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COMPETITIVE ADSORPTION OF Cd(II) AND Zn(II) FROM AQUEOUS SOLUTION ONTO ACTIVATED CARBON

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ABSTRACT

Single and simultaneous Cd(II) and Zn(II) adsorption isotherms from aqueous solution onto activated carbon were determined experimentally. Single isotherms for these ions fit the Langmuir isotherm well, and the maximum molar uptake of Zn(II) averaged 1.6 times that of Cd(II). The simultaneous adsorption isotherms also fit the bisolute Langmuir isotherm modified with an interaction factor. Adsorption of each single ion was reduced by the presence of the other ion, which competed for some of the same active sites. The Zn(II) isotherm proved to be more sensitive to the presence of the competing ion than did the Cd(II) isotherm.

Key Words: Activated carbon; Cadmium; Competitive adsorption; Single adsorption; Zinc

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INTRODUCTION

Adsorption of a metal ion from an aqueous solution onto activated carbon depends on several factors, such as solution pH, metal ion species or hydrocomplexes formed in solution, temperature, physicochemical characteristics of the adsorbent surface, and presence of other metallic ions or compounds that can adsorb on the surface. The influence of competing ions and other solution compounds on adsorption efficiency has been largely ignored, and most reports of metal adsorption from aqueous solution examine only the adsorption isotherm for a single metal. Furthermore, the few studies reporting on metal adsorption isotherms in the presence of other components in solution have provided scantly results with very limited applications.

The adsorption of a component onto activated carbon is affected by the presence of other solution components that may compete for the same adsorption sites. Adsorption of a single component onto a porous solid is generally represented by the adsorption isotherm, which is a mathematical relationship expressing the equilibrium that exists between the moles of the component adsorbed onto the solid surface and the concentration of that component remaining in solution. The most commonly applied isotherm models for solid-liquid systems are the Langmuir and Freundlich isotherms. The adsorption isotherm for a single component allows evaluation of the capacity of the adsorbent for a particular component solely present in the solution. In other words, the adsorption isotherm describes a relationship between 2 variables and is illustrated by a two-dimensional graph.

When 2 or more components are being adsorbed simultaneously from an aqueous solution, the adsorption isotherm is much more complex than when a single component is adsorbed. The adsorption of 2 components may be represented by a three-dimensional isotherm in which the moles adsorbed are plotted against the concentrations of the 2 components. The moles adsorbed may be composed of either or both of the solution species. The adsorption surface is thus obtained. This method of plotting the adsorption isotherm has been used by Volesky et al. (1,2,3) to represent adsorption isotherms for 2 metallic ions onto biosorbents or biomass.

Because of their chemical similarity, cadmium and zinc ions are generally found together in industrial wastewaters. To manage ion removal systems, researchers must evaluate the effect of competitive adsorption between these 2 ions. Few studies of competitive adsorption between Cd(II) and Zn(II) from aqueous solution onto activated carbon have been reported in the literature. Gabaldon et al. (4) measured the percentage of removal for Cd(II) and Zn(II) when both ions were adsorbed simultaneously. They reported a lower percentage of removal for simultaneous adsorption than when each ion was adsorbed independently, and they concluded that the percentage removal of each ion was reduced by the presence of the other. Those authors presented their data as percentage removal for each ion as a function of the initial concentration of the other ion and not as three- dimensional adsorption isotherms.

A number of researchers have studied the multicomponent adsorption of heavy metals from aqueous solution onto activated carbons (4,5), goethite (6), lignite (7), peat (8), and pine bark (9). Several multicomponent adsorption isotherm models have been used to describe the experimental data. The adsorption of Cu(II)-Ni(II), Cu(II)-Cd(II), Cu(II)-Zn(II), and Cd(II)-Zn(II) onto activated carbon were modeled with the surface-complex-formation triple layer model (4,5). The bisolute adsorption of the systems Cu(II)-Cd(II), Cu(II)-Ni(II), and Cd(II)-Ni(II) on pine bark were fitted to the bisolute Langmuir model, extended Freundlich model, Sips model, and the ideal adsorption solution theory model with the single-solute adsorption isotherm constants (9). The competitive adsorption of Cu(II) and Ni(II) on peat was studied by Ho and McKay (8) who reported that their data were described reasonably well with a bisolute Langmuir isotherm modified with an interaction factor.

Our aim was to determine simultaneous isotherms for Cd(II) and Zn(II) adsorbed from an aqueous solution onto activated carbon and to represent these isotherms as adsorption surfaces.

EXPERIMENTAL METHODS

All Cd(II) and Zn(II) solutions were prepared from $Cd(NO_3)4H_2O$ and $Zn(NO_3)6H_2O$, and the solution pH was adjusted with 0.01 mol/L HNO_3 and NaOH solutions. The activated carbon used in this study is commercially known as C carbon and is produced from sawdust by Carbones Mexicanos S. A. C carbon has a surface area of 781 m^2/g, pore volume of 0.397 cm^3/g, average pore diameter of 2.07 nm, particle density of 1.51 g/cm^3, and mean particle diameter of 0.416 mm. The carbon as received from supplier contained dust and was treated to remove it. The carbon was washed with deionized water as many times as needed to remove the dust and was dried in an oven at 110°C for 24 hours. Soon afterward the carbon was stored in plastic containers.

Adsorption isotherm experiments were carried out in a batch adsorber that consisted of an 500 or 1000 mL Erlenmeyer flask. The metal ion solution and granular activated carbon were maintained in contact within the adsorber. The carbon was placed in a nylon mesh basket to avoid formation of fine particles due to attrition between the carbon and adsorber walls. Such fine particles could interfere in the determination of metal ion concentration in aqueous solution. The adsorber was maintained partially submerged in a constant-temperature water bath and the solution within the flask was stirred continuously with a Teflon-coated stirring bar over a magnetic stirrer located under the water bath.

Aqueous concentrations of Cd(II) and Zn(II) were determined using atomic absorption spectrophotometric methods (10). Absorbance of a sample was measured with a double beam Varian Spectra AA-20 atomic spectrophotometer and metal ion concentration was determined by comparing absorbance with a previously prepared, standard calibration curve.

We performed adsorption experiments by adding a known mass of carbon to a specific volume of an aqueous solution of previously determined metal ion concentration. Carbon doses of 1, 2, and 4 g/L were used in the experiments, and the initial concentrations of Cd(II) and Zn(II) varied from 0.04 to 1.5 mmol/L. Metal ion solution and carbon remained in contact until equilibrium was reached. Samples were taken at various times to follow the progress of adsorption, and metal ion concentration in solution was determined as described above for each sample. When 2 consecutive samples showed no change in metal ion concentration, the system was considered to be at equilibrium. Preliminary runs showed that the equilibrium was reached between 5 and 7 days. The molar uptake of the metal ion adsorbed was calculated by performing a mass balance for the metal ion. All adsorption experiments were carried out at pH 7 and 25°C.

RESULTS AND DISCUSSION

Single Adsorption Isotherms for Cd(II) and Zn(II)

We chose C carbon to use in this study because in a previous study (11) both Zn(II) and Cd(II) were reported to adsorb more on C carbon than on other commercial carbons. Single adsorption isotherms for Zn(II) and Cd(II) onto C carbon at 25°C and pH 7 are shown in Fig. 1. Experimental data were fitted to the Langmuir isotherm represented by the following equation:

$$q_{\mathrm{i}} = \frac{q_{\mathrm{m,i}} K_{\mathrm{i}} C_{\mathrm{i}}}{1 + K_{\mathrm{i}} C_{\mathrm{i}}} \tag{1}$$

The constants for this isotherm were obtained by a method of least-squares based on the optimization algorithm of Rosenbrock-Newton. Table 1 presents the values of the isotherm constants and the average percent deviation, which is defined as follows:

$$\%D = \left(\frac{1}{N} \sum_{i=1}^{N} \left| \frac{q_{\mathrm{exp}} - q_{\mathrm{cal}}}{q_{\mathrm{exp}}} \right| \right) \times 100\% \tag{2}$$

As shown by the average percent deviation values reported in Table 1, a reasonable fit to the experimental data was obtained with the application of the Langmuir isotherm. The Freundlich isotherm was also tested, but a better fit was obtained with the Langmuir isotherm. Several authors (12,13) found that the experimental

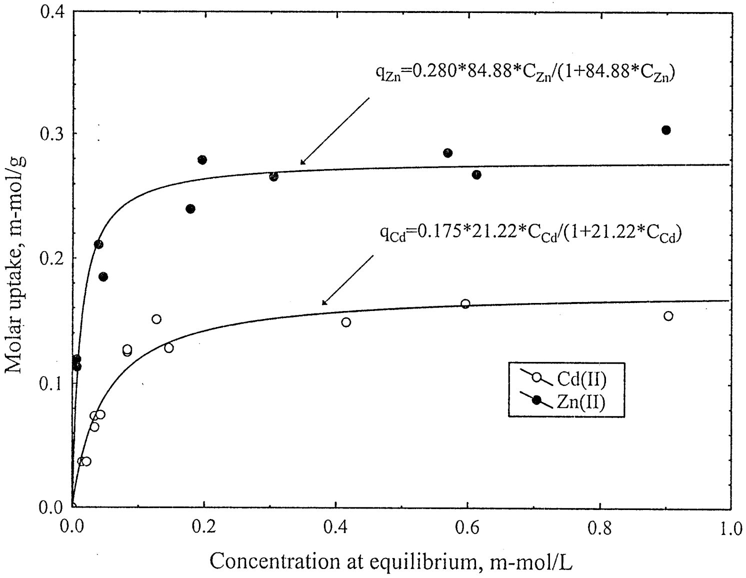

Figure 1. Single adsorption isotherms for Zn(II) and Cd(II) onto C carbon at pH 7 and 25°C.

single-solute adsorption isotherm data for Zn(II) and Cd(II) from aqueous solution onto different types of activated carbon can be represented by the Langmuir isotherm.

Comparison of the individual adsorption isotherms shown in Fig. 1 illustrates that the maximum molar uptake of Zn(II) adsorbed is close to 1.6 times that of Cd(II). In other words, the order of the single-component adsorption capacity is Zn(II) >Cd(II). Gabaldon et al. (4) and Seco et al. (5) reported similar results for the single-component adsorption of Cd(II) and Zn(II) onto some activated carbons.

Zn(II) can be adsorbed more selectively on C carbon than can Cd(II). This result cannot be attributed to accessibility of the internal surface area due to dif-

Table 1. Langmuir Isotherm Constants for Single Cd(II) and Zn(II) Adsorption onto C Carbon at 25°C and pH 7

Ion	$q_{m,i}$ (mmol/g)	K_i (L/mmol)	%*D*
Cd(II)	0.175	21.22	11.21
Zn(II)	0.280	84.88	7.94

ferences in ion sizes because the hydrated radius of Zn(II) (0.430 nm) is very similar to that of Cd(II) (0.426 nm) (14). This difference in adsorption capacity cannot be explained so simply and it is probably related to the electrostatic attraction between the very heterogeneous surface of the activated carbon and the metal ions in solution. In previous studies (15,16), researchers reported that at pH 7 the single adsorption of Cd(II) and Zn(II) is facilitated by the electrostatic attractions because these metals exist as Cd^{2+} and Zn^{2+} in water solution and the surface of C carbon is negatively charged.

Another possible explanation for the relatively high Zn(II) selectivity is related to the ability of Cd(II) and Zn(II) to be adsorbed on one kind of surface sites while Zn(II) can also be adsorbed on other kind of surface sites. This means that the single adsorption isotherm of Zn(II) should be represented by a dual-site Langmuir isotherm, known as the bi-Langmuir isotherm (17). However, a Scatchard plot (q/C vs. q) of the single solute adsorption data of Zn(II) did not suggest that Zn(II) was adsorbed on 2 kinds of sites (17). Thus, the single-site Langmuir isotherm is appropriate to represent the single adsorption of both metal ions.

Simultaneous Cd(II) and Zn(II) Adsorption

As cadmium and zinc ions usually occur together in industrial wastewaters (18), the effect of each ion on the adsorption properties of the other is important information for managing efficient removal techniques. These 2 ions can compete for the same active sites when adsorbed onto C carbon, so that when both ions are present in solution, adsorption of each will be altered depending on the concentration of the other.

The single adsorption isotherm of a specific solute in an aqueous solution is mainly a function of solution pH, temperature, and the characteristics of the adsorbing material. However, in multicomponent systems, the adsorption isotherm of a certain solute is also dependent on the concentration and characteristics of the other solutes in the aqueous solution. The solute of interest may be in competition with other solutes for the same active adsorption sites.

The experimental data for simultaneous Cd(II) and Zn(II) adsorption were interpreted with the bisolute Langmuir isotherm because the Langmuir isotherm best fit the single adsorption data for both Zn(II) and Cd(II) onto C carbon at pH 7 and 25°C. The competitive Langmuir isotherms for Cd(II) and Zn(II) are represented as follows (19):

$$q_{\mathrm{Cd}} = \frac{q_{\mathrm{m,Cd}}\, K_{\mathrm{Cd}}\, C_{\mathrm{Cd}}}{1 + K_{\mathrm{Cd}}\, C_{\mathrm{Cd}} + K_{\mathrm{Zn}}\, C_{\mathrm{Zn}}} \tag{3}$$

$$q_{\mathrm{Zn}} = \frac{q_{\mathrm{m,Zn}}\, K_{\mathrm{Zn}}\, C_{\mathrm{Zn}}}{1 + K_{\mathrm{Cd}}\, C_{\mathrm{Cd}} + K_{\mathrm{Zn}}\, C_{\mathrm{Zn}}} \tag{4}$$

The constants of these 2 isotherms are from the single-solute Langmuir isotherms and are reported in Table 1. This isotherm model successfully fitted the competitive adsorption of Cu(II)-Ni(II) onto pine bark (9) and of Zn(II)-Cd(II), Cu(II)-Cd(II), and Cu(II)-Zn(II) onto a biosorbent prepared from seaweed (1).

The experimental molar uptake values of Cd(II) and Zn(II) were compared to the molar uptake values of Cd(II) and Zn(II) predicted with the bisolute Langmuir isotherm, as plotted in Fig. 2. As shown in this figure, the bisolute Langmuir isotherm overestimated the molar uptake of Zn(II) and the average percent deviation was 94.0%. However, it underestimated the molar uptake of Cd(II) and the average percent deviation was 33.36%. Thus, the binary adsorption data cannot be properly described with the bisolute Langmuir isotherm.

As documented in the literature (9), the bisolute Langmuir model provides a reasonable fit to the multicomponent adsorption data as long as the $q_{\mathrm{m,i}}$ values for each metal evaluated from single-solute Langmuir isotherm are similar to each other. As previously noted, $q_{\mathrm{m,Zn}}$ is approximately 1.6 times greater than $q_{\mathrm{m,Cd}}$.

Jain and Snoeyink (20) assumed that some adsorption occurs without competition because not all sites are available to all solutes. They modified the bisolute Langmuir isotherm for systems in which the $q_{\mathrm{m,i}}$ values of components are different, proposing the following isotherm for the solute with the higher $q_{\mathrm{m,i}}$,

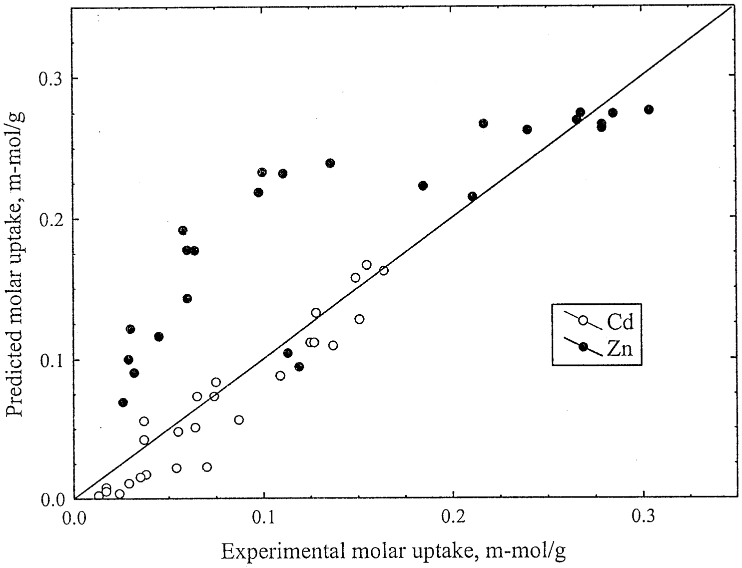

Figure 2. Comparison between the experimental molar uptake and the molar uptake predicted with the bisolute Langmuir isotherm.

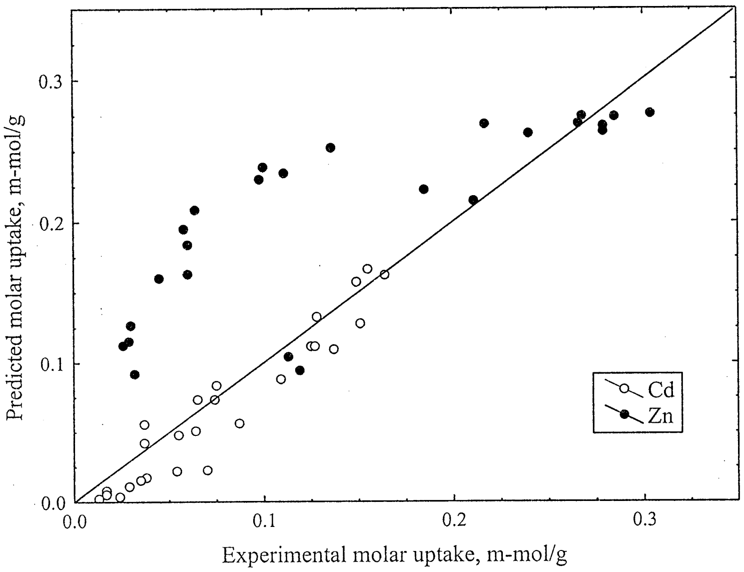

Figure 3. Comparison between the experimental molar uptake and the molar uptake predicted with the modified bisolute Langmuir isotherm.

which in our study is Zn(II):

$$q_{\mathrm{Zn}} = \frac{(q_{\mathrm{m,Zn}} - q_{\mathrm{m,Cd}})\, K_{\mathrm{Zn}} C_{\mathrm{Zn}}}{1 + K_{\mathrm{Zn}} C_{\mathrm{Zn}}} + \frac{q_{\mathrm{m,Cd}} K_{\mathrm{Zn}} C_{\mathrm{Zn}}}{1 + K_{\mathrm{Cd}} C_{\mathrm{Cd}} + K_{\mathrm{Zn}} C_{\mathrm{Zn}}} \tag{5}$$

The isotherm for the solute with the lower $q_{\mathrm{m,i}}$ is the same as that represented in Eq. (3). The constants are from the single-solute Langmuir isotherms and are noted in Table 1. In Eq. (5), the difference between the maximum molar uptakes is the number of sites with noncompetitive adsorption.

The modified bisolute Langmuir isotherm was used by Christophi and Axe (6) to describe the binary adsorption of the systems Cd(II)-Cu(II), Cd(II)-Pb(II), and Cu(II)-Pb(II) onto goethite. This modified bisolute Langmuir isotherm provided a better fit to the experimental data of the 4 binary systems than did the bisolute Langmuir isotherm.

The modified bisolute Langmuir model was applied to the experimental data for simultaneous Cd(II) and Zn(II) adsorption and as shown in Fig. 3, overpredicted the molar uptake of Zn(II), with an average deviation of 112.7%. The modified bisolute Langmuir isotherm failed to predict the molar uptake of Zn(II), and its prediction had a higher average percent deviation than that obtained with the bisolute Langmuir isotherm.

Ho and McKay (8) modified the bisolute Langmuir isotherm with an interaction factor, η, and obtained an excellent fit of the adsorption data of Cu(II) and

Ni(II) onto peat. The model proposed by these authors can be represented as follows:

$$q_{\mathrm{Cd}} = \frac{q_{\mathrm{m,Cd}}\, K_{\mathrm{Cd}} \left(\dfrac{C_{\mathrm{Cd}}}{\eta_{\mathrm{Cd,Cd}}} \right)}{1 + K_{\mathrm{Cd}} \left(\dfrac{C_{\mathrm{Cd}}}{\eta_{\mathrm{Cd,Cd}}} \right) + K_{\mathrm{Zn}} \left(\dfrac{C_{\mathrm{Zn}}}{\eta_{\mathrm{Zn,Cd}}} \right)} \tag{6}$$

$$q_{\mathrm{Zn}} = \frac{q_{\mathrm{m,Zn}}\, K_{\mathrm{Zn}} \left(\dfrac{C_{\mathrm{Zn}}}{\eta_{\mathrm{Zn,Zn}}} \right)}{1 + K_{\mathrm{Cd}} \left(\dfrac{C_{\mathrm{Cd}}}{\eta_{\mathrm{Cd,Zn}}} \right) + K_{\mathrm{Zn}} \left(\dfrac{C_{\mathrm{Zn}}}{\eta_{\mathrm{Zn,Zn}}} \right)} \tag{7}$$

where $\eta_{\mathrm{i,j}}$ is the interaction factor of metal i for the adsorption of metal j. This interaction factor is specific to each metal ion in a given system and depends upon the other metal ions present. This interaction-factor isotherm model was satisfactorily used to represent the multicomponent adsorption equilibrium data of dyes on activated carbon (21).

We obtained the best interaction factor by fitting the isotherm models to the experimental data with a method of least-squares that employs an optimization algorithm. The values of the interaction factors are as follows: $\eta_{\mathrm{Cd,Cd}} = 1.02$; $\eta_{\mathrm{Zn,Cd}} = 3.29$; $\eta_{\mathrm{Cd,Zn}} = 0.089$; and $\eta_{\mathrm{Zn,Zn}} = 1.26$. Figure 4 shows a comparison between

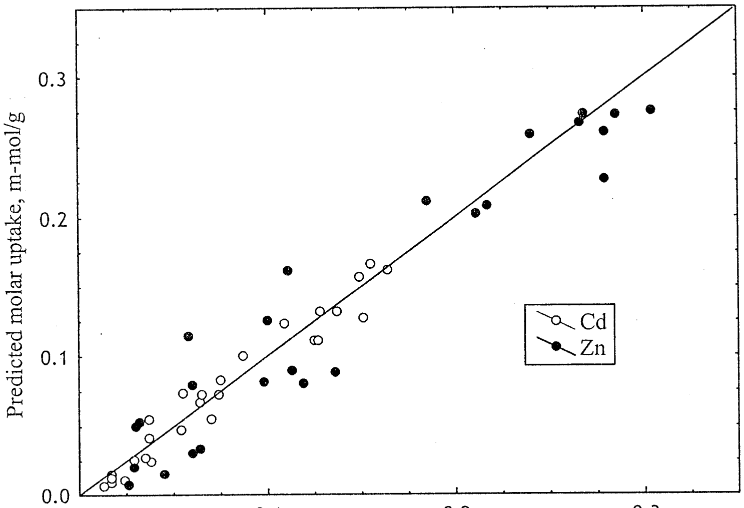

Figure 4. Comparison between the experimental molar uptake and the molar uptake predicted with the bisolute Langmuir isotherm modified with the interaction factor.

the experimental molar uptake and the molar uptake predicted with the bisolute Langmuir isotherm that has been modified with the interaction factor. As shown in Figure 4, this bisolute Langmuir isotherm interprets the experimental data for both ions reasonably well. The average percent deviation was 18.8% for Cd(II) and 31.0% for Zn(II), which are the lowest values for all the models tested in this work.

The experimental data for the competitive adsorption isotherms of Cd(II) and Zn(II) are displayed in Figs. 5 and 6, respectively. The bisolute Langmuir isotherms modified with the interaction factor, determined through the single-solute Langmuir constants (Table 1) and the best values of the interaction factors, are also plotted in these figures. At all Zn(II) concentrations, the Zn(II) adsorption isotherm decreased as the Cd(II) concentration increased. The same behavior was observed for the Cd(II) adsorption isotherm with respect to Zn(II) concentration.

An alternate method for representing the Langmuir isotherms for competitive Cd(II) and Zn(II) adsorption (Eqs. 6 and 7) is to graph the adsorption isotherm of one of the ions while the concentration of the other ion is kept constant. Adsorption isotherms for Zn(II) at different Cd(II) isoconcentrations are shown in Fig. 7. The uptake of Zn(II) was reduced significantly by the presence of Cd(II) with greater reduction at lower Zn(II) concentrations. For example, at a Zn(II) equilibrium concentration value of 0.1 mmol/L, the uptake of Zn(II) adsorbed without

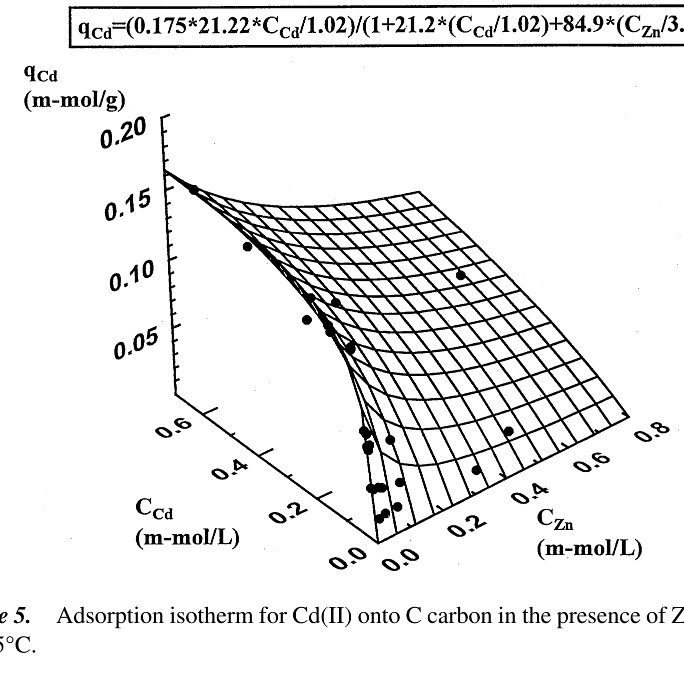

Figure 5. Adsorption isotherm for Cd(II) onto C carbon in the presence of Zn(II) at pH 7 and 25°C.

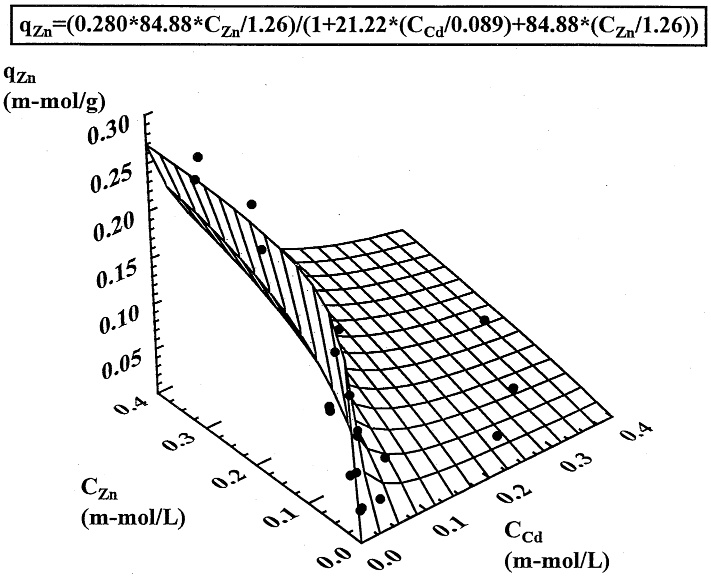

Figure 6. Adsorption isotherm for Zn(II) onto C carbon in the presence of Cd(II) at pH 7 and 25°C.

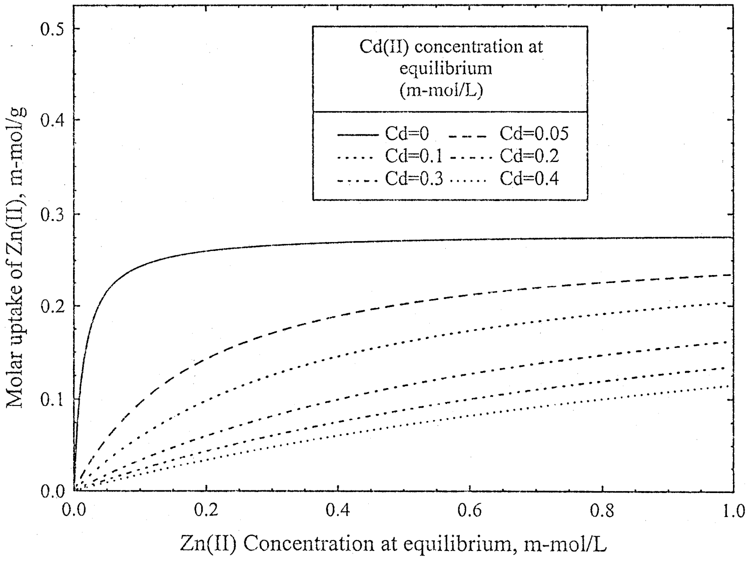

Figure 7. Effect of Cd(II) concentration on the Zn(II) adsorption isotherm onto C carbon at pH 7 and 25°C.

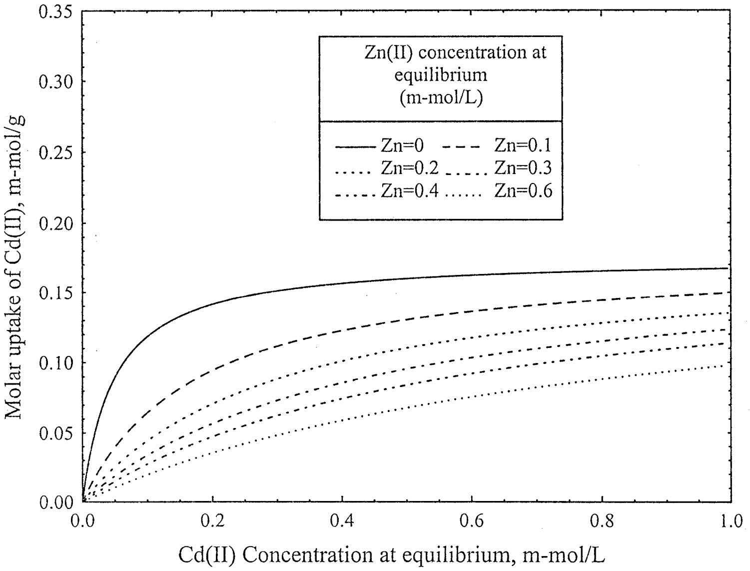

Figure 8. Effect of Zn(II) concentration on the Cd(II) adsorption isotherm on C carbon at pH 7 and 25°C.

Cd(II) was reduced by 60.7, 75.5, 86.1, 90.3 and 92.5% for Cd(II) concentration at equilibrium values of 0.05, 0.1, 0.2, 0.3 and 0.4 mmol/L, respectively.

Cd(II) adsorption isotherms at different Zn(II) isoconcentrations are shown in Fig. 8. The uptake of Cd(II) decreased as concentration of the competitive Zn(II) ion increased. For a Cd(II) concentration at equilibrium of 0.1 mmol/L, the uptake of Cd(II) adsorbed in the absence of Zn(II) decreased by 45.5, 62.5, 71.5 and 77.0% in respective Zn(II) concentration at equilibrium values of 0.1, 0.2, 0.3 and 0.4 mmol/L. In the presence of the competing ion, the uptake of Zn(II) decreased to a greater extent than the uptake of Cd(II). Thus, Zn(II) is more sensitive to the presence of the competing ion than is Cd(II).

CONCLUSIONS

Experimental data for single Cd(II) and Zn(II) adsorption isotherms on C carbon fit reasonably well to the Langmuir isotherm. The maximum molar uptake of Zn(II) was 1.6 times that of Cd(II).

Experimental data for simultaneous Cd(II) and Zn(II) adsorption onto C carbon correlated well to the bisolute Langmuir isotherm modified with an interaction factor. The simultaneous adsorption isotherms for Cd(II) and Zn(II) were always reduced compared to the single adsorption isotherms for these ions. The Zn(II) adsorption isotherm was affected more by the presence of the other ion than was that of Cd(II). The adsorption isotherm for a given ion is always reduced by the presence of the other because the 2 ions compete for some, but not all, of the same active sites.

NOMENCLATURE

C_{Cd}	concentration of Cd(II) in aqueous solution at equilibrium (mmol/L)
C_i	concentration of metal ion in aqueous solution at equilibrium (mmol/L)
C_{Zn}	concentration of Zn(II) in aqueous solution at equilibrium (mmol/L)
K_i	Langmuir isotherm constant for metal i, (L/mmol)
N	number of experimental data
q_{cal}	molar uptake of metal predicted with an isotherm model (mmol/g)
q_{Cd}	molar uptake of Cd(II) adsorbed (mmol/g)
q_{exp}	molar uptake of metal measured experimentally (mmol/g)
q_i	molar uptake of metal adsorbed (mmol/g)
$q_{m,i}$	molar uptake of metal adsorbed once a monolayer is formed (mmol/g)
q_{Zn}	molar uptake of Zn(II) adsorbed (mmol/g)
$\eta_{i,j}$	interaction factor of metal i for the adsorption of metal j

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