration of cadmium from 10.5 mg/dm³ to 201 mg/dm³. The observed dependence of the rate on the initial concentration of cadmium indicates that removal of cadmium from dilute aqueous solution should be achieved much easily, since sorption relative to the initial concentration of cadmium is more rapid in dilute solution, i.e., a greater fraction of the cadmium present will be sorbed in a given period of time (Weber and Morris, 1963).

Table 1 Parameters for the effect of initial cadmium concentration

C_0 , mg/dm ³	q_e , mg/g	k , g/mg min	h , mg/g min	R^2
10.5	0.330	1.12	0.123	0.999
50.4	1.30	0.358	0.609	0.999
100	1.70	0.206	0.593	1.000
151	2.02	0.163	0.660	0.999
201	2.35	0.145	0.805	1.000

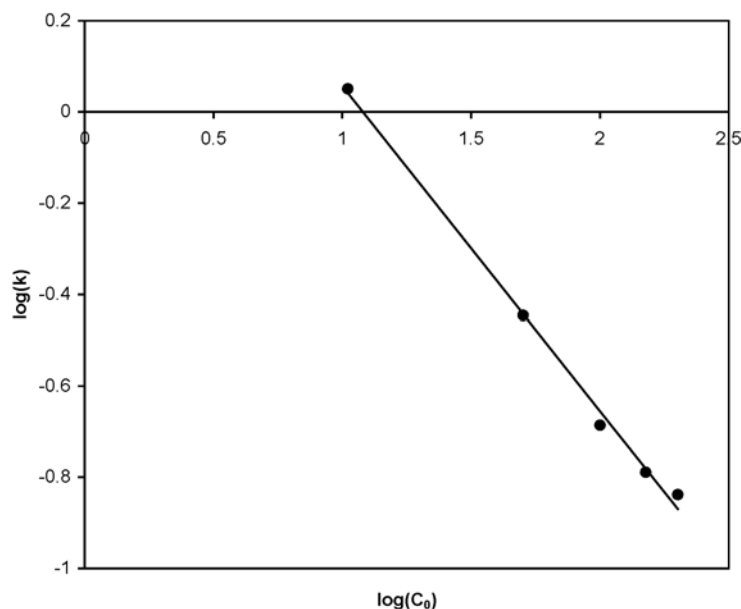
A correlation has been made empirically of the pseudo-second-order rate constant, k , as a function of the initial concentration of lead as follows:

$$k = mC_0^n. \quad (5)$$

McKay et al. (1983) reported that, in the case of sorption of Acid Blue 25 using chitin, n centres around the 0.5 values, confirming that intraparticle diffusion was a prominent factor in the sorption process. In the cases of sorption of larger molecular dyes, the sorption rate was slower, and intraparticle diffusion was the rate-limiting step (Jadhav and Vanjara, 2004; Sun and Yang, 2003). Conversely, in the cases of sorption of metal ions, the sorption rate was faster generally and was not concerted by intraparticle diffusion (Prasad and Saxena, 2004; Ho et al., 2004). The results from this study shown in Figure 2 suggest that the relationship between the pseudo-second-order rate constant, k , and the initial cadmium concentration follows equation (5) with a coefficient of determination of 0.996 as shown in the following equation:

$$k = 5.86C_0^{-0.71}. \quad (6)$$

The value of n value (-0.71) is different from 0.5, based on intraparticle diffusion. This indicates that the kinetics of cadmium sorption using coconut copra meal was described by the pseudo-second-order expression rather than by the intraparticle diffusion, and intraparticle diffusion was not the rate-limiting step. In addition, the negative n value was also found as in the case of sorption of lead onto peat (Ho and McKay, 1998b).

Figure 2 Logarithmic relationship between k and C_0 

3.1 Pseudo-isotherms

Pseudo-isotherms could be obtained using a pseudo-second order kinetic expression constant (Ho, 2004b). Sorption of cadmium onto coconut copra meal was tested by applying the pseudo-Langmuir, pseudo-Freundlich and pseudo-Redlich-Peterson isotherms. The amount of cadmium sorbed at equilibrium, q_e , can be obtained from pseudo-second-order equation, and the equilibrium cadmium concentration can be calculated from the equation below (Ho, 2004b; Ho and Wang, 2004):

$$C_{ep} = C_0 - \frac{q_e m_s}{V}. \quad (7)$$

C_{ep} is the equilibrium cadmium concentration, mg/dm³; C_0 is the initial cadmium concentration, mg/dm³; q_e is the equilibrium sorption capacity obtained from pseudo-second order rate equation, mg/g; V is solution volume, dm³ and m_s is mass of mixed sorbent, g.

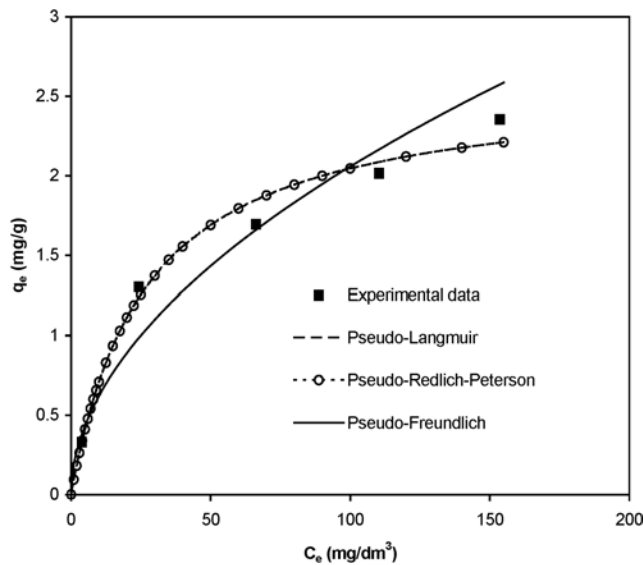
The isotherms and their respective linear forms are shown in Table 2. The coefficient of determination, r^2 , has been used to determine the relationship between the experimental data and the isotherms in most studies. The coefficients of determination were found to be 0.999 for pseudo-Langmuir, 0.996 for pseudo-Redlich-Peterson, and 0.955 for pseudo-Freundlich isotherm, respectively. Figure 3 shows the better-fitting isotherms were pseudo-Langmuir and pseudo-Redlich-Peterson isotherms that were overlapped. The sorption behaviour of the cadmium onto coconut copra meal was examined by using the least squares linear regression analysis. The values of the pseudo-Langmuir isotherm constants were obtained from the slope and intercept of the straight line (Table 2). The saturated monolayer sorption capacity, q_m , and sorption equilibrium constant, K_a , were 2.59 mg/g and 0.0375 dm³/mg for pseudo-Langmuir

isotherm; the pseudo-Freundlich sorption isotherm constant K_F was 0.178 and $1/n$ was 0.521 and the pseudo-Redlich-Peterson isotherm constant A was 0.975, B was 1.82 and g was 1.000. Under the basic condition of this sorption system, it was estimated to be more likely to be approaching the Langmuir form where it can be seen from the data where the value of g was unity. Langmuir isotherm is a special case of Redlich-Peterson isotherm when constant g was unity.

Table 2 Isotherms and their linear forms

Isotherm	Isotherm	Linear form
Langmuir	$q_e = \frac{q_m K_a C_e}{1 + K_a C_e}$	$\frac{1}{q_e} = \frac{1}{K_a q_m} + \frac{1}{q_m} C_e$
Freundlich	$q_e = K_F C_e^{1/n}$	$\log(q_e) = \log(K_F) + 1/n \log(C_e)$
Redlich-Peterson	$q_e = \frac{AC_e}{1 + BC_e^g}$	$\ln\left(\frac{C_e}{q_e} - 1\right) = g \ln(C_e) + \ln(B)$

Figure 3 Theoretical isotherms and experimental data



3.2 Effect of temperature

The effect of increasing temperature on the uptake of cadmium on copra meal is shown in Figure 4. Figure 5 shows excellent agreement between the experimental data and theoretical curves through the use of equation (4); this suggests that the sorption system follows a pseudo-second-order rate model. Table 3 shows that the pseudo-second-order rate constants for the cadmium/copra meal system increased steadily from 0.173 to 0.640 g/mg min for temperature increase from 297 K to 333 K. The sorption capacity decreased from 2.27 mg/g to 0.296 mg/g, and the initial sorption rate decreased from 0.895 mg/g min to 0.0559 mg/g min, when the temperature was increased from 297 K to 333 K. The increase in the equilibrium sorption of cadmium with temperature indicates

that a low temperature favours cadmium removal by sorption on copra meal because an elevation in temperature increases the escaping tendency of cadmium from an interface (Pandey et al., 1984). This effect suggests that an explanation of the sorption mechanism associated with the removal of cadmium onto copra meal involves a physical process. Similar decreases in cadmium capacity with temperature was also observed by using biosorbents such as *Chlorella vulgaris* (Aksu, 2001) and dead *Sargassum* sp. (Cruz et al., 2004) as well as hematite (Singh et al., 1998) and cadmium on chitin (Benguella and Benaissa, 2002). This indicates that the biosorption process of cadmium onto coconut copra meal is exothermic.

Figure 4 Effect of temperature on the sorption of cadmium ions onto copra meal

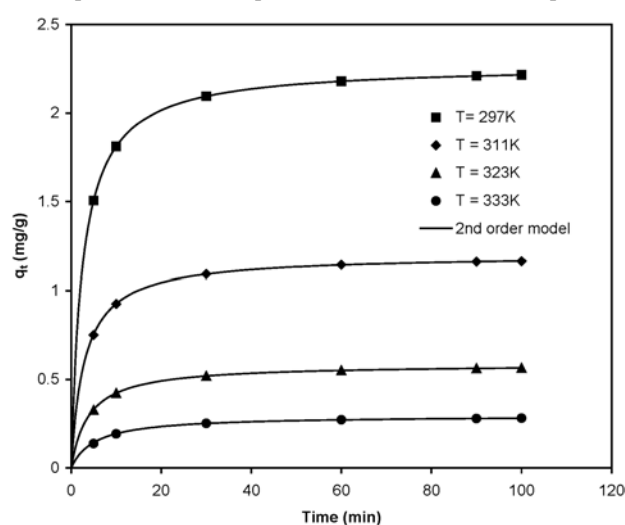


Figure 5 Pseudo-second-order sorption kinetics of cadmium ions onto copra meal at various temperatures

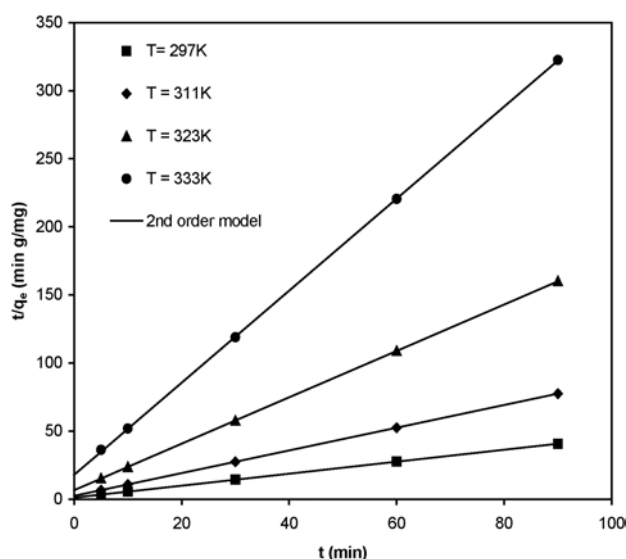


Table 3 The effect of changing temperature on cadmium adsorption at 201 mg/dm³

<i>T</i> , K	<i>q_e</i> , mg/g	<i>k</i> , g/mg min	<i>h</i> , mg/g min	<i>R</i> ²
297	2.27	0.173	0.895	1.000
311	1.20	0.283	0.407	1.000
323	0.587	0.439	0.151	1.000
333	0.296	0.640	0.0559	1.000

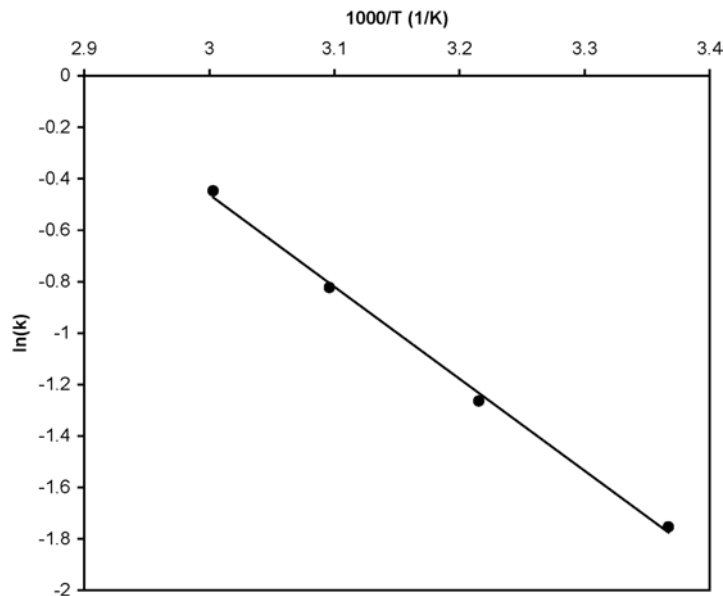
A linear relationship was found to exist between the pseudo-second-order rate constant, *k* and the reciprocal of the reaction temperature ($r^2 = 0.997$). Therefore, the sorption rate constant may be expressed as a function of temperature by the following relationship:

$$k = k_0 \exp\left\{\frac{-E}{RT}\right\} \quad (8)$$

The relationship between *k* and *T* can be represented in an Arrhenius form as

$$k = 2.90 \times 10^4 \exp\left\{\frac{-29.8}{8.314T}\right\} \quad (9)$$

From the equation, the temperature-independent factor, *k*₀, is 2.90×10^4 g/mg min. Furthermore, the activation energy of sorption, *E*, is 29.8 kJ/mole for the sorption system of cadmium on copra meal (Figure 6). Singh et al. (1998) obtained activation energy of 42.2 kJ/mole for sorption of cadmium on hematite. Because the sorption of cadmium onto copra meal is an exothermic process, an increasing solution temperature would be expected to result in a decreased sorption capacity.

Figure 6 Relationship between rate constant and temperature

4 Conclusion

The removal of cadmium ions by coconut copra meal was a function of initial cadmium ion concentration and temperature. The sorption capacity of cadmium ions from solution increased with increasing initial cadmium concentration and decreasing reaction temperature. The kinetics of cadmium ion sorption onto copra meal was based on the assumption of the pseudo-second-order explanation. An activation energy of sorption has been obtained based on the pseudo-second-order rate constants. The Redlich-Peterson and the Langmuir isotherms have higher coefficients of determination than the Freundlich isotherm for the sorption of cadmium onto coconut copra meal. Langmuir isotherm is a special case of Redlich-Peterson isotherm when constant g was unity.

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**CONTENTS, KEYWORDS AND AUTHOR INDEXES
FOR VOLUME 34**

Contents Index**Volume 34, 2008**

Issue Nos. 1/2/3/4**SPECIAL ISSUE: APPLICATION OF BIOSORBENTS ON WASTEWATER
TREATMENT****Guest Editor: Prof. Yuh-Shan Ho****Part I**

- 1 **Bibliometric analysis of biosorption technology in water treatment research from 1991 to 2004**
Yuh-Shan Ho
- 14 **Biosorption study in a mining wastewater reservoir**
Ghinwa Naja, Christian Mustin, Bohumil Volesky and Jacques Berthelin
- 28 **Removal of cadmium from contaminated waters using saltbush (*Atriplex canescens*) biomass: identification of Cd binding sites**
Maather F. Sawalha, Jose R. Peralta-Videa, Jason G. Parsons, Jorge H. Gonzalez and Jorge L. Gardea-Torresdey
- 43 **Treatment of tannery wastewater in a hybrid biological reactor**
Debabrata Mazumder, Somnath Mukherjee and Pradip K. Ray
- 57 **Biosorption of lead onto mimosa tannin resin: equilibrium and kinetic studies**
Harun Türkmenler, Mahmut Özacar and İ. Ayhan Şengil
- 71 **Removal of acid dyes from aqueous solutions using orange peel as a sorbent material**
H. Benaïssa
- 83 **Removal and recovery of uranium (VI) using a fungal based low-cost biosorbent *Ganoderma lucidum***
Mukul Kulshrestha and C. Venkobachar
- 97 **Adsorption of aniline onto hexagonal mesoporous silicate monoliths (HOM-2)**
Sherif A. El-Safty, Fujio Mizukami and Takaaki Hanaoka

- 478 *Contents Index*
- 111 **Adsorption of anionic dyes on Fe(III)-loaded collagen fibre from aqueous solution**
Ying-Chun Gu, Xue-Pin Liao, Yan-Jie Huang and Bi Shi
- 122 **Biosorption kinetics and isotherms of copper (II) ions from aqueous solution and application of the MATLAB**
Recep İleri and Gülümser Çakir
- 140 **Slurry sampling electrothermal atomic absorption spectrometric determination of palladium in water using biosorption with inactive baker's yeast *Saccharomyces cerevisiae***
Beata Godlewska-Żyłkiewicz
- 151 **Biosorption of acid dye on *Pueraria lobata ohwi*. Equilibrium and kinetic modelling**
B. Koumanova, B. Achova and S.J. Allen
- Part II**
- 164 **Biosorption of chlorophenols: a review**
T. Mathialagan and T. Viraraghavan
- 195 **Biosorption of Pb(II) from synthetic wastewater onto *Pseudomonas aeruginosa***
So-Young Kang, Jong-Un Lee and Kyoung-Woong Kim
- 203 **Quaternised biomass as anion exchanger for the removal of fluoride from water**
C. Namasivayam and W.H. Hoell
- 215 **The kinetics of copper sorption onto yohimbe bark wastes**
C. Escudero, N. Fiol, J. Poch and I. Villaescusa
- 231 **Metal ions biosorption from dilute aqueous solution**
K.A. Matis, A.I. Zouboulis, N.K. Lazaridis and Th.D. Karapantsios
- 246 **Ethylenediamine modified rice hull as a sorbent for the removal of Basic Blue 3 and Reactive Orange 16**
C.K. Lee, S.T. Ong and Z. Zainal
- 261 **Adsorption of model textile dyes from aqueous solutions using agricultural wastes as adsorbents: equilibrium, kinetics and fixed bed column study**
Sampa Chakrabarti, Basab Chaudhuri and Binay K. Dutta
- 275 **Use of various agricultural wastes for the removal of heavy metal ions**
Sibel Kahraman, Nukhet Dogan and Sema Erdemoglu
- 285 **Optimisation of cell surface and structural components for improving adsorption capacity of *Pseudomonas putida* 5-x to Cu²⁺**
G.H. Zheng, L. Wang, Q. Zhou and F.T. Li

- 297 **Biosorption of Ag⁺ from aqueous solution by *Streptomyces rimosus* biomass**
M.Z. Bakhti, A. Selatnia and G.A. Junter
- 308 **Adsorption of Acid Yellow 99 onto DEDMA-sepiolite from aqueous solutions**
A. Safa Özcan and Adnan Özcan
- Part III**
- 325 **A methodological approach for dynamic modelling of membrane integrated biosorption: a case study on *Sphaerotilus natans* as biosorbent**
F. Beolchini, F. Pagnanelli, L. Toro and F. Vegliò
- 340 **Biosorption performance of red and green marine macroalgae for removal of trace cadmium and nickel from wastewater**
M.C. Basso and A.L. Cukierman
- 353 **Biosorption of lead(II) by free and immobilised fungal biomass of *Phanerochaete chrysosporium*: a comparative study**
M. Iqbal, A. Saeed and R.G.J. Edyvean
- 364 **Removal of Ni²⁺ from aqueous solution by rice bran**
Xue-Song Wang and Yong Qin
- 374 **Interactions of Pb(II), Cd(II) and Cr(VI) with Neem (*Azadirachta indica*) leaf powder: kinetics and thermodynamics**
Arunima Sharma and Krishna G. Bhattacharyya
- 400 **Effect of sludge age on heavy metal biosorption by selector activated sludge**
Ufuk Alkan and Sevil Çalışkan Eleren
- 412 **Carboxylic acids-modified palm stones as sorbents for metal ions using single and multi-metal aqueous solutions**
Sameer Al-Asheh and Ahmad Belal
- 427 **Innovative use of silvichemical biomass and its derivatives for heavy metal sorption from wastewater**
Hong Yu, Geoff H. Covey and Andrea J. O'Connor
- 451 **Adsorption of C.I. Basic Blue 9 on chitosan-based materials**
Grégorio Crini, Bernard Martel and Giangiacomo Torri
- 466 **Kinetic biosorption study of cadmium onto coconut copra meal as biosorbent**
Augustine E. Ofomaja and Yuh-Shan Ho
-

Indexing is based on the key words and phrases, title and abstract on the first page of each paper. Page references are to the first page of the paper or report

A

acid dye Erionil Marine Blau	151
acid dye	308
activated sludge hybrid reactor	43
activated sludge	400
adsorbent	111, 451
adsorption capacity	285
adsorption kinetics	364
adsorption mechanism	83
adsorption	111, 203, 308, 412, 451
aerobic selector	400
agricultural waste	275
aluminosilicates	97
aniline solute	97
apricot seed	275

B

Basic Blue 3	246
batch processing	297
BDST model	261
biomass	164, 231
biosorbent	231
biosorption kinetics	57
biosorption	1, 83, 122, 140, 151, 164, 195, 275 297, 325, 340, 353, 400, 427

C

C.I. Basic Blue 9	451
cadmium binding	28
cadmium ions	466
CA-PS	412
cations	231
Cd(II)	374
cell culture	285
cell growth age	285
cell pretreatment	285
chitosan	451
chlorophenols	164
chrome tannery	43
citation	1
collagen fibre	111
competition	195

composite wastewater	43
copper	122, 215, 285, 400
copra meal	466
cotton stalk	275
Cr(VI) adsorption	374
D	
desorption	83, 203
dyes	71, 111
dynamic modelling	325
E	
Elovich equation	57
eluant	83
equilibrium studies	83
equilibrium	151, 325
ETAAS	140
ethylenediamine	246
EXAFS	28
F	
Fe(III)	111
Fe(III)-loaded collagen fibre	111
fluoride	203
FTIR-ATR	215
G	
<i>Ganoderma lucidum</i>	83
gram husk	261
groundnut shell	261
H	
heavy metal	275, 325, 400, 427
heavy metals removal	340
hexagonal structures	97
hexavalent chromium	400
I	
ICP/OES	28
immobilised fungal biomass	353
interfering anions	203
ion exchange	215, 427
isotherm	57, 122, 203, 308, 364
K	
kinetic studies	83
kinetic	122, 151, 231, 308, 374, 466
kudzu	151

L	
Lagergren	261
lead	57
lead(II)	353
loofa sponge	353
M	
MA-PS	412
mechanisms	164
membrane processes	325
mesoporous silica	97
metal desorption	14
metal sorption	14
mimosa tannin resin	57
mining wastewater	14
MND-PS	412
modelling	231
monolithic-adsorbents	97
multimetal systems	325
N	
Neem leaf powder	374
Ni ²⁺ removal	364
O	
OA-PS	412
orange peel	71
oxyanions	231
P	
palladium	140
palm stone	412
Pb(II)	195, 374
pH effect	203
pH	164
<i>Phanerochaete chrysosporium</i>	353
potentiometric titration	14
pseudo second order kinetics	261
pseudo-first order equation	215
<i>Pseudomonas aeruginosa</i>	195
pseudo-second order equation	57, 215
pseudo-second-order	466
Q	
quaternised biomass	203

R	
<i>R. arrhizus</i>	122
Reactive Orange 16	246
red and green marine macroalgae	340
removal	71
removal and recovery of uranium	83
removal efficiency	195
rice bran	364
rice hull	246
S	
saltbush	28
SA-PS	412
SCI	1
scientometrics	1
SEM-EDX	215
separation	231
sepiolite	308
shaft-type	43
silver	297
silvichemical biomass	427
sludge age	400
slurry sampling	140
sorption	71, 246
<i>Streptomyces rimosus</i>	297
surfactant	308
T	
tangential flow	325
TA-PS	412
thermodynamic	364, 374
toxicity	275
two-parameter model	246
type of biomass	164
tyre-tube beads	43
U	
ultrafiltration	325
W	
waste sludge	122
waste-water treatment	297
wastewater treatment	427, 451
wastewater	340
water treatments	97

484 *Keywords Index*

X

XANES 28

Y

yeast 140

yohimbe bark wastes 215

Author Index**Volume 34, 2008**

Achova, B.	151	Li, F.T.	285
Al-Asheh, S.	412	Liao, X-P.	111
Alkan, U.	400	Martel, B.	451
Allen, S.J.	151	Mathialagan, T.	164
Bakhti, M.Z.	297	Matis, K.A.	231
Basso, M.C.	340	Mazumder, D.	43
Belal, A.	412	Mizukami, F.	97
Benaïssa, H.	71	Mukherjee, S.	43
Beolchini, F.	325	Mustin, C.	14
Berthelin, J.	14	Naja, G.	14
Bhattacharyya, K.G.	374	Namasivayam, C.	203
Çakir, G.	122	O'Connor, A.J.	427
Chakrabarti, S.	261	Ofomaja, A.E.	466
Chaudhuri, B.	261	Ong, S.T.	246
Covey, G.H.	427	Özacar, M.	57
Crini, G.	451	Özcan, A.	308
Cukierman, A.L.	340	Özcan, A.S.	308
Dogan, N.	275	Pagnanelli, F.	325
Dutta, B.K.	261	Parsons, J.G.	28
Edyvean, R.G.J.	353	Peralta-Videa, J.R.	28
Eleren, S.Ç.	400	Poch, J.	215
El-Safty, S.A.	97	Qin, Y.	364
Erdemoglu, S.	275	Ray, P.K.	43
Escudero, C.	215	Saeed, A.	353
Fiol, N.	215	Sawalha, M.F.	28
Gardea-Torresdey, J.L.	28	Selatnia, A.	297
Godlewska-Żyłkiewicz, B.	140	Şengil, I.A.	57
Gonzalez, J.H.	28	Sharma, A.	374
Gu, Y-C.	111	Shi, B.	111
Hanaoka, T.	97	Toro, L.	325
Ho, Y-S.	1, 466	Torri, G.	451
Hoell, W.H.	203	Türkmenler, H.	57
Huang, Y-J.	111	Vegliò, F.	325
İleri, R.	122	Venkobachar, C.	83
Iqbal, M.	353	Villaescusa, I.	215
Junter, G.A.	297	Viraraghavan, T.	164
Kahraman, S.	275	Volesky, B.	14
Kang, S-Y.	195	Wang, L.	285
Karapantsios, Th.D.	231	Wang, X-S.	364
Kim, K-W.	195	Yu, H.	427
Koumanova, B.	151	Zainal, Z.	246
Kulshrestha, M.	83	Zheng, G.H.	285
Lazaridis, N.K.	231	Zhou, Q.	285
Lee, C.K.	246	Zouboulis, A.I.	231
Lee, J-U.	195		

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