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The Adsorption Characteristics of EDTA–Chelated Copper Ion by Activated Carbon in a Column Test

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ABSTRACT

The adsorption behavior of EDTA–chelated copper on activated carbon is described based on a consideration of electrostatic interaction and complexation of species. In this paper a series of column adsorption experiments is conducted to obtain more design data. Experimental studies indicate the adsorption behavior of chelated copper on activated carbon varies over the entire pH range, depending on the surface charge characteristics of activated carbon and the species distribution of metal and ligand. The extent of adsorption of copper and EDTA is significantly different at lower pH values. The extent of adsorption of EDTA-Cu chelates is higher than that of free copper ions in the solution. One modified model, termed the Bed-Depth-Service-Time (BDST) model, is used to relate the service time of a column to the bed mass. It is shown that the model data adequately describe the adsorption behavior of EDTA–chelated copper on activated carbon. The estimated constants might be used to estimate the service time of carbon dosage, but because deviation exists between the fitting curve and the experimental data obtained at later experimental periods, more study will be necessary to modify the model equation.

Key Words. Copper–EDTA removal; Adsorption; Activated carbon

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INTRODUCTION

Removing heavy metals in the treatment of a variety of industrial wastewaters has become increasingly important, particularly for wastewater that may contain complexing ions such as ethylenediaminetetraacetate (EDTA), tartrate, ammonia, etc. emitted from metal plating and finishing industries. Such ions may inhibit conventional precipitation treatments from removing heavy metals. Hence, adsorption processes have been proposed as an alternative separation technique. Several studies have demonstrated that organic complexing agents in solutions may influence the adsorption of metal species onto a solid surface. Huang et al. (1–4) studied several heterogeneous metal–ligand–solid aqueous systems and indicated that the adsorption of metal species by various adsorbents can be enhanced substantially with complexing agents. They also found adsorbent type, pH, and surface loading to be the most important factors affecting the extent of metal removal. Bhattatcharyya et al. (5, 6) reported that not only will organic ligands enhance the adsorption of heavy metal by activated carbon, but that such adsorption is a function of metal and ligand types, ligand to metal ratio, pH, and metal chelate species distribution. Furthermore, they proposed that the adsorption capacity is related to the electrostatic interaction between the species charge and the surface charge of activated carbon. In contrast, a few investigators noted that the presence of ligand might decrease the adsorption of heavy metals onto solid surfaces. Benjamin and Leckie (7) asserted that complexation by nonadsorbing ligands, such as Cl^- and SO_4^{2-}, decreases trace metal adsorption on oxide solids, such as γ-Al_2O_3, α-SiO_2, etc., by competing with the surface ligand. Peters and Shem (8) found the presence of EDTA decreased the affinity of lead to be adsorbed onto soil, thus EDTA could be used to extract adsorbed lead from soil. Chang and Ku (9) showed that EDTA significantly altered the adsorption behavior of copper on activated carbon due to the formation of copper chelates species in the solution. The results indicated that adsorption isotherms and kinetics are strong functions of solution pH. However, the above studies focused primarily on a batch experimental system. Although these batch experiments can give an indication of the effectiveness of carbon adsorption for removing chelated copper ion, adsorption isotherms cannot provide design data for a granular carbon column system because: 1) adsorption in a granular carbon column is not at equilibrium, 2) granular carbon rarely becomes totally exhausted in commercial processes, and 3) isotherms cannot predict chemical changes occurring in the column. Hence, further column adsorption experiments are usually necessary to obtain more practical design data.

In this study a series of up-flow column adsorption experiments were performed to examine factors governing the mode and extent of adsorption behaviors of copper–EDTA complexes with an activated carbon surface. Based on findings of the adsorption results at different pH values, adsorption mechanisms are proposed on the basis of electrostatic interaction and complexation of species. Then a useful scale-up design model is proposed to investigate its application for a column test at various pH values.

EXPERIMENTAL

The stock solutions used in this investigation were prepared with reagent-grade chemicals and RO-deionized water. Standard acid and base solutions ($HClO_4$ and NaOH) were prepared weekly for pH adjustment. The granular activated carbon Norit PK 3-5 was used as received; its characteristics are shown in Table 1. The pore size distribution of this carbon were determined by the Barrett–Joyner–Halenda (BJH) method using a Micromeritics ASAP 2000 analyzer. The pH was determined by direct titration.

Up-flow column adsorption experiments were performed for all kinetic studies. The laboratory apparatus for up-flow column tests consisted of vertical fixed-bed columns having an inner diameter of 3 cm, 30 cm height, and up to five columns which could be operated simultaneously. Each column could be charged with predetermined quantities of activated carbon. Samples were taken at different time periods. The samples were then filtered for copper and EDTA analyses. Solution feed rates were

TABLE 1
Characterization of Activated Carbon

Physical properties and specification	Norit PK 3-5
Total pore volume[a] (cm^3/g)	1.2
Pore volume of pores[b] between 17.0 and 3000.0 (Å)	0.600
Surface area (BET)[a] (m^2/g)	750
Surface area (BET)[c] (m^2/g)	587
pH value[a]	9–10
pH_{zpc}[d]	8.20 ± 0.05

[a] Data provided by the manufacturer (10).
[b] Determined by BJH cumulative desorption.
[c] Determined by BET.
[d] Determined by the direct titration method.

maintained with a Masterflux pump. The solution temperature was maintained at 25.0 ± 0.5°C for all experiments. The copper concentrations were analyzed by a GBC 904AA atomic adsorption spectrophotometer, and the EDTA concentrations were determined with a Dionex model DX-100 ion chromatograph.

RESULTS AND DISCUSSION

The up-flow column experiments were conducted at several pH values to explain the adsorption behavior of chelated copper in terms of the distribution of charge species and the surface charge of the activated carbon. In addition, the carbon dosage was varied at a fixed pH to investigate the design model for the adsorption of activated carbon.

The Effect of pH

The solution pH value was reported to have a significant effect on the extent of adsorption of chelated copper on the surface of activated carbon. The copper species distribution in the presence of EDTA was calculated and is shown in Fig. 1 as a function of pH based on the mass balances of the reacting species. This figure indicates that the free copper ion (Cu^{2+}), chelated copper species ($CuHEDTA^-$), and the free EDTA ions (H_4EDTA H_3EDTA^-) were all present for a pH less than 2, while the concentrations of negatively-charged $CuHEDTA^-$ and $CuEDTA^{2-}$ species were increasingly dominant when the pH was from 2 to 3. The pH_{zpc} (pH at zero point of charge) of activated carbon was determined by the titration method to be 8.20 + 0.05 as is also shown in Fig. 1. The carbon surface carries more positive charges in acidic solutions and more negatively-charged groups on the surface of activated carbon in alkaline solutions.

Up-flow column experiments were conducted in this study to examine adsorption behavior under various solution pH conditions. Figures 2 and 3 summarize copper and EDTA removal individually from the solution by activated carbon adsorption at different pH values. Based on local regulation of the effluent standard of Cu^{2+}, a 30% breakthrough point was chosen. The extent of adsorption of chelated copper at the 30% breakthrough point was highest at pH 3.27 and lowest at pH 1.82 for these experiments. Negatively-charged $CuHEDTA^-$ and $CuEDTA^{2-}$ chelates are assumed to exhibit a higher affinity for the positively-charged carbon surface for pH 3.27. For a solution pH greater than 8.20, the number of negatively-charged groups on the surface of activated carbon are assumed to increase gradually. Electrostatically, due to the loss of positive surface

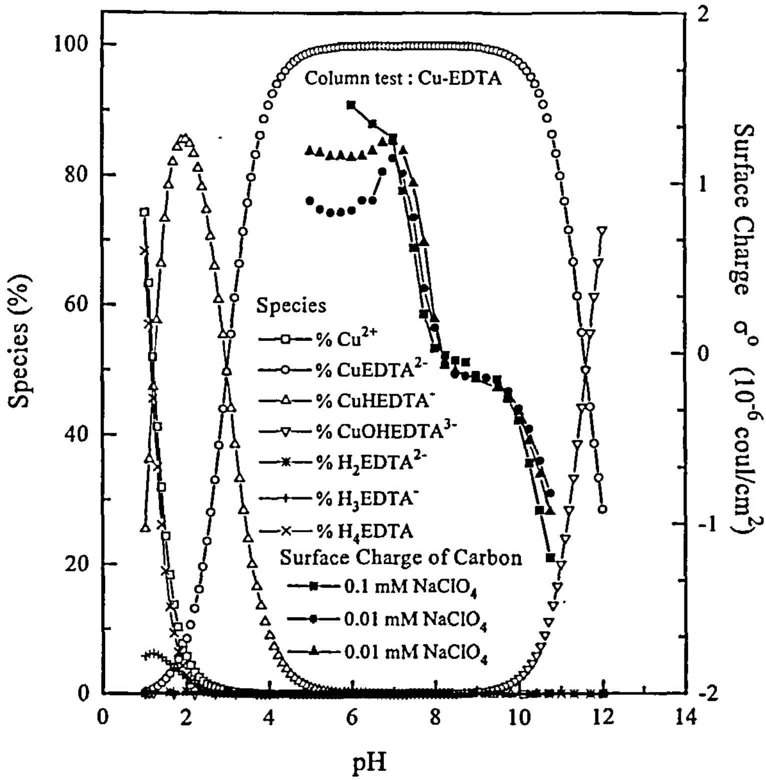


FIG. 1 Species distribution of Cu–EDTA and surface charge characteristics of activated carbon in single electrolyte solution as a function of pH, Cu = EDTA = 0.2 mM, carbon dosage = 4 g/80 mL.

charges and charge repulsion between $CuEDTA^{2-}$ species and these negatively-charged surface groups, the extent of adsorption of chelated copper was reduced at higher pH values. A similar pH effect on the batch adsorption of ligand chelated copper was also obtained for the Cu–NTA–Al_2O_3 system by Elliott and Huang (2) and for the Cu–EDTA–activated carbon system by Chang and Ku (9).

Although the negatively-charged $CuHEDTA^-$ and H_3EDTA^- species are more easily attracted by the highly positively-charged carbon surface at pH 1.82, for the most part the copper ions exist with positively-charged Cu^{2+} species at this pH, and they are assumed to exhibit a lower affinity for the highly positively-charged carbon surface in comparison with the negatively-charged $CuEDTA^{2-}$ chelates for the positively-charged car-

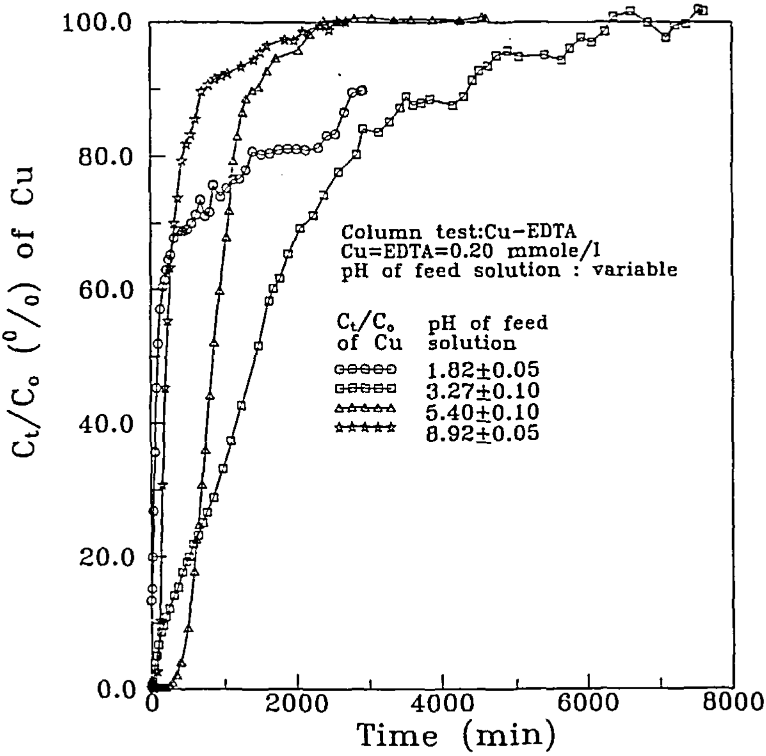


FIG. 2 Breakthrough curves for EDTA-chelated copper at various pH values, flow rate = 264.7 mL/h, carbon dosage = 45 g, 25.0 ± 0.5°C.

bon surface at pH 5.40 and for the slightly positively-charged carbon surface at pH 8.92. Hence, Fig. 2 indicates that for a solution pH of 1.82, the extent of adsorption of copper is lower than that for either pH 8.92 or pH 5.40 during the initial experimental period. However, as the experiment proceeded, the extent of adsorption of copper at pH 1.82 increased and gradually exceeded that at both pH 5.40 and 8.92. Because $CuHEDTA^-$ has a higher pK_a value than H_3EDTA^-, it is assumed that the H_3EDTA^- adsorbed on the surface of activated carbon attracts free Cu^{2+} species to form chelates, and this results in an increase of copper adsorption. The electrostatic interaction on the surface of activated carbon for acidic solutions is

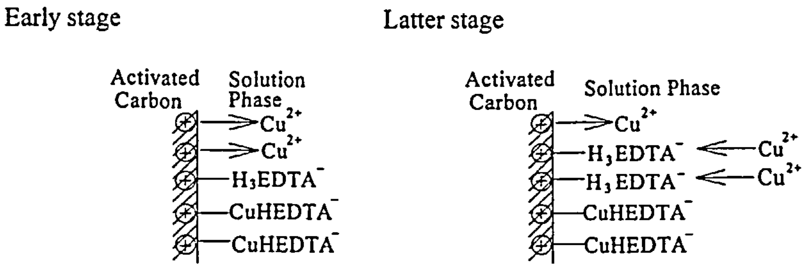


Figure 3 indicates that the extent of adsorption of EDTA at pH 1.82 is lower than that of EDTA at pH 5.40 in the initial experimental period. It was assumed that one of existing EDTA species, H_4EDTA, is neutral at

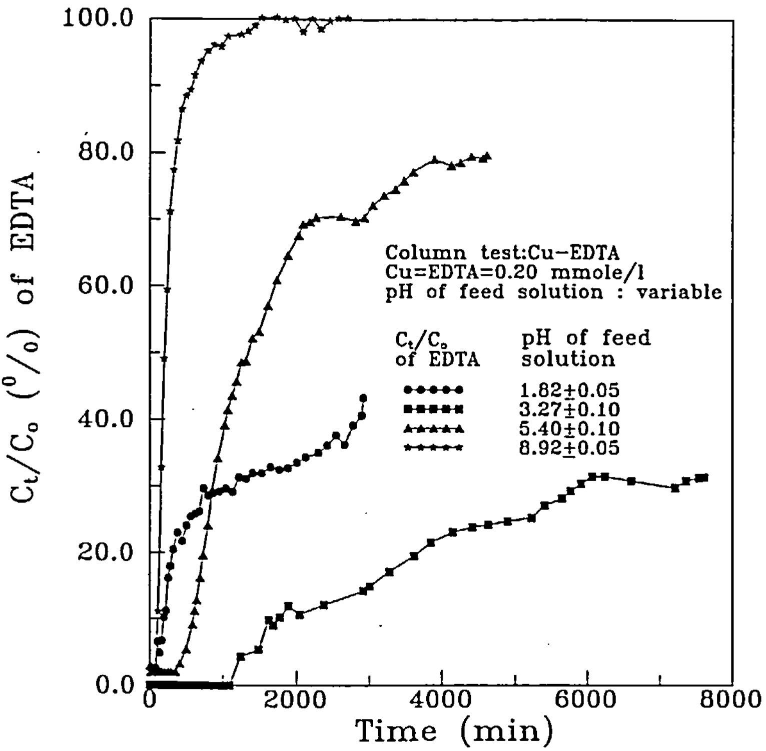


FIG. 3 Breakthrough curves for chelated EDTA at various pH values, flow rate = 264.7 mL/h, carbon dosage = 45 g, 25.0 ± 5°C.

pH 1.82, and that it exhibits a lower rate of adsorption on the highly positively-charged carbon surface in comparison with the negatively-charged $CuEDTA^{2-}$ chelates at pH 5.40. But as the experiment proceeds, the quantity of EDTA species adsorbed on the highly positively-charged surface of activated carbon at pH 1.82 is continuously increasing. On other hand, $CuEDTA^{2-}$ chelates adsorbed on the surface of activated carbon at pH 5.40 may result in the loss of positive surface charges, which decreases the capacity for negatively-charged chelates to be adsorbed on the surface of activated carbon. Consequently, the extent of adsorption of H_3EDTA^-, H_4EDTA, and $CuHEDTA^-$ on a highly positively-charged carbon surface at pH 1.82 might gradually exceed that of $CuEDTA^{2-}$ on a less positively-charged carbon surface at pH 5.40.

The effect of solution pH on metal removal was further studied, and the results are shown in Fig. 4. By viewing the breakthrough and pH

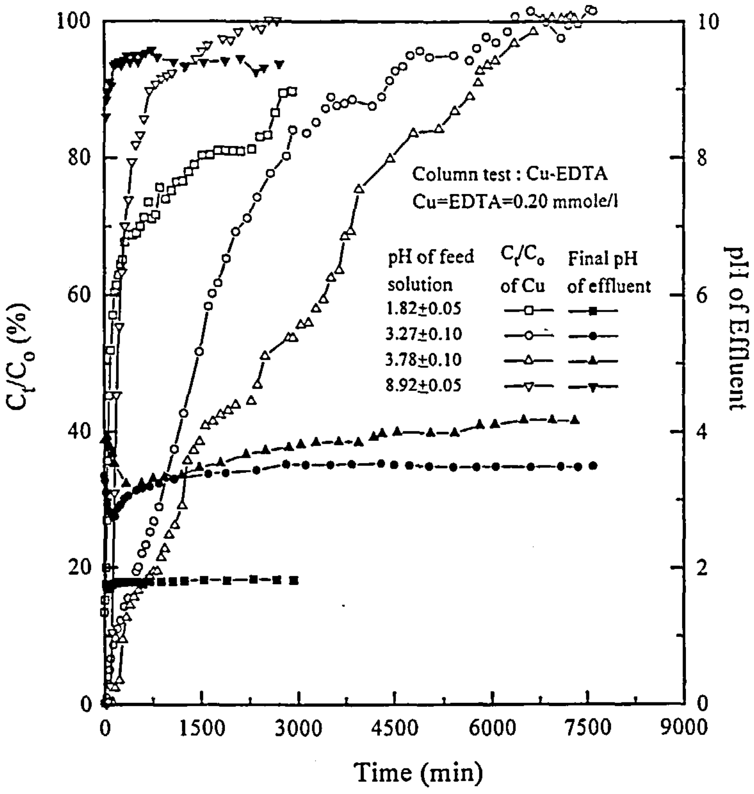


FIG. 4 Effect of initial solution pH on the breakthrough and pH curves, flow rate = 264.7 mL/h, carbon dosage = 45 g, 25.0 ± 0.5°C.

curves for initial pH values of 1.82, 3.27, 3.78, and 8.90, the solution pH was found to decrease in experiments conducted at initial pH values of 1.82, 3.27, and 3.78, and the solution pH was found to increase for pH 8.92 in the initial experimental period, which seems to coincide with the change in the breakthrough curves.

The Extent of Adsorption of Cu and EDTA

By comparing the extent of adsorption of copper and EDTA in the EDTA/Cu system, Fig. 5 combines the adsorption data of copper and EDTA. It shows that the extent of adsorption of copper and EDTA is similar at higher pH values and significantly different at lower pH values. Electrostatically, $CuEDTA^{2-}$ exhibited a very low affinity for the nega-

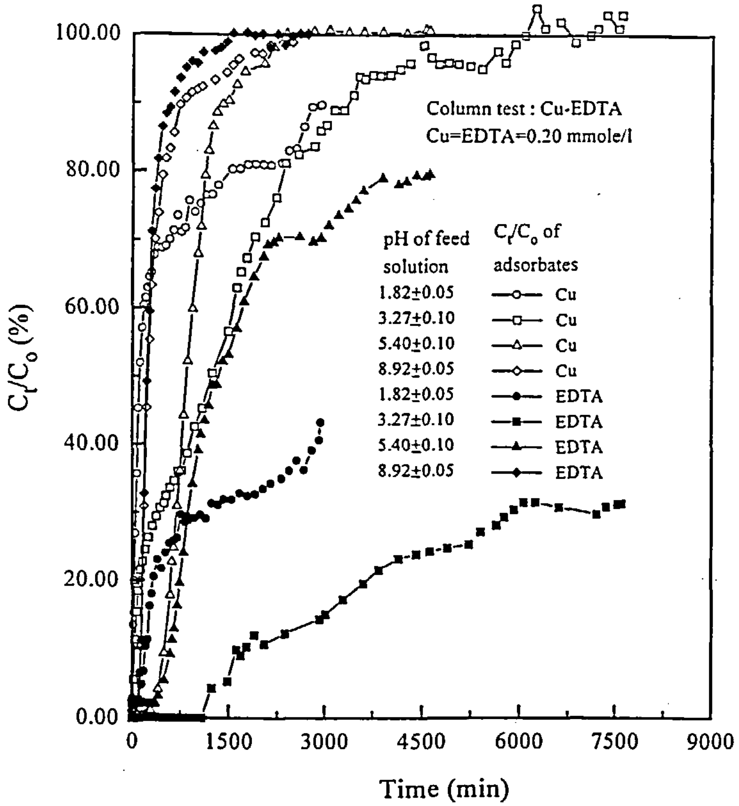
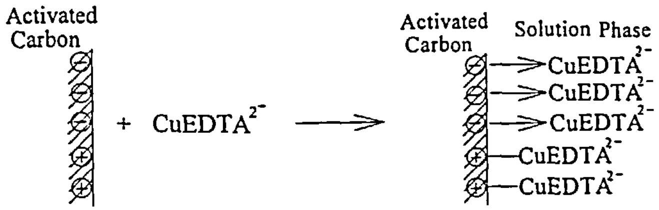


FIG. 5 Effect of solution pH on the extent of adsorption of Cu and EDTA, flow rate = 264.7 mL/h, carbon dosage = 45 g, 25.0 ± 0.5°C.

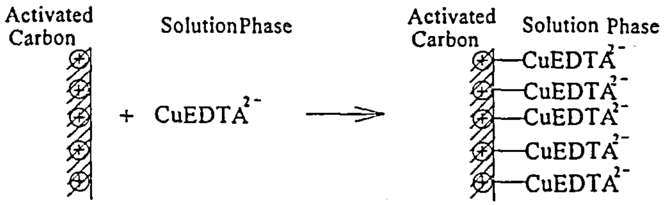
tively-charged surface of activated carbon at higher pH values, and this resulted in similar extents of adsorption of Cu and EDTA. As the solution pH was gradually decreased, more positive surface charges of activated carbon were formed. After operating the column for a certain period of time, there might be a very high concentration of hydrogen ions accumulated on the surface of activated carbon. These concentrated hydrogen ions might be exchanged with the copper ions of the adsorbed $CuEDTA^{2-}$ chelates. This phenomenon became more significant with a decrease in pH value, as shown in Fig. 5. For a pH value lower than 2.0, the positively-charged surface of activated carbon repelled the dominant copper species, Cu^{2+}, in the solution. This might cause the difference between the extent of adsorption of EDTA and copper in the pH range studied. The following reaction scheme is proposed.

Main Reaction at pH 8.92

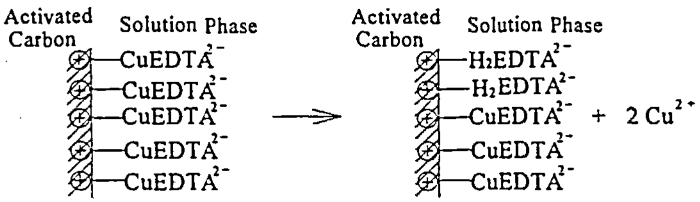


Main Reaction at pH 5.40 and pH 3.27

Early stage

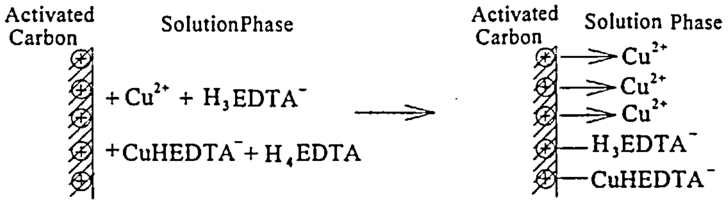


Latter stage

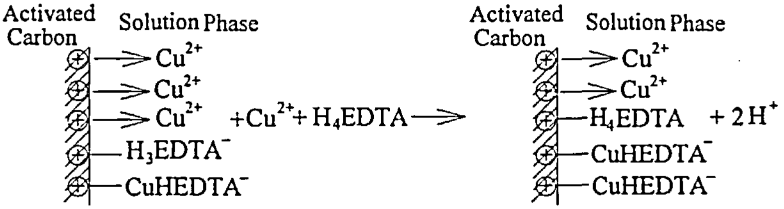


Main Reaction at pH 1.82

Early stage



Latter stage



The above scheme has not been proposed in previous batch studies. It is assumed to be related to the carbon dosage. Thus, a series of column experiments with different carbon dosages was performed, and the experimental results are shown in Fig. 6. The difference between the extent of adsorption of copper and EDTA was observed to decrease with decreasing carbon dosage.

Several further experiments were conducted to examine the extents of adsorption for Cu only, Cu–EDTA chelates, and EDTA system at pH 3.78 as summarized in Fig. 7. Comparison of the experimental results for Cu only and EDTA/Cu = 1 systems in Fig. 7 shows that adding EDTA causes an increase of copper adsorption. Conceptually, a predominant portion of the copper in a solution is assumed to exist as the negatively-charged $CuEDTA^{2-}$ in the EDTA/Cu = 1 system, which exhibits a higher affinity for the positively-charged surface of activated carbon than the positively-charged free copper species. Chang and Ku (9) reported similar findings in a batch system. However, comparing the experimental results for EDTA only and the EDTA/Cu system shows that adding Cu does not seem to result in any increase of the adsorption of EDTA. It could be visualized that the dominant species, $CuEDTA^{2-}$, in the EDTA/Cu = 1 system and H_2EDTA^{2-} in the EDTA only system exhibit a similar affinity for the positively-charged surface of activated carbon at pH 3.78.

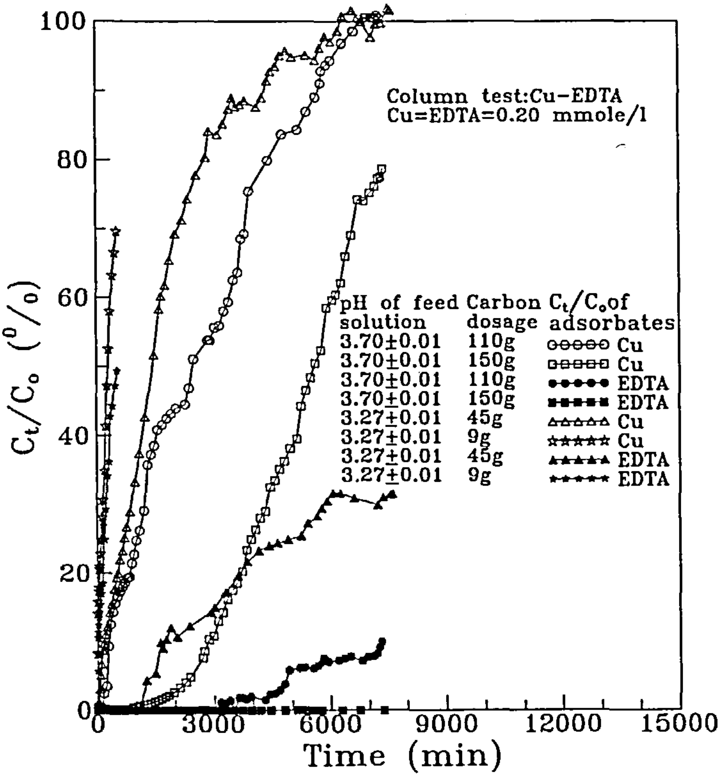


FIG. 6 Effect of carbon dosage on the difference between the extent of adsorption of copper and EDTA, flow rate = 264.7 mL/h, 25.0 ± 0.5°C.

The Bed-Depth-Service-Time (BDST) Equation

A series of kinetic column experiments was performed to obtain more practical design parameters for a scaled-up fixed-bed systems. The key task in designing adsorption columns is to accurately predict the service time of the bed before regeneration or replacement of activated carbon becomes necessary. Efforts have been made over the years by Bohart et al. (11), Erskine et al. (12), and other researchers to correlate the process variables and to develop comprehensive methods of calculating process performance. Based on the experimental results, a family of isoremoval lines was established to correlate the service times with carbon dosages for a fixed adsorption bed at various solution pH values, as shown in Fig.

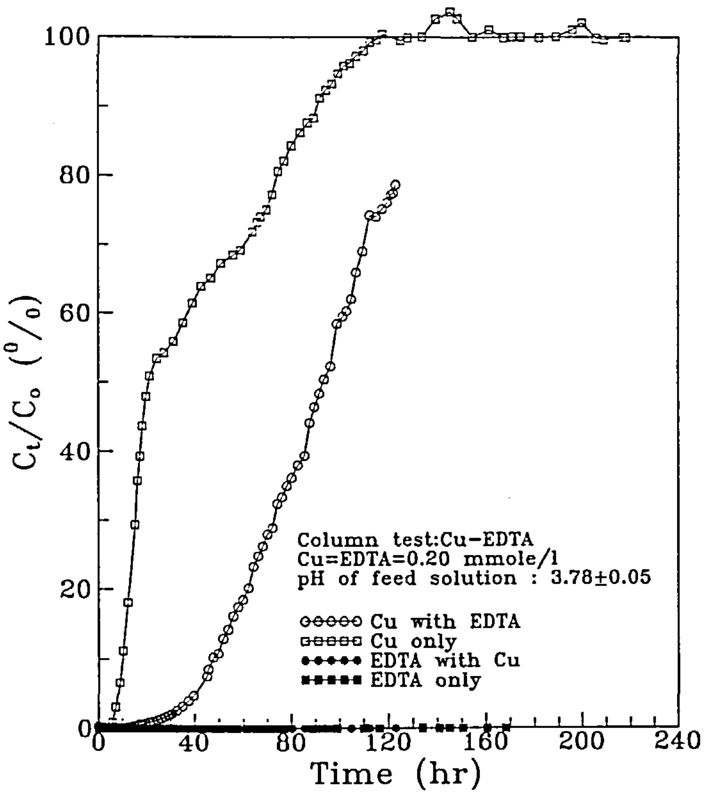


FIG. 7 Comparison of the extent of adsorption for Cu only, Cu-EDTA chelates, and EDTA systems, flow rate = 264.7 mL/h, carbon dosage = 150 g, 25.0 ± 0.5°C.

8 for pH 5.40. The points on the removal lines were obtained from a set of breakthrough curves with different carbon dosages under fixed pH values. By using these lines, the criteria for determining the service time of activated carbon could be established.

Many equations have been derived to correlate the service time of a fixed adsorption bed with operational parameters. One model, termed the Bed-Depth-Service-Time (BDST) model (13), is a modified form of an early theory from Bohart and Adams (10). It relates the service time of an adsorption bed to the height of adsorbent in the bed. Because the bed mass is directly proportional to the bed height, the bed mass was used instead of the bed height in the BDST model. The following equation was

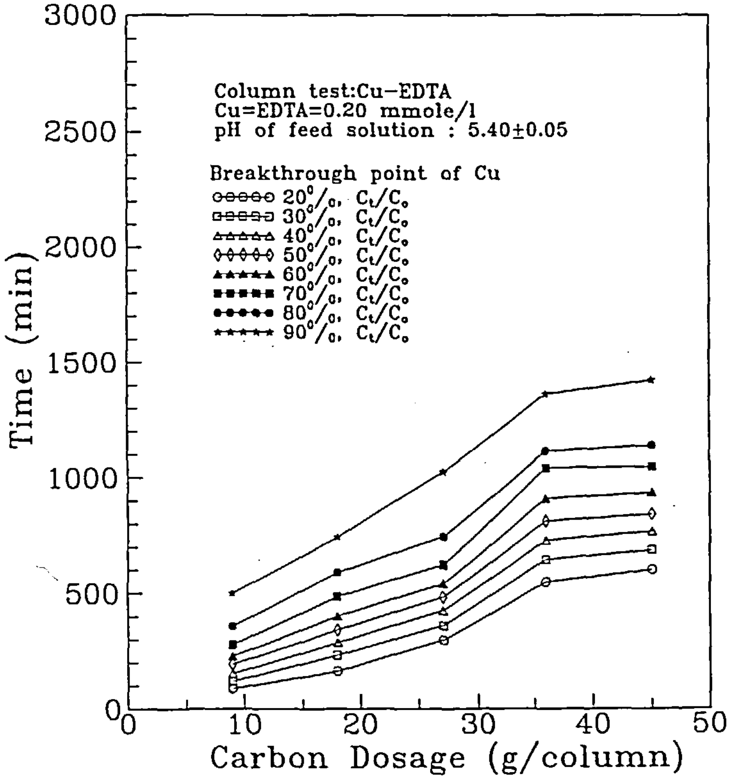


FIG. 8 Isoremoval lines for EDTA-chelated copper at different removal level, flow rate = 264.7 mL/h, 25.0 ± 0.5°C.

the result:

$$t = \frac{q_0}{C_0 V} M - \frac{1}{kC_0} \ln(C_0/C_t - 1)$$

where t = service time (minutes)
q_0 = adsorption capacity (mol/g)
C_0 = initial concentration of adsorbate (mM)
V = constant flow rate (L/min)
M = quantity of activated carbon inside column (g)
k = rate constant of adsorption (L/min/mol)
C_t = effluent concentration of adsorbate (mM)

TABLE 2

The Calculated Constant of the BDST Equation (initial concentration of Cu = EDTA = 0.2 mM; constant flow rate = 264.7 mL/h; carbon dosage = 9, 18, 27, 36, 45 g)

pH of influent	Species	Breakthrough point (%)	$k\left(\frac{\mathrm{L}}{\mathrm{min \cdot mol}}\right)$	$q_0\left[\left(\frac{\mathrm{mol}}{\mathrm{g}}\right)\cdot 10^6\right]$	Correlation coefficient, r
3.27 ± 0.05	Cu	20	50.08	9.94	0.77
		30	43.76	16.01	0.88
		40	22.88	23.30	0.96
		50	—[a]	29.8	0.99
	EDTA	20	5.53	83.48	0.91
		30	3.50	105.20	0.98
5.40 ± 0.10	Cu	20	47.53	15.40	0.96
		30	44.69	16.20	0.97
		40	35.06	17.10	0.96
		50	—[a]	18.12	0.96
		60	246.33	19.31	0.95
		70	67.34	20.62	0.94
		80	36.33	19.84	0.95
		90	35.80	23.32	0.97
	EDTA	20	32.20	20.81	0.96
		30	38.32	21.72	0.97
		40	11.14	27.40	0.97
		50	—[a]	35.72	0.98
		60	—[b]	38.93	0.96
		70	—[b]	76.92	0.99
		80	86.43	73.53	0.97
8.92 ± 0.05	Cu	20	281.88	3.13	0.97
		30	257.54	3.42	0.97
		40	678.04	3.65	0.98
		50	—[a]	3.88	0.98
		60	68.35	4.45	0.98
		70	60.90	4.96	0.95
		80	83.85	6.87	0.96
	EDTA	20	270.87	3.09	0.97
		30	251.27	3.36	0.97
		40	380.36	3.57	0.98
		50	—[a]	3.75	0.98
		60	71.66	4.05	0.98
		70	75.58	4.32	0.94
		80	85.83	5.45	0.95

[a] *k* value cannot be calculated at the 50% breakthrough point.

[b] *k* value is negative.

From the slope and the intercept of the linear plot of t versus M, the adsorption capacity (q_0) and the rate constant of adsorption (k) were calculated individually for various pH values and removals as shown in Table 2. Although the service times at various removals were not linearly correlated with the carbon dosages, the calculated constants still indicated that 1) the adsorption capacity of chelated copper at pH 3.27 was higher than that at other pH values and 2) the adsorption capacity of EDTA was higher than that of copper despite the variation of solution pH values except at pH 8.92. The BDST equation was used to fit the experimental data, and some of the fitting curves (dashed lines) shown in Fig. 9 indicate that the model data adequately described the adsorption behavior of EDTA chelated copper on activated carbon. The best-fit values of k and q_0 calcu-

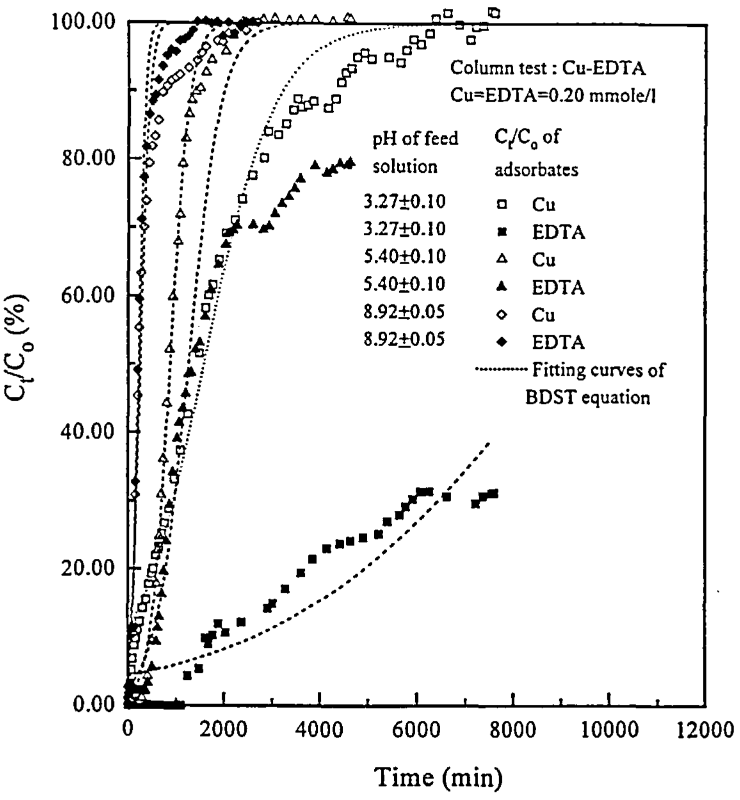


FIG. 9 Direct fitting curves of BDST equation to the breakthrough data of Cu and EDTA, flow rate = 264.7 mL/h, carbon dosage = 45 g, 25.0 ± 0.5°C.

lated from the fitting lines of BDST model are shown in Table 3. By reviewing the scattered parameters in Table 2, it was found that the k and q_0 values shown in Table 3 are similar to the data shown in Table 2 under the condition of lower pH values at the initial stage of an experiment. Hence, it is possible to roughly estimate the exhaust time of a bed with different carbon dosages at a fixed pH value by using the constants k and q_0 obtained from only one set of column test data under certain carbon dosage and pH values. It is also observed in Fig. 9 that for a few cases the modeling results deviate after the experiments have progressed for certain periods, especially for those species which have larger extent of adsorption on activated carbon in the final experimental period. It was realized that one of the basic assumptions for applying the BDST equation is that the reaction should be considered to be monomolecular with respect to the adsorbate, whereas in practice this is not usually the case. Another assumption inherent in the BDST equation is that the rate of adsorption by activated carbon is perfectly uniform, but in this research the adsorbed charged chelated ions seriously affect the surface charges of activated carbon during the adsorption process, and therefore the rate at which the activated carbon surface adsorbs adsorbates varies during the adsorption process. The difference between the assumptions of the BDST equation and this research work might cause deviation of the fitting curves of the

TABLE 3
The Best-Fit Constant of the BDST Equation (initial concentration of Cu = EDTA = 0.2 mM; constant flow rate = 264.7 mL/h)

pH of influent	Species	$k\left[\frac{\text{L}}{\text{min}\cdot\text{mol}}\right]$	$q_0\left[\left(\frac{\text{mol}}{\text{g}}\right)\cdot 10^6\right]$	Carbon dosage (g/column)	χ^2
3.27 ± 0.05	Cu	6.74	30.86	45	22.31
		26.33	31.6	9	8.77
	EDTA	1.78	172.2	45	23.75
		20.88	47.21	9	12.33
5.40 ± 0.10	Cu	25.73	17.07	45	6.01
		36.39	17.80	18	6.88
	EDTA	14.66	24.61	45	12.67
		13.35	32.07	18	92.56
8.92 ± 0.05	Cu	53.09	5.00	45	58.29
		72.16	4.83	9	43.80
	EDTA	73.19	4.37	45	29.32
		90.40	4.02	9	43.97

BDST equation from this experimental work. Hence, further study of the modeling equation is necessary.

CONCLUSION

The adsorption of chelated copper on activated carbon was found to vary over the entire solution pH range, possibly because the electrostatic interactions between the activated carbon surface and the dominant species is highly pH dependent. Based on the column adsorption characteristics of both copper and EDTA species, it is further presumed that the concentrated hydrogen ion on the carbon surface might be exchanged with the copper ion of the adsorbed chelate, $CuEDTA^{2-}$, in acidic solutions. An interesting phenomenon observed was that the adsorption behaviors of copper and EDTA were significantly different for higher carbon dosages. The BDST design model was used and could adequately describe the adsorption behavior of chelated copper species. Although experimental results indicated that further improvement of the BDST equation is necessary, this model offers an alternative method to estimate the exhaust time of activated carbon.

REFERENCES

1. C. P. Huang and F. B. Ostovic, "Removal of Cadmium(II) by Activated Carbon Adsorption," *J. Environ. Eng. Div. Proc., ASCE*, *104*(EE5), 863–875 (1978).
2. H. A. Elliott and C. P. Huang, "The Adsorption Characteristics of Cu(II) in the Presence of Chelating Agents," *J. Colloid Interface Sci.*, *70*(1), 29–45 (1979).
3. C. P. Huang, M. H. Tsang, and Y. H. Hsieh, "The Removal of Cobalt(II) from Water by Activated Carbon Adsorption," *AIChE Symp. Ser.*, *81*(243), 85–98 (1985).
4. O. M. Corapcioglu and C. P. Huang, "Adsorption of Heavy Metals onto Hydrous Activated Carbon," *Water Res.*, *21*(9), 1031–1044 (1987).
5. M. M. Jevtitch and D. Bhattatcharyya, "Separation of Heavy Metal Chelates by Activated Carbon: Effect of Surface and Species Charge," *Chem. Eng. Commun.*, *23*, 191–213 (1983).
6. D. Bhattacharyya and C. Y. Cheng, "Activated Carbon Adsorption of Heavy Metal Chelates from Single and Multicomponent Systems," *Environ. Prog.*, *6*(2), 110–117 (1987).
7. M. M. Benjamin and J. O. Leckie, "Effects of Complexation by Cl^-, SO_4^{2-} and $S_2O_3^{2-}$ on Adsorption Behavior of Cd on Oxide Species," *Environ. Sci. Technol.*, *16*(3), 162–170 (1982).
8. R. W. Peters and L. Shem, "Adsorption/Desorption Characteristics of Lead on Various Types of Soil," *Environ. Prog.*, *11*(3), 234–240 (1992).
9. C. Chang and Y. Ku, "The Adsorption and Desorption Characteristics of EDTA-Chelated Copper Ion by Activated Carbon," *Sep. Sci. Technol.*, *30*(6), 899–915 (1995).
10. Norit Co. Ltd., Data sheet of NORIT PK 3-5, Tokyo, 1988.

11. G. S. Bohart and E. Q. Adams, "Some Aspects of the Behavior of Charcoal with Respect to Chlorine," *J. Am. Chem. Soc.*, *42*, 523–544 (1920).
12. D. B. Erskine and W. G. Schuliger, "Activated Carbon Processes for Liquids," *Chem. Eng. Prog.*, *67*(11), 41–44 (1971).
13. R. A. Hutchins, "New Method Simplifies Design of Activated-Carbon Systems," *Chem. Eng.*, *80*(19), 133–138 (1973).

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