Application of sericite to LWC coatings

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ABSTRACT: Sericite is a mica mineral locally produced from the east coast of Taiwan. It carries high negative charges on its plate surfaces and has a high aspect ratio that might be useful for adjusting water retention values and the viscosity of coating colors. In this study, we partially or totally replaced delaminated clay in a lightweight coating (LWC) formulation with sericite and examined the resulting effects on the optical and printing properties of the coating colors and coated sheets. The water retention value (WRV) of the color decreased with increasing amounts of sericite substitution, reflecting an increase in water retention of the color by about 30%. Low shear viscosities decreased with increasing amounts of sericite substitution, while high shear viscosities showed a V-shaped response, suggesting an increase in the elasticity of the coating color under high shear conditions. Among the coated paper properties, the addition of sericite raised the opacity of the sheet. The brightness of the coated sheets, however, decreased linearly with increasing sericite substitution. When sericite was applied, notable swelling and delamination effects occurred during the preparation of the coating color.

Application: Sericite can effectively alter the properties of coating colors, such as water retention and low and high shear viscosities, and raise opacity (though also decreasing brightness), allowing mills to optimize costs and operation efficiencies of coating color formulations.

In pursuit of lighter grammage, lightweight coating (LWC) has gradually become the mainstay of advertising papers, with consumption increasing yearly. But mills often suffer because LWC products have insufficient opacity and caliper. To resolve these quality problems and to reduce production costs, mills have generally resorted to increasing the proportion of mechanical pulp in the blend, raising the proportion of clay in the coating color formulation (i.e., reducing the use of GCC), or using delaminated clay to replace #1 clay, for instance [1-6].

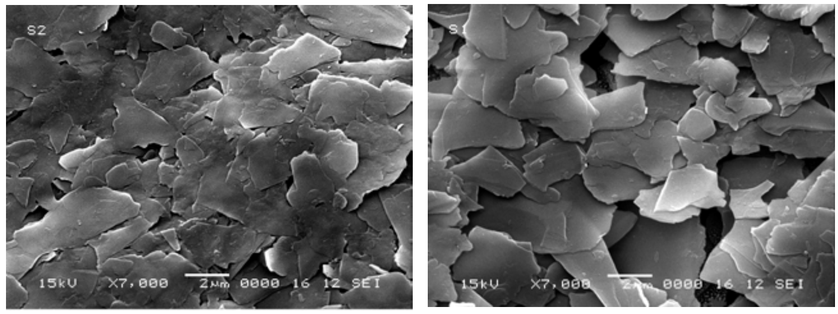
Sericite is a native mica mineral mined in eastern Taiwan. It is a lustrous layered powder composed mainly of silicon dioxide and aluminum oxide, with the empirical formula of $KAl_2(Si_3Al)O_{10}(OH)_2$. The average particle size of the mineral is about 10~14 µm. Typical sericite has a three-layer unit cell consisting of a SiO_2 tetrahedron-Al_2O_3 octahedron-SiO_2 tetrahedron. Between the layers are potassium ionic bonds which hold the layers together. Because the potassium ions are usually not hydrated, it has a relatively high ion exchange capacity (IEC) of about 5 meq/g and is potentially capable of exfoliating or delaminating. However, it is unlikely to delaminate merely through absorbing water and swelling [7-12]. Kaolin clay, on the other hand, is a nonswelling layered mineral [13-15] with two-layer unit cells of a SiO_2 tetrahedron-Al_2O_3 octahedron and an interlayer distance of 0.715 nm, with the platy layers tightly held together by hydrogen bonds. It is nearly devoid of any ion exchange phenomenon.

Table I shows the theoretical and actual measured chemical compositions of water-washed sericite [16]. The table illustrates that water-washed sericite contains various impurities, particularly Fe_2O_3, that markedly affect its brightness. When water-washed sericite at a 30% solids content was leached using a 0.5 M oxalic acid solution for 8 h, and amply washed and dried afterward, its brightness increased from 60% to 70% ISO, while the average particle size decreased from 10.5 to 10.1 µm. **Figure 1** shows scanning electron micrographs of sericite before and after oxalic acid treatment.

Comparisons of the major physical properties of sericite and traditional coating pigments (delaminated clay and wet-ground calcium carbonate [GCC]) are shown in **Table II** [7-19]. Sericite, similar to delaminated clay, has a platy structure, with particle sizes 20~30 times larger than those of delaminated clay and calcium carbonate. It has an aspect ratio 1~2 times that of delaminated

Elemental oxide	Theoretical composition (%)	Actual water-washed sericite (%)
SiO_2	47.37	53.41
Al_2O_3	40.26	30.70
K_2O	12.37	7.06
Na_2O		0.19
CaO		0.03
Fe_2O_3		1.95
MgO		0.30
S		2.20
P		0.47
Total	**100**	**100**

I. Chemical composition of sericite [16].



1. SEM photomicrographs of sericite with (left) and without (right) oxalic acid treatment.

clay and a hardness value roughly comparable to those of delaminated clay and calcium carbonate; thus it should not cause excessive abrasion to forming wires. In terms of optical properties, sericite has a much lower brightness than both delaminated clay and GCC, but a similar refractive index to those pigments. Because particles of sericite are much larger and have greater amounts of impurities, the light-scattering ability may be lower, and the light-absorption ability may be greater. The IEC and particle charge (PCD) values of sericite are far greater than those of delaminated clay and GCC.

The purposes of the study were thus to evaluate the effects of using swelling sericite in LWC coating formulations to partially or totally replace the charge of delaminated clay on the properties of the ensuing coating colors and the optical properties and printability of the coated paper. Thus, we attempted to define the roles sericite plays in coating formulations and to elucidate the properties of sericite-coated LWC paper.

	Sericite	Delaminated clay	GCC, H90
Main ingredients	SiO_2 54% Al_2O_3 31% K_2O 7%	Al_2O_3 45% SiO_2 39%	$CaCO_3$ 98.3%
Crystalline structure	Monoclinic	Triclinic	Rhombic
External appearance	Platy	Hexagonal plates	Spherical
Specific surface area (m^2/g)	57	16	14
Hardness (Mohs)	2.2~2.4	2.0~2.5	3.2~3.5
Range of particle size (µm)	0.8~45.0	< 0.1~5.0	0.1~ 4.0
Average particle size (µm)	10.0~14.0	0.5	0.39~0.41
Specific gravity	2.6~2.7	2.5~2.6	2.67
Refractive index	1.59~1.62	1.56	1.56
Reflectance (%, at 457 nm)	70	??	75
Brightness (% ISO)	65~75	88~89	93~96
pH	7~9	6.5~7.5	8.8~9.6
Aspect ratio	30~40	20[a,b]	1
IEC[c] (meq/g)	5.0[d]	0.5~1.5	–
PCD (meq/g)	-48.4	-1.1	-1.9

[a] Estimated by the J.M. Huber Co.
[b] The value changes with particle size.
[c] IEC, ion exchange capacity.
[d] Most mica minerals have values in the range of 1.6~2.0 meq/g.

II. Comparisons of the physical properties of sericite and traditional coating pigments [7-19].

EXPERIMENTAL

Design

The experimental design simulated a typical gate-roll coating color formulation. The solids content was adjusted to 62%. The control group used a pigment formulation with delaminated clay-GCC content of 50%-50%. The experimental variables were sericite (SA, with oxalic acid treatment; and SB, without oxalic acid treatment) the coating amount (6, 9, and 12 g/m^2), and the amount of sericite that was substituted for the delaminated clay (0%, 25%, 50%, 75%, and 100%). **Table III** shows the experimental formulations. There were nine sets of coating color formulations and 27 sets of coating experiments.

The coating color properties observed included solids content, pH, low and high shear viscosities, and water retention value. Researchers measured the coated paper's optical and printing properties, such as opacity, brightness, gloss, printed gloss, and print density, after passing it through a supercalender stack twice.

Sericite/clay ratio (%)	0/100	25/75	50/50	75/25	100/0
Sericite (part)	0	12.5	25	37.5	50
Delaminated clay (part)	50	37.5	25	12.5	0
GCC H90 (part)	50	50	50	50	50

Color formula: pigments (sericite + clay + H90) 100 parts, lubricant 1 part, latex 14 parts, etherified starch 4 parts, hardener 1 part, melamine resin 2 parts.

III. Coating formulations of the experimental design.

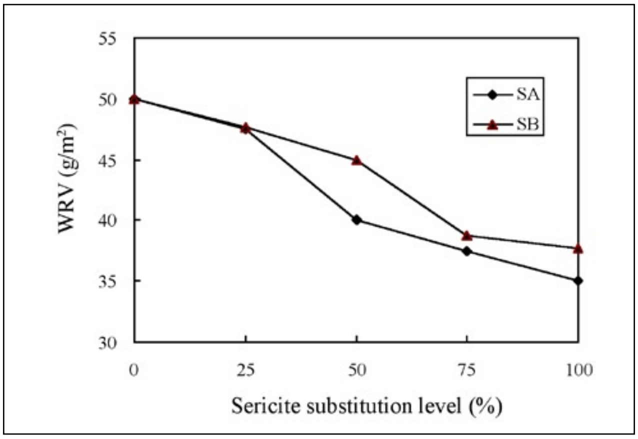
Materials and methods

Specifications of the main ingredients, base paper, pigments, and chemicals used in the experiments are detailed as follows.

The coating base paper was made from bleached hardwood kraft pulp of mixed hardwood (mostly eucalypts), at 60 g/m^2, manufactured by Chung-Hwa Pulp and Paper (Hualien, Taiwan). It had a brightness of 82.3% ISO, color (L*, a*, b*) of 92.30, -0.21, and -0.62; and opacity of 85.1%. The sericite was Sungloss® 11J-038 (plain water-washed) and KF-900-A (oxalic acid treated), from Sunshine Mineral (Taitung, Taiwan). The delaminated clay was Hydraplate® from J. M. Huber (Edison, New Jersey, USA). The ground calcium carbonate (GCC) was H-90 from Omya (Suau, Taiwan). The soybean protein was PC-5000 from DuPont (Wilmington, Delaware, USA). The latex was 780 SF from Shin Foong Chemical Industry (Fongshan, Taiwan).

Instruments used for testing the coating color and coated paper included a Brookfield viscometer (using a no. 3 spindle, at 100 rpm), a Kaltec AA-GWR water retention value meter (1.5 bar, 120 s, and a 5-µm filter), a Hanna pH meter (model 8521), a Hercules high-shear viscometer (1300 g/cm^3 and 4400 rpm), a KRK motor rod-type coater, and a KRK supercalender (model 30FC-200E). The optical instruments included a Zhentner gloss meter (model 60-70 ZLR), an Elrepho brightness meter (model 3000 Datacolor series), a Photovolt opacity meter (model 670), and a Bekk smoothness tester. The printing instruments included an RI printability tester, an IGT printability tester, an IGT Repprotest, and a Techkon printing density meter (model ER 460 eD2B).

The process of coating color preparation was in the order of pigments (clay and GCC) + distilled water (high-speed mixing) → dispersant (high-speed mixing) → sericite (high-speed mix-



2. Influence of sericite on the water retention value (WRV). SA = sericite with acid treatment; SB = sericite without acid treatment.

ing) → defoamer (high- and low-speed mixing) → soy protein/etherified starch (low-speed mixing) → latex/lubricant (low-speed mixing) → hardener/melamine resin (low-speed mixing) → NaOH (low-speed mixing) → distilled water (low-speed mixing). The binder level (latex + starch) was 18 parts pigments.

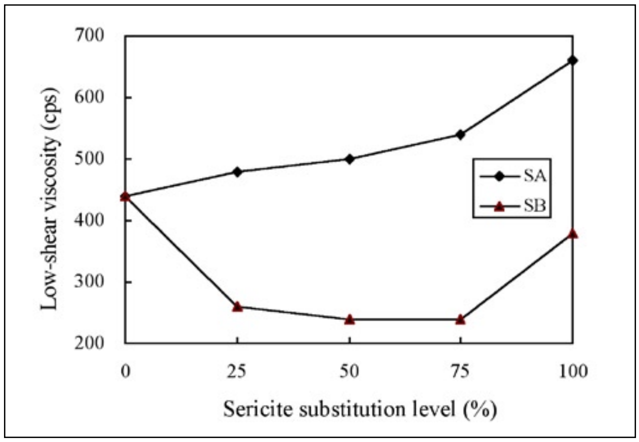
The coating color thus prepared was applied to 21.0 x 29.7-cm base sheets using an automatic bar coater (KRK-motor type). The base sheet was preconditioned for 24 h at 20°C and 65% RH. The coating amount was adjusted by using bars of different numbers. The coated sheets were immediately placed in an oven maintained at 120°C for 30 s to dry. Coated paper was supercalendered in a double pass with a KRK 30FC-200E single-nip supercalender having a linear nip pressure of 9.807 x 10^3 kPa and a temperature of 55°C.

RESULTS AND DISCUSSION

Properties of the coating colors

The coating colors used two varieties of sericite: one was treated with oxalic acid and denoted SA, the other was untreated and was denoted SB. The sericite was added in an increasing series to replace delaminated clay (0%, 25%, 50%, 75%, and 100%), for a total of nine sets of experiments. The coating color solids contents were adjusted to 62%, and ranged from 61.75% to 62.52%. The pH of the coating color was adjusted to 9.5, and ranged from 9.47 to 9.61.

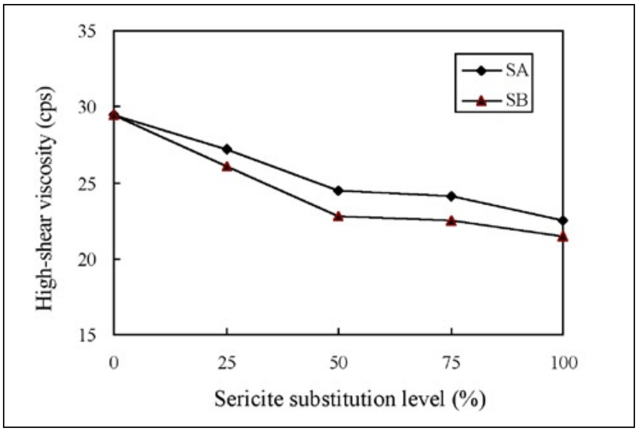
Figure 2 illustrates the effect of sericite addition on the water retention property of the coated colors. The higher a sample's WRV, the poorer its water retention property. With an increase in the amount of delaminated clay replaced by sericite, the WRV of the color linearly decreased. In other words, the addition of sericite enhanced the water retention property. The SA tended to have a more-pronounced water retention property than did the SB, raising the value by approximately 30%. This water retention phenomenon may reflect the expandable nature of sericite; the oxalic acid treatment, which removed some impurities, might have facilitated interlayer expansion. Conceivably, under a high shear environment of color preparation, interstices between the layers of the sericite gradually expand, and some are eventually delaminated. When the interlayer distance is greater than the molecular diameters of smaller particles, such as water or starch [14], these particles tend to migrate into the space through charge attraction, van der Waal forces, capillary action, etc., thus becoming intercalated and leading to increased water retention. Some of the water might be locked inside the matrix of the mineral particles and not be shared among the floc particles. This observation is congruent with that of Huang and Lepoutre [20]. Thus, the flocculent structure is sturdy and the water retention of the color is good when swollen sericite forms a flocculent structure in a mixture of pigment, latex, and a water retention agent, and when the bonding forces among the particle aggregates are greater than the internal coalescing forces of individual particles. Conversely, the flocculent structure is unstable and the water retention property also decreases when the internal coalescing forces of individual particles are greater than the bonding forces among particles.



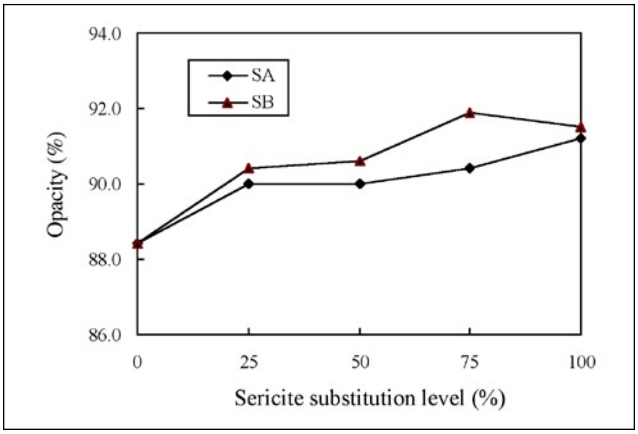
3. Influence of sericite on the low shear viscosity. SA = sericite with acid treatment; SB = sericite without acid treatment.

Figure 3 illustrates the effect of sericite addition on the low shear viscosity of the coating color. This shows that when SA replacement levels were below 75%, the low shear viscosity increased linearly with increasing sericite substitution. At the 100% substitution level, the viscosity increased disproportionally. On the other hand, replacing delaminated clay with SB tended to lower the low shear viscosity, though at 100% substitution a rapid increase in low hear viscosity was observed. These results suggest that, except for the 100% SA replacement, the viscosity of the coating colors could be controlled within the operational range.

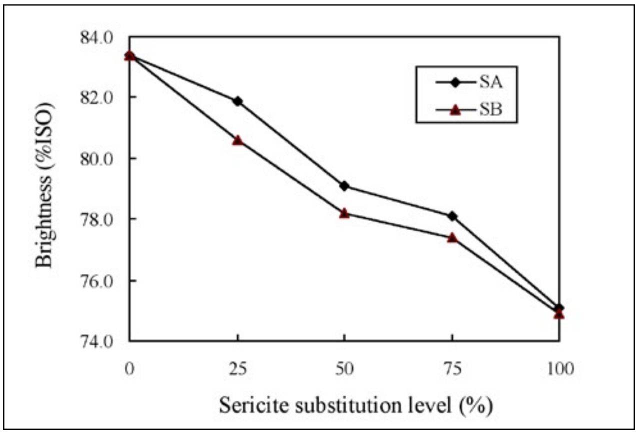
The phenomenon of viscosity behavior may be rationalized as being influenced by the expansion of sericite and by the adsorption of water and polymers (specifically starch) onto the interlayer surfaces. The swelling of sericite increases the viscosity because it has a larger effective volume than the clay it replaced. Some of the water counted in the solid concentration is bound within the sericite matrix. The volume concentration of the pigment is higher than in those formulations without sericite substitution, even though the solids concentration has remained the same. The effective volume fraction of the solids in the dispersion is increased. Intercalation of water and binder into the mineral depletes the aqueous phase and reduces its viscosity. For the SA series, the effect of the mineral's greater



4. Influence of sericite on the high hear viscosity. SA = sericite with acid treatment; SB = sericite without acid treatment.



5. Influence of sericite on the opacity. SA = sericite with acid treatment; SB = sericite without acid treatment.



6. Influence of sericite on the brightness. SA = sericite with acid treatment; SB = sericite without acid treatment.

swelling probably outweighs the absorption of the water, so the low shear viscosity rises. For the SB series, the effects of water depletion in the aqueous phase and mineral swelling are probably about equal, so the low shear viscosity rises only slightly with increasing substitution of delaminated clay.

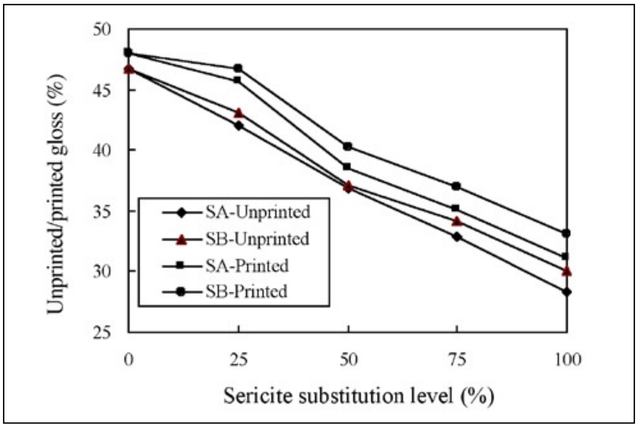
Figure 4 shows the effect of sericite on the high shear viscosity of the coating color. Adding sericite lowered the high shear viscosity in a linear manner. The range of change was between 20 and 30 cps, which is still within the operational range of the coating color viscosity. The SA group showed lower high shear viscosity than the SB group, which was the reverse of the low-shear viscosity phenomenon. Because sericite particles are much larger than clay particles, the viscosity fell at every level of substitution as the number of particles per unit volume decreased. The expandable nature of the particles becomes more important as the proportion of sericite in the blend increases, and this effect results in decreasing viscosity. This observation also suggests that because of the expandable nature of sericite, the expanding interlayer distances and the plate surface adsorption offered an elastic influence on the high shear viscosity. This is expected to effectively improve the operational conditions of the coating process in practice.

Properties of the coated paper

The paper coating experiments entailed applying nine sets of coating colors onto base sheets, with coating amounts of 6, 9, and 12 g/m^2, for a total of 27 runs. Results of different coat amounts tended to show similar trends; hence we opted to use the results of a typical 12 g/m^2 coat weight of LWC for discussion. **Figure 5** shows the effect of sericite substitution levels on the opacity of the coated paper. Opacity increased with increasing sericite substitution in a linear manner. At the maximum level of substitution, the SB group exhibited a 3.1% increase and the SA group showed a 2.8% increase in opacity. The results suggest that oxalic acid treatment, despite its enhancing effect on sericite expansion, might cause a loss of light-scattering effectiveness or the covering power of the pigment. The light scattering ability of the coated layer decreased as delaminated clay was replaced by much-larger sericite particles. This should have caused the opacity to decrease, but opacity increased, probably because the light absorption increased by a greater extent as a result of the lower brightness of sericite. The effect was more pronounced for the lower-brightness SB series.

Figure 6 illustrates the effects of sericite application and substitution levels on the brightness of the coated sheets. The figure shows that sericite lowered the brightness linearly with the added amounts. The SA group showed a slightly less-pronounced brightness-lowering effect than the SB group. The SA group lost 4.3% ISO and the SB group lost 5.2% ISO in brightness at the 50% substitution level. The lower reducing effect of SA might be explained by its higher intrinsic brightness.

We can use the brightness and opacity data of the coated sheets to calculate estimates for the absorption (K) and scattering (S) coefficients based on the Kubelka-Munk equation [21, 22]. Using the 0% and 50% substitution levels, for example, the S and K calculations are shown in **Table IV**. The table illustrates that when substituting small-particle delaminated clay with large-particle sericite, the S value decreased. The S values for 0% and 50% SA and 50% SB were 0.066, 0.063, and 0.0061, respectively. As oxalic acid treatment did not appreciably alter the particle size, the S values of SA and SB were similar. The K value of the low-brightness SB group markedly increased. K values for 0% and 50% SA and 50% SB were 0.00083, 0.0017, and 0.0019, respectively. **Table V** thus explains the ef-



7. Influence of sericite on the unprinted and printed glosses. SA = sericite with acid treatment; SB = sericite without acid treatment.

Optical property	Sericite substitution level (%)		
	0	50-SA	50-SB
Brightness (% ISO)	83.4	79.1	78.2
Opacity (%)	88.4	90.0	90.6
K/S	0.0125	0.0276	0.0304
SW	4.75	4.50	4.40
S	0.0660	0.0625	0.0611
K	0.00083	0.0017	0.0019
W = basis weight (72 g/m²); S = scattering coefficient; K = absorption coefficient.			

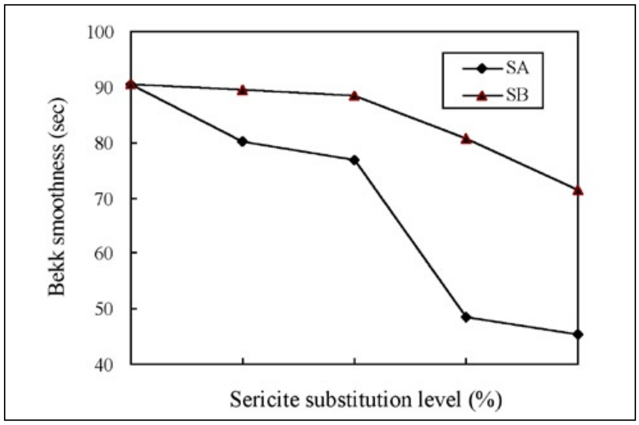
IV. Calculation of the light-scattering coefficient (S) and light absorption coefficient (K) [21, 22].

fect of sericite substitution on the opacity of the coated sheets.

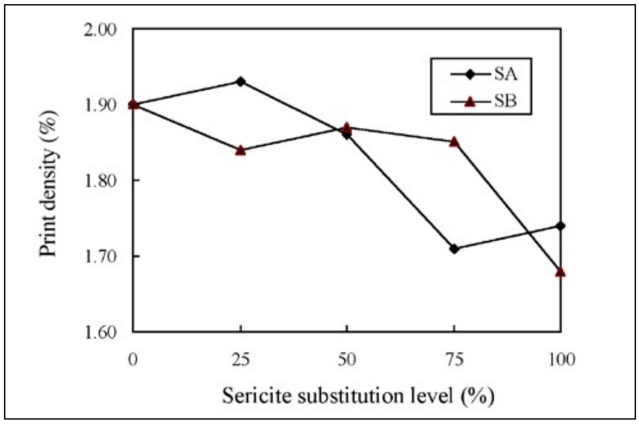
Figure 7 illustrates the effects of sericite substitution on the unprinted and printed glosses of the coated paper. Both unprinted and printed glosses were shown to decrease with increasing sericite levels. Delta gloss was constant with increasing sericite substitution and there were no appreciable differences between the SA and SB groups. This may have been related to the matted nature of the mineral, which despite its lustrous sheen, dissipates light [9]. Oxalic acid treatment had no apparent influence on this characteristic.

Figure 8 shows the effect of sericite substitution on the Bekk smoothness results. The smoothness of the coated paper decreased with sericite substitution. The SB group had a higher smoothness than the SA group. The effects on smoothness might be related to the particle size distribution and the nature of the swelling behavior. Sheets of higher sericite-containing coated paper had higher macro-roughness and micro-roughness values.

The effect of sericite substitution on the IGT printability results was similar. The IGT value was 478 cm/s in the delaminated clay group, and with sericite addition, all IGT values exceeded 500 cm/s. The surface strength of the coated paper effectively increased with sericite substitution. The surface strength increased for a couple of other reasons. The larger sericite particles may have less relative surface area accessible to the latex than the clay particles they replaced; the effective binder to the pigment surface area may have increased; and the coated surface also may have been rougher. These may



8. Influence of sericite on the Bekk smoothness. SA = sericite with acid treatment; SB = sericite without acid treatment.



9. Influence of sericite on the print density. SA = sericite with acid treatment; SB = sericite without acid treatment.

have led to the apparent improvement in surface pick strength. Oxalic acid treatment apparently imparted sericite with facile expansion and delaminating tendencies that, in turn, affected the IGT printability of the coated paper.

Figure 9 illustrates the effect of sericite substitution on the print density of coated paper. Sericite decreased the print density in a linear manner. We speculated that the print density fell because of the surface roughness. As the surface roughness increased with higher sericite substitution levels (Fig. 8), the ink film was increasingly taxed to fill the hills and valleys on the coating surface.

CONCLUSIONS

When swelling platy sericite is used in a coating color formulation for making LWC paper, the water retention properties of the coating color itself can improve by 30%. Except for oxalic acid-treated sericite, which tended to raise the low shear viscosity, all other experimental sets had lower low- and high-shear viscosities; the level of change reached a maximum of 50%. This characteristic can be used to adjust the use of water retention agents (starch, CMC, soy protein, etc.) and rheological modifiers to achieve economical and optimal coating color formulations. As for the properties of the coated paper, using sericite to replace delaminated clay caused a 1.5%~2%

gain in opacity at the 25% level of replacement. Brightness suffered a drop of 0.5%~2% GE, however. The printed gloss of the coated paper will drop 10% when sericite is used, while the print density is largely unaffected. Our experiments also show that during the process of coating color preparation, sericite swells and is delaminated. **TJ**

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INSIGHTS FROM THE AUTHORS


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Sericite, a mica mineral indigenous to Taiwan, is a swelling, layered inorganic material. It has similar characteristics to bentonite and thus has good potential for use in wet-end papermaking and in coating applications. This coating study represents the first step in developing sericite for such applications. We eventually hope to establish an entire application system for the mineral.

The most difficult part of this study was during the preparation of the coating color. The mineral has swelling and water-imbibing characteristics different from traditional delaminated clay and GCC. In the beginning, we either added water in the wrong amounts or at the wrong times, often leading to too high solids contents with the viscosity rising precipitously, and further work was impractical. After numerous trials and errors, we finally established the addition sequence of the coating ingredients and determined how to add suitable amounts of water to reach the desired solids content and minimize experimental errors.

The laboratory study has now established that a small amount of sericite replacing delaminated clay (25% or less) can enhance water retention, improve rheological properties, and raise the opacity of the sheet without appreciably harming other optical and printability properties of the coated paper. Thus it should be useful in improving the quality of LWC paper. In the future, we shall seek suitable LWC paper mills to engage in pilot plant and mill on-line tests to further establish the role of sericite in coating color formulations.

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