Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

Determination of Specific Surface Area of Granular Activated Carbon by Aqueous Phase Adsorption of Phenol and from Pore Size Distribution Measurements

M. K. N. Yenkie[a]; G. S. Natarajan[a]
[a] LAXMINARAYAN INSTITUTE OF TECHNOLOGY NAGPUR UNIVERSITY, NAGPUR, INDIA

To cite this Article Yenkie, M. K. N. and Natarajan, G. S.(1993) 'Determination of Specific Surface Area of Granular Activated Carbon by Aqueous Phase Adsorption of Phenol and from Pore Size Distribution Measurements', Separation Science and Technology, 28: 5, 1177 — 1190

To link to this Article: DOI: 10.1080/01496399308018028

URL: http://dx.doi.org/10.1080/01496399308018028

SEPARATION SCIENCE AND TECHNOLOGY, 28(5), pp. 1177–1190, 1993

Determination of Specific Surface Area of Granular Activated Carbon by Aqueous Phase Adsorption of Phenol and from Pore Size Distribution Measurements

M. K. N. YENKIE and G. S. NATARAJAN
LAXMINARAYAN INSTITUTE OF TECHNOLOGY
NAGPUR UNIVERSITY
NAGPUR 440010, INDIA

Abstract

Adsorption of phenol from the aqueous phase onto six grades of granular activated carbon (GAC) was studied at 35°C. The adsorption equilibrium data for all the GAC–phenol systems were analyzed by Langmuir, BET, and John's adsorption isotherm equations to estimate the specific surface area of each adsorbent. Pore size distribution measurements were also carried out for all the GAC samples. The surface area values obtained by both Langmuir and BET methods are in close agreement in the micromolar concentration range such as those present in wastewater. Pore size distribution measurement shows that for all the GAC samples used in this investigation, more than 97% of the surface area is located in the micropores below 35 Å in radius.

Granular activated carbon (GAC) adsorption is a recognized technology in wastewater and drinking water treatment. Most practical applications of activated carbon require sorbents with a large surface area having a large volume of very fine pores. The presence of micropores substantially influences its sorption properties because the amount adsorbed on the macropore surface is negligible in comparison to that for the micropores and mesopores (*1*). Therefore, characterization of the activated carbon has become one of the most important problems in adsorption technology. In view of growing interest in the use of GAC, there is need to develop some simple method of comparing potential adsorptive capacities of solid adsorbents. One such method is computation of specific surface areas. Estimation of the specific surface areas of GAC and other adsorbents is conventionally based upon measurements of the capacity of the adsorbent for a selected solute having a well-accepted molecular cross-sectional area.

Determinations of surface area are made by fitting either the Langmuir or BET equation to the isothermal equilibrium data. The N_2-BET adsorption method has long been employed for the surface area measurement of adsorbents (*2*), but this value may not be a true indication of the adsorption capacity of GAC during liquid-phase adsorption studies. Rybolt (*3*) determined surface areas for a series of C powders from BET, Langmuir, and virial models applied to the data obtained from N_2 adsorption at 77 K and acetic acid liquid-phase adsorption at 296 K, and observed that the surface area values obtained differed significantly. It is therefore more logical to determine the specific surface area of GAC from liquid-phase adsorption equilibrium data. An effort has been made in the present work to determine the specific surface areas of six grades of GAC.

Phenol is used as a primary reference solute in liquid-phase adsorption studies (*4–6*) and for measuring the specific surface areas of the adsorbents because it meets the requirements suggested by Giles (*7*). The cross-sectional area of the phenol molecule is also well established, and it is taken as 52.2 $Å^2$ for parallel orientation on the surface (*6*). Adsorption of single and mixed phenols on activated carbon has also been extensively studied in order to compare various adsorption theories (*8*, *9*). The literature also indicates that the adsorption of other organic solutes, such as *p*-nitrophenol (*10*), methylene blue (*11*), caffeine (*12*), and halophenols (*13*) from the aqueous phase, can be effectively used for characterizing the chemical nature of the adsorbent surface and for the determination of the specific surface areas of the adsorbents. It is also observed that the surface area measurements are usually carried out in rather concentrated adsorbate solutions (*5*, *12–14*), almost at the saturation concentration of the adsorbate. But since the concentrations of interest in water and wastewaters are micromolar, it appears desirable to examine the surface area values in lower concentration ranges such as those present in wastewater.

In the present investigation, determinations of the surface areas for various grades of GAC are made by fitting the Langmuir, BET, and John's (*15*) equations to the experimental data, and these values are compared with those obtained by the N_2-BET method and from the pore size distribution measurements.

EXPERIMENTAL

AR (BDH) grade phenol, further purified by extraction with petroleum ether (40–60°C) at 50–55°C by vigorous shaking (*16*), was used in this work.

The GAC samples were first sieved to obtain a 12 × 16 mesh (B.S.) fraction (particle diameter = 0.1502 ± 0.009 cm). The uniform fractions were then washed several times with distilled water until no suspended material appeared in the washings, dried in an oven at 100–110°C for 24 hours, and then stored in a $CaCl_2$ desiccator at room temperature until use. The GAC samples used in the present work are summarized in Table 1. The physical properties of the adsorbents used are given elsewhere (*17*).

Experimental Studies

To evaluate the adsorption equilibrium data, the experiments were carried out in a batch system. The experimental setup consisted of a thermostated round-bottom flask and a motor impeller assembly which enabled vigorous stirring of the solid–liquid system by a two-bladed Teflon stirrer. From the preliminary studies it was found that the time to reach equilibrium was independent of the stirring rate for a shaft speed higher than 600 rpm. All the experiments were thereafter carried out at a stirring speed of 800 ± 50 rpm. An electronic stroboscope was used to determine the speed of the agitator assembly. The temperature of the system was kept constant at 35 ± 0.2°C.

For adsorption equilibrium data, 1 L of the adsorbate solution of known concentration was placed in a 2-L round-bottom flask and an accurately weighed quantity of GAC was added to the flask. The adsorbent/adsorbate ratio was kept constant at 0.500 g/L for all the experiments. During the studies on the kinetics of adsorption for the same adsorbent/adsorbate systems, it was observed that about 60 to 80% of the adsorbate was removed in the first hour of contact and the fractional approach to equilibrium took 4 hours. Literature (*4*, *18*) also supports this observation. After about 5 hours of stirring, the adsorbate concentration at equilibrium, C_e, was determined by UV adsorption at 270 nm using a SPEKTROMOM 201 (Hungarian optical works, MOM, Budapest) spectrophotometer. To ensure that equilibrium was attained, some experiments were run for a prolonged period of time (several days) where no change in the equilibrium concentration values could be observed. Some points on the isotherm were also verified for their reproducibility.

Regression analysis of the experimental data of concentration versus optical density for the standard Beer's law plot gave the relation

$$C = [6.8882 \times 10^{-4}\ (\text{optical density})] - (2.28 \times 10^{-6})$$

Adsorption Isotherms

The adsorption equilibrium data for all six GAC–phenol systems are summarized in Table 2. It is possible to conclude from plots of q_e versus C_e that all the isotherms obtained in this work fall under the Type I fa-

TABLE 1
GAC Samples Used in This Investigation

No.	GAC	Origin	N_2-BET surface area (m^2/g)	Manufacturer	Gifted by[a]
1	RRL	Coconut shell	900	Regional Research Laboratory, Hyderabad, India	Regional Research Laboratory, Hyderabad, India
2	CAL	Coal	1090	California Carbon Co., 22610 S. Western Avenue, Torrence, California, USA	Dr. Kenji Hashimoto, Department of Chemical Engineering, Kyoto University, Kyoto, Japan
3	KURARE	—	1020	In Japan	Dr. Kenji Hashimoto, Department of Chemical Engineering, Kyoto University, Kyoto, Japan
4	F-200 (Filtrasorb-200)	Bituminous coal	900	Calgon Corp., A.C. Division, P.O. Box 1346, Pittsburgh, Pennsylvania, USA	Calgon Corp.
5	F-400 (Filtrasorb-400)	Bituminous coal	1100	Calgon Corp., A.C. Division, P.O. Box 1346, Pittsburgh, Pennsylvania, USA	Calgon Corp.
6	LCK	Coconut shell	1025	National Carbon Co., Union Carbide Corp., 270 Park Ave., New York, New York, 10017, USA	Dr. John S. Zogorski, Environmental System Application Center, Bloomington, Indiana 47401, USA

[a] The authors are thankful to the organizations mentioned in this column for their kindness.

vorable isotherm category. The adsorption isotherm plots are given elsewhere (*17*).

The adsorption equilibrium data were fitted into Langmuir, BET, and John's isotherm equations for determining the adsorbent surface area. The Langmuir treatment is based on the assumption that maximum adsorption corresponds to a saturated monolayer of adsorbate molecules on the adsorbent surface, that the energy of adsorption is constant, and that there is no transmigration of adsorbate molecules in the plane of the surface. The equation can be expressed as

$$q_e = \frac{Q^0 b C_e}{1 + b C_e} \tag{1}$$

Rearranging Eq. (1) gives

$$\frac{1}{q_e} = \frac{1}{Q^0 b C_e} + \frac{1}{Q^0} \tag{2}$$

Thus, a plot of $1/q_e$ versus $1/C_e$ should be linear if Langmuir adsorption is operative, permitting calculation of Q^0. Knowledge of Q^0 leads to determination of the surface area of the adsorbent. However, it must be strictly valid over the range of concentrations employed in an investigation. A mathematical analysis of the data will validate this point.

The BET isotherm method usually employs solutions near the saturation concentration of the solute (*5*, *14*), but since the concentrations of interest in wastewater are micromolar, it appeared desirable to examine the surface areas observed for adsorption in lower concentration ranges such as those used in this work. The BET plots for all the GAC–phenol systems were plotted according to the equation

$$\frac{C_e}{q_e(C_s - C_e)} = \frac{1}{Q^0 Z} + \frac{(Z - 1)}{Z Q^0} \frac{C_e}{C_s} \tag{3}$$

From the slope and the intercept, it is possible to obtain Q^0 values.

John's adsorption isotherm (*15*) provides a rapid graphical method for determination of monolayer capacity Q^0. The isotherm equation is expressed as

$$\log \log (C_e/C_s) = n_0 \log q_e + y \tag{4}$$

A plot of $\log \log (C_e/C_s)$ versus $\log q_e$ shows a kink at the point corresponding to Q^0 (Fig. 1). The values of Q^0 and the surface areas for all the

TABLE 2
Adsorption

RRL–phenol			CAL–phenol			KURARE–phenol		
$C_0 \times 10^4$	$C_e \times 10^4$	$q_e \times 10^4$	$C_0 \times 10^4$	$C_e \times 10^4$	$q_e \times 10^4$	$C_0 \times 10^4$	$C_e \times 10^4$	$q_e \times 10^4$
0.50	0.15	0.70	0.28	0.15	0.26	1.25	0.25	2.00
1.05	0.15	1.80	0.90	0.25	1.30	1.80	0.40	2.80
1.35	0.25	2.20	1.35	0.35	2.00	2.30	0.55	3.50
1.50	0.30	2.40	3.35	1.10	4.50	4.05	0.90	6.30
2.50	0.45	4.10	5.20	2.05	6.30	4.75	1.20	7.10
2.55	0.50	4.10	6.20	2.70	7.00	5.00	1.40	7.20
4.25	0.95	6.60	7.65	3.60	8.10	5.50	1.50	8.00
4.90	1.25	7.30	8.75	4.50	8.50	7.60	2.60	10.00
5.20	1.35	7.70	11.80	6.70	10.20	10.10	4.10	12.00
6.65	2.00	9.70	14.15	8.50	11.30	11.75	5.20	13.10
7.70	2.65	10.10	14.70	9.00	11.40	14.00	7.20	13.60
8.35	3.00	10.70				15.50	8.50	14.00
10.50	4.55	11.90				15.95	9.00	13.90
12.90	6.60	12.60						
14.70	8.30	12.80						

GAC–phenol systems determined by using these methods are reported in Table 3.

The specific surface area, S, is related to Q^0 by the relation

$$S = NQ^0A \tag{5}$$

The value of $A = 52.2$ Å^2 for parallel orientation of the phenol molecule on the adsorbent surface (*6*) was used in these calculations.

Pore Size Distribution

GAC is a solid of infinite structure. It is highly porous in character, and both pore and surface characteristics of samples depend on the preparation conditions [type of atmosphere, heating rate, temperature, and degree of carbon burnout (*19*)]. Several workers (*20*, *21*) have shown that rapid adsorption takes place in the macropores, and a portion of the adsorbate finds its way into the micropores over a long period of time.

Pore size distribution is commonly measured by mercury intrusion porosimetry. The method consists of monitoring the volume of mercury that invades pore space when a fixed pressure is applied. Some authors (*22*, *23*) have also used the N_2-adsorption method for estimating pore size distribution over both micropore and mesopore ranges. Comparison of pore

Equilibrium Data

F-200-phenol			F-400-phenol			LCK-phenol		
$C_0 \times 10^4$	$C_e \times 10^4$	$q_e \times 10^4$	$C_0 \times 10^4$	$C_e \times 10^4$	$q_e \times 10^4$	$C_0 \times 10^4$	$C_e \times 10^4$	$q_e \times 10^4$
0.65	0.20	0.90	0.70	0.15	1.10	0.55	0.15	0.80
1.10	0.35	1.50	1.15	0.35	1.60	1.30	0.25	2.10
2.15	0.60	3.10	2.10	0.40	3.40	1.95	0.35	3.20
4.25	1.30	5.90	4.40	1.15	6.50	3.85	0.85	6.00
5.05	1.60	6.90	4.90	1.40	7.00	3.95	0.90	6.10
5.90	2.20	7.40	6.35	2.10	8.50	4.70	1.10	7.20
7.80	3.40	8.80	8.25	3.25	10.00	5.85	1.65	8.40
9.75	4.95	9.60	10.15	4.50	11.30	7.60	2.55	10.10
12.00	6.60	10.80	12.40	6.70	11.40	9.40	3.65	11.50
14.90	8.90	12.00	14.40	8.60	11.60	10.90	5.10	11.60
			15.80	10.00	11.60	12.95	7.20	11.50

size distribution and surface area values obtained from N_2-adsorption and mercury penetration shows that the results are in agreement only in the lower pressure mercury penetration data whereas the data were more consistent with particle degradation at higher mercury penetration pressures (*24*). The literature (*25*) further supports the possibility that during the course of mercury penetration at higher pressures, the adsorbent may undergo either compression or breakdown, resulting in a situation where the pore size distribution determined experimentally may be quite different from that in the original solid. The internal surface area of activated carbons, generally greater than 400 m^2/g, is mainly located in the micropores, i.e., where the pore diameters are less than 20 Å. Whether or not a specific molecule is adsorbed in a pore is determined by the size of both the molecule and the pore. Gonsalves and coworkers (*26*), during their studies on adsorption of N_2 of 77 K and of ethanol and *n*-heptane at 298 K on activated carbon, observed that the accessibility of ethanol and *n*-heptane molecules was restricted in ultramicropores.

Determination of the pore size distribution of GAC samples used in the present work was accomplished with the cooperation of several agencies. The pore size distribution for all grades of GAC except CAL were carried out on a Carlo Erba mercury porosimeter (Alchemi, Bombay, India). CAL pore size distribution was supplied by Dr. Kenji Hashimoto of Kyoto

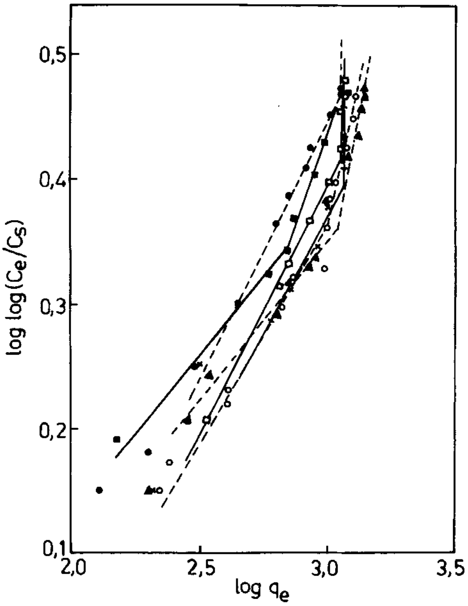

FIG. 1. John's adsorption isotherms: RRL (○ - - -), CAL (● - - -), F-200 (■ ——), KURARE (▲ - - -), F-400 (□ ——), LCK (× ——).

University, Kyoto, Japan, and for Filtrasorb-200 and Filtrasorb-400, measurements were supplied by Mr. T. Hayakawa of Calgon Division, MSD, Tokyo, Japan. These results are presented in Figs. 2–5.

The Carlo Erba mercury porosimeter is not able to detect pores below 35 Å, so one of the samples (KURARE) was investigated for its pore size distribution by an AMINCO porosimeter (R & D, A.C.C. Ltd., Bombay, India) which showed (Fig. 4) that pores of the order of 10 Å are also present.

Obviously, this means that depending upon the type of equipment, pore size distribution is likely to vary in the lower pore diameter range. This is further corroborated by Fig. 3 for F-200 and F-400 GAC samples, where the existence of pores of the order of 6.5 Å is clearly shown, whereas investigations with the Carlo Erba mercury porosimeter indicated a minimum radius of 35 Å. It thus appears that since the estimated value of the pore diameter is likely to vary considerably depending upon the method used to observe it, the surface area calculated from pore size distribution is likely to throw more light on the presence of pores of lower diameters and their abundance. The surface area can be calculated by knowing the

TABLE 3
Comparison of Adsorption Isotherms and Surface Areas

	Langmuir isotherm		BET isotherm				John's isotherm,	Surface area			
System	Equation	$Q^0 \times 10^4$	Slope	Intercept	Z	$Q^0 \times 10^4$	$Q^0 \times 10^4$	Langmuir	BET	John's	N_2-BET
RRL–phenol	$q_e = \frac{11.95C_e}{1 + 7936C_e}$	15.0579	668.46	0.0872	7666.8	14.96	10.00	473.6	471.0	302	900
CAL–phenol	$q_e = \frac{5.7636C_e}{1 + 3985C_e}$	14.4632	699.83	0.1830	3824.8	14.29	11.22	454.9	449.5	353	1090
KURARE–phenol	$q_e = \frac{8.4388C_e}{1 + 4357C_e}$	19.3684	579.05	0.1114	5198.9	17.27	10.96	609.1	543.1	345	1020
F-200–phenol	$q_e = \frac{6.7159C_e}{1 + 4439C_e}$	15.1293	696.00	0.1470	4735.7	14.36	7.08	475.8	451.6	223	900
F-400–phenol	$q_e = \frac{11.3417C_e}{1 + 8517C_e}$	13.3165	764.64	0.0851	8986.2	13.08	11.22	418.8	411.3	353	1100
LCK–phenol	$q_e = \frac{13.1527C_e}{1 + 11009C_e}$	11.9472	745.85	0.0769	9701.2	13.41	11.48	375.7	421.7	361	1025

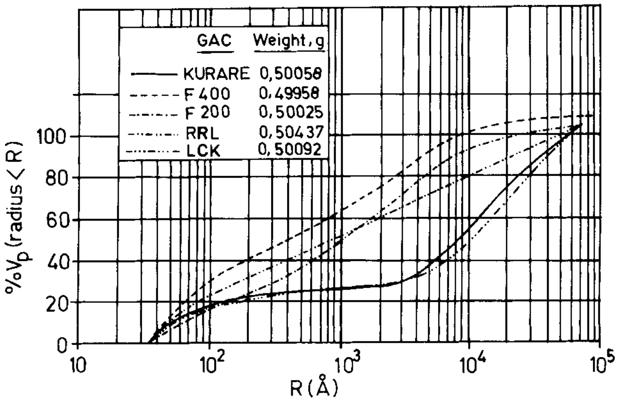

FIG. 2. Pore size distribution of GAC samples.

volume of mercury penetrating into the pore space of a given diameter (*27*):

$$\text{Surface area} = \frac{4(\text{volume of Hg})}{\text{diameter of pore}} \tag{6}$$

The surface areas calculated by using the above relation and Figs. 2 to 5 are summarized in Table 4.

It is obvious from Table 4 that a great many of the pores are likely to be smaller in diameter than estimated by the Carlo Erba porosimeter.

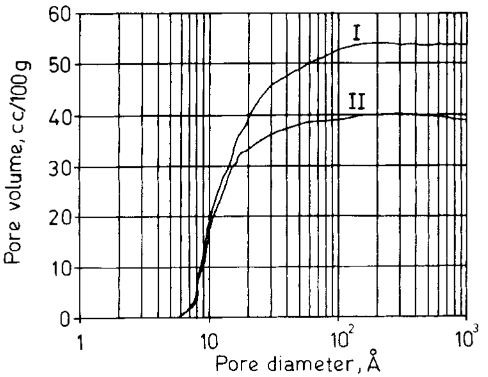

FIG. 3. Pore size distribution of GAC F-200 (II) and F-400 (I).

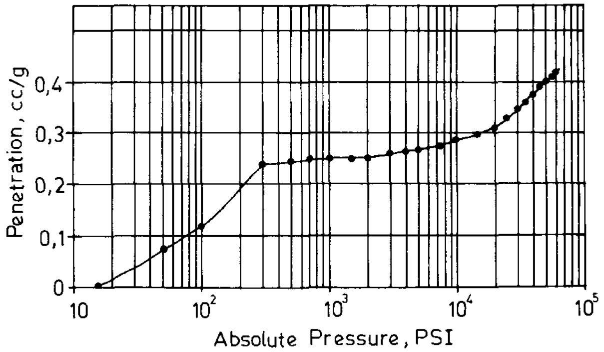

FIG. 4. Pore size distribution of GAC KURARE. Equivalent pore diameter, microns = 175/psi. Contact angle = 130°.

Since the surface area calculated by this method is only around 3% of the surface area calculated by the N_2-BET method, it may be inferred that 97% of the adsorbent surface area exists in pores of less than 35 Å radius, which explains the slow approach to equilibrium following an initial rapid uptake period. The surface areas of GAC samples have also been calculated by using the experimentally determined adsorption isotherms.

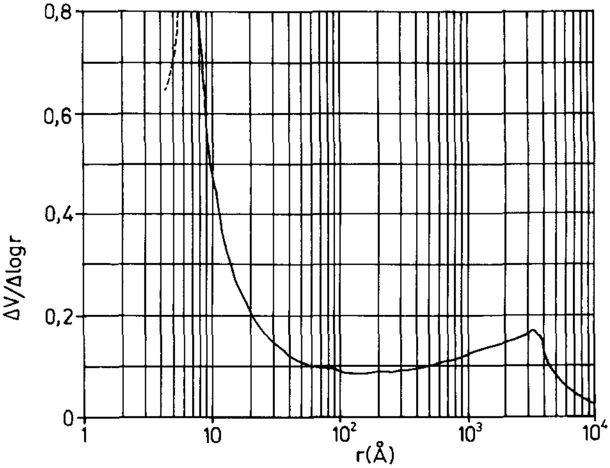

FIG. 5. Pore size distribution of GAC CAL.

TABLE 4
Surface Area (m^2/g) Measurements

GAC	Carlo Erba	AMINCO	N_2-BET[a]
RRL	31.20	—	900
CAL	—	—	1090
KURARE	29.75	977	1020
F-200	17.56	—	900
F-400	48.07	—	1100
LCK	16.53	—	1025

[a] Information provided by the manufacturer.

RESULTS AND DISCUSSION

Table 2 reports the experimental adsorption equilibrium data for all six GAC–phenol systems.

Table 3 reports Langmuir and BET equations for all the systems obtained by regression analysis of experimental equilibrium data. The regression coefficients were above 0.98 in all cases, indicating a very good mathematical fit. Table 3 also reports the Q^0 values obtained by the Langmuir, BET, and John's isotherm methods.

It is not surprising that the surface area values obtained by the Langmuir and the BET equations are fairly close since the BET equation reduces to the Langmuir equation at low concentrations of the adsorbate (*25*). The surface area values determined by John's method are lower than the values obtained by the Langmuir and BET methods by about 30%, probably due to the empiricism of John's method.

The values of the surface area obtained by the N_2-BET method are considerably higher since the adsorbate nitrogen has a low cross-sectional area (16.2 Å^2) compared to the cross-sectional area of the phenol molecule. The reason for the low surface areas observed with phenol is because the finer pores, which should be accessible to the smaller N_2 molecules, are blocked by the larger phenol molecules. The pore-size distribution measurements also indicate that more than 97% of the total surface area exists in micropores of less than 35 Å in radius.

It is also observed that the BET–phenol surface area as determined in concentrated adsorbate solution (*5*, *14*) is about two times larger than the values obtained in the present work. Morris and Weber (*29*) studied the adsorption of phenol on Columbia LCK activated carbon at 30°C and observed that the equilibrium data fitted well to the BET equation in the high concentration range (equilibrium concentration $<$ 300 mmol/L), whereas the data in the lower concentration range (equilibrium concentration < 220 μmol/L) are best described by the Langmuir equation. The BET–phenol surface area was found to be 800 m^2/g and the Langmuir–phenol surface area was 270 m^2/g, roughly one-third of the BET value. Thus, prediction of surface area in the concentrated adsorbate solution may give a high surface area value and not the true picture in the micromolar range of concentrations such as those present in water and wastewater.

It is therefore logical to conclude that surface areas determined by the Langmuir method are the most appropriate ones for wastewater treatment studies where the concentrations are of the micromolar order.

NOMENCLATURE

A	cross-sectional area of phenol molecule (m^2)
b	constant in Eq. (1)
C	concentration of adsorbate in liquid phase (mol/L)
C_0	initial concentration of adsorbate (mol/L)
C_e	equilibrium concentration of adsorbate (mol/L)
C_s	saturation concentration of phenol in water at 35°C (0.97 mol/L) (*6*)
q	adsorbent phase concentration (mol/g)
q_e	adsorbent phase concentration at equilibrium (mol/g)
Q^0	monolayer capacity of the adsorbent (mol/g)
N	Avogadro's number
n_0	constant in Eq. (4)
S	adsorbent surface area (m^2/g)
y	constant in Eq. (4)
Z	constant in Eq. (3)

Acknowledgments

M.K.N.Y. wishes to thank the University Grants Commission for providing financial support for carrying out this work. Thanks are also due to Dr. G. D. Nageshwar, Director, Laxminarayan Institute of Technology, Nagpur University, Nagpur, for providing facilities.

REFERENCES

1. M. M. Dubinin, *Carbon, 21,* 359 (1983).
2. E. Robens, *TIZ Int. Mag. Powder Bulk, 115*(7/8), 304 (1991) (in German).
3. T. R. Rybolt, *J. Tenn. Acad. Sci., 61*(3), 66 (1986).
4. C. O. M. Miller and C. W. Clump, *AIChE J., 16*(2), 169 (1970).
5. N. D. Drozhalina and N. O. Bulgakova, *Zh. Prikl. Khim., 47*(2), 298 (1974).
6. B. R. Puri, in *Activated Carbon Adsorption of Organics from Aqueous Phase,* Vol. 1

(I. H. Suffet and M. J. McGuire, Eds.), Ann Arbor Science, Ann Arbor, Michigan, 1980, p. 353.

7. C. H. Giles, T. H. MacEwan, S. Nakhwa, and D. Smith, *J. Chem. Soc.*, p. 3973 (1960).
8. E. Worch and R. Zapke, *Acta Hydrochim. Hydrobiol.*, *14*(3), 305 (1986) (in German).
9. E. Costa, G. Calleja, and L. Marijuan, *Adsorp. Sci. Technol.*, *5*(3), 213 (1988).
10. J. V. Ibarra and J. J. Lazaro, *Tec. Lab.*, *11*(137), 266 (1987) (in Spanish).
11. H. G. Linge, *Fuel*, 68(1), 111 (1989).
12. H. F. Stoeckli, M. Fraguiere, S. Huguenin, M. Depraz, and L. Bellerini, *Carbon*, *26*(6), 915 (1988).
13. A. V. Mamchenko and T. I. Yakimova, *Zh. Fiz. Khim.*, *60*(8), 1993 (1986) (in Russian).
14. B. R. Puri, S. S. Bhardwai, and U. Gupta, *J. Indian Chem. Soc.*, *53*, 1095 (1976).
15. P. T. John, T. A. K. Ghori, and K. C. Nagpal, *Indian J. Technol.*, *18*, 261 (1980).
16. P. H. Mars, *Pharm. Weekbl.*, *88*, 319 (1953).
17. M. K. N. Yenkie and G. S. Natarajan, *Sep. Sci. Technol.*, *26*(5), 661 (1991).
18. J. S. Zogorski and S. D. Faust, in *Removal of Phenols from Polluted Waters*, Project A-003NJ, New Jersey Water Resources Research Institute, 1974.
19. W. E. White, C. H. Bartholomew, W. C. Hecker, and D. M. Smith, *Adsorp. Sci. Technol.*, *7*(4), 180 (1990).
20. F. E. Bernardin Jr., *Chem. Eng.*, *83*, 77 (1976).
21. R. G. Peel, A. Benedek, and C. M. Crowe, *AIChE J.*, *27*(1), 26 (1981).
22. S. Sato, *J. Chem. Eng. Jpn.*, *21*(5), 534 (1988).
23. N. A. Seaton, J. P. R. B. Walton, and N. Quirke, *Carbon*, *27*(6), 853 (1989).
24. D. R. Milburn, B. D. Adkins, and B. H. Davis, *Stud. Surf. Sci. Catal.*, *39*, 501 (1987).
25. I. B. Paul, T. N. Roy, and P. N. Mukherjee, *Indian J. Technol.*, *20*, 441 (1982).
26. G. da Silva, M. Amelia, V. A. M. Soares, J. C. Calado, and M. Brotas de Carvalho, *J. Chem. Soc., Faraday Trans.*, *87* (23), 3799 (1991).
27. J. M. Coulson and J. F. Richardson, in *Chemical Engineering*, Vol. III., Pergamon Press, New York, 1980, p. 476.
28. J. W. Weber, *Physicochemical Process for Water Quality Control*, Wiley-Interscience, New York, 1972.
29. J. C. Morris and W. J. Weber Jr., in *Adsorption of Biochemically Resistant Materials from Solution*—1, AWTR-9, Report 999-WP-11, U.S. Department of Health, Education and Welfare, 1964.

Received by editor November 8, 1991
Revised June 1, 1992