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Application of the Theory of Multimolecular Adsorption to the Experimental Data

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The validity of BET equation has been proved by applying it successfully to the data on the adsorption, on coal, of the acid and water from an aqueous solution of acetic acid as obtained by Ostwald and Izaguirre in 1922,

INTRODUCTION

The BET equation[1] was introduced on the assumption that adsorption does not stop with the formation of a monolayer at the interface but also include formation of multilayers. Since the assumption on which the equation has been built up is of the most general nature, it is quite likely that adsorption of a multilayer type should also be possible in quite a good number of cases, not referred to by Brunauer, Emmett and Teller or other workers. It was of interest, therefore, to test the possibility of the applicability of BET equation to the data already published before the introduction of the BET equation. Such data having been accumulated even before the BET equation was thought of, should have no special bias in proving the applicability of BET equation and as such can be taken to be independent and impartial.

From the data collected, a random choice was made out of a number of cases studied, one case of adsorption, on coal, of the acid and water from an aqueous solution of acetic acid, as studied by Ostwald and Izaguirre[2], is quoted in the present communication.

DISCUSSION

Calculation of Data : The data of observations, as reported by the workers[2], are presented in (Table 1) :

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TABLE 1

Percentage of acetic acid (a)	Acetic acid (gm) adsorbed on 10 gm of coal (v)	Water (gm) adsorbed on 10 gm of coal (v′)	Percentage of water (by difference) (a′)
0.000	—	7.780	100.000
0.006	0.159	7.778	99.994
0.033	0.325	7.774	99.967
0.130	0.579	7.770	99.870
0.438	0.968	7.730	99.562
1.262	1.500	7.640	98.738
3.935	2·420	7.340	96.065
8.513	3.345	6.790	91.487
14.000	4.130	6.280	86·000
23.800	5.160	5.360	76.200
35.220	6.060	4.420	64.780
57.230	7.440	2.740	42.770
73.710	8.080	1.730	26.290
80.000	8.570	1.200	20.000
90.000	9.110	0.610	10.000
95.000	9.210	0.310	5.000
100.000	9.420	0.000	0.000

Application of BET equation to adsorption of acetic acid only : From the plot of a vs. v, the values of v were determined for various convenient values of $x = a/100$ (vide Table 2). If the number of layers adsorbed = $n = 1$, then $x = \pi(nx) = \Delta(nx)$, where[3]

$$\pi(nx) = x \cdot \frac{(1 - x^n)}{(1 - x)} \qquad \cdots(1)$$

and

$$\Delta(nx) = x \cdot \frac{(1 - x^n) - nx^n(1 - x)}{(1 - x)^2} \qquad \ldots(2)$$

TABLE 2

x	v	For n = 1		For n = 2		For n = 3	
		$\pi(nx)$	$\Delta(nx)/v$	$\pi(nx)$	$\Delta(nx)/v$	$\pi(nx)$	$\Delta(nx)/v$
0.1	3.60	0.100	0.028	0.110	0.031	0.111	0.034
0.2	4.76	0.200	0.042	0.240	0.059	0.248	0.064
0.3	5.66	0.300	0.053	0.390	0.085	0.421	0.100
0.4	6.40	0.400	0.063	0.560	0.113	0.624	0.143
0.5	7.00	0.500	0.071	0.750	0.143	0.875	0.197
0.6	7.60	0.600	0.079	0.960	0.174	1.176	0.260
0.7	8.00	0.700	0.088	1.190	0.210	1.531	0.339
0.8	8.57	0.800	0.093	1.440	0.243	0.952	0.413
0.9	9.11	0.900	0.099	1.710	0.277	2.439	0.517

The curve obtained by plotting $\pi(nx)$ vs. $\Delta(nx)/v$ (for $n = 1$) is not a straight line (cf. Fig. 1); i.e., the BET equation is not applicable[3] for $n=1$. For $n = 2$ and 3, the corresponding values of $\pi(nx)$ and $\Delta(nx)/v$ are calculated using equations 1 and 2 (Table 2, cf. Fig. 2).

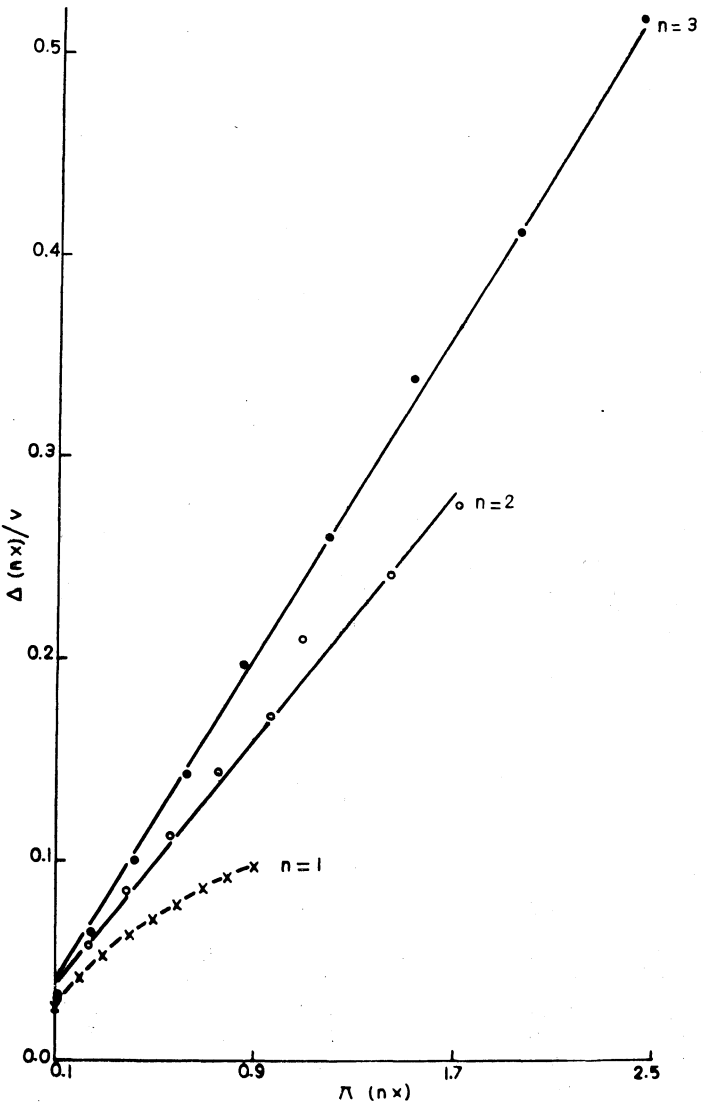

Fig. 1

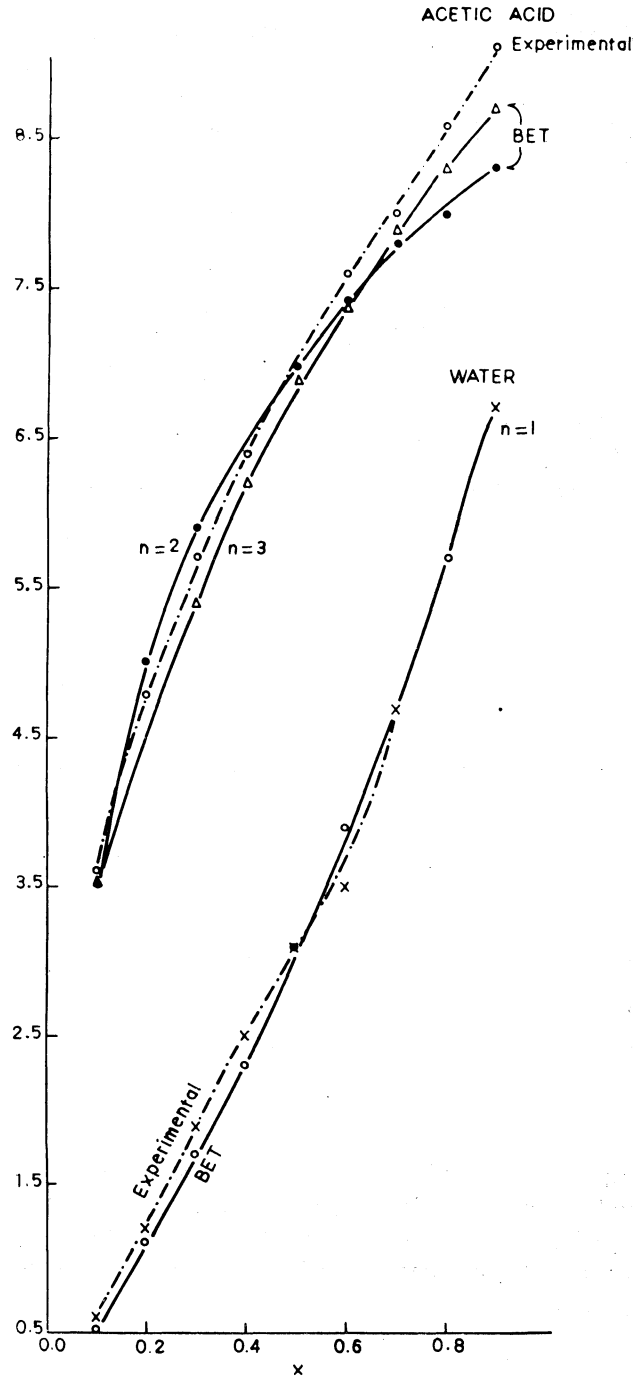
ACETIC ACID
Experimental
BET
WATER
n=1
n=2
n=3
Experimental
BET
8.5
7.5
6.5
5.5
4.5
3.5
2.5
1.5
0.5
0.0
0.2
0.4
0.6
0.8
x

Fig. 2

The slope ($1/v_m$) of the straight line obtained by plotting $\pi(nx)$ vs. $\Delta(nx)/v$ and the intercept ($1/v_m \cdot c$) give the values of v_m, and c, where v_m is the amount of adsorbate adsorbed when the entire adsorbent surface is covered with a complete unimolecular layer[1,3], and c is the concentration of adsorbate in equilibrium with the adsorbent[1,3]. These values have been used to calculate theoretically the amount of acetic acid (v) that should have been adsorbed in accordance with thc BET equation[1], where,

$$v = \frac{v_m \cdot c \cdot x}{(1-x)}\left[\frac{1-(n+1)x^n + nx^{n+1}}{1+(c-1)x - cx^{n+1}}\right] \quad \ldots(3)$$

The values of v were calculated by using the BET equation 3 (Table 4).

Applicability of the BET equation to the adsorption of water only : From the plot of v′, i. e., water adsorbed on 10 gm of coal, against a′, the percentage of water (Table 1), the values of v for various convenient values of x were determined (Table 3).

TABLE 3 (for n = 1)

x	v	$\pi(nx)$	$\Delta(nx)/v$
0.1	0.61	0.10	0.164
0.2	1.20	0.20	0.167
0.3	1.90	0.30	0.158
0.4	2.45	0.40	0.I63
0.5	3.10	0.50	0.161
0.6	3.84	0.60	0.156
0.7	4.72	0.70	0.148
0.8	5.70	0.80	0.140
0.9	6.70	0.90	0.134

The curve obtained by plotting $\pi(nx)$ vs. $\Delta(nx)/v$ is a straight line. The slope and intercept of this straight line give the values[1,3] of v_m and c. These values have been used to calculate theoretically the amount of water (v′) that should have been adsorbed in accordance with the BET equation 3 (Table 4).

TABLE 4

x	gm of acetic acid adsorbed (v)			gm of water adsorbed (v^1)	
	Experimental	By BET equation For n = 2	By BET equation For n = 3	Experimental	By BET equation For n = 1
0.1	3.600	3.519	3.527	6.700	6.750
0.2	4.760	4.995	4.910	5.700	5.700
0.3	5.660	5.895	5.404	4.720	4.717
0.4	6.400	6.403	6.236	3.840	3.845
0.5	7.000	7.030	6.852	3.100	3.048
0.6	7.600	7.425	7.392	2.450	2.333
0.7	8.000	7.733	7.891	1.900	1.687
0.8	9.570	8.041	8.341	1.200	1.071
0.9	9.110	8.275	8.740	0.610	0.514

The data and the subsequent calculations, therefore, indicate that the adsorption of acetic acid is concentration dependent, forming a bimolecular layer at lower concentrations and a termolecular layer at higher concentrations, whereas the adsorption of water monolayer and therefore independent of its concentration. Thus, the BET equation is applicable to the acetic acid-water system.

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