

Viscoelastic structure properties of the polymer binder affecting the processability, structure and properties of dry surface treated papers

KAISA PUTKISTO, JUHA MAIJALA, JOHAN GRÖN, AND MIKAEL RIGDAHL

ABSTRACT: In dry surface treatment (DST), coating processability and the properties of the coated paper are significantly affected by the temperature-dependent viscoelastic properties of the latex binder. It is beneficial to have a polymeric binder of high gel content to improve the surface strength of the final product and to reduce the likelihood that coating will stick to the heated roll during the fixation step. DST papers are comparable in many respects to conventional wet-coated papers. The DST coating structure appears to be more open, however, especially with latexes of high gel content.

Application: Dry surface treatment is an on-line, water-free, and noncontact method of coating a paper surface.

The dry surface treatment (DST) of paper and board is a combined coating and calendering process. It involves the application of dry coating particles and a thermomechanical fixing of the coating layer onto the substrate [1, 2]. An electric field is used to direct the charged particles to the paper surface, onto which the deposited layer initially attaches by electrostatic forces. Fixing takes place in a heated roll nip.

The two-component powdery coating materials contain pigments and thermoplastic polymeric binders. These binders deform during the thermomechanical treatment to allow spot-bonding of particles. This surface treatment is a noncontact process in which the drying phase is avoided. It may offer possibilities for reducing the costs of energy, equipment, and raw materials.

In processes like DST, a fast response of the polymeric binder to heat and stress is critical to achieve coherent and porous structure. However, if the fixation temperature is too high, the coating could stick to the heated roll [1–5]. The component materials selected and the method used for preparing the coating powders affect their performance in the DST process and influence the distribution of components in the coating layer.

Consequently, we studied the relationships between the processability in DST and the temperature-dependent viscoelastic properties of binders that affect the surface strength, the pore structure, the surface smoothness, and the depth of penetration of the coating. The size of the particles in the coating powders was also considered.

FIXATION BACKGROUND

Although the dwell time is short, it must be sufficient for the polymer to change its state and for the film to form. The binder should soften at a relatively low temperature, and there should be a distinct change in the elastic (storage) modulus. Effective softening and spreading of the polymer must be combined with appropriate viscoelastic properties at elevated temperatures. In other words, the stiffness has to be adjusted, which is controlled by the gel properties. Good binding ability is usually associated with a low glass-transition temperature, or a low film-formation temperature [6].

The WLF (Williams, Landel, Ferry) time-temperature superposition principle can be used to estimate the viscoelastic properties of a polymer or a paper coating [7, 8] at any particular processing temperature and speed if the same properties are known at other conditions:

$$\log a_T = \frac{-C_1(T - T_r)}{C_2 + T - T_r} \quad (1)$$

$$\omega_1 a_{T_1} = \omega_2 a_{T_2} \quad (2)$$

where

a_T = shift factor at the temperature T
 C_1, C_2 = constants at given conditions
 T_1, T_2 = temperatures in States 1 and 2
 T_r = reference temperature to which the values of a_T refer
 ω_1, ω_2 = frequencies of deformation in States 1 and 2.

However, temperature and pressure are not constant; instead, they form a gradient thorough the coating layer in the

nip. Furthermore, the void volume and the interactions between the binder and the pigment particles modify the stress transfer. The inclusion of pigment particles can be expected to have a reinforcing effect on the mechanical properties of the polymer film, or rather the coating layer. Therefore, the results of dynamic mechanical analysis (DMA) can be used only to indicate the deformation phenomena during processing.

MATERIALS AND METHODS

We produced two-component DST coatings from freeze-dried particles that contained 100 pph ground calcium carbonate (GCC) as the pigment and different grades of carboxylated latexes. These latexes, defined in Table I, were based on styrene-butadiene copolymers. The latex contents were 10 and 20 pph, and the concentration of the dispersant was 0.04 pph. The latex binders are denoted by the glass transition temperature (T_g) and the relative gel content (L, M, and H for low, medium, and high).

We used DMA to characterize the viscoelastic properties of the binder films (ASTM D5279-01). We measured the shear storage modulus G' and the shear loss modulus G'' in the glass transition region and at elevated temperatures in the shearing mode in the linear region at a frequency of 1 Hz. We also derived the mechanical loss factor ($\tan \delta = G''/G'$).

The coating particles were prepared by freeze-drying and subsequent cryogenic grinding [3]. We determined the particle size distribution of the produced coating particles using a

LATEX	AVERAGE PARTICLE SIZE, μm	GEL CONTENT, T_g , °C	%
5-H	0.15	5	87
24-L	0.15	24	<30
24-H	0.11	24	86
45-M	0.19	45	58
51-H	0.18	51	83

I. Latexes used in the DST coatings.

low-angle laser-light-scattering instrument (LALLS, Malvern series 2600c, UK).

Powder deposition and fixing

The powders were applied onto the paper substrate with a laboratory-scale application unit [1]. A coat weight of 6 g/m² was applied onto 45-g/m² paper having a moisture content of 3% (SCAN-P 4:63). The coating was fixed in a laboratory calender.

For some samples, a fixing pretreatment consisted of one pass in a roll nip consisting of a heated roll with a Teflon-based cover and a polymeric backing roll. The roll surface temperature was 200°C, the speed was 10 m/min, and the linear load was 30 kN/m, corresponding to a dwell time of 60 ms.

For the actual calendering treatment, a "reinforced fixing" procedure was used for all samples. The roll nip consisted of a heated, hard-chromed roll and a polymeric backing roll with hardness of 90 ShoreD. The roll surface temperature was 200°C, and the speed was 10 m/min. Linear loads of 20 kN/m and 100 kN/m were used, corresponding to dwell times of 20 ms and 22 ms. All samples were treated in two nip passes at 20 kN/m and in two subsequent passes at 100 kN/m.

The reference paper was a commercially available blade-coated, soft-calendered MFC paper (machine finished coated) with a total grammage of 54 g/m² with a coat weight of approximately 6.5 g/m² per side.

Analysis

The coated papers were examined with scanning electron microscopy (SEM) with a solid-state backscatter electron detector (BSE). The cross-sections of the papers were studied after the papers were embedded in an epoxy resin and coated with gold. The coating coverage was assessed visually from micrographs obtained with environmental SEM (ESEM).

The porosity of the coating layers was determined by mercury porosimetry (Pore Sizer 9300, Micromeritics, Norcross, Ga.). A number of paper prop-

LATEX	CONC., pph	PARTICLE SIZE DISTRIBUTION, μm	
		D(90)	D(50)
5-H	10	72.6	26.6
5-H	20	127.2	49
24-L	10	60.1	17.3
24-L	20	109	33.6
24-H	10	37.4	10.4
24-H	20	47.7	17
45-M	10	9.5	2.7
45-M	20	14	5.5
51-H	10	11.3	2.3
51-H	20	12	5.1

II. Particle size distributions with 100 pph GCC and different binders.

erties were measured for the coated papers according to standard testing methods: grammage and coat weight (SCAN-P 6:75), moisture content of the base paper (SCAN-P 4:63), Bendtsen roughness (ISO 8791-2), PPS-s10 surface roughness (ISO 8791-4), and a IGT/Prüfbau AIC2-5 device was used with standard medium viscosity testing oil to determine (IGT) surface strength (SCAN-P 63:90). Each result given is an average of four measurements. Surface oil absorption properties were examined with a nozzle applicator method (KCL, Finland). All measurements were performed on specimens that had been conditioned according to SCAN-P 2:75 at room conditions.

RESULTS AND DISCUSSION

Viscoelastic properties

The latexes we used were divided into three classes based on the T_g : low, medium, and high. The gel content affected the magnitude and the temperature dependence of the shear storage modulus G' (Fig. 1) and the mechanical loss factor (not shown).

For latexes with a high gel content, the value of the shear storage modulus G' levelled out at a constant level on the order of 10⁵ Pa at elevated temperatures (the rubber plateau). Simultaneously the loss factor $\tan \delta$ decreased to relatively low values at temperatures above the glass transition temperature. For latexes with a low gel content, G' decreased more markedly with increasing temperature above T_g . The loss factor remained at a rather high level or even increased after the peak value, as noted with Latex 24-L, which exhibited excessive flow with increasing temperature.

LATEX	C_1	C_2 , °C
5-H	2.1	40.0
24-L	3.8	73.9
24-H	1.4	16.1
45-M	2.1	24.9
51-H	1.3	27.5

III. Constants C_1 and C_2 of the WLF equation.

Coating particle size distribution

The freeze-drying of the pigment-latex suspensions resulted in aggregated particles. With high- T_g and high-stiffness latexes, the rigidity and brittleness of the dried aggregates made it easier for them to break during grinding. These aggregates broke into finer coating particles with the narrowest size distributions. Determined by LALLS, D(90) and D(50) in Table II denote that 90% and 50%, respectively, of the particles were smaller than the size given.

Increasing the binder content increased the average particle size, or aggregate size. The size of coating particles affects not only the application and deposition results in DST [9] but also the coverage, component distribution, and pore structure of DST coatings [3-5].

Coating processability

Provided that the coating particles were rather small and the fixing conditions sufficient for binder softening, we obtained porous and abrasion-resistant coating layers. At the same coat weight, the surfaces of the DST papers appeared more porous than the reference surface, but the surface fibers were well covered, as Fig. 2 shows.

The surface of DST coatings appeared to vary in density with the surface roughness of the base paper. (To put it more technically, there was differential compression of the layer as a result of high and low fiber areas). DST papers exhibited a very low degree of coating penetration into the paper surface, reducing the effect of mechanical interlocking on coating adhesion, as shown in the cross-section micrographs in Fig. 2.

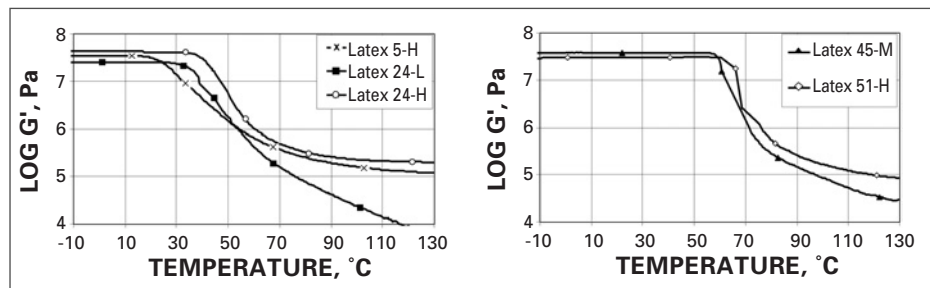
Sticking. High fixation temperatures and several nip passes with prolonged dwell times were used to promote binder flow and the bonding of the powder-formed coatings. However, high fixation temperatures in DST can also cause drastic degradation of the surface by increasing the tendency of the binder to stick. For example, pronounced sticking

was observed with coatings containing Latexes 24-L and 45-M, which also exhibited a reduced G' at elevated temperatures (Fig. 1). The sticking was emphasized at the higher binder content.

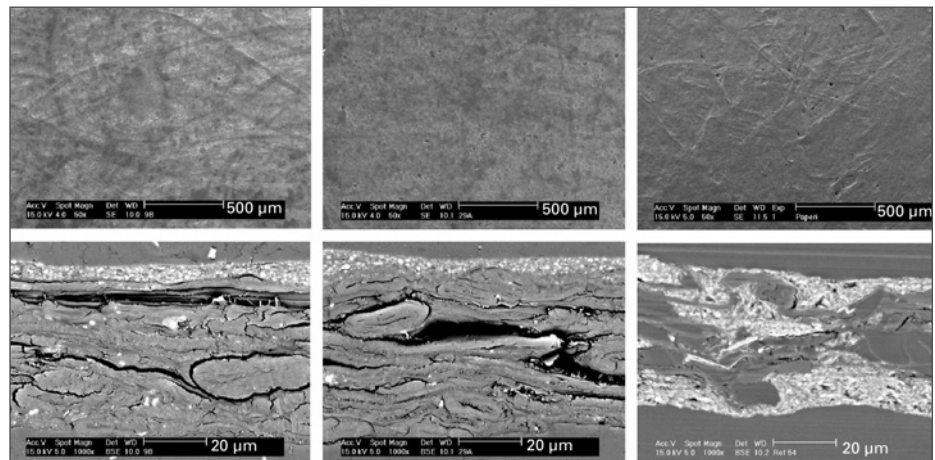
A reduced sticking tendency was attributed to the high-gel latexes and to the lower binder content, provided that the degree of aggregation was also low. This relation between calender runnability and latex G' is in agreement with the results reported by Wikström *et al.* [6].

In some cases, the sticking tendency at the first fixation nip could be reduced by pretreatment, as seen in Fig. 2 and Fig. 3 for the coating containing 10 pph of Latex 45-M. On the other hand, the sticking tendency can also increase with high-gel latexes with higher polymer contents. In some cases, judging from excessive dusting with powders with large particles, adhesion to the substrate was also poor (Fig. 3, second left micrograph) [2–5]. This problem may be related to the relatively low area available for adhesion with aggregated or large coating particles.

Homogeneity. During the preparation of coating particles, the distribution of binder may also become inhomogeneous, which may have been the case during freeze-drying. Prior to the freezing, the coating suspension is laid on a drying tray, where stirring is not possible and the components may settle unevenly. Grinding may be expected to make the coating powder more homogeneous. The grinding operation we used was not efficient in refining highly aggregated materials, and in most cases the particle sizes were larger than those of the pigment and latex in a suspension. For a coherent coating layer to form with aggregated particles, the binder would have to flow



1. Shear storage modulus G' of Latexes 5-H, 24-L, and 24-H (left) and 45-M and 51-H (right) as a function of temperature at a frequency of 1.0 Hz.



2. SEMs of the surface (upper row) and cross-sections (lower row) of the reference paper (right) and DST papers with 10 pph of Latex 45-M (left) or 10 pph of Latex 51-H (middle). The fixing pretreatment was used for the paper containing Latex 45-M.

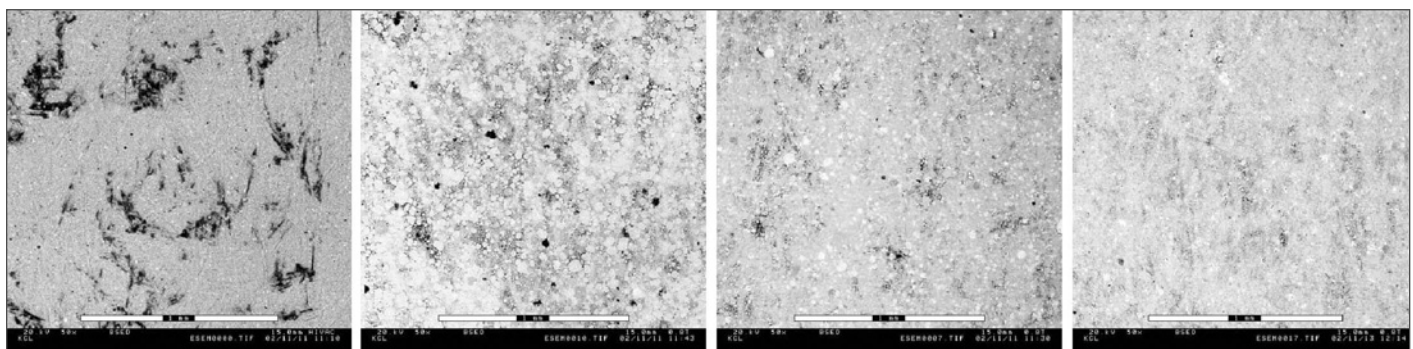
extensively to homogenize the composition and structure.

Nip conditions. Some effects of a varying nip dwell time and linear load on the fixation and on the properties of coated paper were reported elsewhere [5]. Our aim was to study the differences in the performance of different latexes by keeping the fixation conditions unchanged.

Aggregates. Low- T_g and film-forming latexes can be related to a high binding ability [6]. In this study, the aggregation of the coating particles overshadowed the possible advantages of a low- T_g

binder. Coatings containing Latex 5-H exhibited a low binding ability, although the sticking of the coating to the roll surface was avoided, probably at least partly because of its high gel content.

Thus, with regard to runnability in DST and the defect density in the formed coating surface, not only is the gel content of the latex important, but the size of the primary coating aggregates is also important. With smaller aggregates, the detached areas became smaller (Fig. 3, third micrograph), and the coverage became more uniform (right micrograph).



3. SEMs of the surface of the reference paper (left) and DST papers with 10 pph of Latex 24-H (second), 10 pph of Latex 51-H (third), and 10 pph of Latex 45-M (right). At a coat weight of approximately 6 g/m², the average coverage values were 88%, 98%, 98%, and 100%, respectively.

LATEX	CONC., pph	PRE-TREATMENT	IGT SURFACE STRENGTH, m/s	
			MEAN	STD. DEV.
5-H	10	—	—	—
5-H	20	—	—	—
24-L	10	yes	0.12	0.02
24-L	10	—	0.13	0.02
24-L	20	—	—	—
24-H	10	yes	0.15	0.02
24-H	10	—	0.15	0.03
24-H	20	yes	0.18	0.03
24-H	20	—	0.17	0.03
45-M	10	yes	0.20	0.02
45-M	10	—	0.22	0.01
45-M	20	—	—	—
51-H	10	yes	0.18	0.01
51-H	10	—	0.16	0.02
51-H	20	yes	0.35	0.02
51-H	20	—	0.33	0.01

IV. IGT surface strengths of DST papers (averages of four measurements in each case).

Obviously DST can give better coating coverage than can suspension coating with blade metering. This observation supports earlier results obtained by Putkisto *et al.* [3–5].

Flow properties. To estimate differences in the flow properties of the polymers during fixation, we used the WLF time-temperature superposition principle and the DMA results. The values of the constants C_1 and C_2 in the WLF equation were determined in the way reported by Richard [10]. For this purpose, modification of Eq. 1 yields

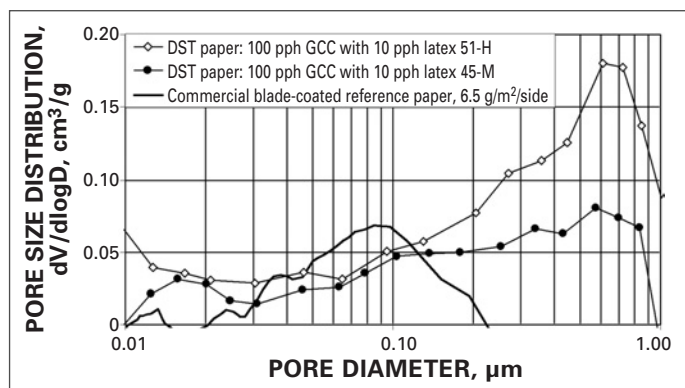
$$\frac{1}{\log a_T} = -\frac{C_2}{C_1(T-T_r)} - \frac{1}{C_1} = -\frac{C_2}{C_1} \frac{1}{T-T_r} - \frac{1}{C_1} \quad (3)$$

This relationship yields a straight line when $(1/\log a_T)$ is plotted as a function of $1/(T - T_r)$, where the slope of the line is $-C_2/C_1$ and the intercept with the y axis is $-1/C_1$. The shift factor a_T was calculated using G' obtained for each latex at temperatures higher than the $\tan\delta$ peak temperature (denoted in this context as T_p). In our case, this gave C_1 values between 1.3 and 3.8 and C_2 values between 16.1°C and 73.9°C, as shown in Table III. These values are consistent with results for styrene-butadiene copolymer latexes [10, 11].

Let us assume that the surface layer of the coating reached a temperature close to the roll surface temperature of 200°C (T_2). Let us also assume that the dwell time was 20 ms (corresponding to an angular frequency, ω_2 , of 50 s⁻¹).

By comparing the DMA results ($\omega_1 = 1.0$ s⁻¹), we see that the sticky polymers were considerably above the main transition in the nip, or the temperature region where G' fell below 10⁵ Pa and 10⁴ Pa and $\tan\delta$ was close to unity. For those latexes with a high degree of crosslinking, these fixation conditions corresponded approximately to the transition region, where G' was of the order of 10⁶ Pa.

Consequently, there is an appreciable difference in the deformability and flow properties between the latexes during processing. This difference can partially account for the observed differences in sticking tendency. However, in this analysis, we did not take into account the coating pigment nor the compression pressure during fixation. Obviously, more detailed studies would be required on the heat transfer in the nip and on the thermomechanical properties of the pigment-containing coating layer.



4. The coating pore size distribution of the reference paper and DST papers with 10 pph of Latex 45-M or 10 pph of Latex 51-H.

Properties and structure of the coating

Surface strength. In most of the cases, the IGT surface strength of the DST papers was below that of the reference paper (0.35 m/s), ranging from 0.12 m/s to 0.35 m/s, as shown in Table IV. The proposed causes are aggregation related to the low- T_g latexes and, on the other hand, the low deformability of the layer with high- T_g binders.

Aggregation combined with an insufficient deformation during the fixing phase may have led to a smaller contact area for adhesion to the paper surface. However, additional contributing factors may be the rather low coating penetration and the rupture of the layer during fixation as a result of binder sticking.

Each coating layer that had a surface strength that was too low to measure is noted with a dash in Table IV. Pretreatment was found to have no notable effect on the surface strength.

With stiff latexes, an increased latex content improved surface strength. Surface strengths comparable to that of the reference were achieved with coatings containing 20 pph of Latex 51-H.

Mechanical performance. The factors that directly affect the mechanical performance of the papers of low binder contents include the pore structure of the coating layer [12], interactions between the binder and the pigment particles [13], the binder distribution, and the mechanical properties of the coating layer constituents [8, 13–15].

The uniformity of the pore structure [16, 17] certainly affects the coating interaction with the applied IGT testing oil and thus the surface strength. For the DST papers, the particle aggregation often resulted in coating voids with sizes in the micrometer range, which were irregularities that we also observed in the SEM micrographs. These irregularities can certainly be expected to reduce the picking strength.

Pore size and porosity. The coating layers we produced by DST exhibited broader pore size distributions than did the reference coating. Furthermore, the dominant pore size of the reference coating was smaller, as measured by mercury porosimetry and as shown in Fig. 4.

The coating porosity values were evaluated in the pore size range of 0.02–0.5 μm. The porosity of the reference coating (37%) was lower than that of the DST coatings. With the stiff latexes, 24-H and 51-H, porosities were 45–55%. With the soft latexes, 24-L and 45-M, porosity values were closer to 40%. Significant differences in the dominant pore size were not found with different coating compositions: the size was about 0.5–0.8 μm (as opposed to about 0.1 μm in the reference coating).

An increase in the binder content resulted in a dense coating layer in the submicron range, but the larger voids remained. It appeared difficult to reduce the aggregation-induced porosity in the larger pore size region and at the same time enhance the

porosity in the region of pore sizes close to 0.1 μm . Obviously the lower porosity and pore size is likely to contribute to the higher IGT surface strength of the reference paper.

Pore structure and surface strength.

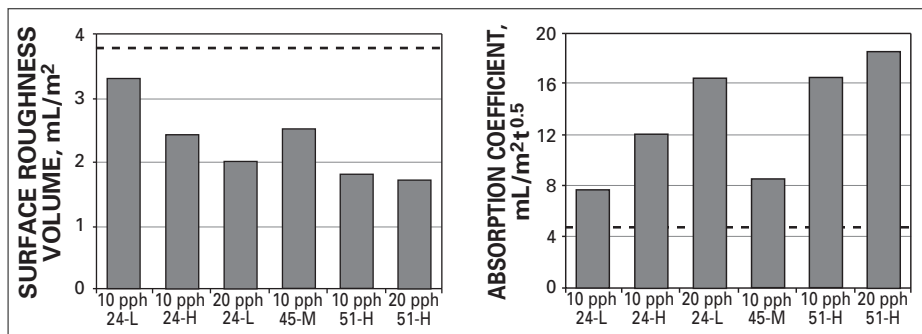
The effects of the temperature-dependent viscoelastic properties of the binder on the coating pore structure have been discussed elsewhere [7, 14, 18–22]. The general conclusion is that soft binders contribute to form coatings with low void volumes and high surface strengths.

When DST papers are produced with dried coating particles, a low degree of aggregation can be related to high- T_g and high-gel latexes, which lead to a more homogeneous coating structure than when softer latexes are used. This outcome, combined with sufficient deformation and sticking resistance during calendering, may have led to the higher surface strength of coatings containing stiff latexes. However, using fine coating particles with a low- T_g binder could be beneficial to the surface strength of DST papers.

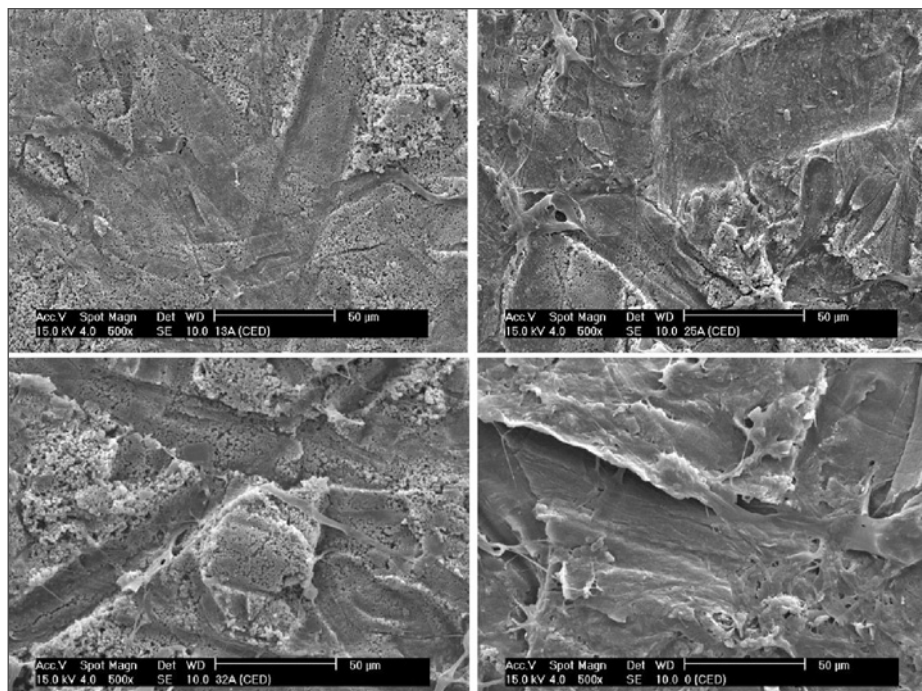
Sintering vs. consolidation. The “consolidation” of the DST coating pore structure can be compared with the corresponding course of events for wet-laid and dry-coalesced coatings.

In wet-laid coatings, a sintering treatment takes place after the coating dries below the binder softening temperature. This sintering effect results in a more porous structure and larger pore sizes than those of wet-coalesced layers, which undergo more shrinkage as a result of the capillary effect of water [23–25]. The sintering of the DST coating binder during the thermomechanical treatment may be one reason for the large pores in the coatings, but the primary aggregates, the type of pigments, and the compression during the fixation also play roles.

Slightly smaller pore sizes were observed after an increase in the nip dwell time and an increase in the linear load; this decrease in pore size usually reduced the porosity of DST coatings [5]. As with wet-laid coatings, the dominant pore size and the size distribution might be tunable with the size and the shape of the pigment [26], but the most important issue here is avoiding the particle aggregation.



5. Surface roughness volume (left) and absorption coefficient (right) of DST papers. Reference values were 3.8 mL/m² and 4.7 mL/m²·t^{1/2}, respectively.



6. Coating reverse side structure of three DST papers and the blade-coated reference paper (lower right). The DST coatings contained 10 pph of Latex 51-H (upper left), 10 pph of Latex 45-M (upper right), or 20 pph of Latex 51-H (lower left).

Oil absorption. The oil absorption behavior of the papers at short contact times was determined using a moving nozzle applicator (KCL, Finland). Rapid initial uptake of oil and a low steady-rate absorption indicate a porous surface and dense interior parts of the coating [27, 28]. This kind of absorption behavior was observed with the reference coating, represented in Fig. 5 by the lines with dashes.

In contrast, DST papers had relatively sealed surfaces and interiors that were strongly absorbing. The thermomechanical fixation was likely more effective in densifying the coating surface than was the soft calendering of the commercial reference paper.

The coating composition of the DST papers had a clear effect on the absorption characteristics. With low-gel latexes, the more pronounced flow and the possible sticking both contributed to a higher surface pore volume. An increase in

the latex content reduced the surface absorptivity, as expected. The more open inner coating structure achieved with high-gel binders was reflected in higher values of the steady-rate absorption coefficient. With stiff latexes, an increase in the latex content increased the degree of aggregation, which presumably resulted in larger voids in the interior regions than in the finer structure achieved with a low binder content.

Surface roughness. The Bendtsen surface roughness of the DST papers was consistent with the surface roughness volumes. The roughness values ranged from 15 mL/min to 25 mL/min, being significantly lower than for the reference at 61 mL/min. The roughness values according to PPS-s10 ranged between 1.05 and 1.25 μm for the DST papers, with the reference having a value of 2.95 μm . Altogether, the variation of the surface roughness was less

for each of the DST papers.

Coating penetration into the paper surface. With wet-laid coatings, an important factor in the coating-paper adhesion is the formation of an interphase region. In this region, some amount of the coating penetrates the sheet surface, depending on the roughness and openness of the base sheet [28, 29]. In DST, the anchoring of the layer to the base substrate by material penetration is diminished (Fig. 2).

Therefore, the surface properties of the substrate, such as its surface pore structure and surface energy, are not as critical as they are for wet-laid coatings. During fixing, the flow of the polymeric binder material to contact the rough and porous fibrous surface results in mechanical interlocking of the coating layer. The rheological properties of the binder affect the degree of penetration and the contact area between the adhering materials.

We also studied the coating-paper interface by exposing the reverse side of the coating structure. We dissolved the fibers in a CED (cupriethylene diamine) solution [30] and used SEM to make observations. On the reverse side surface of the reference coating (the lower right micrograph in Fig. 6), the layer forms connections at the fiber boundaries, originating from interlocking arms. The layer also exhibited relatively high thickness variations. With DST coatings, the topographical changes were mainly caused by fiber depressions in the surface.

At 10 pph of Latex 51-H, the reverse side of the coating was smoother than that of the reference, indicating a lower degree of mechanical interlocking. With a softer latex, 45-M, the "replica" of the fibrous surface on the coating reverse side was rougher and more dense, indicating that the latex penetrated deeper and formed a film. An increase in the amount of the stiff latex in the coating seemed to increase the coating-paper contact area (lower left micrograph in Fig. 6).

A high interfacial roughness corresponded to some extent with high surface strength of the papers. The surface strength of a DST coating containing Latex 51-H increased from 0.18 m/s to 0.35 m/s as the latex content was increased from 10 pph to 20 pph. A surface strength comparable to that of the reference (0.35 m/s) was obtained with a coating containing 20 pph of Latex 51-H. Still, the reverse side of the DST-coating

appears more porous and less coarse on the micrometer scale. This outcome suggests that it is possible in DST coatings to enhance additional interactions to compensate for the low penetration.

At a low binder content, when aggregation was a less of a problem, the DST fixation process seemed efficient in smoothing and densifying the coating surface. With soft latexes, a more open surface was formed as a result of an enhanced polymer film formation in the vicinity of the heated roll, which is consistent with the behavior of wet-laid coatings. However, soft latexes produce a denser coating interior and a denser coating-paper interface than formed with stiff latexes.

CONCLUSIONS

The processability in DST as well as the properties of the final coated paper are markedly affected by the temperature-dependent, viscoelastic properties of the latex binder used in the coating formulation. The conditions in the fixation phase of DST should have a strong bearing on the choice of the binder.

The binder also affects the degree of aggregation of the dried coating particles. In general, a polymeric binder with a low T_g induced higher degrees of aggregation, which may overshadow the improved binding properties associated with these binders. The preparation of coating particles through drying may cause variations in the binder distribution of the DST coating layer compared to the coating distribution in the conventional process.

With a high degree of aggregation, the fixed layer appeared aggregated as well, since the calendering was more efficient in compacting the surface of the coating than in compacting the interior parts. With finer coating particles, associated with high- T_g latexes in this study, the coating layer may deposit more homogeneously with the electrostatic application and may be further compacted during calendering. The resulting coating layer may thus be expected to have a higher degree of integrity and a better resistance against surface picking.

In general, using latex binders with a high gel content (corresponding to a high stiffness at elevated temperatures) reduced sticking deposits on the hot fixing roll. This finding is in agreement with

observations reported in conjunction with the calendering of conventionally coated papers at elevated temperatures. The surface strength of the papers produced by DST was favored by a high gel content and a high T_g of the latex binder, both of which seem to be beneficial in obtaining fine coating powders.

Comparisons between the DST papers and conventionally coated papers revealed some structural differences attributed to the properties of the coating particles. In most cases, a surface smoothness similar to that of the reference was attainable, but the coating structure produced by DST was more open in certain respects than that of conventional wet-coated papers. This greater openness is reflected in the larger pore size, the higher porosity of the layers, and a higher oil absorption coefficient. Binders with high gel content and high stiffness during fixation gave structures that were more open.

The coating penetration into the paper substrate was significantly less for the coated papers produced by DST than for the conventionally coated reference paper. Improving the cohesion and the adhesion properties of DST pigment coatings requires small coating particles and binders with enhanced film-forming ability and elasticity (or a sufficiently low T_g and a sufficiently high gel content). **TJ**

ACKNOWLEDGMENT

We thank the National Technology Agency (Finland), Metso Paper Inc., and Dynea Chemicals Inc. for their financial support of this work.

LITERATURE CITED

1. Maijala, J., Putkisto, K., and Grön, J., *TAPPI 2002 Coating Conference Proceedings*, TAPPI PRESS, Atlanta, p. 501.
2. Maijala, J., Putkisto, K., and Grön, J., *11th International Coating Science and Technology Symposium Proceedings*, ISCST, 2002, p. 67.
3. Putkisto, K., Maijala, J., Grön, J., and Rigdahl, M., *Nordic Pulp Paper Res. J.* 18(2): 226(2003).
4. Putkisto, K., Maijala, J., Grön, J., and Rigdahl, M., *TAPPI 2003 Advanced Coating Fundamentals Symposium Proceedings*, TAPPI PRESS, Atlanta, Session 1:2.
5. Putkisto, K., Maijala, J., Grön, J.,

- and Rigdahl, M., *5th International Paper Coating and Chemistry Symposium 2003 Preprints*, PAP-TAC, Montreal, QC, Canada, p. 117.
6. Wikström, M., Carlsson, R. and Salminen, P., *20th PTS Streicherei-Symposium Proceedings*, PTS, München, Germany, 2001, p. 42-1.
 7. Kan, C.S., Kim, L.H., Lee, D.I., and van Gilder, R.L., *TAPPI J.* 80(5): 191(1997).
 8. Mikkilä, J., Järvinen, H., Starck, et al., *20th PTS Streicherei-Symposium Proceedings*, PTS, München, Germany, 2001, p. 41-1.
 9. Majjala, J., Keitamo, J., Grön, J., and Kettunen, L., *TAPPI 2003 Advanced Coating Fundamentals Symposium Proceedings*, TAPPI PRESS, Atlanta, Session 1:1.
 10. Richard, J., *Polymer* 33: 562(1992).
 11. Ferry, J.D., *Viscoelastic Properties of Polymers*, John Wiley, New York, 1980, p. 264.
 12. Lohmander, S., "The influence of particle shape of coating pigments on their packing ability and on the flow properties of coating colors," Ph.D. thesis, KTH Royal Institute of Technology, Stockholm, Sweden, 2000.
 13. Parpaillon, M., Engström, G., Pettersson, et al., *J. Appl. Polym. Sci.* 30: 581(1985).
 14. Rigdahl, M., Lason, L., Hagen, et al., *J. Appl. Polym. Sci.* 63: 661(1997).
 15. Heiser, E.J., *Pulp & Paper* 55(5): 66(1981).
 16. Engström, G., Rigdahl, M., Kline, J., and Ahlroos, J., *TAPPI J.* 74(5): 171(1991).
 17. Kumano, A., Higuchi, A., Watanabe, T., and Matsui, H., *TAPPI 1993 Coating Conference Proceedings*, TAPPI PRESS, Atlanta, p. 45.
 18. LePoutre, P.F., and Rezanowich, A., *TAPPI J.* 60(11): 86(1977).
 19. Watanabe, J., and LePoutre, P.F., *TAPPI 1982 Coating Conference Proceedings*, TAPPI PRESS, Atlanta, p. 181.
 20. Yamawaki, K., Sasagawa, Y., and Tsuji, A., *TAPPI 1991 Coating Conference Proceedings*, TAPPI PRESS, Atlanta, p. 199.
 21. LePoutre, P.F., and Rigdahl, M., *J. Matl. Sci.* 24: 2971(1989).
 22. Jud, C., Hahn, N., and Kan, C., *TAPPI 2000 Coating Conference Proceedings*, TAPPI PRESS, Atlanta, p. 431.
 23. Al-Turaif, H., and LePoutre, P.F., *J. Appl. Polymer Sci.* 82: 968(2001).
 24. LePoutre, P.F., and Alince, B., *J. Appl. Polym. Sci.* 26: 791(1981).
 25. Stanislawski, A., and LePoutre, P.F., *TAPPI J.* 79(5): 117(1996).
 26. Johnson, R.W., Abrams, L., Maynard R.B., and Amick, T.J., *TAPPI J.* 82(1): 239(1999).
 27. Groves, G., Penson, J.E., and Ruggles, C., *TAPPI 1993 Coating Conference Proceedings*, TAPPI PRESS, Atlanta, p. 187.
 28. Dickson, R., Forsström, U., and Grön, J., *TAPPI 2000 Coating Conference Proceedings*, TAPPI PRESS, Atlanta, p. 167.
 29. Huang, T., and LePoutre, P.F., *TAPPI J.* 81(8): 145(1998).
 30. Dickson, R., and LePoutre, P.F., *TAPPI J.* 80(11): 149(1997).

Received: May 14, 2003
Revised: February 20, 2004
Accepted: March 11, 2004

Presented at the TAPPI 2003 Advanced Coating Fundamentals Symposium, May 2003, Chicago, Illinois.

This paper is also published on TAPPI's web site <www.tappi.org> and summarized in the June *Solutions! for People, Processes and Paper* magazine (Vol. 87 No. 6).

INSIGHTS FROM THE AUTHORS

Dry surface treatment is a novel technique that was generated in 1997 and has undergone the major technical and material development during the last few years. As far as we know, there are no other research groups reporting on this topic in the pulp and paper field. The results of this study on DST support earlier findings of our group. In essence, optimizing the formation and the properties of the coating layer requires a balance between the properties of the coating materials and the fixation conditions.

Obtaining coating particles of suitable fine size and achieving sufficient repeatability of the coating operation were the most difficult aspects of this research. We prepared the particles by drying coating suspensions, which is a route that is not feasible on any larger scale than laboratory experiments. Avoiding the drying-related aggregation is one of the key issues, however, because the size of the coating particle aggregates strongly influences the uniformity of the coating layer.

Next, we will concentrate on applying the powder in better controllable ways and double-sided, and on evaluating different ways to manufacture the coating powders in the dry state.

On a larger scale, an increase in the speed may bring up new challenges with respect to the electrostatic deposition and the fixation. Evaluation of the overall feasibility of this novel approach requires consideration of the different stages: the materials preparation, the integration to the papermaking line, and the surface finishing.

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