Bull. Environ. Contam. Toxicol. (1990) 45:170-176

Bulletin of Environmental Contamination and Toxicology

Adsorption Removal of Cresol by Granular Activated Carbon for Medical Waste Water Treatment

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In medical facilities, disinfectants were frequently used for handwashing and disinfection of instruments, and operating rooms. Waste water from medical facilities contain large quantities of disinfectants. In general, the activated sludge method has been used in waste water treatment; however, its drawback is that a constituent of disinfectants hinder the purification abilities of aerobic bacteria (Kiba et al 1972). Therefore, another treatment method is desirable. It is effective to use a solid adsorbent such as activated carbon for removing organic substances in waste water (Urano 1975).

Little work on the advanced treatment of waste water containing disinfectants has been done so far. In this work, we discussed the characteristics of adsorption removal of cresol, one of the disinfectants, onto granular activated carbon for medical waste water treatment.

MATERIALS AND METHODS

Saponated cresol solution was of the Japanese pharmacopoeia grade (Yamazen Chemical Co., Ltd.) and the concentration of cresol was indicated as 50%. Activated carbons used were commercial products, and its particle size was 4-8 mesh. Before use, the activated carbon was washed by distilled water until no fine carbon particle was observed. It was dried at 110 °C for 48 hrs, then kept in a desiccator. The physicochemical properties of activated carbon are shown in Table 1.

Adsorption equilibrium isotherms were obtained for the cresol-water/adsorbent system. It took 8 days to attain equilibrium adsorption at 25°C with constant shaking. After equilibration, the cresol concentration in the supernatant solution was measured at 275 nm with a spectrophotometer (Hitachi model 101).

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Table 1. Properties of Activated Carbon Used.

Adsorbent No.	Surface Area (m^2/g)	Pore Volume (mL/g)			pH
		$r<20$ Å	$20<r<100$ Å	$r<100$ Å	
1	990.91	0.491	0.066	0.557	6.82
2	876.84	0.418	0.064	0.482	10.28
3	979.76	0.408	0.178	0.586	3.06
4	931.08	0.484	0.108	0.592	8.82
5	674.11	0.350	0.063	0.413	10.20
6	1145.97	0.554	0.069	0.623	7.13
7	692.18	0.378	0.076	0.424	10.19
8	1220.54	0.584	0.087	0.671	6.41
9	813.69	0.420	0.065	0.485	6.37

Adsorption rate was measured as follows; four grams of activated carbon were placed in a flask containing distilled water, and the suspension was boiled for 1 hr to displace the air in the pores of the activated carbon with water. The activated carbon impregnated with water was placed in 800 mL of a stirred cresol solution (ca. 100 ppm). A solution of 20 mL was taken from the suspension at regular intervals, and immediately an adsorbate solution with a concentration of ca. 1000 ppm and distilled water were added into the suspension until its concentration and its volume became 100 ppm and 800 mL, respectively.

RESULTS AND DISCUSSION

The adsorption isotherms of cresol onto activated carbon at 25°C were of the Freundlich type. In general, the Freundlich equation can be applied to the adsorption of organic compounds onto activated carbon in a liquid phase. The greater the k value, the more adsorbate is adsorbed. It is easy for adsorbate to be adsorbed when the $1/n$ value is in the range of 0.1 to 0.5 (Hassler 1963). Table 2 shows the Freundlich constants and amount adsorbed at each equilibrium concentration. The amount adsorbed was determined by isotherm experiments. The k value of activated carbon No.3 was about 2.9 times greater than that of No.7. All $1/n$ values of cresol onto activated carbon were in range of 0.152 to 0.320. The amount adsorbed at each equilibrium concentration (10, 100 and 1000 ppm) of No.6 indicated the greatest value, and Nos.1, 3 and 5 indicated great amount of cresol adsorbed. The weight of activated carbon for 90 % removal of cresol is shown in Table 3. In the initial concentration of 1000 mg/L, the weight of activated carbon was large for No.7 and small for Nos.1,3 and 5. When the initial concentration was 10 mg/L, the weight of No.3 for 90% removal was less than

Table 2. Freundlich Constants and Equilibrium Amount of Cresol Adsorbed.

Adsorbent No.	Freundlich Constant		Amount Adsorbed (mg/g)		
	1/n	k (mg/g)	Q_∞ at 10ppm	Q_∞ at 100ppm	Q_∞ at 1000ppm
1	0.222	51.34	85.63	142.82	238.21
2	0.320	25.99	54.33	113.57	237.40
3	0.152	66.72	94.67	134.35	190.65
4	0.247	39.94	70.47	124.33	219.37
5	0.243	46.50	81.34	142.28	248.90
6	0.262	52.18	95.28	173.99	317.73
7	0.306	22.90	46.32	93.73	189.64
8	0.260	38.06	69.33	126.28	230.00
9	0.213	46.16	75.45	123.34	201.63

Table 3. Weight of Activated Carbon for 90% Removal for Cresol.

Adsorbent No.	Weight of Activated Carbon (g)		
	1000ppm——100ppm	——10ppm	——1ppm
1	6.302	1.051	0.175
2	7.925	1.657	0.346
3	6.699	0.951	0.135
4	7.239	1.277	0.225
5	6.325	1.107	0.194
6	5.173	0.945	0.172
7	9.602	1.943	0.393
8	7.127	1.298	0.236
9	7.297	1.193	0.195

about 1/3 as much as that of No.7. From the standpoint of amount adsorbed, activated carbon No.3 and No.6 were the preferable adsorbent for cresol adsorption in the range of lower equilibrium concentrations of 1 ppm or less and higher equilibrium concentrations of 10 ppm, respectively.

In order to elucidate which properties of adsorbent dominated the amount of cresol adsorbed mainly, the correlation coefficients between amount adsorbed and physicochemical properties of activated carbon were calculated, and the results were shown in Table 4. At low equilibrium concentrations of 1 and 10 ppm, significant differences ($p<0.01$, $p<0.05$) were recognized between the amount adsorbed and the surface pH of activated carbon. It was concluded that the amount of cresol

Table 4. Correlation Coefficients between Properties of Adsorbent and Amount of Cresol Adsorbed.

Amount Adsorbed	Surface Area	Pore Volume			pH
		r<20Å	20<r<100Å	r<100Å	
at 1 ppm	+0.332	+0.125	+0.547	+0.399	-0.802**
at 10 ppm	+0.425	+0.181	+0.345	+0.453	-0.679*
at 10^2ppm	+0.503	+0.448	-0.023	+0.454	-0.353
at 10^3ppm	+0.433	+0.498	-0.436	+0.300	+0.155

Significantly Different; **p<0.01, *p<0.05.

adsorbed in low equilibrium concentrations was mainly affected by the surface pH of activated carbon.

The adsorption capacity of activated carbon for organic substances is dominated by pore structure (Giusti et al 1974, Abe et al 1976, Puri et al 1976). The greater the number of pores with a minimum diameter that the adsorbate can invade, the greater the amount adsorbed (Abe et al 1976). Activated carbon frequently contains hetero atoms such as oxygen, nitrogen and hydrogen. These hetero atoms may affect the adsorption capacity of activated carbon. Moreover, it is known the existence of surface oxygen groups on carbon, and the effect of surface oxygen groups on amount adsorbed in not neglected (Coughlin and Ezra 1968). It is reported that the contents of surface oxygen groups on carbon are related to surface pH (Sugiura et al 1985). Graham (1955) reported that acidic chemisorbed oxygen on carbon can play a very significant role in adsorption from an aqueous solution. According to our results, the amount of cresol adsorbed in low equilibrium concentrations is significantly affected by the surface pH of activated carbon.

In the case of liquid phase adsorption, intraparticle diffusivity is of major importance since it is the rate determining step in the adsorption process. Keii (1970) reported the following equation for calculating the intraparticle diffusivity (Di'). It refers to the concentration of adsorbate in a solid adsorbent for liquid phase adsorption under nonlinear equilibrium:

$$\mathrm{Log}(1-F^2)=-(\pi^2 Di'/2.303R^2)t \quad (1)$$

$$F=Qt/Q_\infty \quad (2)$$

where F is the adsorption ratio, R is the particle radius of adsorbent, t is the adsorption time, Qt is the amount of adsorbate adsorbed in the solid adsorbent at an elapsed time, and Q_∞ is the equilibrium amount adsorbed. In order to calculate the value of the intraparticle diffusivity by using equation (1), the amount adsorbed must be measured at regular intervals and at a fixed concentration. In this case, the amount adsorbed at an elapsed time was measured at a fixed concentration of 100 ppm and the equilibrium amount adsorbed was obtained from the adsorption isotherm at an equilibrium concentration of 100 ppm.

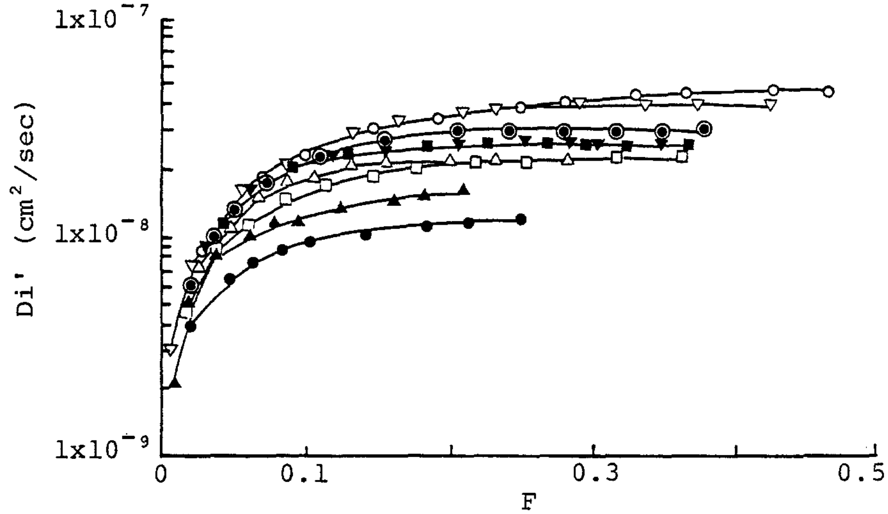

Figure 1. Change in Intraparticle Diffusivity with Adsorption Ratio of Cresol.
●:No.1, ◉:No.2, ▲:No.3, △:No.4, ■:No.5,
□:No.6, ▼:No.7, ▽:No.8, ○:No.9.

Fig. 1 shows the change in intraparticle diffusivity with the adsorption ratio. The curves of the logarithm of Di' against F became concave toward F and steadily approached their constant values. Activated carbon of No.8 and No.9 indicated large values of Di', and No.1 and No.3 indicated small values. From the standpoint of adsorption rate, activated carbon No.8 and No.9 were the preferable adsorbent. Yagi et al (1965) reported that for the adsorption of organic solvents on silica gel, the intraparticle diffusivity is small for small adsorbent mean pore radii. Urano and Kiyoura (1970) assumed that the intraparticle diffusivity of phenol onto coal was dominated by surface diffusion, because the pore radius of coal was small. Therefore, the intraparticle diffusivity is closely related to the pore structure of the adsorbent. Although we discussed the correlation between the value of Di' and the physicochemical properties of activated carbon, no significant correlation was found.

The treatment of the effluent from medical facilities is difficult because of the variety in the kinds of pollutants. Especially, the effluent from medical facilities contains a large amount of disinfectants. Sayama(1980,1981) reported on the treatment of the effluent containing disinfectant by an activated sludge method. It is difficult to decompose the disinfectant completely by activated sludge method. Moreover, trace amounts of undecomposed phenols react with chlorine in water and produce chlorophenols, a malodorous substance. Therefore, the advanced treatment of the waste water treatment containing disinfectants is desirable. In this investigation, we discussed the adsorption removal of cresol, one of the disinfectants, by activated carbon adsorption. It was found that the adsorption capacity of cresol in low concentration is mainly dominated by the surface pH of activated carbon.

REFERENCES

Abe I, Hayashi K, Kitagawa M (1976) Adsorption of nonionic surfactants. Yukagaku 25:145-150

Coughlin RW, Ezra FS (1968) Role of surface acidity in the adsorption of organic pollutants on the surface of carbon. Environ Sci Technol 2:291-297

Giusti DM, Conway RA, Lawson CT (1974) Activated carbon adsorption of petrochemicals.J Water Pollut Contr Fed 46:947-965

Graham D (1955) The separate effects of pore size and surface acidity on the adsorbent capacities of activated carbons. J Phys Chem 59:896-900

Hassler JW (1963) Activated Carbon. Chemical Publishing Company, New York

Keii T (1970) Kyuchaku(Adsorption). Kyoritsu Publishing Company, Tokyo

Kiba T, Terashima Y, Homma M (1972) Treatment of waste water from hospitals. Japan Analyst 21:1408-1417

Puri BR, Bhardwa SS, Gupta U (1976) Adsorption of phenol from aqueous solutions by carbons in relation to their specific surface areas. J Indian Chem Soc 53:1095-1098

Sayama N (1980) Basic experimental studies on the treatment of cresol, phenol and formalin. Jpn J Hyg 34:733-742

Sayama N (1981) Treatment of benzalkonium chloride and chlorhexidine. Jpn J Hyg 35:869-873

Sugiura M, Esumi K, Meguro K, Honda M (1985) Surface treatment of meso-carbon microbeads by oxygen plasma. Bull Chem Soc Jpn 58:2638-2640

Urano K (1975) Waste water treatment by activated carbon. Yuki Gosei Kagaku Kyokaishi 33:333-341

Urano K, Kiyoura R (1970) Adsorption characteristics of the coals in aqueous solutions. Kogyo Kagaku Zasshi 73:1921-1927

Yagi T, Shimizu K, Uchimoto K, Maeda S (1965) Adsorption kinetics of various hydrocarbons on silica gels. Kogyo Kagaku Zasshi 68:335-338

Received July 13,1989; accepted January 29,1990.