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SELECTIVE REMOVAL OF LEAD IONS BY POLYETHYLENE GLYCOL METHACRYLATE GEL BEADS CARRYING CIBACRON BLUE F3GA

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ABSTRACT

Polyethylene glycol methacrylate (PEG-MA) gel beads carrying Cibacron Blue F3GA (42.6 μmol/g polymer) were prepared for the removal of Pb(II), Cu(II), and Cd(II) from aqueous solutions that contained different amounts of these ions (10–100 mg/L) and at different pH values (2.0–7.0). Adsorption rates were high, and adsorption equilibria were reached within 20 minutes. Adsorption of these metal ions on the unmodified PEG-MA gel beads was zero. The maximum adsorptions of heavy metal ions onto the Cibacron Blue F3GA–attached microbeads from single solutions were 23.3 mg/g (112.4 μmol/g) for Pb(II), 12.4 mg/g (110.3 μmol/g) for Cd(II), and 7.0 mg/g (110.2 μmol/g) for Cu(II). When the heavy metal ions competed (in the case of the adsorption from their mixture) the amounts of adsorption were 14.96 mg/g (72.2 μmol/g) for

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Pb(II), 0.72 mg/g (11.3 μmol/g) for Cu(II), and 1.10 mg/g (9.8 μmol/g) for Cd(II). Under competitive conditions, the system showed high selectivity for Pb(II) ions. The formation constants of Cibacron Blue F3GA–metal ion complexes were investigated through the application of the Ružić method. The calculated values of formation constants were 8.5×10^5 L/mol[1] for Pb(II) dye, 2.5×10^5 L/mol for Cu(II) dye, and 8.2×10^4 L/mol for the Cd(II) dye complex. PEG-MA gel beads carrying Cibacron Blue F3GA can be regenerated through washing with a solution of nitric acid (0.1 mol/L). The maximum regeneration value was as high as 98.5%. These PEG-MA gel beads are suitable for more than 3 adsorption-desorption cycles without experiencing considerable loss of adsorption capacity.

Key Words: Polyethylene glycol methacrylate; Gel beads; Cibacron Blue F3GA; Pb(II) removal; Formation constants

INTRODUCTION

Heavy metals, such as lead, mercury, copper, and cadmium, are highly toxic when absorbed into the body. The symptoms of heavy metal toxic effects may vary widely at the physiological level, but the basic toxicity mechanisms at the molecular level may be limited. The toxicities of heavy metals may be caused by mechanisms that include blocking essential functional groups of biomolecules such as proteins and enzymes, displacing essential metal ions from biomolecules, disrupting the integrity of biomembranes, or binding with bioanions to result in a decreased level of essential bioanions. For example, copper and cadmium ions can replace the native zinc ion from many proteins and enzymes to a degree that depends on their affinities. The lead ion also inhibits a variety of enzymes. The toxicities of heavy metals have been reviewed extensively in the literature [1,2]. Millions of tons of water containing transition, lanthanide, and actinide metals are generated annually. Removal of these heavy metals has been the focus of extensive research. In many cases, reduction of heavy metals to low levels is possible. However, most technologies to remove heavy metals are relatively nonselective. When wastewaters are treated, the metals are removed as a multimetal mixture that is very difficult to separate into individual components. The end result is an unusable product that must be placed in containment systems.

In response to this need, we developed in our laboratories a new sorbent system for the selective removal of contaminant heavy metal species from aquatic systems. We investigated the selective removal of Pb(II), Cu(II), and Cd(II) ions by polyethylene glycol methacrylate gel (PEG-MA) beads that carry Cibacron Blue F3GA.

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EXPERIMENTAL

Preparation of PEG-MA Gel Beads

PEG-MA (Aldrich Chemical Co, St. Louis, USA) and ethylene glycol dimethacrylate (EGDMA) (Röhm and Haas, Darmstadt, Germany) was used as the monomer and the cross-linker, respectively. Benzoyl peroxide (BPO) (Fluka Chemie A. G., Buchs, Switzerland) and poly(vinyl pyrrolidone) (PVP, average molecular weight: 360.000) (Sigma Chemical Co, USA) were selected as the initiator and the stabilizer, respectively. The spherical gel beads were prepared by a suspension polymerization procedure. A typical procedure for the preparation of gel beads may be given as follows: The monomer phase, including PEG-MA (4 mL), EGDMA (0.8 mL), and 0.10 g of BPO, were charged into the polymerization reactor that included 0.5 g of PVP K-90 in 50 mL of distilled water at room temperature. The polymerization was conducted in a sealed cylindirical polymerization reactor at 75°C for 4 hours and at 85°C for 2 hours at a 400 rpm stirring rate. The gel beads were extensively washed with ethylalcohol and then by water to remove unreacted monomer and were stored in distilled water until use.

Cibacron Blue F3GA Immobilization

For the immobilization of Cibacron Blue F3GA on the PEG-MA gel beads, the following procedure was applied: 100 mL of 500 mg Cibacron Blue F3GA (Sigma Chemical Co, USA) solution was poured into 50 mL of distilled water that contained gel beads, and then 5.0 g of NaOH was added. The medium was then heated in a sealed reactor at 80°C for 4 hours at a stirring rate of 400 rpm. Under these experimental conditions, a chemical reaction took place between the Chloride groups of Cibacron Blue F3GA, and the hydroxyl groups of the PEG-MA gel beads resulting in with the elimination of NaCl, and covalent attachment of Cibacron Blue F3GA to the PEG-MA gel beads. The Cibacron Blue F3GA–immobilized gel beads were removed and washed with distilled water and methanol several times until all the physically adsorbed and/or absorbed dye molecules were removed.

Characterization of Gel Beads

Equilibrium Water Swelling Ratio

To determine the equilibrium swelling ratio of gel beads, approximately 3 g of dry polymer sample was put into a cylindirical tube. The height of the bed

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formed by the dry beads (H_d) was measured. Then, 50 mL of buffer solution at a certain pH and ionic strength was added into the tube. The sealed tube was shaken on a rotater at 30 rpm for 24 hours. At the end of this period, the height of the bed formed by the swollen gel beads (H_s) was recorded. The equilibrium water swelling ratio was calculated based on the following expression:

$$\text{Equilibrium Water Swelling Ratio} = (H_s/H_d) \times 100 \quad (1)$$

Elemental Analysis

The amount of Cibacron Blue F3GA immobilized to the PEG-MA gel beads was determined from an elemental analysis device (Leco, CHNS-932, USA).

Optical Micrograph

An optical photograph of PEG-MA gel beads was obtained by an optical microscope. (Nikon, Alphaphot Y.S, Japan)

Heavy Metal Adsorption/Desorption

Pb(II), Cu(II), and Cd(II) adsorption from the single metal aqueous solutions was investigated in batch adsorption-equilibrium experiments. Effects of the initial concentration of Pb(II), Cu(II), and Cd(II) and pH on the adsorption capacity were studied. Twenty milliliters of aqueous solutions with different concentrations (in the range of 10–80 mg/L) were treated with the Cibacron Blue F3GA–immobilized PEG-MA gel beads at room temperature. Adsorption flasks were stirred magnetically at 600 rpm. The suspensions were brought to the desired pH by the addition of sodium hydroxide and nitric acid. The pH was maintained in a range of $\pm$ 0.1 units until equilibrium was attained. Investigations were made for pH values in the range of 2.0 to 7.0. In all experiments, the polymer concentration was kept constant at 50 mg/20 mL. Blank trials (without polymer gel beads) were performed for each tested metal concentration. After the predetermined adsorption time (60 minutes), the gel beads were separated from the adsorption medium, and analyses for Pb(II), Cu(II), and Cd(II) in the aqueous phase was performed using a graphite furnace atomic absorption spectrophotometer (Unicam, 939 AAS, UK) with deuterium background correction. The respective hollow cathode lamps (Unicam, UK) for each metal ion were used. The spectral band width was 0.5 nm. The working currents/wavelengths for Pb(II), Cu(II), and Cd(II) were 10 mA/283.3 nm, 5 mA/324.8 nm, and 8 mA/228.8 nm respectively.

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The instrument response was periodically checked with known metal solution standards. For each set of data present, standard statistical methods were used to determine the mean values and standard deviations. Confidence intervals of 95% were calculated for each set of samples to determine the margin of error.

Data Treatment

After post-adsorption equilibrium determination of metal ion concentrations in the supernatant was achieved, C (mg/L), the concentration of adsorbed metal ions was calculated through the use of the following equation:

$$Q = [(C_i - C)/m_a]\, V \tag{2}$$

Here, Q is the metal ion adsorption (mg/g polymer); C_i is the initial metal ion concentration (mg/L); m_a is the quantity of adsorbent (g); and V is the suspension volume (mL).

To determine the reusability of the Cibacron Blue F3GA–immobilized PEG-MA gel beads, 3 consecutive adsorption-desorption cycles were repeated with the same affinity gel beads. Desorption of heavy metal ions was performed with a 0.10 mol/L HNO_3 solution. The PEG-MA gel beads were loaded with heavy metal ions, placed in this desorption medium, and stirred at 600 rpm for 30 minutes at room temperature. The final metal ion concentration in the aqueous phase was determined through the use of an atomic absorption spectrophotometer. The desorption ratio was calculated from the amount of metal ions adsorbed on the gel beads and the final metal ion concentration in the desorption medium. The desorption ratio was calculated from the following expression:

$$\text{Desorption ratio} = \frac{\text{(Amount of metal ions desorbed to the elution medium)}}{\text{(Amount of metal ions adsorbed onto the gel beads)}} \times 100 \tag{3}$$

RESULTS AND DISCUSSION

Properties of Polyethylene Glycol Methacrylate Gel Beads

PEG-MA is a hydrophilic polymer and very inert toward microbial degradation and resistant to many chemicals. PEG-MA gel beads create cross-linked structures. It is not soluble in water but swells in aqueous media; the degree of swelling depends on the cross-linking ratio. The equilibrium water swelling ratio of PEG-MA used in this study was 222% (vol/vol). The PEG-MA gel beads are transpar-

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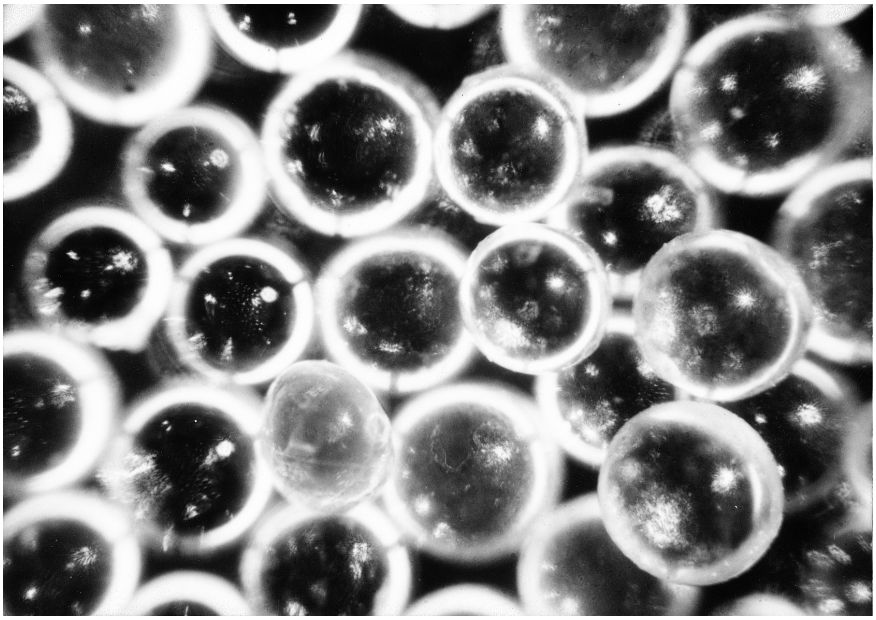

Figure 1. A representative photograph of the PEG-MA gel beads (taken at × 1000 and reduced at a proper ratio).

ent when the equilibrium water content was less than approximately 40% and opaque when the equilibrium content was higher than 60%. A representative photograph of PEG-MA gel beads is shown in Fig. 1. Elemental analysis of the unmodified PEG-MA and Cibacron Blue F3GA–immobilized PEG-MA gel beads was done, and the immobilization of the Cibacron Blue F3GA was found through the use of nitrogen and sulfur stoichiometry to be 42.6 μmol/g polymer.

Cibacron Blue F3GA release from the PEG-MA gel beads was also monitored continuously. Dye was not released in any of the adsorption and desorption media, which assured us that the cleaning procedure used for removal of physically adsorbed Cibacron Blue F3GA molecules from the PEG-MA gel beads was satisfactory.

Adsorption Capacity

Adsorption Rate

Figure 2 shows equilibrium adsorption times of the metal ions on the Cibacron Blue F3GA–immobilized PEG-MA gel beads from aqueous solutions at a constant pH of 5.5. Note that these batch experiments were performed with single solutions that each contained the ion of interest. As can be seen from the figure, metal ion adsorption capacity increased with time during the first 20 minutes

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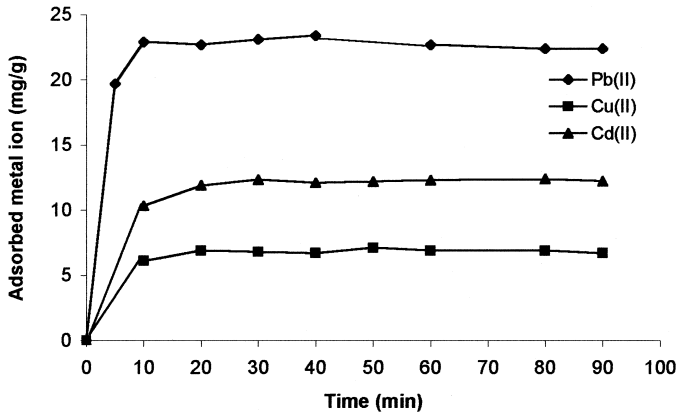

Figure 2. Adsorption rates of Pb(II), Cu(II), and Cd(II) on the Cibacron Blue F3GA–immobilized PEG-MA gel beads. Initial concentrations of heavy metal ions: 70 mg/L for Pb(II), 70 mg/L for Cd(II), and 30 mg/L for Cu(II): 20°C, pH: 5.5.

and then leveled off as the equilibrium adsorption capacity was reached. This is due to the high complex formation rate between the metal ions and the Cibacron Blue F3GA molecules on the surface of PEG-MA gel beads. Mass transfer limitations were also overcome by high driving force, which in the case of high metal concentration was the concentration difference of the metal ions between the liquid and the solid phases.

Data on the adsorption kinetics of heavy metal ions by various polymer sorbents have shown a wide range of adsorption rates. Dev and Rao studied sorption of different metal ions, including Cu(II), Cd(II), Co(II), Ni(II), Pb(II), Zn(II), and Mn(II), by polystyrene divinylbenzene–based macroreticular resin functionalized with bis(*N,N*'-salicylidene) 1,3 propanediamine ligands, and they reported that the kinetics of the resin-metal interaction was sufficiently rapid for most of the metal ions. Sorption reached equilibrium within 60 minutes (3). Roozemond et al. studied Cu(II) and Cd(II) uptake of 3,5-dimethyl-1-hydroxymethyl pyrazole–attached *p*-aminomethyl substituted poly(styrene-co-divinylbenzene) chelating polymer. They showed that adsorption was rather slow; after 2 days the resin appeared to reach equilibrium (4). Ebrahem and Hamdi studied various divalent ions, including Ni(II), Cu(II), Zn(II), and Cd(II), on a phenol formaldehyde polymer that contained poly(salicyl aldoxime 3,5-diylmethylene) and reported a 10 hour equilibrium adsorption time (5). Latha et al. studied ethylenediamine functionalized polyacrylamide resin for extraction of several metal ions, such as Fe(III), Fe(II), Cu(II), and Ni(II), and they reported that complexation reaction proceeds very slowly (equilibrium time was 5 hours) (6). Konishi et al. studied sorption of Zn(II), Cd(II), and La(III) by biopolymer gel beads of alginic acid, and they reported high adsorption rates in which equilibrium was achieved in approximately

2 hours (7). Many experimental and structural parameters determine the adsorption rate, such as stirring rate in the aqueous phase, structural properties of sorbent (e.g., porosity, surface area, topography, and swelling degree), amount of sorbent, ion properties (e.g., hydrated ionic radius, coordination complex number), initial concentration of heavy metal ions, chelate-formation rates between the complexing ligand and the metal ions, and existence of other metal ions that may compete with the metal ions of interest for the same active complexation sites. Therefore a comparison of the adsorption rates reported is too difficult to achieve. However, the adsorption rates obtained with the Cibacron Blue F3GA–immobilized PEG-MA gel beads produced seem to be good.

Effect of Initial Concentration of Metal Ions

Adsorption capacities of the metal ions investigated on the Cibacron Blue F3GA–immobilized PEG-MA gel beads are given in Fig. 3 as a function of the initial concentration of the ions within the aqueous phase. Metal ion adsorption capacity of the Cibacron Blue F3GA–immobilized gel beads increased first with increasing initial concentration of the ions, then it reached a saturation level. The maximum adsorption capacities of the Cibacron Blue F3GA–immobilized PEG-MA gel beads were 23.3 mg/g (112.4 μmol/g), 12.4 mg/g (110.3 μmol/g), and 7 mg/g (110.2 μmol/g) for Pb(II), Cd(II), and Cu(II), respectively. These capacity levels may be due to the presence of functional chelating groups on the gel beads. The specificity of the metal-chelating ligand (i.e., Cibacron Blue F3GA molecules) may also contribute to this high adsorption capacity. According to elemental analysis results, the PEG-MA gel beads carry 42.8 μmol Cibacron Blue F3GA.

A wide variety of sorbents with a wide range of adsorption capacities for heavy

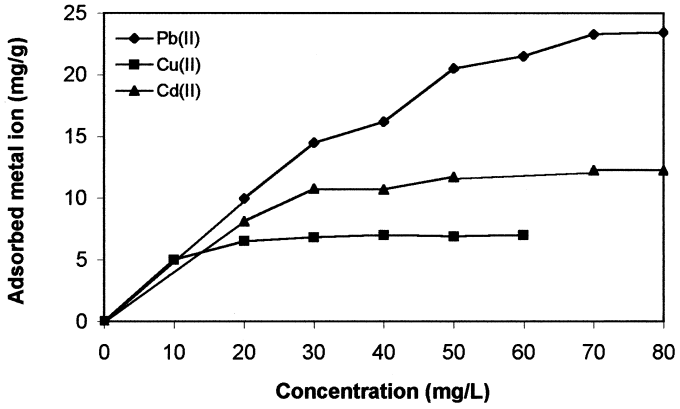

Figure 3. Adsorption capacities of Cibacron Blue F3GA–immobilized gel beads for Pb(II), Cd(II), and Cu(II) at 20°C and pH 5.5.

metal ions has been reported. Shreedhara-Murty and Ryan found 3.9–14.4 mg Cd(II)/g and 4.8–27 mg Cu(II)/g removal by cellulose-dithiocarbamate resins (8). Shambhu, Theodorakis, and Digenis immobilized polyamines onto polystrene, and the sorbents reached an adsorption value of 33 mg Cu(II)/g polymer (9). Janus, Morcellet and Delporte used poly(vinyl amine) sorbent for copper adsorption and reported an adsorption capacity of 0.5 mg Cu(II)/g (10). Denizli, Salih, and Piskin used Alkali Blue 6B–attached poly(EGDMA-HEMA) sorbents in which the maximum adsorption capacities were 2.3 mg Cu(II)/g, 5.5 mg Cd(II)/g, and 125 mg Pb(II)/g (11). Dev and Rao reported 29.2 mg Cu(II)/g, 51.7 mg Cd(II), and 80.8 mg Pb(II)/g adsorption capacities for polystyrene-vinylbenzene macroreticular resin functionalized with bis-(*N,N*'-salicylidene) 1,3-propanediamine (3). From the data that we obtained in this study, we concluded that the Cibacron Blue F3GA–attached PEG-MA microbeads are promising tools for removal for metal ions from aqueous media.

Effect of pH

Due to the protonation and deprotonation of the acidic and basic groups of the metal complexation ligand (dye molecules), the sorption behavior for metal ions is influenced by the pH value, which affects the surface structure of sorbents, the formation of metal ions, and the interaction between sorbents and metal ions. Therefore, the pH dependence of the adsorption for metal ions was investigated in detail. Figure 4 shows the pH profile data for metal ion adsorption. This figure shows that complexation behavior of metal ions is sensitive to pH changes, especially at lower pH region. The inhibition of metal chelation with a decrease in pH

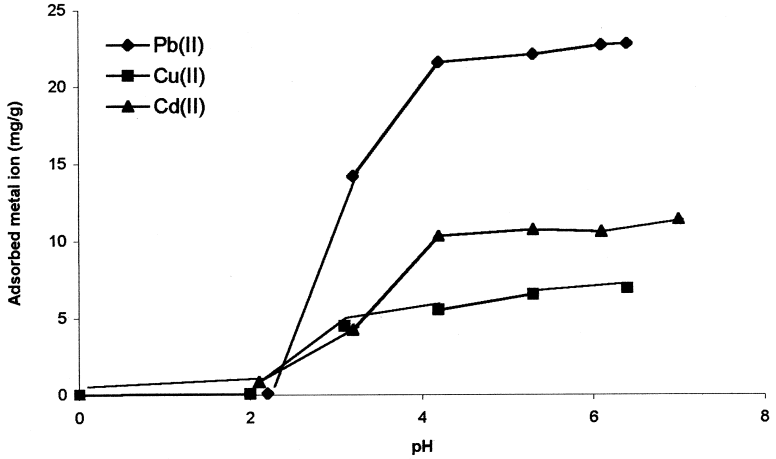

Figure 4. Effect of pH on adsorption of metal ions on the Cibacron Blue F3GA–immobilized gel beads. Cibacron Blue F3GA loading: 42.6 μmol/g; initial concentrations of metal ions were 70 mg/L for Pb(II), 40 mg/L for Cd, and 20 mg/L for Cu(II).

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was observed by several authors and in different sorbents (12,13). Thus, protons and metal ions appear to compete for the same binding sites. The adsorption capacities increased with increasing pH, reaching plateau values at around pH 5.5. High adsorption at relatively high pH values implies that metal ions interact with Cibacron Blue F3GA (unprotonated) groups by chelation (11,14,15).

Regeneration of Gel Beads

Regeneration was carried out in 0.1 mol/L HNO_3 solution. Cibacron Blue F3GA–immobilized gel beads loaded with maximum amounts of metal ions were placed within the desorption medium and the amount of metal ions desorbed in 30 minutes was measured. The maximum regeneration values were 98% for Pb(II), 98.5% for Cd(II), and 97.5% for Cu(II). When HNO_3 was used as a regeneration agent, the coordination spheres of the chelated metal ions were disrupted and subsequently metal ions were released from the Cibacron Blue F3GA–immobilized PEG-MA gel bead surface into the desorption medium. Results show that these gel beads can be used repeatedly without significantly losing their adsorption capacities for the metal ions studied.

Competitive Adsorption

The heavy metal ion adsorption capacities of Cibacron Blue F3GA–immobilized gel beads under competitive conditions (adsorption from solutions containing all heavy metal ions) for Pb(II), Cd(II), and Cu(II) are given in Table 1. This table shows the amounts of adsorption for each metal ion both in mass and molar basis, which were 14.96 mg (72.2 μmol) for Pb(II), 0.72 mg (11.3 μmol) for Cu(II), and 1.10 mg (9.8 μmol) for Cd(II). The adsorption capacity of Cibacron Blue F3GA–attached PEG-MA microbeads for all metal ions studied when adsorption was measured from single metal ion solutions was higher than was competitive adsorption capacity of the ions. When they existed in the same concentrations (0.63 μmol/L each), the ions compete for the same attachment sites and the system shows high selectivity to Pb(II).

Table 1. Competitive Adsorption of Pb(II), Cu(II), and Cd(II) on the Cibacron Blue F3GA–immobilized PEG-MA Gel Beads

Ions	mg Adsorbed Ion/g Polymer	μmol Adsorbed Ion/g Polymer
Pb(II)	14.96 ± 0.64	72.2 ± 3.1
Cu(II)	0.72 ± 0.10	11.3 ± 1.5
Cd(II)	1.10 ± 0.15	9.80 ± 1.3

Initial concentration of Pb(II), Cu(II), and Cd(II) was 0.63 μmol/L.

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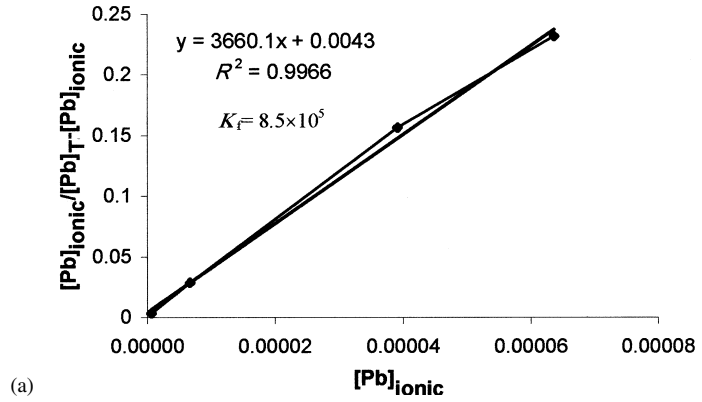
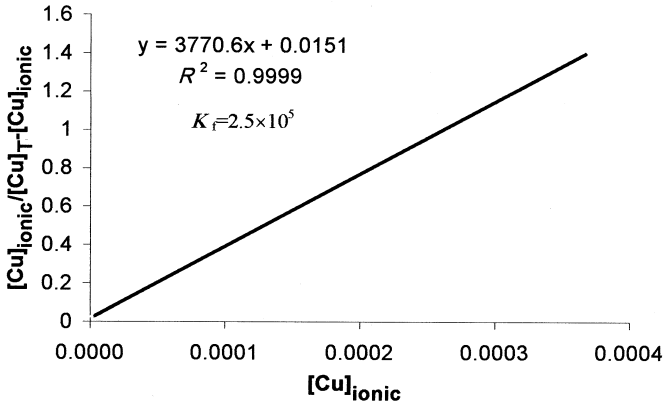
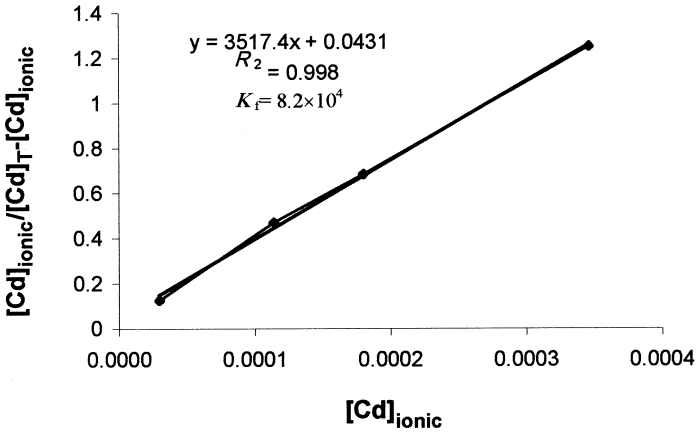


(a)



(b)



(c)

Figure 5. Plot of free to complexed Pb(II), Cu(II), and Cd(II) concentration ratios versus free metal concentration.

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The Stability Constants of Metal Ion–Dye Complex

The formation constants of Cibacron Blue F3GA–metal ion complex are of great importance. The formation constants of dye–metal ion complex were investigated by the application of the Ružić method (16). According to this data treatment, a plot of $[M]_{ionic}/[M]–[M]_{ionic}$ vs. $[M]_{ionic}$ should yield a straight line if one type of complex is predominant $[M]_T$ is the total metal concentration and $[M]_{ionic}$ is the concentration of the labile metal species). From the slope, the metal binding capacity of the ligand (C_L) is determined. From the intercept, determined according to the following equation, the apparent concentration stability constant (K') is defined:

$$\frac{[M_{ionic}]}{[M] - [M_{ionic}]} = \frac{1}{K'C_L} \tag{4}$$

The straight-line relationship of $[M]_{ionic}/[M]_T–[M]_{ionic}$ vs. $[M]_{ionic}$ indicates that the metal ions studied with Cibacron Blue F3GA interacted via one type of functional group, i.e., the SO_3H groups of the dye molecule (Fig. 5).

The K_f values obtained, 8.5×10^5 L/mol, 2.5×10^5 L/mol, and 8.2×10^4 L/mol for Pb(II), Cu(II), and Cd(II), respectively, clearly explain the results of competitive adsorption of these ions.

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