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Two layer slot coating: die configuration and frequency response analysis

Danmer Maza Quinones
Márcio da Silveira Carvalho
Department of Mechanical Engineering Pontifical Catholic University of Rio de Janeiro - PUC-Rio
danmerm@puc-rio.br, msc@puc-rio.br

Abstract. *Coating uniformity requirements is becoming more severe as new products come into the market. Multi-layer slot coating has to be designed not only based on the steady state operation, but also taking into account how the flow responds to ongoing disturbances, such as flow rate, gap or vacuum pressure oscillations. These disturbances may lead to thickness variation on each deposited liquid layer that may be unacceptable for product performance.*
This study extends available transient analysis of single layer slot coating to determine the amplitude of the oscillation of each individual coated layer obtained by two layer slot coating in response to small periodic variation on the coating gap, web speed and flow rate as a function of process conditions and frequency of the perturbation. The predictions are obtained by solving the complete transient Navier-Stokes equations for free surface flows. The results show how the thickness variations on each deposited liquid layer can be minimized by changing the geometry of the die.

Keywords: *Multilayer coating, Periodic disturbances, Galerkin/FEM, Transient response, Free surface flow.*

1. INTRODUCTION

Multilayer slot coating is one of different coating methods largely used in the manufacturing process of many different products. This method is used when complex functionality of each layer is required and it help to reduce the production cost because it is capable of applying multilayer in a single pass and solidify them together. Specifically in two layer slot coating the process consists in depositing two thin uniform liquid layers onto a moving substrate through different feed slots. The two liquid phases are separated by an inter-layer attached to the die surface as shown in Fig. 1. The flow into the coating bead is strongly affected by the operating conditions such as coating gap, flow rate of each layer, web speed, vacuum pressure, liquid properties and die configuration.

The region in the space of operating parameters of a coating process where the delivered liquid layer is adequately uniform is usually referred to as coating window. Knowledge of coating windows for different coating methods is needed in order to predict in steady state regime whether a particular method can be used to coat a given substrate at a prescribed production rate. There has been a lot of works in the past both theory and experiments trying to understand the boundary of this process to see at which conditions is possible to have a two dimensional flow, steady state flow and stable flow.

Cohen (1993) in his experimental analysis shows that the inter-layer start at the separation point located somewhere in the mid die block. Taylor and Hrymak (1999) in their analysis on two miscible liquids show that diffusional and convective mixing occurs between layers under certain operating condition. Sartor (1990) observed that the ideal location of the separation point is the downstream corner of the mid die lip that leads to desired steady flow. Musson (2001) concluded that the inter-diffusion zone for miscible liquids can be considered as an inter-layer with zero interfacial tension because the residence time in the coating bead is extremely small. Nam and Carvalho (2009, 2010) analyzed by theory the stability of two layer slot coating and the location of the "separation point" at different operation condition.

A lot is known about steady-state operation and limits of operability of slot coating process but coating uniformity requirements is becoming more severe as new products come into the market. In optical products for example the film thickness variations should be typically less than 1% of the film thickness. Multi-layer slot coating has to be designed not only based on the steady state operation, but also taking into account how the flow responds to ongoing external disturbances. These disturbances may lead to thickness variation on each deposited liquid layer that may be unacceptable for product performance.

In a manufacturing plant, there are inherent periodic disturbances at different frequencies that influence the uniformity of the coated layer. In particular case of two-layer slot coating process, the external disturbances are usually periodic variations on the coating gap given by roll run out or mechanical vibration or non-uniform thickness of the substrate, periodic variation of the flow rate given by fluctuation of the liquid supply or pressure, vacuum pressure variations by pumping fluctuations or acoustic vibration, and web speed variation by drive imperfections as shown in Fig. 1.

It is important to know how sensitive is the steady flow to these external disturbances, even if the flow is stable with respect to them and to determine many different sets of operating conditions inside the coating window for a given product specification, which one will produce a more uniform final deposited layer. Once the flow response is known, the process may be designed to minimize the coating thickness variation.

Katagiri and Scriven (1996) analyzed the response of the flow that occurs in slide coating process to oscillations on flow rate by this approach. They found two natural frequencies of the system at which the response was locally amplified.

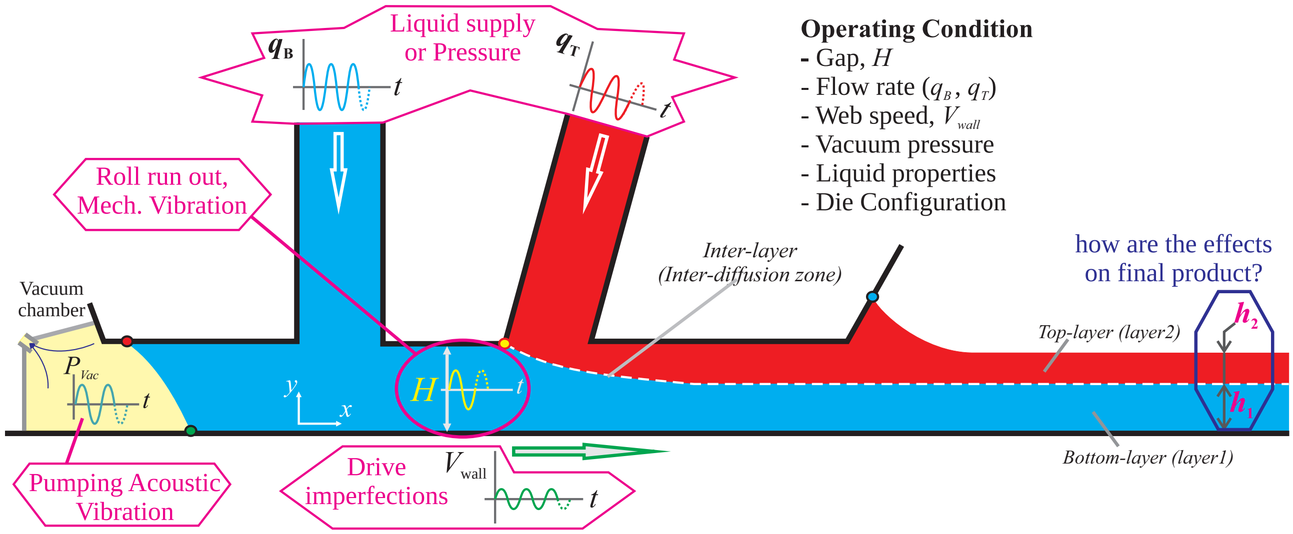

Figure 1. Schematic view of a two layer slot coating and its external disturbances.

Sensitivity of single-layer slot coating flows to periodic disturbance was analyzed experimentally by Joos (1999). The flow was excited by imposing an oscillatory variation on the flow rate and vacuum pressure at different frequencies and the down-web variation of the coating thickness was measured at each condition. Results show how the amplitude of the film thickness variation changes with the frequency of the imposed disturbance.

Cho and Lee (2003) also used this approach to analyze the dynamics of the flow in a baffled fuel tank in order to determine the best baffle configuration that minimizes the sloshing effect.

Romero and Carvalho (2008) solved the transient flow to analyze the film thickness oscillation in single layer coating process due to periodic variation on the flow rate fed and on the coating gap. The analysis showed the most dangerous frequencies for each type of disturbance and the die configuration may be altered in order to reduce the sensitivity of the flow to periodic disturbances.

Perez and Carvalho (2011) have used the predictions of the transient flow to evaluate the objective function of a bound-constrained optimization problem in order to determine the values of vacuum pressure and coating gap, the two easiest parameters to control in a coating line, that minimize the amplitude of the film thickness oscillation at a fixed web speed and flow rate.

Previous works, however, did not discus in how those external disturbances can affect the final thickness of each layer in multilayer slot coating process and which available coating die design could reduce better those perturbations and finally see the best process condition to be operated.

In this work, we extend available transient analysis of single layer slot coating to determine the amplitude of the oscillation of each individual coated layer obtained by two layer slot coating in response to periodic variation on the coating gap, web speed and flow rate as a function of process conditions and frequency of the perturbation.

2. Mathematical Model

The mathematical formulation of the transient slot coating flow was presented in detail by Romero and Carvalho (2008) and Nam and Carvalho (2010); it is only briefly summarized here.

The velocity v and pressure p fields of the transient, two-dimensional, incompressible flows are governed by the continuity $\nabla \cdot v = 0$, and momentum $\rho_i(\partial v/\partial t + v \cdot \nabla v) - \nabla \cdot \mathbf{T}_i = 0$, equations for each layer. Where ρ_i is the liquid density. The total stress tensor for Newtonian liquids is $\mathbf{T}_i = -p + \mu_i[\nabla v + (\nabla v)^T]$ where μ_i is the liquid viscosity. Here, subscript i defines the two liquid phases, $i = 1$ for the top layer and $i = 2$ for the bottom layer. Because of the small dimensions of the flow, body forces are usually neglected in coating flows. Boundary conditions are needed to solve the Navier-Stokes system. In two-layer slot coating flow, the domain is bounded by inflow and outflow planes, solid walls and free surfaces (gas-liquid interfaces) and the surface that separates the two liquids, the inter-layer, as shown in Fig. 2.

Initial condition is needed in order to solve the transient flow. In this work, the steady state solution of the flow was used as the initial condition for the transient analysis $v(t = 0) = v_0$, $p(t = 0) = p_0$ this initial condition had to be computed at each set of operating parameters. The periodic disturbance on the flow rate leads to a transient response of the flow. The thickness of the each deposited liquid layer $h_i(t)$ varies periodically around the steady state value $h_i(t = 0) = h_{i0} = q_{i0}/V_{wall}$ leading to a non uniform film along the downweb direction h_{i0}. The amplitude of the oscillation h_{im} and the phase lag of the thickness response ϕ_i are unknown and need to be determined for each condition.

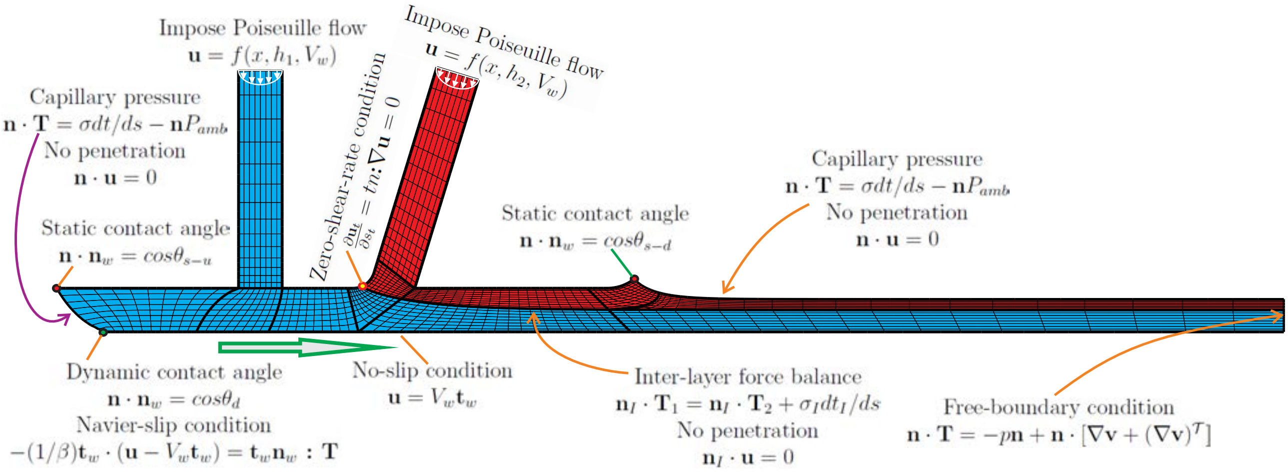

Figure 2. Boundary Conditions and the finite element mesh.

The ratio of the relative amplitude of the film thickness oscillation to the imposed operating parameters disturbance (λ) is called the amplification factor defined as $\alpha_{ij} = [h_{im}/h_{i0}]/\lambda_j$ where λ_j is associate with the source of the disturbances and represent the relative amplitude of the disturbance parameter. Here, subscript j defines different disturbances, $j = q$ for the flow rate, $j = H$ for the gap and $j = w$ for the web speed oscillation. For example $\lambda_H = H_m/H_0$.

Flows with free surfaces and inter-layer give rise to a free-boundary problem. The flow domain is unknown a priori, and it is part of the solution. To solve a free-boundary problem by means of standard techniques for boundary value problems, the set of differential equations and boundary conditions posed in the unknown physical domain have to be transformed to an equivalent set defined in a known, fixed computational domain. This transformation is made by a mapping $\mathbf{x} = x(\boldsymbol{\xi})$ that connects the two domains. The physical domain is parameterized by the position vector $x = (x, y)$ and the reference domain by $\boldsymbol{\xi} = (\xi, \eta)$. The mapping used here is the one described by de Santos (1991).

The system of governing equations together with the appropriate boundary conditions and initial condition was solved by Galerkin's method with quadrilateral finite elements. The temporal discretization of the set of ordinary differential-algebraic equations follows the first-order fully implicit Euler method. A mesh with 1,353 elements (26,760 degrees of freedom) was as shown in Fig. 2 considered satisfactory and was used to obtain the solutions reported here. To keep the error less than 2%, a time step of $\Delta t \approx 70\omega$ was adopted in all computations, i.e. 140 time steps were used per cycle of the imposed periodic perturbation.

3. Results

As mentioned before, the initial condition for the transient analysis was set at a given parameters, as shown in the table 1, that allows a steady-state flow and stable flow. Once a steady-state solution was obtained, a periodic variation on the flow rate with a prescribed amplitude and frequency was imposed. It is important to establish the spectrum of frequency of this typical mechanical system. For computational purpose we use values of frequency from $< 0.1 - 1000 >$ Hz.

3.1 Pinned and unpinned effect: Downstream static contact line

In the literature, many authors in the past have been generally assumed that the downstream static contact line (DSCL) pinned at the edge of the slot as shown in Fig. 2. Sartor (1990) examined experimentally the flow structure for various fluid and geometrical parameters where he shows the meniscus can climb along the die shoulder or recede into the bead along the downstream die according to certain operation conditions. But, Sartor (1990) in his theoretical analysis was consider the case with the meniscus pinned on the corner. Kapur (1999), was one of first in present numerical results with the downstream meniscus free to climb the die of the knife coating where the free surface are parameterized by their location along conveniently placed spines (Kistler and Scriven (1983)). Romero and Carvalho (2006) made a theoretical analysis on rounded edge effect at different radius of curvature of the downstream die and shoulder where the local contact angle is treated as a specified equilibrium value. These previous analysis were focus on steady state regime. Romero and Carvalho (2008) and Perez and Carvalho (2011) solved the transient flow of the slot coating of single layer without consider the free movement along the die shoulder. For a dynamic response analysis is necessary to consider more realistic condition: rounded edge and downstream static contact angle able to move along the die. Figure 3 shows the map of the amplification factor when the vacuum pressure is perturbed. At low frequency the rounded and unpinned effect is not important. At intermediate frequency the bottom layer response with a natural frequency at 80 Hz approximately and α_{1Vac} is amplified

Table 1. Parameters for a typical two layer slot coating and dimensionless parameters

Operating parameters

Parameter	Unit	Symbol	Range	Base Case
Gap width	μm	H	50 - 300	250
Web speed	ms^{-1}	V_w	0 - 5	1
Pressure difference	kPa	P_{vac}	0 - 10	2
Dynamic contact angle	rad	θ_d	2.4 - 2.8	2.44
Upstream static contact angle	rad	θ_{s-u}	0.35 - 1.57	1.05
Downstream static contact angle	rad	θ_{s-d}	0.35 - 1.57	1.57
Interfacial tension	mN/m	σ_I	0 - 1	1
Density	Kgm^{-3}	$(\rho_1 = \rho_2)$	0.9 - 1.3	1.2
Viscosity	$mPa.s$	$(\mu_1 = \mu_2)$	0.5 - 100	25
Surface tension	mN/m	$(\sigma_1 = \sigma_2)$	20 - 70	60

Dimensionless Parameters

Group	Definition	Base Case
Reynolds number	$Re = \rho V_w H/\mu$	12
Capillary number	$Ca = \mu V_w/\sigma$	0.42
Vacuum Pressure	$Vac = P_{vac} H/\sigma$	6.25
Bottom-layer thickness ratio	$G_1 = H/h_1$	2
Top-layer thickness ratio	$G_2 = H/h_2$	4

in 20% related to pinned condition. The bottom layer at higher frequency shows no difference between those effects. So, it doesn't matter if it's pinned or not. The top layer response with two resonances. At pinned condition the natural frequency is around 200 Hz and for unpinned effect this frequency is anticipated at 160 Hz approximately reducing α_{2Vac} in 17%. At higher frequency the rounded and unpinned effect reduce α_{2Vac} in 50% the second resonance frequency at 716 Hz. In principle, the free movement of the downstream static contact line has an effect to change the curvature of the downstream meniscus and it change the capillary pressure that tends to stabilize the bead reducing the perturbation as we can see in the top layer.

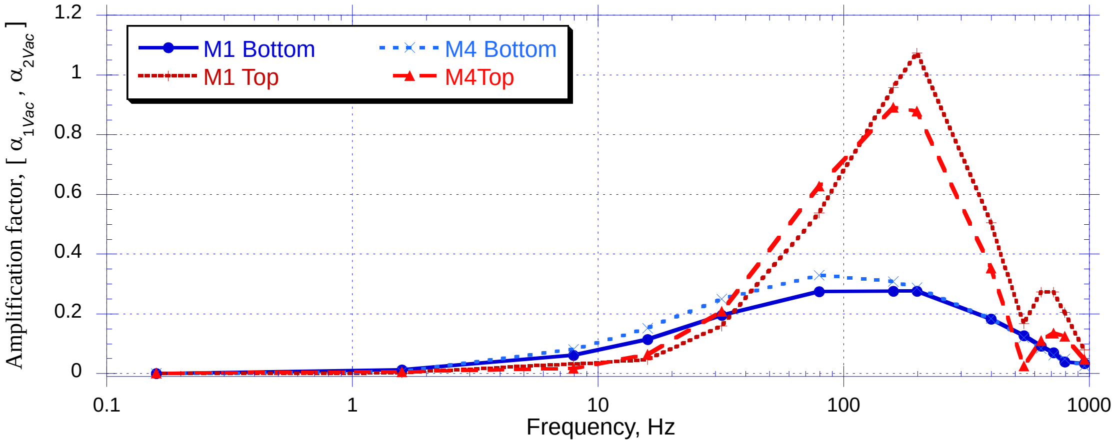

Figure 3. Comparison between M1(DSCL pinned) and M4(DSCL free to move along the die shoulder)

3.2 Die Design and dynamic response analysis to external disturbances

Large number of die lip configuration disclosed in the patent literature in order to improve the production rate and maintain an stable flow. Sartor *et al.* (1998) adjusted the die geometry to control the pressure gradients in the coating bead as we can see in Fig. 4. Comparison between those three different die configurations was considered to see which configuration reduce better external disturbances. Those external disturbances are represented by periodic variation of bottom and top flow rate, gap distance, vacuum pressure and web speed. The base case was analyzed in previous work by Maza and Carvalho (2012).

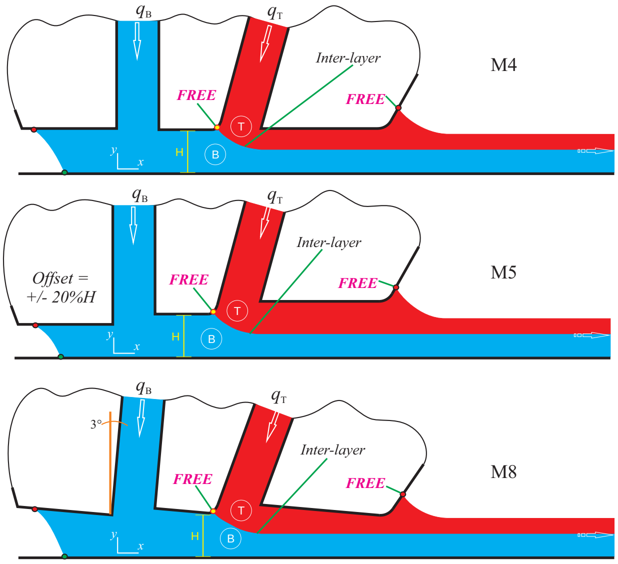

Figure 4. Comparison between a base case M4 (up-side), a underbite die lip M5 (middle) and a convergent die lip M8 (down-side) configuration given by Sartor *et al.* (1998)

3.3 Sensitivity to bottom flow rate variations

At low frequencies, in Fig. 5, i.e. $f \leq 10$ Hz the coated film thickness of the bottom layer h_1 is proportional to the imposed flow rate q_1 and consequently the amplification factor is equal to $\alpha_{1q} \approx 1$ and the film thickness of the top layer h_2 keep going constant in the down-web direction, in this layer the oscillation was completely damped. At frequency $f \geq$ 10 Hz, the diffusion of momentum occurs in a time scale comparable to the imposed perturbation and the amplification factor of the bottom layer decrease monotonically as the frequency rises with -0.45/decades of slope. On the other hand, the film thickness variation of the top layer increase as a frequency rises until it reaches a maximum $\alpha_{2q} \approx 1.8$ at natural frequency $f \sim 200$ Hz approximately. At the conditions reported here, this oscillation would cause a variation of the top layer thickness along the downweb direction almost 1.8 times for this flow rate oscillation, which may be unacceptable for many different products. However, after this natural frequency prediction shows the amplification factor α_{2q} decrease as the frequency rises and then appear another small resonance around 750 Hz. Result shows that the frequency of the flow rate of the bottom layer should be kept below 40 Hz or above 300 Hz in order to avoid the resonance. The pump system needs to work at high frequency that means at high rotations rate to damp perturbations. For practical reason the selection of the pump size and pump rotation should be such as to avoid these critical frequencies during operation. Those three configurations don't have effect for this disturbance as we can see in Fig. 5. The amplitude of perturbation was set to $q_{1m} = 0.1q_{10}$.

Comparing this prediction with single layer coating analysis obtained by Romero and Carvalho (2008) we can conclude that the bottom layer has a qualitative behavior similar to flow rate disturbance in single layer and the top layer behavior is similar to gap disturbance in single layer coating. However for this last case the qualitative behavior is just in appropriate frequency range. In single layer the gap disturbances at high frequency the gain oscillates around 1 and it does not dumb perturbation.

3.4 Sensitivity to top flow rate variations

Figure 6 shows the sensitivity to flow rate of top layer. The amplitude perturbation was set to $q_{2m} = 0.1q_{20}$. It is clear that the bottom layer flow is insensitive to this disturbance. The top layer decrease monotonically as the frequency rises and the bottom layer remains insensitive at different frequencies. For this case, the diffusion momentum occurs at 20Hz and -0.75/decade of slope and it is higher than the bottom layer in previous case showed at Fig. 5. However, there is no relevant effect using those different configurations at this perturbation.

3.5 Sensitivity to gap oscillation

In the present analysis we can see the system is really sensitive to gap perturbations as shown in Fig. 7. Around 240 Hz it excites a maximum amplitude ratio of 11.3 for the configuration M8 and 9 for configuration M5 in the top layer. At lower frequencies until get 16 Hz the response is less than 0.75 after that increase as the frequency rises the first natural frequency at 240 Hz. After that, decrease until get 540 Hz and then increase until obtain the second natural frequency in

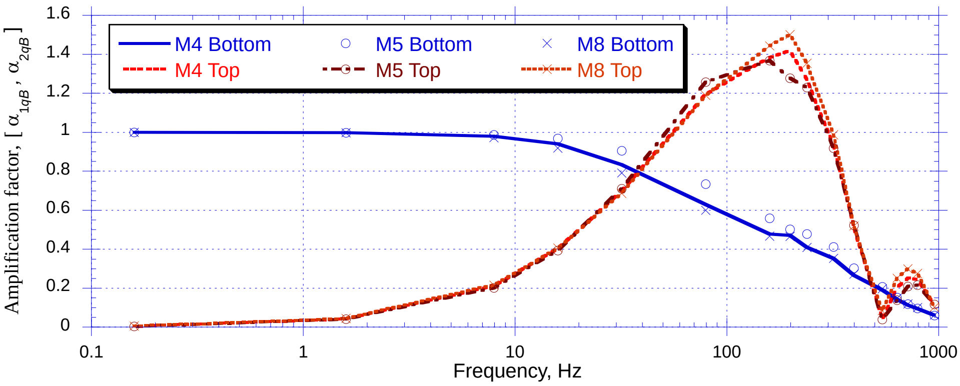

Figure 5. Comparison between base case M4, a underbite die lip M5 and convergent die lip M8 for bottom flow rate oscillation

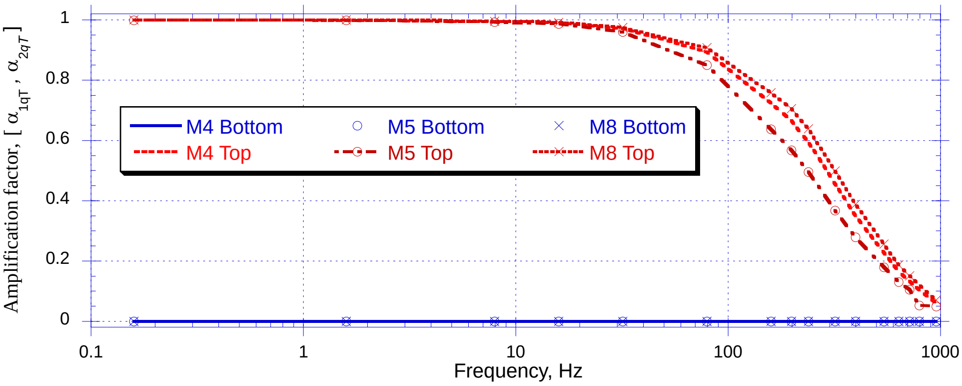

Figure 6. Comparison between base case M4, a underbite die lip M5 and convergent die lip M8 for top flow rate oscillation

approximately 716 Hz and then decrease as the frequency rises.

The other hand, the bottom layer shows a similar behavior than gap disturbance in single layer achieving the maximum gain at 320 Hz and then it decrease as frequency increase until get the amplitude ratio of 1. In this case the configuration M8 has a good response at around the natural frequency but in lowest and highest frequencies the configuration M5 is the best. When the web vibrates the final coated thickness is immediately affected because there is only small quantity of liquid on the web. The amplitude of perturbation was set to $h_m = 0.01H$.

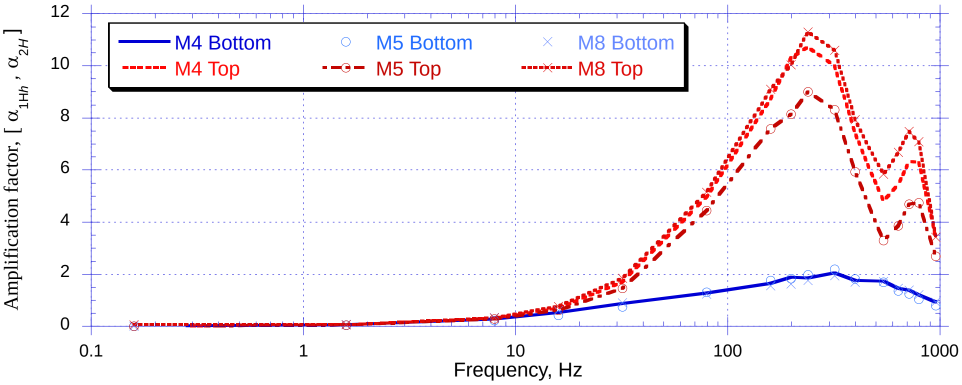

Figure 7. Comparison between base case M4, a underbite die lip M5 and convergent die lip M8 for gap perturbation

3.6 Sensitivity to vacuum pressure variations

The amplitude of the periodic pressure difference across the bead was set to $P_m = 0.1P_{vac}$. At this disturbance the system at low frequency is insensitive in both layers. Above 10 Hz the top layer increases rapidly until get the maximum amplitude of disturbance at the natural frequency around 200 Hz as shown in Fig. 8. The configuration M5 reduces significatively the amplitude at this resonance frequency. The bottom layer gets maximum amplitude at 80 Hz for configuration M4 and M5 the other hand for configuration M8 achieve the natural frequency at 200 Hz. Therefore, the flow into the bead is insensitive to this perturbation. When vacuum fluctuation occurs the bead acts as a reservoir absorbing the perturbation.

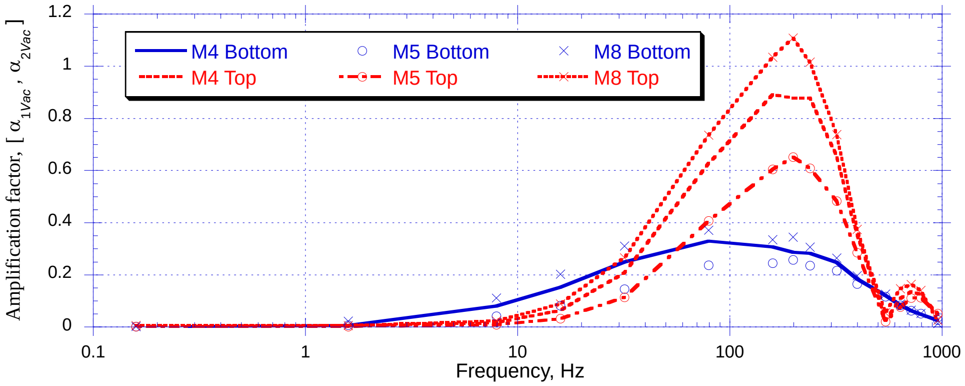

Figure 8. Comparison between base case M4, a underbite die lip M5 and convergent die lip M8 for vacuum oscillation

3.7 Sensitivity to web speed variations

At low frequency the amplitude of disturbance is proportional to wet film thickness variation in both layers as we can see in Fig. 9 but after 10 Hz the diffusion momentum occurs into the flow of the bead region and the Bottom layer start to be damped as a frequency rises with -0.8/decades and after 160 Hz decrease monotonically. The top layer is excited until get the maximum amplitude at 80 Hz. The source of this disturbance start close to the substrate and the interface does not feel this perturbation at high frequency. The configuration M8 absorbed better this perturbation comparatively to the others. For this disturbance the configuration M5 was not the best option.The amplitude of perturbation was set to $V_m = 0.1V_w$

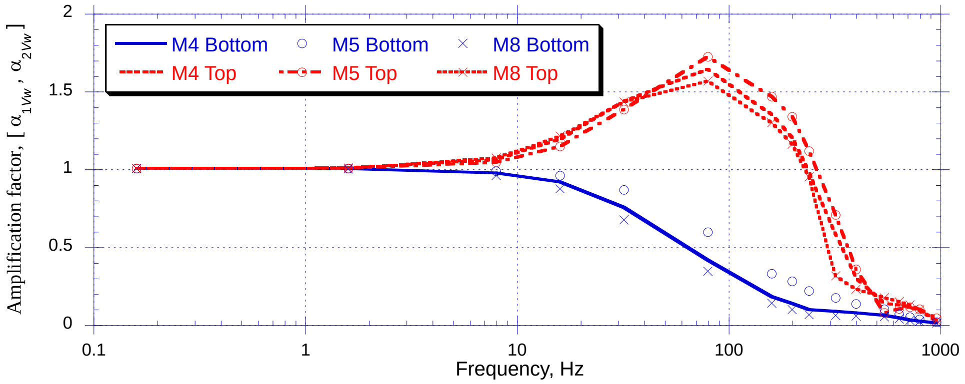

Figure 9. Comparison between base case M4, a underbite die lip M5 and convergent die lip M8 for web speed oscillation

4. Final Remarks

Therefore, Sensitivity to periodic disturbances should be taken into account in designing coating equipment, e.g. coating die, pumps, etc., in order to avoid resonance frequencies. Small periodic oscillation may lead to unacceptable down-web coating thickness variation. At the conditions explored, the top layer thickness is very sensitive to all imposed disturbances, but more strongly to gap perturbation. Different die configuration can be adjusted to reduce effect of periodic perturbations.

5. Acknowledgements

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