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and

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Organized by

*The Society of Physical Chemists of
Serbia*

in co-operation with

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and

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INVESTIGATION OF SORPTION BEHAVIOUR OF Cu(II) AND Pb(II) ONTO A NOVEL EDTA MODIFIED COPOLYMER

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ABSTRACT

A new metal-chelating glycidyl methacrylate based macroporous copolymer with ethylenediaminetetraacetic acid functions was tested as a potential Cu(II) and Pb(II) ions sorbent from aqueous solutions. The sorption was studied in non-competitive batch experiments, by varying initial pH and metal concentration, contact time and temperature. Pseudo-first and pseudo-second order model, as well as Bangham, intra-particle and liquid film diffusion models were used to determine the nature of sorption kinetics and the rate of limiting step. Equilibrium sorption data were analyzed with Langmuir, Freundlich, Temkin and Elovich isotherm models.

INTRODUCTION

Lead and copper are the common heavy metal ions discharged from electroplating industries and battery manufacturing [1]. Polymer-based Cu(II) and Pb(II) sorbents are good alternative in comparison to clays, activated carbon and biosorbents, due to their large surface area, appropriate particle size distribution, chemical and mechanical strength [3]. As known, EDTA forms stable chelates with metal ions via metal-oxygen carboxylate and metal-nitrogen bonds [4]. The purpose of this study was to investigate possible synergetic effect of incorporation of multidentate ligand into macroporous polymer and adjustable porous structure on the sorption of Cu(II) and Zn(II) from single aqueous solutions.

EXPERIMENTAL

Sorption of Cu(II) and Pb(II) onto chelating sorbent was carried out in non-competitive batch conditions with a dose of sorbent of 10 g/L. The effect of initial pH (1 – 5), temperature (298 – 343 K), contact time (0 – 180 min) and initial metal concentration (0,0005 – 0,15 mol/dm³) was studied. For pH adjustment, 1 mol/dm³ HCl and 1 mol/dm³ NaOH solutions were used. At the predetermined time intervals sample aliquots were withdrawn, and analyzed with ICP-OES (Perkin Elmer, Model ICP 400). All experiments were repeated in triplicate or more if necessary.

RESULTS AND DISCUSSION

Figure 1 and 2 summarize the influence of initial pH and concentration, contact time and temperature on the Cu(II) and Pb(II) ions sorption. For all investigated sorption parameters, considerably higher affinity of sorbent towards Pb(II) than Cu(II) was observed. These results are a consequence of the Pb(II) smaller hydrated radius.

As seen from Figure 1a, both Cu(II) and Pb(II) sorption capacities are strongly dependent on the pH value with the maximum Q_t values at pH 4 and pH 5 for Cu(II) and Pb(II), respectively. The Q_t-t (Figure 1b) dependence for Pb(II) clearly shows two stages of the sorption process. Up to 30 minutes, a rapid Q_t increase was observed, followed by a gradually increase up to ca. 1350 mg/g after 30 minutes. On the otherside, Cu(II) sorption capacity gradually increased, reaching equilibrium value of ca. 450 mg/g after 30 minutes.

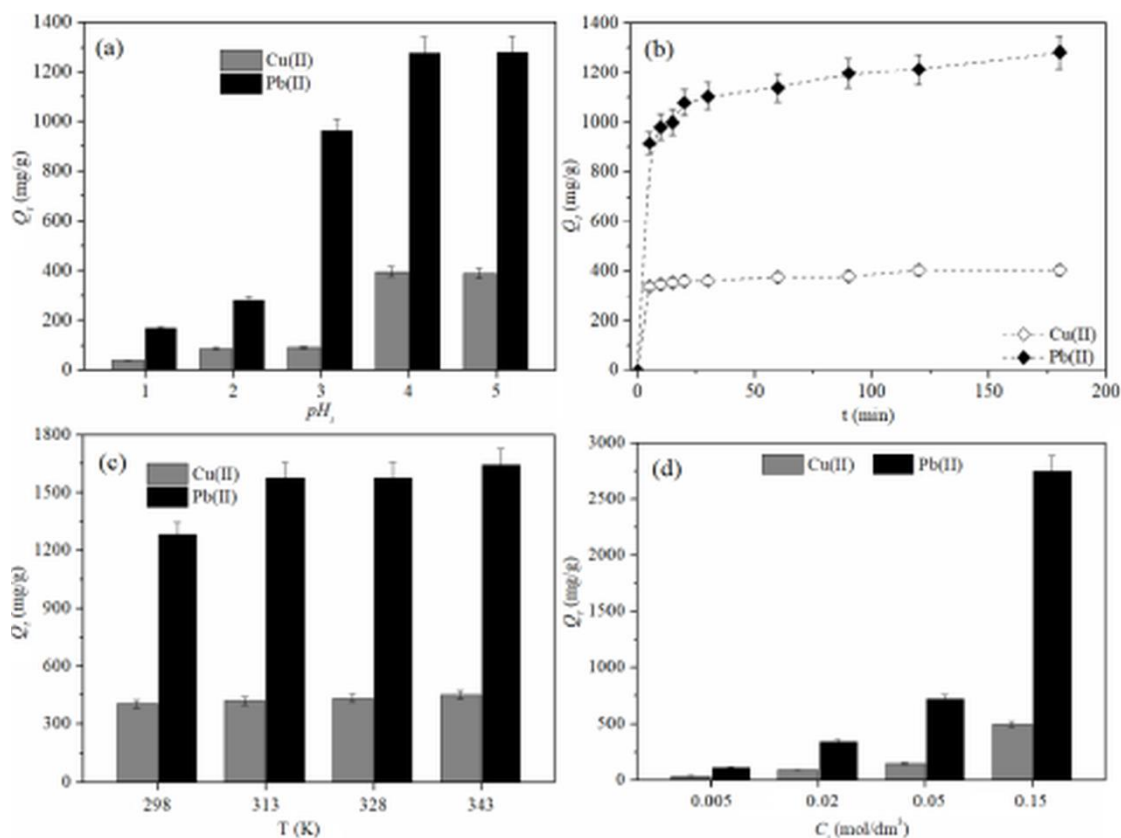


Figure 1. Effect of (a) initial pH, (b) contact time, (c) temperature and (d) initial concentration on the sorption capacity of Pb(II) and Cu(II)

On the otherside, Cu(II) sorption capacity gradually increased, reaching equilibrium value of ca. 450 mg/g after 30 minutes. It was observed (Figure 1c) that the capacity of sorption increased slightly along with the increasing temperature. The sorption capacity also increased with increasing the initial metal concentration (Figure 1d). The observed behavior can be attributed to the increase in the number of metal ions to the fixed quantity of adsorbents used in this study.

For analysis of the kinetic data, pseudo-first order model, pseudo-second order model, as well as Bangham, intra-particle and liquid film diffusion model were used [4]. The results are presented in Table 1. The highest correlation coefficient ($R^2 > 0.99$) and the Q_e^{calc} value (400 and 1282 mg/g) approximating to Q_e^{exp} (405 and 1284 mg/g), indicate the pseudo-second order model is the best fit for Cu(II) and Pb(II) sorption. Also, the high R^2 values (> 0.9) for intra-particle diffusion and liquid film diffusion model suggest the simultaneous influence of pore and film diffusion. This was

expected, since chelating sorbent has a high value of pore diameter corresponding to the half of pore volume ($d_{V/2} = 294$ nm).

Table 1. Kinetic parameters for Cu(II) and Pb(II) sorption onto chelating sorbent

	Cu(II)	Pb(II)
Q^{exp} (mg/g)	405	1284
Pseudo-first order model		
$k_1 \cdot 10^2$ (1/min)	1.49	1.375
Q_e^{calc} (mg/g)	84.58	323.35
R^2	0.961	0.944
Pseudo-second order model		
$k_2 \cdot 10^3$ (g/mg min)	0.07	0.17
Q_e^{calc} (mg/g)	400	1282
R^2	0.999	0.998
Bangham diffusion model		
α	0.09	0.15
$k_B \cdot 10^3$ (g/mg min)	1.78	1.69
R^2	0.952	0.979
Intra-particle diffusion model		
k_{1id} , (mg/g min ^{0.5})	10.12	67.75
$C_{1id} \cdot 10^2$ (mg/g)	302.36	762.53
R^2	0.995	0.942
k_{2id} (mg/(g min ^{0.5}))	6.851	22.32
C_{2id} (mg/g)	314.14	978.60
R^2	0.983	0.988
Liquid film diffusion model		
k_{fd} , (1/min)	0.02	0.01
C_{fd} ,	-1.57	-1.38
R^2	0.962	0.944

Table 2. Isotherm parameters for Cu(II) and Pb(II) sorption

	Cu (II)	Pb(II)
Langmuir isotherm model		
Q_{max} (mg/g)	1011	5000
$K_L \cdot 10^4$ (L/mg)	2.65	3.07
R^2	0.864	0.977
Freundlich isotherm model		
K_F (L ⁿ mg ¹⁻ⁿ /g)	0.94	32.36
N	1.31	1.28
R^2	0.998	0.997
Temkin isotherm model		
$K_T \cdot 10^3$ (L/mg)	6.88	10.13
b_T , (kJ/mol)	17.95	3.80
R^2	0.876	0.879
Elovich isotherm model		
K_E (L/mg)	0.99	1.00
Q_{mE} (mg/g)	667	3135
R^2	0.816	0.970

In this study, Langmuir, Freundlich, Temkin and Elovich isotherm models were used for equilibrium data analysis. The results are shown in Table 2. According to the high R^2 values (>0.99), the Freundlich model provided a better fit than the other used models. These results suggested that reversible sorption played a significant role in the Cu(II) and Pb(II) sorption process.

CONCLUSION

The sorption behavior of Cu(II) and Pb(II) onto chelating sorbent was investigated from aqueous solution. All the investigated sorption parameters have more pronounced effect on Pb(II) than Cu(II) sorption. The sorption of both metal ions followed the pseudo-second order kinetic model, with unambiguous influence of pore and film diffusion. Equilibrium data fitted best with Freundlich isotherm model.

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