A study of how surface wetting may affect the coating layer when coating paper
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A study of how surface wetting may affect the coating layer when coating paper

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Abstract

Light Weight Coated (LWC) paper and many other paper grades are made in coating units producing 100 000 ton paper/year or far more. The paper prize for LWC during the last years has been less than 700 euros/ton paper and falling, and much more for extraordinary coated paper grades. Single industrial coating units are considered to be a critical step controlling final quality and hence one of the most essential components when controlling annual turnovers greater than 60-70 millions euro/year/100 000 ton capacity. Falling prizes can not be compensated anymore with the scale of economics. This calls for advanced and reliable multi-disciplinary models, to assist high-tech product innovation, in the field of paper coating. General opinion is that the final pore structure of the coated layer is very much dependent of the dryer section of the coater. It is therefore considered that drying, which includes drainage, of the coating layer is the process step that has direct consequences for the final product quality.

Advanced drying studies have further shown that drying couples together product engineering and process engineering controlling energy efficiency, productivity and quality considerably (e.g. paper ink interaction, print mottle etc.). Our work will therefore aim to illuminate the knowledge of dynamic drainage, which is needed to enhance conventional or novel surface treatment techniques (sym-sizing, plasma, curtain coating, etc.), that are dependent of fine tuning of the in-plane and out-of-plane drainage mechanisms such as surface wetting.

Keywords: drying, porous media, colloidal system, wetting, sorption

Introduction

Industrial coating

Industrial coaters may need less than one second to coat and dry (cure) the coated layer. The thickness of the coating layer for LWC varies between 1-10 μm, giving a mean coating thickness of less than 10 μm, see figure 1.
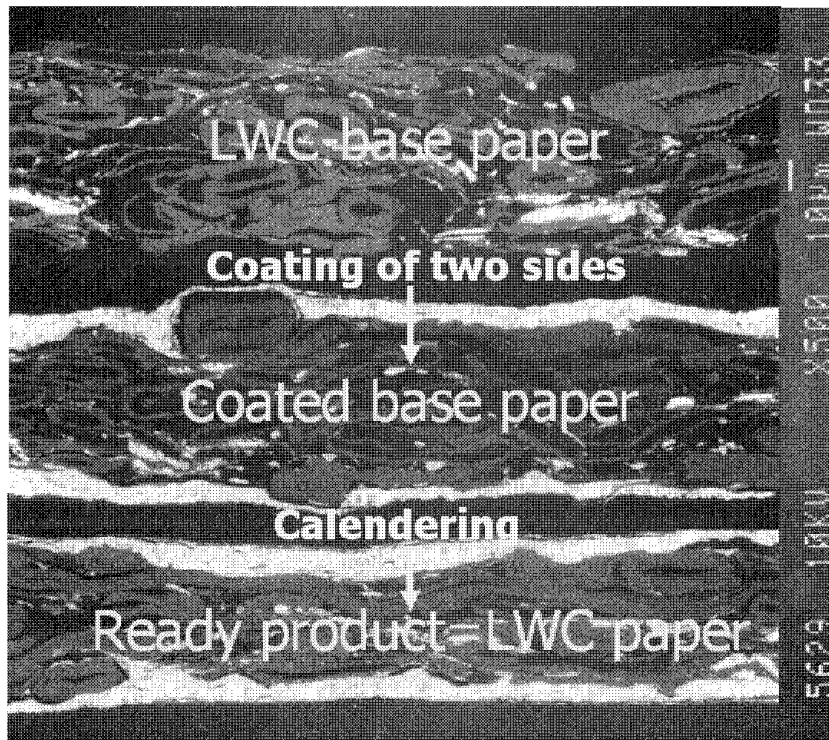

Fig 1. Scanning electron micrograph of a cross section of LWC paper and the structural changes during production. The final calendered product may have a basis weight of w_b=62 g/m2, and ρ=1530 kg/m^3, [4].

The paper coating process

The formulation of a high-performance paper coating requires a strong understanding of the underlying interfacial and colloidal interaction mechanisms dominating initially in the wet state and then during dewatering and consolidation. There are simultaneous interactions between pigments, polymers (stabilizers, binders and thickeners) and other coating components. The distribution of components within the dry coating layer and especially at its surface determines the optical and mechanical properties and printability of the final product.

The extremely fast and harsh drying process of coated paper is impossible to measure. The objective of this work will therefore be to explore and illuminate basic liquid state and movement already in the initial phase when coating color is brought to the surface of the hygroscopic paper (i.e. coating solid contents from 55 % to 65 %). The initial phase includes physicochemical property changes at fiber surfaces and time

spans that are less than 100 milliseconds. Drying of coatings are said to have a consolidation phase where the coating is immobilized. This often takes place in the dryer at a coating solid content between 73 % and 85 % [5]. A common way to define consolidation is to set it between the FCC (first critical concentration) point where the pigment matrix starts to form and the SCC (second critical concentration) point where the three dimensional matrix is fixed. Previous research has indirectly shown that the amount of drained water, during initial drying, can affect product quality considerably (print mottle etc.). We will penetrate this issue further in our paper by connecting drained water to the amount of liquid that is left to participate in film formation (or skinning) of the surface of the coating layer [2, 3].

We feel that the systematic design of paper coatings is hindered by the lack of reliable laboratory techniques that assists simulation of the complexity of phenomena occurring in a coating color after application of a coating layer or after application of a printing color. Existing methods tend to fall into two classes, namely industry standards limited to relative, and often only qualitative, comparisons, and scientific techniques with little relevance to the process realities of pressure, temperature, speed etc.

Novel non-contact application methods have gained a lot of interest recently in paper coating research community. For example, the emerging non-contact curtain coating method gives new possibilities for innovative coatings made of new raw materials, which have not been possible with traditional coating methods. The operational window of traditional coating methods has been controlled e.g. by viscosity, to minimize pressure pulses during application. Curtain coating and spray coating methods highlight now the role of surface wetting and surface chemistry, mainly due to small application pressures. There is therefore an increased need to understand the development of both coating systems and measurement methods when we are trying to fulfill the need of the end consumer.

We will therefore in this work illuminate the dynamics of observed in-plane changes of process variables that are induced by the drying process. The knowledge presented in this article is needed when e.g. new nanostructured mesoporous silica pigments for premium matte inkjet coatings are emerging. New mesoporous silica pigments might have mono-disperse pores that are adjustable from 2 nm to 16 nm. These pigments have shown superior print quality and drastically reduced demand of binder compared to commercially available high-end silica gel products. It is hence expected that nanostructured materials will bring many new adjustable multi-functionalities to paper coatings.

Dimensional changes

Water-induced dimensional changes in paper can be in-plane (Cross Direction and Machine Direction) and out-of-plane (Z-Direction/Thickness), see figure 2.
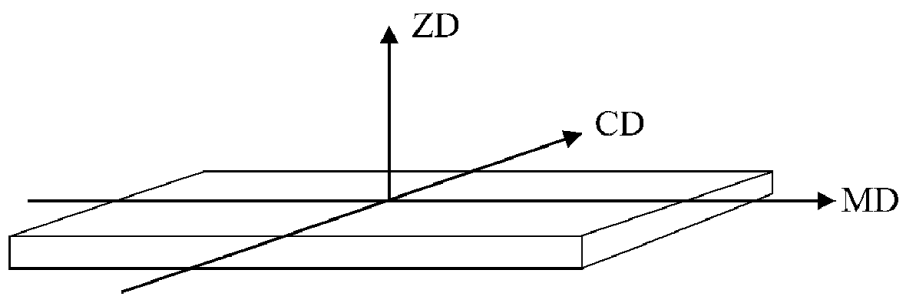
Fig 2. Definition of in-plane and out-of plane.

Hygroexpansion is the name given to dimensional changes in the paper caused by vapor normally after a change in the relative humidity or temperature. Hydroexpansion is on the other hand the name for dimensional changes in the paper caused by a direct contact with an aqueous film or water drops [8]. Hydroexpansion is a more rapid phenomenon than hygroexpansion and this makes it a greater problem in the printing process. The rate of the dimensional changes is most likely related to the rate of water drainage.

Enomac et al. [9] found that the roughening by moisture occurs mainly due to a change in the bulk. The diffusion of water molecules into the fibre wall expands the hemicelluloses in the cross section, and debonding of fibers changes the sheet structure. When fibers swell they affect the structure of the coating color, see figure 3.
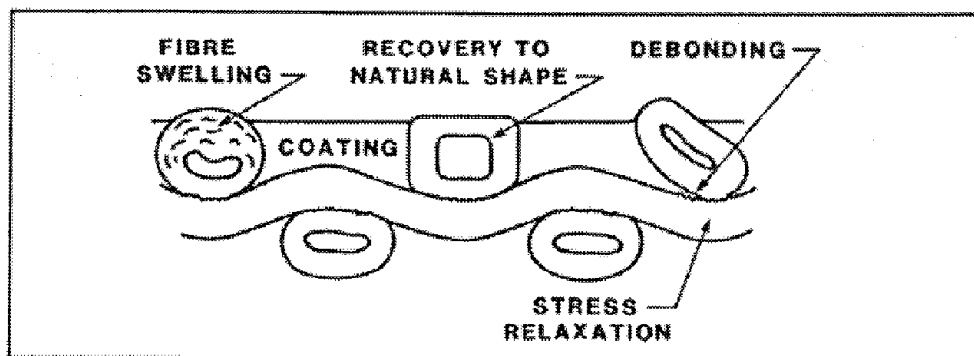

Fig 3. Mechanisms behind surface roughness increase [11].

The surface roughening of a paper and the attachment of coating to the paper depends on many factors; the type of wood, the pulping method, the degree of delignification and the method of drying and calendering the paper [15]. Paper from mechanical pulp roughens more than paper from chemical pulp, [12]. Chemical pulp fibers collapse in the forming and drying stages in the paper machine. Mechanical pulp fibers, on the other hand, usually reach a collapsed state after the compression in the calendaring step, se e.g. figure 1.

Water transport in the base paper

Water transport into paper can be divided into four basic mechanisms:
1. Liquid penetration through the pores by capillary flow

2. Liquid movement through the pores by surface diffusion
3. Vapor phase movement through the pores
4. Liquid and vapor movement through the fibers with analogous mechanisms to 1-3

Lucas-Washburn's equation and Fick's diffusion equation both predict that the rate of liquid penetration in paper should be a function of the square root of time. This means that the sorption time is a linear function of the square root of time at least during short time intervals.
There are however exceptions from the linear dependence due to: the dynamic character of the capillary pressure, the expansion of the fiber network, liquid transport mediated by the vapor phase and whether an external pressure is applied, [13].

Surface treatment methods

Internal sizing:
To reduce the water uptake, chemical agents are added to the paper in the manufacturing step (ASA, AKD, etc.). Their purpose is to minimize the capillary pressure and reduce the capillary penetration by making the surfaces more hydrophobic, see figure 4.
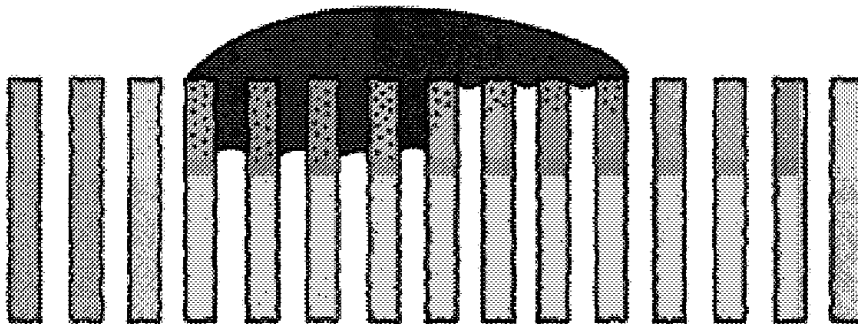

Fig 4. The hydrophilic part of the paper is on the left and the hydrophobic part of the paper is to the right [10].

The driving potential for water penetration according to the Young-Laplace equation is reduced and capillary penetration is minimized. The possible water transport mechanisms being left are surface diffusion and diffusion through the pores. If an external pressure exists, the driving potential is the sum of the capillary pressure and external pressure and the resistance associated with the surface chemistry decreases. The purposes of surface sizing (ASA, AKD, etc.) is to strengthen the surface and reduce water penetration in non-contact printing such as ink-jet or to protect the printed product against moisture or unwanted contact with water. Our measurements and calculations later on in this work show that usage of carboxymethyl cellulose (CMC) in coatings is a chemical agent that reduces water drainage but not necessarily surface wetting when added to coatings.

Novell external sizing
An emerging an interesting technique is plasma treatment of paper surfaces which may have the capability to enhance or even replace several conventional paper surface treatment techniques. It seems very likely that an activation, hydrophilisation or hydrophobisation of surfaces will become more and more important when making tailor made paper in future paper machines aiming for considerable cost advantages. Pre-surface treatment, either by surface sizing, impregnation or plasma, is particularly needed in combination with extrusion coating and flexographic printing of PE and PP-coated paperboard using water-based inks, as well as with emerging novel paper surfaces processing methods such as curtain coating, dry and spray paper coating [19,20]. Knowledge of dynamic drainage is hence needed to enhance conventional or novel surface treatment techniques, which are dependent on the fine tuning of the in-plane and out-of-plane drainage mechanisms such as surface wetting.

Defining surface wetting

We start by defining that a homogenous flux of water through any surface may be written in the following form,

$$\frac{\dot{m}_w}{A} = w_w \rho_w \frac{V_w}{V} \tag{1}$$

When, using simultaneously for moist matter the following definition, $X = m_w / m_{bd}$, and defining local porosity as, $\varepsilon = (V_a + V_W)/V$, gives then for a porous volume V the following waterless bone dry density,

$$\rho_{mbd} V = \rho_{bd} V (1 - \varepsilon) \tag{2}$$

Using, equation (2) in equation (1) transforms our calculations into the following form,

$$\frac{\dot{m}_w}{A} = w_w \rho_w \rho_{mbd} \frac{V_w}{V \rho_{bd}(1 - \varepsilon)} = w_w \rho_{mbd} \frac{m_w}{m_{bd}} = w_w \rho_{mbd} X \tag{3}$$

Rewriting, equation (3) for a movement in space described by a distance, Δy gives, in continuum, the following,

$$\frac{-\dot{m}_w}{A} = w_{w1} \rho_{mbd1} X_1 - w_{w0} \rho_{mbd0} X_0$$
$$= \frac{\beta_{w1} \rho_{mbd1} X_1 - \beta_{w0} \rho_{mbd0} X_0}{y_1 - y_0} \tag{4}$$

Where, β is the mass transfer coefficient and the differential distance, $\Delta y = y_1 - y_0$, defines the direction of the movement in space, and hence also the sign for those components that have flow vectors, e.g. when needed a minus sign for the mass flux. Equation (4) can now be used to describe pure diffusion, which is mass transfer dominated by molecular movement, for this we obtain the following,

$$\frac{\dot{m}_w}{A} = \frac{\beta_w}{\Delta y}(\rho_{mbd0}X_0 - \rho_{mbd1}X_1) \tag{5}$$

Where, β according to molecular diffusion theory is based on the average diffusion velocity. Using for moist air the following definition, $x = m_v / m_a$, and assuming that the ideal gas law is valid, transforms equation (5) at constant pressure into the following form,

$$\frac{\dot{m}_v}{A} = \frac{\beta_v}{\Delta y}(\varepsilon_0 \rho_{a0} x_0 - \varepsilon_1 \rho_{a1} x_1)$$
$$= \frac{\beta_v}{\Delta y}(\varepsilon_0 (\frac{p_0 M_a}{RT_0})x_0 - \varepsilon_1 (\frac{p_1 M_a}{RT_1})x_1) \tag{6}$$

Where $\varepsilon p = p - p_v$, and defining,

$$x = \frac{M_v}{M_a}\frac{p_v}{p - p_v} \quad \text{or} \quad \frac{p_v}{p} = \frac{1}{M_v/(xM_a)+1} \tag{7}$$

We insert equation (7) in equation (6) obtaining,

$$\frac{\dot{m}_v}{A} = \frac{\beta_v}{\Delta y}(\varepsilon_0 \rho_{a0} x_0 - \varepsilon_1 \rho_{a1} x_1) = \frac{\beta_v}{\Delta y}\frac{M_v}{R}(\frac{p_{v0}}{T_0} - \frac{p_{v1}}{T_1}) \tag{8}$$

Equation (8) is a common equation for describing mass transfer in moist air [1]. We may now stress, when having in mind equation (4), that when the pure mass transfer dominated molecular diffusion diminishes we may need to rewrite the mass transfer equation in a form where the mass transfer coefficients are in a more general form, i.e.,

$$\frac{\dot{m}_v}{A} = \frac{1}{\Delta y}\frac{M_v}{R}(\beta_{v0}\frac{p_{v0}}{T_0} - \beta_{v1}\frac{p_{v1}}{T_1}) \tag{9}$$

From this we see that we may even have a case where $p_{v0} = p_{v1}$ and still have mass transfer, as long as $\beta_{v0}/T_0 \neq \beta_{v1}/T_1$, such special cases are, in molecular systems, e.g. thermo diffusion etc.
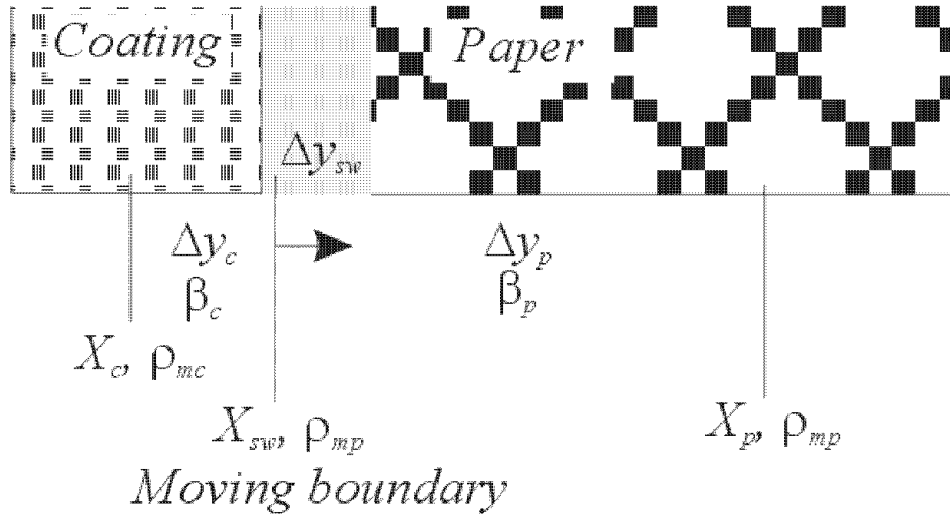
Fig 5. Problem formulation and notation used when solving the stated problem, where Δy_{sw} is the surface-wetting layer. This layer could also be called a boundary storage zone.

We will now be using equation (5) and refer to figure 5 when defining the set of differential equations, which are solved in this work,

$$\overbrace{\frac{dm_{wc}/A}{d\tau}}^{\text{calculated flux}} = \frac{\beta_c}{\Delta y_c}(\rho_{mc}X_c - \rho_{mp}X_{sw}) \tag{10}$$

$$\underbrace{\frac{dm_{wsw}/A}{d\tau}}_{\text{stored flux}\,=\,\text{surface wetting}} = \frac{\beta_c}{\Delta y_c}(\rho_{mc}\frac{X_{c,0}+X_c}{2} - \rho_{mp}X_{sw}) \tag{11}$$

$$- \frac{\beta_p}{\Delta y_p}(\rho_{mp}X_{sw} - \rho_{mp}X_p)$$

Where we use, $(X_{c,0}+X_c)/2$ to account for the surface wetting caused flows of water within the control volume (internal circulation+storage). In this work we define Δy_c and Δy_p, by postulating the following,

$$\Delta y_c = \Delta y_{c,0} + \Delta y_{bl} \quad \text{where we have}: \Delta y_{bl} = \Delta y_{sw}(\frac{\tau}{\tau_{max}})^n \tag{12}$$

$$\Delta y_p = \Delta y_{p,0} - \Delta y_{bl} \tag{13}$$

Where, the exponent $n \leq 1$ varies as a function of how the surface wetting is taken place (e.g. physicochemical properties change from product to product, which in turn changes the water movement at boundaries, [14]). We have now defined, how the

surface-wetting boundary moves within the surface-wetting layer, Δy_{sw}. In the context of diffusion, the potential to be used have to be connected in some way to the partial molar Gibbs free energy of the component of interest. In our work this is done by the terms describing local concentration, ρ_{mX}, of water and by the term describing diffusion velocity, $\beta / \Delta y$. Changes in physicochemical fiber properties, i.e. hornification and geometry changes will be seen as changes in the term describing diffusion velocity, $\beta / \Delta y$ and surface-wetting layer, Δy_{sw}.

Study of surface wetting – In-plane observations

A study of how surface wetting may affect the properties of the coated layer, when coating paper, is now illuminated by including figures 6 to 8. In these figures the dynamic changes in surface wetting is made visible by including a blue color in the coating color when applying air impingement drying. The blue color moves with the water phase and shows how even the water movement has been during drying of the coated paper, see figures 6 and 7.
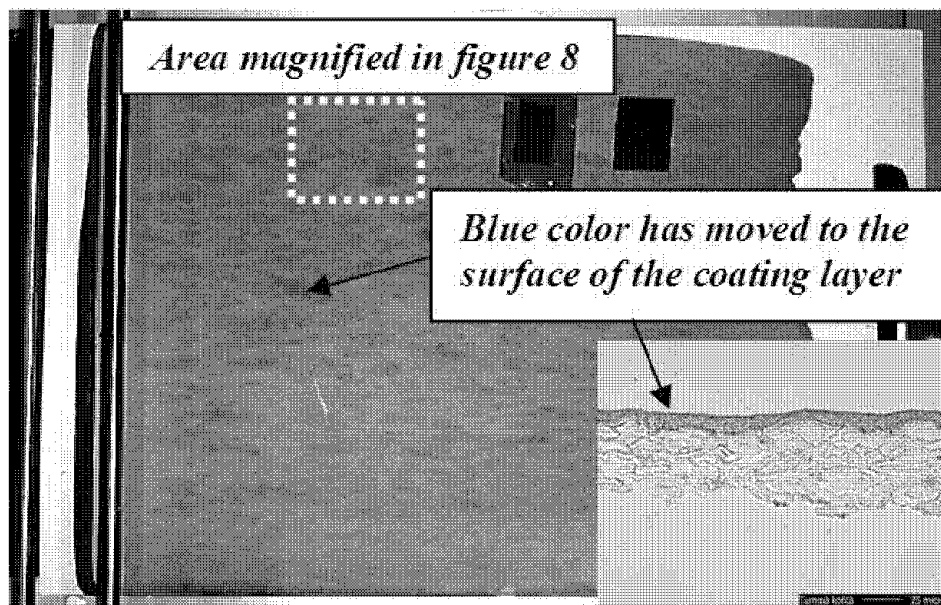

Fig 6. Impingement dried (4 cm/s, 200°C and drying time less than 2 seconds). The surface color variations are not evened out due to surface wetting which influences the amount of liquid that is left to flow to the surface and to participate in film formation, skinning or such.
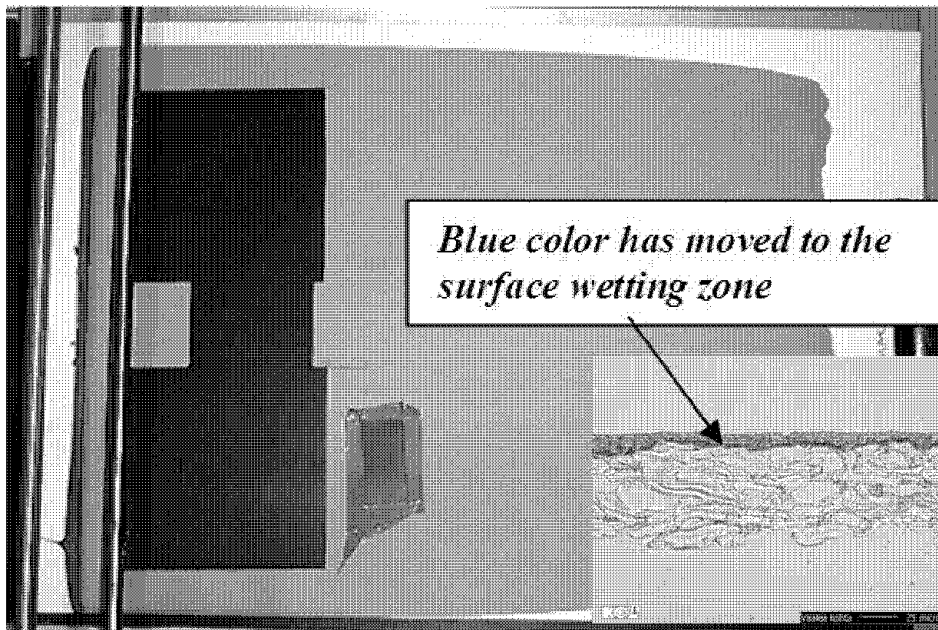
Fig 7. Free air convection dried paper (mainly sorption). Drying time (mainly sorption) was more than 3 minutes. The surface color variations have been wiped out.

At this point we stress that an in-plane color transition from light blue to dark blue, within one or two millimeters, corresponds to, many g/m^2 change in the local mass drained. When this is recalculated into drainage rates we find that the drainage rate might vary within a millimeter range from 1 to 10 g/m^2s, (i.e. 3,6 to 36 kg/m^2h)! It is known that strong surface tension gradients results from compositional changes during drying and spatial temperature gradients [16, 17]. The big variations in drainage rates are therefore most likely influencing the film formation process and can even induce cellular flows in the coating layer. This seems to argument for studies of Marangoni like flows in thin coating layers.
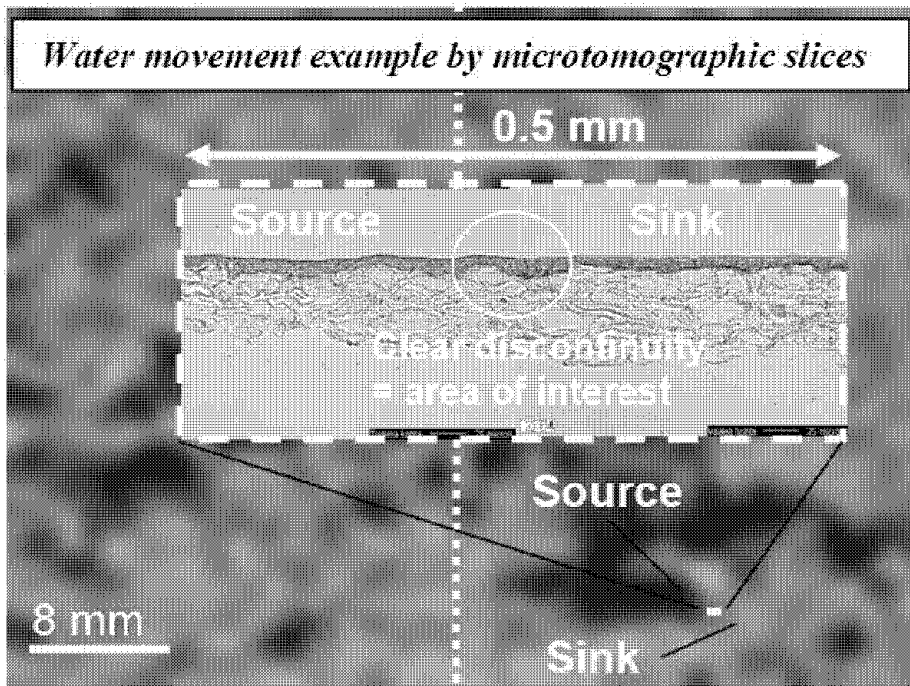
Fig. 8. Enlargement of figure 6. The mosaic drying pattern reminds of print mottle, see right half reedited in figure 9.

Figure 9 is added to show similarities of the mosaic drying pattern and the quality defect called print mottle.
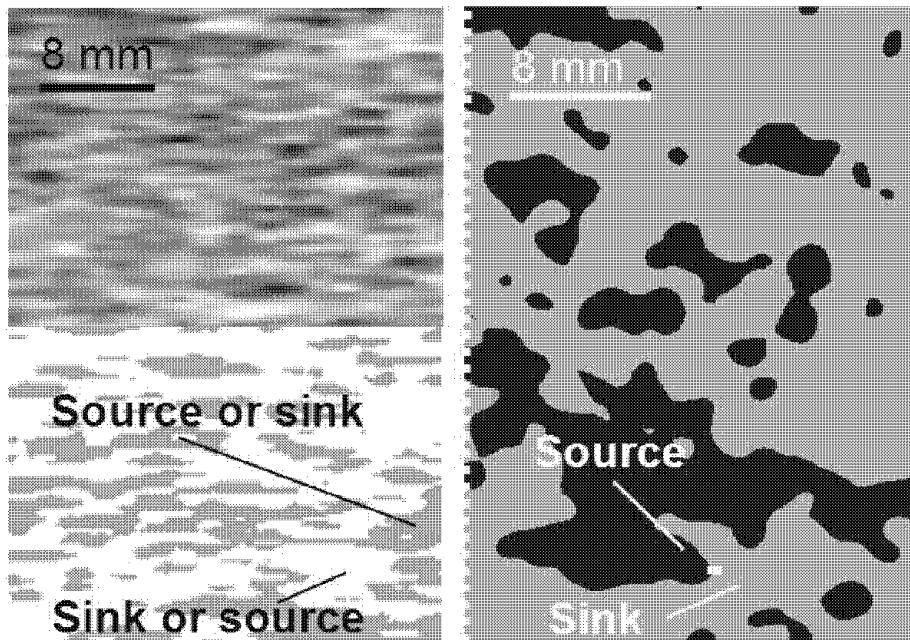

Fig 9. A general example of print mottle is included in the left upper corner in gray scale, in the left lower corner same picture is reduced into white and light gray color and on the right hand is figure 8 right hand side reduced into light and dark blue color.

Figure 9 shows that color editing into two basic colors brings forward the mosaic drying pattern. The mottling pattern in figure 9 seems to be very similar to the mosaic drying pattern observed in figure 6 and 8. This makes us to conclude that print mottle has to be very strongly connected to a sink and source mechanism that controls the water drainage during coating or printing and affects the final quality. Print mottle can, however, give an "inverse print" (i.e. photo negative) of the mosaic drainage pattern in figure 6, while print mottle is depending on hydrophilicity or hydrophobicity of the printing color.

Calculated surface wetting

A set of differential equations that includes a very rough model for sinks and sources are solved using the equations (10-13). The boundary conditions are written into the calculated figures 10 to 14, these being: paper temperature, t_p, coating temperature, t_c, measured and calculated drainage m_{wc}/A, measured and calculated mass flux $\dot{m}_{wc}/A$, the moving boundary Δy_c, the calculated moisture content of the surface wetting zone X_{sw}, the calculated moisture content of the coating color X_c, the dryness of the coating color Y_c, the mass transfer coefficients, β_c, β_p, and geometrical boundary conditions, $\Delta y_p,0$ and Δy_{sw}. The experimental set-up is described in Berg et al. [18].
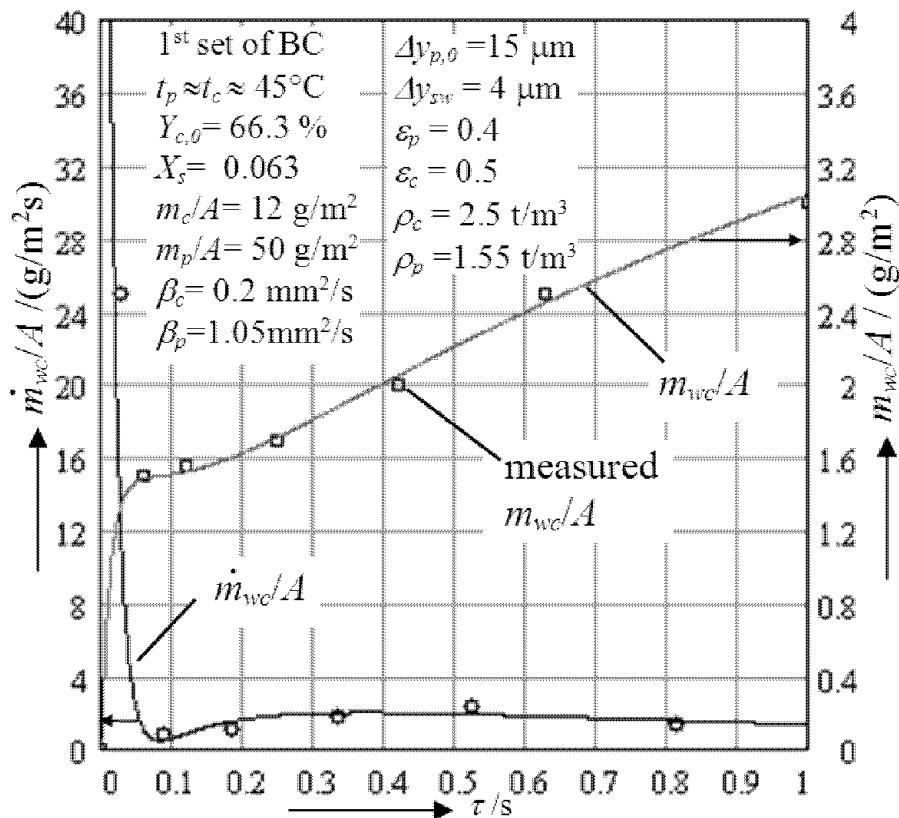

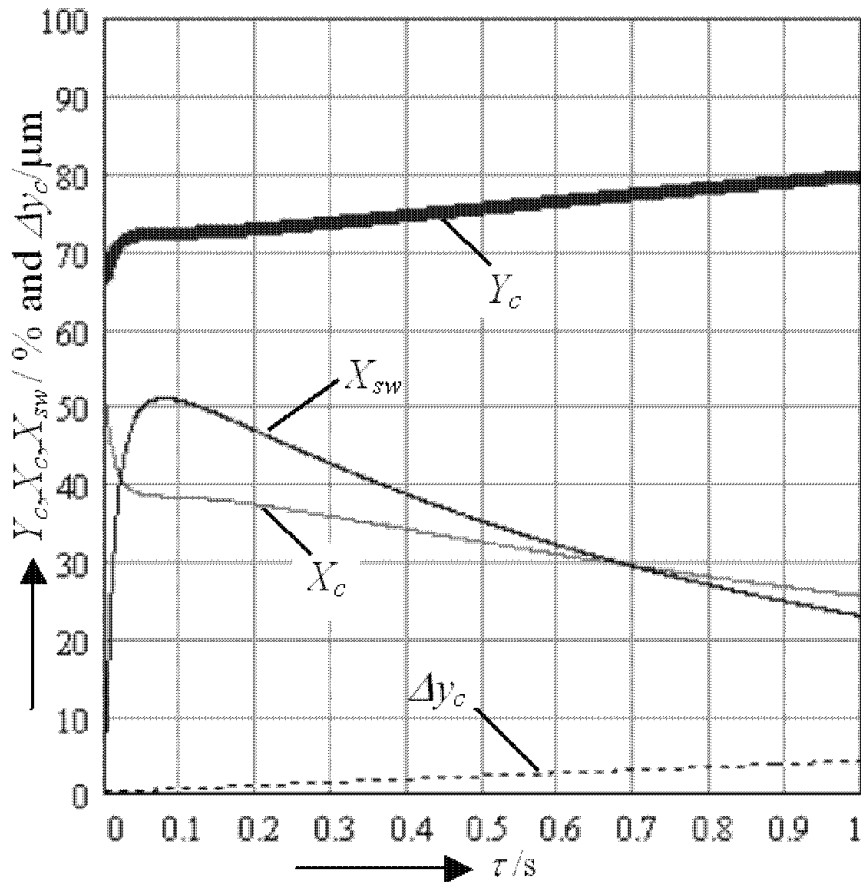Fig10. Mass flux and drainage, of LWC-paper, as a function of contact time, τ, and equations (10-13).

Fig 11. "Process variables" for figure 10 as a function of contact time, τ.

Our calculations show that the surface wetting layer may change from $\Delta y_{sw} = 1.6 \mu m$ to $\Delta y_{sw} = 5.2 \mu m$. When combining this knowledge with the mass flux values in figures 10 to 14, we draw the conclusions that,

• surface-wetting occurs for contact times that are smaller than 0.1 to 0.2 seconds, and that this phenomenon is not the same as wetting delay or dynamic wetting line [6].

• a point of minimum mass transfer might occur after the initial surface-wetting phase, and this can be "negative" The absolute value depends of measurement method, and it is found in the time interval 0.05 to 0.3 seconds.

• diffusion out of the coating seems to be the remaining mechanism controlling liquid transfer, for contact times longer than 0.2 to 0.3 s.

• surface-wetting is a function of hornification and topographic changes during rewetting and this seems to affect the liquid drainage after 0.3 to 0.6 seconds [7].

Can we change the way water moves? To answer this we added an example of using the binder and thickener carboxymethyl cellulose (CMC) to control water movement, see figures 12 and 13.
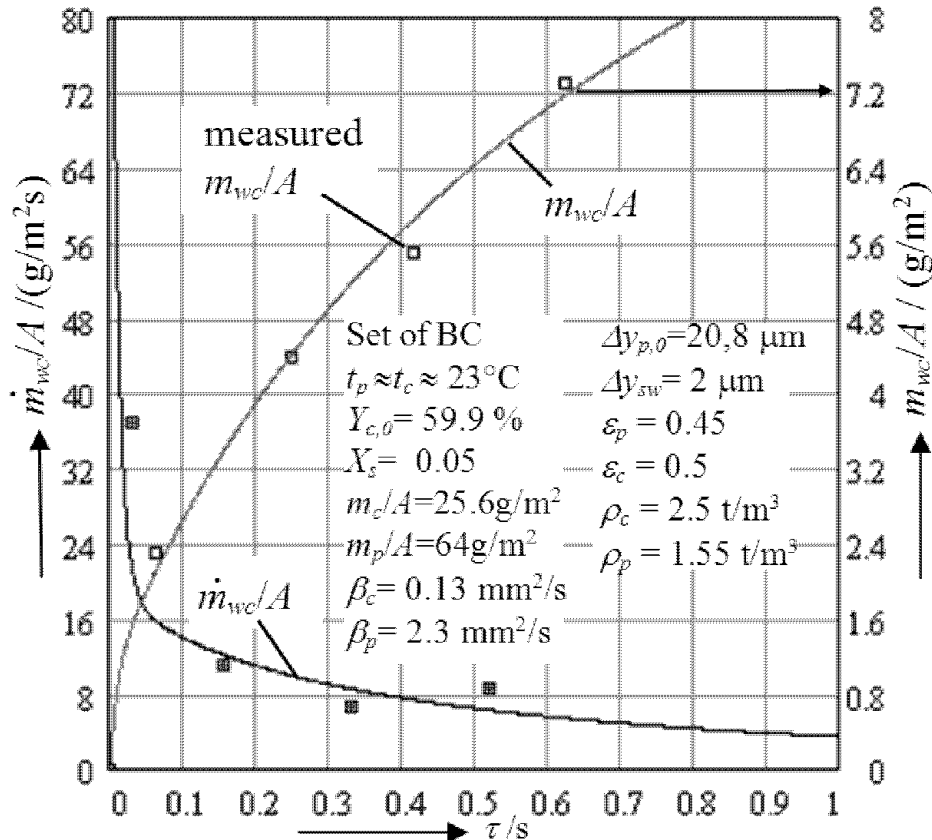
Fig. 12. Mass flux and drainage, as a function of contact time, τ. Varkaus (WF) fine paper with carbonate coating (S3B, 100 pp carbonate, 12 pp latex and no CMC, [18]).

The effect of CMC is clearly seen when comparing the results in figures 12 and 13 (i.e. carbonate coating with and without CMC). The difference in drained water is 1.5 g/m^2 after 0.25 s, which corresponds to 6 g/m^{2}s! Starch is another drainage regulating chemical that affects surface wetting, see e.g. [1]. Our calculations tend to show that the reduction for CMC is mainly due to a reduction of liquid drainage but not necessarily a reduction in surface wetting (the surface wetting layer can be held constant in the calculations). We therefore find that CMC is an example of a chemical that tunes the liquid release of the coating color and affects the mosaic pattern shown in figure 6 and hence also print mottle.

CMC raises the viscosity of the water phase and will hence influence the internal liquid flows of the coating and hence both the liquid release properties of the coating and the structure of the final coating color. CMC is also a dispersing agent, which means that it affects both coating coverage as well as in-plane liquid drainage.

We therefore end by concluding that both our calculations and pictures show that there is a clear need to control surface properties when making multifunctional surfaces of uniform quality.
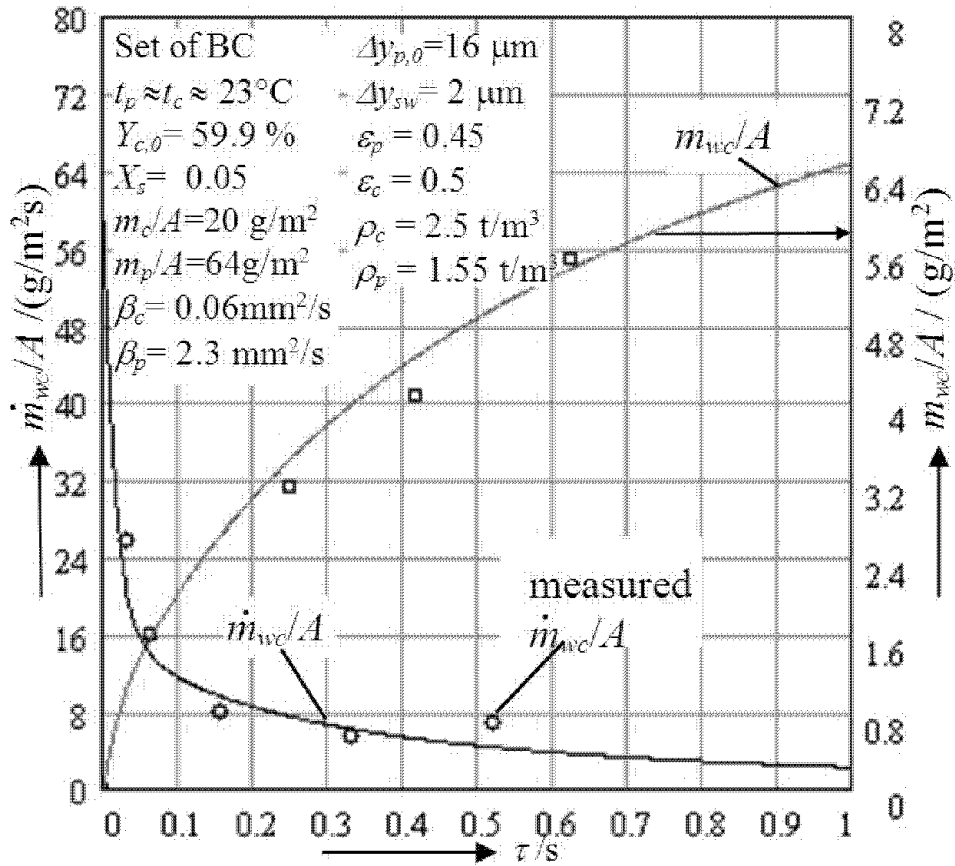
Fig. 13. Mass flux and drainage, as a function of contact time τ. Same coating as in fig 12 but 0.5 pp CMC, [18].

The most extreme drainage behavior was obtained for art paper, see figure 14. These measurements show backflows of liquid making a calculated negative minimum at roughly 0.05 s, which as previously stated is explained by the sink and source mechanisms during drainage.
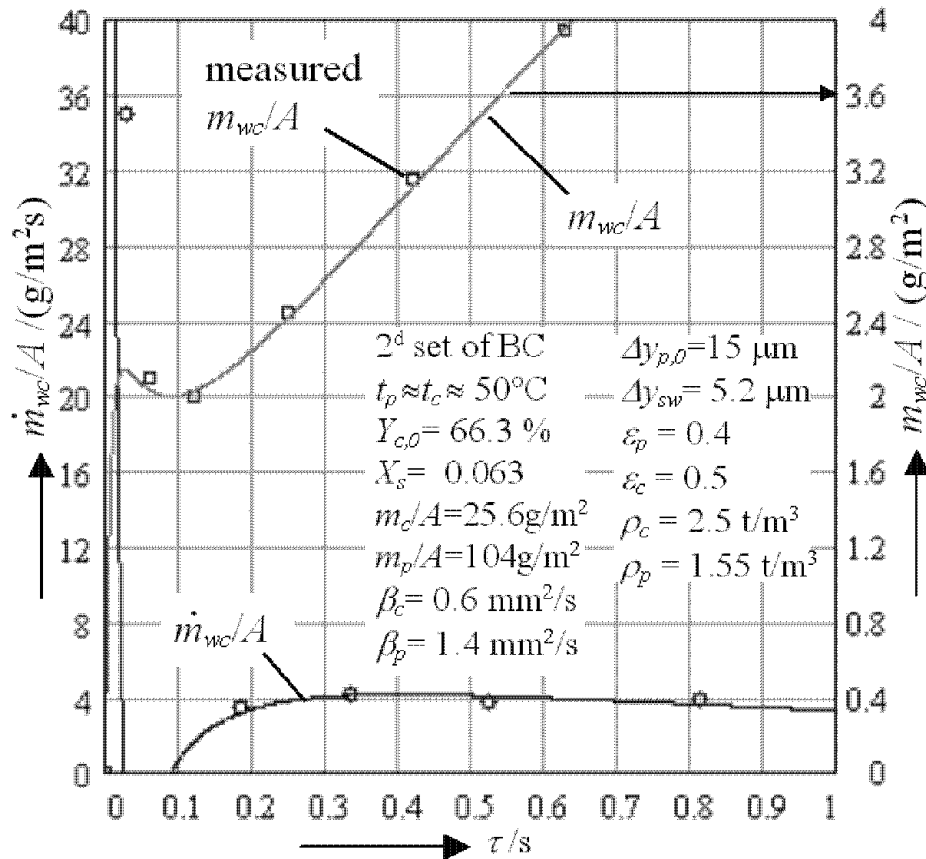
measured
m_{wc}/A

m_{wc}/A

2^d set of BC $\Delta y_{p,0}=15\ \mu m$
$t_p \approx t_c \approx 50°C$ $\Delta y_{sw}= 5.2\ \mu m$
$Y_{c,0}= 66.3\ \%$ $\varepsilon_p = 0.4$
$X_s= 0.063$ $\varepsilon_c = 0.5$
$m_c/A=25.6 g/m^2$ $\rho_c = 2.5\ t/m^3$
$m_p/A=104 g/m^2$ $\rho_p = 1.55\ t/m^3$
$\beta_c= 0.6\ mm^2/s$
$\beta_p= 1.4\ mm^2/s$

$\dot{m}_{wc}/A$

τ / s

Fig 14. Mass flux and drainage, of art paper, as a function of contact time τ, i.e. equations 10 to 14 solved with applied boundary conditions, see also ref. [1].

Conclusions

We find that future studies have to look at the basic phenomena that are significantly influencing liquid movement at fiber-coating boundaries and enforcing local changes of the structure of the coating layer. The result of such work will be an increased knowledge of the formation of the coating layer. This knowledge is needed when developing existing and new products for the paper industry that have multifunctional processing properties (surface chemistry, roughness, specific energy etc.). This is accomplished, for example, by producing with novel equipment and nanoparticles a multifunctional LWC-paper that could be used both in the rotogravure and offset printing press. Controlled surface treatment (sizing, plasma etc.) is in turn particularly needed for emerging novel paper surface processing methods such as curtain coating, dry coating or spray coating.

Knowledge of dynamic drainage is hence needed to enhance conventional or novel surface treatment techniques, which are dependent on the fine tuning of the in-plane and out-of-plane drying mechanisms, i.e. surface wetting, drainage and evaporation.

We believe drying knowledge in this work is bridging the product engineering with the process concept, as in the figure below.
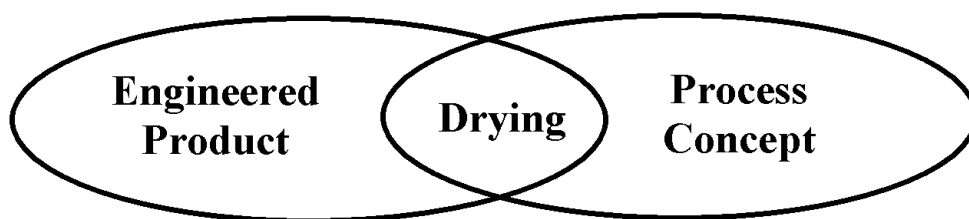
Drying knowledge seems to be especially important when introducing new technologies, e.g. nanoparticles and plasma surface treatment due to its bridging capability in the product formation process. This work may also be useful for other products where quality is dependent on initial surface wetting and fast liquid transfer rates during remoistening (e.g. paper ink interaction, dried food, etc.).

Notation

A	m^2	area
β	m^2/s	mass transfer coefficient
ε		porosity
$m, \dot{m}$	kg, kg/s	mass, flow of mass
M	kg/mole	molar weight
n	mole	moles
p	kPa	pressure
$q, \dot{q}$	kJ, kJ/s	heat, flow of heat
R	kJ/mol K	ideal gas law constant
ρ	kg/m3	density of matter or fluid
τ	s	time
T, t	K, °C	temperature
x	kgH2O/kgda	air humidity
X	kgH2O/kgbd	moisture content dry basis
Y	kgbd/kgtot	dryness
$y, \Delta y$	m	length normal to paper
a		air
bl		boundary layer
c		coating
da, bd		dry air, bone dry
m	max	mean value, maximum value
p		paper
sw		surface wetting
v, w		vapor, water
$0, 1$		0=start value, place or time

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