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Single and multielementary isotherms of toxic metals in aqueous solution using treated coconut shell powder

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ABSTRACT

Solid residues of the agroindustries are abundantly available at low cost and, because of their adsorption properties, they could be advantageously used for the treatment of industrial effluents. In this work, base-treated green coconut shell powder was applied to remove toxic metals from synthetic aqueous effluents. It was shown that Langmuir and Freundlich isotherms describe the adsorption features in single and multicomponent systems.

Keywords: Adsorption; Green coconut shell powder; Toxic metals; Adsorption isotherms

1. Introduction

Residues from industrial activities often contain toxic metals [1], in particular aqueous industrial effluents [2]. Environmental dispersion and bioaccumulation in the human body cause great concerns [3,4] in view of the toxic and carcinogenic properties associated to some metals [5] and to all transition metals [6–8]. Most techniques to remove the polluents are expensive [9,10], while methods based on adsorption on cheap natural and renewable materials offer interesting alternatives [11–13]. Most research efforts have highlighted the low cost, the easy applicability and the low environmental impact [14–16].

In particular, the use of residues derived from agricultural practices for the treatment of industrial wastewater seems feasible [17] and residues from various sources including carrots, peanut shell, rice husks, walnuts, sugarcane bagasse and neam leaf have been successfully used for the purpose [18–22].

Most studies have been carried out on the removal of single or binary metal ions from aqueous solution by diverse types of low cost adsorbents [23–26]. In the practice, the adsorption metal ions from wastewater generally involve the simultaneous presence of metal ions in wastewater promoting a competition between different metal ions by adsorption site. However, study of equilibrium modeling of multi-metal ions is essential for understand the real system, but usually are neglected [27].

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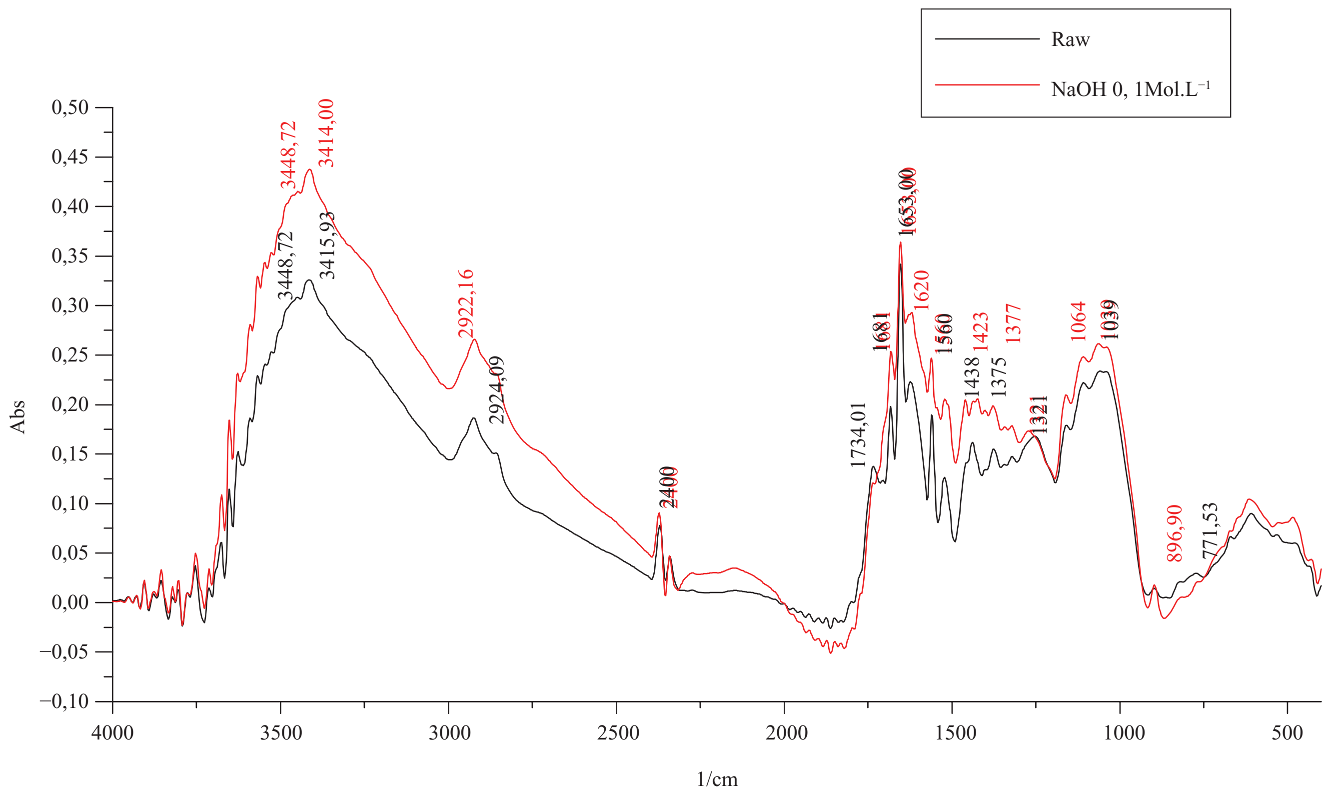

Fig. 1. IR spectrum (KBr) of green coconut shell powder treated with NaOH 0.1 mol L^{-1}/3 h and raw green coconut shell powder.

In this work, the application of green coconut shell powder (*Cocos nucifera*) pre-treated with sodium hydroxide to remove toxic metals from aqueous industrial effluents has been studied and we wish to report the result herein. Equilibrium isotherms of single and multicomponent of Cd^{2+}, Cu^{2+}, Ni^{2+}, Pb^{2+} and Zn^{2+} on the green coconut shell were modeled. Furthermore, the competitive adsorption effects of the metal ions on the green coconut shell showing the best effectiveness for the uptake of the metal ions was investigated using the extended Langmuir model.

2. Materials and methods

2.1. Materials

Stock solutions 1 g L^{-1} of selected metal ions were prepared from the corresponding salts, namely $Cu(NO_3)_2{\cdot}4H_2O$, $Zn(NO_3)_2{\cdot}6H_2O$, $Cd(NO_3)_2{\cdot}4H_2O$, $Pb(NO_3)_2$ and $Ni(NO_3)_2{\cdot}6H_2O$ (Merck, São Paulo, Brazil), using deionized water and a single and multicomponent standard solution (Cd^{2+}-Cu^{2+}-Ni^{2+}-Pb^{2+}-Zn^{2+}) 100 mg L^{-1} at pH 5.0) was prepared as well. The concentrations of the ions were determined by atomic absorption spectrophotometry (AAS, GBC 933 *plus* model) in an air-acetylene f1ame.

2.2. Preparation of the adsorbent

The green coconut shell used in this work were supplied by Embrapa Tropical Agroindustry – CE (EMBRAPA/CE). The method used in the production of the green coconut shell powder was described by Sousa [12]. The material was characterized by infrared spectroscopy (FTIR, Shimadzu, Prestige) and the morphological characteristics were determined by scanning electron microscopy (MEV, Philips, SEM - XL30).

2.3. Adsorption experiments

Batch experiments were performed at ambient temperature (28°C) in erlenmeyer flasks, containing 0.40 g green coconut shell powder treated with 10 mL of an aqueous single and multicomponent solution (Cd^{2+}-Cu^{2+}-Ni^{2+}-Pb^{2+}-Zn^{2+}) at pH 5.0. Metal concentrations varied between 10.0 and 1.0 g L^{-1}. After shaking the flasks and filtration, the filtrate was analyzed by atomic absorption spectrophotometry (AAS).

The adsorption capacity, Q_e (mg metal g^{-1} adsorbent) was determined based on the difference of the concentrations of the dissolved metal ions using following equation [28,29]:

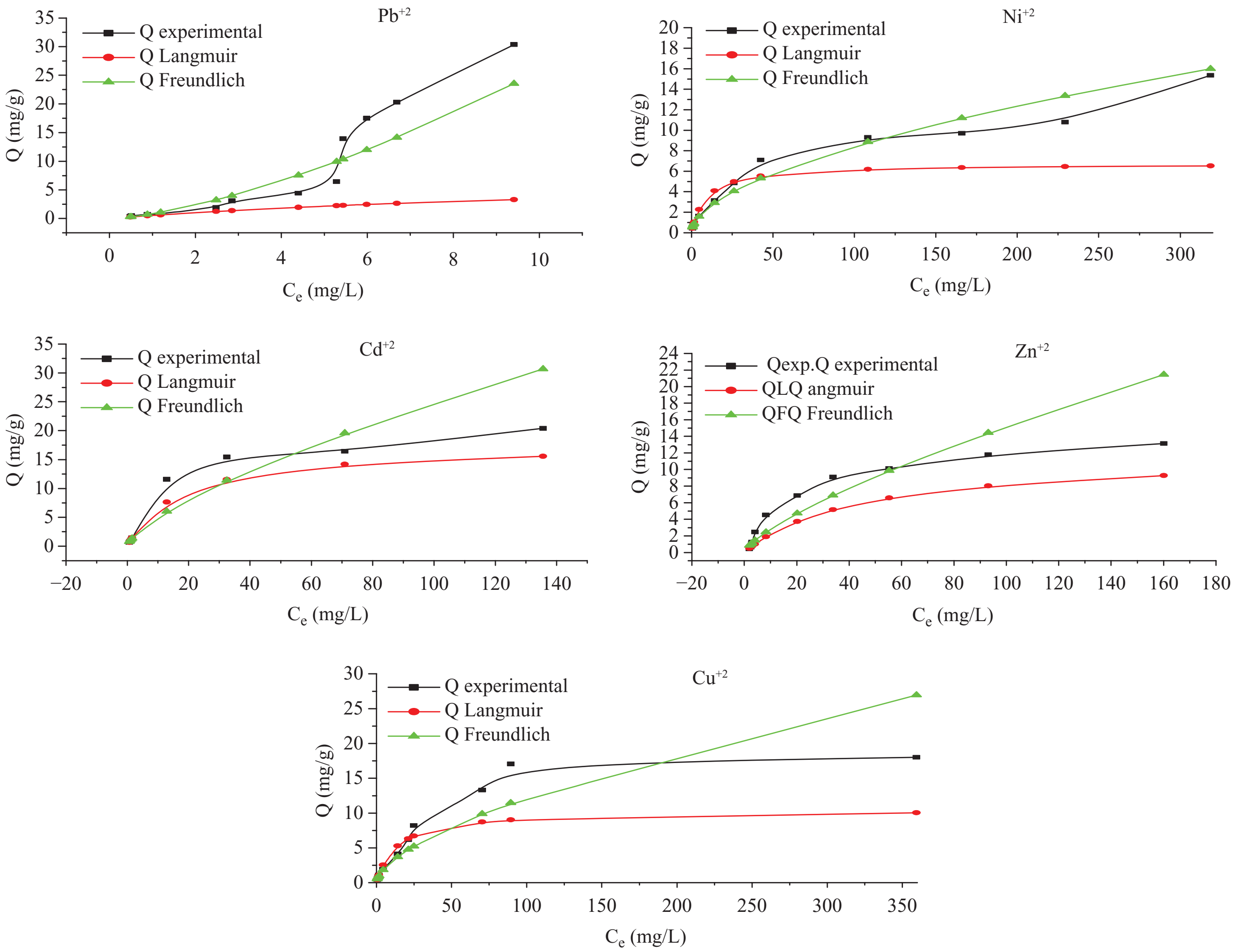

Fig. 2. Comparison of the Langmuir and Freundlich models with the experimental isotherm for Pb^{2+}, Ni^{2+}, Cd^{2+}, Zn^{2+} and Cu^{2+} on green coconut shell powder pre-treated with NaOH 0.1 mol L^{-1}/3 h, pH 5, $C_{\text{biomass}} = 40$ g L^{-1}, particle size 60–99 mesh, ambient temperature (28 $\pm$2°C).

$$Q_e = \frac{(C_0 - C_e)V}{m}, \quad (1)$$

where C_0: the initial concentration (mg L^{-1}); C_e: the equilibrium concentration of adsorbate (mg L^{-1}); V: the solution volume (L) and m: mass of the adsorbent (g).

The adsorption equilibrium isotherms (relationship between adsorption capacity and metal ion concentration at equilibrium) were obtained using the linearized Langmuir (2) and Freundlich (3) equations [28–30].

$$\text{Langmuir Equation}: \quad \frac{1}{Q_e} = \frac{1}{Q_{\max}} + \left(\frac{1}{Q_{\max}K}\right)\left(\frac{1}{C_e}\right), \quad (2)$$

$$\text{Freundlich Equation}: \quad \text{Log}Q_e = \text{Log}P + 1/n\text{Log}C_e, \quad (3)$$

where Q_e (mg g^{-1}) the amount of metal ion adsorbed, expressed in mg metal ion/g adsorbent; C_e (mg mL^{-1}) the equilibrium concentration of the metal ion in solution; $Q_{\max}$ (mg g^{-1}) and K (L mg^{-1}): Langmuir constants; P (mg g^{-1}) and $1/n$ Freundlich constants related to the adsorption capacity and to the surface heterogeneity, respectively.

3. Results and discussion

3.1. Characterization of the adsorbent

Scanning electron microscopy (MEV) of the adsorbent showed porous material having an irregular surface with tabular hole distribution [12]. Infrared spectroscopy provided evidence of weak and strong bands between 3,448 and 607 cm^{-1} indicating the presence of carbonyls, aromatics, and alcohols (Fig. 1).

Table 1
Langmuir and Freundlich parameters with the correlation coefficients (R^2) single solution

	Langmuir			Freundlich		
Ion	K (L mg^{-1})	Q_{max} (mg. g^{-1})	R^2	K (L. mg^{-1})	*1/n*	R^2
Pb^{2+}	0.086	8.32	0.986	0.814	1.492	0.925
Ni^{2+}	0.110	6.71	0.986	0.685	0.547	0.986
Cd^{2+}	0.085	17.51	0.979	1.315	0.669	0.880
Zn^{+2}	0.023	11.78	0.865	0.526	0.731	0.883
Cu^{2+}	0.072	10.45	0.994	0.767	0.614	0.958

Table 2
Values of the separation factor (R_L) and ionic radius for the ions studied

Ion	R_L	Ionic radius (Å)
Pb^{2+}	0.056–0.001	1.32
Ni^{2+}	0.339–0.010	0.69
Cd^{2+}	0.865–0.154	0.95
Zn^{2+}	0.697–0.060	0.74
Cu^{2+}	0.441–0.013	0.73

The bands at 3,448 and 3,415 cm^{-1} are characteristic of axial stretching of alcohols or phenols; the bands at 2,922 and 2,924 cm^{-1} show hydrocarbon nature; the band at 1,734 cm^{-1} is for carbonyl groups; the bands at 1,681 and 1,653 cm^{-1} indicate of the presence carboxyl functions; the bands at 1,560 and 1,520 cm^{-1} are for amines and amides; the bands at 1,423 and 1,377 cm^{-1} indicate axial deformations of CN groups in amines and amides; the bands at 1,064 and 1,039 cm^{-1} are for C–N and C–O bonds. The treatment with NaOH shows no chemical modification of the green coconut, but by other hand there was an increase of the intensities of several bands after the chemical treatment. The assignments are in agreement with the findings by Pino [31], who studied the adsorption of cadmium on green coconut shell powder without any pre-treatment.

3.2. Adsorption isotherms for single metal ions

The adsorption equilibrium were investigated using Langmuir and Freundlich models [28,29]. The adsorption isotherms of Pb^{2+}, Ni^{2+}, Cd^{2+}, Zn^{2+} and Cu^{2+} on green coconut shell powder pre-treated with NaOH 0.1 mol L^{-1} are shown in Fig. 2. The isotherms according to the Giles classification are of the type L [30].

The Langmuir and Freundlich isotherms for all metal ions are shown in Fig. 2. The parameters determined for each model are given in Table 1. The experimental data clearly show that the Langmuir-type isotherms describe well the adsorption mechanism for Pb^{2+}, Cd^{2+} and Cu^{2+}, while Zn^{2+} is of the Freundlich-type. On the other hand, Ni^{2+} agrees well with both models. The adsorption capacity of the metals studied followed the order: $Cd^{2+} > Zn^{2+} > Cu^{2+} > Pb^{2+} > Ni^{2+}$, which can be correlated to the ionic radius of Pauling, except for Pb^{2+} and Ni^{2+} (Table 2) [32–35].

Table 3
Langmuir and Freundlich parameters with the correlation coefficients (R^2) in a multimetal solution

	Langmuir			Freundlich		
Ion	K (L mg^{-1})	Q_{max} (mg g^{-1})	R^2	K (L. mg^{-1})	*1/n*	R^2
Pb^{2+}	0.112	7.89	0.972	0.812	0.497	0.886
Ni^{2+}	0.451	1.72	0.967	0.580	0.245	0.900
Cd^{2+}	0.962	3.24	0.991	1.04	0.315	0.759
Zn^{2+}	0.045	13.3	0.896	0.877	0.341	0.903
Cu^{2+}	0.227	5.09	0.969	0.834	0.581	0.977

The separation factor (R_L) is used to predict the affinity between the adsorbent and the adsorbate and is given by equation:

$$R_L = 1/(1 + KC_0), \qquad (4)$$

where: K: Langmuir constant; C_0: initial concentration of the solute in solution.

In accordance to Ngah [36], values of R_L between 0 and 1 show favorable adsorption, which is the case for all ions studied (Table 3).

3.3. Adsorption isotherms for multimetal ions

The adsorption isotherms for ions an aqueous solution for five-component (Cd^{2+}-Cu^{2+}-Ni^{2+}-Pb^{2+}-Zn^{2+}) on pre-treated green coconut shell powder are shown in Fig. 3. The isotherms according to the Giles classification are of the type L [30].

The Langmuir and Freundlich isotherms for all metal ions are shown in Fig. 3. The parameters determined for each model are given in Table 3. The experimental data

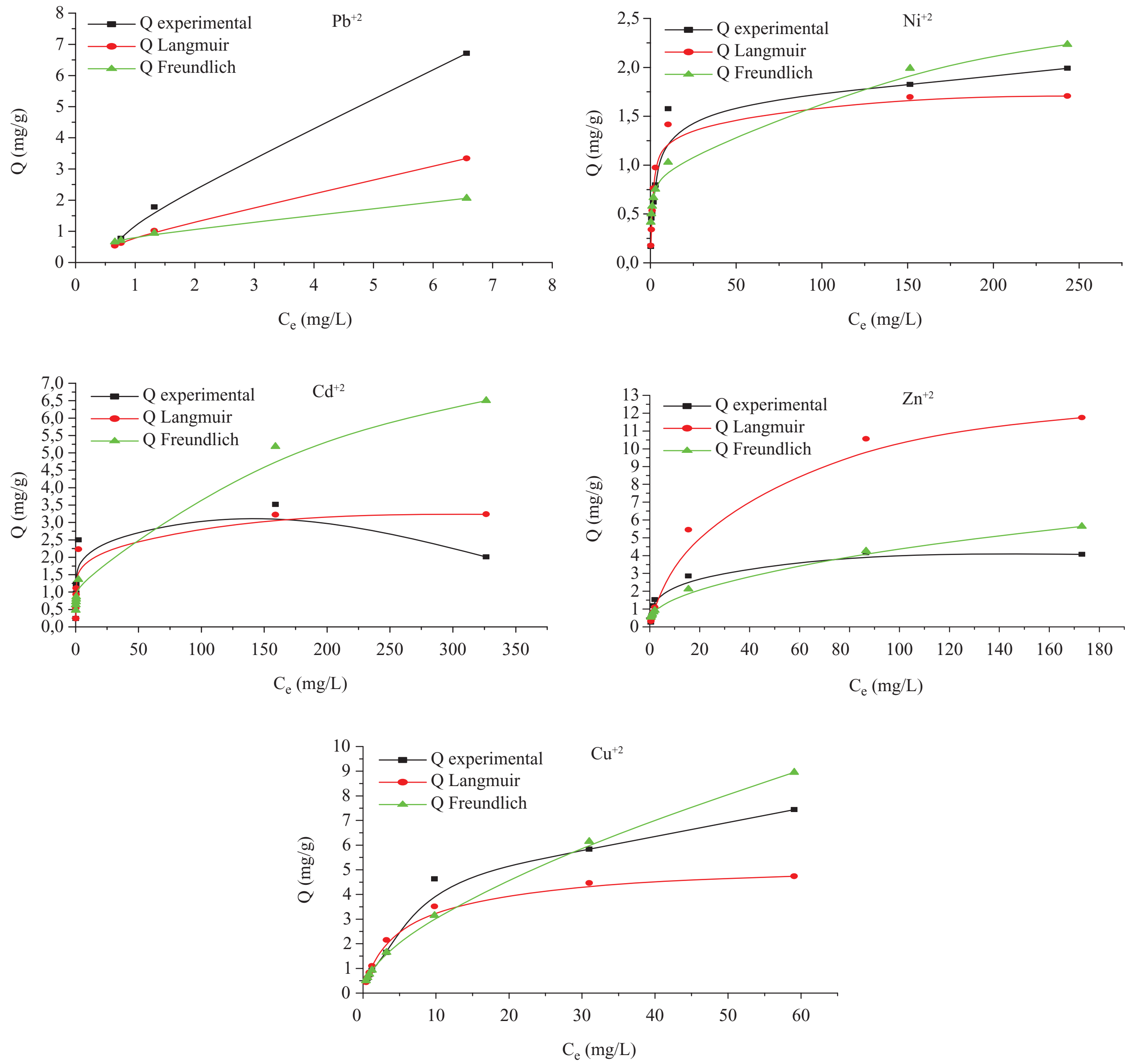

Fig. 3. Comparison of the Langmuir and Freundlich models with the experimental isotherm for Pb^{2+}, Ni^{2+}, Cd^{2+}, Zn^{2+} and Cu^{2+} in a multimetal solution on green coconut shell powder pre-treated with NaOH 0.1 mol L^{-1}/3 h, pH 5, $C_{biomass} = 40$ g L^{-1}, particle size 60–99 mesh, ambient temperature (28 ± 2°C).

clearly show that the Langmuir-type isotherms describe well the adsorption mechanism for Pb^{2+}, Ni^{2+} and Cd^{2+}, while Zn^{2+} and Cu^{2+} are of the Freundlich-type as can be observed by the values of correlation coefficients (R^2). The adsorption capacity of the metals studied followed the order: $Zn^{2+} > Pb^{2+} > Cu^{2+} > Cd^{2+} > Ni^{2+}$. This can be attributed to the specificity of active sites, to varying affinities for adsorption or to competitive effects. Sekhar [34] referred that multicomponent systems, the complex interactions of several factors such as ionic charge and

Table 4
Values of the separation factor (R_L) for the ions studied in a multimetal solution

Ion	Range of R_L
Pb^{2+}	0.560–0.008
Ni^{2+}	0.241–0.003
Cd^{2+}	0.098–0.001
Zn^{2+}	0.677–0.025
Cu^{2+}	0.354–0.004

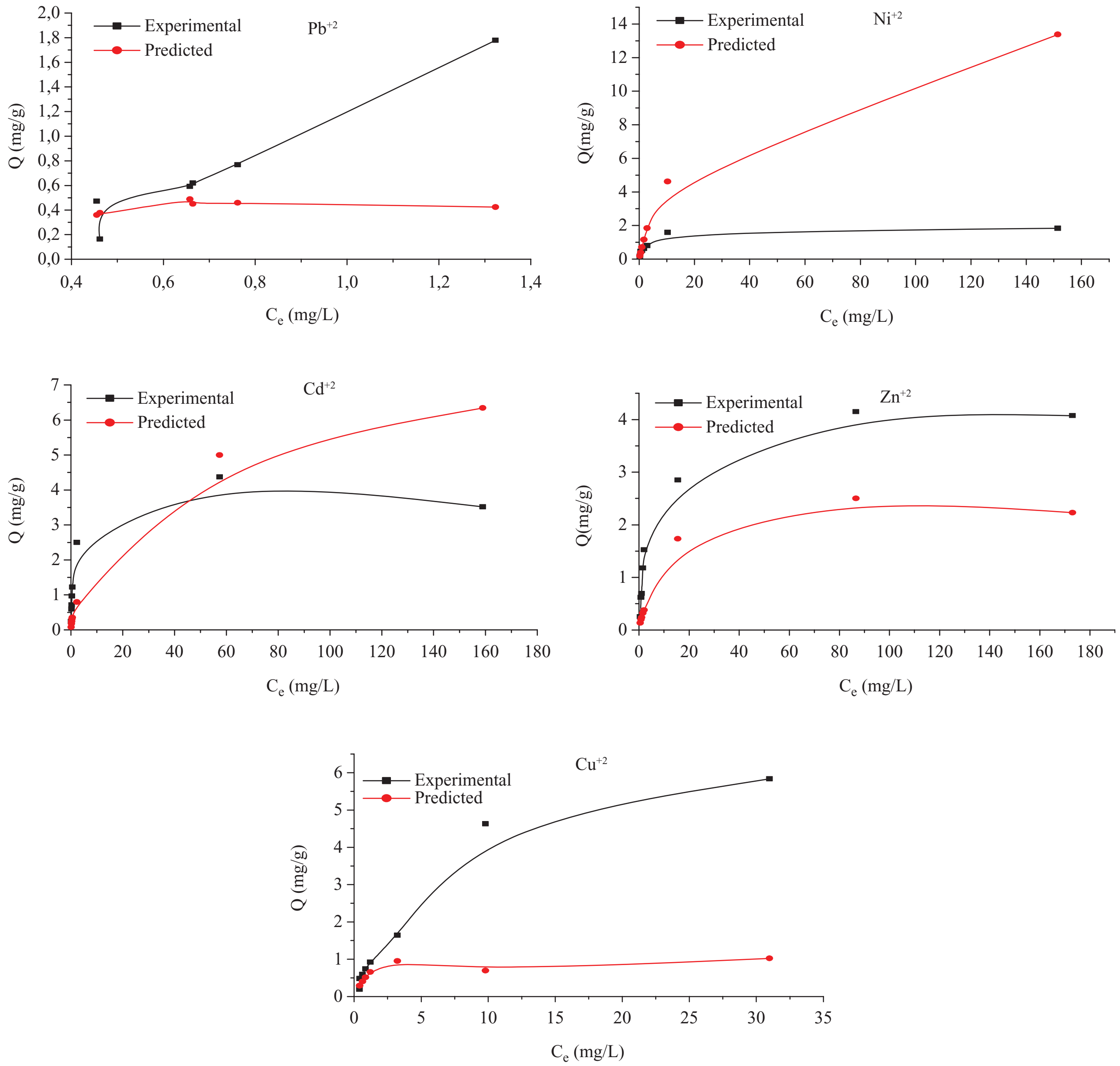

Fig. 4. Comparison between the experimental and predicted adsorption models for Pb^{2+}, Ni^{2+}, Cd^{2+}, Zn^{2+} and Cu^{2+} on green coconut shell powder pre-treated with NaOH 0.1 mol L^{-1}/3 h, in a multimetal solution. Conditions: pH 5, $C_{biomass} = 40$ g L^{-1}, particle size 60–99 mesh, ambient temperature (28 ± 2°C).

ionic radii will account for the differences in the metal removal capacity of the adsorbent. As a result, ordering of the metal ions based on a single factor is very difficult.

The values of R_L are all between 0 and 1 indicating that adsorption on green coconut shell powder is efficient also in the multimetal system (Table 4).

Competitive adsorption effects were investigated using the extended Langmuir equation (Eq. (5)) [27,29,37].

$$q_i = \frac{Q_i K_i C_i}{1 + \sum_{j=1}^{n} K_j C_j}, \qquad (5)$$

where Q_i and K_i are the maximum adsorption capacity (mg. g^{-1}) and Langmuir constant for each component i in its single system.

The results (Fig. 4) prove competition for binding sites from comparison of the experimental and

Table 5
Comparison of the adsorption capacities Pb^{2+}, Ni^{2+}, Cd^{2+}, Zn^{2+} and Cu^{2+} ions on various natural adsorbents and on charcoal

	Q_{max} (mg. g^{-1})								
Adsorbent	Pb^{2+}	Ni^{2+}	Cd^{2+}	Zn^{2+}	Cu^{2+}	pH	T (°C)	$C_{Biomass}$ (g L^{-1})	Reference
Green coconut shell powder pre-treated (single metal ion)	8.32	6.71	17.51	11.78	10.45	5.0	30	40	This work
Green coconut shell powder pre-treated (multimetal ions)	7.89	1.72	3.24	13.26	5.09	5.0	30	40	This work
Green coconut shell powder	–	–	285.7	–	–	7	27	5	[31]
Carrot residue	–	–	–	29.61	32.74	4.5	25	10	[18]
Marine macroalgae	–	181.5	–	–	–	5	30	70	[39]
Sugar cane bagasse	–	0.001	–	–	–	8	30	1	[10]
Rice husk	–	–	8.58	–	–	6.6-6.8	28-30	10	[40]
Papaya wood	–	–	17.35	14.44	19.9	5	30	5	[14]
Carbon prepared from coconut shell powder	26.51	–	–	–	–	4.5	45	1	[41]
Charcoal	–	31.08	–	–	–	8	30	1	[10]
Bagasse fly ash	–	0.03	–	–	–	8	30	1	[10]
Carbon prepared from sugar cane bagasse (single metal ions)	–	–	38.03	31.11	–	4.5	25	6	[33]
Carbon prepared from sugar cane bagasse (multimetal ions)	–	–	29.77	19.02	–	4.5	25	6	[33]

predicted data. Thus, adsorption is not specific and limited to a maximum binding capacity.

The effects of ionic interactions on adsorption are expressed by the ratio of the adsorption capacity for a specific metal ion in the presence of other metal ions [38].

For $Q^{Mix}/Q > 1$: adsorption is promoted by the presence of other metal ions;
For $Q^{Mix}/Q = 1$: net interaction is absent;
For $Q^{Mix}/Q < 1$: adsorption is suppressed by the presence of other metal ions.

The results reveal that adsorption of Ni^{2+}, Cd^{2+} and Cu^{2+} ions is suppressed in the multimetal solution ($Q^{Mix}/Q < 1$), whereas adsorption of Pb^{2+} and Zn^{2+} ions is promoted by presence of the other metal ions ($Q^{Mix}/Q > 1$).

Using the results of the Langmuir-model presented in Table 5, the adsorption capacities of several natural adsorbents and charcoal were compared [10,14,18,31,33,39–41]. However, because the varying experimental conditions do not permit to extract reliable conclusions.

4. Conclusion

The green coconut shell powder pre-treated with NaOH 0.1 mol L^{-1} during 3 h is a residue from natural and renewable sources and it may represent a viable low-cost alternative to remove toxic metal ions, for example, from industrial contaminated wastewater. The Langmuir-model describes well the adsorption mechanism for Pb^{2+}, Cd^{2+} and Cu^{2+} ions, while the Freundlich-model is more effective for Zn^{2+} ions. Adsorption of Ni^{2+} ions agrees well with both adsorption models. In a multimetal solution, competitive effects were noted for all ions. The Langmuir-model is apt to explain the adsorption mechanism for Pb^{2+}, Ni^{2+} and Cd^{2+} ions, while adsorption of Zn^{2+} and Cu^{2+} ions follow the Freundlich-model.

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