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Study on the Adsorption of Pharmaceuticals by Filter aids

—Adsorption of caffeine, niacinamide, pyridoxine hydrochloride and thiamine hydrochloride by Celite 545 and Dawsonite—

*Yona Woo Hyun, *Shin Hong Min, *Shang Hi Rhee *Yong Bae Kim

緩衝液에서의 caffeine, niacinamide, pyridoxine hydrochloride, thiamine hydrochloride의 濾過助劑에 依한 吸着현상을 比較檢討하였다. 實驗에 使用한 vitamin類의 吸着은 *Langmvir* 吸着式의 pattern에 따랐으며 caffeine은 거의 吸着되지 않았다.

濾過助劑中 Dawsonite는 Celite 545 보다 큰 吸着현상을 나타냈으며 pH 變化에 따른 影響은 niacinamide가 가장 예민하였고 pH의 增加는 吸着量을 현저히 감소시켰다.

A filter aid is a material which serves to obtain or improve clarification or increase the filtration rate, or both. A filter aid must be free from objectionable impurities such as soluble salts, organic matter, coloring material, taste or order imparting bodies, gritty particles, etc.

It must be also inert or unreactive in the material being filtered. The proper choice of filter aid depends upon the type and size of suspended material to be iltered out.

The quality factor of the filtrate and its suitability for further treatment is frequently more important than the filtration rate[1]. The synthesized Dawsonite [2-4], $NaAl(OH)_2CO_3$, was used as filtration aid, decolorizer, deacidification agent and cation exchanger.

The Dawsonite showed adsorption activity in the dilution solution of calcium and barium ions, and heavy metals at low temperature, but at the high temperature, it showed cation exchange activity[4].

The results of those experiments showed a greater activity than carbon, diatom earth, which are used commonly.

From time to time papers[5,6] have been presented which dealt with various

*Dong-A Pharm. Co., Ltd.

phases of clarification with diatomaceous silica filter aids, the most widely used type of filter aid, but for the most they have given a chemical or physical interpretation.

In the present study the adsorption of caffeine, niacinamide, pyridoxine hydrochloride and thiamine hydrochloride on diatom earth and Dawsonite from aqueous solution was investigated for revealing the adsorption features of filter aids.

EXPERIMENTAL

Materials.——Dawsonite (Korea Dawsonite Co., Ltd.), 120 mesh; Celite 545 (Hayashi pure chemicals), 120 mesh; caffeine monohydrate (KP Ⅱ); niacinamide, m.p. 130°; pyridoxine hydrochloride, m.p. 205°; thiamine hydrochloride, m.p. 249°;

Equipment.——Constant temperature water bath equipped with a mechanical stirrer, Beckman DU-2 spectrophotometer, 30ml. glass stoppered bottle, 1cm. quartz cell.

Procedure.——About 200mg. of Dawsonite and Celite 545 were suspended in 30ml. bottles of various concentrations (usually between 1×10^{-3} M and 1×10^{-4}M) of caffeine, niacinamide, pyridoxine hydrochloride and thiamine hydrochloride solution in USP XVIII standard buffer of pH 3,5,7 respectively and shaken in a thermostat at 5° or 30° with mechanical stirrer for 24 hours.

A series of control bottles in which contained buffer solution rather than filter aids were also prepared.

After equilibration the supernatant liquid was removed and isolated by centrifugation or filtration, and concentration of caffeine and vitamins was determined.

Method of analysis.——The test soluiton was diluted with distilled water and the spectrophotometric analysis method was employed to determine the concentrations of the drugs at the following wave lengths; caffeine, 272 mu; niacinamide, 260mu; pyridoxine hydrochloride, 325mu; thiamine hydrochloride, 274 mu.

RESULTS AND DISCUSSION

The adsorption isotherms on filter aids apparently fitted the *Langmuir* type equation,

$$\frac{1}{r}=\frac{1}{n}+\frac{K}{nA}$$

where rrepresents the amount of an adsorbate adsorbed on a unit weight filter aids, n the maximum of r, K the constant which indicates the strength of the adsorption, and A the equilibrium concentration of the adsorbate in the solution.

The values of n and K were obtained utilizing the graph and the apparent heat of adsorption, $\Delta H_{app.}$ and the apparent entropy change, $\Delta S_{app.}$, were calculated. The values of these constants are listed in Table 1.

Table 1. Constants for adsorption of vitamins on filter aids

Vitamin/Filter aid	pH	Temp. (°C)	n(M/G× 10^3)	K(L/M× 10^{-3})	$\Delta G_{app.}$ (K cal/M)	$\Delta H_{app.}$ (K cal/M)	$\Delta S_{app.}$ (e.u.)
Niacinamide	3	5	1.13	0.0508	−2.18	1.93	14.7
		30	1.13	0.0678	−2.55		
Dawsonite	5	5	1.13	1.184	−3.93	1.08	18.0
		30	1.13	1.391	−4.38		
	7	5	1.13	3.683	−4.56	1.11	20.3
		30	1.13	4.340	−5.07		
Celite 545	3	5	1.00	0.963	−3.82		
Pyridoxine hydrochloride	3	5	0.90	0.130	−2.70	3.87	23.6
		30	0.90	0.231	−3.29		
Dawsonite	5	5	0.90	0.225	−3.01	1.82	17.3
		30	0.90	0.295	−3.44		
Thiamine hydrochloride							
Dawsonite	7	5	0.15	0.320	−3.20		
Celite 545	7	5	0.15	0.397	−3.32		

The values of n for thiamine hydrochloride were about one tenth as large as those for other vitamins, but there was not so large difference between $\Delta G_{app.}$ for the vitamins.

Contrary to ordinary cases of adsorption, the values of $\Delta H_{app.}$ and $\Delta S_{app.}$ for filter aid were positive.

Fig. 1 shows the adsorption isotherms of pyridoxine hydrochloride from pH 3.

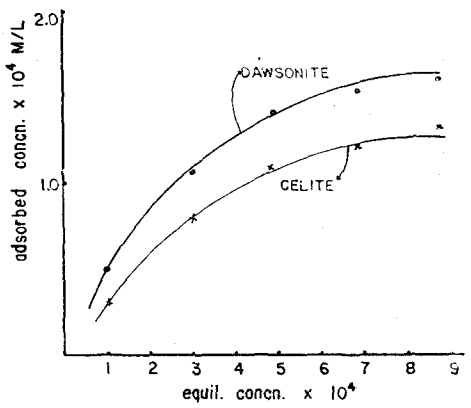

Fig. 1. Adsorption isotherms of pyridoxine hydroclhoride from pH 3 buffer solution by Celite 545 and Dawsonite at 5°C.

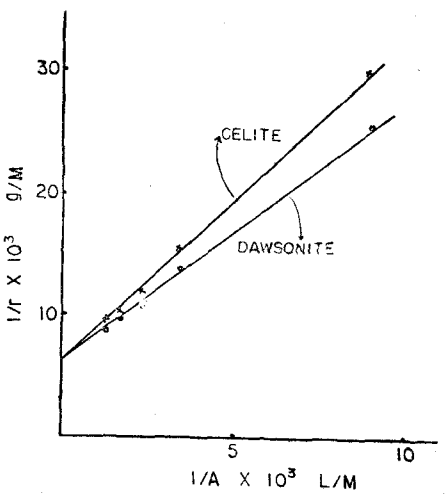

Fig. 2. The *Langmuir's* plot for adsorption of thiamine hydrochloride on Celite 545 and Dawsonite in pH 7 buffer solution at 5°C.

The adsorption tendency of vitamins by filter aids decreased with the temperature increase and the adsorption degree by Dawsonite was larger than that of the adsorption by Celite 545.

Adsorption of caffeine was scarecely observed in the presence of filter aids. Fig. 2-4 indicates the *Langmuir's* plot for adsorption of niacinamide and thiamine hydrochloride.

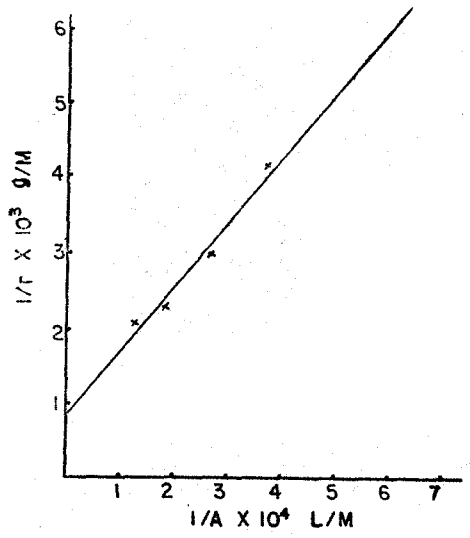

Fig. 3. The *Langmuir's* plot for adsorption of niacinamide on Celite 545 in pH 3 buffer solution at 5°C.

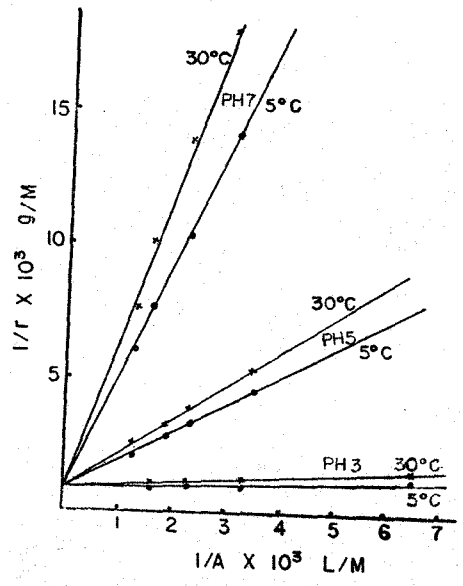

Fig. 4. The *Langmuir's* plot for adsorption of niacinamide on Dawsonite.

As shown in Fig. 5 the degree of the repression by filter aids was examined at various pH. The pH dependence of the repression was not so large in case of pyridoxine hydrochloride but the adsorption of niacinamide on Dawsonite sharply decreased with the pH increase.

CONCLUSION

The adsorption of caffeine, niacinamide, pyridoxine hydrochloride and thiamine hydrochloride on Celite 545 and Dawsonite from buffer solutions of pH 3, 5, and 7 was investigated at 5° and 30°. A definite adsorption occurred with all of the vitamins examined in Dawsonite

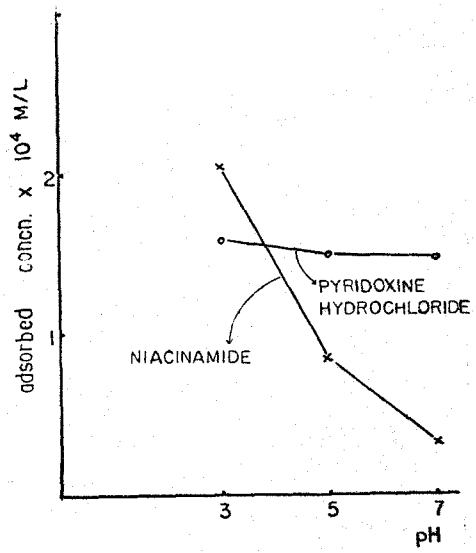

Fig. 5. Influence of pH on the adsorption of niacinamide and pyridoxine hydrochloride on Dawsonite.

solution, but the adsorption tendency by Celite 545 was small in comparison with Dawsonite.

The adsorption of caffeine was negligible. The adsorption isotherms apparently fitted the *Langmuir* equation and the adsorption tendency of vitamins by filter aids such as Celite 545 and Dawsonite decreased with the temperature increase.

REFERENCES

1. A.B. Cummins, *Ind. Eng. Chem.*, **34**, 398(1942)
2. A.J. Bruch, Jr. and J.P. Golightly, *Can. Min.*, **1**, 51-56(1968)
3. C.K. Huang and P.F. Kerr, *Am. Min.*, **45**, 311(1960)
4. S.W. Kwon and Ishikawa, *Bulletin of scientific engeneering research laboratory, Waseda university*, vol **41**, p-45 (1968)
5. A.B. Cummins and L.E. Weymouth, *Ind. Eng. Chem.*, **34**, 392(1942)
6. A.B. Cummins and M.S. Badollet., *Ind. Eng. Chem. Anal. Ed.*, **5**, 328(1933)