Fundamentals of Slot Coating Process

Prof. Marcio Carvalho

http://carvalho.usuarios.rdc.puc-rio.br/

msc@puc-rio.br

DEPARTAMENTO DE
ENGENHARIA
MECÂNICA
PUC - RIO

PONTIFÍCIA UNIVERSIDADE CATÓLICA
DO RIO DE JANEIRO

Introduction

Coating process is the main manufacturing step
for many different (old) products…

Adhesive tapes

Magnetic tapes and disks

Paper

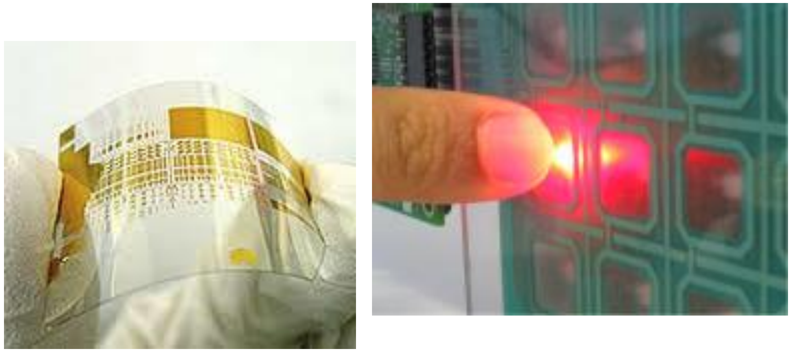
Needs:

Higher yields and faster production speed;

Process improvement and optimization.

Coating process is the main manufacturing step for many different (new) products…

Flexible and transparent electronics



Thin / flexible displays:
Plasma, LCD, OLED, …

Needs:

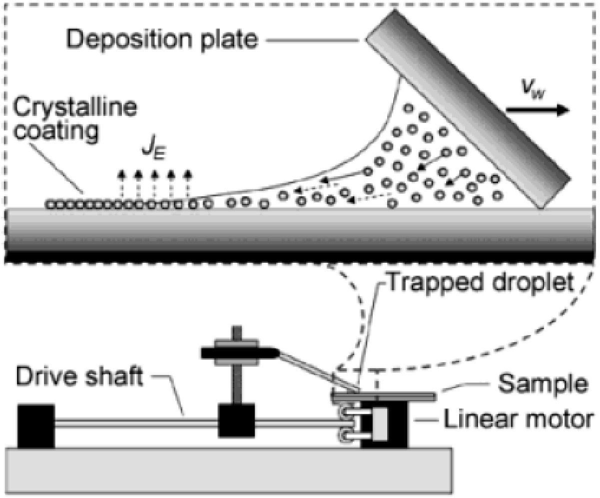
Uniformity requirements are extremely tight;

May need 3D features;

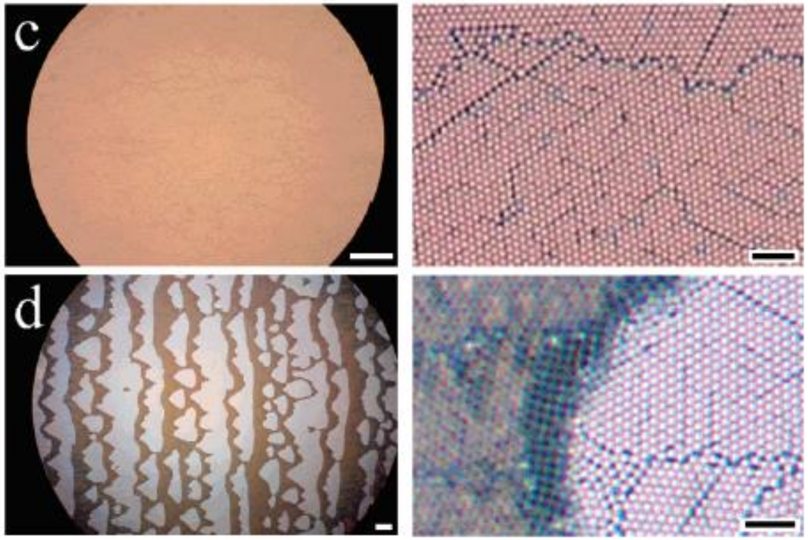
Process optimization to minimize film thickness variation.

… and “is the most promising approach to practical fabrication of nanoparticle structures”

(Fujita & Yamaguchi, 2006)



(Prevo and Velev, 2007)



Needs:

Complex internal microstructure of each layer;

Industrial production speeds (prototype speeds are microns/sec)

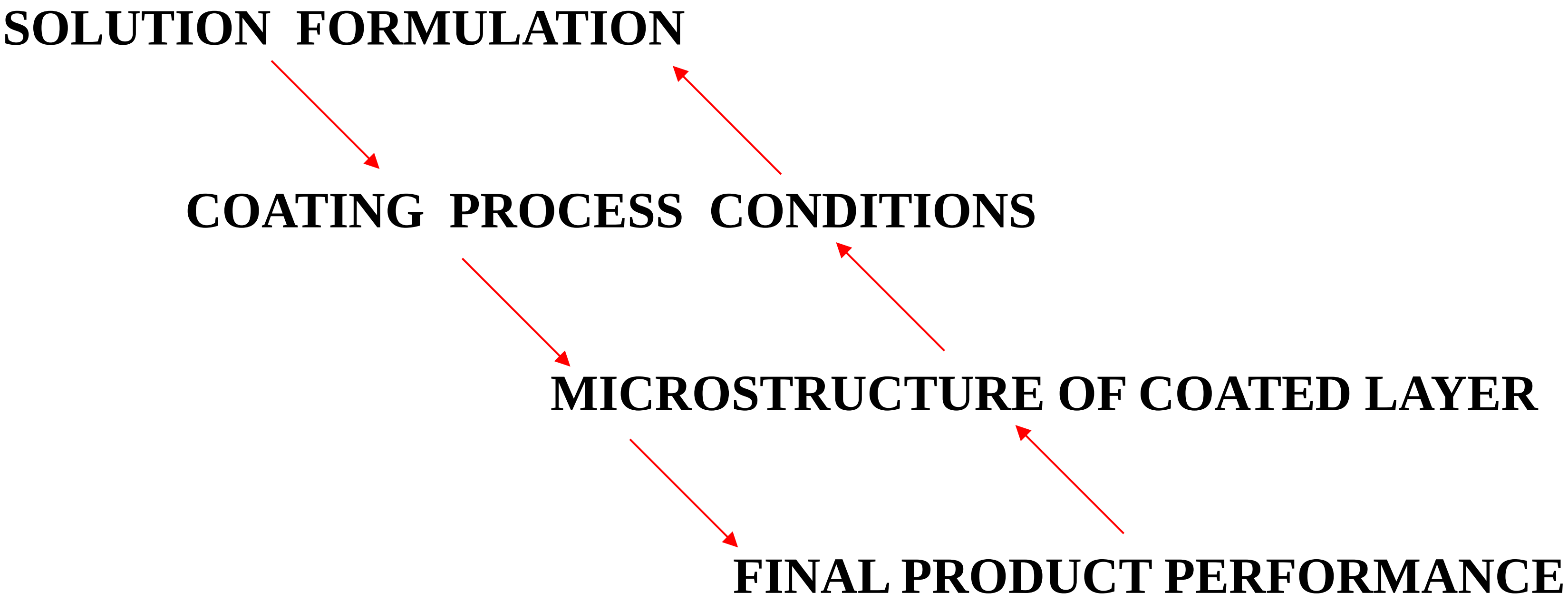
Process development.

FUNCTIONAL COATINGS AND FILMS

Coatings and films produced by ***depositing*** a liquid layer and subsequently ***solidifying*** it are vital ingredients of many different kind of products.

The interior of many coatings and films has to have particular ***microstructure*** or ***nanostructure*** in order to function as intended, whether ***optically***, ***photo-chemically, electronically*** or ***mechanically***.

Fundamental understanding of all steps of the product development and manufacturing is crucial for the product & process optimization



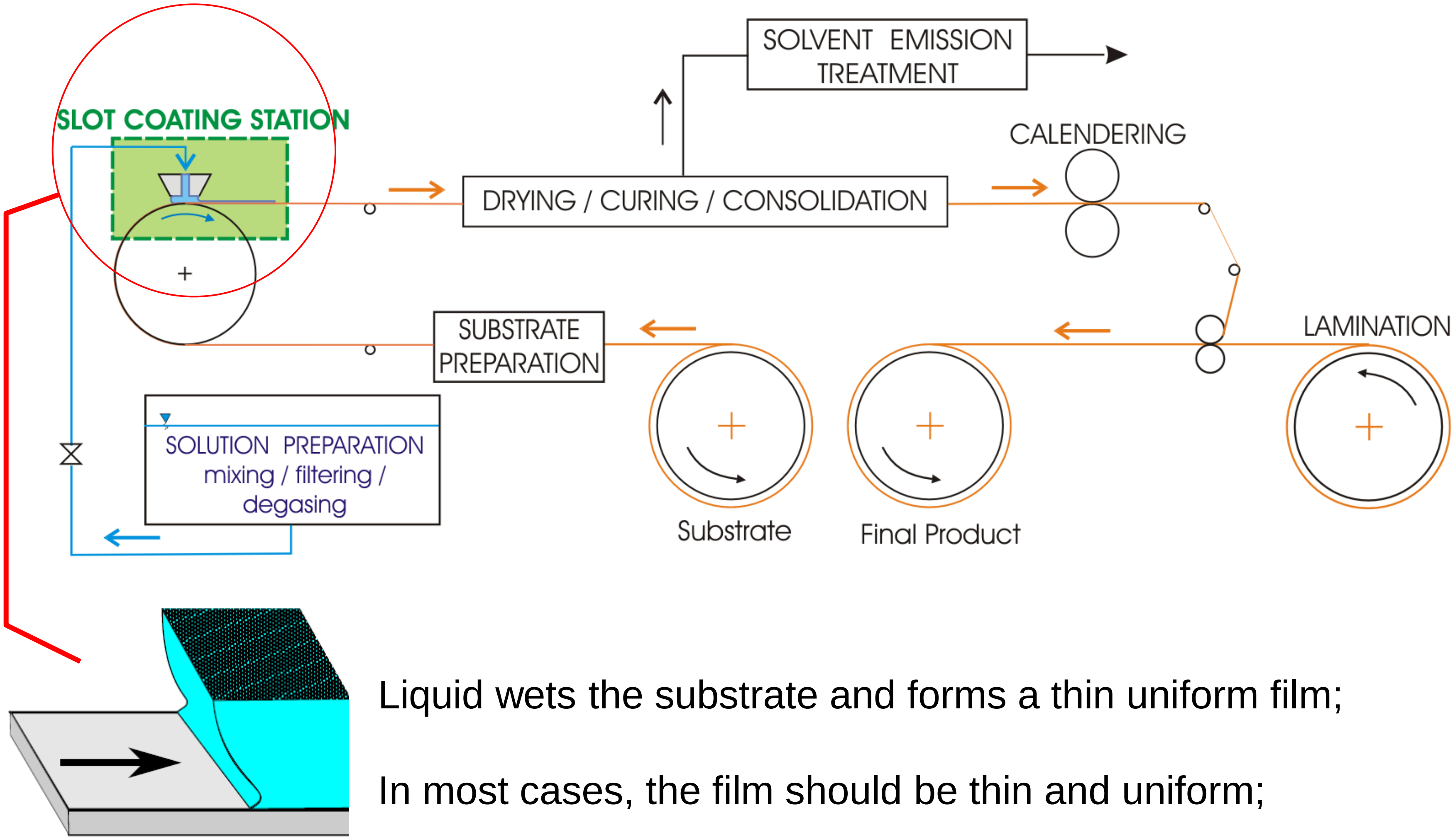
SOLUTION FORMULATION

COATING PROCESS CONDITIONS

MICROSTRUCTURE OF COATED LAYER

FINAL PRODUCT PERFORMANCE

Unit Operations of a typical coating line



SLOT COATING STATION

SOLVENT EMISSION TREATMENT

DRYING / CURING / CONSOLIDATION

CALENDERING

SUBSTRATE PREPARATION

LAMINATION

SOLUTION PREPARATION
mixing / filtering /
degasing

Substrate

Final Product

Liquid wets the substrate and forms a thin uniform film;

In most cases, the film should be thin and uniform;

Limitations on how fast this process can be run.

Development of Coating Technology

- DEVELOPMENTS WERE RESTRICTED TO EACH INDUSTRY SEGMENT
- FIRST PART OF 20TH CENTURY, COATING TECHNOLOGY WAS AN ART
- COATING IS A MULTI-DISCIPLINARY SUBJECT

  WETTING, SPREADING;
  ADHESION;
  FLUID MECHANICS AND RHEOLOGY;
  CHEMISTRY, INTERFACIAL SCIENCE; …

- FROM 1940'S, MATHEMATICAL MODELING AND CAREFUL EXPERIMENTS (NOT ONLY PILOT TRIALS) STARTED TO BE USED TO DEVELOP AND IMPROVE COATING PROCESSES
- COMPETITIVE PRESSURE DRIVES THE TECHNOLOGY, IMPORTANT TO ANALYZE THE PHYSICAL MECHANISMS THAT DETERMINES THE SUCCESS OR FAILURE OF THE PROCESS

Technological Challenges in the Coating Industry

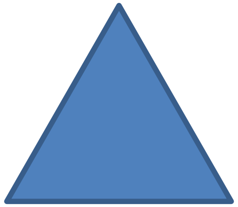
- Thinner and more uniform wet coating layer;
- Reduction of emission of organic solvents –
  more concentrated solutions;
- New coating liquid formulations and
  more complex product structure;
- Discrete and non-continuous coating;
- Increase in line speed and yields;
- Adapting existing coating lines for new products;

⋮

Coating Fundamentals Research

Move from not only Know-how (process developement)
to also Know-why (process understanding)

Need fundamental understanding of the basics mechanisms involved
in all phases of the process: liquid preparation, coating,
and solidification.



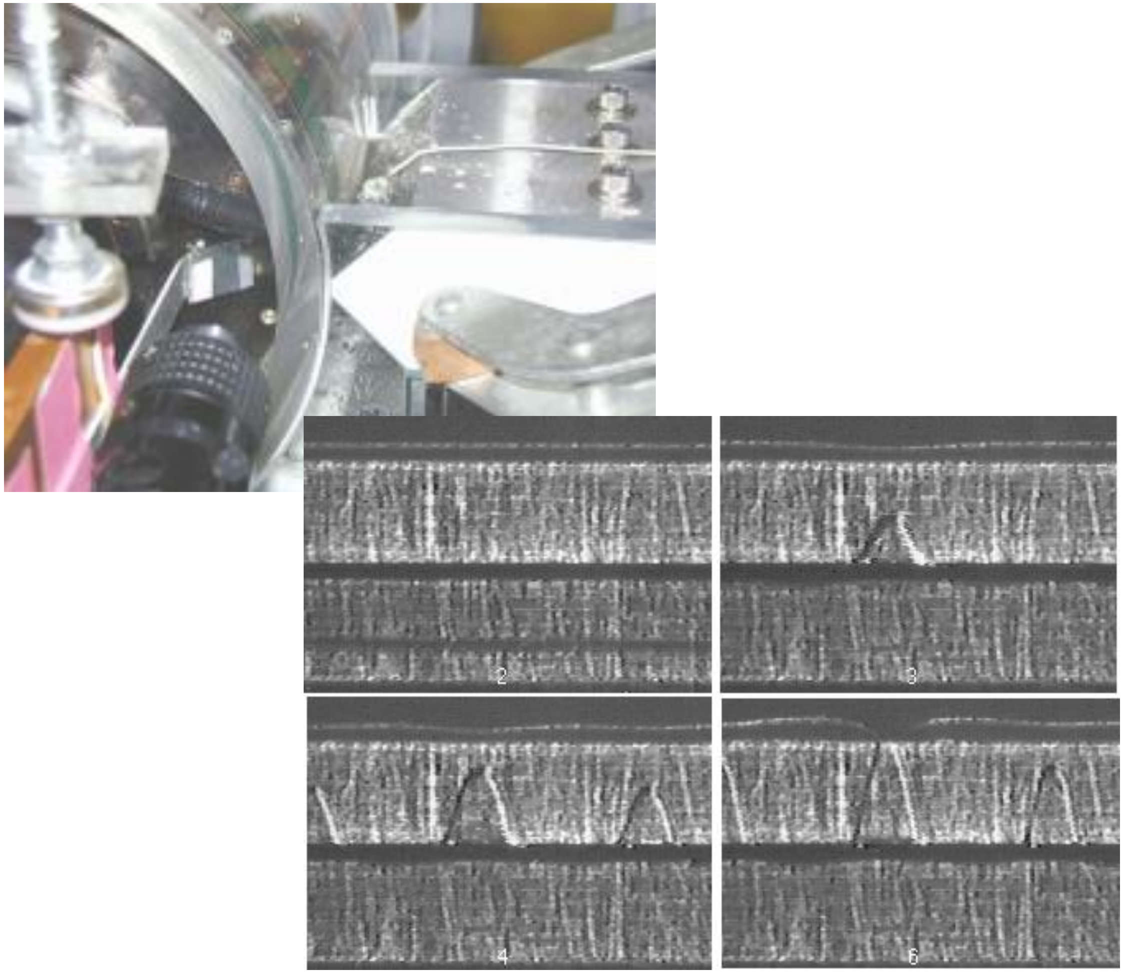
Theory

Experiments

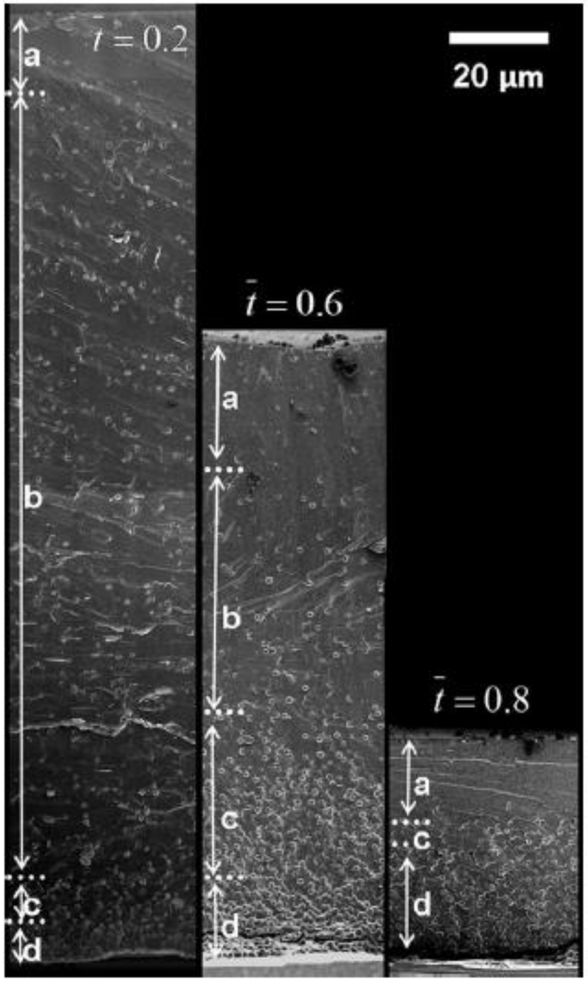
Numerics

Need specially developed experimental and numerical tools
to be able to study in detail all the mechanisms involved.

Flow and microstructure development visualization



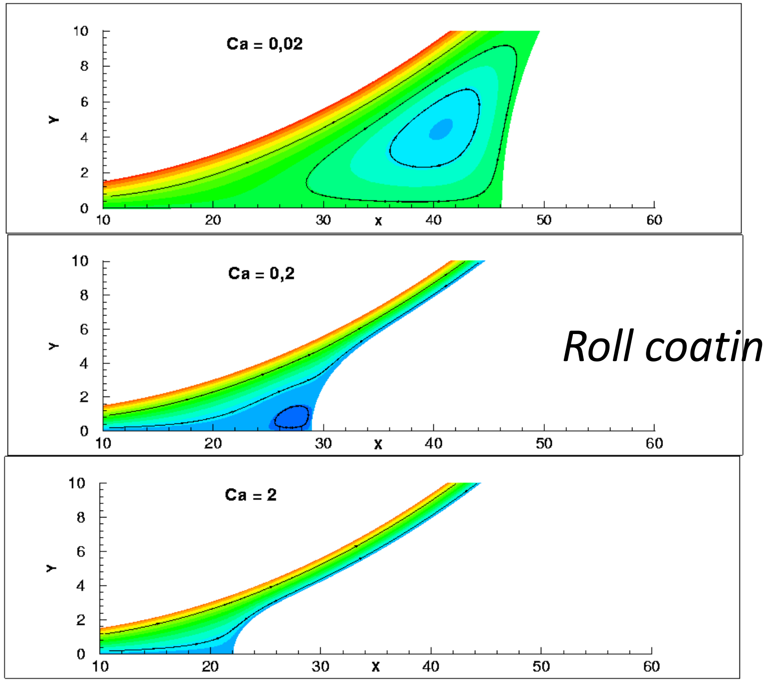
Slot coating visualization: analysis of bead breakup mechanisms – Romero and Carvalho (2004)



Micro structure development During drying – Cardinal and Francis, AIChE J (2011)

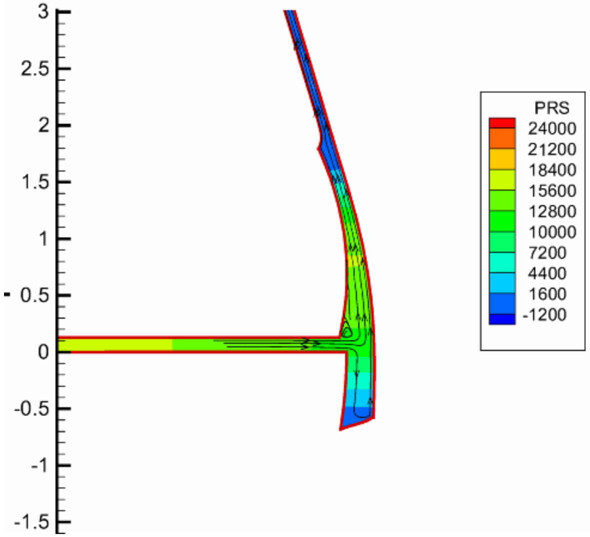
Computer-aided theory for flow prediction

Prediction of failure mechanisms and coating window

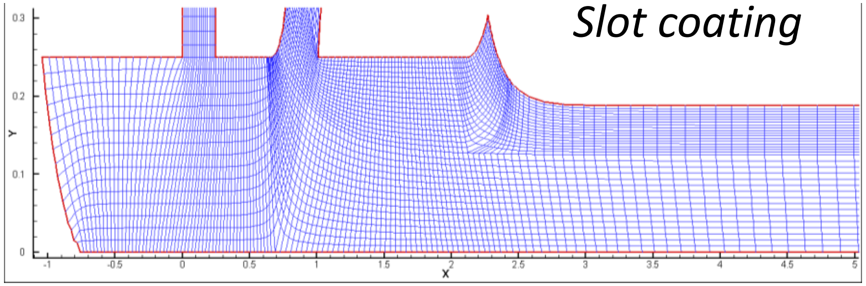


Roll coating

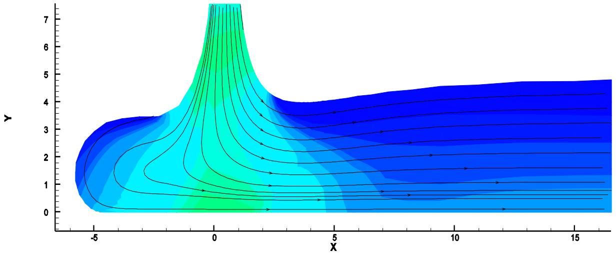
Tensioned Web coating



Slot coating



Curtain coating

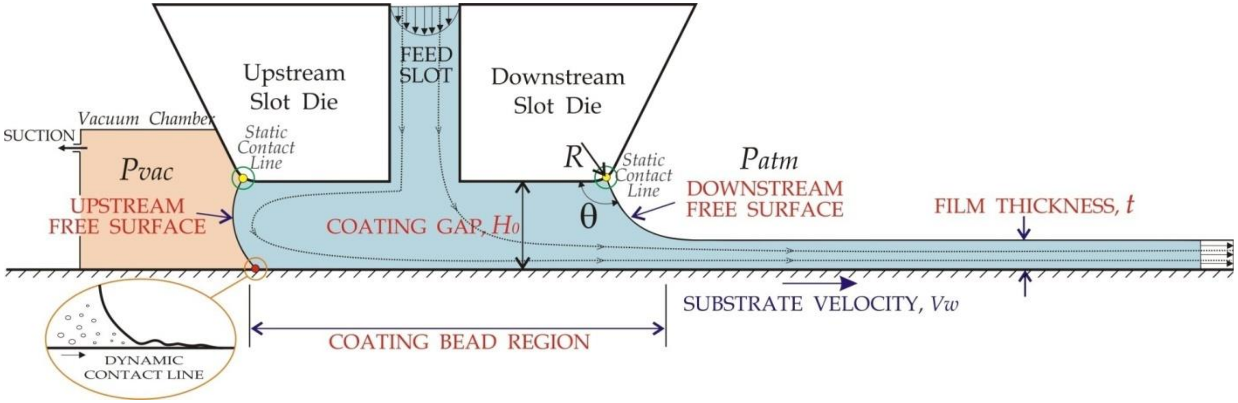


Slot Coating Process – Fundamentals

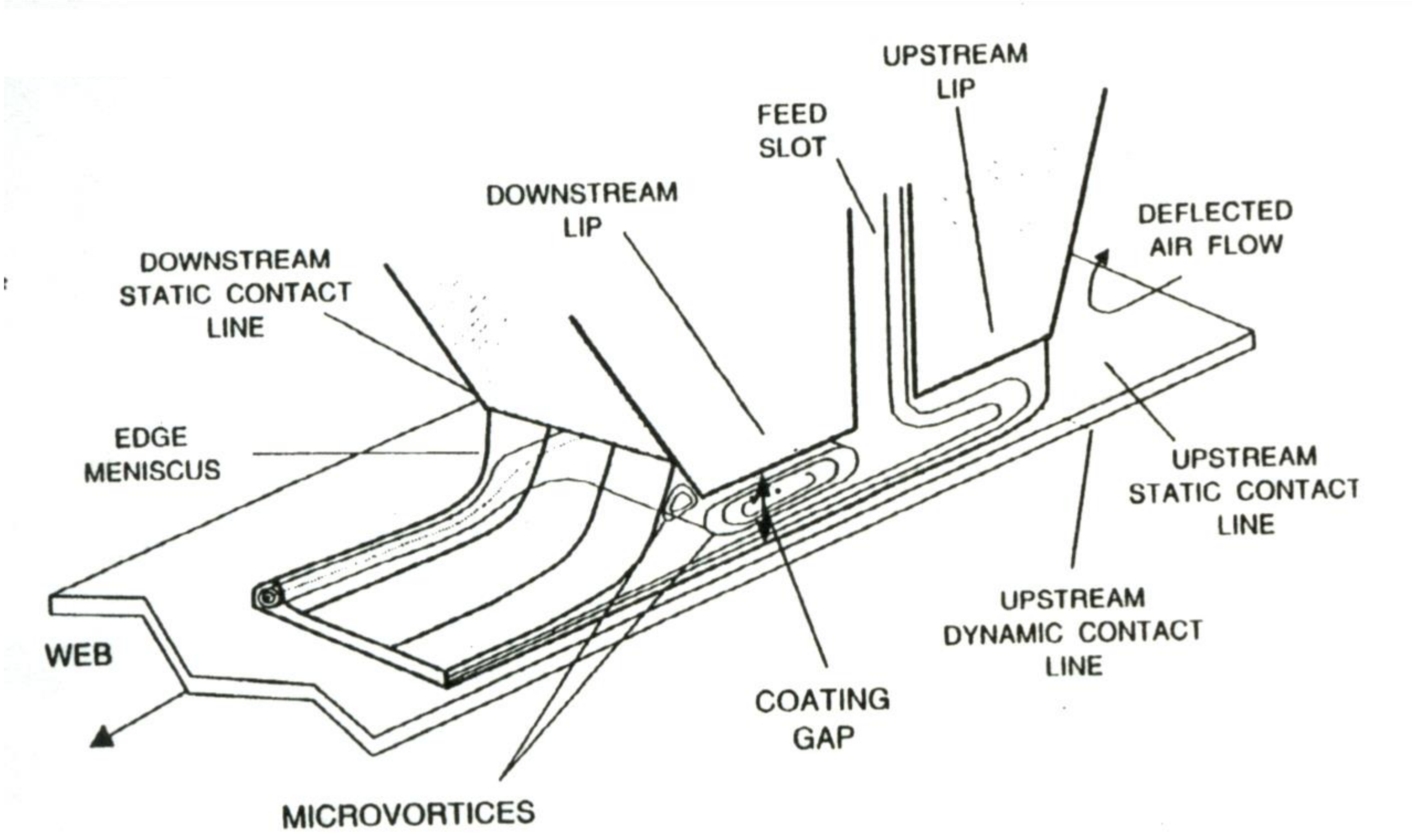
- SLOT COATING IS USED IN THE MANUFACTURING PROCESS OF MANY DIFFERENT PRODUCTS

- PRE-METERED METHOD: THICKNESS IS SET BY FLOW RATE

- FLOW UNIFORMITY IN THE COATING BEAD IS STRONGLY AFFECTED BY OPERATING PARAMETERS



FUNDAMENTAL ASPECTS OF SLOT COATING



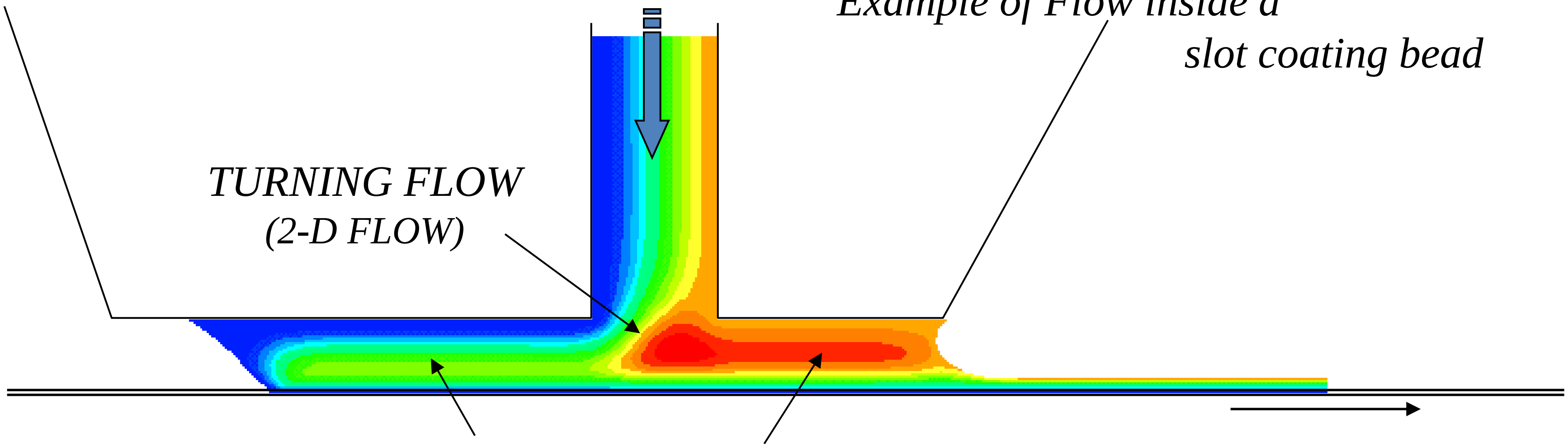
OPERATING CONDITIONS

GAP
FLOW RATE (OR FILM THICKNESS)
WEB SPEED
DIE CONFIGURATION (GEOMETRY)
LIQUID PROPERTIES

SUCH THAT, FLOW IS

TWO-DIMENSIONAL
STEADY STATE
STABLE TO SMALL DISTURBANCES

FORCES ARE THE KEY TO UNDERSTAND FLOW CONDITIONS



Example of Flow inside a slot coating bead

TURNING FLOW
(2-D FLOW)

ALMOST RECTILILNEAR FLOW
(DRAG + PRESSURE GRADIENT)

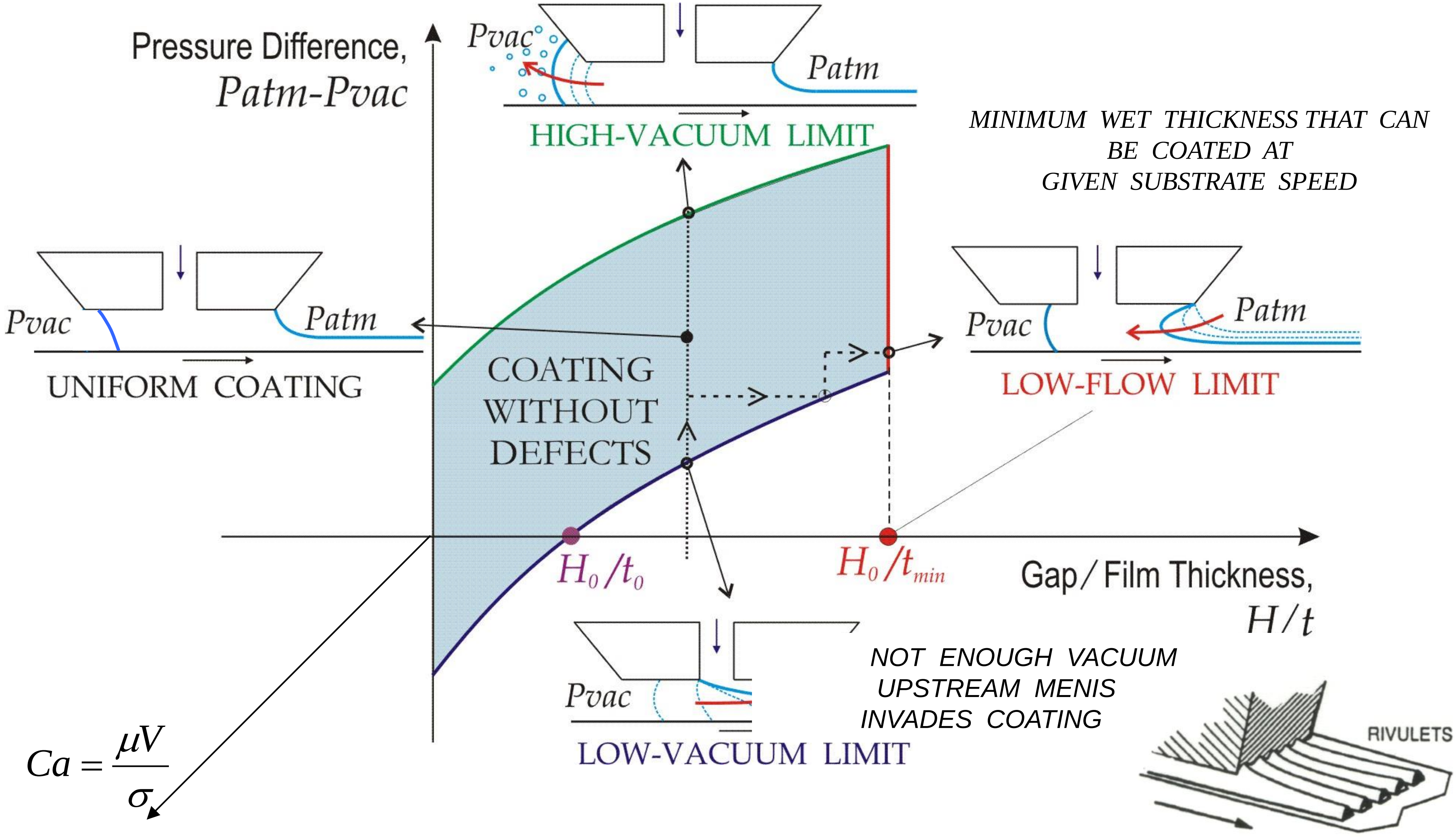
PRESSURE, VISCOUS, SURFACE TENSION AND INERTIAL FORCES MUST BALANCE TO PERMIT STEADY, 2-D FLOW

IF FORCES ARE NOT IN BALANCE, COATING BEAD WILL BREAK INTO A 3-D FLOW (RIVULETS, RIBBING,…) OR TRANSIENT FLOW

CONCEPT OF COATING WINDOW

THE COATING WINDOW IS BORDERED BY DEFECTS

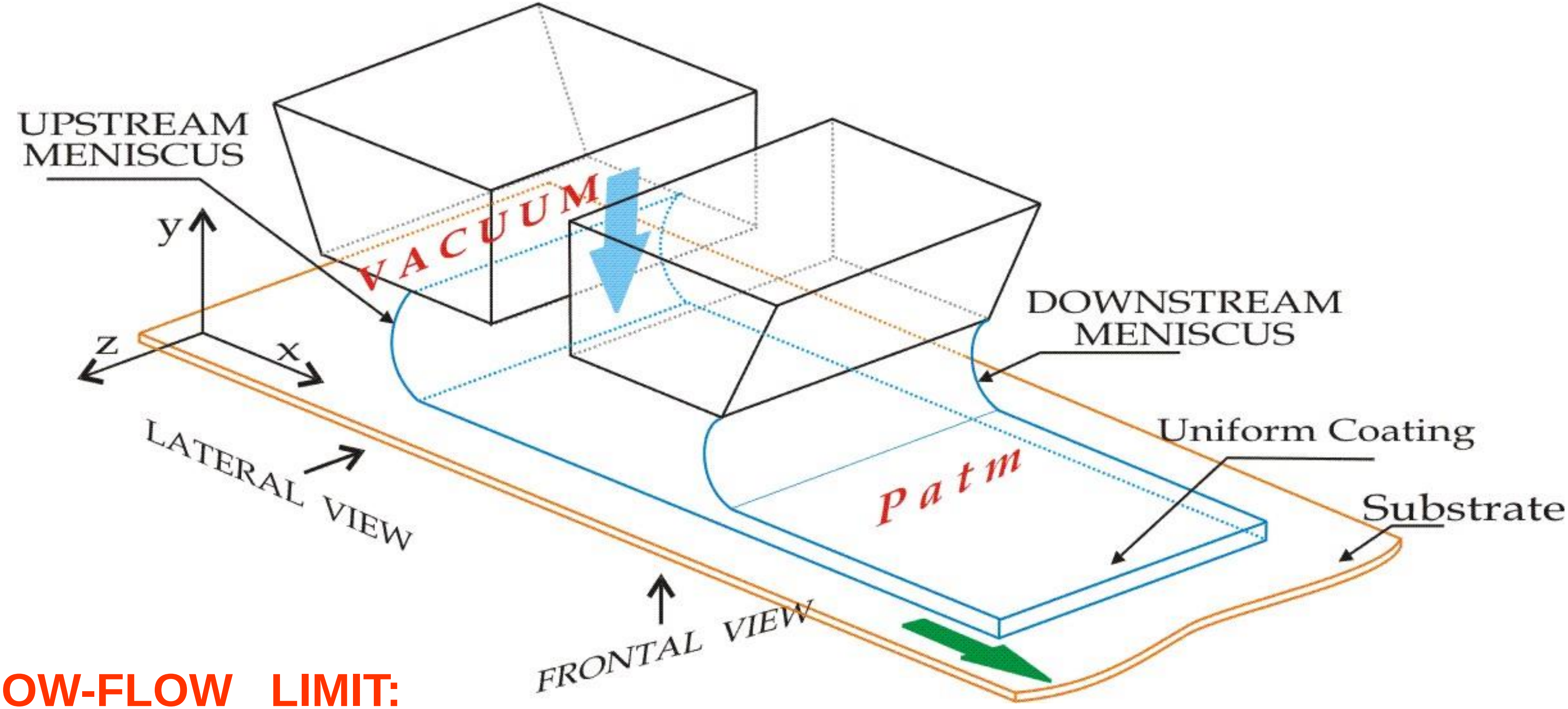
UPSTREAM PRESSURE TOO LOW
LIQUID INVADES VACUUM CHAMBER
PREMETERED ACTION IS LOST



MINIMUM WET THICKNESS THAT CAN
BE COATED AT
GIVEN SUBSTRATE SPEED

NOT ENOUGH VACUUM
UPSTREAM MENIS
INVADES COATING

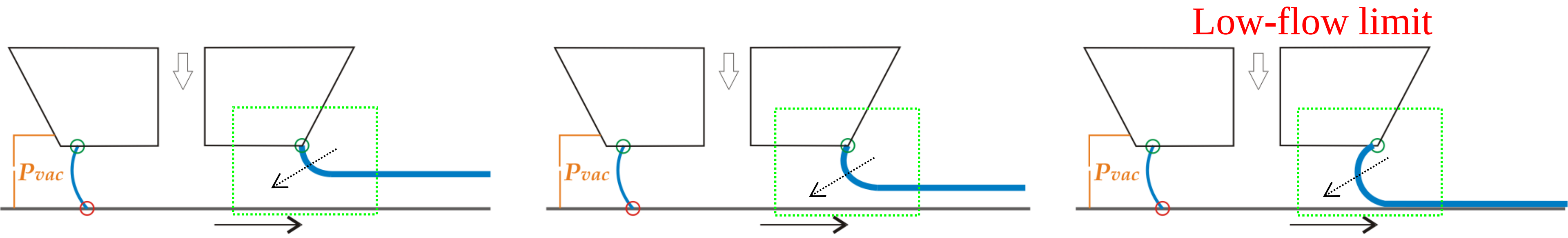
$$Ca = \frac{\mu V}{\sigma}$$

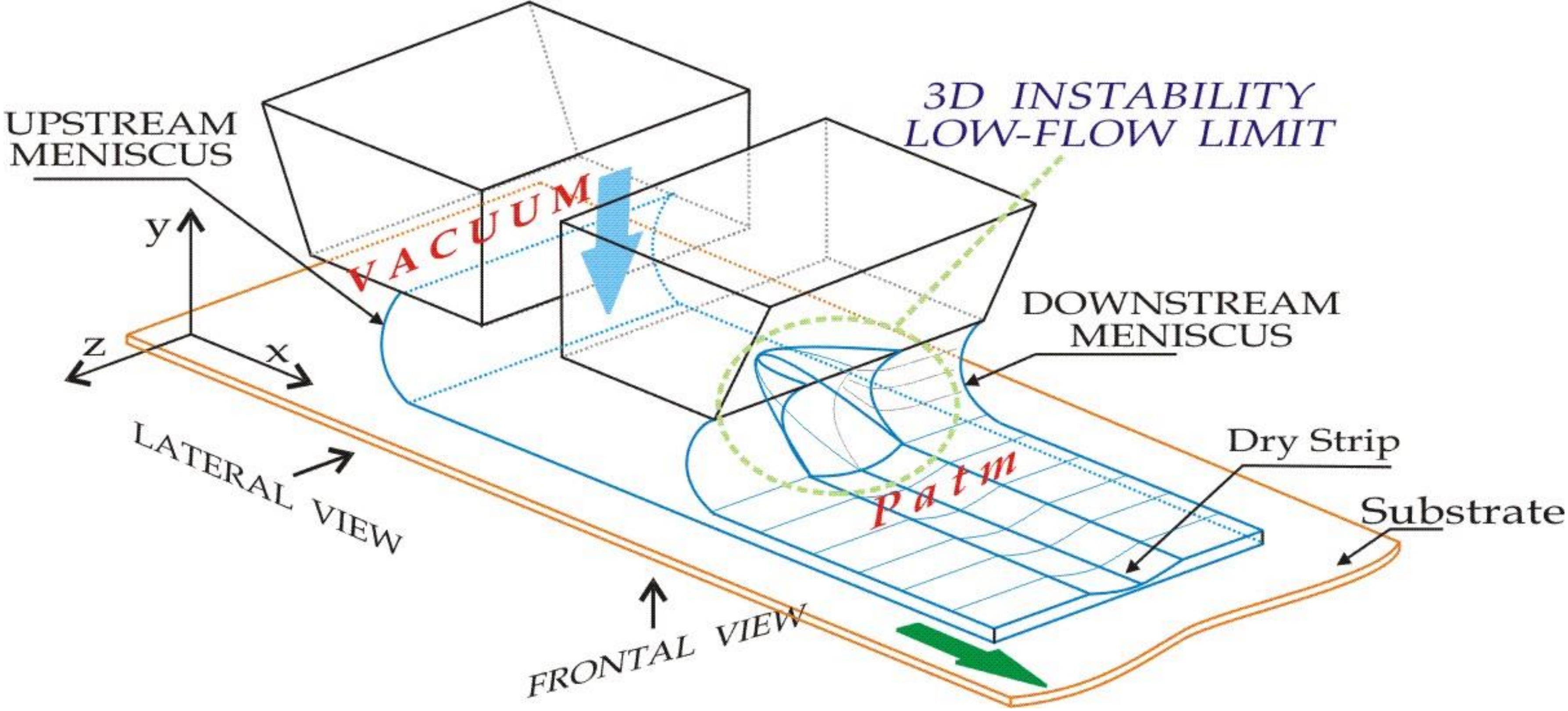


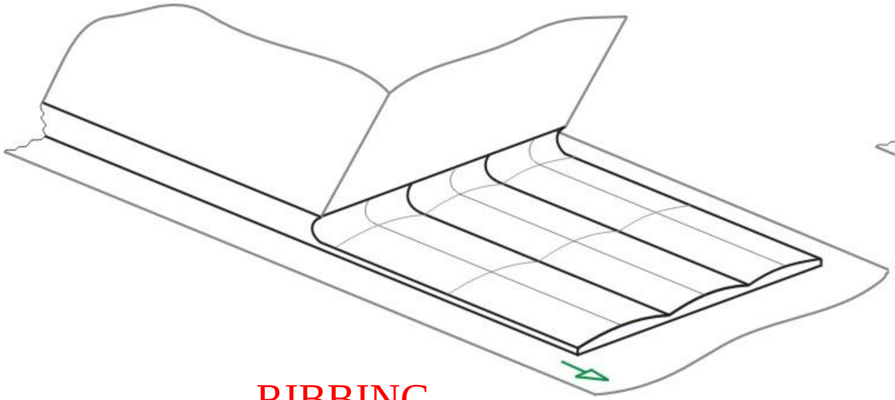
LOW-FLOW LIMIT:

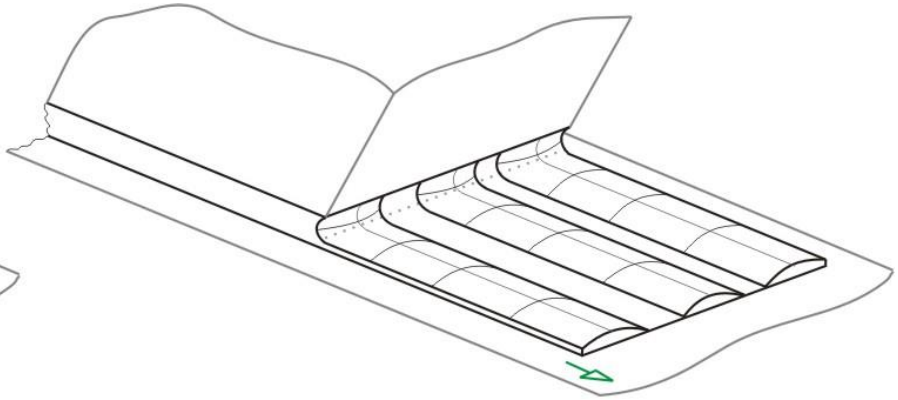
MINIMUM COATING THICKNESS AT A GIVEN SUBSTRATE SPEED ;

MAXIMUM SUBSTRATE SPEED AT A GIVEN COATING THICKNESS .

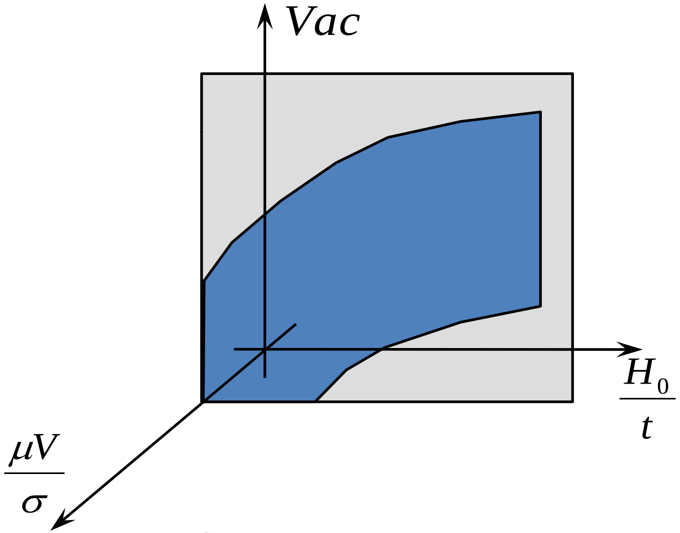




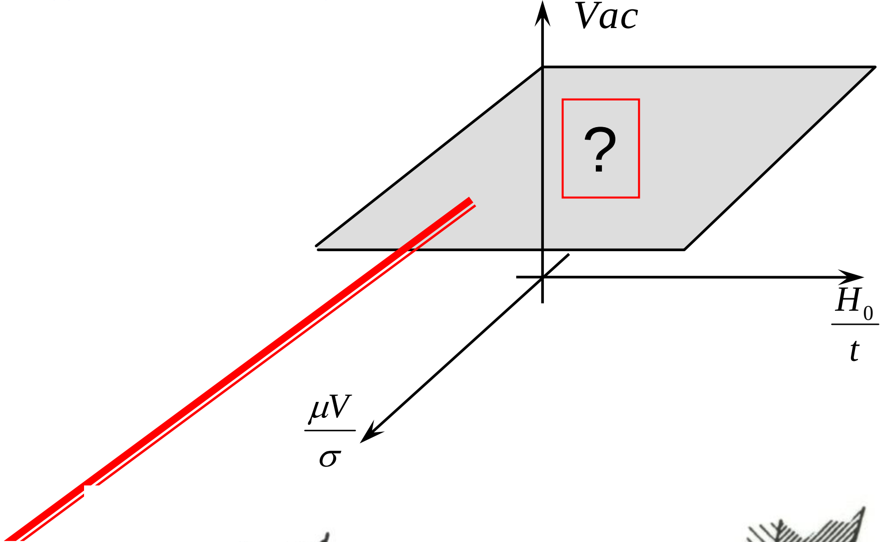

RIBBING

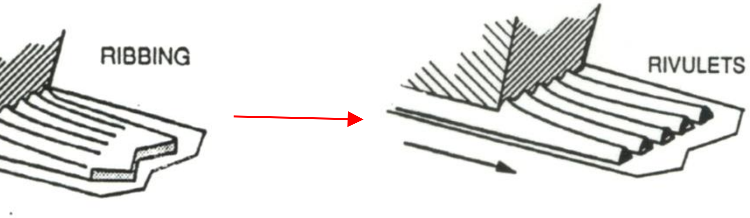

RIVULETS

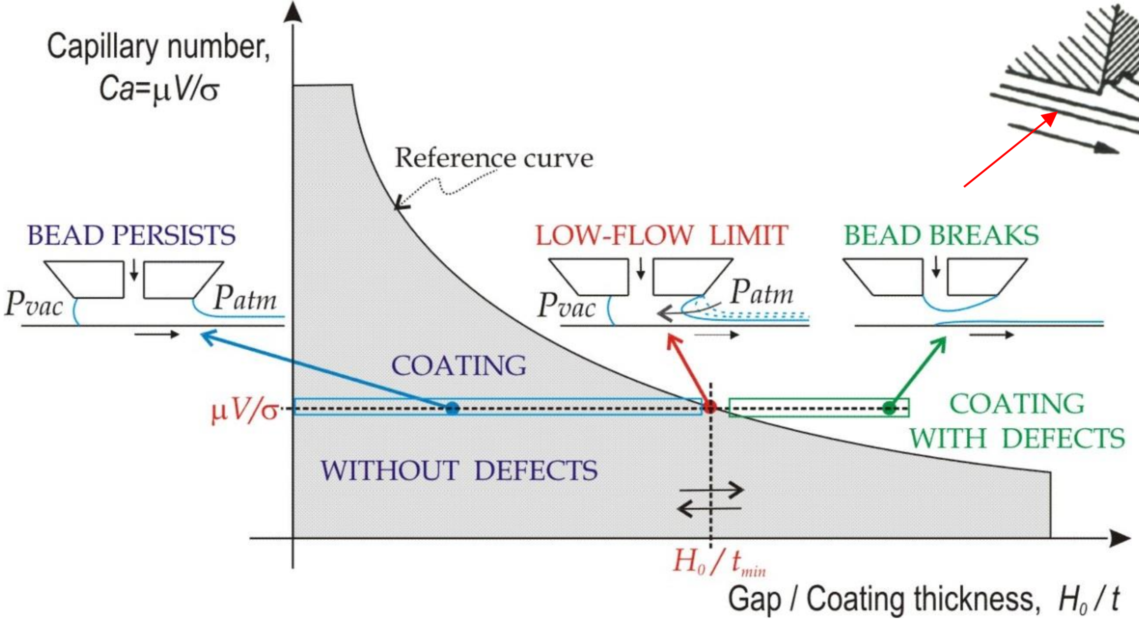
COATING WINDOW IN PLANE OF VACUUM VS THICKNESS



COATING WINDOW IN PLANE OF WEB SPEED VS THICKNESS



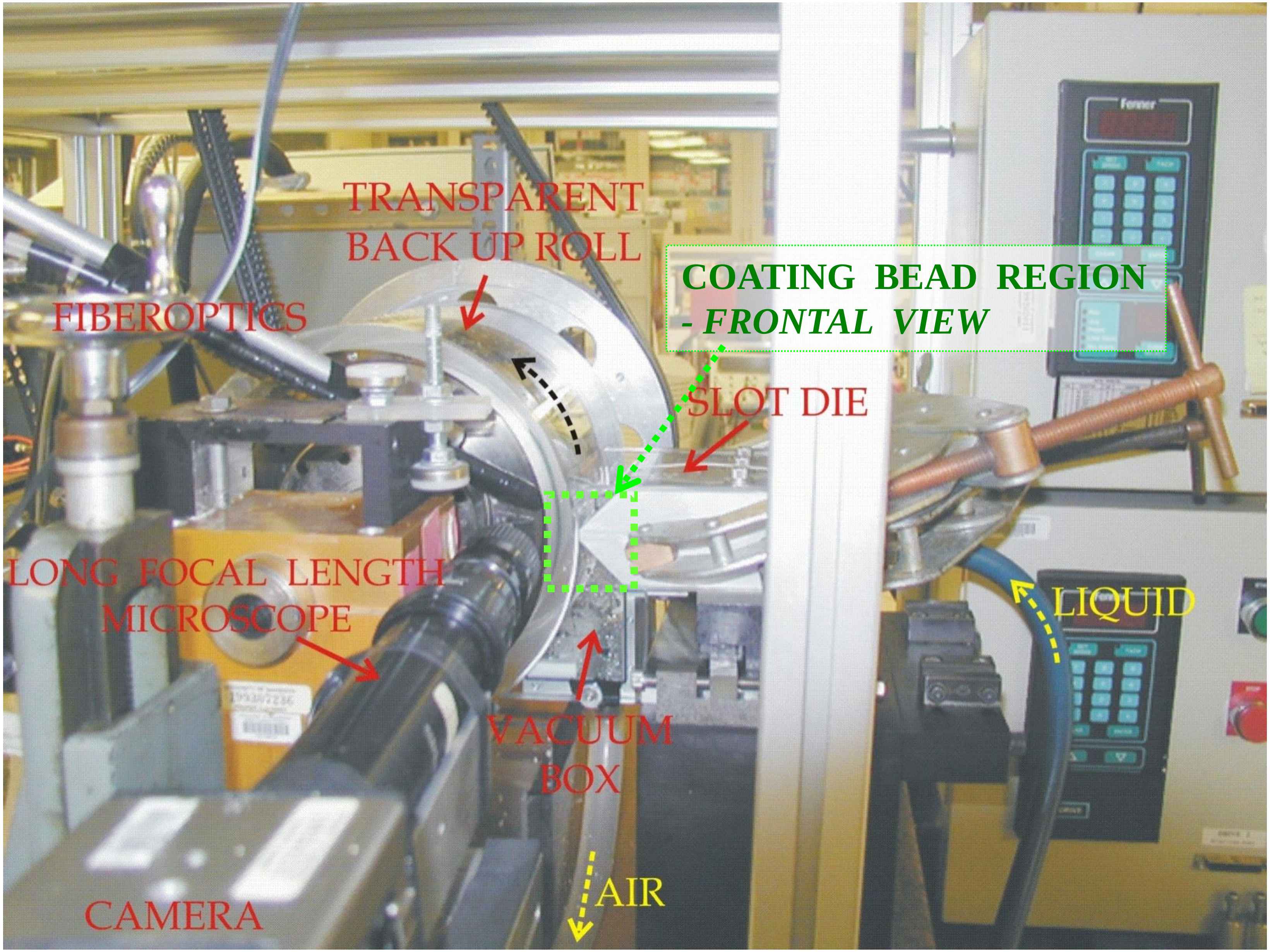


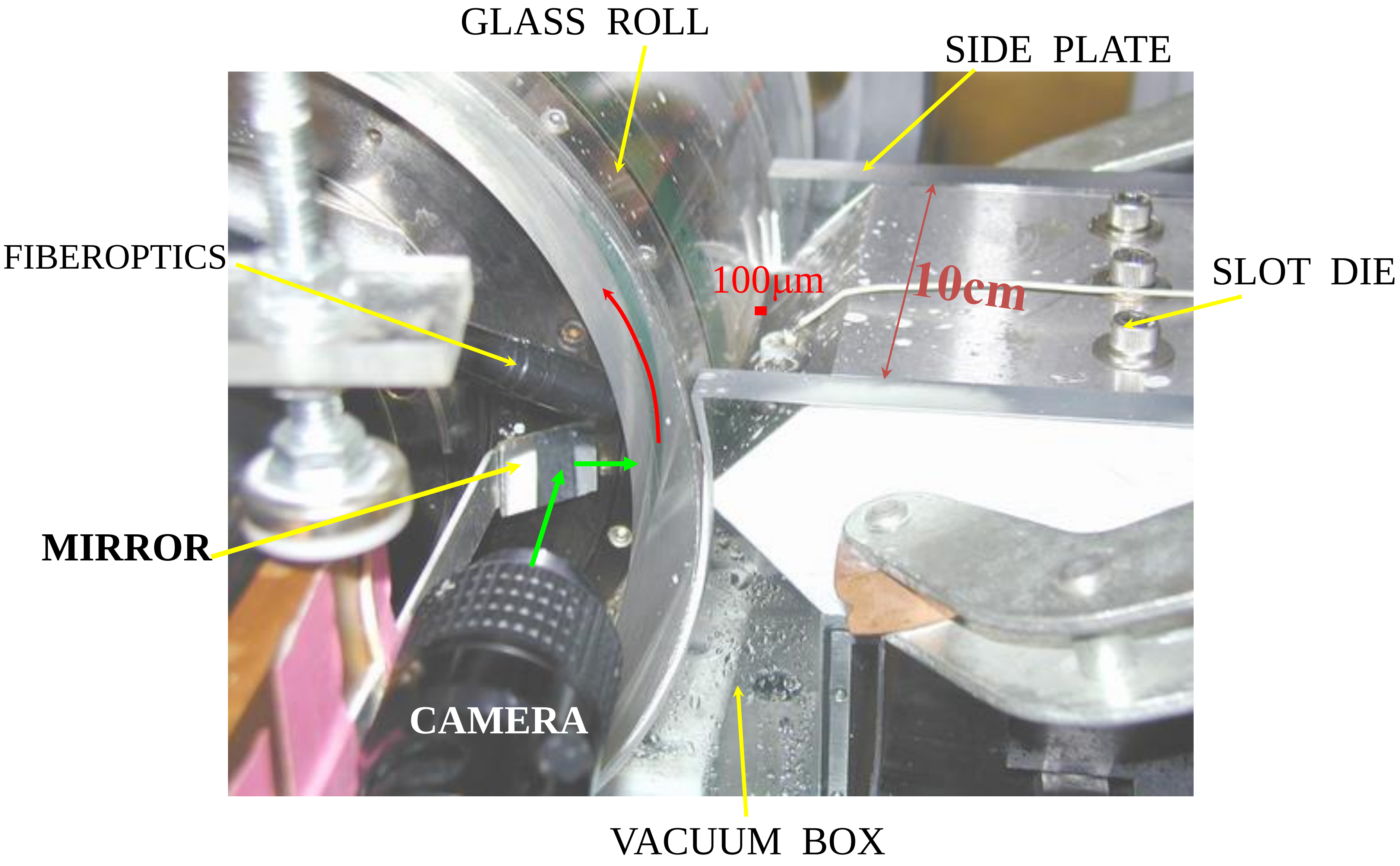


THE FASTER THE WEB SPEED,
THE LARGER IS THE
MINIMUM WET THICKNESS

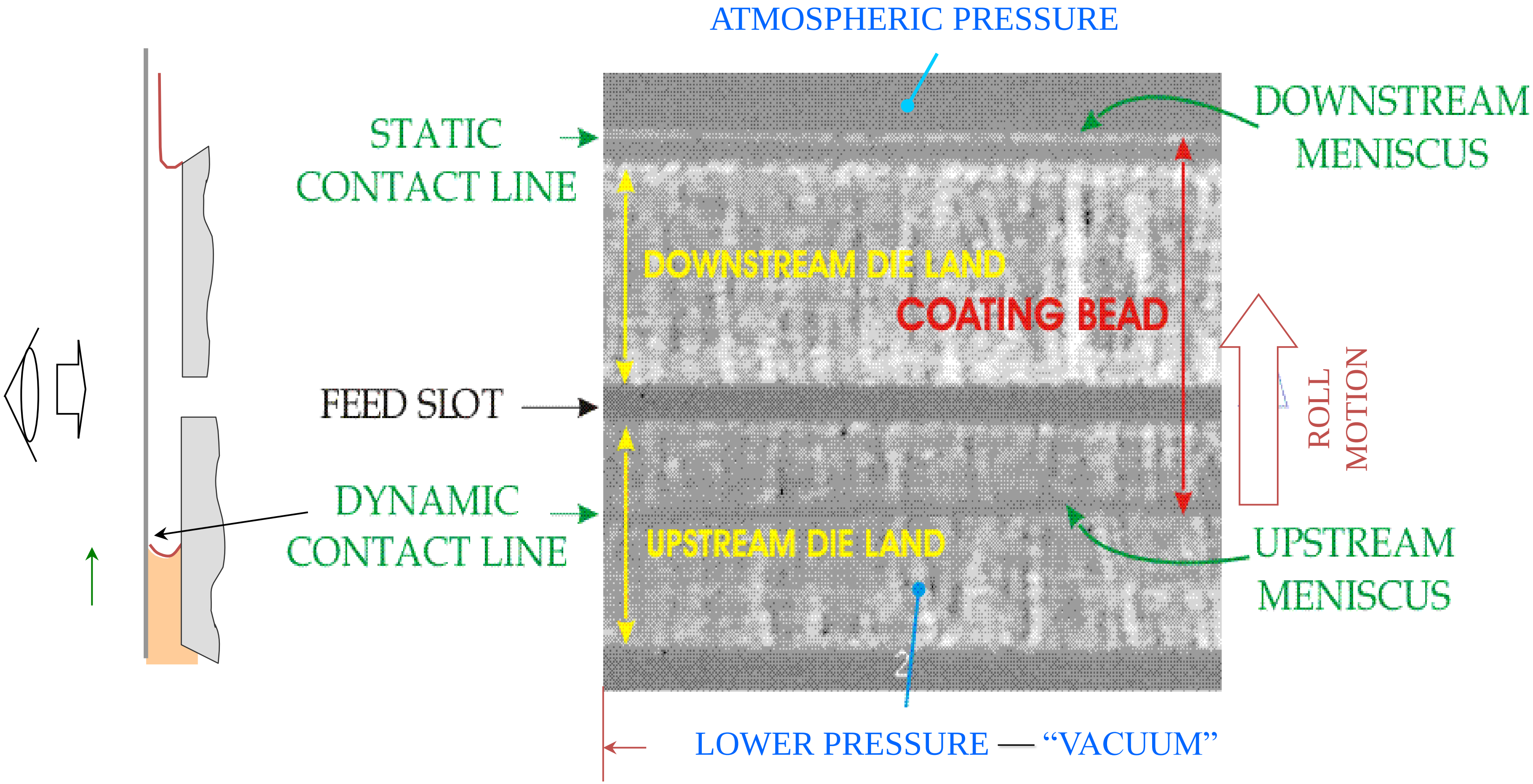
VISUALIZING THE DIFFERENT MECHANISMS OF BEAD BREAKUP

Romero, Scriven and Carvalho (JNNFM, 2004)




GLASS ROLL
SIDE PLATE
FIBEROPTICS
100µm
10cm
SLOT DIE
MIRROR
CAMERA
VACUUM BOX

FLOW VIEWED THROUGH GLASS ROLL



ATMOSPHERIC PRESSURE

STATIC CONTACT LINE

DOWNSTREAM MENISCUS

DOWNSTREAM DIE LAND

COATING BEAD

FEED SLOT

ROLL MOTION

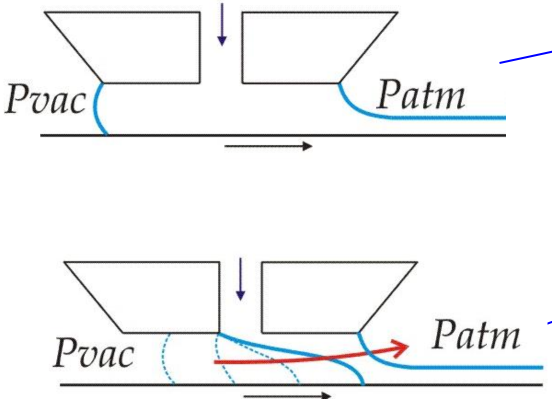
DYNAMIC CONTACT LINE

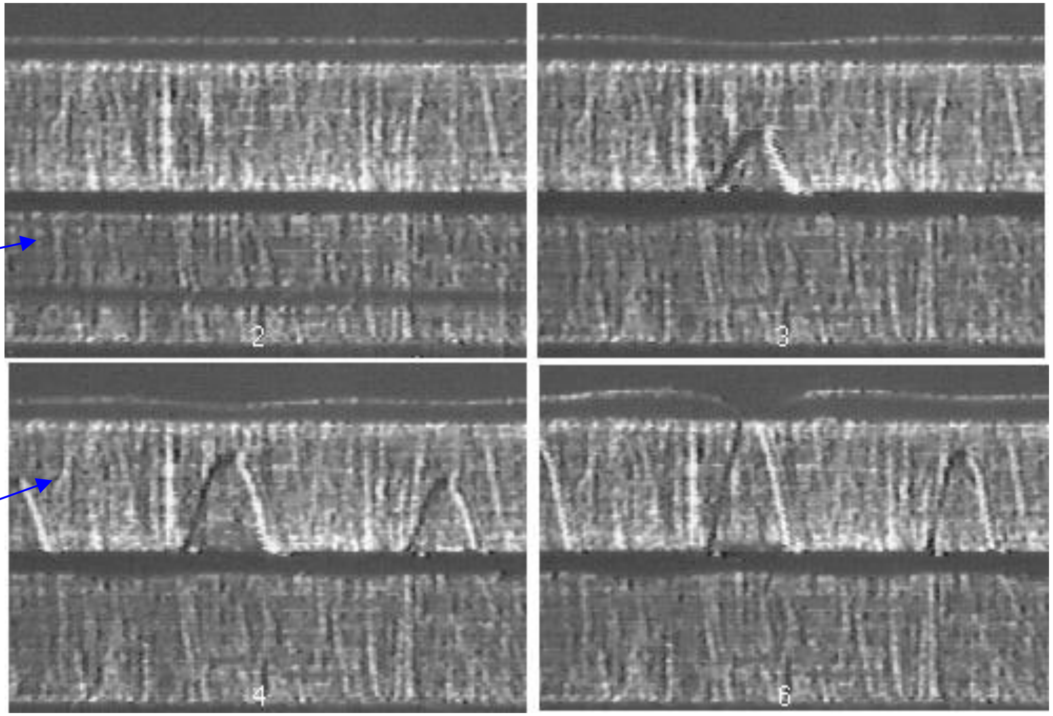
UPSTREAM DIE LAND

UPSTREAM MENISCUS

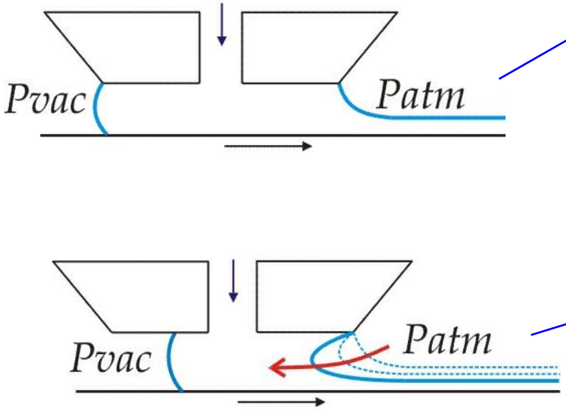
LOWER PRESSURE — “VACUUM”

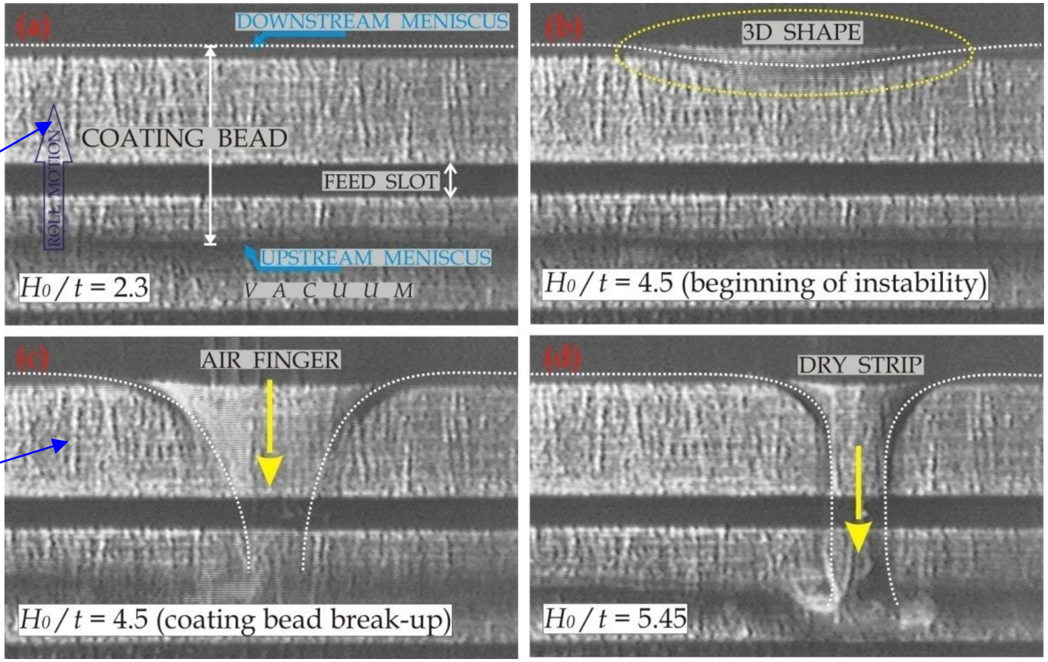
INVASION OF THE UPSTREAM MENISCUS



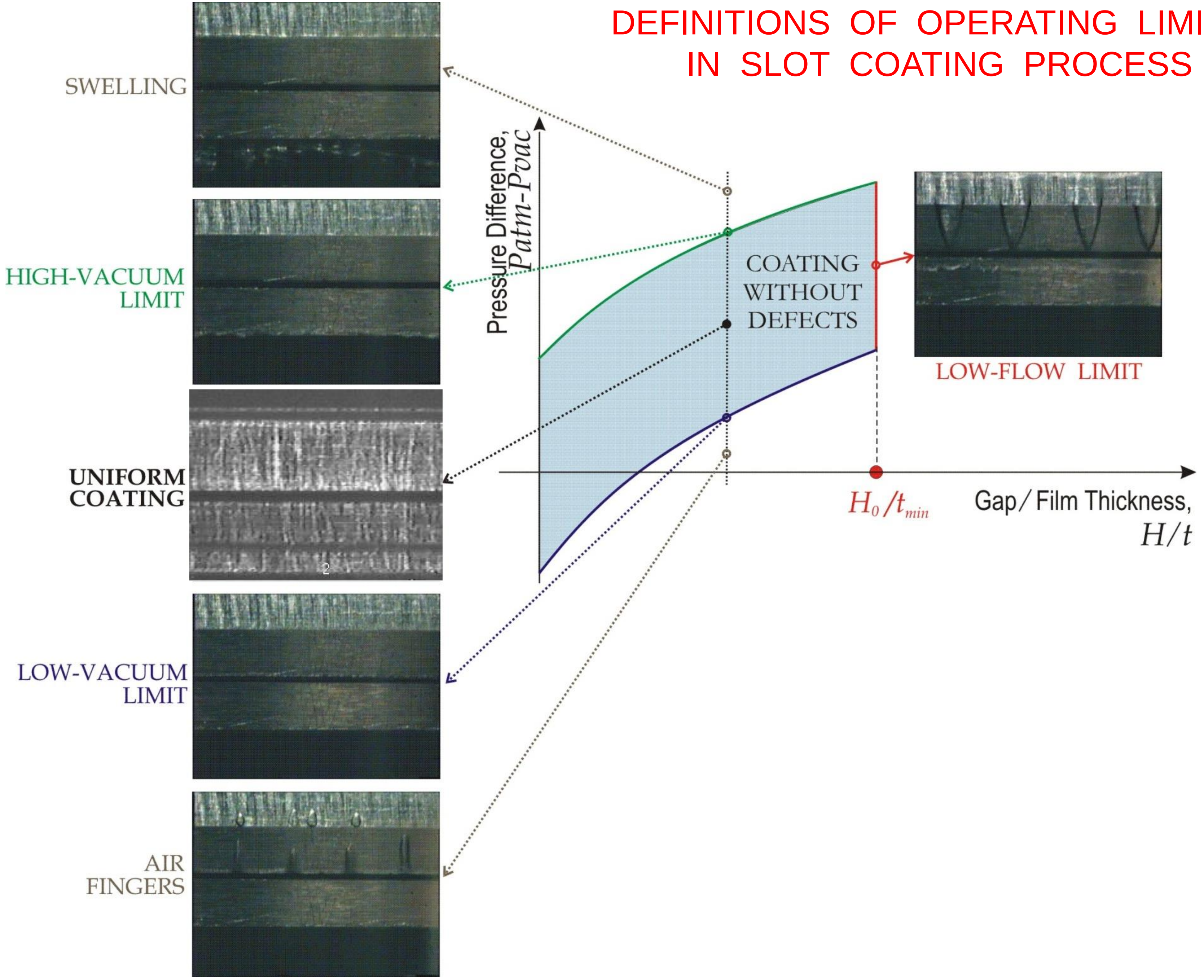


INVASION OF THE DOWNSTREAM MENISCUS

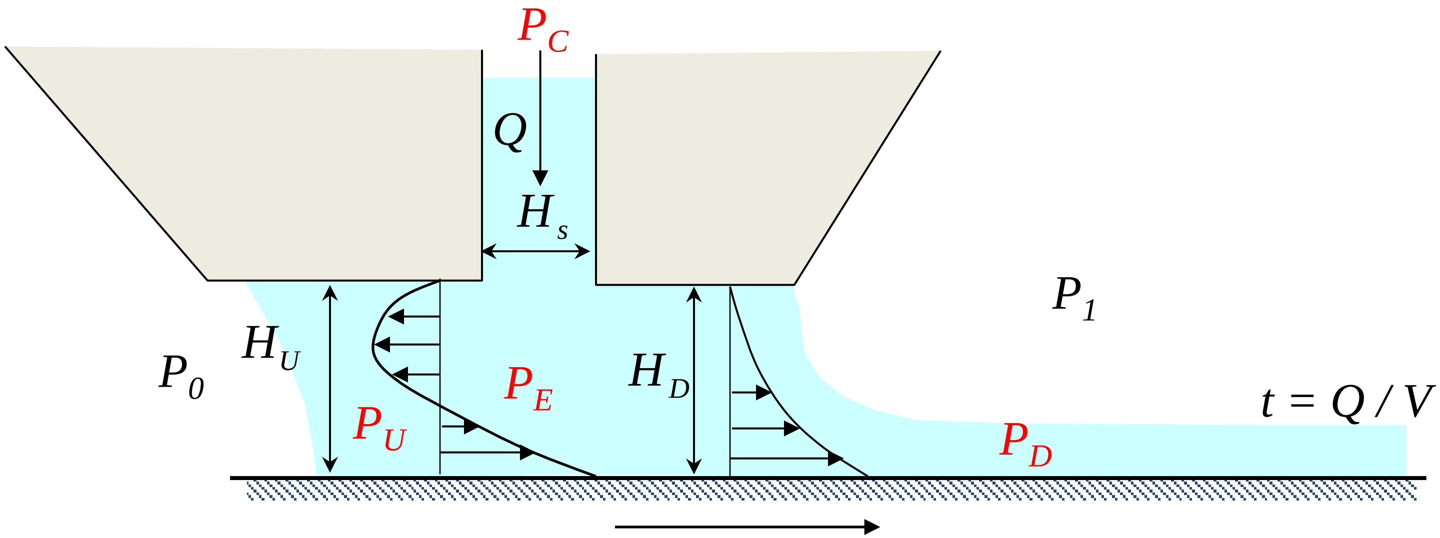




DEFINITIONS OF OPERATING LIMITS IN SLOT COATING PROCESS



LUBRICATION APPROXIMATION MODEL – Rectilinear Flow



FLOW IN FEED SLOT

$$Q = \frac{H_s^3}{12\mu}\frac{P_C - P_E}{L_S}$$

FLOW UPSTREAM

$$Q_U = 0 = \underbrace{\frac{H_U^3}{12\mu}\frac{P_U - P_E}{L_U}}_{<0} + \frac{VH_U}{2}$$

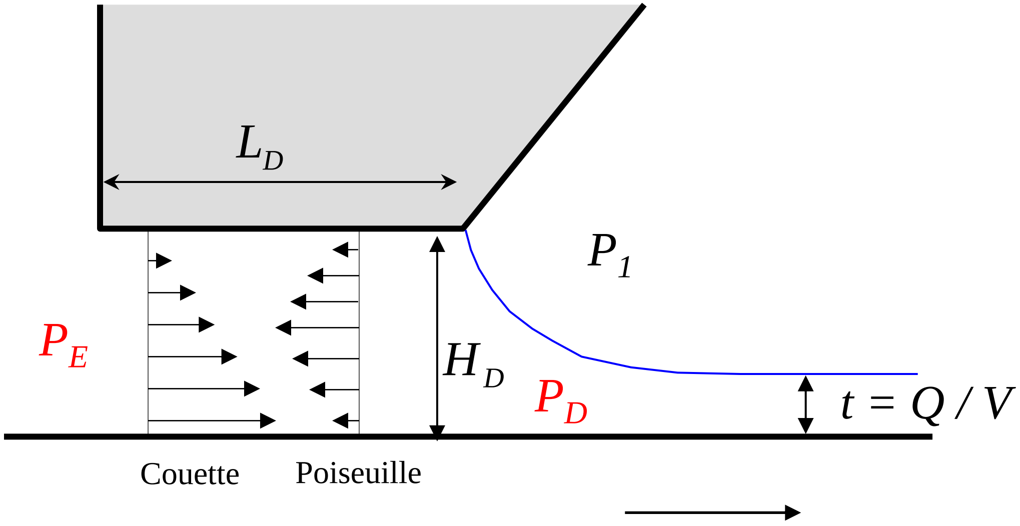
$$<0 \quad \Rightarrow \quad P_U \approx P_0 < P_E$$

VISCOUS FLOW IN SLOT COATING – *CONT.*

FLOW DOWNSTREAM

$$Q = Vt = \frac{H_D^3}{12\mu}\frac{P_E - P_D}{L_D} + \frac{VH_D}{2}$$

$$\Rightarrow \quad t = \frac{H_D}{2} + \frac{H_D^3}{12\mu V}\frac{P_E - P_D}{L_D}$$



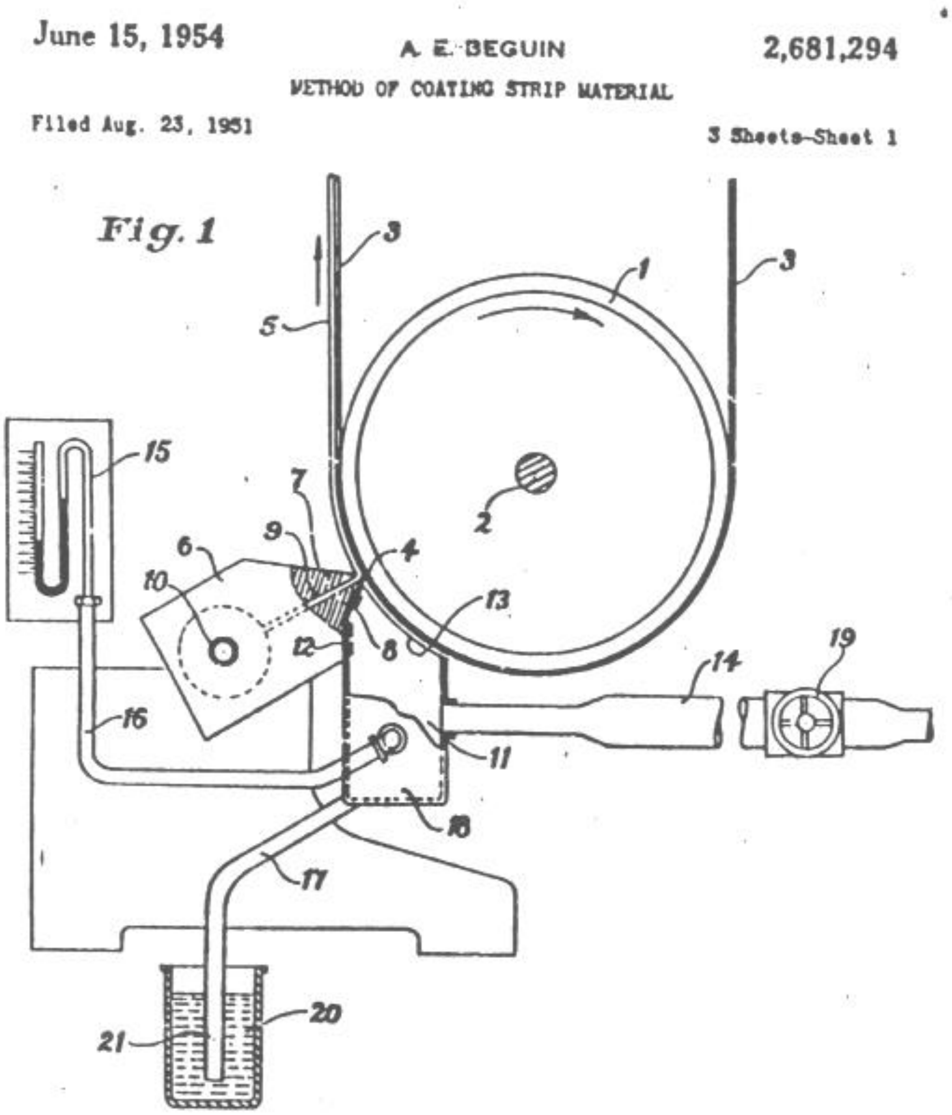
IF $\boxed{t > \frac{H_D}{2} \quad \Rightarrow \quad P_E > P_D \;\; (\text{for} \;\; L_D > 0)}$

IF $\boxed{t < \frac{H_D}{2} \quad \Rightarrow \quad P_E < P_D \approx P_1 \;\; (\text{for} \;\; L_D > 0)}$ $\Rightarrow \quad P_0 \approx P_U < P_E < P_D \approx P_1$

IF $P_1 = P_{amb} \quad \Rightarrow \boxed{P_0 < P_{amb}}$

VACUUM IS NEEDED, OTHERWISE FLOW BREAKS INTO RIVULETS

IMPROVING BEAD STABILITY BY VACUUM APPLICATION
(BEGUIN, 1954, US PATENT 2,681,294)



June 15, 1954 A. E. BEGUIN 2,681,294

METHOD OF COATING STRIP MATERIAL

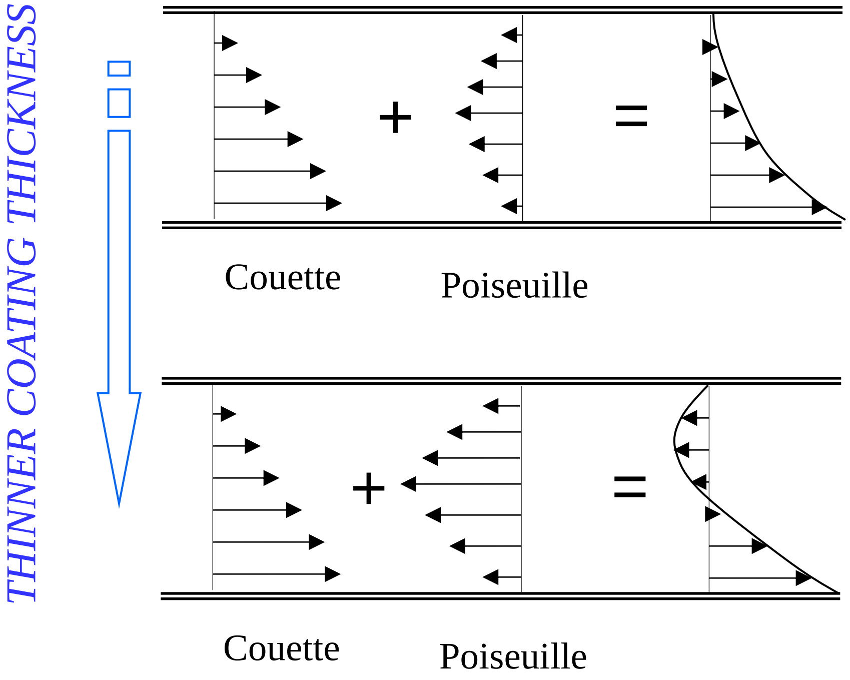
Filed Aug. 23, 1951 3 Sheets-Sheet 1

Fig. 1

FLOW DOWNSTREAM – *CONT.*

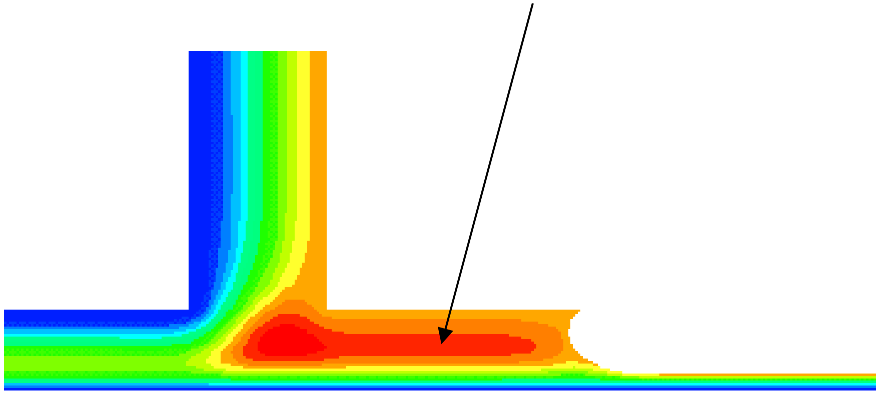
FLOW IS A COMBINATION OF DRAG (*COUETTE*) AND PRESSURE DRIVEN (*POISEUILLE*)

THE THINNER THE COATING THICKNESS, THE STRONGER THE POISEUILLE CONTRIBUTION

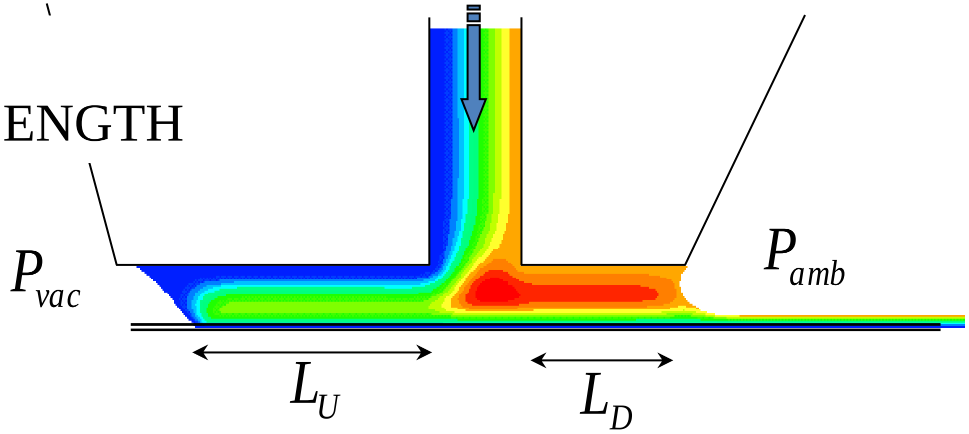


IF $t < \dfrac{H_D}{3}$

RECIRCULATION APPEARS UNDER DOWNSTREAM DIE LIP



RELATION BETWEEN VACUUM AND BEAD LENGTH



$$Vac = P_{amb} - P_{vac} = \frac{12\mu V L_D}{H_D^3}\left(\frac{H_D}{2} - t\right) + \frac{6\mu V L_U}{H_U^3} H_D$$

THE GREATER THE VACUUM, THE GREATER THE UPSTREAM BEAD LENGTH

IN THE LIMIT OF NO UPSTREAM COATING BEAD

MINIMUM VACUUM NEEDED FOR GIVEN COATING THICKNESS t

$$Vac_{\min} = P_{amb} - P_{vac} = \frac{12\mu V L_D}{H_D^3}\left(\frac{H_D}{2} - t\right)$$

THIS REGIME IS UNSTABLE – IMPOSSIBLE TO MAINTAIN AS STEADY, TWO-DIMENSIONAL FLOW

2-D FLOW BREAKS INTO PARALLEL RIVULETS ON THE WEB

THE GREATER THE UPSTREAM BEAD LENGTH, THE GREATER THE STABILITY AGAINST RIVULET FLOW, AND THE GREATER THE ABILITY OF THE COATING BEAD TO ACCOMMODATE FLUCTUATIONS.

CONCERNS WITH RECIRCULATION, IF PRESENT.

COATING DIES HAVE A FIXED UPSTREAM LIP LENGTH

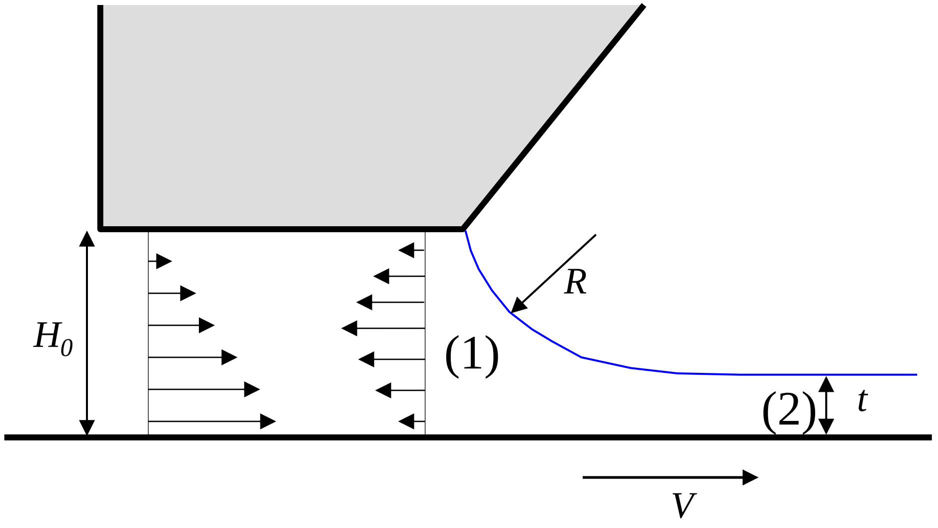
THERE IS A MAXIMUM VACUUM THAT CAN BE APPLIED BEFORE THE UPSTREAM BEAD BECOMES TOO LONG AND INVADES THE VACUUM BOX – *WEEPING*

PREMETERING IS LOST

THE RANGE OF VACUUM OVER WHICH SLOT COATER CAN BE OPERATED GIVES DEFINES THE COATING WINDOW

COATING WINDOW – *LOW FLOW LIMIT*

BASIC MECHANISM WELL DESCRIBED BY VISCOCAPILLARY MODEL



$$\mathbf{Q = V\,t = Q_{couette} - Q_{Poiseuille}}$$

$$Q_{couette} = V\,H_0 / 2$$

$$Q_{poiseuille} \propto P_2 - P_1 = \sigma / R$$

AT A FIXED WEB SPEED, MINIMUM THICKNESS OCCURS WHEN $Q_{poiseuille}$ IS MAXIMUM ➔ *R* IS MINIMUM

$$R_{\min} = \frac{H_0 - t}{2} \quad ➔ \quad (P_2 - P_1)_{\max} = \frac{2\sigma}{H_0 - t}$$

FROM FILM-FLOW EQUATION $P_2 - P_1 = 1.34 Ca^{2/3} \dfrac{\sigma}{t}$

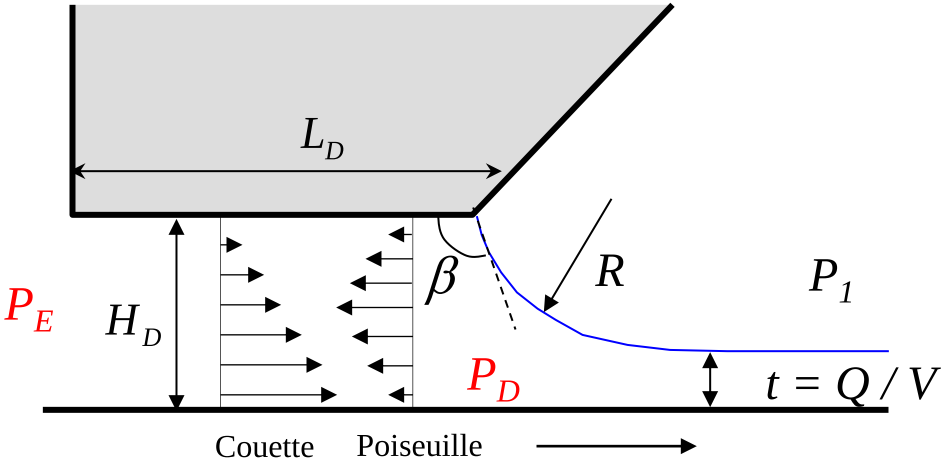
AT THE ONSET OF LOW-FLOW LIMIT $Ca \equiv \dfrac{\mu V}{\sigma} = 0.65 \left(\dfrac{2}{H_0/t - 1} \right)^{\frac{3}{2}}$

LUBRICATION MODEL CAN BE USED TO PREDICT THE RANGE OF OPERABILITY FOR DIFFERENT PARAMETERS

FLOW NEAR DOWNSTREAM FREE SURFACE

Landau-Levich eq.

$$\boxed{P_1 - P_D = -P_D = 1.34\left(\frac{\mu V}{\sigma}\right)^{2/3}\frac{\sigma}{t}} \quad (1)$$



Young-Laplace eq.

$$\boxed{P_1 - P_D = -P_D = \frac{\sigma}{R}} \quad (2)$$

Geometric relation (meniscus is an arc of circle).

$$\boxed{\beta = \arccos\left(\frac{H_D - t}{R} - 1\right)} \quad (3)$$

Flow under downstream die lip

$$Q = Vt = \frac{H_D^3}{12\mu}\frac{P_E - P_D}{L_D} + \frac{VH_D}{2} \quad \Rightarrow \quad \boxed{P_E = P_D + \frac{12\mu V L_D}{H_D^3}\left[t - \frac{H_D}{2}\right]} \quad (4)$$

FLOW NEAR UPSTREAM FREE SURFACE

Flow under upstream die lip

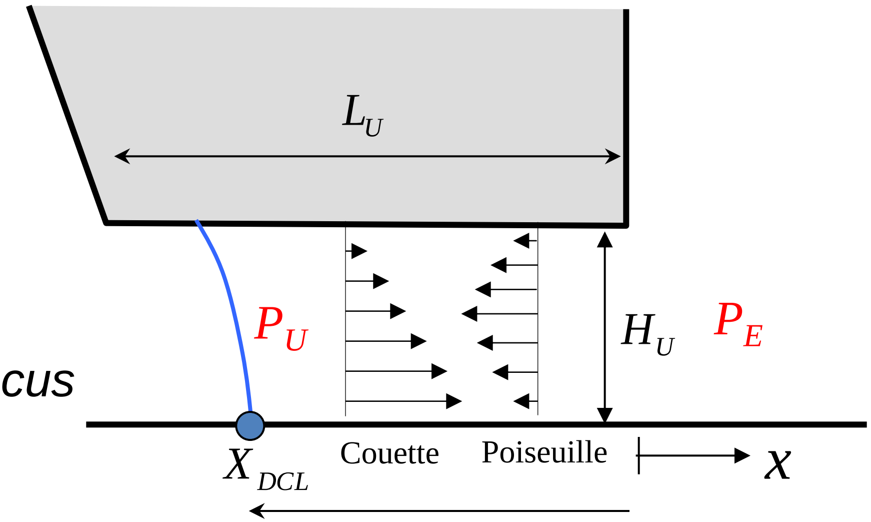
$$Q_U = 0 = \frac{H_U^3}{12\mu}\frac{P_U - P_E}{(-X_{DCL})} + \frac{VH_U}{2}$$

↘ x_{DCL} *is < 0*

Neglect capillary effect on upstream meniscus

$$P_U \approx P_0 = P_{VAC}$$

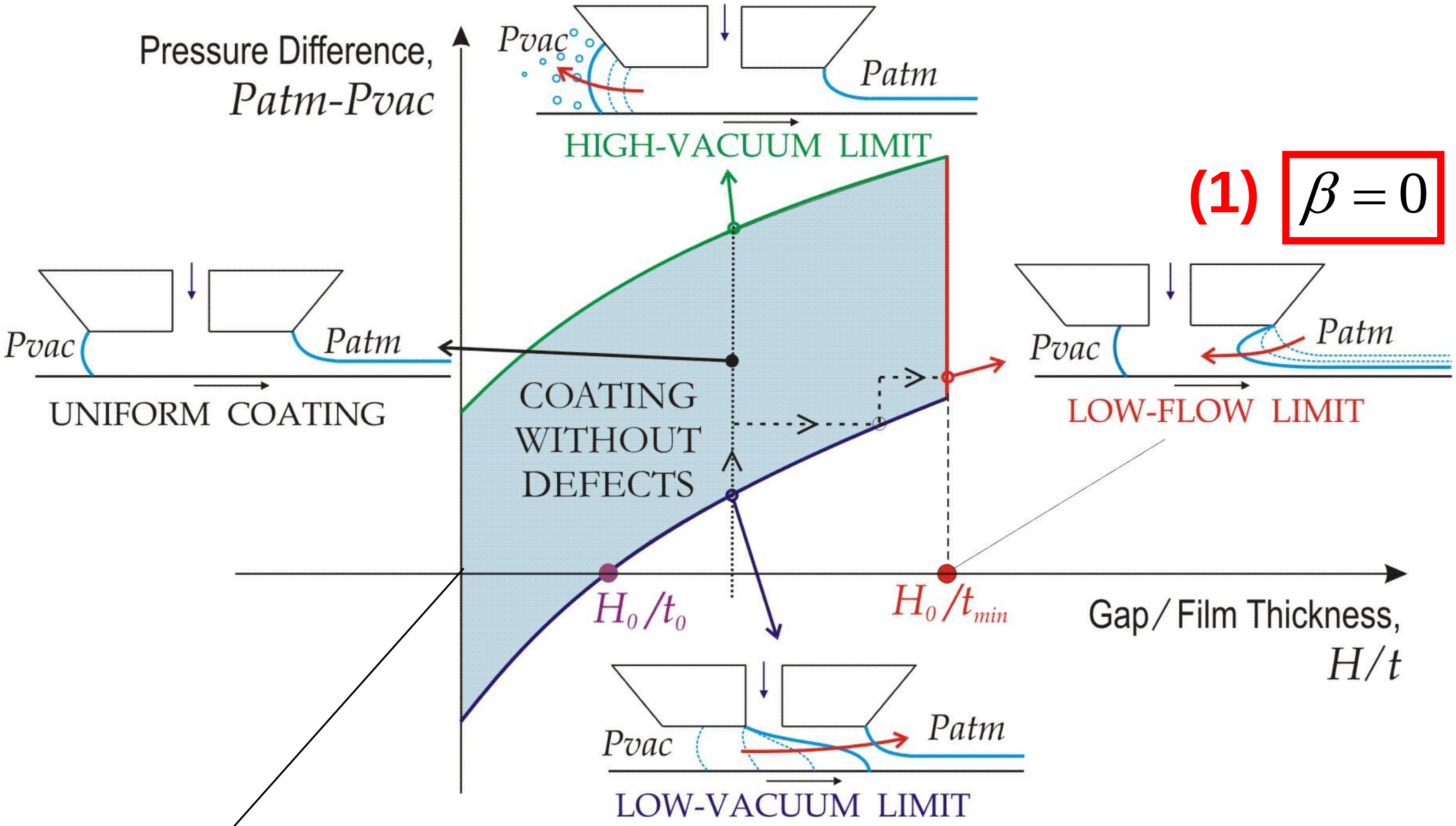
$$\boxed{X_{DCL} = \frac{P_E - P_{VAC}}{6\mu V} H_U^2} \quad (5)$$



FAILURE MECHANISMS

(3) $X_{DCL} < -L_U$

(1) $\beta = 0$



(2) $X_{DCL} > 0$

Capillary number $\frac{\mu V}{\sigma}$

Free variable: FILM THICKNESS t

Maximum film thickness

Failure mechanism **(3)** : Bead invades the vacuum box

$$X_{DCL} = -L_U \quad \xrightarrow[\text{Eq. (5)}]{} \quad P_E = P_{VAC} + \frac{6\mu V L_U}{H_U^2}$$

$$\xrightarrow[\text{Eq. (4)}]{} \quad P_D = P_{VAC} + \frac{6\mu V L_U}{H_U^2} - \frac{12\mu V L_D}{H_D^3}\left[t - \frac{H_D}{2}\right]$$

$$\xrightarrow[\text{Eq. (1)}]{} \quad P_D = -1.34\left(\frac{\mu V}{\sigma}\right)^{2/3}\frac{\sigma}{t}$$

$$\Bigg\} \Longrightarrow$$

$$\boxed{\frac{12\mu V L_D}{H_D^3}t - 1.34\left(\frac{\mu V}{\sigma}\right)^{2/3}\sigma\frac{1}{t} - \left[P_{VAC} + \frac{6\mu V L_U}{H_U^2} + \frac{6\mu V L_D}{H_D^2}\right] = 0} \qquad (6)$$

$$\boxed{t_{MAX} \quad \text{is the root of Eq.(6)}}$$

Minimum film thickness

Failure mechanism **(1)** : Downstream meniscus invades coating bead
Or
Failure mechanism **(2)** : Upstream meniscus invades coating bead

Failure mechanism **(1)**

$$\left.\begin{array}{l} \beta = 0 \xrightarrow[\text{Eq. (3)}]{} \left(\dfrac{H_D - t}{R} - 1\right) = 1 \Rightarrow R = \dfrac{H_D - t}{2} \\[2ex] \xrightarrow[\text{Eq. (1) and (2)}]{} 1.34\left(\dfrac{\mu V}{\sigma}\right)^{2/3}\dfrac{\sigma}{t} = \dfrac{\sigma}{R} \end{array}\right\} \Longrightarrow$$

$$\boxed{t_{MIN}^{(1)} = \frac{1.34\left(\frac{\mu V}{\sigma}\right)^{2/3} H_D}{2 + 1.34\left(\frac{\mu V}{\sigma}\right)^{2/3}}} \qquad (7)$$

Failure mechanism **(2)**

$$X_{DCL} = 0 \xrightarrow{\text{Eq. (5)}} P_E = P_{VAC}$$

$$\xrightarrow{\text{Eq. (4) and (2)}} P_{VAC} = -1.34\left(\frac{\mu V}{\sigma}\right)^{2/3}\frac{\sigma}{t} + \frac{12\mu V L_D}{H_D^3}\left[t - \frac{H_D}{2}\right] \longrightarrow$$

$$\boxed{\frac{12\mu V L_D}{H_D^3}t - 1.34\left(\frac{\mu V}{\sigma}\right)^{2/3}\sigma\frac{1}{t} - \frac{6\mu V L_D}{H_D^2} - P_{VAC} = 0} \quad (8)$$

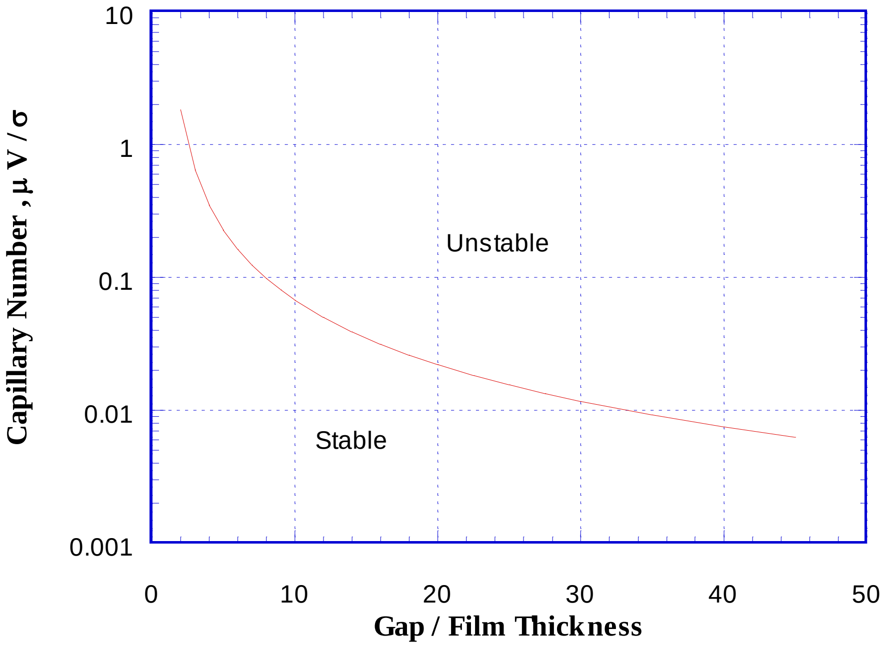
$t_{MIN}^{(2)}$ is the root of Eq.(8)

$$\boxed{t_{MIN} = \max\left(t_{MIN}^{(1)}, t_{MIN}^{(2)}\right)}$$

LOW FLOW LIMIT

MAXIMUM WEB SPEED AT A GIVEN FILM THICKNESS

MINIMUM FILM THICKNESS AT A GIVEN WEB SPEED



$$Ca \equiv \frac{\mu V}{\sigma} = 0.65\left(\frac{2}{H_0/t - 1}\right)^{\frac{3}{2}}$$

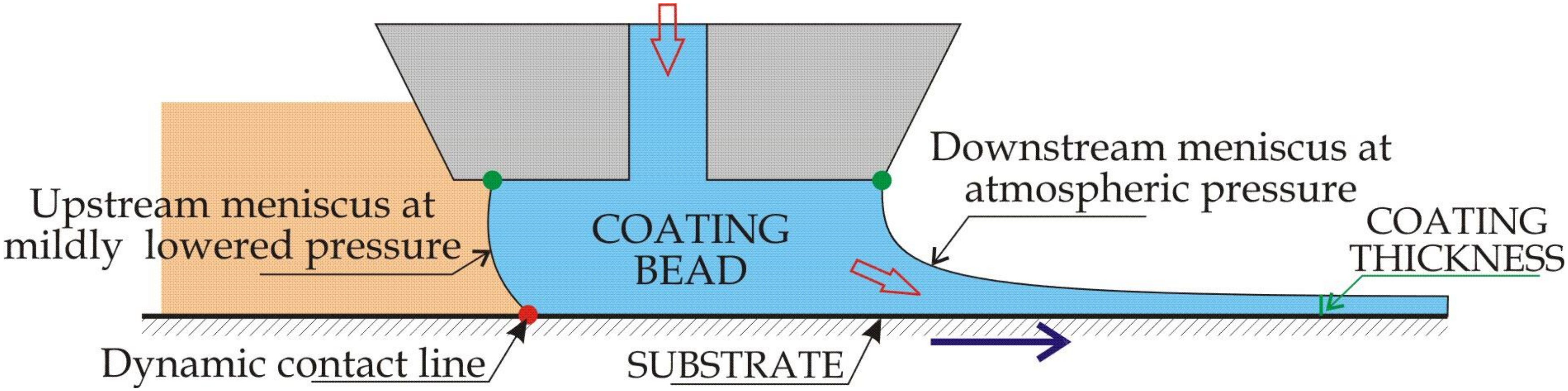
THE MAXIMUM POSSIBLE WEB SPEED FALLS AS THE WET FILM THICKNESS DECREASES

FOR $t = 0.6\,\text{mils};\ H_0 = 4\,\text{mils};\ \mu = 20\,\text{cP};\ \sigma = 25\,\text{dyn/cm}$ ➔ *V_{max} = 30 ft/min*

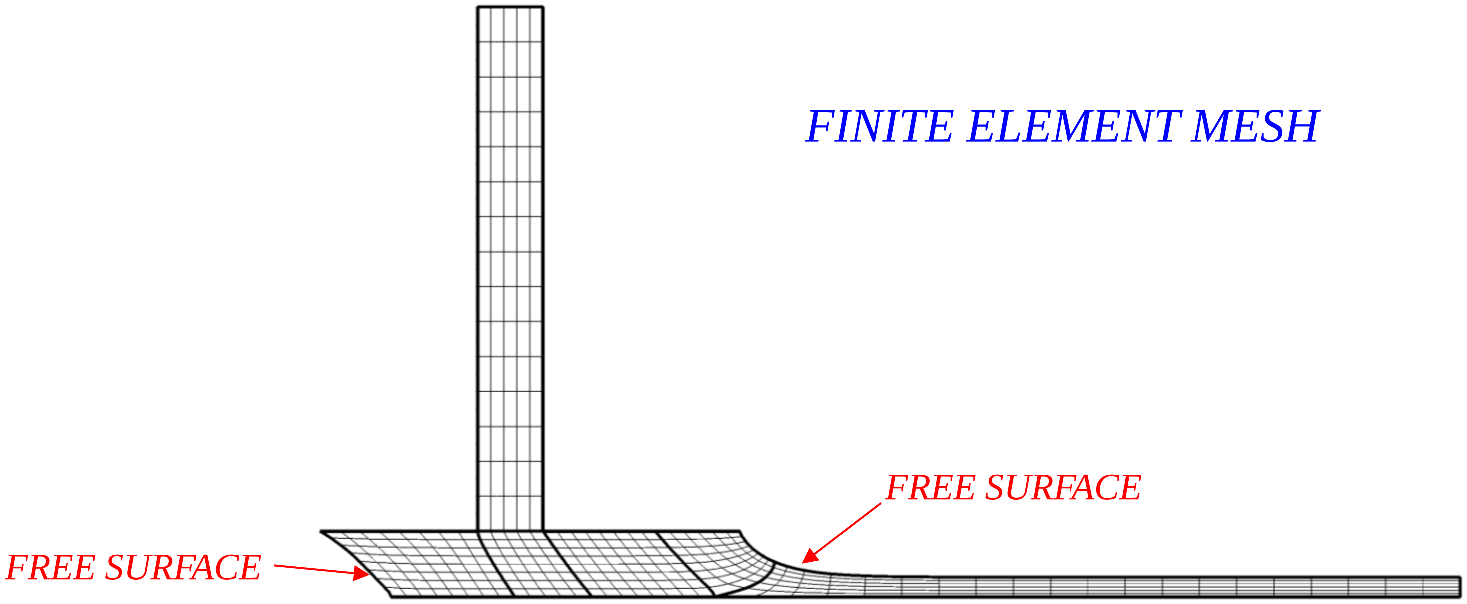
VISCOCAPILLARY MODEL VALID ONLY AT LOW CAPILLARY NUMBER

EXPERIMENTS HAVE SHOWN EXAMPLES WHERE THE MODEL FAILS TO PREDICT THE CORRECT MAXIMUM SPEED

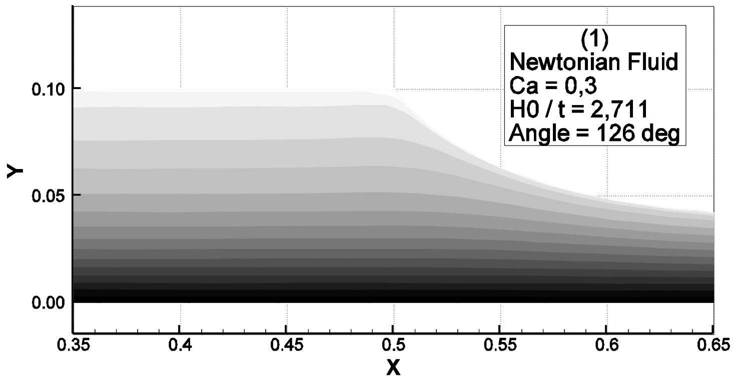
TWO-DIMENSIONAL MODEL – Navier-Stokes equations

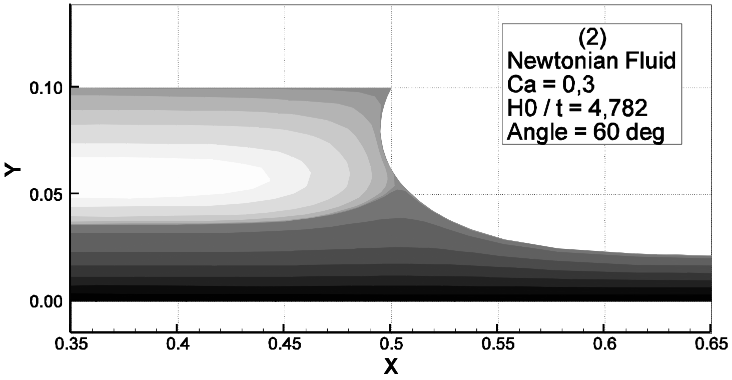


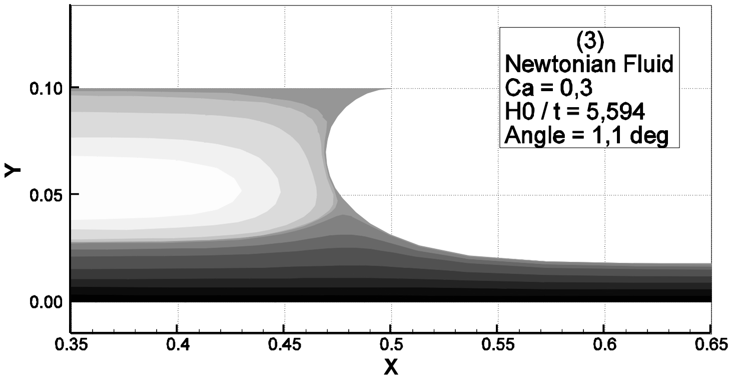
FINITE ELEMENT MESH



THINNER COATING THICKNESS





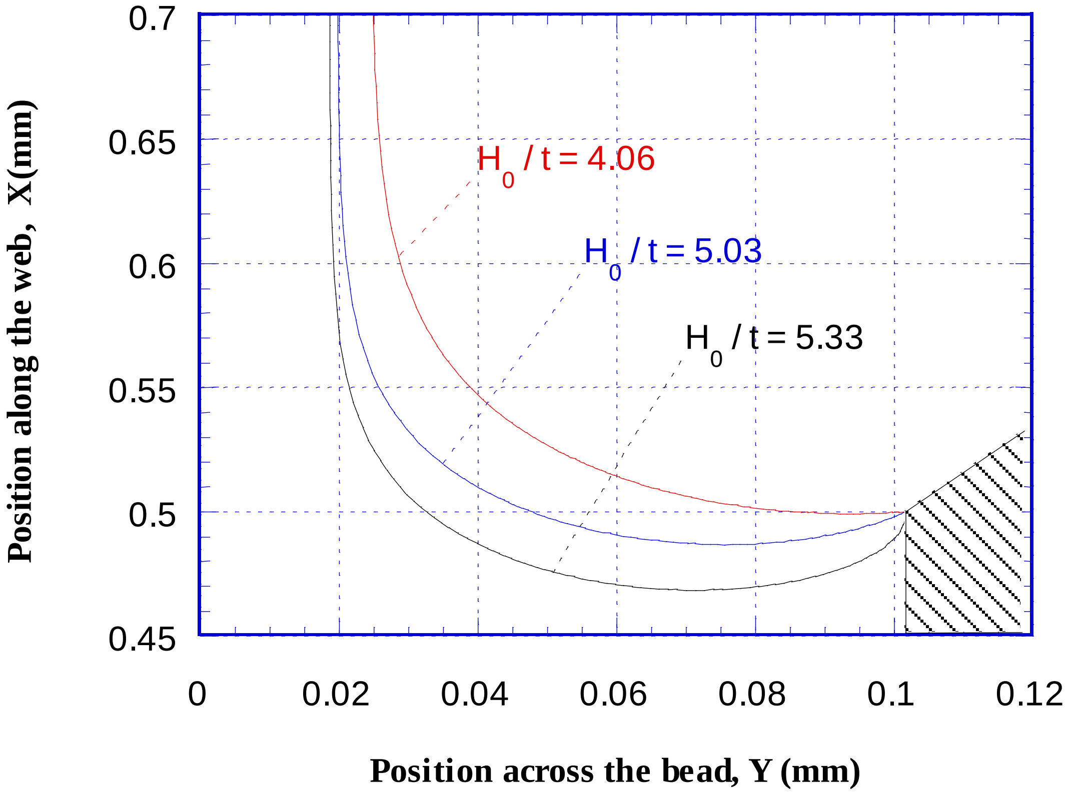


✓ THICK COATING:
NO RECIRCULATION

✓ RECIRCULATION ATTACHED TO
FREE SURFACE APPEARS
AT $H_0 / t \approx 3$

✓ MENISCUS BECOMES MORE
CURVED AS THICKNESS FALLS

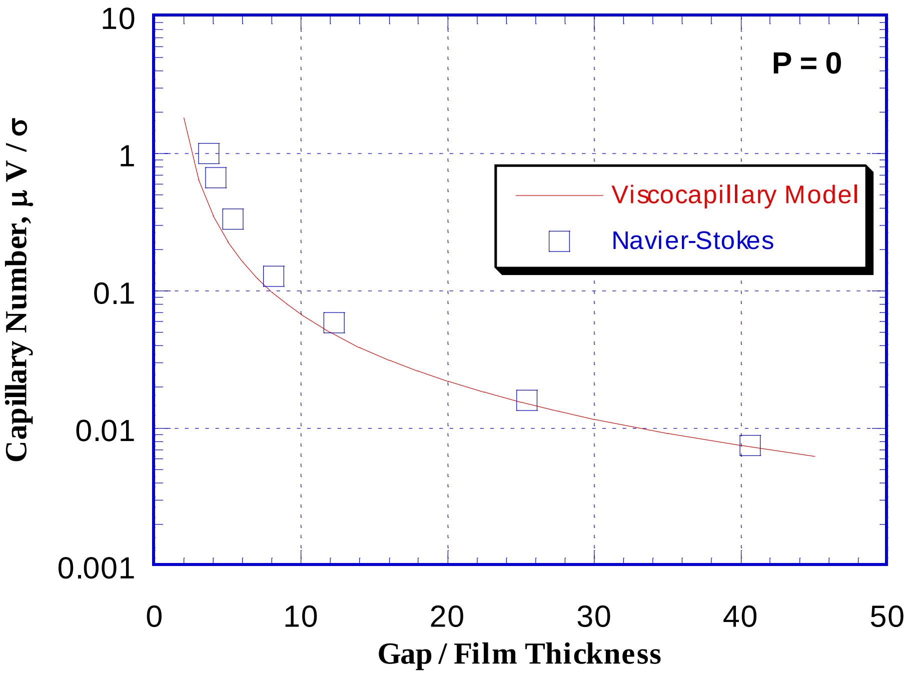
DETAIL OF MENISCUS CONFIGURATION AS THICKNESS FALLS



✓ AS THE GAP-TO-THICKNESS RISES, THE MENISCUS BECOMES MORE CURVED

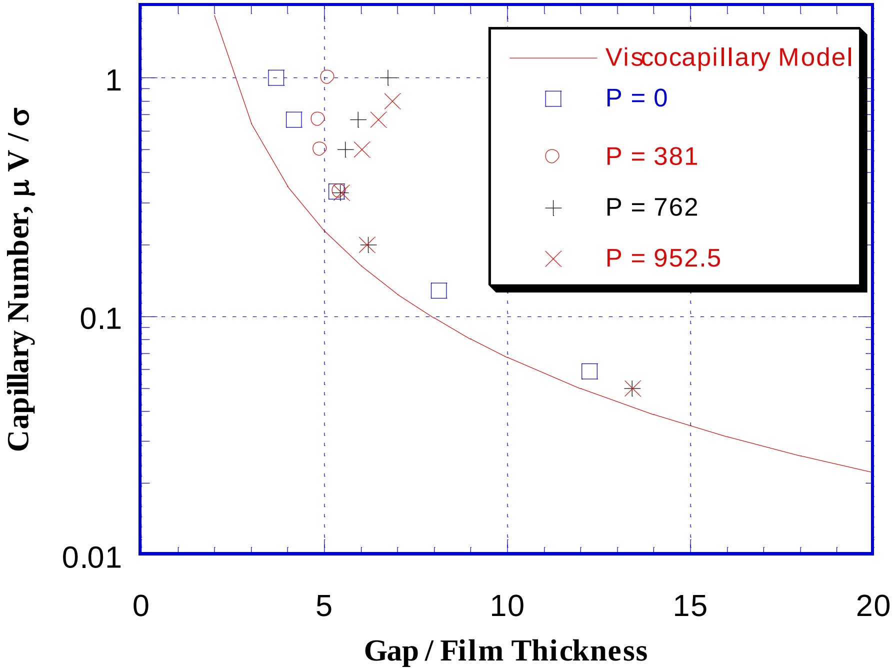
✓ AT THE TURNING POINT ($H_0 / t = 5.33$), THE ANGLE BETWEEN THE DIE AND THE FREE SURFACE IS ALMOST ZERO

BEAD BREAKS INTO RIVULETS

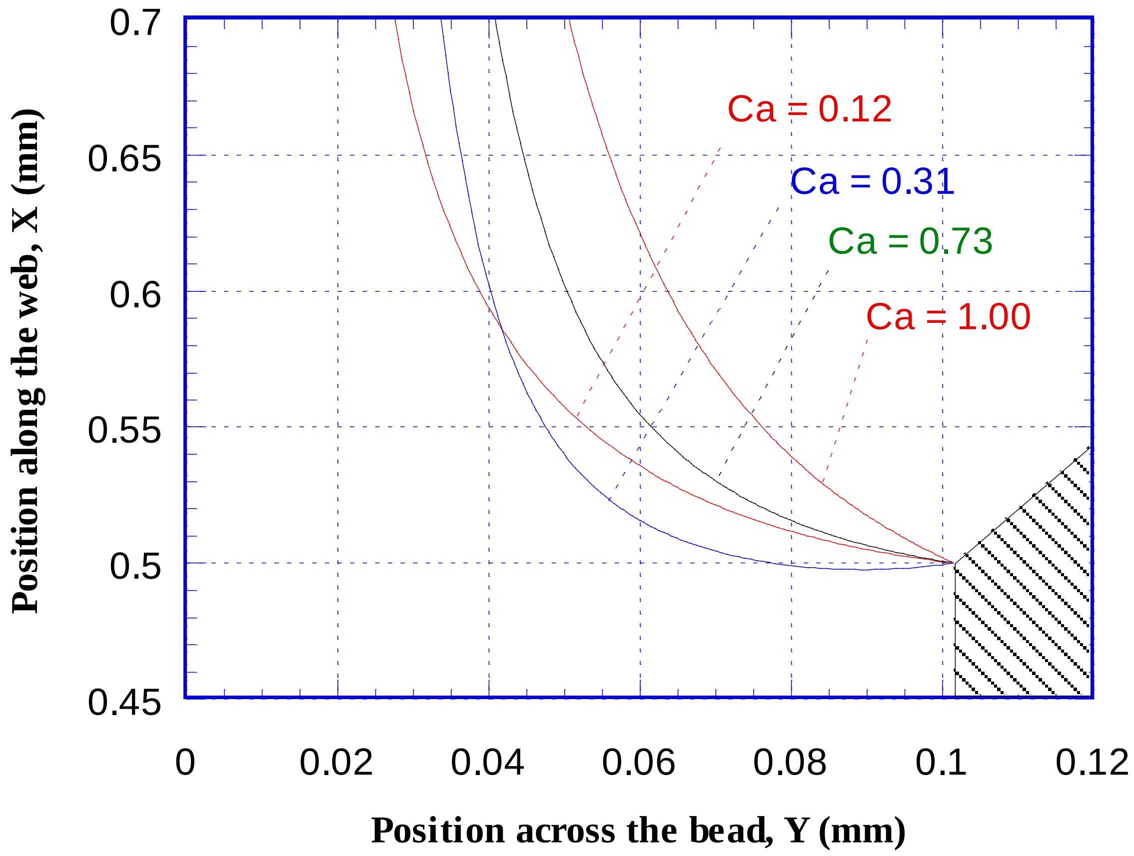


NO INERTIAL EFFECTS

EFFECT OF INERTIA



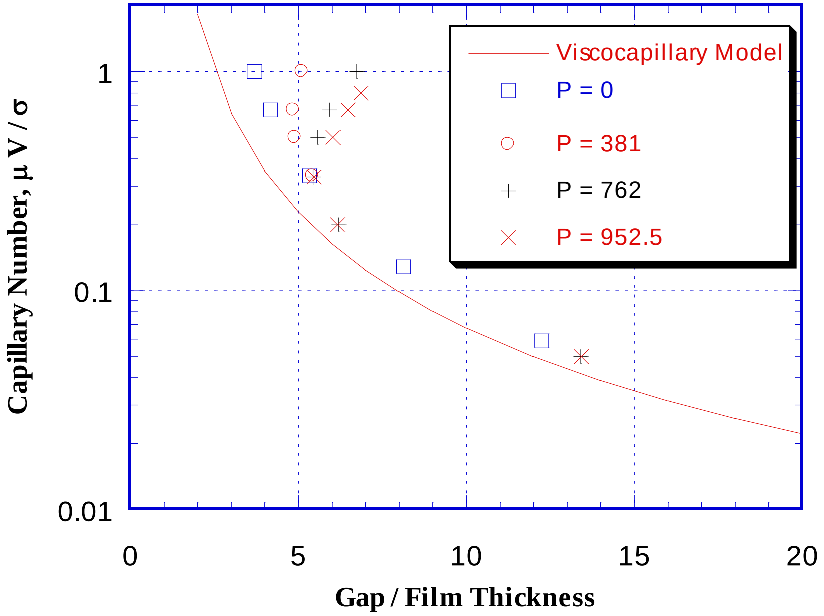
DETAIL OF MENISCUS CONFIGURATION AS COATING SPEED RISES



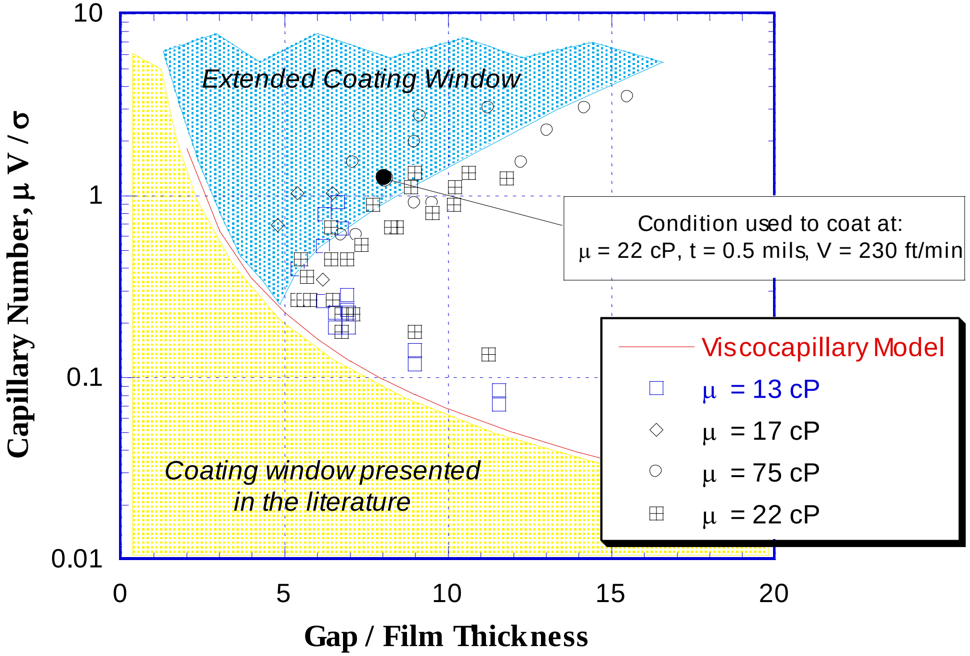
INERTIA CAN BE USED TO COUNTERACT THE RECEDING ACTION OF THE DOWNSTREAM MENISCUS AND DELAY THE ONSET OF THE LOW FLOW LIMIT

(Carvalho and Keshghi, 2000)

MODEL



EXPERIMENTS



- ✓ COATING WINDOW OF THE PROCESS IS LARGER THAN THE ONE REPORTED PREVIOUSLY IN THE LITERATURE

- ✓ CAN COAT THINNER BY GOING FASTER !

Current Coating Fundamentals Challenges

- Minimization of film thickness variation for more uniform films;
- Better understanding of coating of particulate suspensions for more complex film structures;
- Better understanding of multilayer coating process for more complex film structures;
- Discrete and patch coating;

Examples of recent advances and how they can help the coating industry...

Current Coating Fundamentals Challenges

- Minimization of film thickness variation for more uniform films;
- Better understanding of coating of particulate suspensions for more complex film structures;
- Better understanding of multilayer coating process for more complex film structures;
- Discrete and patch coating;

Examples of recent advances and how they can help the coating industry...

Transient Response of Coating Flow

Romero e Carvalho (CES, 2008), Perez e Carvalho (JEM, 2011)

Production lines are subjected to perturbations (even if very small…)

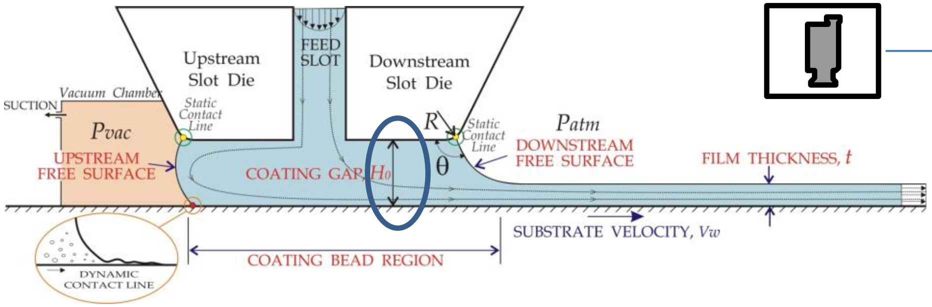
Gap oscillation

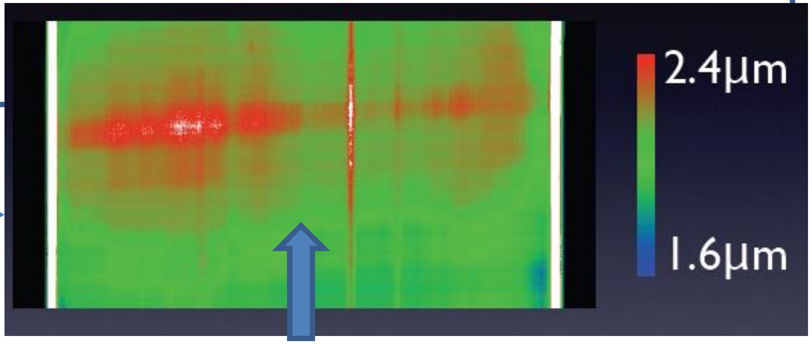
$$H(t) = H_0 + H_m \sin(\omega t)$$

Roll radius is not constant,
Mechanical vibrations, …

Coating thickness oscillation

$$h(t) = h_0 + h_m \sin(\omega t + \phi)$$

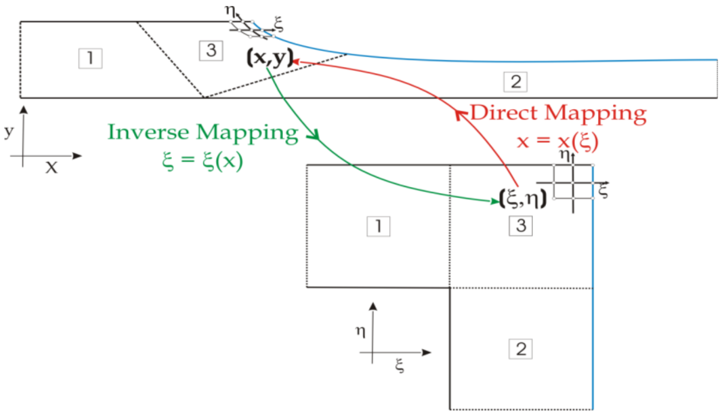




Goal

Optimize slot coating process to minimize film thickness variation due to ongoing disturbances of process conditions;

Solution Method



$$\rho\left[\overset{\circ}{\mathbf{v}} + (\mathbf{v} - \overset{\circ}{\mathbf{x}}) \cdot \nabla \mathbf{v}\right] - \nabla \cdot \mathbf{T} = 0.$$

$$\nabla \cdot (D_\xi \nabla \xi) = 0 \qquad ; \qquad \nabla \cdot (D_\eta \nabla \eta) = 0.$$

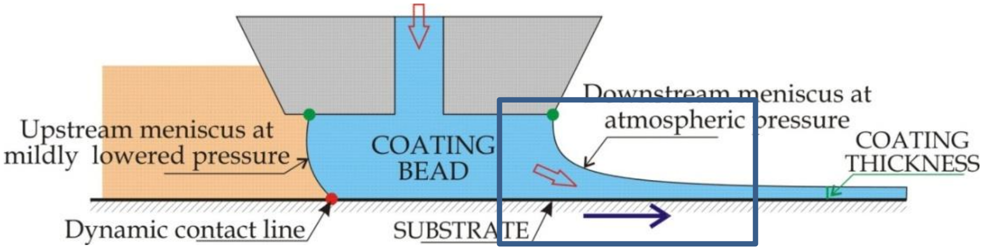
Finite Element Method

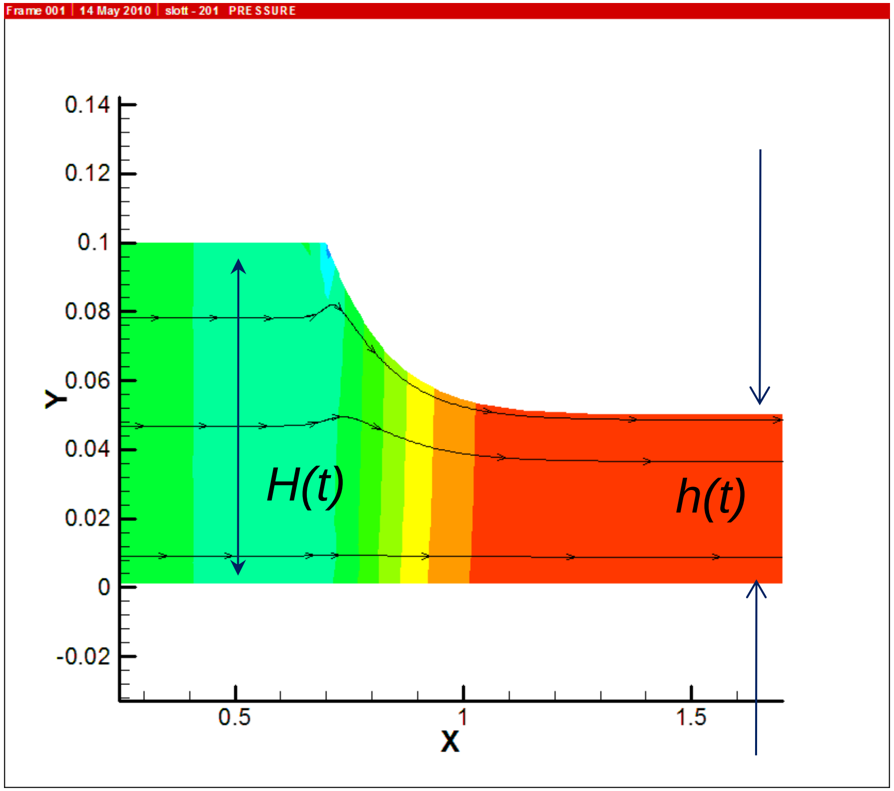
$$\mathbf{v} = \sum_j^M \tilde{\mathbf{v}}_j(t)\, \varphi_j^{\mathbf{v}}(\xi,\eta), \quad p = \sum_j^N \tilde{p}_j(t)\, \varphi_j^{p}(\xi,\eta), \quad \mathbf{x} = \sum_j^M \tilde{\mathbf{x}}_j(t)\, \varphi_j^{\mathbf{x}}(\xi,\eta).$$

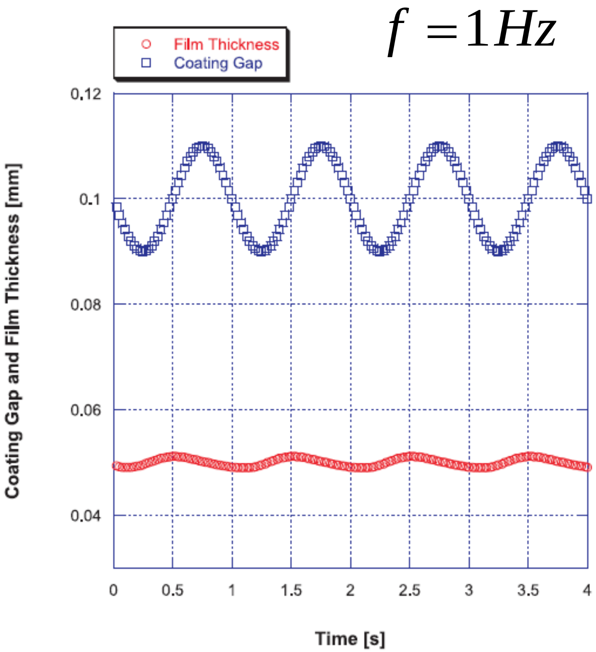
Implicit time integration –
Newton's method

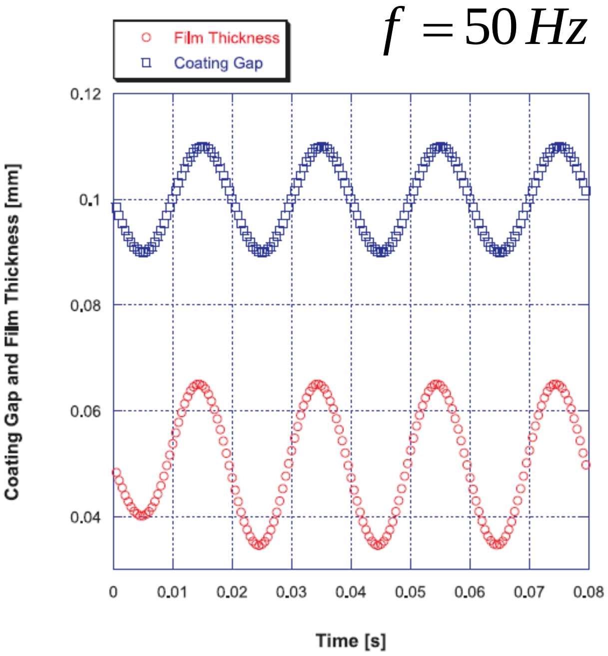
$$\left(\frac{1}{\Delta t}\mathbf{M} + \mathbf{J}\right)\, \delta\mathbf{u}^{(k+1)} = -\mathbf{R}(\mathbf{u}^{(k+1)}, \mathbf{u}^{k}),$$
$$\mathbf{u}^{(k+1)} = \mathbf{u}^{k} + \delta\mathbf{u}^{(k+1)},$$

Transient Response



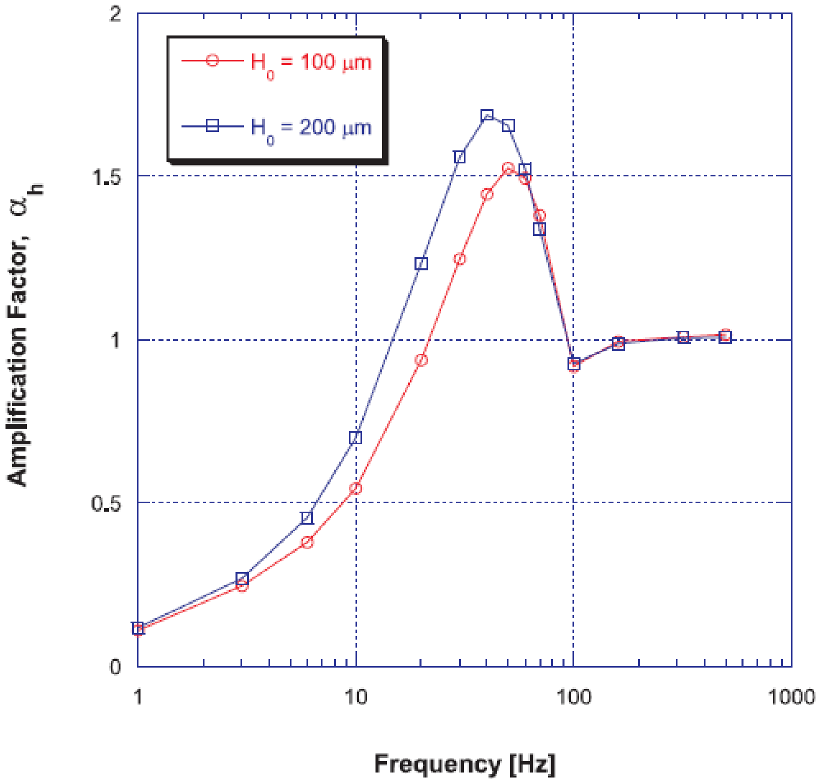




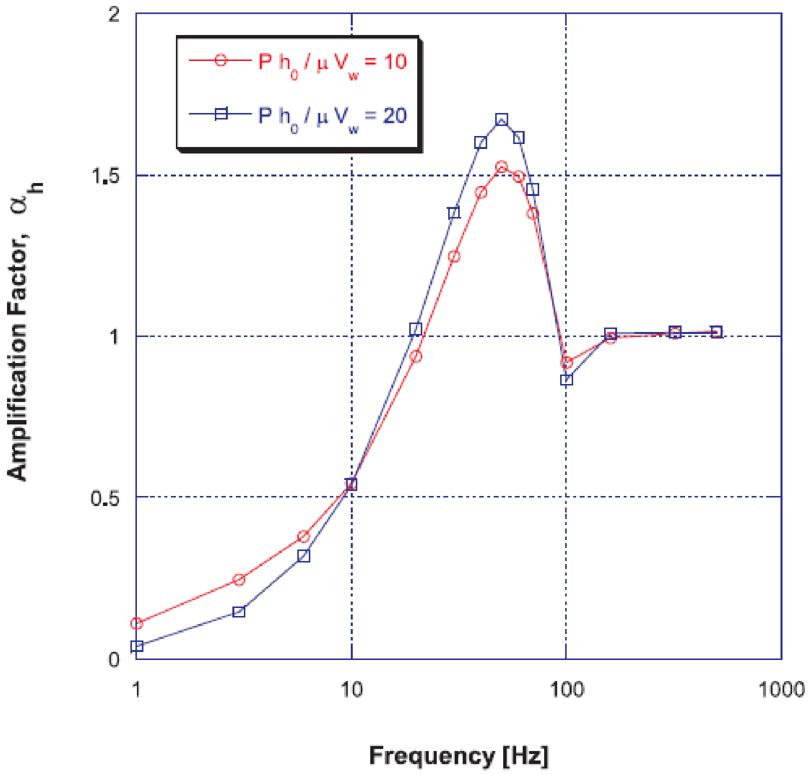


Amplification factor is a function of

frequency of the imposed disturbance
process conditions
geometry of die lip

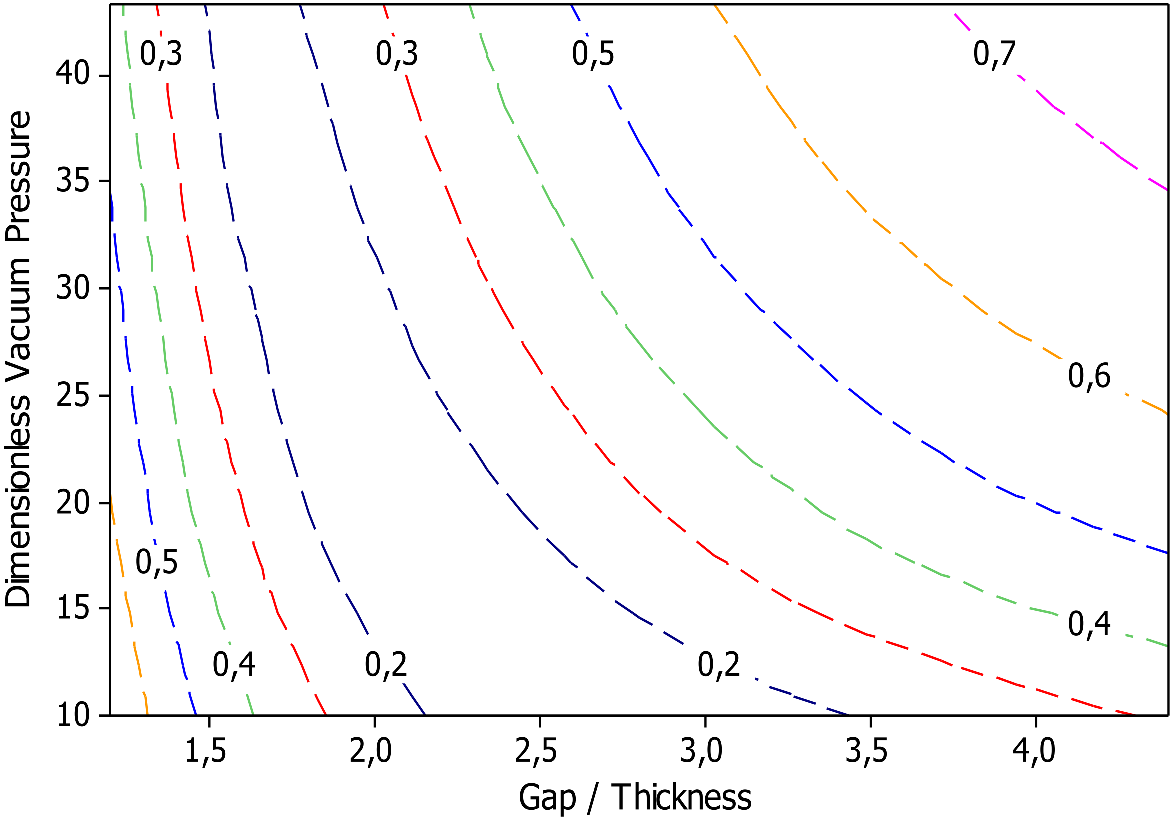


Effect of Coating Gap



Effect of Vacuum pressure

Amplification factor at a given frequency can be mapped as a function of process conditions.



$$f = 3\,Hz$$

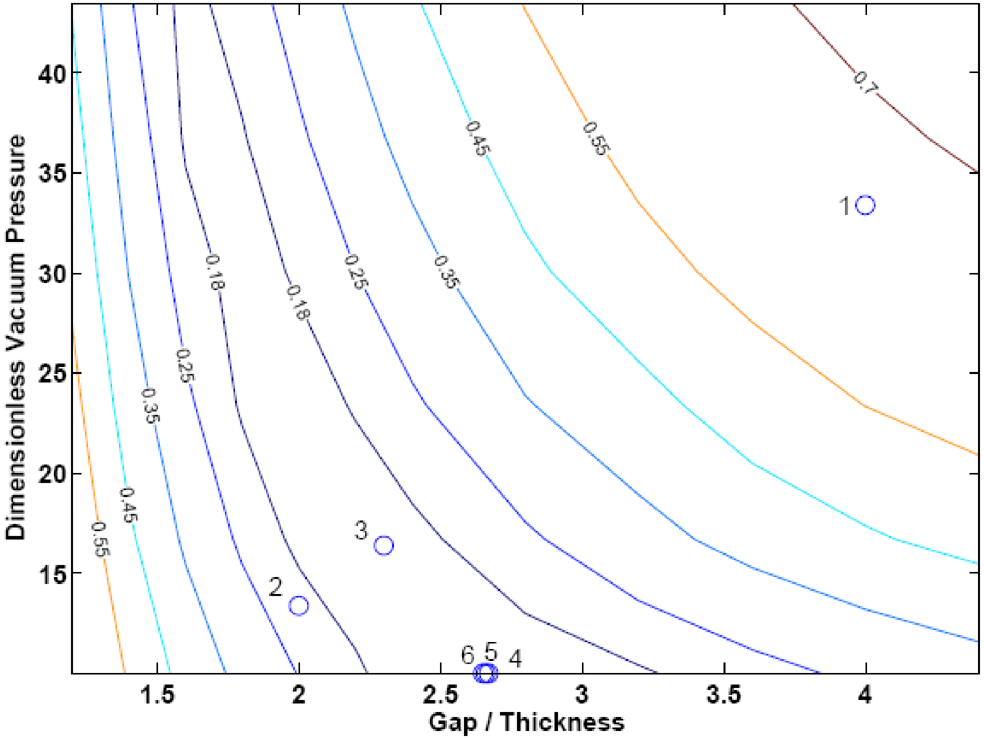
Amplitude of film thickness oscillation may be reduced by a factor of 5 just by adjusting process conditions.

Boundary Constraint Optimization algorithm.

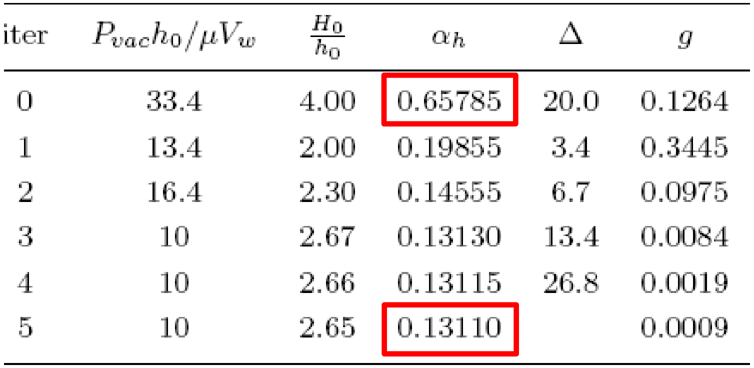
$$q(x-x_0)=\frac{1}{2}(x-x_0)^T H(x-x_0)+b^T(x-x_0)+f(x_0)$$

$$H=\nabla^2 f\ \ (x_0) \qquad b=\nabla f(x_0)$$

Contour plot of amplification factor as a function of gap and vacuum pressure



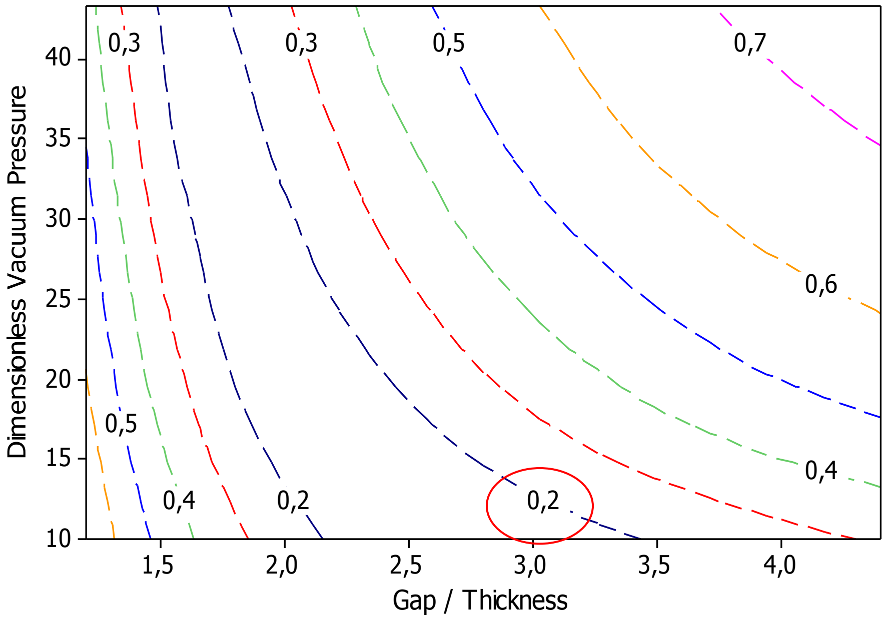
$$f=3Hz$$

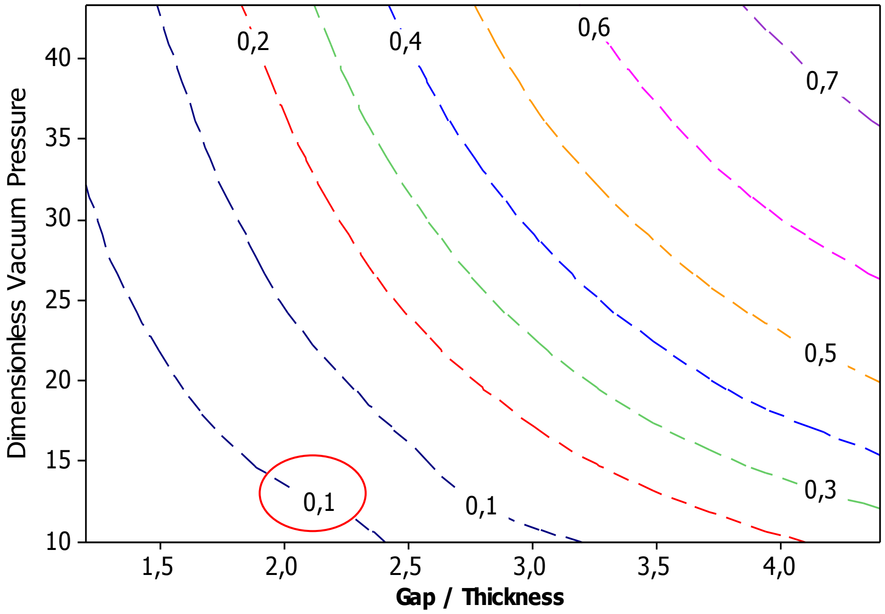


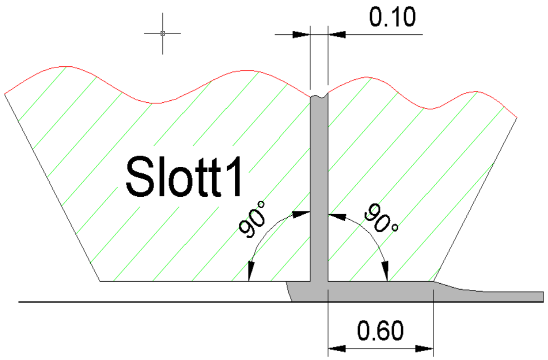
iter	$P_{vac}h_0/\mu V_w$	$\frac{H_0}{h_0}$	α_h	Δ	g
0	33.4	4.00	0.65785	20.0	0.1264
1	13.4	2.00	0.19855	3.4	0.3445
2	16.4	2.30	0.14555	6.7	0.0975
3	10	2.67	0.13130	13.4	0.0084
4	10	2.66	0.13115	26.8	0.0019
5	10	2.65	0.13110		0.0009

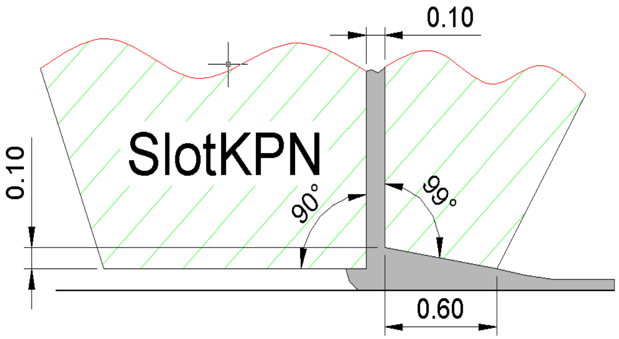
Solution has been implemented in a production line at Fuji Film, Japan.

Amplification factor map is a strong function of die lip geometry.









Die lip geometry may also be optimized to reduce film thickness oscillation.

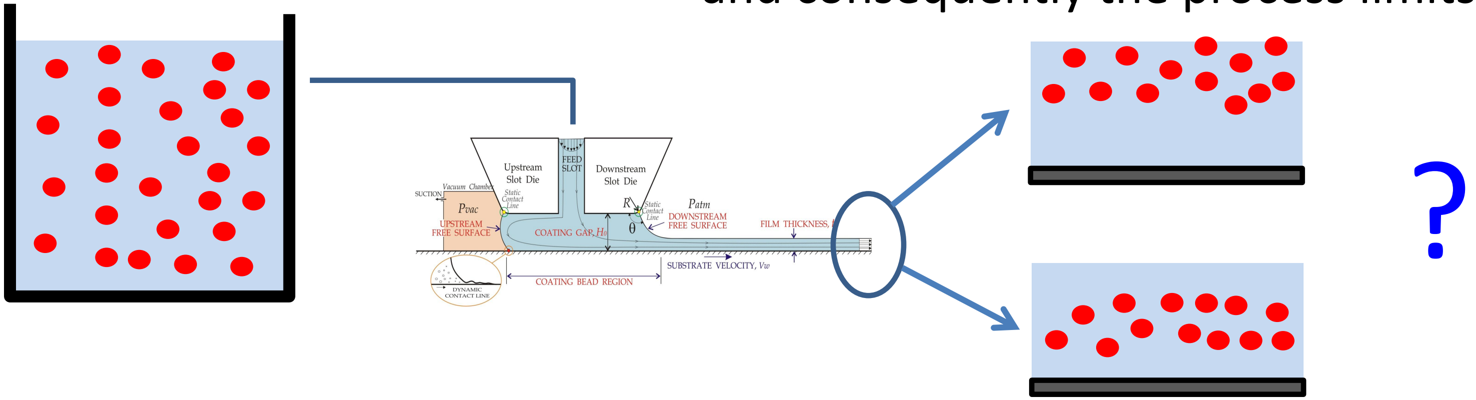
Slot Coating of Particle Suspensions

In many applications, coating liquid is a particle suspension.

Common approach is to study the flow as Newtonian or non-Newtonian with the liquid viscosity evaluated based on the avarage particle concentration.

Experimental evidences show that suspensions of particles assume very non-uniform concentration distributions in nonhomogeneous shear flow.

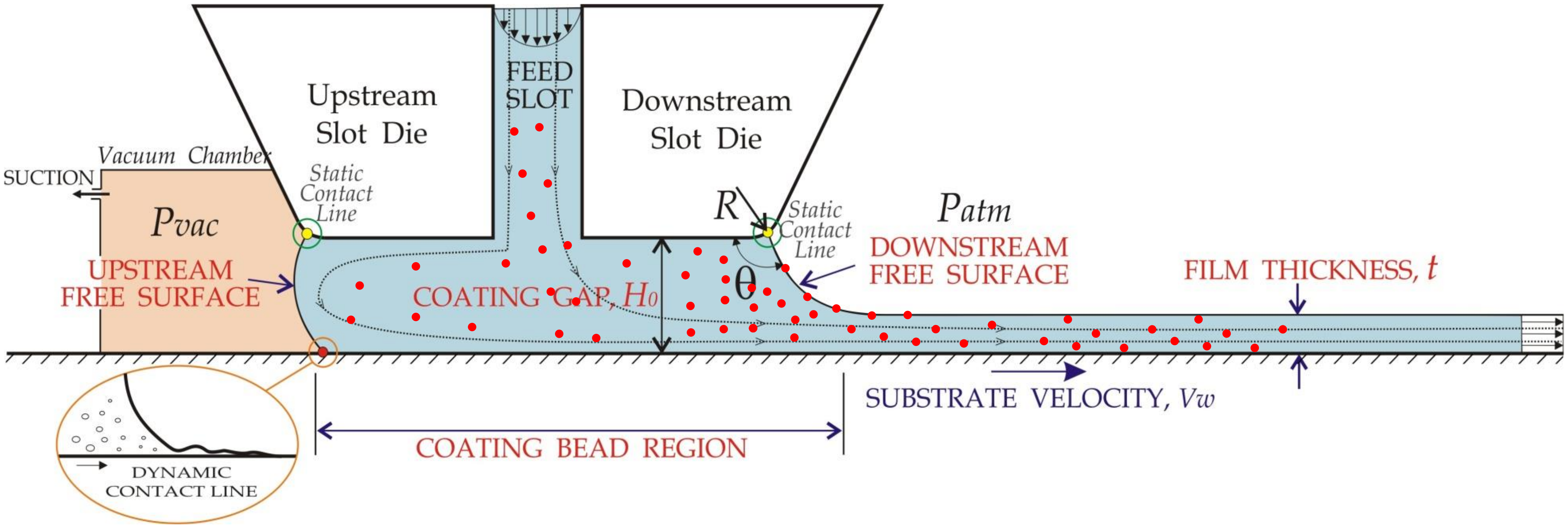
Local variation of viscosity and surface tension may change the flow pattern and consequently the process limits.



Particle suspension *Coating process* *Coated film*

Final particle distribution on the coated film may not be uniform and have a strong effect on the drying process and product performance.

MATHEMATICAL MODEL OF COATING FLOW



Momentum conservation
$$\rho\,\mathbf{v}\cdot\nabla\mathbf{v} - \nabla\cdot\left[-p\mathbf{I}+\mu(c)\left(\nabla\mathbf{v}+(\nabla\mathbf{v})^{T}\right)\right]=0$$

Mass conservation
$$\nabla\cdot\mathbf{v}=0$$

Particle Transport
$$\mathbf{v}\cdot\nabla c+\nabla\cdot\mathbf{N}=0$$

BCs along interface
$$\mathbf{n}\cdot\mathbf{v}=0$$
$$\mathbf{n}\cdot\mathbf{T}=\sigma(c_s)\kappa\,\mathbf{n}+\nabla_s\overbrace{\cdot\,\sigma(c_s)}^{\sigma(c_s)=\sigma}$$
$$\mathbf{n}\cdot\mathbf{N}=0$$

as first approximation

Particle Transport / Bulk

$$\mathbf{v}\cdot\nabla c+\nabla\cdot\mathbf{N}=0$$

Total flux of particles due to different migration mechanisms

Assume neutrally bouyant spherical, rigid particles;

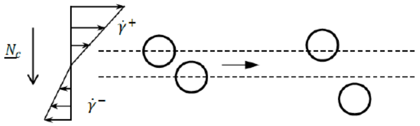
Neglect Brownian diffusion – particle size > 0.5 μm;

Diffuse flux model for particle migration proposed by Phillips et al (*PF, 1992*);

Particle migration by two mechanisms: $\mathbf{N}=\mathbf{N}_c+\mathbf{N}_\eta$

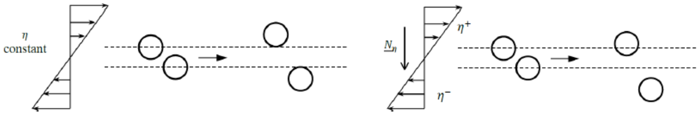
1. Spatially varying particle-particle interaction frequency

$$\boxed{\mathbf{N}_c=-k_c a^2 c\nabla(\dot{\gamma}c)}$$



2. Spatially varying liquid viscosity

$$\boxed{\mathbf{N}_\eta=-k_\eta\dot{\gamma}c^2\left(\frac{a^2}{\eta}\right)\nabla\eta=-k_\eta\dot{\gamma}c^2\left(\frac{a^2}{\eta}\right)\frac{d\eta}{dc}\nabla c}$$



Viscosity Model

Empirical viscosity model for concentrated suspension developed by Krieger:

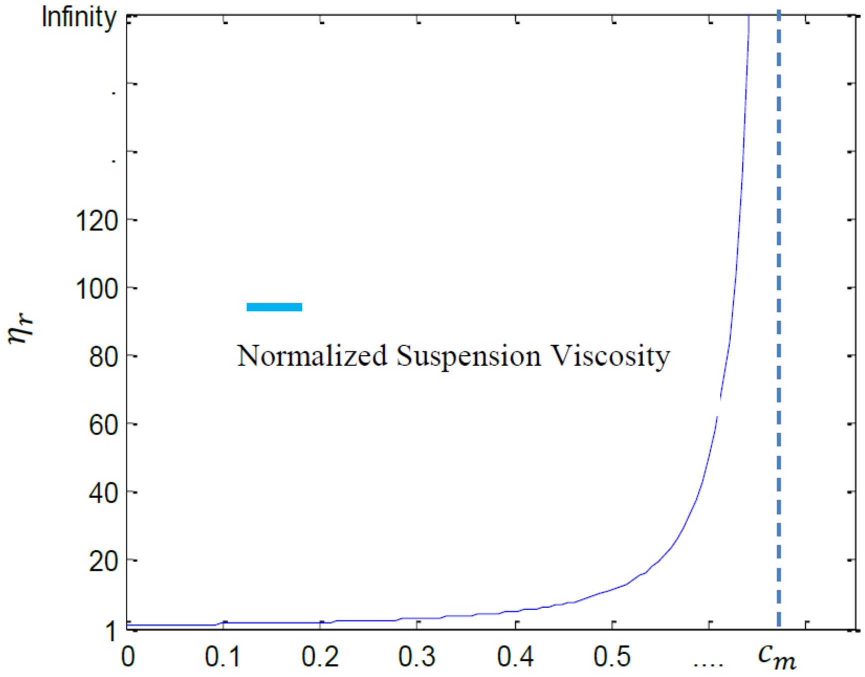
$$\eta = \eta_S \left(1 - c/c_m\right)^{-1.82}$$

η - suspension viscosity.

η_s - continuous phase viscosity.

c - volume fraction of particles.

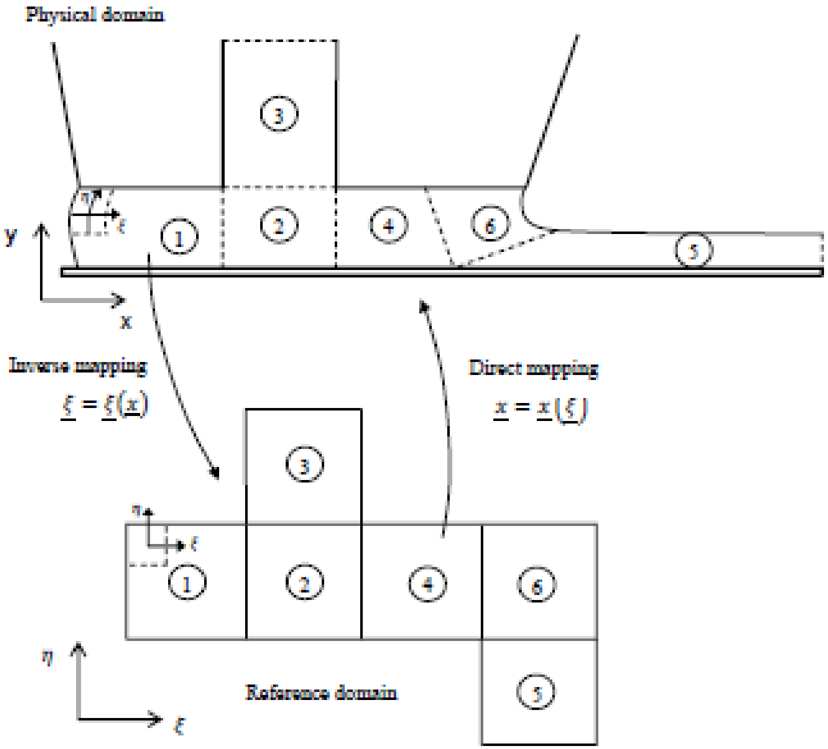
c_m - maximum packing fraction of particles.



In this work: $\eta_s = 12\,\text{cP};\quad c_m = 0.68$

$$c_0 = 0.4 \quad \Rightarrow \quad \eta(c_0) \approx 60\,\text{cP}$$

SOLUTION METHOD



Unknown physical domain is mapped
to a fixed reference domain;

Mapping is described by a set of
differential diffusion equations:

$$\nabla \cdot (D_\xi \nabla \xi) = 0, \quad \nabla \cdot (D_\eta \nabla \eta) = 0$$

The set of PDE is solved by Galerkin´s / Finite Element Method;

Need to modify system in order to compute derivative of shear rate
(second derivative of velocity field)

Deformation rate tensor is treated as an independent field that is also
expanded in terms of finite element basis functions:

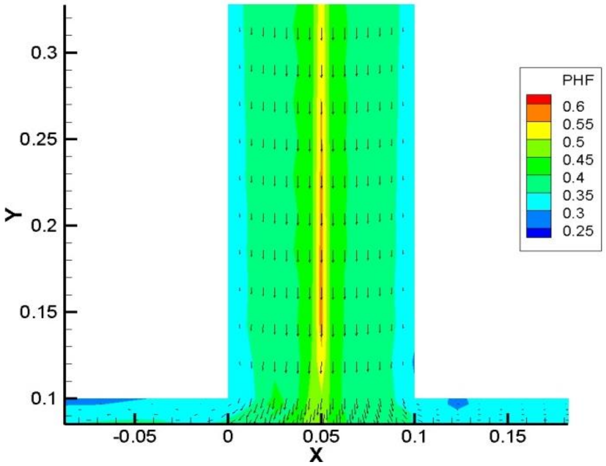
$$\mathbf{G} = \nabla \mathbf{v} - \frac{\nabla \cdot \mathbf{v}}{tr(\mathbf{I})} \mathbf{I}$$

RESULTS

$V = 0.1\,\mathrm{m/s}\,; \quad \sigma = 60\,\mathrm{dyn/cm}\,; \quad H = 100\,\mu m\,; \quad \eta_s = 12\,\mathrm{cP}\,; \quad k_c = 1.2\,; \quad k_\eta = 1.8$

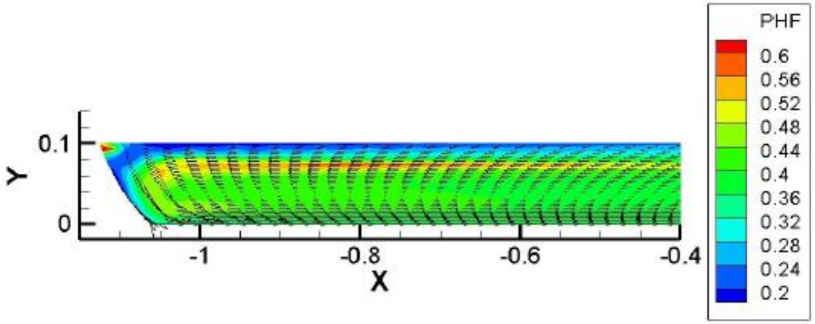
Inlet condition: uniform concentration profile: $c(\mathbf{x}) = c_0 = 0.4$

Feed Slot: Particles migrate towards the middle of the feed slot (zero shear rate)



Low particle concentration at the walls;
Flow is lubricated;
Possible particle agglomeration.

Upstream gap: Particles migrate towards zero shear rate layer.



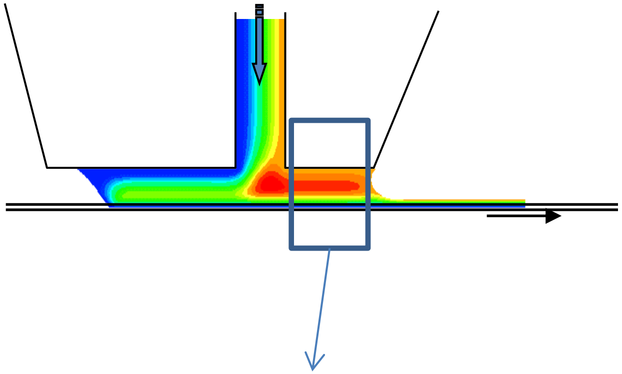
Lower particle concentration at die lip;
Flow is lubricated;
Upstream meniscus position is shifted;
Effect on low vacuum limit.

Film thickness (flow rate) has a strong effect on

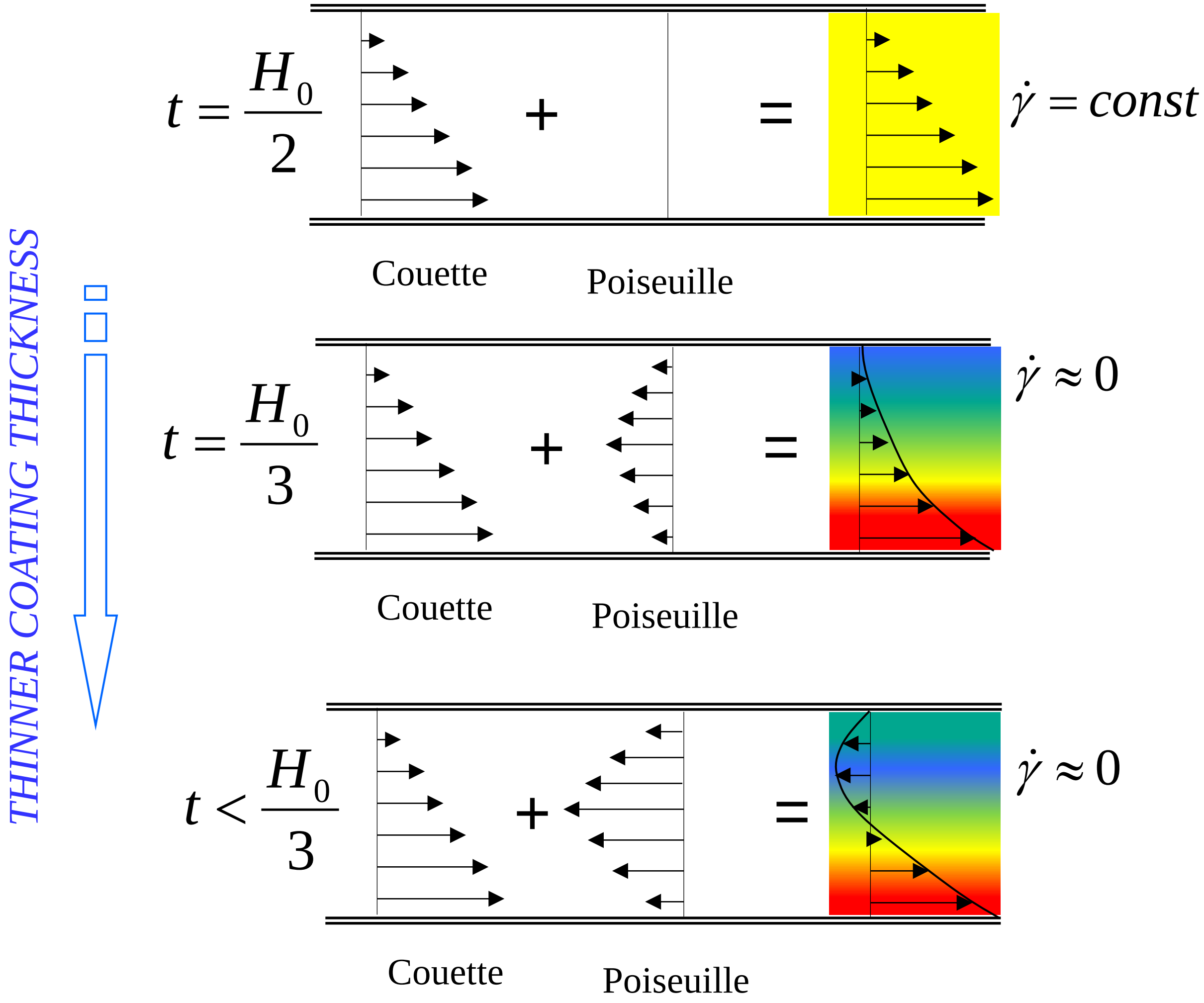
deformation rate distribution under the die lip;

Consequently, it has a strong effect on

the particle concentration in the coating bead and final coated layer;



Almost rectilinear flow
Couette (drag) +
Poiseuille (pressure gradient)



$t = \frac{H_0}{2}$ + = $\dot{\gamma} = const$

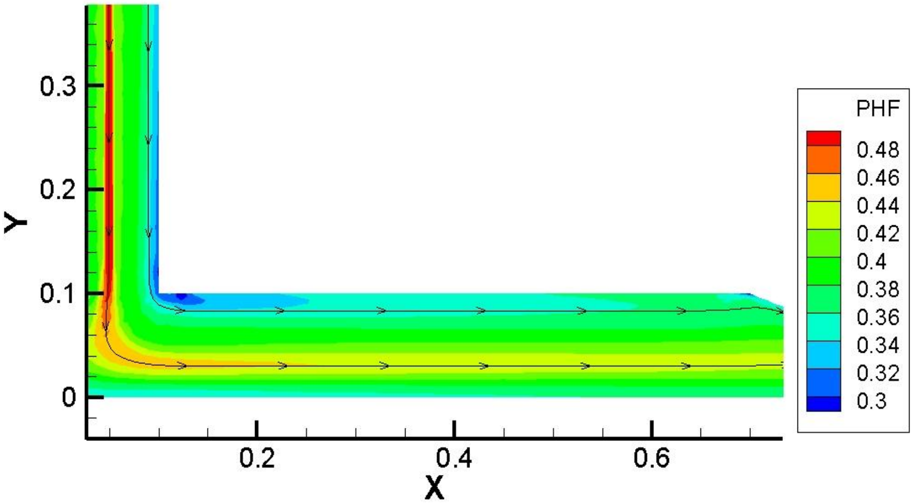
$t = \frac{H_0}{3}$ + = $\dot{\gamma} \approx 0$

$t < \frac{H_0}{3}$ + = $\dot{\gamma} \approx 0$

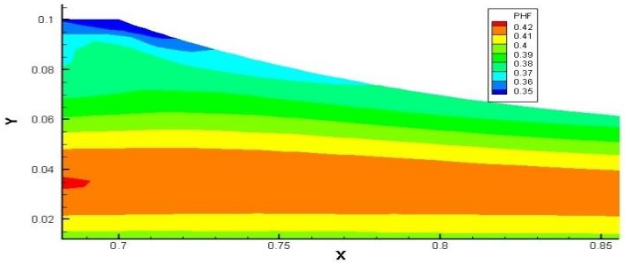
Film thickness equal to half of the gap $t = 50\mu m = \frac{H_0}{2}$

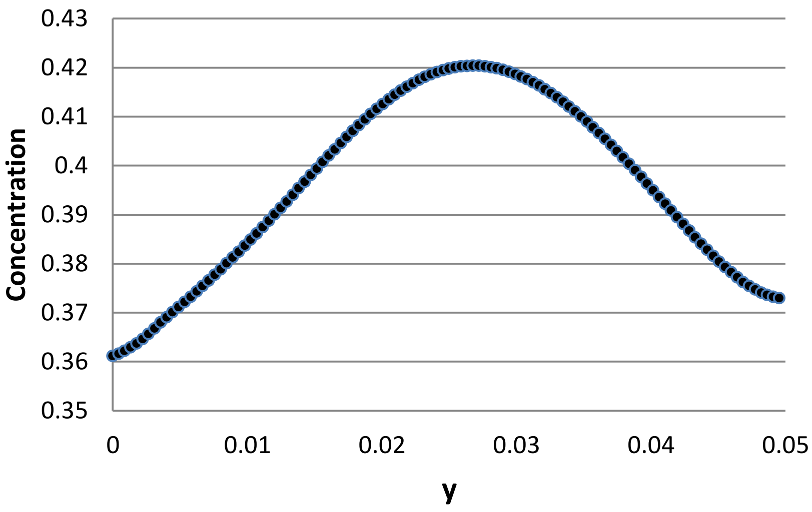
Flux related to shear rate gradient is zero. Weak particle migration after feed slot.

High particle concentration at center of feed slot is convected to final coated layer.



Concentration field at coated layer





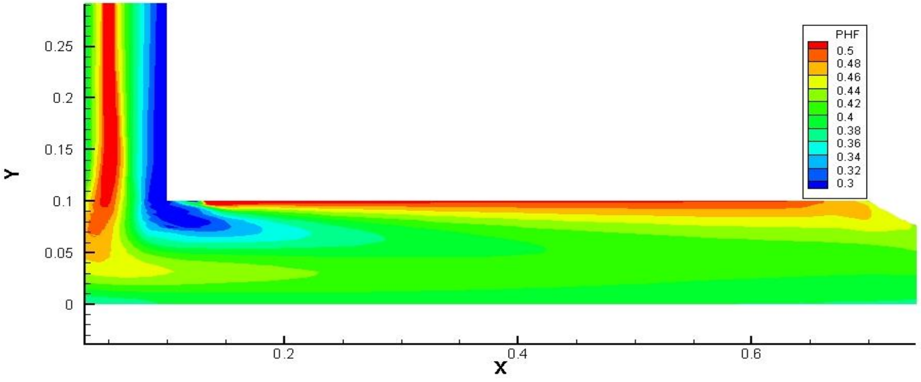
Region of high particle concentration in the middle of the coated layer.

Possible effect on final structure and drying process.

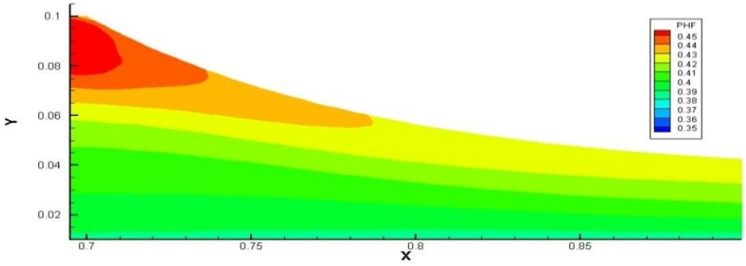
Film thickness close to one-third of the gap $t = 37\mu m \approx \frac{H_0}{3}$

Strong flux towards the zero-shear rate layer attached to the die lip;

High particle concentration attached to the die lip is convected to top of the coated film.

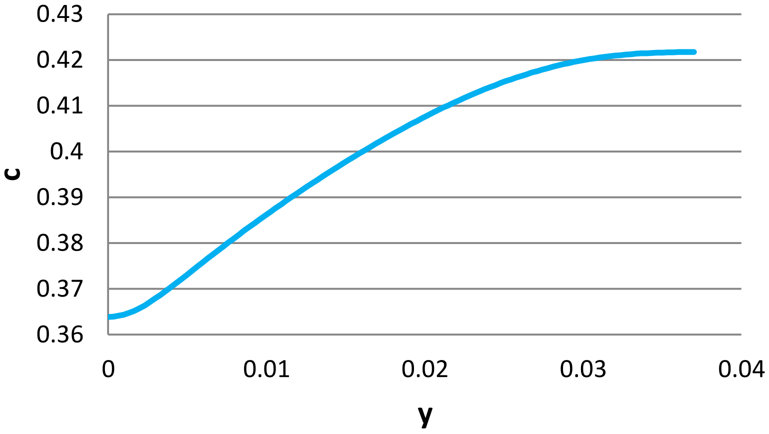


Concentration field at coated layer



Region of high particle concentration on the top of the coated layer.

Possible effect on final structure and drying process.

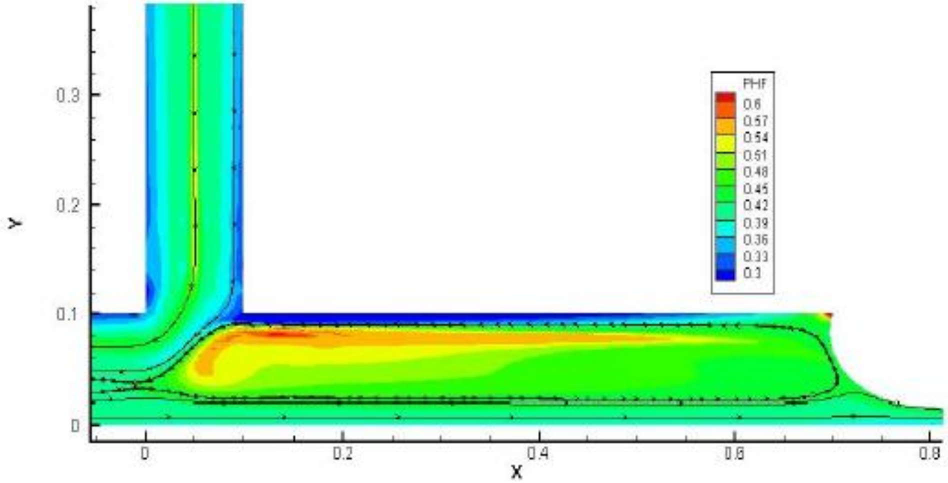


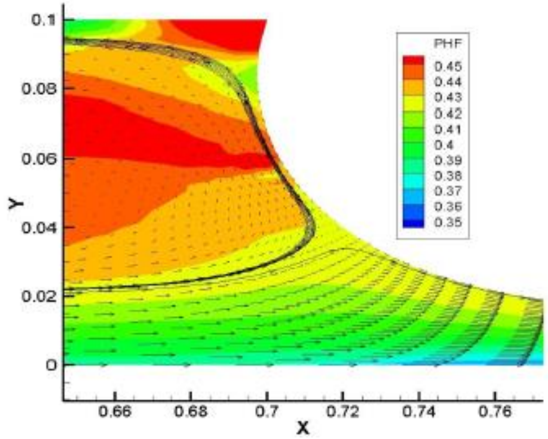
Film thickness less than one-third of the gap $t = 14\mu m < \frac{H_0}{3}$

Recirculation under the the die lip;

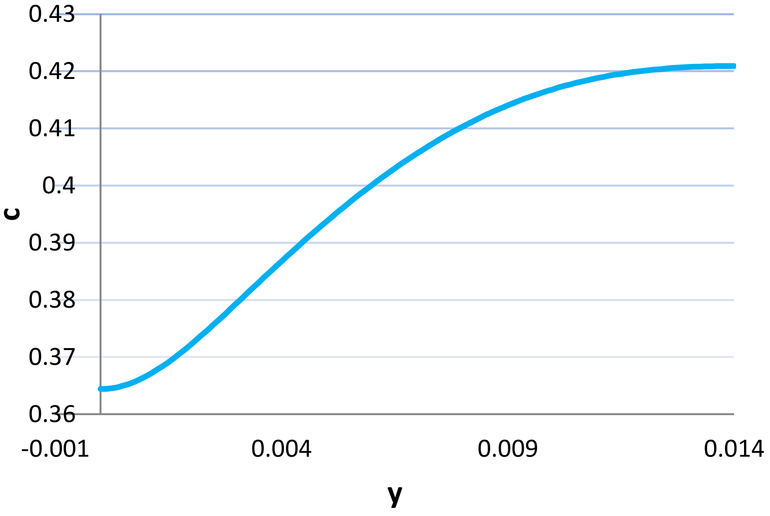
High concentration inside the recirculation (near close packing) – particle agglomeration?;

High concentration gradient in the free surface – Strong Marangoni effect ?





Region of high particle concentration on the top of the coated layer.



Final Remarks

- ✓ Slot coating fundamentals is well understood for two-dimensional, steady-state operation – coating window studies;

- ✓ Fundamental understanding pays off

  objectives need to be well defined for industrial use

- ✓ Coating research is addressing current and more complex issues faced by the coating industry;

Microscale Free Surface Flows of Complex Liquids

Prof. Marcio Carvalho's Research Group

Thank you!

You are welcome to visit PUC and Rio de Janeiro

ISCST

International Society of Coating Science & Technology

17th International Coating Science and Technology Symposium

September 7-10, 2014

Sheraton Carlsbad Resort, San Diego, CA

Highlights

- Interaction across industrial sectors and between academia and industry
- Special sessions focused on energy
- Vendor Exhibit
- Welcome Reception
- Networking Sessions
- Short Course.
- Extended Abstract book

www.iscstsymposium.org

Sponsored by: The International Society of Coating Science and Technology (www.iscst.org)

In Cooperation with: The European Coating Symposium and The Japan Coating Symposium

The ISCST Symposium provides a forum for researchers with both academic and industrial perspectives on coating science and technology to discuss the latest research on the deposition and solidification of thin liquid films. The Symposium features contributions on both fundamental and applied research by many of the experts in the field from Europe, Asia, and the Americas. The Symposium format is designed to provide opportunities for networking and for the exchange of information between scientists and engineers who are working on coating process and materials development and manufacturing.

Symposium Chair: Prof. Marcio Carvalho, PUC-Rio, msc@puc-rio.br
Symposium Co-Chair: Dr. Brent Bell, W.L. Gore & Associates Inc., brbell@wlgore.com
ISCST President: Prof. Andrew Hrymak, The U. of Western Ontario, ahrymak@uwo.ca
Symposium Facilitator: Ms. Ashley Wood, AIMCAL, ashley@aimcal.org