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Drying air-induced disturbances in multi-layer coating systems

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ABSTRACT

A range of new experimental techniques is developed to quantify drying-air induced disturbances on low viscosity single and multi-layer coating systems. Experiments on prototype slide-bead coating systems show that the surface disturbances take the form of a wavelike pattern and quantify precisely how its amplitude increases rapidly with wet thickness and decreases with viscosity. Heat transfer measurements show that the redistribution of water to form an additional lower viscosity carrier layer while increasing the solids concentration of the upper layer or layers enables the maximum drying rate, for which drying-air induced surface disturbances are acceptably small, to be increased with significant commercial benefits.

Key words: films, fluid mechanics, heat transfer, optimisation, polymer processing, visualisation.

1. INTRODUCTION

Multi-layer coating systems are widely used throughout the inkjet media and photographic manufacturing industries, offering significantly higher process efficiency than traditional multi-pass single layer coating systems. The factors leading to defects in the wet coating phase have recently been considered by Gaskell et al. (1999), Noakes et al. (2002a) and Ikin et al. (2007). This paper focuses on the defects that arise in the drying phase of, for example, multi-layer curtain or slide-bead coating systems, when the drying air emerges from an array of slot nozzles arranged perpendicular to the machine direction and disturbs the surface of the wet coating. The key issues are highlighted by studying drying defects in the slide-bead process from the standpoint of the photographic industry, where the coatings are typically relatively dilute melts containing gelatine, silver halide dispersions, polymeric thickeners and cross-linking agents. Fig. 1 shows a sketch of the process for a double-layer system; note that in practice the number of layers can be much larger, Ikin et al. (2007). (Fig. 1 inserted here).

The stability dynamics of thin films when defects are induced by, for example, substrate topography or near moving contact lines has received a great deal of attention in the recent scientific literature, the most common approach being that of modelling the stability of the film flow by performing a linear stability analysis of the thin film lubrication equations. Davis & Troian (2005), for example, have revealed the key role of recirculating streamlines on the stability of free surface disturbances. Wet, low viscosity coatings typically found in multi-layer coating
systems are also particularly prone to becoming disturbed by drying-air, Cohen & Gutoff (1992). In contrast, little previous work on air induced defects has been reported and until very recently theoretical models have been restricted to idealised cases with small pressure fluctuations where linear theory is applicable, Ruschak (1987), Alleborn & Raszillier (2004). Recent work on gas-jet wiping, with larger pressure disturbances is of greater relevance to the cases considered here, Laconette et al. (2006), Gosset & Buchlin (2007). These showed that theoretical analysis of such problems is more complicated than those cited above since it is necessary to either measure or predict the shear stress and pressure profiles caused by the turbulent air flow above the film.

This paper is the first to focus on the experimental quantification of drying-air induced disturbances on low viscosity single and multi-layer coating systems, an effect that is often referred to as mottle, Cohen & Gutoff (1992). Important previous experimental studies of mottle include those of Cohen & Gutoff (1992) who studied how this effect could be reduced or eliminated by decreasing the air speed or increasing the concentration of the coating solutions and Bell et al. (2000) who found that the latter approach is often particularly beneficial since it increases viscosity and reduces wet thickness. The traditional approach adopted in the photographic industry is to chill the wet coating to about 2°C to 4°C between the coating and drying phases. Owing to the properties of the gelatine used as binder for photo-sensitive coatings, this leads to a very significant increase in viscosity before the surface is exposed to the drying air, thus increasing its robustness to air induced disturbances. Other important studies have shown that mottle can be eliminated by directing air parallel to the web surface with the air speed matching that of the web, Bell et al. (2000), while Iwado (2003) proposed the use of two dryers, in the first of which air speeds are limited while viscosity (and hence robustness against disturbance) increases, and the second uses high speed air to increase solvent evaporation rates.

In setting up experimental methods to study drying-air induced disturbances, it is important to ensure continuous, constant relative speed between the coating sample and the dryer in order to eliminate the banding effect illustrated in Fig. 2. (Fig. 2 inserted here). Here, a single layer of viscosity 20 mPas comprising Polyvinyl Alcohol (PVOH) and some additional magenta dye was coated at a wet thickness of 100µm onto a paper substrate and the wet sample transported into the dryer and stopped beneath the jets while being dried. The bands, imaged by reflected light, form due to the air from the jets sweeping the fluid away from the impingement zones and in the direction of air flow. This requirement poses difficulties in a laboratory where equipment is necessarily of finite length. Experimental methods for overcoming these difficulties are described and results are given which quantify drying-air induced

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disturbances in multi-layer coating systems for the first time and which demonstrate their dependence on the coating layer viscosity and thickness distributions. Since drying capacity is often the key limitation on production rate in industrial coating systems, the effect of surface disturbances on overall drying capacity is also investigated. It is shown that drying speed can be enhanced significantly by optimising the water distribution within a multi-layer coating assembly and highlights, in particular, the benefits of introducing a bottom “carrier” layer on overall drying speed.

2. EXPERIMENTAL METHODS

2.1 Initial Rotary Dryer Tests

Initial visualisations of air-induced disturbances were obtained using a rotary dryer system, sketched in Fig. 3, which was designed to mimic the industrial situation and allows continuous relative motion between a stationary wet coating and an array of air impingement nozzles. (Fig. 3 inserted here). With 18 nozzles mounted on the periphery of a hollow cylindrical drum, air is fed into the drum and constrained by a pair of stationary baffles to emerge through a maximum of three nozzles and impinge on the web; brushes are mounted at ends of the baffles to minimise air leakage while allowing free movement of the rotor. The rotary dryer could achieve relative peripheral speeds between the nozzles and coating up to 3.3m/s.

A CCD camera was used to record sequences indicating the build-up of disturbances to the coating surface under the rotating dryer wheel. A light box housing four fluorescent tubes was mounted behind a diffusing screen to illuminate the sample near grazing incidence, while the camera was mounted to receive specularly reflected light via a mirror, as shown in Fig. 3. A mask comprising equi-spaced diagonally mounted opaque strips was set up in a zebra-stripe pattern as a visual aid for observing the break-up of the wet surface into waves. The camera was fitted with a low powered lens and set up at a distance such that the depth of field embraced both the mask and the drying zone. The resultant sequences revealed distortions to the image of the zebra stripe pattern due to the departure of planarity of the wet coating. These took the form of equispaced wavy bars of increased coating thickness generally aligned perpendicular to the machine direction. Fig. 4 was obtained by directly imaging a fully dried single layer coating of a three-layer photo-emulsion whose properties are summarized in Table 1. The severe surface waves resulted on increasing the nozzle air pressure to 900Pa, the speed of the nozzles being 3.3 m/s relative to the web and the minimum nozzle-to-web gap 10mm. The velocity \( V \) of the air emerging from the nozzle in this case was 36.3 m/s - estimated from equation (1) assuming a discharge coefficient \( C_d \) of 0.98, \( P \) being the pressure drop across the nozzles,, \( T \) the temperature and \( \rho_{NPT} \) the density of air measured at 20°C and 1 bar. (Fig. 4 inserted here).

\[
V = C_d \left( \frac{2P(273+T)}{(293\rho_{NPT})} \right)^{1/2}
\]  

\( (1) \)
The effect of excessive air motion is to cause surface waves similar to those seen by Craik (1966) on wind-blown water films, by Özgen et al. (2002) on thin layers deposited on the lower wall of a wind tunnel section and by Gosset & Buchlin (2003) when wiping a thin film coating on a rotating cylinder with an impinging slot jet.

### 2.2 Visualisation of Surface Waves Under a Static Linear Pilot Dryer

The rotary dryer, although useful for simulating continuous high speed drying, has the disadvantage that the gap between each nozzle and the web varies as the former traverses the sample. This becomes more significant as the minimum gap is reduced and light used to illuminate the sample necessarily becomes interrupted as each nozzle traverses the beam. Moreover, the small sample size covers only typically 25 cycles whereas many tens of cycles are needed to smooth out statistical variations to obtain an accurate assessment of mean wave amplitude. Studies using a stationary American pilot dryer incorporating similar nozzles demonstrated that these limitations could be overcome for web speeds as low as 0.1 m/s. Fig. 5 shows disturbances in the surface of a wet PVOH coating of thickness 100 µm and viscosity 257mPas when subjected to air from this dryer, the speed of the air jets being steadily increased from 22 m/s to 70 m/s during the time it took for the sample to traverse the dryer. (Fig. 5 inserted here). It will be seen that the disturbances are remarkably similar in both appearance and wavelength to those from the rotary dryer - Fig.4. This motivated further work using a second static air impingement pilot dryer located in the laboratory made available to the authors.

Achieving good web speed control throughout this second pilot dryer was vital to ensuring accurate waveform analysis. Speed control systems designed for high coating speeds, employing feedback from a speed measuring device, often become unstable at very low speeds. Here, web speed uniformity to ±1% down to 0.1 m/s was achieved using open loop control combined with a rubber covered web tracking wheel for monitoring speed. Visualising the disturbances in the restricted space between the web and nozzles was also challenging, previous relevant studies having either focussed on quantifying waves near a single jet or along the lower surface of a wind tunnel. In the former, Gosset and Buchlin (2003) investigated methods for imaging disturbances by both reflected and transmitted light. The methods developed here extend their light absorption technique in two important respects: (i) to highlight how surface waves grow and become established and to determine the minimum web speed for further studying these waves without incurring unwanted banding; (ii) to profile the wave forms at a fixed point beneath the dryer.

#### 2.2.1: Recording the Formation of Waves

The apparatus for recording surface waves in the pilot dryer is shown in Fig. 6. (Fig. 6 inserted here). A Cohu model 4912 CCD camera was mounted on a carriage free to move along rails pre-aligned with the machine direction and...
the zoom lens adjusted to achieve a field of view of 60mm. The carriage control speed was calibrated to enable the camera to move at the same speed as the web, namely the lower limit of 0.12 m/s. The first two nozzles in the dryer were blocked off to ensure the camera captured the true start of drying as it traversed the dryer. The web was of transparent film base and pre-marked with a line drawn cross-width to act as a reference for checking phase shifts in the waveform as the sample progressed through the dryer. The lower outward surfaces on the jet box were rendered matt white to achieve adequate image brightness when reflecting light from a fluorescent tube through the coated web. The web was supported at the edges on flat horizontal metal rails and by narrow cross members at intervals of 0.3m in order to prevent the web sagging downwards between the rails under the action of the impinging air.

The PVOH coating solution used for the study contained a green dye and a magenta filter was installed at the camera aperture to enhance contrast. Waveform profiles were obtained by first digitizing the intensity along a line in the machine direction using the OPTIMAS image analysis software produced by Bioscan Inc. Unwanted slowly varying density components were removed by computing the Fast Fourier Transform using Mathcad, discarding all frequencies below a threshold frequency and transforming back to the space domain by determining the inverse Fast Fourier Transform.

2.2.2: Profiling Film Thickness at a Fixed Point Beneath the Pilot Dryer

The triangulation method incorporated into commonly available displacement meters (for example as supplied by Keyance Corporation) utilises a laser that is focused onto the test surface and the back scatter received at a fixed angle by a position sensitive detector. This was found to be over-sensitive to inherent vibrations of the web caused by the impinging turbulent air so a light absorption technique was used instead. A light source comprising a HeNe laser operating at 2mW and emitting a wavelength of 633nm was mounted on the same carriage as used for recording wave build-up. Cost considerations dictated the use of a paper web instead of film base used in the rotary dryer experiments. The increased opacity of the paper web necessitated the use of a photomultiplier tube and the development of a highly sensitive densitometer system. A green absorbing dye was again used to gain sensitivity to wet thickness variations.

A DC amplifier circuit was constructed to gain sensitivity to subtle variations in transmitted light while ensuring that the output signal remained within the input range acceptable to the digital adaptor (Thurlby) used for recording waveforms. A calibration was first run by coating three samples, one of the target coating weight and the others ± 10% relative to the target weight. A windowing function was applied to 1024 consecutive coating thickness assessments in order to remove end effects. The data were subjected to a form of digital filtering using Mathcad in order to extract surface wave signals from the noise due to variations in thickness of the paper substrate. The fast fourier transform was computed to determine
the predominant spatial wavelengths. These were isolated by multiplying the spectral information with a suitable pass-
band filter and the inverse transform taken to transfer the information back into the space domain to yield a final
assessment of rms amplitude made for comparison purposes, Schweizer (1997).

2.3. Measurement of Heat Transfer Coefficient

The influence of coating formulation and drying air speeds on the maximum drying rates, for which surface
disturbances remain tolerably small, was also investigated. The heat transfer apparatus used is shown in Fig. 7. (Fig.
7 inserted here). The heat flux sensor was a solid state heat pump (MELCOR) utilising the Peltier effect. The sensor
was bonded to a heat sink cooled by water flowing at 1 litre/min and controlled at 20°C. The sensor assembly was
transported at constant speed beneath the nozzles using a translating table. The sensor was calibrated using the
method developed by Noakes et al. (2002b) for determining the heat transfer characteristics of an experimental air
flotation dryer.

The bulk temperature of the air impinging onto the sensor assembly was determined using a thermocouple mounted
3mm above the sensor. Having calibrated the sensor, its thermal conductivity was obtained by machining a conical
cup having a base of cross-section exactly matching that of the sensor, Ikin (2005).

3. RESULTS

Having established that the pilot dryer was capable of reproducing the high speed disturbances seen from the rotary
dryer, the pilot dryer was used to investigate wave formation and heat transfer in greater detail, as described below.

3.1. Drying-air induced surface wave profiles

The mechanism for the surface disturbance was investigated as follows. The carriage containing a CCD camera was
set in motion to track a freshly coated sample as it entered the pilot dryer. Drying speed was controlled to 0.12 m/s
by reverting to open loop control to minimise instabilities and reduce the risk of distorting the response to waves
induced in the liquid. The coating comprised a single layer PVOH solution with the addition of green dye to enable
visual contrast and was laid down to yield a wet thickness of 75µm at a coating speed of 0.25 m/s in order to achieve
good coating stability and uniformity. The viscosity of the solution prior to coating was 9.9 mPas when measured at
23°C. Dynamic surface tension was measured using an inclined plane apparatus described by Tricot (1997) as a
function of surface age.. The timescales over which surface tension gradients are likely to affect wave growth and
damping are in the range of 1 to 10 s. The lower limit is dictated by the slow speed at which the fluid moves in
response to air flow and the upper limit by the duration of the experiment. Values over these timescales vary
between 33.0 and 36.5mN/m. The finding that surface tension varies only slightly with surface age indicates that
surfactant is diffusing quickly in this case. The work of Valentini et al (1991) suggests that this could lead to large surface elasticities which would in turn increase the damping of free surface disturbances produced by external pressure disturbances. This is borne out here by related experiments which shows that the coating levels quickly when it is extracted prematurely from the dryer.

The speed of the sample was reduced to 0.12 m/s prior to entering the dryer and maintained at this value throughout the dryer. Air was supplied to the nozzles at 250Pa resulting in an air impingement speed of approximately 19.4m/s. The impingement nozzles were equi-spaced at intervals of 73.5mm and at a height of 9mm above the web. Further analysis using a well-established drying model, Cary & Gutoff (1991), suggests that the temperature falls rapidly during the period investigated in this experiment to equilibrate to a wet bulb temperature of 16.4°C. The estimated temperatures for the four recordings shown in Fig.8a are 20.8°C, 18.1°C, 16.4°C and 16.4°C respectively. The corresponding estimated viscosities, taking into account the limited amount of evaporation and hence increase in solids concentration predicted by the drying model are 11.7 mPa.s, 12.9 mPa.s, 13.9 mPa.s and 14.1 mPa.s. The waveform captured by the 50mm field of view of the camera is shown in Fig. 8(a). The disturbances are initially random but grow by displacement of liquid generally parallel to the machine direction away from thinner areas and towards thicker areas until a wavelike pattern emerges. The phase of the waveform then remains stationary relative to the web. Between jets 10 and 11, the wavelength of the disturbance varies between 5 and 7mm, a range of values that is consistent with that typically found in Gosset and Buchlin’s (2003) experimental study of air induced disturbances in gas-jet wiping systems. The rate of wave amplitude growth is shown in Fig. 8(b) and is found to be proportional to $t^2$ for the time range 0-6 seconds covered by the experiment as determined by the web speed and finite length of rail, where time, $t$, is in seconds. (Fig. 8 inserted here).

In many coating applications, in the absence of external pressure disturbances, it is the characteristic flow Capillary number (ratio of destabilising viscous to stabilising surface tension stresses) that determines the dominant wavelength of the free surface disturbance and, ultimately, the overall stability of the flow, Weinstein & Ruschak (2004). In this example the Capillary number, $Ca=(viscosity \times web\ speed)/surface\ tension$, is small so that the dominant destabilising mechanism is clearly the external pressure fluctuation. The overall complexity of this flow, which depends on the interaction of turbulent air flow with complex free surface phenomena, such as surface elasticity and surface tension gradients caused by the presence of surfactants, Tricot (1997), complicates any simple analysis of the flow dynamics observed here. However, the recent work of Buchlin and co-workers [Laconette et al. (2006), Gosset & Buchlin (2007)] suggests that important insight may be provided by the development of a numerical model of the instability mechanism where turbulent pressure profiles are coupled into numerical solutions.
of the thin film lubrication equations. The results presented here provide valuable data for validating any such model, the development of which presents an interesting topic for future research.

An important issue when measuring the onset of waves is that, as illustrated in Fig. 2, too prolonged a period under an array of nozzles discharging air causes the fluid to continually travel away from each impingement zone and hence gather into bands. Experiments carried out here showed that the onset of waves is dictated by the maximum air speed from the nozzles rather than the web speed and that bands are avoided simply by ensuring that the residence time under the nozzles is smaller than a critical value. For the experiments carried out here to study and quantify wave amplitude, the lower machine speed limit of 0.12 m/s used here was found to be large enough to avoid the development of bands.

3.2. Dependence of Wave Amplitude on Viscosity and Wet Thickness

Having established that the web speed selected for the study, 0.12 m/s, gave representative effects, surface disturbances for two green dyed PVOH coating solutions were studied, with viscosities 10 mPas and 20 mPas at 23°C. Three wet thicknesses were evaluated for each viscosity, the lower limit being dictated by coating constraints and the upper limit by excess movement under the dryer when operating at the minimum achievable air pressure. Results are shown in Figs. 9 and 10 for the 10 mPas and 20 mPas coatings respectively. Running the drying model of Cary and Gutoff (1991) indicates that the temperature of the fluid would have fallen for each recording to close to the wet bulb temperature of 16.5°C by the time the sample had arrived at the point where the waves were recorded. The resultant viscosities are included in the caption for each graph shown in Figs. 9 and 10. The rms wave amplitude indicates the deviation in coating weight normalised with respect to the mean value. The results clearly show that the amplitude of the surface waves increases with both air speed and wet thickness in both cases and decreases as viscosity increases. (Figs. 9 and 10 inserted here).

3.3: Benefits of Optimising Water Distribution using a Carrier Layer

Further pilot drying experiments were conducted for typical industrial conditions with a gelatine/cellulose mixture of viscosity 43.9 mPas and a wet thickness of 96.6 µm. (Figs. 11 and 12 inserted here).

Figs.11 and 12 are illustrative of how a coating engineer typically sets up an industrial dryer to achieve the highest possible drying speed without incurring damage to the product due to surface waves. Fig.11(a) shows how the experimentally determined heat transfer coefficient increases with air speed and thus indicates the advantage of
increasing the applied pressure at any zone to the maximum that can be sustained by the coating without unacceptable surface disturbances. Fig.11(b) shows how temperature is typically increased to attain higher evaporation rates as the surface becomes more robust against surface disturbances. The pressure applied at any instant throughout drying is then varied by trial and error so as to achieve a maximum without incurring visible surface waves - finally yielding an optimum profile as typically shown in Fig. 12(a). Figs. 12(b) and 12(c) show residual water content and evaporation rate respectively as determined from the predictions of the drying model as set up by Cary and Gutoff (1991). The initial rising evaporation rate corresponds to the constant rate period. This is followed by a steep decline owing to the evaporation then being dictated by diffusion of water to the surface of the coating.

Fig.13 shows the results of an experiment aimed at reducing surface waves during the critical initial part of the drying process shown in Figs.11 and 12. Here, the effect is examined when replacing the formulation A, which is effectively a single layer, with a double layer formulation B, the properties of which are given in Table 2. (Fig. 13 inserted here).

Formulation B is arranged to deposit the same overall solids content as formulation A but, as suggested by Dittman & Rozzi (1977) and (Choinski (1978), having a diluted lower “carrier” layer to increase coatability. Water is removed from its upper layer in order to reduce the overall drying load. Fig. 13 shows the surface wave amplitudes for a range of air pressures and hence air speeds for the two formulations shown in Table 2. It clearly demonstrates that the use of a carrier layer enables significantly higher air speeds to be used without incurring unacceptable damage due to surface waves. In order to investigate the mechanism for this improvement, it was subsequently found impossible to coat the upper more viscous layer on its own using the slide bead process available in the laboratory. On replacing the lower layer with one comprising the same solution as used for the upper layer and controlling the wet thickness to that of the original carrier layer in formulation B, a simple visual check indicated that waves only became visible as the pressure exceeded 500 Pa. This suggests that the dominant mechanism for the improved robustness of formulation B is due to the increased viscosity of the upper layer.

For the particular case of a jet pressure of 400 Pa generating an air speed of 24.4 m/s, for example, the surface wave amplitude is an order of magnitude smaller for the carrier layer formulation. Moreover, the value of the heat transfer coefficient for the array of nozzles at 250 Pa working pressure was found to be $169 \text{ Wm}^{-2}\text{K}^{-1}$ at a drying gap of
Fig. 13 shows that the carrier layer formulation enables the working pressure to be increased from 281 Pa to 697 Pa if the target rms wave amplitude is set to the arbitrarily small value of 0.007. The effect of the increased pressure on heat transfer coefficient can be determined using Cary and Gutoff’s (1991) estimate that heat transfer coefficient varies according to the expression 

\[ h = h_0 \left( \frac{\Delta P}{\Delta P_0} \right)^{0.39} \]

where \( h_0 \) is the heat transfer coefficient at the reference pressure difference, \( \Delta P_0 \). Since the evaporation rate during the early drying phase, when water molecules are free to migrate to the surface, is proportional to \( h \) it can be estimated that the carrier layer formulation enables the evaporation rate to be enhanced by a factor of \( \left( \frac{697}{281} \right)^{0.39} \), i.e. a 43% increase. This is another significant benefit in addition to the overall reduction in water load.

4. CONCLUSIONS

Although several previous studies have considered the stability dynamics of thin films where defects are induced by, for example, substrate topography or the presence of moving contact lines, comparatively few have studied the important class of air-induced surface disturbances to which low viscosity coating systems are particularly susceptible. A range of new experimental techniques has been developed which enable these disturbances to be quantified and related to the viscosity and wet thickness of the coatings. Experiments show that the surface disturbances take the form of a wavelike pattern which travels at the same velocity as the web and whose amplitude increases rapidly with wet thickness and decreases with viscosity in much the same way as predicted by Ruschak (1987) for forced disturbances from air issuing from parallel rows of holes. Recent modelling approaches applied to the related problem of gas-jet wiping suggest that further insight into the physical mechanisms controlling the onset and growth of drying-air induced disturbances may be provided by a coupled turbulent-lubrication numerical flow model. The results presented here provide useful data for the validation of such models.

Heat transfer measurements for multi-layer coating systems show that redistributing water within a single layer coating to form a lower “carrier” layer and a more concentrated upper layer can lead to significant commercial benefits in the drying phase in terms of reduced drying load and increased drying rates. These are in addition to the well-known benefits of carrier layers that have been reported by Dittman & Rozzi (1977) and Choinski (1978) in terms of increased coating speeds and robustness to defects in the wet coating phase.

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6. REFERENCES


FIGURE CAPTIONS

Figure 1: A double-layer slide-bead coating system.
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Figure 13: Surface wave instability for Formulations A and B.
### NOTATION

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<thead>
<tr>
<th>Term</th>
<th>Unit</th>
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<td>Heat transfer coefficient</td>
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<tr>
<td>Reference heat transfer coeff.</td>
<td>$h_0$ (W/m$^2$K)</td>
</tr>
<tr>
<td>Pressure difference</td>
<td>$\Delta P$ (Pas)</td>
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<td>Reference pressure difference</td>
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