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# Congo Red-Attached Poly(EGDMA-HEMA) Micro beads for Removal of Heavy Metal Ions

Bekir Saḷh[a]; Adil Denizli[b]; Erhan Piskin[c]
[a] DEPARTMENT OF CHEMISTRY, HACETTEPE UNIVERSITY BEYTEPE, ANKARA, TURKEY [b] DEPARTMENT OF CHEMICAL ENGINEERING, HACETTEPE UNIVERSITY BEYTEPE, ANKARA, TURKEY [c] BIOENGINEERING DIVISION, HACETTEPE UNIVERSITY BEYTEPE, ANKARA, TURKEY

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# Congo Red-Attached Poly(EGDMA–HEMA) Microbeads for Removal of Heavy Metal Ions

BEKIR SALİH
DEPARTMENT OF CHEMISTRY

ADIL DENİZLİ and ERHAN PİŞKİN*
DEPARTMENT OF CHEMICAL ENGINEERING AND BIOENGINEERING DIVISION

HACETTEPE UNIVERSITY
BEYTEPE, ANKARA, TURKEY

## ABSTRACT

In this study we investigated a new sorbent system, Congo Red-attached poly(EGDMA–HEMA) microbeads, for removal of heavy metal ions from aqueous solutions. Poly(EGDMA–HEMA) microbeads were prepared by suspension copolymerization of ethylene glycol dimethacrylate (EGDMA) and hydroxyethyl methacrylate (HEMA) by using poly(vinyl alcohol), benzoyl peroxide, and toluene as the stabilizer, the initiator, and the diluent, respectively. Congo Red molecules were then covalently attached to these microbeads. Microbeads (150–200 μm in diameter) with a swelling ratio of 55% and carrying 14.5 μmol Congo Red/g polymer were used in the adsorption/desorption studies. Adsorption rate and capacity of the microbeads for selected metal ions, i.e., Cd(II), Cu(II), Zn(II) and Pb(II), were investigated in aqueous media containing different amounts of these ions (1–500 ppm) and at different pH values (1.5–7.5). Very high adsorption rates were observed at the beginning, and adsorption equilibria were then gradually achieved in about 5–90 minutes. The maximum adsorptions of metal ions onto the Congo Red attached microbeads were 18.3 mg/g for Cd(II), 2.9 mg/g for Cu(II), 53.8 mg/g for Zn(II), and 165 mg/g for Pb(II). However, when the metal ions competed (in the case of adsorption from their mixture), the amounts of adsorption for Cd(II), Zn(II), and Pb(II) were quite close. Desorption of metal ions was studied by using 2 M NaCl (pH 7.0) for Cd(II) ions and 0.1 M $HNO_3$ (pH 1.0) for Cu(II), Zn(II),

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* To whom correspondence should be addressed at Hacettepe University, P.K. 716, Kizilay, 06420 Ankara, Turkey. Telephone and FAX: (90) 312 2352330 or (90) 312 4406214.

and Pb(II) ions. High desorption ratios (more than 85%) were achieved in all cases. Adsorption/desorption cycles showed the feasibility of repeated use of this novel sorbent system.

*Key Words.* Congo Red; Poly(EGDMA–HEMA) microbeads; Heavy metal ions

## INTRODUCTION

The presence of heavy metal ions in the environment is a major concern due to their toxicity to many life forms. Treatment of wastewaters containing heavy metal ions requires concentration of the metals into a smaller volume followed by recovery or secure disposal. Heavy metal ions can be removed by adsorption on solid carriers. Nonspecific sorbents, such as activated carbon, metal oxides, silica and ion-exchange resins have been used (1–3). But recently, specific sorbents are used have been considered as one of the most promissing techniques (4–6). Specific sorbents consist of a ligand (e.g., ion-exchange material or chelating agents) which interacts with the metal ions specifically, and a carrier matrix which may be an inorganic material (e.g., aluminum oxide, silica, or glass) or polymer microbeads [e.g., polystyrene, cellulose, poly(maleic anhydride), or polymethyl methacrylate] (7–10).

As carrier matrices, polymer microbeads have attracted the most attention because they may be easily produced in a wide variety of compositions and modified into specific sorbents by introducing a variety of ligands. Synthetic polymers have almost entirely displaced inorganic carriers, with a few exceptions, for the separation of heavy metal ions (11–14). Nonporous or porous polymer microbeads with average diameters of more than 100 μm are usually used in these applications. In order to increase the active surface area available for attachment of the ligands, and therefore for specific adsorption, porous polymeric microbeads are preferentially utilized. There are commercially available polymer-based porous sorbents which exhibit surface areas 100–250 $m^2$ or even larger per gram of sorbent (15). One of the most critical points in the use of porous sorbents is their pore structure because the pore diffusion and the surface area in the pores determine adsorption rate and capacity, respectively. Sorbents with highly open pore structures are needed for high adsorption rates. However, the highly active surface area of porous sorbents is mainly due to the fine pores in the matrix which are not available for large molecules. In other words, large molecules cannot penetrate within these fine pores and therefore cannot use the active surface area there, which means that large molecules have low adsorption capacities. In summary, the optimization of pore structures of carrier matrices is very important for achieving both high adsorption rates and high adsorption capacities.

In our recent studies we prepared water-swellable poly(ethylene glycol dimethacrylate–hydroxyethyl methacrylate) microbeads with different sizes and swelling abilities (16). For heavy metal ions removal, we proposed covalently attaching a new ligand, i.e., Congo Red, to these microbeads (17). The preparation and characterization of these sorbents, and the results of preliminary adsorption/desorption studies with a model ion (i.e., cadmium ions), were reported in our previous papers (17–19). Here we are presenting our further studies to show the selectivity and useability of these sorbents for the removal of other important heavy metal ions, i.e., Cd(II), Cu(II), Zn(II), and Pb(II), which are often responsible for environmental pollution.

## EXPERIMENTAL

### Congo Red-Attached Poly(EGDMA–HEMA) Microbeads

Details of the preparation and characterization of plain poly(EGDMA–HEMA) microbeads are reported elsewhere (16). The comonomers, ethylene glycol dimethacrylate (EGDMA) (Röhm and Haas, Germany) and 2-hydroxyethyl methacrylate (HEMA) (Sigma Chemical Co., USA) were copolymerized in suspension by using benzoyl peroxide (BPO) and poly(vinyl alcohol) (PVAL; MW: 100,000, 98% hydrolyzed, Aldrich Chemical Co., USA) as the initiator and the stabilizer, respectively. Toluene (Merck, Germany) was included in the recipe as the diluent (as a pore former). Table 1 gives the recipe and polymerization conditions to obtain copolymer microbeads with a swelling ratio of 55% and in the size range of 150–200 μm (in swollen form).

Preparation and characterization of the Congo Red-attached poly(EGDMA–HEMA) microbeads were reported in detail in our previous paper (17). In order to prepare microbeads containing 14.5 μmol Congo Red/g polymer, which was the maximum amount of dye attachment that we have reached, the following procedure was applied: 10 mL of an aqueous solution containing 300 mg Congo Red (BDH Ltd., UK) was poured into 90 mL of a suspension of poly(EGDMA–HEMA) microbeads in distilled water (containing 3.0 g of the microbeads), and then 4.0 g NaOH was added. The medium was heated in a sealed reactor for 4 hours at a stirring rate of 400 rpm and a constant temperature of 80°C. The dye-attached poly(EGDMA–HEMA) microbeads were filtered and then washed with distilled water and methanol several times until all of the physically ad-

TABLE 1
Recipe and Polymerization Conditions for Preparation of Plain Poly(EGDMA–HEMA) Microbeads with a Swelling Ratio of 55% and in the Size Range of 150–200 μm

- Aqueous dispersion phase:
  - 50 mL distilled water
  - 0.2 g PVAL
- Organic phase:
  - 8 mL EGDMA
  - 4 mL HEMA
  - 12 mL toluene
  - 0.06 g BPO
- Polymerization conditions:
  - Reactor volume: 100 mL
  - Stirring (mechanical) rate: 600 rpm
  - Temperature and time: first at 65°C for 4 hours and then at 90°C for 2 hours

sorbed and/or absorbed dye molecules were removed (17). The amount of Congo Red molecules attached covalently to the plain poly(EGDMA–HEMA) microbeads was evaluated from the elemental analysis data of the dried samples obtained by using an elemental analysis device (Leco, CHNS-932, USA) (17).

## Adsorption/Desorption Studies

### *Adsorption*

Adsorption of heavy metal ions from aqueous solutions was investigated in batch adsorption-equilibrium experiments. Effects of the initial concentration of metal ions and pH of the medium on the adsorption rate and capacity were studied. Aqueous solutions (20 mL) containing different amounts of metal ions (in the 1–500 ppm range) were treated with the sorbents (i.e., the plain and the dye-attached poly(EGDMA–HEMA) microbeads) at different pH values (in the 1.5–7.5 range) (adjusted with universal buffer solution) at room temperature in flasks agitated magnetically at an agitation speed of 600 rpm. After adsorption, the polymeric microbeads were separated from the polymerization medium by filtration. The concentrations of the metal ions in the aqueous phases after the desired treatment periods were measured by using an Atomic Absorption Spectrophotometer (GBC 932 AA, Australia). The amount of adsorption per unit mass of the microbeads was evaluated by using the following expression:

$$Q = [(C_0 - C)V]/m \qquad (1)$$

Here, $Q$ is the amount of metal ions adsorbed onto a unit mass of the microbeads (mg/g); $C_0$ and $C$ are the concentrations of the metal ions in the initial solution and in the aqueous phase after treatment for certain period of time, respectively (mg/mL); $V$ is the volume of the aqueous phase (mL); and $m$ is the amount of poly(EGDMA–HEMA) microbeads used (g).

Competitive adsorption of Cd(II), Cu(II), Zn(II), and Pb(II) from their mixture was also investigated batchwise. A solution (20 mL) containing 10 ppm of each metal ion was treated with 100 mg of the Congo Red-attached poly(EGDMA–HEMA) microbeads at a pH of 7.0 at room temperature in flasks agitated magnetically at an agitation speed of 600 rpm. After adsorption, the concentrations of the metal ions in the supernatant were measured by using an Atomic Absorption Spectrophotometer. The amount of adsorption per unit mass of the microbeads was evaluated by using Eq. (1).

### Desorption and Reuse

Desorption of heavy metal ions was studied in buffer solutions containing 2 M NaCl at pH 7.0 for Cd(II) ions and 0.1 M $HNO_3$ at pH 1.0 for Cu(II), Zn(II), and Pb(II). The microbeads carrying metal ions were placed in this desorption medium and stirred (at a stirring rate of 600 rpm) for 2 hours at room temperature. The final metal ion concentration in the aqueous phase was determined by using an Atomic Absorption Spectrophotometer. In order to prevent interference of NaCl during the atomic absorption measurements for Cd(II) analysis, 4000 ppm $NH_4NO_3$ matrix modifier was added to the sample solution (1:1 ratio). The desorption ratio was calculated from the amount of metal ions adsorbed on the microbeads and the final metal ions concentration in the desorption medium by using the following equation:

$$\text{Desorption ratio} = \frac{\text{amount of metal ions desorbed to the elution medium}}{\text{amount of metal ions adsorbed on the microbeads}} \times 100 \qquad (2)$$

In order to obtain the reuseability of the Congo Red-attached poly(EGDMA–HEMA) microbeads, the adsorption–desorption cycle was repeated 5 times by using the same sorbent.

## RESULTS AND DISCUSSION

The poly(EGDMA–HEMA) microbeads used in this study were prepared and characterized in our earlier studies (16–19). These microbeads are crosslinked hydrophilic matrices, i.e., hydrogels. Therefore, they do not dissolve in aqueous media but do swell, depending on the degree of crosslinking and on the hydrophilicity of the matrix. The equilibrium swelling ratio (the ratio of the volumes of the microbeads before and after swelling) of the microbeads used in this study, which were prepared with the recipe given in Table 1, is 55% (17). These swollen microbeads have an average diameter within the 150–200 μm range. It should be noted that these microbeads are quite rigid and strong due to their highly crosslinked structure, and are therefore suitable for fixed-bed or fluidized-bed column applications.

The dry poly(EGDMA–HEMA) microbeads are opaque (white in color), which is an indication of the porosity in the matrix, as a result of the diluent (i.e., toluene) used in the polymerizations (16). However, the opacity of the microbeads significantly decreases and the color of the microbeads becomes light blue when the microbeads are swollen in water (17). After attachment of the dye (i.e., Congo Red), the size of the swollen microbeads did not change but the color became light red, which is a clear indication of the incorporation of dye molecules in the structure of the poly(EGDMA–HEMA) microbeads (17).

As shown in our previous paper, the dye molecules were attached to the poly(EGDMA–HEMA) microbeads by covalent bonding via hydroxyl groups (17). Poly(EGDMA–HEMA) microbeads containing 14.5 μmol dye/g polymer, which was the maximum amount of dye attachment that we reached, were used in this study.

The adsorption rate and capacity of these sorbents for the heavy metal ions, namely Cd(II), Cu(II), Zn(II), and Pb(II), and their reuseabilities are presented and discussed separately in the following sections.

## Adsorption Rate

Figure 1 shows the adsorption rates of heavy metal ions by Congo Red-attached poly(EGDMA–HEMA) microbeads as a function of time. Note that these batch experiments were performed by using single (not together) solutions of the ions of interest. The initial concentrations of the ions within the aqueous phase were changed between 1 and 500 ppm. The medium pH values were 6.8 for Cd(II), 4.1 for Cu(II), 6.8 for Zn(II), and 7.0 for Pb(II). As seen from Fig. 1, high adsorption rates are observed at the beginning of adsorption, and then plateau values (i.e., adsorption equilibria) are gradually reached within 5–90 minutes for all metal ions. The adsorption of metal ions was quite fast, especially when the metal ion concentration was high. This may be due to high driving force, which is the metal ion concentration difference between the liquid (i.e., adsorption

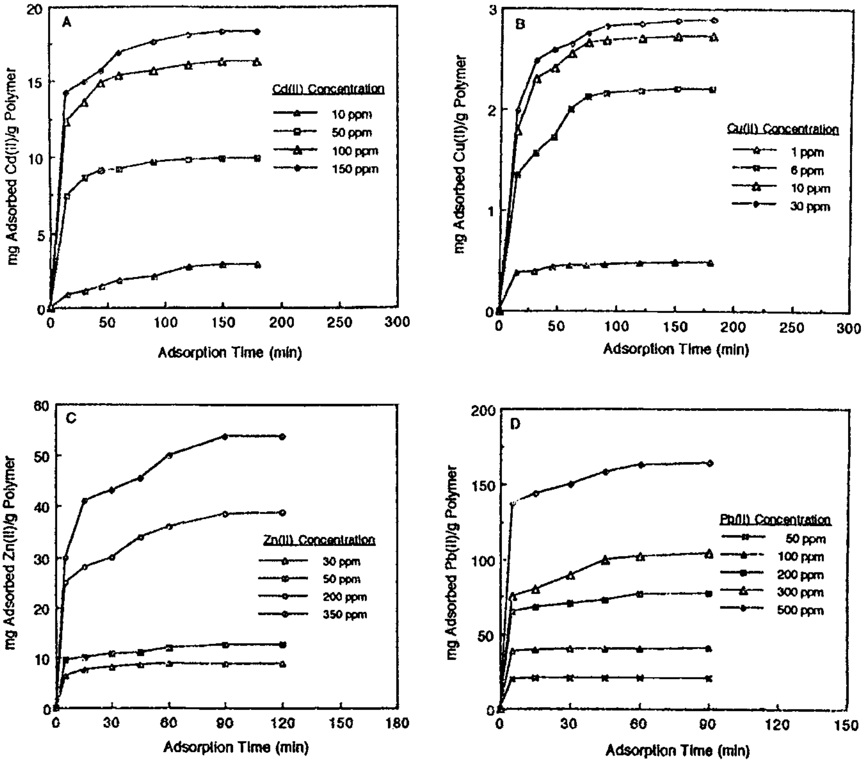

FIG. 1 Adsorption rates of heavy metal ions on the Congo Red-attached poly(EGDMA-HEMA) microbeads at 20°C: (A) for Cd(II), (B) for Cu(II), (C) for Zn(II), and (D) for Pb(II).

medium) and the solid (i.e., the microbeads) phases in the case of a high metal ion concentration.

It can be seen that Pb(II) and Zn(II) were adsorbed much faster than Cd(II) and Cu(II) due to the much higher affinity of immobilized Congo Red molecules to these ions. The order of adsorption rate is Pb(II) > Zn(II) > Cd(II) > Cu(II).

Experimental data on the adsorption kinetics of heavy metal ions by various sorbents have shown a wide range of adsorption rates. For example, Reed and Matsumoto considered 6 hours to be a short equilibrium time in their cadmium adsorption kinetic studies in which they used activated carbon as the sorbent (3, 20). Konishi et al. investigated the recovery of zinc, cadmium, and lanthanum by biopolymer gel particles of alginic acid. In their recent paper they reported very high adsorption/desorption rates for lanthanum by alginic acid, in which equilibrium was achieved in about 60–90 minutes (21). Shreedhara-Murthy and Ryan investigated mercury, copper, cadmium, lead, and uranium adsorption on cellulose–dithiocarbamate resins and reported that the adsorption rates were very slow (22). Egawa studied uranium adsorption on polyacrylonitrile fibers containing amidoxime groups and reported a 7-hour equilibrium adsorption time (23). Note that in such an adsorption process there are several parameters which determine the adsorption rate: agitation (or flow) rate in the aqueous phase, sorbent structural properties (e.g., size, porosity, surface area), amount of sorbent, metal ion properties (e.g., hydrated ionic radius), initial concentration of metal ions, chelate-formation rate, and, of course, the existence of other ions which may compete with the ions of interest for the active adsorption sites. All individual experimental studies published in the literature have been performed under different conditions. Consequently, it is almost impossible to compare the adsorption rates reported.

## Adsorption Capacity

### *Effect of Initial Concentration of Metal Ions*

Adsorption capacities of heavy metal ions of the Congo Red-attached poly(EGDMA–HEMA) microbeads are presented as a function of the initial concentration of metal ions within the aqueous phase in Fig. 2. This figure was prepared by using the plateau values shown in Fig. 1. Note that the adsorption capacities for Cu(II) ions were much lower than for the others; therefore, an enlarged graph for these ions is also given in Fig. 2.

The amount of metal ions adsorbed per unit mass of the polymer (i.e., adsorption capacity) first increased with the initial concentration of metal ions, then reached a plateau value which represents saturation of the active points (which are available for specific metal ions) on the microbeads. These plateau values were 125, 20, 300, and 350 ppm for Cd(II), Cu(II), Zn(II) and Pb(II), respectively. The maximum adsorption capacities of the Congo Red-attached poly(EGDMA–HEMA) microbeads are 18.3, 2.9, 53.8, and 165.0 mg per gram of the polymer for Cd(II), Cu(II), Zn(II), and Pb(II), respectively. The order of affinity is Pb(II) > Zn(II) > Cd(II) > Cu(II).

It should be noted the nonspecific adsorptions [adsorptions on the plain poly(EGDMA–HEMA) microbeads] of heavy metal ions were relatively low, about 0.93, 0.36, 1.47, and 12.3 mg/g of the polymer for Cd(II), Cu(II), Zn(II), and Pb(II), respectively. There is neither ion-exchange nor com-

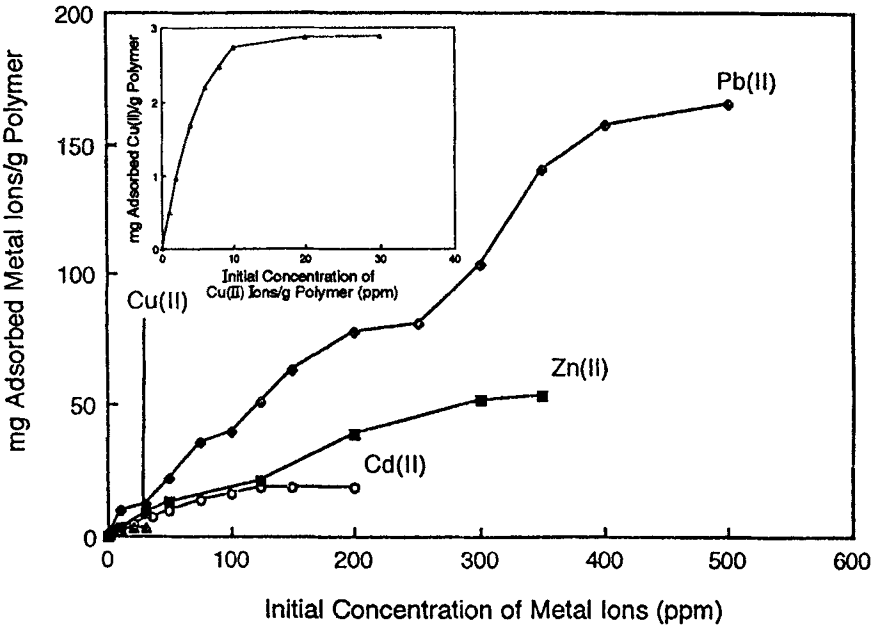

FIG. 2 Adsorption capacity of Congo Red-attached poly(EGDMA–HEMA) microbeads for heavy metal ions.

plex forming groups onto the plain poly(EGDMA–HEMA) microbeads. This adsorption may be due to diffusion of metal ions into the swollen matrix of the microbeads.

Some previous data on the sorption equilibria of cadmium, zinc, and lanthanum for aqueous solutions by natural (chitin and chitosan) and synthetic (alginic acid gels) polymers were compared by Konishi et al. [21]. These data and other reported values (3, 5, 20, 21, 24, 25) were again in a very wide range (4.8–96.3 mg cadmium/g, 2.35–17.6 mg zinc/g, 2.36 mg lanthanum/g), in which the initial concentration of the metal ions in solution was in the 0.056–2.81 mg/L range. Shreedhara-Murthy and Ryan reported 4.8–27 mg copper/g and 3.9–14.4 mg cadmium/g values with cellulose–dithiocarbamate resins (22). Roozemond et al. reported 40 mg cadmium/g, 20 mg zinc/g, and 0.3–31.9 mg copper/g with pyrazole containing poly(styrene–divinylbenzene) sorbents (26). The extent of adsorption increased with the initial metal ion concentration as expected, similar to our data shown in Fig. 2.

### *Effect of pH*

It is well known that metal ion adsorption on both nonspecific and specific sorbents is pH-dependent (3, 5, 20, 21, 24). In the absence of complexing agents, the hydrolysis and precipitation of the metal ions are affected by the concentration and the form of soluble metal species. The solubility of metal ions is governed by the hydroxide or carbonate concentration. As discussed in detail by Boomhower (27) and Reed and Matsumoto (3), hydrolysis of metal ions becomes significant at approximately pH 8.5, 6.5, 7.0, and 7.5 for Cd(II), Cu(II), Zn(II), and Pb(II), respectively. The theoretical and experimental precipitation curves indicate that precipitation begins above these pH values, which also depends on the concentration of metal ions in the medium. Therefore, in our study, in order to establish the effect of pH on the adsorption of metal ions onto the Congo Red-attached poly(EGDMA–HEMA) microbeads, we repeated the batch equilibrium studies at different pH values in the 1.5–7.5 range. In this group of experiments the initial concentration of metal ions were 125, 30, 350, and 500 ppm for Cd(II), Cu(II), Zn(II), and Pb(II), respectively. Figure 3 shows the specific adsorption [i.e., adsorption by chelating with the

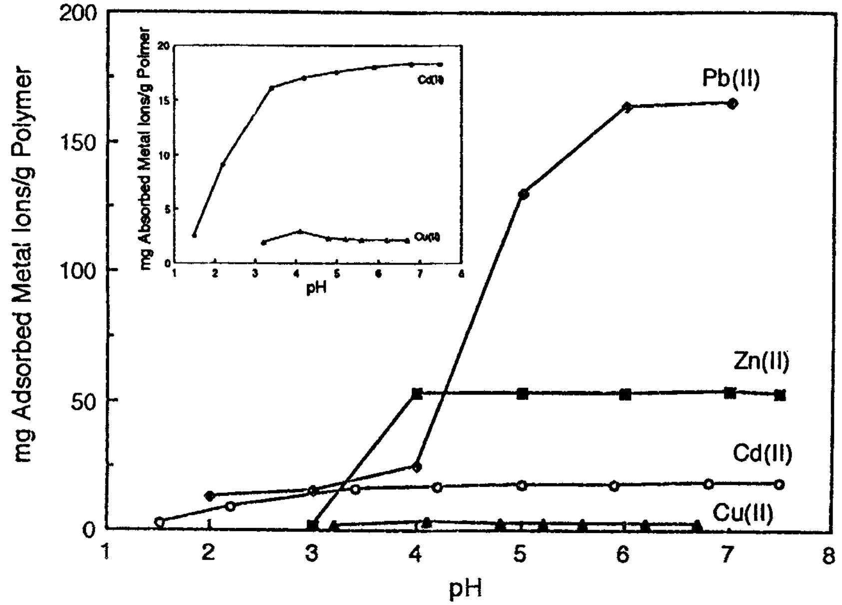

FIG. 3 Effect of pH on adsorption of heavy metal ions on Congo Red-attached poly(EGDMA–HEMA) microbeads.

Congo Red molecules attached on the poly(EGDMA–HEMA) microbeads] of metal ions. Note that the adsorption capacities for Cu(II) and Cd(II) ions were much lower than the others, therefore an enlarged graph for these ions is also given in Fig. 3.

As seen in Fig. 3, the adsorptions of Zn(II) and Pb(II) ions first increases linearly with increasing pH and almost reaches a plateau value around pH 4 for Zn(II) and 6.0 for Pb(II). High adsorption at pH values around 5.0–7.0 implies that metal ions interact with Congo Red not only through nitrogen atoms by chelating, but also through —$SO_3H$ groups by cation exchange, which are unprotonated at these pH values. There was a slight increase in the adsorption capacity for Cd(II) ions with pH for the pH values studied. There was almost no significant effect of pH on the adsorption of Cu(II) ions.

## *Competitive Adsorption*

Competitive adsorption of the metal ions evaluated in this study, namely, Cd(II), Cu(II), Zn(II), and Pb(II), were also investigated. Solutions (20 mL) containing 10 ppm of each metal ion were treated with 100 mg of the microbeads at pH 7.0 at room temperature. Figure 4 shows the amount of adsorption for each metal ions, which were 3.52 mg for Cd(II), 0.33 mg for Cu(II), 3.73 mg for Zn(II), and 4.36 mg for Pb(II). It is quite interesting to note that the adsorption capacity of the Congo Red-attached poly(EGDMA–HEMA) microbeads for Pb(II) ions (in the case of adsorption from single metal ions solutions) was much higher than for other ions

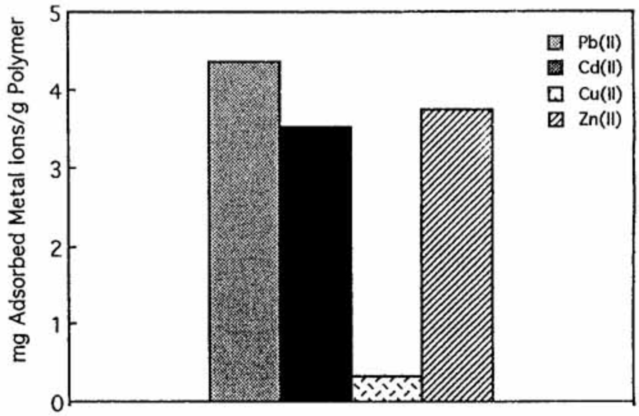

FIG. 4 Competitive adsorption of heavy metal ions on Congo Red-attached poly(EGDMA–HEMA) microbeads.

[especially from Cd(II) and Cu(II) ions]. However, when they exist in the same concentrations, they (except copper ions) do compete for the same active points, as shown in Fig. 4. It should be noted that the extent of adsorption of each type of metal ion is strongly dependent upon its relative concentration within the mixture. Therefore, it is almost impossible to generalize the order of adsorption or to define the extent of adsorption for each metal ion when they compete.

## Desorption and Repeated Use

Desorption of the adsorbed metal ions from the Congo Red-attached poly(EGDMA–HEMA) microbeads was also studied in a batch experimental set-up. Dye-attached microbeads carrying the maximum amounts of the respective metal ions were placed within the desorption medium

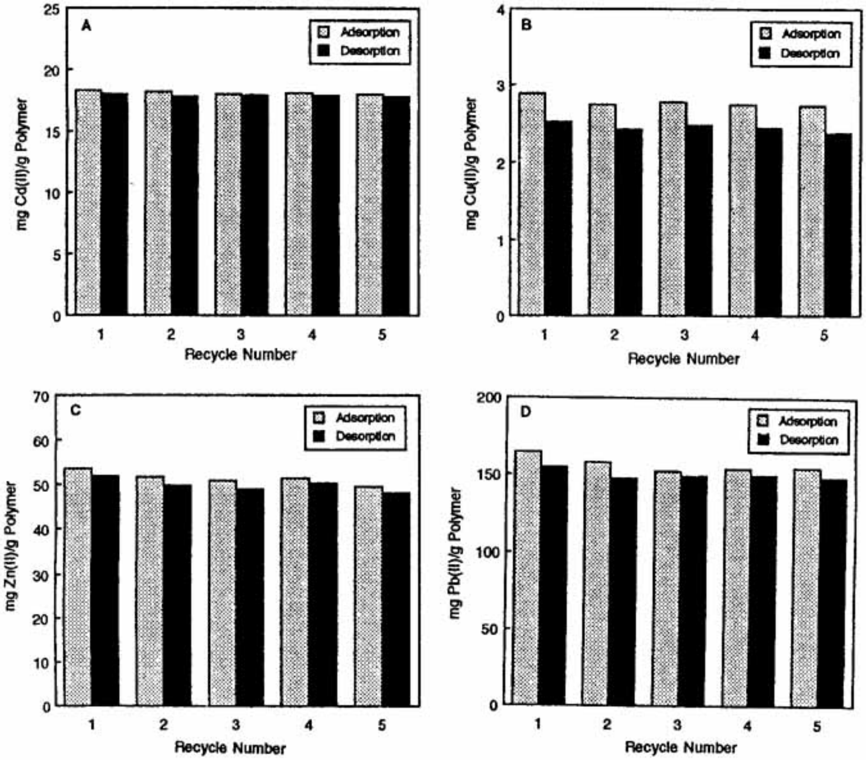

FIG. 5 Adsorption/desorption and repeated use of Congo Red-attached poly(EGDMA–HEMA) microbeads: (A) for Cd(II), (B) for Cu(II), (C) for Zn(II), and (D) for Pb(II).

containing 2 M NaCl at pH 7.0 for Cd(II) ions and 0.1 M $HNO_3$ at pH 1.0 for Cu(II), Zn(II) and Pb(II) ions, and the amount of metal ions desorbed in 2 hours was measured. The desorption ratio was then calculated by using the expression given in Eq. (2). Recoveries (or desorption ratios) were very high (up to 85%) with the eluant system and conditions used for all heavy metal ions, as also observed in Fig. 5.

In order to obtain the reuseability of the Congo Red-attached poly(EGDMA–HEMA) microbeads, the adsorption–desorption cycle was repeated 5 times by using the same sorbent. Figure 5 shows these data. As seen here, the resorption capacity of the sorbent for all metal ions did not change significantly during repeated adsorption–desorption operations.

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