Print mottle effect of binder migration and latex film formation during coating consolidation

Kenichi Yamazaki, Toshiyasu Nishioka, Yoshihiko Hattori, and Kazuyo Fujita

ABSTRACT: The authors studied the effects of binder migration and latex film formation on print mottle. In the first stage they examined binder migration using experiments on a laboratory scale. The second stage was a pilot coating to confirm the initial results. There were some differences in print mottle tendency between the stages. In addition to binder migration latex film formability also affected print mottle.

KEYWORDS: Adhesive migration, film, formability, glass transition temperature, latex, mottle, print quality, starch.

Binders used in paper coatings migrate toward the base sheet and toward the coated surface during the consolidation process of a coating operation *(1–9)*. This migration has a strong influence on the printability of the final coated product *(1–8, 10)*. The study of binder migration is therefore very important.

Starch and latex are the most popular binders for paper coating. One usually uses them together in color coatings. Although starch and latex are both aqueous binders, they are not exactly similar. Starch is a water soluble binder, and latex is not. Latex is a water dispersed binder because the latex particle itself is not soluble in water. Considering these different characteristics of starch and latex, one would not expect them to migrate in the same fashion.

There are many studies on binder migration *(1–12)*. Few of these address the difference of the migration behavior between starch and latex *(11–13)*. The reason for this seems to be the difficulty of the measurement of the starch and the latex in the coated layer.

Recently studies used ESCA for this purpose *(11–14)*. Two studies briefly reported the difference of starch and latex migration behaviors *(11, 12)*. Although both studies used ESCA for the measurement of the surface starch and latex, their results did not agree. One group of researchers measured the depth profile of the starch to latex ratio. They concluded that latex migrates toward the coated surface rather than the starch. Another researcher measured the surface starch and latex contents of commercial coated paper. He showed that the starch content in the coated surface is 168% higher than that in the color coating. The latex content in the coated surface is 123% higher than that in the color coating.

Binder migration

Our study here first examines the difference of the migration behavior between starch and latex. Binder migration consists of two stages. The first occurs between the coater and the dryer. Here the binder migrates toward the base sheet in conjunction with the penetration of water. The second stage occurs in the dryer when the binder migrates toward the coated surface during the evaporation of water. Considering that both of these processes transpire, the interval period between coating and drying should be one of the important factors which strongly affects the binder migration.

This work closely studies the influence of the interval between coating and drying. It also includes the influence of the glass transition temperature (Tg) of the latex because film formation and adhesion of the particles could affect latex migration.

To determine printability, we evaluated the print mottle of the test samples. We also examined the relationship between binder migration behavior and print mottle.

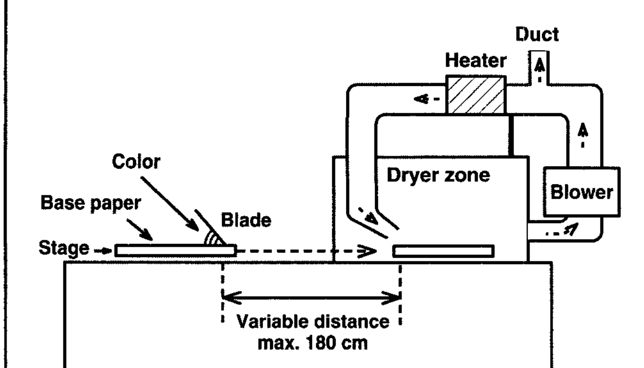
ESCA is a satisfactory technique for this purpose. It is a relative method, however. The preparation procedure of the samples influences the resulting data. We therefore used direct

All of the authors are associated with Latex Research Center, Sumitomo Dow Ltd., 2–4, Nakanoshima 3–chome, Kita–ku, Osaka, Japan 530

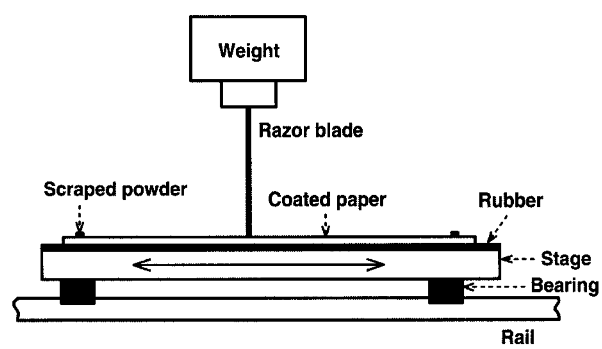
I. Coating formulations for laboratory experiments (L series) and pilot coater experiments (P series) in dry weights

	L–1	L–2	L–3	L–4	L–5	P–1	P–2	P–3	P–4
Ingredient									
No. 1 Kaolin clay	80	80	80	80	80	80	80	80	80
Finely ground calcium carbonate	20	20	20	20	20	20	20	20	20
Sodium polyacrylate	0.16	0.16	0.16	0.16	0.16	0.16	0.16	0.16	0.16
Sodium hydroxide	0.25	0.25	0.25	0.25	0.25	0.25	0.25	0.25	0.25
Modified starch	6	2	6	6	6	6	6	6	2
Latex A, Tg = 5°C	11	11	7	11	0	0	11	0	11
Latex B, Tg = -10°C	0	0	0	0	11	0	0	0	0
Latex H, Tg = 40°C	0	0	0	0	0	11	0	0	0
Latex L, Tg = -25°C	0	0	0	0	0	0	0	11	0
Solids content, %	63	63	63	66	63	63	63	63	66
Setting time, s	16.0	8.5	15.2	12.1	17.2	-	-	-	-

1. Multipurpose laboratory coater



2. Surface scraping apparatus



measurements of powder scraped from the coated samples for the analysis of the surface starch and the latex.

Laboratory experiments

In the laboratory scale experiments, we studied the influences of the Tg of the latex and the interval period between coating and drying on the binder migration behavior as well as the relationship between the surface binder content and print mottle.

Coating formulations

Table I shows the formulations of colored coatings containing clay and calcium carbonate as pigments along with starch and latex binders. The formulations for this series have the letter L as the prefix. There were variations in starch levels, latex levels, latex Tg, and solids contents.

Coating and drying

A new laboratory coater shown in **Fig. 1** applied the formulations onto 46 g/m^2 wood-free basestock at a coating weight of 8 g/m^2 using a blade and at 17 g/m^2 using a wire wound rod at a speed of 45 m/min. This coater also controlled the interval period between coating and drying. Drying conditions for preparation of the coated samples were an air velocity of 35 m/s, an air amount of 1 m^3/min, an air temperature of 110°C at the coated surface, and a drying time of 6 s. Drying intervals ranged from 0.3 s to 1.1 s. After conditioning, we supercalendered the coated samples at 60°C.

Testing

The setting time method measured the time during which the coating retained water. This technique involves applying the formulations with a wire wound rod onto a smooth and uniform base

II. Surface starch and latex contents for laboratory experiments in parts per hundred

Formulation	Coating weight, g/m²	Interval period, s: 0.3 Starch	0.3 Latex	0.7 Starch	0.7 Latex	0.9 Starch	0.9 Latex	1.1 Starch	1.1 Latex
L–1	8	6.4	10.4	4.7	10.2	4.7	10.0	4.4	10.5
L–2	8	2.2	8.6	1.9	7.9	1.6	8.6	1.8	9.1
L–3	8	6.5	5.7	5.3	5.5	4.9	5.7	4.8	5.8
L–4	8	6.0	11.1	5.4	10.7	4.8	11.2	5.0	10.7
L-5	8	5.9	10.5	4.9	10.0	4.6	10.4	5.2	10.2
L–1	17	10.2	12.9	9.9	12.1	-	-	9.1	11.7
L–5	17	10.1	10.5	9.5	10.2	-	-	8.8	10.6

stock. A stop watch records the time until the gloss of the coated surface disappears. This is the setting time.

The second test determined the starch and latex content at the coated surface. Using the surface scraping apparatus of our own design shown in **Fig.** 2, we scraped the coated surface of the sample to a depth of 0.3 μm to collect powder. We extracted the starch from this with hot water and then determined the starch content by absorbance measurement using a starch–iodine solution. Pyrolysis gas chromatography at a temperature of 590°C allows direct measurement of the latex content in the scraped powder.

A trapping test measures the print mottle tendency. In this test, we printed coated paper samples with a yellow ink with the first roll followed by an overcoating with a magenta ink with the second roll. Evaluation of the uniformity of the magenta ink provides a measurement of print mottle tendency.

Results and discussion

Table II shows the values for the surface starch and latex contents for the samples. The data indicate that the migration behaviors of starch and latex are not identical. The surface starch content increases with a decreasing drying interval period for all five coatings. This tendency is very apparent when the interval period varies from 0.3 s to 0.7 s. Conversely the latex contents in the coated surface are independent of the drying interval period as seen in **Fig.** 3.

The surface starch contents of the samples prepared with the shortest interval period of 0.3 s are slightly higher than the starch contents in the color coatings. In these samples, the starch is therefore migrating toward the coated surface. On the other hand, the surface starch contents of the samples prepared with interval periods longer than 0.3 s are lower than the starch content in the color coatings. In these cases, the starch is therefore migrating toward the base sheet rather than the coated surface.

The surface latex contents are slightly lower than the latex contents in the color coatings in every interval period. There is therefore no migration toward the surface but slight migration toward the base sheet.

To study the influence of coating weight, we prepared higher coating weight samples of 17 g/m². Since it was difficult to achieve this by blade coating, we coated these samples by using a wire wound rod.

Table II also shows the surface starch and latex contents of 17 g/m² coated samples prepared with two color coating samples. In these examples, the surface starch content decreased with an increase of the interval period similar to that for the lower coating weight samples.

In the case of the samples prepared with L–1, the surface latex content decreases with an increase of the interval period. For the samples prepared with L–5, however, the surface latex content is independent of the interval period as shown by **Fig. 4.** The difference between these two formulations is the Tg level of the latex used. L–1 uses latex A whose Tg value is 5°C, and L–5 uses latex B whose Tg value is -10°C. This means that the migration of latex depends on its Tg level. High Tg latex migrates more easily than low Tg latex. This observation agrees with previous work *(15)*.

Although the surface starch and latex contents increase as the coating weight increases, the dependence of the increase ratio on coat weight is much higher for starch than for latex as indicated in **Fig.** 5. The increase ratio is the ratio of the starch or latex content in the coated surface to the starch or latex content in the color coating.

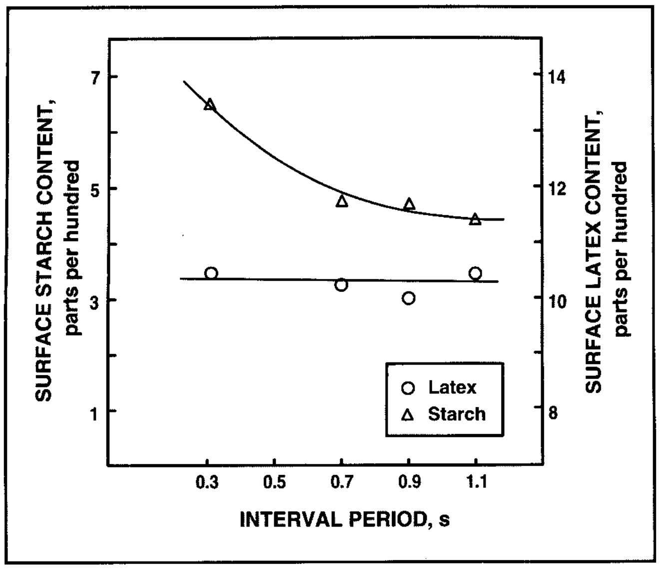
In Fig. 5, there is also an indication that the distribution of surface starch content tends to become nonuniform more easily than that of surface latex content as a result of the uneven distribution of coating weight.

All the data show that starch migrated more easily than latex either toward base sheet or coated surface.

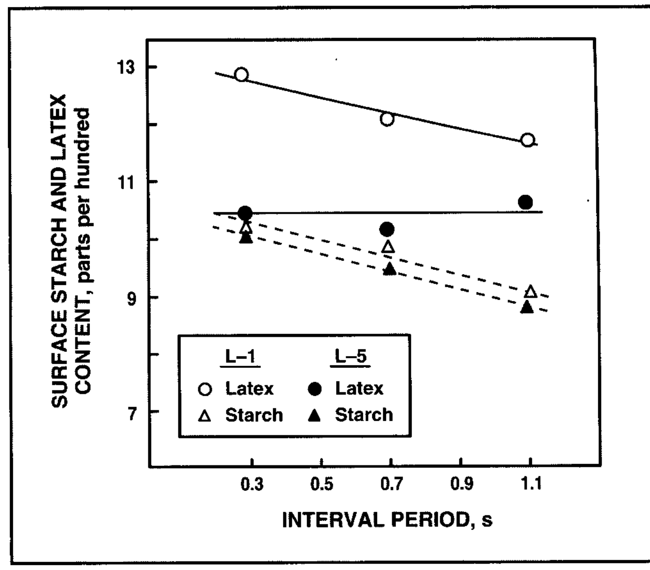
Our results can explain the disagreement of the previous studies *(11, 12)*. When migration toward base sheet occurs, the ratio of surface starch to latex is lower than that in the color coating. For migration toward the coated surface, however, the ratio of surface starch to latex is higher than that in the color coating.

Table III shows the print mottle tendency of the blade coated samples. In every formulation, the print mottle improves with an increasing interval period between coating and drying. This mottle tendency seems to have some correlation with the migration behavior of starch. Although there is no obvious relationship between the mottle tendency and the surface starch

3. Interval period between coating and drying for various surface starch and latex contents using 8 g/m² coated samples of formulation L–1



4. Interval period between coating and drying for various surface starch and latex contents using 17 g/m² coated samples of formulations L–1 and L–5



contents, samples in which the starch migrates toward the coated surface show severe mottle. We presume the reason for this is the nonuniform starch distribution in the coated surface which arises from the nonuniform distribution of coating weight seen in Fig. 5.

There is a critical interval period in which the mottle disappears in every formulation. Comparing the mottle tendency of the samples prepared with L-1, L-2, and L-3, the critical interval period decreases with a decrease of the ratio of starch to latex in the color coating.

The critical interval period decreases with an increase of solids content and decreases with the latex Tg.

Since the factors which affect the critical interval period also affect the print mottle, one might conclude that the factors which decrease the critical interval period reduce the print mottle as well.

Pilot coater study

Although there were numerous interesting results in the laboratory experiments, the factors of blade geometry, coating speed, and drying condition using a laboratory coater differ significantly from those with a mill coater. Pilot coating results are therefore necessary to confirm any conclusions from lab coating. Accordingly, we evaluated print mottle of coated samples printed with a commercial sheet–fed offset press.

III. Print mottle tendency for laboratory experiments

Formulation	Interval period, s 0.3	0.7	0.9	1.1
L–1	Severe	Severe	Severe	None
L–2	Severe	Severe	None	None
L–3	Severe	Severe	Some	Severe
L–4	Severe	Some	None	None
L–5	Severe	Some	None	None

Coating formulations

To study the effect of latex Tg level in detail, we prepared three formulations. These formulations were identical to those of L-1 except that the Tg levels of the latexes differed. In addition to latex A, there were two additional commercial latexes designated H and L. Latex H is harder than latex A and latex L is softer than latex A. Tg values of these latexes are 40°C and -25°C respectively. We also prepared the formulation using high solids content and low starch level as a mottle free formulation. Table I contains these formulations.

IV. Surface starch and latex contents for pilot coater experiments in parts per hundred

Formulation	Interval period, s: 0.2 Starch	0.2 Latex	0.6 Starch	0.6 Latex
P–1	5.4	10.3	5.1	10.5
P–2	4.8	10.5	4.2	10.1
P–3	4.8	10.4	4.2	10.5
P–4	1.6	10.3	1.5	10.1

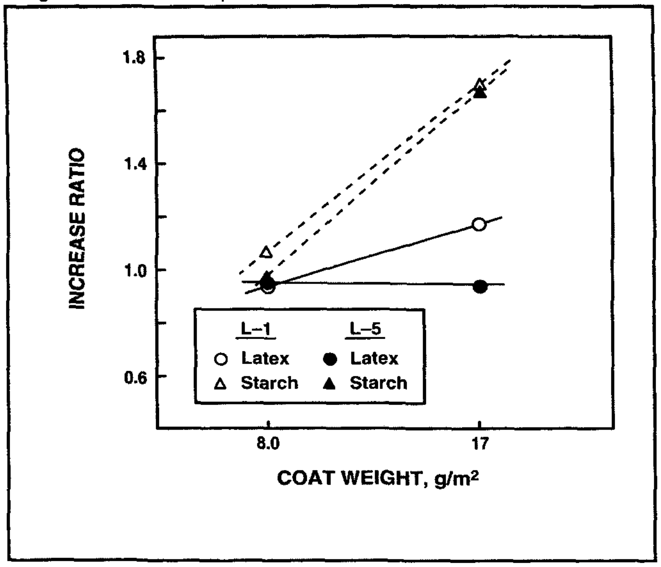
Coating and drying

We coated the formulations on 64 g/m² wood-free basestock at 10 g/m² using the pilot SDTA coater shown in **Fig. 6.** We coated each formulation twice using two different interval periods between coating and drying. Selection of the dryers allowed control of the interval period.

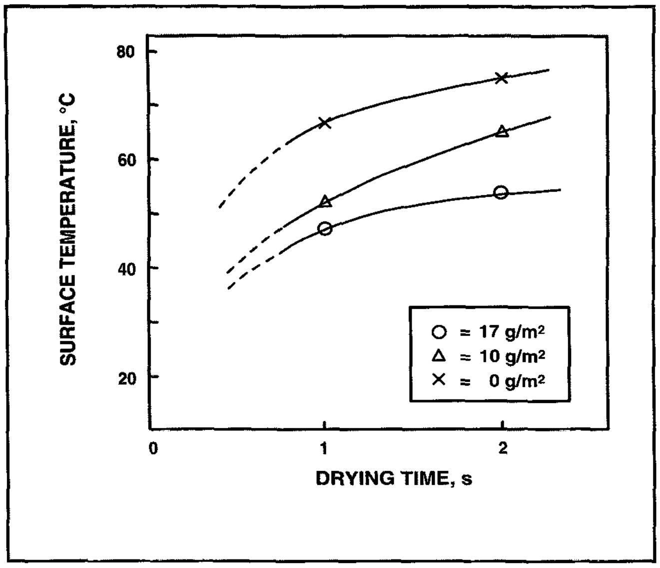
The pilot coater had five flotation dryers. We used three of these to vary the interval period between coating and drying. Using the first three dryers gave a short interval period. Using the last three dryers gave a long interval period.

Because of drying capacity limitations, the coating speed of this experiment was only 700 m/min with interval periods of 0.2 s and 0.6 s.

5. Variation of the ratio of starch or latex content in the coated surface to the starch or latex content in the color coating compared to coating weight with an interval period of 0.3 s



7. Effect of drying time on surface temperature



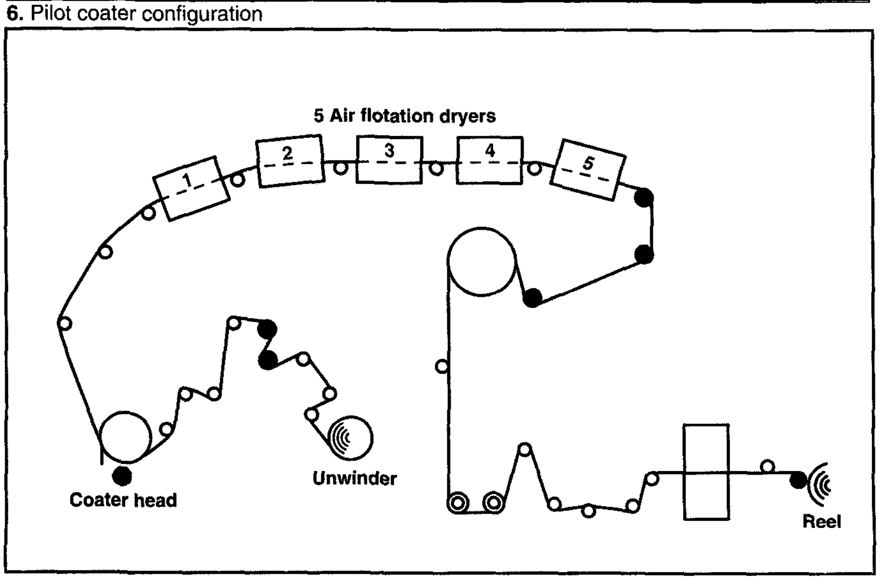
V. Print mottle tendency for pilot coater experiments

Form-ulation	Interval period, s: Test printer 0.2	Test printer 0.6	Commercial printer 0.2	Commercial printer 0.6
P–1	Worst	Worst	Severe	Worst
P–2	Severe	Severe	Some	Some
P–3	Severe	Some	Severe	Some
P–4	None	Some	None	Some

VI. Coated surface temperatures during pilot coating

Form-ulation	Interval period, s	Coated surface temp., °C: Before dryer	After dryer
P–1	0.2	-	64.8
P–1	0.6	40.3	65.1
P–2	0.2	-	64.6
P–2	0.6	43.7	67.9
P–3	0.2	-	65.6
P–3	0.6	40.7	65.7
P–4	0.2	-	60.4
P–4	0.6	36.5	60.8

6. Pilot coater configuration



Testing

We measured the surface starch and latex contents using the same procedure as in the laboratory experiment. A print tester and commercial four–color sheet–fed offset press printed the coated samples with a speed of 6000 sheets/h with a color sequence of black, blue, magenta, and yellow.

Results and discussion

Table IV shows the surface starch and latex contents of the pilot coated samples. In pilot coating, the surface starch content increases with a decrease in the interval period between coating and drying. Surface latex content is almost the same in all samples. This tendency is the same as that shown in Table II for the laboratory experiment.

The surface starch and latex content are both lower than the starch and latex contents in the color coatings. Even in the shortest interval period, the migration occurs toward the base sheet rather than toward the coated surface. It seems that in the pilot coating the penetration force is higher than that in the lab coating.

The effect of latex Tg on the binder migration did not occur in pilot coating. This may be due to the low coating weight of the pilot coated samples as well as the difference in the penetration force.

Table V provides the print mottle results for all the pilot coated samples. Sample P-4 showed the least tendency to mottle. The print mottle of the samples prepared with low Tg latex did not meet our expectations. Furthermore, the effect of the interval period between coating and drying on print mottle differed between the pilot coating and the laboratory coating. These differences may arise from the differences in migration behavior of the starch between the coatings. The migration of starch toward the base sheet is stronger in pilot coating than in the laboratory coating.

The worst mottle occurs in the samples prepared with the formulation using the high Tg latex. Since there is no particular binder migration in these samples, one would expect that the reason for this is not due to the binder migration. The Tg level of the latex in formulation P-1 is at a high level. The mottle of the sample prepared with this formulation becomes worse as interval period increases. We therefore think the reason for this bad mottle was uneven film forming of the latex in the coating.

The measurement of surface binder shows there is no binder migration toward the coated surface. This fact suggests that the coating is already in an immobile state as the coated sample comes into the dryer.

The temperature at which a latex begins to form a film is usually some degrees above the Tg level of the latex. Accordingly, we measured the temperature of the coated surface during the pilot coating process to provide the results in **Table VI**. This table shows that the temperature of the coated surface just before the dryer is almost the same as the Tg level of latex H, 40°C. After coating, the consolidation of the color coating is therefore progressing at a temperature near the Tg level of latex H. Latex H then is partly film forming.

Figure 7 shows the relation between coating weight and surface temperature using various drying times. It shows that surface temperature differs with coating weight at the same drying time. It further shows that nonuniformity of coating weight causes an uneven surface temperature during the drying process. If the coating temperature is near the Tg level of the latex in the formulation, the coated sample will exhibit mottle because of the nonuniform film formation of the latex during the consolidation process.

Conclusion

From the information we have collected in our experiments using both laboratory tests and pilot-scale trials, we can conclude the following:

- Migration behaviors of starch and latex during the consolidation process of coating are not identical. Starch migrates more easily than latex toward either the base sheet or the coated surface.
- Coating weight and interval period between coating and drying change the starch content in the coated surface more easily than the latex content. The starch therefore has more influence on print mottle of the coated paper than the latex.
- Tg level influences the migration behavior of latex. High Tg latex migrates more easily than low Tg latex. This tendency is especially clear in high coating weight samples.
- The tendency for print mottle decreases with decreasing starch level and increasing solids content of the coating formulation, because there is a reduced influence from starch migration.
- The film formation characteristics of the latex have an influence on print mottle, as well. If the temperature of the coating is near the Tg level of the latex during the consolidation process, a coated sample shows mottle because of nonuniform film forming of the latex during coating.

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