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Liquid-Phase Adsorption of Phenol and Its Derivatives on Activated Carbon Fibers

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ABSTRACT

The adsorption of three phenolic compounds from aqueous solutions on activated carbon fibers was measured in the 2–5 mol/m^3 concentration range at 303 K. High adsorption capacities were obtained for the substituted phenols. Several isotherm equations were tried in order to find an equation which correlates the equilibrium data best. Among the equations tried, the three-parameter equation of Jossens et al. derived thermodynamically and based on a heterogeneous surface adsorption theory was found to be the most satisfactory over the entire range of concentrations. A rate parameter was obtained and used to describe the adsorption process on a quantitative basis. This rate parameter could be successfully correlated against the initial solute concentration.

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INTRODUCTION

Phenolic compounds widely exist in industrial effluents such as those from oil refineries and the coal tar, plastics, leather, and steel industries (1). Because they are highly toxic and are, in general, not amenable to biological degradation, new methods of treatment are continuously being modified and developed. Possible methods include chemical oxidation, coagulation, solvent extraction, liquid membrane permeation, and adsorption. Of all the methods, adsorption appears to offer the best prospects for overall treatment, especially for effluents with moderate and low concentrations. Because granular or powdered activated carbon (GAC or PAC) has a good capacity for the adsorption of organic compounds, it has become the most widely used adsorbent for this purpose (2–8). However, this process requires a container, and careful filling is generally necessary; replacement of the adsorbent is by no means a simple procedure. Also, the use of PAC for liquid solutions involves subsequent filtration.

Activated carbon fiber (ACF) used in this study can obviate these disadvantages (9, 10). In fact, ACF has been extensively used for the adsorption of gaseous species such as acetone, benzene, isopropanol, water vapor, hexane, chloroform, carbon tetrachloride, dichloroethane, ethanol, N_2, CO_2, HCN, NH_3, NO, NO_2, and SO_2 (9–11), as well as of liquid species such as fatty acids, benzene, phenol, benzoic acid (22), tri- and tetrachloroethylene, organic and inorganic phosphates, Cr(VI) and Hg(II) ions, and iodine (10, 12, 13).

For many adsorption applications, ACF provides greater ease of use than GAC because ACF is not granular but can be manufactured in the form of a cloth. The surfaces of ACF can be regenerated in-situ with electrical resistance heating and can be chemically tailored to allow preferential adsorption of certain compounds. The pore sizes of ACF typically lie over a narrow range in the micropore region (pore diameter < 2 nm), which allows use of ACF as a molecular sieve in some applications (9). In contrast to GAC, moreover, ACF typically exhibits higher adsorption capacities due to larger surface area per unit volume and higher adsorption rates due to a shorter diffusion path (9, 11). The faster kinetics with ACF is expected to be more beneficial to liquid-phase adsorption in comparison to gas-phase adsorption, since the effect of diffusion is more remarkable in liquid-phase adsorption.

Although ACF adsorption of a species from aqueous solution has been widely investigated as stated above, to our knowledge systematic studies on the adsorption of phenol and its derivatives have rarely been made.

In the present work the equilibrium and kinetics for the batch adsorption of phenol, 4-chlorophenol, and 4-nitrophenol on ACF were measured at 303 K. Various two- and three-parameter isotherm equations were tested in order to find an equation which correlates the equilibrium data best. The rate parameters for these adsorption processes were also obtained and are discussed.

EXPERIMENTAL

Reagents, ACF, and Solutions

All phenolic compounds used in this study, including phenol, 4-chlorophenol (4-CP), and 4-nitrophenol (4-NP), were offered by Merck Co. as analytical grade. They were used without further purification. The ACF, traded as Charsob CP-1300, was supplied by Yih Hsin Technical Co., Taipei, Taiwan, and was used as received. It was manufactured by carbonizing polyacrylonitrile at 200–300°C in air and activating it at 800–1200°C. Its physical properties are listed in Table 1. The aqueous solutions were prepared by diluting different phenolic compounds with deionized water to the required concentrations (2–5 mol/m^3). The deionized water employed in this work was produced by a Millipore Milli-Q Water System.

Procedure

In adsorption equilibrium runs, a fixed amount of ACF (0.1–0.2 g) and 100 cm^3 of an aqueous solution were placed in a 250-cm^3 glass-stoppered flask and shaken at 130 rpm for 3 days using a thermostated shaker bath (Firstek Model B603, Taiwan). Preliminary experiments had shown that the adsorption studied was complete after 1 day. After equilibrium, the concentrations of phenolic compounds in the aqueous phase were ana-

TABLE 1
Physical Properties of the Investigated ACF

Property	Value
Material	Polyacrylonitrile
Thickness (mm)	0.61
Weight (g/m^2)	80
Mean micropore volume (cm^3/g ACF)	0.586
Micropore radius (nm)	0.8–1.0
BET specific surface area (m^3/g)	1380

lyzed using a Hitachi UV/visible spectrophotometer (Model U-2000) at each suitable wavelength. Each experiment was duplicated under identical conditions.

The contact-time experiments were made in a batch mode using several 250-cm^3 Erlenmeyer flasks. In each flask, a slice of ACF (0.1 g) of 18 × 18 mm was placed into which an aqueous solution (100 cm^3) was then poured. The timing was started upon the addition of the aqueous solution. The solution was agitated by using a magnetic stirrer at a speed of 190 rpm, since after which the stirring speed has no effect on the adsorption process. At each preset time interval, mixing in one flask was stopped and a sample was immediately taken. The concentrations of phenol, 4-CP, and 4-NP in the aqueous solutions were similarly analyzed as described above.

RESULTS AND DISCUSSION

Equilibrium Adsorption

Figure 1 shows the equilibrium adsorption; that is, the relationships between the amount of phenolic compound adsorbed per unit mass of ACF (q_e) and its remaining concentration in the aqueous solutions (C_e). The adsorption capacity, defined as the amount adsorbed at the plateau of the isotherm, has the following order: 4-CP > 4-NP > phenol, i.e., high capacities are obtained for the adsorption of the substituted phenols. Streat et al. (7) also found that the capacity is higher for 4-CP than for phenol when using GAC, wood, shell, rubber, and coal as adsorbents.

It is generally accepted that the capacity for GAC adsorption is higher when the adsorbate has a high molar mass or a smaller solubility in water (2, 3, 7). This is not the case here since the molar mass (g/mol) and the water solubility (mol/dm^3) are reported to be 94.1 and 0.9 for phenol, 128.6 and 0.2 for 4-CP, and 139.1 and 0.1 for 4-NP, respectively (2). On the other hand, Mattson et al. (8) indicated that the adsorption capacity of phenols on activated carbon is affected by hydrogen bonding between adsorbates and adsorbents. There are many conjugated π-bonds on the ACF surface, and they are nonlocalized and highly active. After activation, a number of oxygen-containing functional groups exist in the crystal lattice of the surface, including electron-acceptor (e.g., carbonyl and carboxyl) and electron-donor (e.g., hydroxyl) groups. It is hence probable that hydrogen bonds are formed between the electron-acceptor groups on the ACF surface and the hydrogen atoms on phenolic compounds (8). Judging from the electrophilic nature of the investigated phenols, the ca-

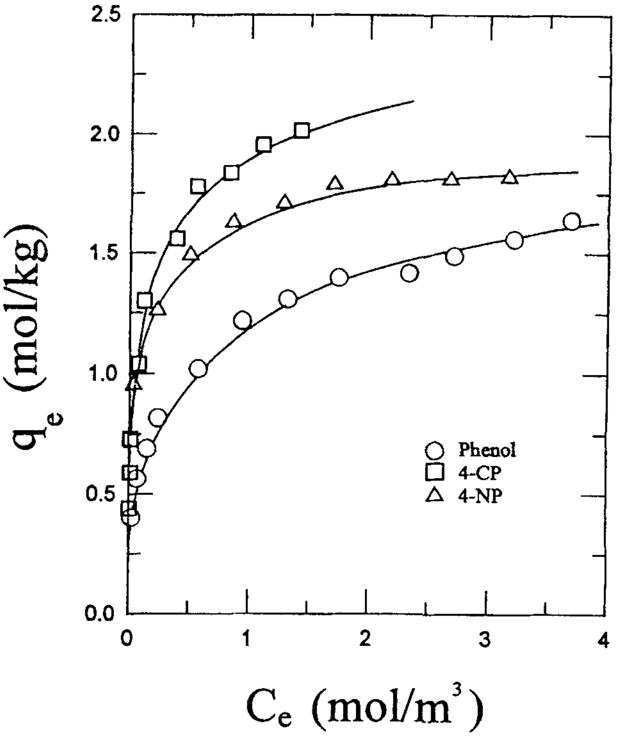

FIG. 1 Equilibrium adsorptions of phenol, 4-CP, and 4-NP on ACF at 303 K.

pacity would likely decrease in the order 4-NP > 4-CP > phenol. This is in some disagreement with the experimental findings. The present ordering of adsorption capacity is somewhat different from what has been observed using other adsorbents. For example, Caturla et al. (2) found the ordering for GAC adsorption to be phenol > 4-CP > 4-NP, as expected, opposite to that of the molecular volume of phenols. Jossens et al. (5) found the following order: phenol > 4-NP > 4-CP for their adsorption on GAC. Itaya et al. (14) observed that the ordering is 4-CP > phenol > *p*-cresol on Amberlite XAD-4 macroreticular resin and *p*-cresol > 4-CP > phenol on XAD-7 resins.

Such discrepancies are probably due to the differences in pore structure among ACF and other adsorbents. GAC is essentially characterized by a broad pore size distribution containing micro- (<2 nm), meso- (2–50 nm), and macropores (> 250 nm). It is believed that the major adsorption sites on GAC are located in micropores rather than in mesopores where only weaker adsorption is observed (13). The adsorption on GAC must proceed through a sequence of diffusion steps from the bulk phase into mesopores and then to micropores. Thus, the adsorption capacity would be lower for an adsorbate with a larger molecular volume (2). However, for ACF the micropores with a depth less than the radius of a fiber (about 6 μm) are directly exposed on the external surface (12, 13). It is thus deduced that micropore size and its distribution may play an important role in the adsorption of phenols (11). Perhaps the effect of hydrophobic interaction between polyacrylonitrile fibers and adsorbates should not be ignored (14).

Isotherm Equations

Basically, adsorption isotherms are important to describe how adsorbates will interact with ACF and so are critical in optimizing the use of ACF as an adsorbent. In this regard, the correlation of equilibrium data using either a theoretical or empirical equation is essential to practical adsorption operation. Four two- and three-parameter isotherm equations are tested in the present work.

(1) Langmuir Equation

$$C_e/q_e = (1/K_L q_{mon}) + (1/q_{mon})C_e \tag{1}$$

where q_{mon} denotes the amount adsorbed per unit mass of adsorbent corresponding to complete monolayer coverage. K_L is the Langmuir constant.

(2) Freundlich Equation

$$\ln q_e = \ln K_F + (1/n) \ln C_e \tag{2}$$

where n and K_F are constants for a given adsorbate–adsorbent system.

(3) Redlich–Peterson Equation (15)

$$q_e = a_R C_e/(1 + b_R C_e^{\beta}) \tag{3}$$

where β lies between 0 and 1. This equation gave a good fit for the adsorption of phenol from dilute aqueous solution on Amberlite XAD-8 (16) as well as on XAD-4 and XAD-7 resins (14). In a slightly different form, it was used by Radke and Prausnitz (6).

(4) Equation of Jossens et al. (5)

For a highly heterogeneous surface, the highest energy sites are filled first so that the heat of adsorption declines rapidly with increased surface coverage. In this respect, the following isotherm equation is derived on a thermodynamic basis (5).

$$C_e = (q_e/H)\exp(K_J q_e^p) \qquad (4)$$

where H corresponds to Henry's law constant and K_J is a function of temperature only. The value of p is characteristic of the adsorbent only, irrespective of temperature and the nature of adsorbates. For example, $p = 0$ corresponds to an uniform surface for which all sites have the same energy. A good representation of equilibrium data using this equation was reported for the adsorption of phenolic compounds from aqueous solutions on GAC (5) and on Amberlite XAD-4 and XAD-7 resins (14).

The two parameters in the Langmuir and Freundlich equations can be graphically determined. On the other hand, the three parameters in the Redlich–Peterson equation and the equation of Jonnens et al. are obtained by using a least-squares fitting procedure to minimize the deviation between calculated and experimental equilibrium data. In order to compare the validity of the isotherm equations more definitely, a normalized percent deviation, Δq, is calculated and defined as follows:

$$\Delta q\ (\%) = (100/N_l)\sum_{i=1}^{N_l}\{|q_{e,i}^{\text{expt}} - q_{e,i}^{\text{calc}}|/q_{e,i}^{\text{expt}}\} \qquad (5)$$

where the superscripts "calc" and "expt" are the calculated and experimental values, respectively, and N_l is the number of measurements. The parameters in all isotherm equations obtained for the investigated systems, together with Δq, are listed in Table 2.

It follows from Table 2 that Δq for the Langmuir equation is very large (7.19–16.4%). It is well known that the Langmuir equation is intended for a homogeneous surface, although it obeys Henry's law at extremely low concentrations. Therefore, the poor fit of the adsorption data considered here can probably be attributed to the nature of the highly heterogeneous surface of ACF (11).

The Freundlich fit is somewhat better (Δq = 2.62–6.08%) than the Langmuir fit. In fact, the Freundlich equation, which is suitable for a

TABLE 2
Parameters and Normalized Percent Deviations Obtained Using Various Isotherm Equations

No.	Isotherm	Parameter	Phenol	4-CP	4-NP
1.	Langmuir	q_{mon} (mol/kg)	1.65	2.05	1.89
		K_L ($\mathrm{m^3/mol}$)	4.00	16.7	10.2
		Δq (%)	12.9	16.4	7.19
2.	Freundlich	n	3.64	3.67	6.68
		K_F ($\mathrm{mol}^{n+1}/\mathrm{kg \cdot m}^{3n}$)	1.17	2.00	1.62
		Δq (%)	3.25	6.08	2.62
3.	Redlich–Peterson	β	0.779	0.843	0.950
		a_R ($\mathrm{m^3/kg}$)	55.8	115.2	34.8
		b_R ($\mathrm{m^3/mol})^{\beta}$	45.8	59.3	19.7
		Δq (%)	2.78	3.53	5.18
4.	Jonnens	p	0.977	0.977	0.977
		H ($\mathrm{m^3/kg}$)	41.3	214.4	1365
		K_J ($\mathrm{kg/mol})^{p}$	2.90	2.49	4.16
		Δq (%)	2.06	2.96	2.10

highly heterogeneous surface, often gives a good representation of typical adsorption data over a restricted range of concentrations (for example, the use of piecewise fitting) (4, 8). It does not reduce to Henry's law at concentrations approaching zero. It often fits the adsorption data at high and moderate concentrations, and it provides a poor fit for adsorption data at low concentrations (5).

The value of β in the Redlich–Peterson equation ranges from 0.779 to 0.910, which lies exactly between 0 and 1. It should be noted that the Redlich–Peterson equation (Δq = 2.78–5.18%) does not give any improvement for the representation of adsorption data compared to the Freundlich equation. In the case of the equation of Jossens et al., p is found to be 0.977 ± 0.062. It is now found that Δq is always less than 2.96%. That is, this equation gives a best fit of isotherm data over the entire range of concentrations for all systems.

As shown in Table 2, H increases in the order phenol < 4-CP < 4-NP. Indeed, Jossens et al. (5) found that the values of H have the following order: 4-CP < phenol < 4-NP for GAC adsorption. It is worth noting that in their work the values of p obtained using identical GAC are not constant, ranging from 0.25 for phenol to 0.80 for 4-CP. On the other hand, the phenol < *p*-cresol < 4-CP ordering for *H* was observed for their adsorption on Amberlite XAD-4 and XAD-7 resins (14). This is opposite to both the observations of Jossens et al. and the solubility of the adsorbates in water (14). The explanation is that hydrophobic interaction between adsorbates and macroreticular resins plays an important role in adsorption onto macroreticular resins.

Adsorption Rates

Contact-time experiments can be used to establish the time dependence of the system. Figures 2–4 show the measured results. As expected, for a given amount of ACF the dimensionless aqueous concentrations decrease

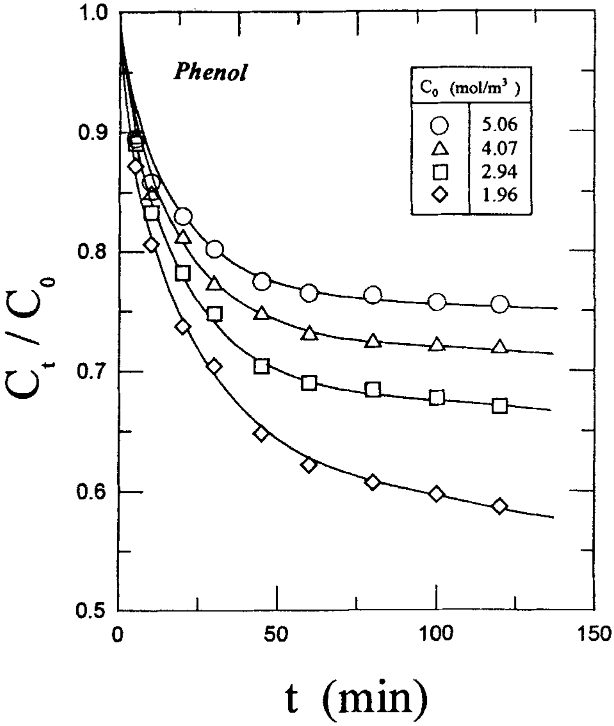

FIG. 2 Rate of phenol adsorption on ACF at different initial concentrations.

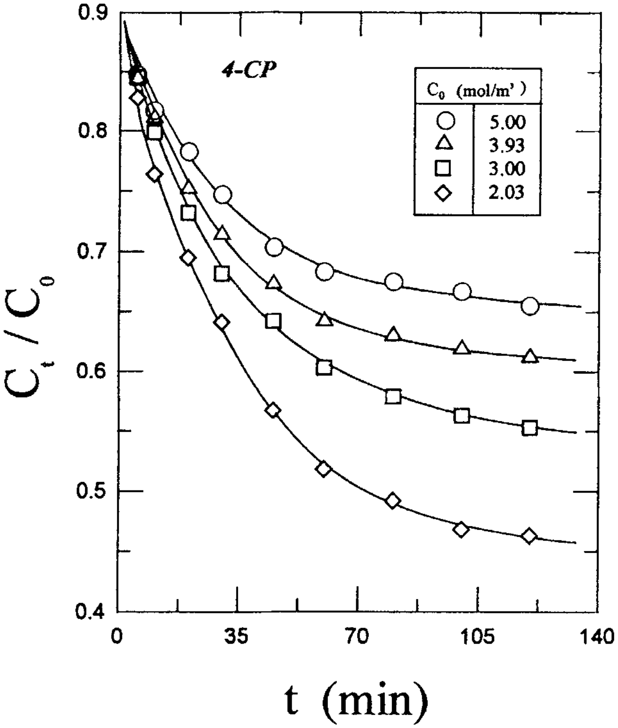

FIG. 3 Rate of 4-CP adsorption on ACF at different initial concentrations.

more quickly at lower initial concentrations. It is also seen that the adsorption rate decreases in the order 4-NP > phenol > 4-CP under comparable conditions.

In general, the adsorption mechanism can be determined by plotting the fraction of adsorption Y against the normal function of time $t^{1/2}$ (17–23), where $Y = (q_t/q_e) = (C_0 - C_t)/(C_0 - C_e)$. Figures 5–7 show the results. It is evident that these plots have the same general shape, i.e., three-stage linear sections, but the position of each transition depends on the initial concentration and the type of solutes. The first, sharper portion is the external surface adsorption stage (21) or instantaneous adsorption stage (23). The second, gradual linear portion is the gradual ad-

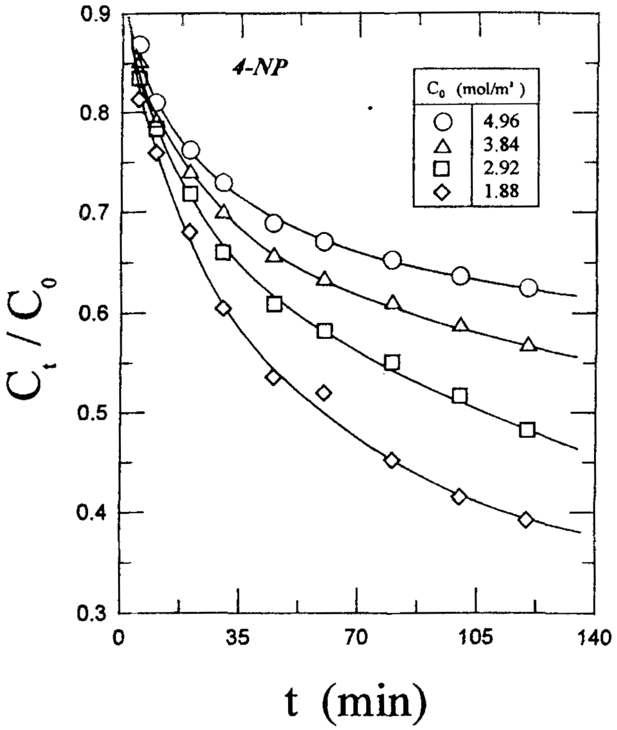

FIG. 4 Rate of 4-NP adsorption on ACF at different initial concentrations.

sorption stage (23). The third portion is the final equilibrium stage (22, 23).

Although ACF used in this study was in the form of a slice, from a microscopic viewpoint it may be considered to be a collection of many microcrystallines with a graphite random structure (10). In this model the microcrystalline particles are considered to be surrounded by a boundary layer film through which the solutes must diffuse prior to external adsorption on the ACF surface, and this step is responsible for the first section in Figs. 5–7. The second section of the plots, between about 30–70% solute adsorption, is due to intraparticle diffusion as the predominant rate-controlling step. As the bulk solute concentration and the surface solute

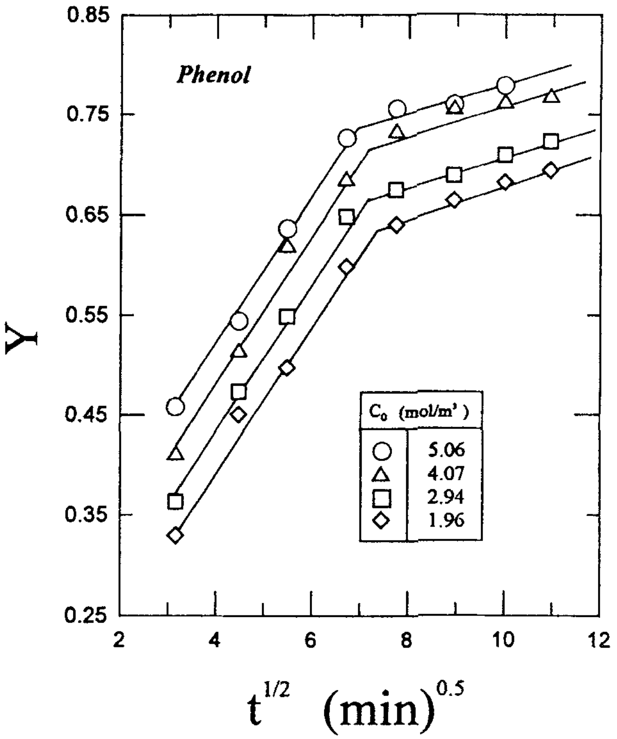

FIG. 5 Time dependence of the fraction of phenol adsorption on ACF.

concentration start to decrease, the third section of the figures is attributed to a decrease in the rate of diffusion. In fact, Alexander et al. (17) studied the adsorption of dyes on silica and also found three stages in equivalent plots. However, the shape is different; that is, an initial curved portion, a linear section, and a final curved portion.

The intercepts of the second and third stages in Figs. 5–7 can be adopted to calculate the rate parameters k_2 and k_3, respectively, as was also done earlier (22). They are expressed as

$$Y_j = a_j t^{1/2} + k_j \qquad (j = 2, 3) \tag{6}$$

The value of k_j can be linearly related to initial solute concentration (Fig. 8), indicating that increasing C_0 increases the driving force of phenols

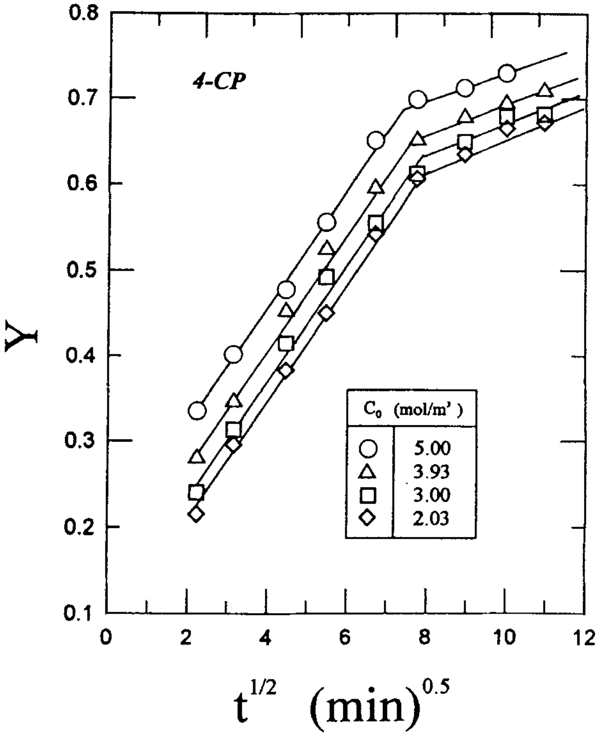

FIG. 6 Time dependence of the fraction of 4-CP adsorption on ACF.

from the bulk on and into the solid fibers (22). Consequently, we have

$$k_j = b_j C_0 + d_j \qquad (j = 2, 3) \tag{7}$$

The values of a_j, b_j, and d_j obtained are listed in Table 3 for comparison, together with the normalized percent deviations, defined as

$$\Delta Y(\%) = (100/N_2) \sum_{i=1}^{N_2} \{|Y_i^{\text{expt}} - Y_i^{\text{calc}}|/Y_i^{\text{expt}}\} \tag{8}$$

The agreement between the measured and calculated results is sound. It follows from Table 3 that the value of b_j, a measure of the adsorption rate related to the effect of C_0, decreases in the order 4-NP > phenol > 4-CP, as indicated in Figs. 2–4.

In practice, the linear plot of Y vs $t^{1/2}$ has been reported by Ruthven (24) on a theoretical basis. For the batch adsorption of solutes on spherical

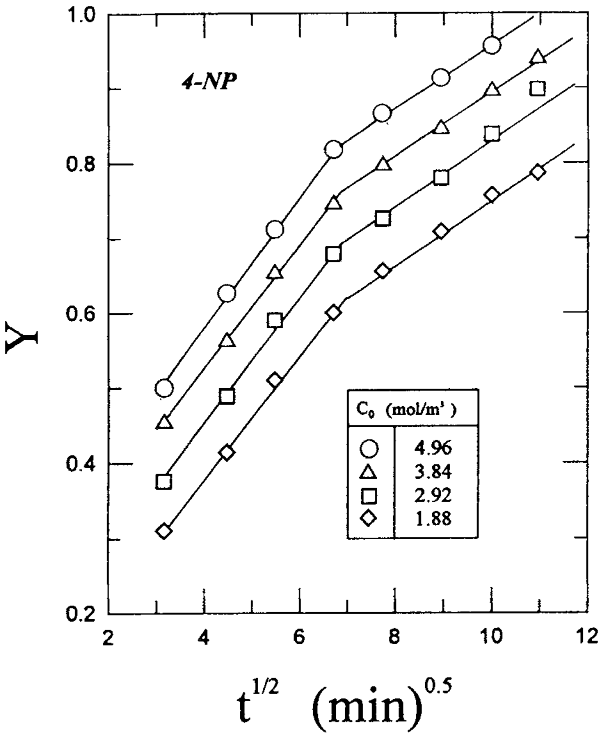

FIG. 7 Time dependence of the fraction of 4-NP adsorption on ACF.

TABLE 3
Parameters in Eqs. (6) and (7) and the Normalized Percent Deviations

	Second stage				Third stage			
Solute	a_2	b_2	d_2	ΔY (%)	a_3	b_3	d_3	ΔY (%)
Phenol	0.075	0.040	0.022	1.47	0.014	0.035	0.471	0.99
4-CP	0.073	0.037	−0.022	1.83	0.016	0.028	0.427	1.12
4-NP	0.086	0.066	−0.085	0.40	0.046	0.066	0.177	1.33

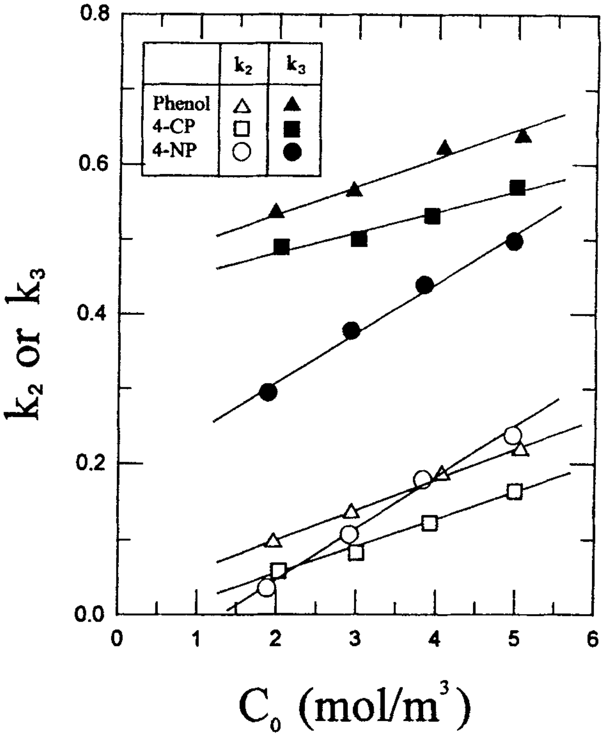

FIG. 8 Relationship between rate parameters of the second and third stages and initial solute concentrations.

particles, he indicated that for $Y < 0.3$ the solution of diffusion equations based on Fick's second law can be reduced to

$$Y \approx (2A/V)(D_p t/\pi)^{1/2} \tag{9}$$

where A/V is the ratio of external area to particle volume and D_p is the intraparticle diffusivity. In the case of a high fraction of adsorption, however, the linear relationship is merely an empirical observations at this stage (22, 23).

CONCLUSIONS

Equilibrium and kinetic studies on the adsorption of phenol, 4-chlorophenol (4-CP), and 4-nitrophenol (4-NP) from aqueous solutions using

ACF have been carried out in the concentration range of 2–5 mol/m^3 at 303 K. The adsorption capacity is found to decrease in the order 4-CP > 4-NP > phenol. The Langmuir equation gives the poorest overall fit due to its homogeneous monolayer nature. The Freundlich equation yields a somewhat better fit, but still deviates considerably for 4-CP. The normalized percent deviation (Δq) obtained by the Redlich–Peterson equation ranges from 2.78 to 5.18%. Of the equations tested, the equation of Jossens et al. gives the best fit over the entire range of concentrations and yields Δq less than 2.96%. Plots of the fraction of adsorption against $t^{1/2}$ give three linear sections, which represent the instantaneous adsorption stage, the gradual adsorption stage, and the final equilibrium stage in sequence. It is shown that the rate parameter, and hence the fraction of adsorption obtained here, can be satisfactorily related to the initial solute concentration and contact time.

NOTATIONS

a_R	parameter in Eq. (3) (m^3/kg)
b_R	parameter in Eq. (3) (m^3/mol)$^\beta$
C_e	equilibrium solute concentration in the aqueous solution (mol/m^3)
C_t	solute concentration in the aqueous solution at time t (mol/m^3)
C_0	initial solute concentration in the aqueous solution (mol/m^3)
H	Henry's law constant in Eq. (4) (m^3/kg)
k_j	rate parameter defined in Eq. (6) (—)
K_F	parameter in Eq. (2) (mol^{n+1}/kg·m^{3n})
K_J	parameter in Eq. (4) (kg/mol)^p
K_L	parameter in Eq. (1) (m^3/mol)
n	parameter in Eq. (2) (—)
p	parameter related to the distribution of energy sites on the surface in Eq. (4) (—)
q_e	equilibrium amount adsorbed on ACF (mol/kg)
q_{mon}	equilibrium amount of monolayer adsorption defined in Eq. (1) (mol/kg)
q_t	amount adsorbed at time t (mol/kg)
Y	fraction of solute adsorption (—)

Greek Letter

β	parameter in Eq. (3) (—)

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