

## **Adsorption of Cationic Methylene Blue and Anionic Methyl Orange by Crude Drug Starches**

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**Abstract:** In order to use effectively the starch from the extracted residue of crude drugs, adsorption characteristics of cationic methylene blue (MB) and anionic methyl orange (MO) were investigated. Starches (S) were prepared from the roots of *Panax ginseng* C.A. Meyer (PG) and *Panax notoginseng* (Burk.) F.H. Chen (PN), the rhizomes of *Pinellia ternate* (Thunb.) Breitenbach (PT) and *Alisma orientale* Juzepczuk (AO), and the seeds of *Coix lacrymajobi* Linné var. *ma-yuen* Stapf (CL). The adsorption isotherm was evaluated using Akaike's information criterion (AIC) value, and the adsorption of cationic MB and anionic MO by the starches was classified into the Langmuir type and Freundlich type, respectively. The starches adsorbed more MB than MO. The amounts adsorbed were affected not by the particle size or the content of cationic minerals, but by the phosphorus content. Cationic MB was adsorbed more strongly than anionic MO on the negatively charged surface of the starches because of the presence of phosphorus. S-CL was superior in adsorption capacity for both cationic MB and anionic MO to S-PG, S-PN, S-PT, S-AO, and S-ST (potato starch) at 5 and 25°C. S-CL had a porous and stripe structure on the granular surface, and contained a small amount of phosphorus.

**Key words:** cationic methylene blue, anionic methyl orange, crude drug starch, applicability of adsorption equation, reuse of crude drug residue

Crude drugs are used as raw materials for powdered crude drugs or extracts at pharmaceutical factories. In 1988, the amounts of PG, PT, AO and CL consumed in Japan were 954, 670, 247 and 330 tons, respectively, an increase of approximately 3 to 7 fold compared to the year 1978.<sup>1)</sup> In 2002, the quantitative imports of PG, PT, AO and CL were 876, 250, 200 and 6972 tons, respectively.<sup>2)</sup> More consumption of crude drugs is estimated in the field of health foods or cosmetics. After a crude drug is extracted with a solvent, its residue now is reused as a compost or incinerated. The crude drugs PG, PN, PT, AO and CL contain 5.5–12.9% starch by weight.<sup>3)</sup> However, there has been almost no research on the adsorption of dyes by crude drug starches except for kudzu starch.<sup>4)</sup> The adsorption properties of crude drug starches were investigated in an attempt to make profitable the use of starch from the extracted residues.

The adsorption of dye by starch is important for the coloring of bean-jam cakes and starch confectioneries. The isotherm for the adsorption of anionic dyes by cereal starches<sup>5)</sup> and that of ferric chloride (cationic) by potato,

corn and cassava starches<sup>6)</sup> were found to fit the Langmuir equation. Both the Langmuir and Freundlich equations were applicable to the isotherm of anionic dye.<sup>4)</sup> However, the adaptability of an adsorption equation has been evaluated based on a partial or uniform weighting of the amount adsorbed and residual concentration.<sup>4–6)</sup> In the present study, a statistical analysis on the isotherm was performed using Akaike's information criterion (AIC) value<sup>7)</sup> to solve this problem.

The extent of the adsorption of anionic dyes by starches was related to the ratio of amylopectin to amylose<sup>5)</sup> or the amount of phosphorus content.<sup>4)</sup> In this study, the factors responsible for the difference in amounts of cationic methylene blue (MB) and anionic methyl orange (MO) adsorbed were discussed, and a crude drug starch superior in the capacity to adsorb both cationic and anionic dyes was identified.

### MATERIALS AND METHODS

**Materials.** The crude drug starches (S) of PG, PN, PT, AO and CL and potato starch (S-ST) were the same as those used previously.<sup>2)</sup> The starch was deproteinized by the method of Suzuki *et al.*<sup>8)</sup> and defatted by the method of Schoch.<sup>9)</sup> The starch was screened with 400 mesh or less and kept in tight containers. Reagent-grade MB and MO were obtained from Wako Pure Chemicals, Osaka, Japan. All other reagents were of the highest grade commercially available.

**Adsorption procedure.** About 150 mg of starch was weighed accurately and added to a hermetic glass con-

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Abbreviations: MB, methylene blue; MO, methyl orange; S, starch; PG, root of *Panax ginseng* C.A. Meyer; PN, root of *Panax notoginseng* (Burk.) F.H. Chen; PT, rhizome of *Pinellia ternate* (Thunb.) Breitenbach; AO, rhizome of *Alisma orientale* Juzepczuk; CL, seed of *Coix lacrymajobi* Linné var. *ma-yuen* Stapf; ST, potato; AIC, Akaike's information criterion; V, amount adsorbed; C, residual concentration;  $\Delta G^\circ$ , adsorption free energy.

tainer (30 mL) containing the dye solution (5.0 mL), and the slurry was incubated in a water bath shaker at 5 and 25°C for 30 min, since this was previously determined to be the time required to ensure equilibrium. The initial concentrations of the MB and MO solutions were 0.076, 0.153, 0.306, 0.458, 0.611, 0.916, 1.222, 1.833, 2.444 and 3.055 mmol/L. After the incubation, the suspension was centrifuged for 3 min at 3000 rpm, and the absorbance of MB and MO supernatant fluids was measured at 650 and 420 nm, respectively. The amount adsorbed was calculated from the decrease in absorbance of the supernatant, using a calibration curve.

**Statistical analysis.** A statistical analysis on the adsorption isotherm was performed using the nonlinear least-squares regression program, WinNonlin (Pharsight Co., Mountain View, CA, USA), to evaluate the adaptability of the Langmuir and Freundlich equations to the adsorption data. AIC<sup>7)</sup> was used to decide the adsorption equation that provided the best fit of the amount adsorbed ( $V$ )-residual concentration ( $C$ ) curve, without any weighting of  $V$  and  $C$ .  $AIC = n \ln SS + 2m$ ;  $n$  is the number of experimental data,  $SS$  is the sum of residual squares, and  $m$  is the number of parameters in the adsorption equation. The equation giving the lower AIC value was considered to be more applicable.<sup>7)</sup> The line of the isotherm was smoothed using the equation yielding the lower AIC value (Table 1).

**Particle size distribution.** The particle size distribution was determined with a laser diffraction particle size analyzer SALD-2100 (Shimadzu Seisakusho, Co., Ltd., Kyoto, Japan), after dispersing the starch in a 0.01% neutral detergent (Ekiserin, Kao Corporation) solution.

**SEM observation.** Starch was scattered on a double-faced tape stuck on the sample holder. After sputtering with gold ion using a cryounit, the sample was observed with an electron probe microanalyzer-8705 (Shimadzu

Seisakusho, Co., Ltd., Kyoto, Japan) at 10 kV of acceleration voltage at a magnification of 2000 times.

**Inorganic element.** Starch was decomposed with a nitric acid-perchloric acid solution according to the wet digestion method.<sup>10)</sup> The decomposed solution was diluted with ultra-pure water in a volumetric flask. Determination of P, Mg and Ca was performed with an ICPS-2000 sequential plasma spectrometer (Shimadzu Seisakusho, Co., Ltd., Kyoto, Japan). Starch was ashed at 500°C for 2 h in a porcelain crucible according to the dry combustion method.<sup>10)</sup> The ash was dissolved in a 1 M-hydrochloric acid solution. The solution was diluted with ultra-pure water in a volumetric flask. Determination of Na and K was performed with an AA-6800 atomic absorption-flame emission spectrophotometer (Shimadzu Seisakusho, Co., Ltd., Kyoto, Japan).

## RESULTS AND DISCUSSION

### Qualitative analysis of adsorption isotherm of MB and MO.

The adsorption data were analyzed statistically to elucidate more precisely the adsorption equation applicable to the isotherms. Usually, the applicability of the Langmuir equation ( $V = K_a V_m C / (1 + K_a C)$ ) or Freundlich equation ( $V = kC^{1/n}$ ) to an isotherm has been evaluated from the linearity of the plots of  $C/V$  vs.  $C$  or  $\log V$  vs.  $\log C$ , respectively.<sup>4-6)</sup> In the standard calculation,  $V$  in the Langmuir equation is partially weighted as  $C/V$ , and  $V$  and  $C$  in the Freundlich equation are uniformly weighted as logarithmic values.<sup>4-6)</sup> No weighting of  $V$  and  $C$  should be added to compare the adaptability of two equations under the same conditions. The present evaluation of the nonlinear equation with the AIC value solves this problem.<sup>7)</sup> Table 1 shows AIC values evaluated from a simultaneous curve. The isotherms of MB and MO could be fitted ( $p < 0.05$ ) to both the Langmuir and Freundlich equations. However, the adsorption of MB by all the starches except S-CL and the adsorption of MO by all except S-PN showed lower AIC values in the Langmuir equation and Freundlich equation, respectively. There were no significant differences in AIC values of the two model equations applied to the adsorption of MB by S-CL and the adsorption of MO by S-PN. Since the equation yielding a lower AIC value was considered to be more applicable,<sup>7)</sup> the isotherms of cationic MB and anionic MO for the starches were classified as the Langmuir type and Freundlich type, respectively.

The adsorption of anionic dyes by cereal starches was found to fit the Langmuir equation,<sup>5)</sup> being inconsistent with our results. It is not suitable to evaluate the applicability of an equation from data in a narrow range of residual concentrations up to 1.3 mmol/L,<sup>5)</sup> because the isotherm fits both the Langmuir and Freundlich equation in a narrow range of residual concentrations.<sup>11)</sup> The applicability in this study was evaluated using a wide range of residual concentrations up to 2.7 mmol/L (Figs. 1 and 2). That is, since the isotherms were obtained over a range of concentrations at which the MB and MO solutions are almost saturated, the present evaluation is more reliable.

The lines of isotherms of MB and those of MO were

**Table 1.** Comparison of AIC<sup>a)</sup> values of Langmuir (L) and Freundlich (F) equations applied to the isotherms for adsorption of methylene blue and methyl orange by starches at 25°C.

Starch	Adsorption of methylene blue		Adsorption of methyl orange	
	AIC of L eq.	AIC of F eq.	AIC of L eq.	AIC of F eq.
S-PG	19.2*	73.7**	22.7*	20.1*
S-PN	24.3*	33.3***	25.4*	26.5*
S-PT	61.7*	69.6*	60.8*	40.8*
S-AO	42.2*	50.4**	42.2*	32.4*
S-CL	42.5*	40.2*	82.0*	66.9*
S-ST	63.4*	92.1*	— <sup>b)</sup>	43.5*

<sup>a)</sup>Akaike's information criterion,  $AIC = n \ln SS + 2m$ ;  $n$ , number of experimental data;  $SS$ , sum of residual squares;  $m$ , number of parameters in the isotherm equation. <sup>b)</sup>Modeling unsuccessful. \* $p < 0.001$ , \*\* $p < 0.01$ , \*\*\* $p < 0.05$ . S-PG, starch from the root of *Panax ginseng* C.A. Meyer; S-PN, starch from the root of *Panax notoginseng* (Burk.) F.H. Chen; S-PT, starch from the rhizome of *Pinellia ternata* (Thunb.) Breitenbach; S-AO, starch from the rhizome of *Alisma orientale* Juzepczuk; S-CL, starch from the seed of *Coix lacryma-jobi* Linné var. *ma-yuen* Stapf; S-ST, starch from the rhizome of *Solanum tuberosum* Linné.

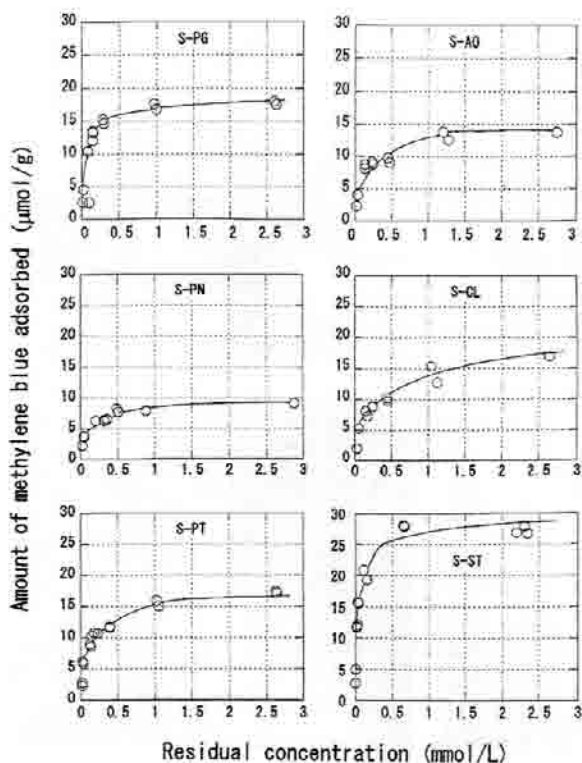


Fig. 1. Isotherms for adsorption of methylene blue by starches at 25°C.

For abbreviations of starches see Table 1. The lines were smoothed using the Langmuir equation yielding the lower AIC value in Table 1.

smoothed using the Langmuir and Freundlich equation, respectively, in Figs. 1 and 2. The adsorption of MB by crude drug starches and potato starch (S-ST) corresponds to the formation of a unimolecular layer (Fig. 1). The shapes of the curves for the adsorption of MO may be explained by the formation of several physically adsorbed layers (Fig. 2).

**Amounts of cationic MB and anionic MO adsorbed by crude drug starches.**

The amounts adsorbed were measured to compare the adsorption capacities of five kinds of crude drug starches (Tables 2 and 3). The constants,  $V_m$  (Table 2) and  $k$  (Table 3), show the amount adsorbed by the monolayer (Fig. 1) and the amount adsorbed at a residual concentration, 1.0 mmol/L (Fig. 2), respectively. The amounts of

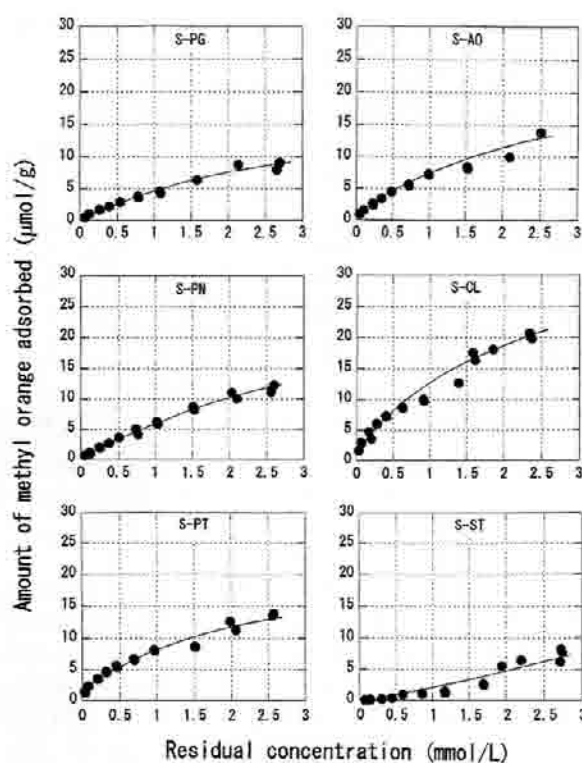


Fig. 2. Isotherms for adsorption of methylene orange by starches at 25°C.

For abbreviations of starches see Table 1. The lines were smoothed using the Freundlich equation yielding the lower AIC value in Table 1.

MB adsorbed ( $V_m$ ) by the starches at 25°C were 8.39 to 18.14  $\mu\text{mol/g}$ , being lower than the amount adsorbed (27.41  $\mu\text{mol/g}$ ) by S-ST (potato starch). The amounts of MO adsorbed ( $k$ ) by the starches at 25°C were 4.45 to 11.87  $\mu\text{mol/g}$ , being 1/4 to 4/5 of the amounts of MB adsorbed at 1.0 mmol/L in Fig. 1.

Sago starch adsorbed more MO than MB<sup>12)</sup> according to an evaluation of color (%), completely contradicting our results. Schoch and Maywald<sup>13)</sup> showed that potato starch adsorbed cationic dyes strongly but did not adsorb anionic dyes. Since the crude drug starches adsorbed more MB than MO (Figs. 1 and 2, Tables 2 and 3), it is suggested that the surfaces of the starches are negatively charged.<sup>13,14)</sup>

The amounts of MB adsorbed were larger at 5°C than at 25°C, and the differences ( $\Delta V_m$ ) in amounts adsorbed by S-PT and S-CL at between 5°C and 25°C were greater

Table 2. Langmuir constants and apparent adsorption free energy for methylene blue at 25°C and 5°C.

Starch*	Adsorption at 25°C			Adsorption at 5°C		
	$V_m$ ( $\mu\text{mol/g}$ )	$K_a$ (1/mmol)	$\Delta G^\circ$ (kcal/mol)	$V_m$ ( $\mu\text{mol/g}$ )	$K_a$ (1/mmol)	$\Delta G^\circ$ (kcal/mol)
S-PG	18.14	17.41	-5.78	19.32	22.78	-5.54
S-PN	8.39	16.33	-5.74	9.24	15.25	-5.32
S-PT	16.73	9.77	-5.44	21.96	3.96	-4.58
S-AO	13.08	12.35	-5.58	16.42	8.01	-4.96
S-CL	16.69	5.01	-5.04	23.61	2.21	-4.25
S-ST	27.41	35.29	-6.21	28.16	37.57	-5.82

\*For abbreviations see Table 1.  $V_m$  and  $K_a$ , Langmuir constants;  $\Delta G^\circ$ , apparent adsorption free energy.



**Table 3.** Freundlich constants of adsorption of methyl orange by starches at 25°C and 5°C.

Starch*	Adsorption at 25°C		Adsorption at 5°C	
	$k^{**}$ ( $\mu\text{mol/g}$ )	$n$	$k^{**}$ ( $\mu\text{mol/g}$ )	$n$
S-PG	4.45	1.41	7.02	1.21
S-PN	5.97	1.33	7.96	1.37
S-PT	8.09	1.92	8.51	1.92
S-AO	6.83	1.52	9.48	1.31
S-CL	11.87	1.66	13.31	1.62
S-ST	1.33	0.57	3.28	0.82

\*For abbreviations see Table 1. \*\*The value of  $k$  shows the amount of methyl orange adsorbed at a residual concentration, 1 mmol/L in Fig. 2.

than those for other starches (Table 2).  $K_a$ , a constant in the Langmuir equation, is a measure of the partitioning of the dye molecule and water molecule between the surface and the bulk solution.<sup>15)</sup> Negative correlations were found ( $p < 0.05$ ) between  $\Delta V_m$  and  $K_a$  at 5°C or 25°C. The smaller the  $K_a$  value, the higher the porosity of adsorbent.<sup>16)</sup> Since the diffusion and adsorption of adsorbate are affected by adsorption temperature in the pores rather than on the plain surface<sup>17)</sup>, it is suggested that S-PT and S-CL with small  $K_a$  value (Table 2) have a porous structure.

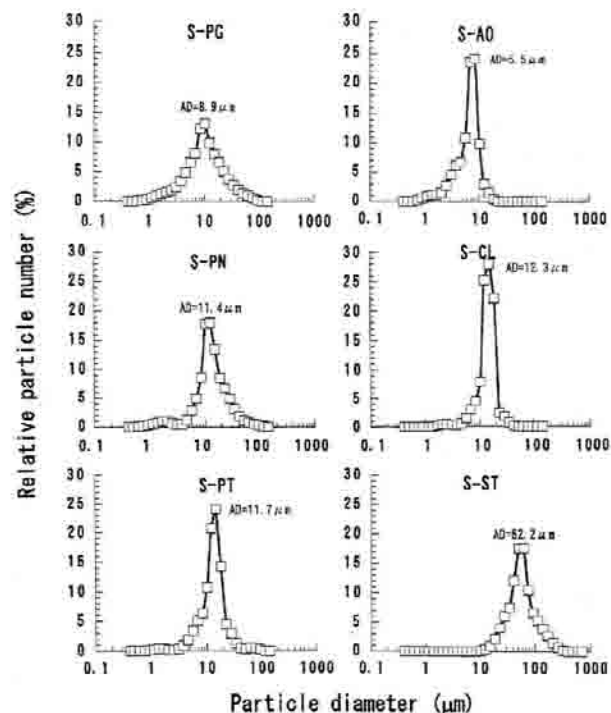
The apparent adsorption free energy ( $\Delta G^\circ$ ) was obtained using the following equation:  $\Delta G^\circ = -RT \ln K_a$  ( $R$ , gas constant;  $T$ , absolute temperature). The  $\Delta G^\circ$  values in Table 2 indicate that MB is physically adsorbed<sup>18)</sup> by the crude drug starches. Crini proposed that the mechanism by which dyes were taken up by starches was physical adsorption from the magnitude of the  $\Delta G^\circ$  value.<sup>19)</sup> Based on  $\Delta G^\circ$  values at 25°C and 5°C, the starches ranked in decreasing order as follows: S-ST, S-PG, S-PN, S-AO, S-PT and S-CL. The  $\Delta G^\circ$  value was larger at 25°C than 5°C. The larger the  $\Delta G^\circ$  value, the stronger the binding force of the dye.<sup>19)</sup> MB was adsorbed more strongly by S-ST (potato starch) than by the crude drug starches. It is considered that less MB is adsorbed at 25°C than at 5°C, and that the binding force of MB is a little stronger at 25°C than at 5°C.

The amounts of MO adsorbed were larger at 5°C than 25°C (Table 3). The differences (0.42–2.65  $\mu\text{mol/g}$ ) in amount of MO adsorbed ( $k$ ) due to the difference in temperature were as much as those (0.76–2.50  $\mu\text{mol/g}$ ) in amounts of MB adsorbed at 1.0 mmol/L in Fig. 1.

S-PG, S-PT and S-CL adsorbed cationic MB more than S-PN and S-AO at 5 and 25°C (Table 2). On the other hand, S-CL adsorbed anionic MO more than the other crude drug starches at 5 and 25°C (Table 3). It was concluded that S-CL was, therefore, superior in its capacity to adsorb both cationic MB and anionic MO to the other four crude drug starches.

#### Relationship between physicochemical properties of starch and amounts adsorbed.

The particle size distributions of the starches were measured to elucidate the relation between the amounts adsorbed and particle size (Fig. 3). The average diameters

**Fig. 3.** Particle size distribution of starches.

For abbreviations of starches see Table 1. AD, average diameter.

of S-PG, S-PN, S-PT, S-AO and S-CL granules were 8.9, 11.4, 11.7, 5.5 and 12.3  $\mu\text{m}$ , respectively. Their granular sizes closely corresponded with those from the observation of SEM (Fig. 4).

An apparent positive correlation ( $p < 0.05$ ,  $n = 6$ ) was found between the  $V_m$  of MB and average diameter (Table 5), whereas the correlation coefficient for 5 crude drug starches other than S-ST (potato starch) was 0.060. The result indicates that two variables of S-ST are largely localized in this distribution. It was, therefore, concluded that the  $V_m$  of MB was not correlated with the average diameter. There was no correlation between the  $k$  of MO and average diameter (Table 5). The saturated amounts of ferric chloride adsorbed by 4 kinds of starches were not correlated to particle size.<sup>5)</sup> There was no correlation between amounts of MO adsorbed by 7 kinds of starches and their specific surface area,<sup>3)</sup> which were measured using the nitrogen adsorption method. It is suggested that the amounts adsorbed are not dominated by the surface area of the crude drug starch.

S-PG, S-PN, S-PT, S-AO and S-CL granules were broken into a few pieces when they were prepared, and they display irregular shapes (Fig. 4). The angulate and wrinkled shapes like rice starch<sup>20)</sup> and oval and spherical shapes were observed in the S-PG and S-PN granules. The small holes on spherical surface and the stripe structure on fractured sections of S-CL granules were observed, whereas no holes were observed on the surface of S-PT granules.

The contents of phosphorus and cation minerals (Na+K+Mg+Ca) were analyzed to clarify the contribution of mineral contents to the amounts adsorbed (Table 4). S-ST (potato starch) contained the largest amounts of phosphorus (590  $\mu\text{g/g}$ ) and cation minerals (459  $\mu\text{g/g}$ ). S-PG contained a higher amount of phosphorus (139  $\mu\text{g/g}$ ) and the smallest amount of cation minerals (95.6  $\mu\text{g/g}$ ). S-

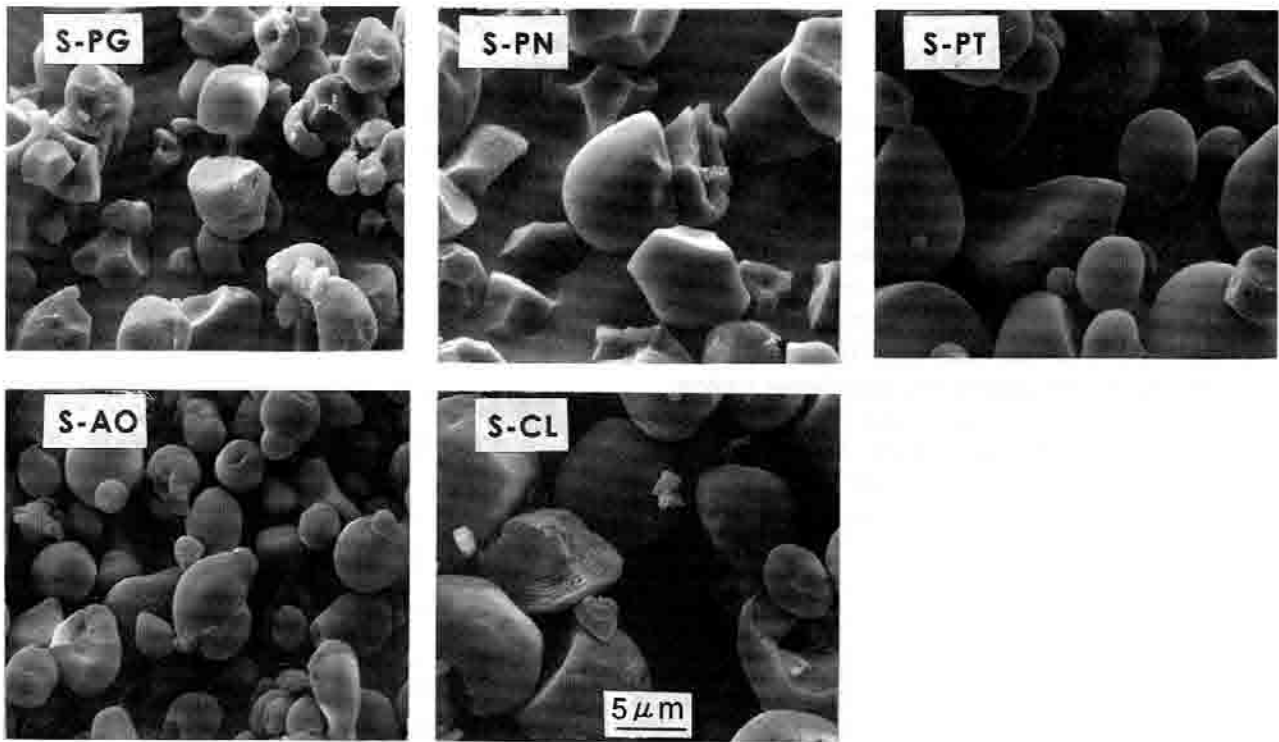


Fig. 4. Scanning electron micrographs of starches.

For abbreviations of starches see Table 1.

Table 4. Phosphorus and cation contents in starches.

Starch*	Mineral content ( $\mu\text{g/g}$ )					
	P	Na	K	Mg	Ca	Na+K+Mg+Ca
S-PG	139	9.3	1.3	57	28	95.6
S-PN	57	60	1.4	33	110	204.4
S-PT	94	12	29	40	58	139
S-AO	67	24	23	25	35	107
S-CL	57	13	53	53	130	249
S-ST	590	29	300	59	71	459

\*For abbreviations see Table 1.

PN, S-PT, S-AO and S-CL contained phosphorus, 57–94  $\mu\text{g/g}$  and cation minerals, 107–249  $\mu\text{g/g}$ .

There was a positive correlation ( $p < 0.05$ ) between the  $V_m$  of MB and phosphorus content, and a negative correlation ( $p < 0.05$ ) between the  $k$  of MO and phosphorus content (Table 5). It was, therefore, confirmed that one of the factors responsible for the difference in amounts of cationic and anionic dyes adsorbed by the starches was phosphorus content. The amounts of cationic butyric acid and anionic chlorogenic acid adsorbed by 4 kinds of starches were correlated positively ( $r = 0.947$ ,  $p < 0.05$ ) and negatively ( $r = -0.974$ ,  $p < 0.05$ ), respectively, to phosphorus content,<sup>6</sup> from the results of our calculation. It is suggested that phosphorus induces the adsorption of MB, and interferes with the adsorption of MO. The potato starch which was not defatted adsorbed no MO,<sup>5</sup> the defatted potato starch, however, adsorbed a small quantity of MO (S-ST in Fig. 2). S-CL which adsorbed more anionic and cationic dyes than the other starches, contained a small amount of phosphorus. The fact that S-CL adsorbed

Table 5. Correlations between amount adsorbed and mineral content of starches.

Amount adsorbed	Mineral content	
	Content of phosphorus* <sup>1</sup>	Content of cation minerals (Na+K+Mg+Ca)* <sup>1</sup>
$V_m$ of MB* <sup>2</sup>	0.873**	0.681
$k$ of MO* <sup>3</sup>	-0.814**	-0.411

\*<sup>1</sup>Table 4, \*<sup>2</sup> $V_m$  at 25°C of Table 2, \*<sup>3</sup> $k$  at 25°C of Table 3, \*\* $p < 0.05$ .

a larger amount of MB in spite of the small content of phosphorus may be affected by the structure of spherical surface and fractured section of granules.

There were no correlations between the sum of cationic minerals (Na+K+Mg+Ca) and  $V_m$  of MB or  $k$  of MO (Table 5). If the cationic minerals contribute electrostatically to the amount adsorbed by the charged dyes,<sup>12</sup> the amounts of cationic MB and anionic MO adsorbed are estimated to be correlated negatively and positively, respectively, with the sum of cationic minerals. The experimental results were completely unexpected (Table 5). The correlation between the amount of anionic chlorogenic acid adsorbed by 4 kinds of starches and the sum of cationic minerals (Fe+K+Mg+Ca)<sup>5</sup> was calculated by us; however, no correlation between them could be found. It is, therefore, concluded that the cationic minerals did not affect the adsorption of dyes. It was suggested that the amounts adsorbed were affected not by the surface area or the content of cationic minerals, but mainly by the phosphorus content.

## CONCLUSIONS

The isotherms for the adsorption of MB and MO by 5 crude drug starches were measured at 5 and 25°C in an attempt to use more effectively the starches from extracted residues. The adsorption isotherms of cationic MB and anionic MO were classified using the AIC value into the Langmuir type and Freundlich type, respectively. MB and MO were physically adsorbed by the starches. The starches adsorbed more MB than MO. The amounts adsorbed were mainly affected by the phosphorus content. S-CL was superior in adsorption capacity for both cationic and anionic dyes to S-PG, S-PN, S-PT, S-AO and S-ST (potato starch). S-CL had a porous and stripe structure and contained a small amount of phosphorus.

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### 生薬澱粉によるカチオン性メチレンブルーと アニオン性メチルオレンジの吸着

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生薬の抽出残渣に含まれている澱粉を有効利用するため、カチオン性メチレンブルー (MB) とアニオン性メチルオレンジ (MO) の吸着特性を調べた。澱粉 (S) は、*Panax ginseng* C.A. Meyer の根、ニンジン (PG)、*Panax notoginseng* (Burk.) F.H. Chen の根、サンシチ (PN)、*Pinellia ternate* (Thunb.) Breitenbach の塊茎、ハンゲ (PT)、*Alisma orientale* Juzepczuk の根茎、タクシャ (AO)、*Coix lacrym-jobi* Linné var. *ma-yuen* Stapf の種子、ヨクイニン (CL) から調製した。吸着等温線は AIC 値を用いて評価した (Table 1)。その結果、カチオン性 MB は Langmuir 型 (Table 1, Fig. 1)、アニオン性 MO は Freundlich 型 (Table 1, Fig. 2) に分類された。澱粉は、MO より MB を多く吸着した (Table 2, 3)。吸着量に影響を及ぼす要因は、粒子径やカチオン性無機質の含量でなく、リン含量であった (Table 5)。カチオン性 MB がアニオン性 MO よりも澱粉表面に強く吸着したが、それは含有リンの陰イオン性のゆえであった。ヨクイニン澱粉 (S-CL) は、5°C と 25°C いずれの温度でも、カチオン性 MB とアニオン性 MO の両方に対し、S-PG, S-PN, S-PT, S-AO, S-ST (バレイショ澱粉) より、優れた吸着力を示した (Table 2, 3)。S-CL は、その表面に多孔性ならびに縞状構造が観察され (Fig. 4)、リン含量は低かった (Table 4)。