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The Reactivity of Tetradecene with Mica Varieties of Different Particle Sizes and Characteristics of Mica Modified with Tetradecene

Tsuneo SUHARA*, Kyoko NAGAYA**, Fukuji SUZUKI* and Hiroshi FUKUI*

Abstract

Mica was modified with a tetradecyl group, and the products were characterized in various respects. 1,3,5,7-Tetramethylcyclotetrasiloxane (TMCTS) was deposited on mica at 80°C by chemical vapor deposition and was polymerized catalytically to form a poly(methylsiloxane) layer on the surface. Mica coated with TMCTS was prepared in four varieties with different mean particle sizes. The coated amount of TMCTS and the amount of Si-H groups decreased with increasing mean particle size. Mica modified with a tetradecyl group was synthesized from silicone coated mica by hydrosilylation with 1-tetradecene. The added amount of tetradecyl groups decreased with increasing mean particle size. The water contact angles and the yield value and the viscosity of disperse systems of mica modified with the tetradecyl group were dependent on the added amount of tetradecyl groups.

Key-words: *Chemical vapor deposition, Hydrosilylation, 1,3,5,7 - Tetramethylcyclotetrasiloxane, Tetradecyl group, Viscosity*

1. Introduction

We recently reported that powders could be coated with polymerized 1,3,5,7-tetramethylcyclotetrasiloxane (TMCTS) by chemical vapor deposition (CVD)¹⁾. The surface of the powders thus obtained did not have catalytically active sites and appeared to be inert. The thickness of the poly(methylsiloxane) (PMS) layer was approximately 0.5 to 1.5 nm, which corresponds to that of a monolayer structure.

Furthermore, the active Si-H group present in PMS can react with various functional groups by hydrosilylation²⁻⁹⁾. The dispersibility of titanium dioxide modified with olefins⁴⁾ or alcoholic hydroxyl groups^{5,8)} differed from that of the original titanium dioxide. Powder modified with ionic diethylamino groups could be utilized as a novel anionic exchanger²⁾. Silica gel modified with diethylamino groups or octadecyl groups by the hydrosilylation method showed strong stability in alkaline solution because of the surface hydrophobicity^{2,6)}. Fine silica powder modified with a quaternary ammonium group using *N,N*-dimethyl-*n*-alkyl amine changed gradually from hydrophilic to hydrophobic with an increasing number of carbon atoms in the alkyl chain⁷⁾. The alkyl chain of the quater-

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* Shiseido Cosmetic Product Research Laboratories, 1050 Nippa-cho, Kohoku-ku, Yokohama 223-8553, Japan.

** Shiseido Basic Research Laboratories, 1050 Nippa-cho, Kohoku-ku, Yokohama 223-8553, Japan.

nary ammonium group on the surface was presumably oriented outward⁷⁾. The zeta potentials of this powder as a function of pH were constantly positive over the range of pH 2 - 10, and no isoelectric point was observed¹⁰⁾. The adsorption behavior of an anionic surfactant on this powder was different depending on the number of carbons in the alkyl chain¹¹⁾. This powder showed bactericidal activity against gram positive¹²⁾. As previously mentioned, the onset of the effect of the powder modified with the functional group required a greater added amount of the functional group. In other words, the surface of the powder needed a greater added amount of the active Si-H groups for hydrosilylation. The thickness of the surface coating layer of TMCTS on the powder became more

In general, particles range from small to large, and the plate-shaped particle, which changes in aspect ratio with mean particle size, differs in surface potential between the edge part and the flat face part¹³⁾. The silicone structure and the property of the coated surface on the powder may be influenced by these characteristics. This study selected the mica of the plate-shaped particle, and this mica was classified into four varieties with different mean particle sizes. These four varieties of mica were modified by the TMCTS coating (CVD method). The products were subjected to a subsequent reaction with 1-tetradecene by hydrosilylation. The reactivity and other characteristics (contact angle and viscosity behavior) of the modified

mica will be discussed.

2. Experimental

Chemicals

Mica was of cosmetic grade. The reagents used were as follows: TMCTS (specific gravity: 0.992) (Shin-Etsu Kagaku Co., Ltd.), 1-tetradecene (Mitsubishi Chemical Co., Ltd.), and chloroplatinic(IV) acid hexahydrate($\text{H}_2\text{PtCl}_6 \cdot 6\text{H}_2\text{O}$) (N. E. Chemcat Co., Ltd.). Squalane (viscosity: 18 mPa s, SQ) was of cosmetic grade. The water used in this study was purified through a Milli-Q Reagent water system. Micas of the four varieties with different mean particle sizes (Mica- m , $m=A, B, C$, and D) were prepared by classification (Nisshin Engineering Co., Ltd.). **Table 1** shows the mean particle sizes and the specific surface area of the four varieties of mica.

Reaction

Because the amount of TMCTS coated on the powder was influenced by the specific surface area, the added amount of TMCTS was controlled. 300 g of Mica-A to D were coated with polymerized TMCTS. The supplied amount of TMCTS was 2 mg m^{-2} (low, L-type) or 5 mg m^{-2} (high, H-type). CVD was carried out in a small reactor held at 80°C for 16 hours. The product (L-type, LP m or H-type, HP m ; $m=A, B, C$, or D) was dried at 120°C for 4 hours under atmospheric pressure to remove excess TMCTS.

The scheme of the hydrosilylation procedure is shown in **Figure 1**. A mixture of 100 g of LPA-LPD, 4.9 g, 4.7 g, 4.0 g, or 3.2 g of 1-tetradecene, or that of 100 g of HPA-HPD, 18.

Table 1 Properties of Mica-A to Mica-D.

Specimen	Mean particle size / μm	Specific surface area / $\text{m}^2 \text{g}^{-1}$	Specific volume / $\text{cm}^3 \text{g}^{-1}$
Mica-A	6.2	8.7	2.45
Mica-B	9.4	7.2	2.10
Mica-C	13.0	6.6	1.85
Mica-D	18.4	4.5	1.65

3 g, 14.7 g, 12.2 g, or 6.0 g of 1-tetradecene, 150 cm³ of ethanol, and 2.5 mg of chloroplatinic(IV) acid hexahydrate was reacted at 60°C for 4 hours. The LPA-LPD or HPA-HPD derivative containing the tetradecyl group (LC m or HC m ; m =A,B,C, or D) was washed with ethanol to remove excess 1-tetradecene and the catalyst and then dried at 110°C for 16 hours *in vacuo*.

Measurements

Infrared spectra were obtained by a Fourier-transform infrared spectrophotometer FTIR-8100M (Shimadzu Co., Ltd.) using a diffuse reflectance method with 4 cm⁻¹ resolution in the range of 4000 to 1100 cm⁻¹.

Samples for elemental analysis were prepared by drying at 120°C for 4 h, and the analysis was carried out by a CHN2400 elemental analyzer (Perkin Elmer Co., Ltd.).

The amount of Si-H group was measured by the following method. About 5 g of LPA-D or HPA-HPD was suspended in 20 cm³ of ethanol at 25°C with stirring, and 5 cm³ of 10% sodium hydroxide solution was added. Five minutes later, the volume of hydrogen that had been evolved was measured with a buret. The amount of Si-H groups, N_H (mmol g⁻¹), was calculated by eq. 1;

$$N_H = PV / 0.08205 TW \quad (1)$$

where P is pressure (atm), V is volume of hydrogen (cm³), T is temperature (K), and W is sample weight (g). The cross-linking ratio of the PMS films on the mica was evaluated by eq. 2;

Crosslinking ratio (%) = $(1 - N_H / N_o) \times 100$ (2)
where N_o is the amount of TMCTS obtained

Fig. 1 Scheme of hydrosilylation.

from the elemental analysis (mmol g⁻¹) (eq. (1)).

The thickness of the polymerized TMCTS coating on LPA-LPD or HPA-HPD was calculated by eq. 3;

$$\text{Thickness (layer)} = S S_M / N_o A v \quad (3)$$

where $A v$ is Avogadro's number, S_M is the molecular occupied area of $-(H)Si(CH_3)_2O-$ ($S_M = 0.13 \text{ nm}^2$), and S is the specific surface area (m² g⁻¹).

Water contact angle was measured by the following method. Pellets were prepared by a press of the powders at 750 kg cm⁻² and dried in an oven at 100°C for 16 hours *in vacuo*. Water contact angle was measured using an automatic contact angle apparatus CA-Z (Kyowa Kaimen Kagaku Co., Ltd.).

Viscosity-shear rate measurement was performed at 25°C using a CSL2 100 (Carri-Med Co., Ltd.) controlled stress rheometer, provided with 4 cm diameter parallel plates. The volume fraction of the dispersed powder was 0.5.

3. Results and Discussion

1. Mica covered with TMCTS

Figure 2 shows the diffuse reflectance infrared spectra of the original Mica-A and HPA. The absorption peaks of the C-H asymmetric stretching vibration, the Si-H stretching vibration, the C-H asymmetric deformation vibration, and the C-H symmetric deformation vibration were observed at 2970, 2160, 1408, and 1265 cm⁻¹, respectively. The surface of the mica was found to be covered with the polymerized TMCTS. The intensities of these peaks, which decreased from HPA to D, were greater than those for LPA-LPD.

The characteristics of LPA-LPD and HPA-HPD are shown in **Table 2** in which the amount of the polymerized TMCTS on the surface has been converted to TMCTS. The coated amounts of TMCTS and the amounts of the Si-H group on LPA-LPD were 250-108 $\mu\text{mol g}^{-1}$ and 124-82 $\mu\text{mol g}^{-1}$, respectively. The

coated amounts of TMCTS and the amounts of the Si-H group on HPA-HPD were 621-216 $\mu\text{mol g}^{-1}$ and 467-154 $\mu\text{mol g}^{-1}$, respectively. The coated amounts of TMCTS and the amounts of the Si-H group decreased with increasing mean particle size. The thickness of the polymerized TMCTS on LPA-LPD or HPA-HPD was therefore calculated by considering the specific surface area. The thickness of the polymerized TMCTS coating on LPA-LPD was consistently 2 - 2.5 layers and

Fig. 2 Diffuse reflectance infrared spectra of mica.
(a) Mica-A and (b) HPA

was independent on the mean particle size of the mica. On the other hand for HPA-HPD, when the mean particle size of the mica increased, the thickness of the polymerized TMCTS coating decreased from 5.6 layers to 3.8 layers. The coated amount of TMCTS was found to increase with decreasing mean particle size. Furthermore, the structure of the polymerized TMCTS coating could be estimated by the crosslinking ratio. The crosslinking ratio for LPA-LPD and HPA-HPD ranged between 25-50% and 22-28%, respectively. The crosslinking ratio of LPA-LPD decreased with increasing mean particle size of the mica. In the case of LPD, the network polymer of the polymerized TMCTS was thought to be less linear compared with LPA-LPC. The crosslinking ratio of HPA-HPD was independent of the mean particle size at a consistent value of about 25%. It is known that the crosslinking ratio, which is very high in the initial stage of the crosslinking reaction, decreases with further supply of TMCTS^{1,8,14}. As previously mentioned, the structure of the polymerized TMCTS coating of LPA-LPC was thought to be different from that of LPD. Because the mica is a plate-shaped particle, the mean particle size of the mica will depend on the ratio of S_F/S_E (S_F is the surface area of the flat face and S_E is the surface of the edge). The ratio of S_F/S_E increased with increasing

Table 2 Characterization of LPA-LPD and HPA-HPD.

Specimen	Coated amount of TMCTS / $\mu\text{mol g}^{-1}$	Residual amount of Si-H group / $\mu\text{mol g}^{-1}$	Crosslinking ratio / %	Thickness / layer	Water contact angle / deg
LPA	250	124	50	2.3	125
LPB	242	121	50	2.6	125
LPC	179	104	42	2.1	124
LPD	108	82	25	1.9	125
HPA	621	467	25	5.6	121
HPB	520	375	25	5.4	123
HPC	400	313	22	4.8	123
HPD	216	154	28	3.8	122

mean particle size. At a low amount of TMCTS supplied this ratio was influenced by the structure of the polymerized TMCTS, but at a high amount of TMCTS supplied it was thought to be influenced by the thickness. The water contact angles on LPA-LPD and HPA-HPD were very high, about 125° , and were independent of the coated amount of TMCTS and the mean particle size. This is because the surface of LPA-LPD and HPA-HPD contains the methyl group of the polymerized TMCTS, and the surface tension of the methyl group was very low.

2. LPA-LPD and HPA-HPD modified with tetradecene

Figure 3 shows the diffuse reflectance infrared spectra of HCA and HPC. The absorption peaks of the C-H symmetric stretching vibration of the tetradecyl group were observed at 2960, 2923, 2854, and 1465 cm^{-1} . The intensities of these absorption peaks, which decreased from HCA to D were greater than those of LCA-LCD. The peak of the Si-H symmetric stretching vibration was observed

Fig. 3 Diffuse reflectance infrared spectra of mica: (a) HPA and (b) HCA.

at 2170 cm^{-1} even after hydrosilylation of HCA-HCD. However, the peak intensity of HCA-HCD at 2170 cm^{-1} was less than that of HPA-HPD. This phenomenon occurs from the fine titanium dioxide modified with alcoholic hydroxyl group appearing to retain the Si-H group after hydrosilylation⁵⁾. On the other hand, none of LCA-LCD exhibited the peak of the Si-H group.

The confirmation of the chemical bond between LPA-LPD or HPA-HPD and 1-tetradecene was examined using TG-DTA measurement. **Figure 4** shows the TG-DTA curves of HCA. TG-DTA curves were observed to have an exothermic peak with weight loss above 241°C . The intensities of those peaks which decreased from HPA to D were greater than those of LPA-LPD. The weight loss observed above 600°C was due to the dehydration of the interlayer in mica. On the other hand, the TG-DTA curve of the physically mixed mica covered with the polymerized TMCTS and glycerol- α -monoallylether was observed to have an endothermic peak with weight loss at 50°C . The endothermic peak was thought to be due to the physical adsorption between the glycerol- α -monoallylether and the mica covered with the polymerized TMCTS. For the mica modified with glycerol- α -monoallylether by hydrosilylation, an exothermic peak was ob-

Fig. 4 TG-DTA curves of HCA: solid line (DTA), broken line (TG).

served; the surface of the mica covered with the polymerized TMCTS was thought to be modified with glycerol- α -monoallylether by a chemical bond³⁾. As previously mentioned, for the TG-DTA curves of LCA-LCD and HCA-HCD in which the exothermic peak was the same as that of the mica modified with glycerol- α -monoallylether, the surface of LPA-LPD and HPA-HPD was thought to be modified with 1-tetradecene by a chemical bond.

Table 3 shows the characteristics of LCA-LCD and HCA-HCD. The added amount of the tetradecyl group of LCA-LCD and HCA-HCD decreased with increasing mean particle size. That tendency was the same as that of the coated amount of TMCTS, because the added amount of the tetradecyl group depended on the specific surface area (the coated amount of TMCTS and the residual amount of Si-H groups) of the mica. The occupied surface area of the tetradecyl group on LCA-LCD, by considering the specific surface area, was consistently about 0.33 nm². On the other hand, that of the tetradecyl group on HCA-HCD increased from 0.06 nm² to 0.11 nm² with increasing mean particle size. Furthermore, the reaction ratio of LCA-LCD and HCA-HCD were about 30% and 45%, respectively. The reaction ratio of HCA-HCD was greater than that of LCA-LCD, and the reaction ratio,

in general, decreased with increasing Si-H group due to the steric hindrance of the modified group. However, in this reaction, the orientation of the Si-H group between LPA-LPD and HPA-HPD was thought to be different with the amount of TMCTS supplied on the mica. The orientation of the Si-H group on LPA-LPD caused the reaction between the Si-H group and tetradecene to proceed with difficulty. This is because the polymerized TMCTS tightly coated the surface of the mica when present as a thin layer. In the case of HPA-HPD, the direction of the Si-H group allowed the reaction to proceed easily. In this case the thick layer of polymerized TMCTS loosely coated the surface of the mica.

Figure 5 shows the water contact angle of LCA-LCD and HCA-HCD. The water contact angle of LCA-LCD, which was consistently about 125°, and showed hydrophobicity, was independent of the coated amount of TMCTS, or the added amount of tetradecyl groups. Since the tetradecyl group moiety of LCA-LCD is orientated in a standing state so that the terminal methyl groups are present at the external surface, the water contact angles of LCA-LCD were very high, the same as that of LPA-LPD. On the other hand for HCA-HCD, where the water contact angle increased with increasing mean particle size, the

Table 3 Characterization of LCA-LCD and HCA-HCD.

Specimen	Added amount of tetradecyl group / $\mu\text{mol g}^{-1}$	Reaction ratio / %*	Occupied surface area / nm ²
LCA	43	35	0.33
LCB	34	28	0.35
LCC	31	30	0.35
LCD	24	29	0.32
HCA	235	50	0.06
HCB	184	49	0.06
HCC	121	41	0.08
HCD	70	46	0.11

* Added amount of tetradecyl group / Residual amount of Si-H ratio

tetradecyl group moiety was orientated at random, and the surface of HCA-HCD contained not only the methyl group and but also the ethylene chain. This occurred because the occupied surface area of the tetradecyl group in HCA-HCD was smaller than that of LCA-LCD. Because the surface tension of the methyl group was lower than that of the ethylene chain¹⁵⁾, the water contact angle of HCA-HCD was smaller than that of LCA-LCD. Furthermore, the appearance of the ethylene chain on the surface decreased with increasing mean particle size, and the water contact angle of HCA-HCD was thought to increase with increasing mean particle size.

3. Characteristics of LPA-LPD, HPA-HPD, LCA-LCD, and HCA-HCD in a disperse system

The dispersion evaluation of LPA-LPD, HPA-HPD, LCA-LCD, and HCA-HCD was done using viscosity measurements in a powder-oil disperse system. The volume fraction of the dispersed LPA-LPD, HPA-HPD, LCA-LCD, and HCA-HCD was 0.5. (The flow curves of SQ showed Newtonian flow.) **Figure 6** shows the flow curves in LCA-LCC and HCA-

Fig. 5 The relation ship between the water contact angle on LCA-LCD and HCA-HCD and the mean particle size: ○, LCA-LCD; ●, HCA-HCD.

HCD / SQ disperse systems. However, Mica-A to D could not be measured at this volume fraction because they could not be slurred, and the flow curve of LCD could not be exhibited on this scale because of the very high yield value. The flow curves for the LCA-LCD and HCA-HCD did not go through the origin of the coordinate axes, and the shear stress increased with increasing shear rate. Therefore, the flow behavior for LCA-LCC and HCA-HCD seems to be Casson flow. The flow curves of the LPA-LPC and HPA-HPC / SQ disperse system showed the same flow curves as the Casson flow in Figure 6. However, the flow curves of LPD, HPD, and LCD do not show the Casson flow. The yield value and the viscosity in the Casson equation are given by eq. 4 ;

$$\tau^{1/2} = \tau_0^{1/2} + \eta_\infty^{1/2} D^{-1/2} \quad (4)$$

where D (s^{-1}) is the shear rate, τ_0 (Pa) is the yield value, τ (Pa) is the shear stress, and η_∞ (Pa s) is the residual viscosity of suspension¹⁶⁾.

Figure 7 shows the yield value of the LPA-

Fig. 6 Casson plot of LCA-LCC (upper) (○, LCA; ●, LCB; □, LCC) and HCA-HCD (lower) (○, HCA; ●, HCB; □, HCC; ■, HCD) dispersed in SQ (The volume fraction of the dispersed powder was 0.5.)

LPC, HPA-HPC, LCA-LCC, and HCA-HCD / SQ dispersion systems (except for LPD, HPD, and LCD). The yield value between LPA-LPB and HPA-HPB was the same value, but that of LPC and HPC rapidly increased, and that of LPD and HPD was several hundred pascals. This tendency which was not influenced by the coated amount of TMCTS, depended on the mean particle size. The yield value of LCA-LCC, which increased with increasing mean particle size, decreased compared with LPA-LPC because the affinity between the tetradecyl group on LCA-LCC and SQ was greater than that between the methyl group on LPA-LPC and SQ. The yield value of HCD, which was higher than that of HCA-HCC, was influenced by the mean particle size. The yield value of HCA-HCD was lower than that of LCA-LCD, because the affinity between the tetradecyl group and SQ was influenced by the added amount of tetradecene. This can be explained by the decreasing coagulation force between particles due to the hydrophobic interaction between the tetradecyl group and SQ. The yield value is known to increase with increasing coagulation force between the dispersed particles. Gillespie¹⁷⁾ advanced the

yield value (τ_0) in eq. 5;

$$\tau_0 = K\phi^2 E \quad (5)$$

where ϕ is the volume fraction of the dispersed particles, E is the coagulation energy between particles, and K is the constant determined for the disperse system. The τ_0 was confirmed to vary in proportion to ϕ^2 ¹⁸⁾. When ϕ and K were constant values in this experimental system, the difference in the yield value between the LCA-LCC or HCA-HCD particles was presumed to be the difference in the coagulation energy between LCA-LCC or HCA-HCD particles. Furthermore, the plate-shaped particle, which is known to differ in surface potential between the flat face and the edge, formed the card-house structure in the suspension, and the yield value and the viscosity increase¹⁹⁾. However, the surface of LPA-LPD, HPA-HPD, LCA-LCD, and HCA-HCD coated with polymerized TMCTS was not influenced by the surface potential, and the increase in the yield value depended on the mean particle size.

Figure 8 shows the viscosity of the LPA-LPC, HPA-HPC, LCA-LCC, and HCA-HCD / SQ dispersion systems (except for LPD, HPD, and LCD). The viscosity among LPA-LPB, HPA-HPB, and LCA-LCD was the

Fig. 7 Plots of yield value against the mean particle size for LPA-LPC (○), HPA-HPC (●), LCA-LCC (□), and HCA-HCD (■) dispersed in SQ.

Fig. 8 Plots of viscosity against the mean particle size for LPA-LPC (○), HPA-HPC (●), LCA-LCC (□), and HCA-HCD (■) dispersed in SQ.

same value, but that of LPC, HPC, and LCC rapidly increased, because these viscosities, which were independent of the methyl group or the tetradecyl group, may be dependent on the mean particle size. On the other hand, the viscosity of HCA-HCC was consistently 0.05 Pa s, while that of HCD rapidly increased. The affinity between the tetradecyl group moiety of HCA-HCC and SQ was greater than that between the tetradecyl group moiety of LCA-LCC and SQ. This is because the added amount of 1-tetradecene on HCA-HCC was more than that of LCA-LCC, and the high viscosity of HCD depended on the mean particle size. Because the yield value and the viscosity of HCA-HCC were low, HCA-HCC were found to be easy to disperse in SQ.

4. Conclusion

At a low amount of TMCTS supplied, the coated amount of TMCTS decreased with increasing mean particle size, but the thickness of TMCTS on LPA-LPD, by considering the specific surface area, was consistently 2 – 2.5 layers. In contrast, at a high amount of TMCTS supplied, the thickness of TMCTS on HPA-HPD decreased with increasing mean particle size. This phenomenon may be influenced by the ratio between the surface area of the flat face and the edge.

The added amount of tetradecene on LCA-LCD and HCA-HCD decreased with increasing mean particle size. However, the occupied surface area of the tetradecyl group, by considering the specific surface area, was consistently about 0.33 nm² in LCA-LCD but it increased from 0.06 nm² to 0.11 nm² in HCA-HCD. These occupied surface areas were dependent on the residual Si-H group on contained in the polymerized TMCTS.

The water contact angles on LPA-LPD, HPA-HPD, and LCA-LCD, which were consistently about 125°, were independent of the mean particle size, the coated amount of

TMCTS, or the added amount of tetradecene. However, the water contact angle on HCA-HCD, which increased with increasing mean particle size was dependent on the added amount of tetradecene. The flow behavior of the powder – SQ disperse system showed Casson flow (except for LPD, HPD, and LCD). The viscosity and the yield value were independent of the coated amount of TMCTS and depended on the added amount of tetradecene and the mean particle size.

References

- 1) H. Fukui, T. Ogawa, M. Nakano, M. Yamaguchi, and Y. Kanda: *"Controlled Interphases in Composite Materials"*, Elsevier Science Publishing Co., New York, pp. 469–478 (1990).
- 2) T. Suhara, H. Kutsuna, H. Fukui, and M. Yamaguchi: *Colloid Polym. Sci.*, **271**, 486 (1993).
- 3) T. Suhara, H. Fukui, M. Yamaguchi, Y. Kawajiri, S. Kumagai, and M. Takahashi: *J. Soc. Cosmet. Chem. Jpn.*, **28**, 359 (1995).
- 4) T. Ogawa, T. Suhara, H. Fukui and M. Yamaguchi: *Preprints of Annual Technical Meeting, Shikizai Kyokai (Jpn. Soc. Colour Mater.)*, pp.170 (1990).
- 5) T. Suhara, H. Kutsuna, H. Fukui and M. Yamaguchi: *Shikizai Kyokaishi (J. Jpn. Soc. Colour Mater.)*, **65**, 264 (1992).
- 6) Y. Ohtsu, H. Fukui, T. Kanda, K. Nakamura, O. Nakano, and Y. Fujiyama: *Chromatographia*, **24**, 380 (1987).
- 7) T. Suhara, T. Kanemaru, H. Fukui, and M. Yamaguchi: *Colloids Surf.*, **95**, 1 (1995).
- 8) T. Suhara, H. Fukui, and F. Suzuki: *Shikizai Kyokaishi (J. Jpn. Soc. Colour Mater.)*, **68**, 597 (1995).
- 9) T. Suhara and H. Fukui: *Shikizai Kyokaishi (J. Jpn. Soc. Colour Mater.)*, **70**, 75 (1997).
- 10) T. Suhara, H. Fukui, and M. Yamaguchi: *Colloids Surf.*, **101**, 29 (1995).
- 11) T. Suhara, H. Fukui, and M. Yamaguchi: *Colloids Surf.*, **119**, 15 (1996).
- 12) T. Suhara, F. Shimano, Y. Sato, H. Fukui, and M. Yamaguchi: *Colloids Surf.*, **119**, 105 (1996).
- 13) A. W. Flegmann, J. W. Goodwin, and R. H. Ottewill: *Proc. Brit. Ceram. Soc.*, **13**, 31 (1969).
- 14) T. Suhara, H. Fukui, and K. Esumi: *Shikizai Kyokaishi (J. Jpn. Soc. Colour Mater.)*, **70**, 369

- (1997).
15) H. W. Fox, E. F. Hare, and W. A. Zisman: *J. Colloid Sci.*, **8**, 194 (1953).
16) T. Sato: *Shikizai Kyokaishi (J. Jpn. Soc. Colour Mater.)*, **68**, 441 (1993).
17) T. Gillespie: *J. Colloid Sci.*, **15**, 219 (1960).
18) D. Heath, Th. F. Tadros, P. K. Thomas, and R. P. Warrington: In "Advances in Pesticide Formulation Technology", H. B. Scher, Ed., ACS Symposium Series 254, American Chemical Society, Washington, DC, pp. 29-46 (1984).
19) S. Okuda: *Hyomen*, **4**, 2 (1966).

マイカのテトラデセンによる修飾反応に及ぼす粒子径の影響と修飾マイカの表面物性

須原常夫*・長屋京子**・鈴木福二*・福井 寛*

*資生堂製品開発センター 神奈川県横浜市港北区新羽町 1050 (〒223-8553)

**資生堂基盤研究センター 神奈川県横浜市港北区新羽町 1050 (〒223-8553)

要 旨

平均粒子径の異なるマイカに1, 3, 5, 7-テトラメチルシクロテトラシロキサン(TMCTS)を用いて化学気相蒸着法(CVD)によりシリコン被覆を行い, さらにこの粉末表面上にヒドロシリル化反応によりテトラデセン修飾を行い, 反応性およびその表面物性について検討を行った。

シリコン被覆量, Si-H 基量およびテトラデセン修飾量は, 平均粒子径が増加するにしたがって減少する傾向を示した。

水に対する接触角, 油に分散させた時の粘度および降伏値は, 平均粒子径が増加するにしたがって増加する傾向を示すことがわかった。