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## PEER-REVIEWED **DRY SURFACE TREATMENT**

# *Polymer coating of paper using dry surface treatment: Coating structure and performance*

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**ABSTRACT:** What effects do fixation conditions and the polymer thermomechanical properties have on the structure and performance of coatings prepared in a laboratory-scale DST prototype? At low coat weights, the prerequisite for homogeneous layers was a fine particle size distribution. At high coat weights, it was important to enhance polymer flow during fixation to obtain a coherent and uniform film. Small particle size, high polymer deformability, and high coat weight were beneficial in sealing the paper substrate. Prolonging the dwell time in the DST nip to enhance polymer flow is not a promising solution because of the problem of sticking. Instead, a better way to reach a balance between polymer characteristics and fixation conditions is to obtain better adapted polymers. Nevertheless, DST provides an alternative approach for manufacturing coated and laminated structures.

**Application:** Dry surface treatment is a solventless, noncontact process that should let papermakers use materials not practical to use in extrusion and wet-laid coating.

Polymer coating of paper, paperboard, plastic films, and metallic foils is intended to improve the protective prop-

erties and the appearance of the package material. Conventional off-line coating and lamination techniques make it possi-

ble to form different types of converting products, although these techniques have limitations in capacity and flexibility.

The performance of the converting product must meet the requirements on cost-effectiveness during manufacture and use and during recycling and disposal. Therefore, a compromise must often be found between the costs and the desired characteristics of the materials [1].

The development of the dry surface treatment (DST) has been aimed at more compact, flexible, cost-effective converting techniques. With DST, the coating and surface smoothening are accomplished in a combined process. Dry fluidized coating particles are deposited electrostatically onto the substrate, followed by thermomechanical fusing in a heated roll nip.

Whereas previous studies with the DST method have concentrated on pigment-coated papers [2-5], we used the same principles in our laboratory experiments to produce thermoplastic polymer coatings. To some extent, the same hypothesis put to work for the pigment coatings holds for the polymer coatings produced with DST: a small particle size is beneficial in charging and in the initial electrostatic attachment [5-8]. Furthermore, during fixation, a small particle size and an even layer thickness help to make compaction uniform. Material homogenization and film formation during the transient fixation stage help to eliminate density variations and voids in the layer [9, 10].

Our aim was to evaluate the feasibility of using the DST process to apply polymer coatings to paper. We compared polyethylene-based coatings produced by DST and by extrusion coating in terms of some structural characteristics and the coating performance. Our focus was on different grades of low-density polyethylene with different particle sizes and melt flow characteristics. The results should be regarded as being preliminary in nature. Furthermore, the comparison with extrusion-coated substrates is valid only for the material used here and cannot be claimed to have broad validity.

## ADVANTAGES OF THE DST TECHNIQUE

The DST technique may allow papermakers to process materials that are not manageable in the conventional processes. In extrusion coating, the material-related factors that limit the coating

speed and efficiency include the melt strength, the neck-in, and the adhesion properties of the film drawn from the melt. In DST, the noncontact application of coating may help reduce the web stress, and the coating can be fixed at a low elongational shear rate compared to the stretching of the melt in extrusion coating.

Temperature-sensitive materials that may suffer from changes in density or composition during the melt processing in extrusion [11] may be suitable for DST. In DST, the nip dwell time may be as short as a few milliseconds, and the compression pressure compensates for the lower temperatures. In short, DST should prove to be less dependent on the material than extrusion coating is.

Dispersion and solvent-based coating methods enable papermakers to produce paper with low coating weights and to use various materials that are not extrudable. The drawbacks in these methods include an energy-intensive evaporation and drying stage and the need to handle media in liquid and gaseous forms. One restriction in using liquid media is that the base substrate may be chemically or physically sensitive. The DST process is more or less solvent-free, and no drying is required after the coating is applied.

In extrusion as well as in DST, the processability of coating materials is affected by the solid feed properties (friction and bulk density), the specific thermodynamic properties (specific heat, heat of fusion, melt density, and thermal diffusivity), the viscoelastic properties, and the chemical nature of the polymer [1, 8]. The DST heat treatment is ideally used for controlling the rheological behavior of the polymer to form an adequate film through sintering and molecular diffusion and to allow the polymer to penetrate the porous surface of the paper and anchor in it. Furthermore, the fusing conditions should allow the stress in the coating layer to relax, since structural defects,

|           | Density,<br>g/m <sup>3</sup> | Melting<br>temp.,<br>°C | Melt<br>index,<br>g/10 min | Particle<br>Powder<br>preparation<br>method | size<br>range,<br>µm |
|-----------|------------------------------|-------------------------|----------------------------|---------------------------------------------|----------------------|
| LDPE      |                              |                         |                            |                                             |                      |
| A         | 0.92                         | 124                     | 0.95                       | A                                           | 20-500               |
| B         | 0.92                         | 115                     | 1.77                       | A                                           | 100-500              |
| C         | 0.92                         | 123                     | 2.62                       | A                                           | 20-100               |
| D         | 0.92                         | 111                     | 3.81                       | B                                           | 5-50                 |
| E         | 0.92                         | 111                     | 3.56                       | B                                           | 5-50                 |
| Polyamide | 1.19                         | 127*                    | —                          | C                                           | 20-100               |

\*Glass transition temperature.  
A: Obtained in a particulate form from a reactor and ground.  
B: Obtained by drying a polymer dispersion after emulsion polymerization.  
C: Ground from granulates.

## I. Properties of the coating polymers and particles.

such as a weakening of the coating-substrate interface, may appear otherwise [8]. An effective heat-induced softening of the polymer must be combined with adequate mechanical properties at elevated temperatures to keep the polymer from sticking to the hot roll surface [4].

Multilayer coatings can be produced with the DST in stepped application stages, with or without an intermediate fixation. The degree to which the polymers mix and penetrate the corresponding interphase region may be adjusted by the thermomechanical treatment. In extrusion, the production of multilayers requires either several successive extruders or coextrusion of the layers. In some cases, the disturbance of the coextrusion process may cause the layers to rearrange [12], impairing the structure's barrier properties.

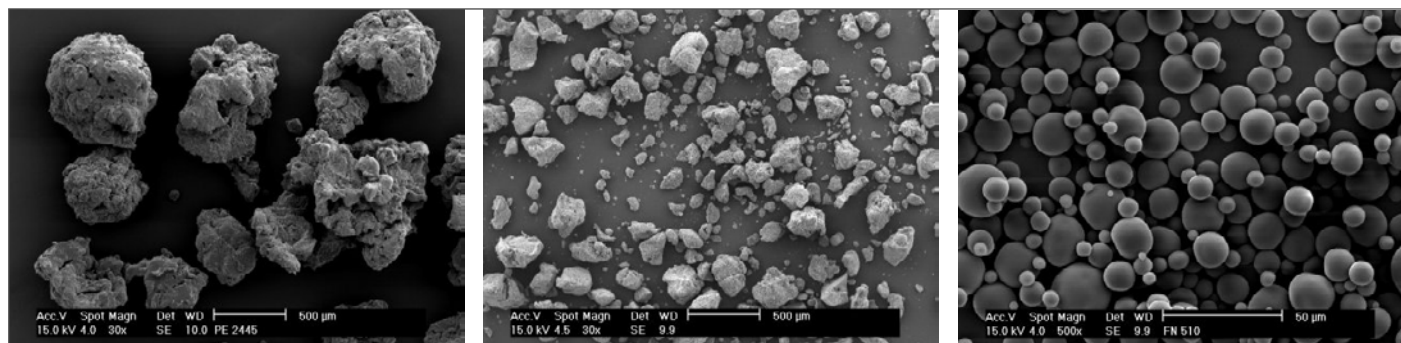
## MATERIALS AND METHODS

### Coating components

Table I lists the properties of the coating polymers and the particles used in the DST trials. The coating particles were of three origins. Three grades were obtained in a particulate form from a reactor and ground (A), two were obtained by drying a polymer dispersion after emulsion polymerization (B), and one was ground from granulates (C). The low-density polyethylene (LDPE) and amorphous polyamide (PA) granulates were ground in a high-power laboratory grinder in liquid nitrogen.

The glass transition temperature,  $T_g$ , of the polyamide and the melting temperatures,  $T_m$ , of the LDPE polymers were evaluated by differential scanning calorimetry with a heating rate of 10 °C/min. The melt index (MFR) of the

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1. SEM micrographs of LDPE-C particles before grinding (left, scale bar of 500 µm), LDPE-A particles after grinding (middle, 500 µm), and LDPE-D particles prepared by drying a polymer dispersion (right, 50 µm).

| Paper substrate | Basis weight, g/m <sup>2</sup> | Coat weight, g/m <sup>2</sup> |
|-----------------|--------------------------------|-------------------------------|
| Woodfree        | 70                             | 30                            |
|                 | 80                             | 12                            |
|                 | 90                             | 10                            |
| Kraft           | 70                             | 23                            |
| Laminated kraft |                                |                               |
| Board           | 200                            | 25                            |
| Paper           | 140                            | 13                            |

## II. Extrusion-coated reference papers.

polyethylenes was determined according to ISO 1133 (190°C, 2.160 kg). The particle size range was assessed with scanning electron microscopy (SEM).

Figure 1 shows particles from different sources: particles directly from a reactor and before grinding (left), a ground powder (middle), and particles after dispersion drying (right). Polymers with a higher melting temperature appeared to produce finer particles during grinding, although the size distribution was broad in all cases. The  $T_m$  (or  $T_g$ ) might thus indicate the brittleness of the polymer even at low temperatures. The difference between grades LDPE-D and LDPE-E was that LDPE-E contained a few weight percents of a flow-enhancing additive, a flaky mineral pigment with an average particle size of about 1 µm. This flow-enhancing pigment reduced the aggregation of particles and improved the fluidity of the powder in handling and in the fluidization process.

### Dry surface treatment of papers

Three woodfree, filled papers were used as the substrates, with basis weights of 80, 90, and 140 g/m<sup>2</sup> and PPS-s20 surface roughness values (SCAN-P 21:67) of 6.5 µm, 6.5 µm, and 5.6 µm, respectively. In application, the moisture content of the papers (SCAN-P 4:63) were 4.5%, 4.5%, and 6%, respectively.

The coating particles were applied onto the paper with a laboratory-scale application unit [2, 3]. A 95 kV negative voltage with a maximum current of 100 mA was used to charge the particles and to form the electric deposition field (Sure Coat, Nordson Corp.). The average coat weights were 10, 12, 25, and 30 g/m<sup>2</sup>. The coatings were fixed with a laboratory calender with one pass through a roll nip consisting of a heated roll (at 200°C) with a Teflon-based cover and a polymeric backing roll, at a speed of 10 m/min and a linear load of 45 kN/m (a dwell time of 60 ms). The coated paper was cooled in air after the fixation nip, and no chilling or after-treatment was used.

A few 90 g/m<sup>2</sup> papers, with an average coat weight of 12 g/m<sup>2</sup> of LDPE-E, were fixed with two passes in the nip. A two-layer polymer coating was formed on 90 g/m<sup>2</sup> paper by first fixing 12 g/m<sup>2</sup> of LDPE-E and subsequently 6 g/m<sup>2</sup> of PA. The fixing conditions were similar to those for the coat weights of 10, 12, 25, and 30 g/m<sup>2</sup>.

### Examination of the coating layers

The coated surfaces and the cross-sections of the polymer-coated papers were examined with SEM. Cross-sections were obtained by fracturing in liquid nitrogen. Before examination, the specimens were coated with gold.

Because of the low basis weight of the base papers and the low coat weights, the adhesion between the coating and the substrate was impossible to examine with a standard peeling test (such as a Houndsfield tester). However, we achieved some degree of classification between the samples by using a tape test (modified ASTM D 3359). Additionally, we examined the degree of mechanical interlocking by separating the coating layer from the paper according to a method

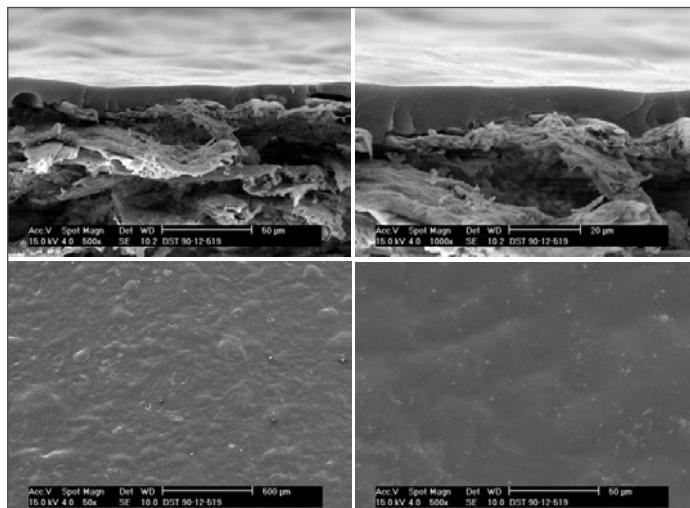
described by Dickson and LePoutre [13] and then using SEM. Dissolving the fibers in a cupriethylene diamine (CED) solution exposed the coating reverse side structure and made it possible for us to examine the interface between the paper and the coating.

We determined the existence of pinholes in the coating layers according to a modified version of PTN 8-70 (Packforsk, Sweden), using colored pinene turpentine. A thin layer of the solution was applied with a brush onto the coated surface. After being allowed to dry at room temperature for 15 min, the surface was wiped dry. We then examined the penetration of the liquid through the paper to the reverse side. If any well-defined pinholes were discerned, we counted the number within an area of 50 cm<sup>2</sup>. The frequency of pinholes was given as an average number per square meter. Three samples were examined for each determination.

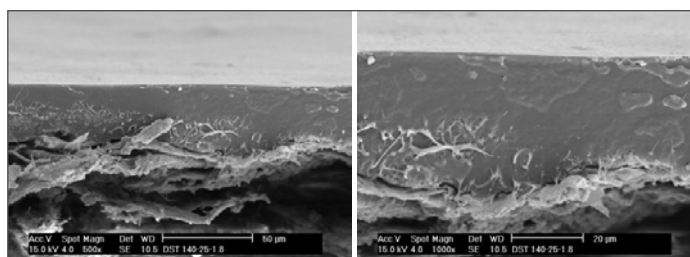
The water vapor transmission rate (WVTR) of the coated materials was determined at 23°C and a relative humidity of 50% (SCAN-P 22:68). The area for vapor transmission in each sample was approximately 40 cm<sup>2</sup>. The average WVTR value for three samples was calculated from the gravimetric uptake of water at 24 h and 48 h after each sample was exposed to the controlled conditions. The result for each coating represents an average weight of (transmitted) water per unit area within 24 h.

### Reference materials

As references, we used a set of the polyethylene extrusion-coated papers, as listed in Table II. These papers were produced from different types of pulp and had different basis weights and coat weights. The properties of the reference base papers and the properties of the polymer grades were not available.



**2. The cross section (upper row, scale bars of 50 and 20 µm) and the surface (lower row, scale bars of 500 and 50 µm) of a DST paper with an average coat weight of 12 g/m² of LDPE-E on a paper with a basis weight of 90 g/m².**



**4. The cross section (scale bars of 50 and 20 µm) of a DST paper with an average coat weight of 25 g/m² of LDPE-B on a paper with a basis weight of 140 g/m².**

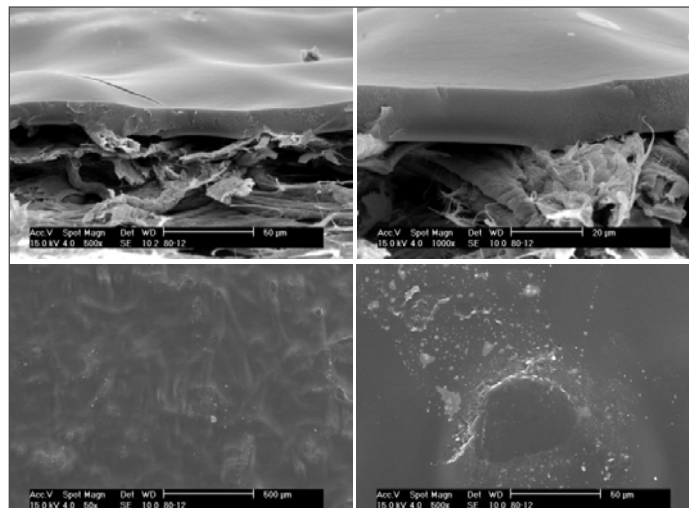
However, the PE papers were sufficient as references against which to compare the coating characteristics produced in the different processes.

## RESULTS AND DISCUSSION

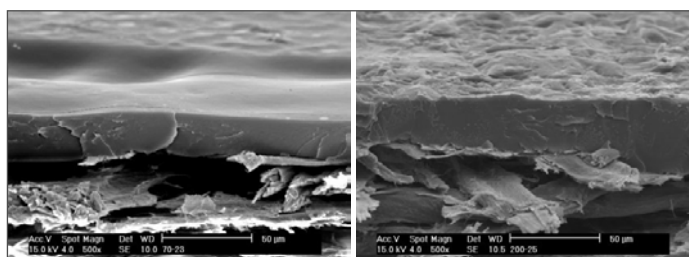
### Coating surfaces and SEM cross sections

The very small particle size of polymers LDPE-D and LDPE-E enabled us to reach low coat weights in DST coatings. The coating film formed by LDPE-E seemed to be continuous and homogeneous at a coat weight of 12 g/m² and appeared to form close contact with the base paper (Fig. 2). However, the coating surface was rather rough. This roughness was probably caused by incomplete spreading of the particles and insufficient homogenization of the film in some regions and by the roughness of the base paper (where the shapes of the fibers “showed through” the coating layer).

The extrusion-coated reference paper at the same average coat weight of 12 g/m² also appeared to be rough, which was associated with the roughness of the base paper. Figure 3 shows this roughness, where distinguishable fiber shapes can be seen. In addition, the surface of the extrusion-coated reference suffered from fibers pointing out from the surface and from other surface irregularities. These irregularities can be seen in the lower right micrograph in Fig. 3. The extrusion-coated film seemed to have only a few contact points with the substrate, in



**3. The cross section (upper row, scale bars of 50 and 20 µm) and the surface (lower row, scale bars of 500 and 50 µm) of an extrusion-coated reference with an average coat weight of 12 g/m² on a paper with a basis weight of 80 g/m².**



**5. The cross-sections (scale bars of 50 µm) of extrusion-coated references with an average coat weight of 23 g/m² on a paper with a basis weight of 70 g/m² (left) and with an average coat weight of 25 g/m² on a board with a basis weight of 200 g/m² (right).**

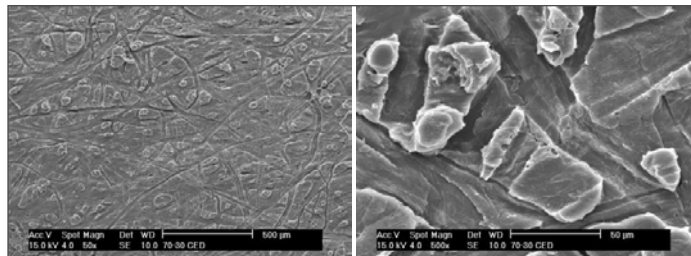
which areas the polymer had covered the fiber surfaces. The film thickness was relatively constant, which indicates that the interfacial region between the coating and the paper surface contained voids.

At higher coat weights, the irregularly shaped particles of ground polymer were also found to be suitable for the DST process. Additionally, the higher coat weights reduced the effect of the roughness of the base substrate on the roughness of the coating surface. The fixing conditions seemed adequate for forming a film of a relatively thick layer of viscous polymer LDPE-B (with a MFR of 1.77 g/10 min), as shown in Fig. 4.

Compared to LDPE-A, the more enhanced flow of LDPE-B and LDPE-C during fixation apparently improved the uniformity of the coating layer. A high stiffness and a large average particle size are likely to cause uneven coverage and therefore uneven thickness of the coating layer. Variation in coat weight is associated with the filling out of the surface irregularities of the substrate, and the structure of the DST coatings resembled barrier coatings produced by dispersion coating [14, 15].

The surface of the extrusion-coated reference papers at a higher coat weight was found to be rather rough, as Fig. 5 indicates. The surface may appear smooth on a small scale but wavy on a larger scale, or it may also appear rough on a smaller scale,

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**6. SEM micrographs of the reverse side of an extrusion-coated reference paper (scale bars of 500 and 50 µm) with a basis weight of 70 g/m<sup>2</sup> and a coat weight of 30 g/m<sup>2</sup>.**

as noted with the two reference papers. Again, judging from Fig. 5 (left), the contact of the coating layer with the base paper seems more localized.

## Adhesion between coating and the substrate

Although the degree of adhesion was assessed only qualitatively with the tape test, some general observations emphasize the differences between the coatings produced by extrusion coating and those produced by DST. In general, the extrusion-coated references exhibited higher adhesion than the cohesion of the paper itself, resulting in delamination of the paper.

The DST papers with coat weights of 12 g/m<sup>2</sup> and 25 g/m<sup>2</sup> for LDPE-D and for LDPE-E exhibited qualitatively lower adhesion than the extrusion-coated references showed. At the lower coat weight, the adhesion strength was sufficient to tear off the surface fibers at the beginning of the peel, but the coating cohesion generally seemed insufficient (with rupture of the polymer film). At 25 g/m<sup>2</sup>, the whole coating layer was peeled off with the tape from the very beginning of the peel, indicating incomplete fusion of a thick layer. The DST papers fixed with two passes in the nip (coat weight of 12 g/m<sup>2</sup> of LDPE-E) exhibited a fiber-tear type of adhesion to the paper surface, indicating that the contact with the substrate improved with the longer heating time. However, enhancing the flow of the polymer must be balanced against the tendency for blocking to occur.

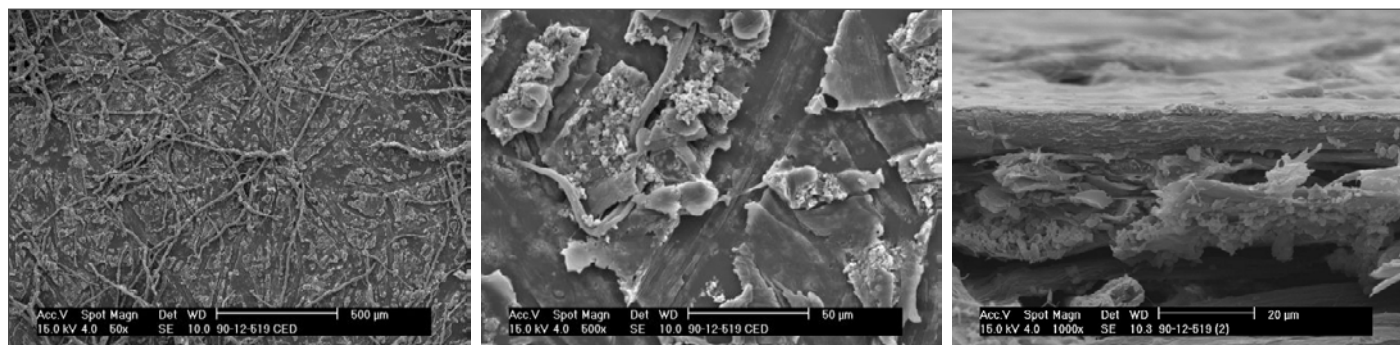
Some of the DST papers and the extrusion-coated reference papers were treated with a solution of CED, and the structure of the exposed coating on the reverse side was examined by SEM. For the reference in Fig. 6, this coating structure was a very rough replica of the paper surface, having connective arms

deep between the fibers. The mechanical interlocking to the substrate would be expected to be significant.

The coating reverse side structure of the DST papers was different from that of the references. The extraction of the fibers (and fillers) was not complete, indicating some areas of high adhesion between the coating and the fibers, as shown in the left-hand and middle micrographs in Fig. 7. The example DST paper was fixed with two passes in the nip. (With only one pass through the nip, the sample preparation with CED failed because the cohesion of the polymer film was too low). The polymer had formed a film, and the attached fibers and fillers indicate a rough interface between the coating and the paper surface. Another factor was that the prolonged dwell time in the nip may cause the surface layer of the coating to rupture because the tendency to stick was enhanced. The consequence of a local failure of coating cohesion was a rough coating surface and irregularities in the film, as shown in the right-hand micrograph in Fig. 7.

The contribution from the mechanical interlocking to the adhesion is obviously important [13]. A possible reason for the somewhat lower adhesion of DST coatings may be a weak boundary layer in the surface regions of the paper arising from a low compatibility between the coating and the fibers. However, in some cases, the failure was related to the cohesion of the film, where a more efficient fusing of the polymer particles should then reduce the problem. Thus, insufficient penetration of the polymer into the paper surface may not be the sole factor responsible here. The chemical characteristics of the coating polymer and the surface to be coated are also important with regard to the wettability properties [16]. Consequently, it is possible to enhance the contact and penetration if necessary.

Functional groups can be incorporated into the polymer, either in the preparation phase or during the coating process (e.g., oxidation) to promote the “chemically related” adhesion to paper-based substrates. In extrusion, the oxidation of the polymer [17] and the treating or priming of the substrate (such as corona or flame treatments) are methods used to improve both chemical bonding and the surface area available for mechanical interactions [16, 17]. Most of the reference papers we used were produced with a type of discharge pretreatment, which could contribute to the observed difference. The ozone created in the application stage in the DST process may have the effect of chemically priming the substrate to some extent.



**7. SEM micrographs of a coating reverse side (left and middle, scale bars of 500 and 50 µm) and a cross section (right, scale bar of 20 µm) of a DST paper with a basis weight of 90 g/m<sup>2</sup> and a coat weight of 12 g/m<sup>2</sup> of LDPE-E, fixed with two passes in the nip.**

| Substrate  | Basis weight, g/m <sup>2</sup> | Coating polymer | Coat weight, g/m <sup>2</sup> | Pinhole count, per m <sup>2</sup> |           | WVTR, g/(m <sup>2</sup> 24 h) |           |
|------------|--------------------------------|-----------------|-------------------------------|-----------------------------------|-----------|-------------------------------|-----------|
|            |                                |                 |                               | Avg.                              | Std. dev. | Avg.                          | Std. dev. |
| Base paper | 90                             | —               | —                             | ∞                                 | n.a.      | 620                           | 60        |
| Base paper | 140                            | —               | —                             | ∞                                 | n.a.      | 670                           | 60        |
| Reference  | 90                             | PE              | 10                            | 1200                              | 1100      | 12                            | 1.2       |
| Reference  | 80                             | PE              | 12                            | 1000                              | 850       | n.a.                          | n.a.      |
| Reference  | 70                             | PE              | 23                            | 100                               | 140       | 2.3                           | 0.3       |
| Reference  | 200                            | PE              | 25                            | 0                                 | 0         | 2.7                           | 0.4       |
| DST        | 80                             | LDPE-A          | 30                            | n.a.                              | n.a.      | 30                            | 15        |
| DST        | 140                            | LDPE-B          | 25                            | ∞                                 | n.a.      | n.a.                          | n.a.      |
| DST        | 90                             | LDPE-D          | 10                            | ∞                                 | n.a.      | 98                            | 110       |
| DST        | 90                             | LDPE-D          | 12                            | ∞                                 | n.a.      | 52                            | 57        |
| DST        | 140                            | LDPE-D          | 25                            | 0                                 | 0         | 5.4                           | 1.1       |
| DST        | 90                             | LDPE-E          | 10                            | ∞                                 | n.a.      | 58                            | 24        |
| DST        | 90                             | LDPE-E          | 12                            | ∞                                 | n.a.      | 15                            | 7.6       |
| DST        | 140                            | LDPE-E          | 12                            | 3500                              | 700       | 14                            | 2.6       |
| DST        | 140                            | LDPE-E          | 25                            | 200                               | 100       | 5.2                           | 3.0       |

∞ = pinhole count of highly permeable materials, >10,000 m<sup>2</sup>.

### III. The pinhole and WVTR values of the DST base papers, the references, and the coated DST papers.

Other grades of polyethylene could probably improve the mechanical performance of the DST papers. The melt index of the grades used was quite low compared to that of grades that are sometimes used [18]. Furthermore, using grades with higher melt indices can have a positive influence on the penetration of the melt into the fiber structure as well as on the homogeneity and strength of the film.

#### Layer homogeneity and barrier properties

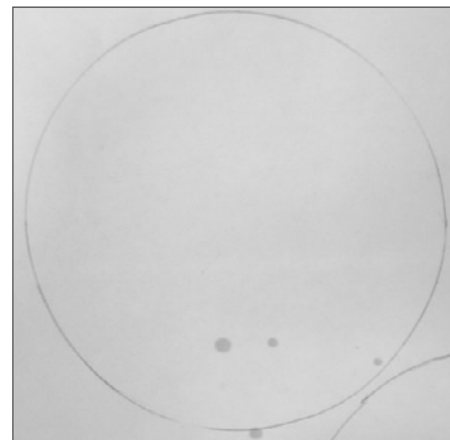
The sealing effect of the higher coat weight, examined by the pinhole frequency and the water vapor transmission rate, was notable for both the extrusion-coated and the DST papers. By testing on the uncoated backside of the reference samples and on the DST base papers, we were assured that the substrates were permeable to the pinhole test liquid. A direct comparison between the different types of coatings is limited, however, because the base substrates are different. In this sense, it was easier to compare the coatings with the WVTR values, which were more independent of the base substrate in indicating the permeability of the coatings. Furthermore, neither of these tests takes into account the size of the irregularities in the layer. Nevertheless, corresponding results were obtained in the two measurements: coatings that gave a lower pinhole frequency usually performed better in the WVTR analysis, as reflected in Table III.

With the reference papers, an increase in the coat weight from 10 g/m<sup>2</sup> to 25 g/m<sup>2</sup> reduced the number of pin-

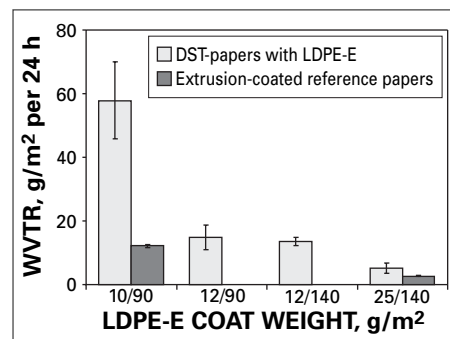
holes observed, from about 1000–2000 per square meter to zero. Comparing the extrusion-coated materials with average coat weights of 23 g/m<sup>2</sup> and 25 g/m<sup>2</sup>, we can expect the higher basis weight of the substrate of the latter to contribute to the lower pinhole count. The standard deviation was high in the cases in which pinholes were detected. This finding can be explained, at least to some extent, by the rather rough scale of the analysis and the small sample area. An example of a pinhole measurement sample is shown in Fig. 8.

The DST coatings exhibited different results with regard to the pinhole frequency, which was associated mainly with the average coat weight and the deformability of the polymers. On average, the pinhole count ranged from 3500 to over 10,000 per square meter for papers with coat weights of 10 g/m<sup>2</sup> and 12 g/m<sup>2</sup> for LDPE-D and for LDPE-E.

For average coat weights of 25 g/m<sup>2</sup> for LDPE-D and for LDPE-E on a paper with a basis weight of 140 g/m<sup>2</sup>, the average pinhole counts were 0 and 200 per square meter, respectively, for the DST papers. At high coat weights, the DST papers thus compared better against the extrusion-coated references. However, in cases in which the film formation appeared incomplete, typically with polymers LDPE-A, LDPE-B, and LDPE-C, there were visible voids in the coatings even at high coat weights. This finding emphasizes the importance of optimizing the polymer and particle properties with respect to the transient fixation process. Here, a small average particle



8. An example of pinhole measurement for an extrusion-coated paper with an average coat weight of 12 g/m<sup>2</sup> and a basis weight of 80 g/m<sup>2</sup> (reverse side of the paper with an analyzed area of 50 cm<sup>2</sup>).

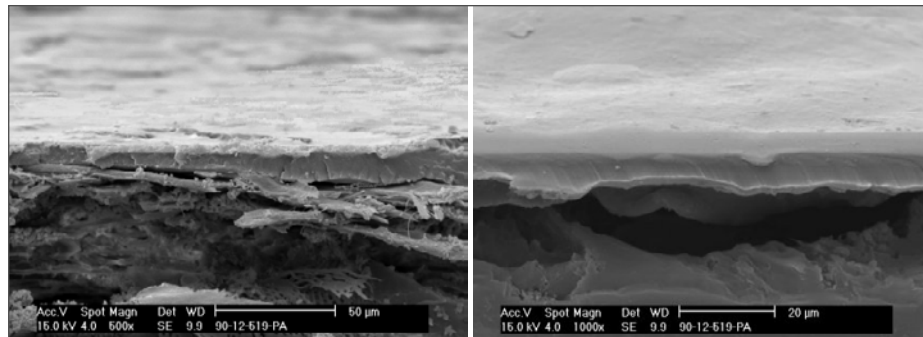


9. The average and standard deviation of WVTR values of DST papers with LDPE-E and two extrusion-coated reference papers as a function of coat weight. (On the horizontal axis, both the coat weight and the basis weight are in g/m<sup>2</sup>.)

size, a low melting temperature, and a high melt flow rate were beneficial to the uniformity of the DST coating layer.

With regard to WVTR, LDPE-E performed better than LDPE-D, which may perhaps be associated with the presence of the flaky filler particles in LDPE-E, which increase the tortuosity of the structure [16]. At an average coat weight of 10–12 g/m<sup>2</sup>, the DST papers exhibited much higher WVTR values (and more experimental scatter) than exhibited by the extrusion-coated reference at 12 g/m<sup>2</sup> per 24 h, although the rate was significantly lower than that of the uncoated base paper (Table III). At a coat weight of 12 g/m<sup>2</sup> of LDPE-E, the WVTR value was rather independent of the base paper (Fig. 9), but the paper with the

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**10. Cross section of a DST paper (scale bars of 50  $\mu\text{m}$  and 20  $\mu\text{m}$ ) with a two-layer coating consisting of a 12-g/m<sup>2</sup> layer of LDPE-E and a 6-g/m<sup>2</sup> layer of PA, fixed separately. The base paper had a basis weight of 90 g/m<sup>2</sup>.**

lower basis weight exhibited a higher standard deviation.

At higher coat weights, DST papers compared better with the references. The reference values at coat weights of 23–25 g/m<sup>2</sup> were around 2.5 g/m<sup>2</sup> per 24 h, whereas average values ranging from 5 to 30 g/m<sup>2</sup> per 24 h were obtained with DST, depending on the polymer. Here again, better barrier properties were attributed to a smaller particle size (better packing in the layer), a lower  $T_m$ , and a higher MFR of the polymer. As a reference, a typical WVTR value for a 25  $\mu\text{m}$  thick film of low-density polyethylene is approximately 4 g/m<sup>2</sup> per 24 h [1].

With this limited series of polymers, it was difficult to explicitly differentiate between the effects of polymer characteristics, particle size, coat weight, and the base substrate since these could not be changed independently. Grinding resulted in finer coating powders with high- $T_m$  polymers, but the stiffness may be expected to hinder the film formation in the nip. An increase in the coat weight and thickness set higher requirements for fixation. A higher roll surface temperature or a higher linear load may be advantageous in increasing the compression of the layer and enhancing the heat transfer. The use of grades with a higher melt index, and possibly a higher crystallinity [1, 19], could be a viable route to improving the sealing of the paper.

## **Multilayer coating with DST**

Difficulties in multilayer coating in coextrusion or in successive extrusion processes may be related to different flow behaviors of the layered polymers and may also be related to their mutual affinity [1, 20]. As an example of the capa-

bility of the DST process, we used powders of polymers with different thermo-mechanical characteristics, LDPE-E (underlayer, 12 g/m<sup>2</sup>) and PA (top layer, 6 g/m<sup>2</sup>), to produce a two-layer coating on a paper substrate (Fig. 10).

The surface of the PA coating was relatively rough because of the large variation in the particle size (Fig. 10, left). However, the bonding between the different polymers appeared to be uniform (Fig. 10, right). Thus, DST may constitute an interesting way to produce multilayered structures in a rather straightforward manner.

## **CONCLUSIONS**

DST appears to be a feasible approach that provides good contact between a polymeric coating and paper. The sealing capability of DST coatings improved with increasing coat weight, furthermore polymer properties such as a small particle size and a high melt index favor improved compaction and uniformity of the applied layer.

DST papers with low coat weights (10–12 g/m<sup>2</sup>) exhibited sealing capability only when the coating particle size was small (average at a maximum of 15–20  $\mu\text{m}$ ) and the deformability of the molten polymer was high enough for the film to form under the fixation conditions. The permeability of DST coatings was generally higher than that of extrusion-coated materials at equal coat weights. As with extrusion-coated papers at low coat weights, the roughness of the base paper is the limiting factor for the uniformity and integrity of the coating layer. At high coat weights, DST coatings produced with high-melt-index polymers

compared better with their extrusion-coated counterparts with regard to pin-hole frequency and water vapor permeability. DST fixation seemed adequate for the film formation and surface smoothing of the applied layers.

The mechanical performance of the DST coatings (most importantly adhesion to the paper and cohesion) require optimization, coupled with the adoption of selected process conditions. This optimization relates to particle size distribution, which should be as fine as possible, and to the melt index, which probably should be higher than the one used in this study. The particle size distribution is critical not only for particle packing in the deposited layer but also for compaction and homogenization of the melting material. If the powdered coating materials are prepared by grinding, the stiffness of the polymer may affect the particle size distribution.

A high melt index, corresponding to a lower molecular weight or a tailored molecular weight distribution, would promote film formation in the fixation step and enhance polymer penetration. The thermal and viscoelastic properties of polymers need to be in balance with the fixation conditions to achieve mechanical interlocking and to prevent the polymer from sticking to the heated roll. Polymers with a small particle size and a high melt index would improve the surface smoothness of the coated material.

DST appears to be a flexible method for producing multilayered structures and for processing different materials without major adjustments of the process. It can reduce or eliminate some of the limitations encountered with other techniques. Further studies are required, however, to confirm its feasibility for industrial production. **TJ**

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## INSIGHTS FROM THE AUTHORS

It would be advantageous to develop solventless, high-speed coating processes with less impact on the coating materials and the base substrate. Powder-based coating obtained through noncontact application in dry surface treatment (DST) offers a potential route. The DST coating processability and properties have a strong bearing on the properties of the coating particles and on the viscoelastic character of the coating polymers.

The DST technique has been studied since 1997, but there are no other research groups reporting on this topic. The main work on DST has been done with pigment coatings. However, work has also been done with polymeric coatings where, to a large extent, the powder-based layer corresponds in structure and calendaring behavior to conventional extrusion-coated and suspension-coated layers.

The most difficult aspect of this research was to obtain coating particles of suitably fine size. We prepared the particles by grinding bulk polymers or by drying polymer dispersions. For any larger scale production, more efficient preparation methods must be sought. The size of the coating particles and the melt viscosity strongly influence the uniformity of the coating layer. Thus, optimizing the materials for DST is one of the key issues if this approach is to be developed further.

The laboratory-scale experiments indicated a potential for high-coverage coatings at low coat

weights. In many respects, the coated papers were comparable to their extrusion-coated counterparts. However, the technique needs to be evaluated for its feasibility on a larger scale, where an increase in speed could introduce new challenges for electrostatic deposition and fixation. Compared to extrusion coating and solvent-base processes, the DST process is more compact and could reduce material costs. Next, we plan to evaluate ways to manufacture the coating powders and ways to improve control in applying the powder.

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