SAND2021-15554C

Process Modeling and Simulation Tools for Roll-to-Roll Nanomanufacturing

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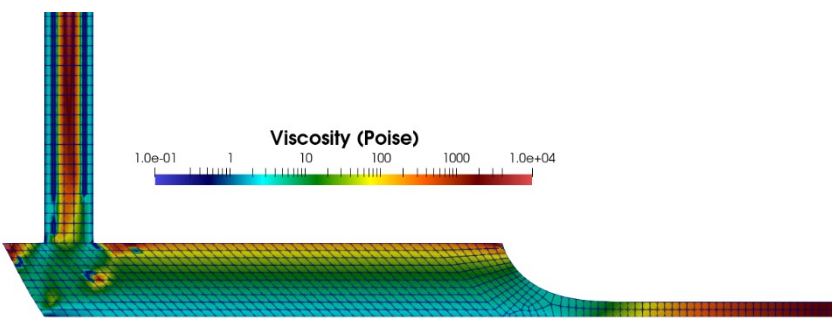

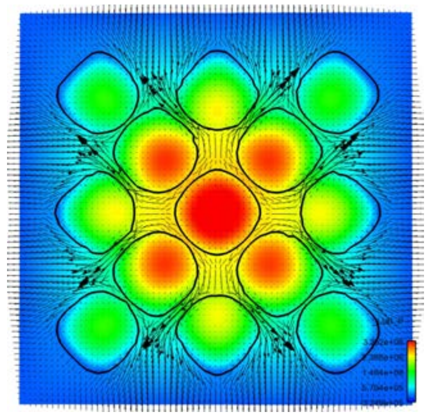

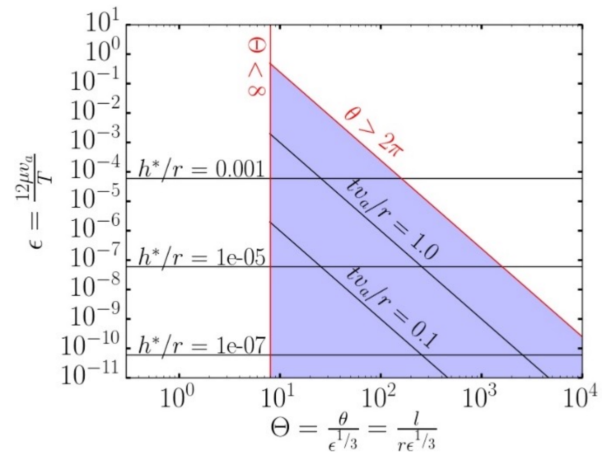

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† * Sandia National Laboratories

Motivation

- Validated models and robust modeling tools for process design and scale-up
  R2R industry has greatly benefitted from continuum scale models, and now data-driven models

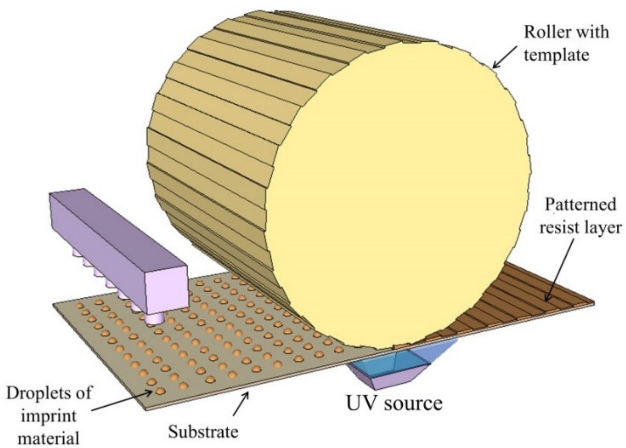
- Guide tool development → reduce iterations
  Assist NM-Fab tool and process design

- Construct defect-free operating conditions
  Operability limits and associated underpinning physical mechanisms

- Foundational understanding of the physics and its underpinning competitions….
  Drives innovation

Challenges in Modeling R2R Nanomufacturing Processes

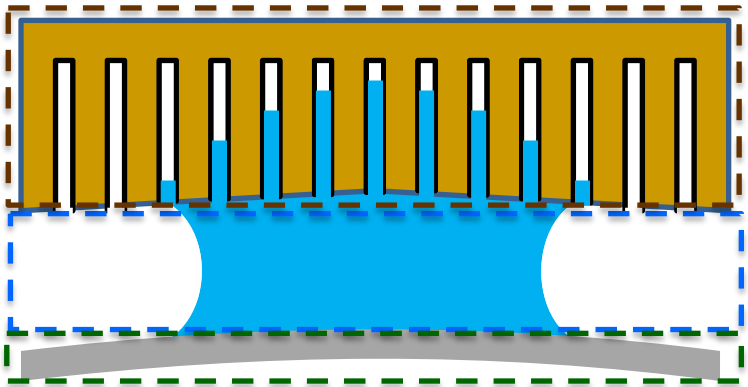
Example: Nanoimprint Lithography



10,000+ drops

Not drawn to scale!

Feature length ~ 50 – 100 nm
Substrate width ~ 1 cm



Template
- Feature flow
- Poroelastic

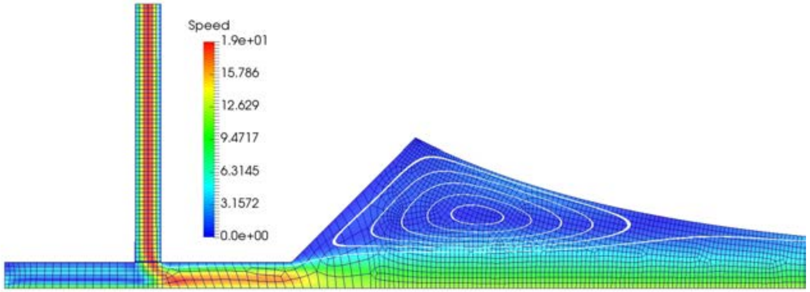
Flow in Gap
- Multiphase flow
- Gas dissolution
- Capillary forces

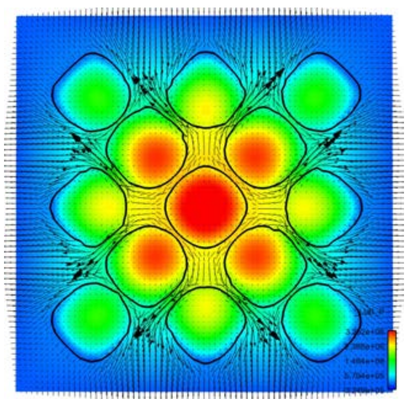
Substrate
- Web tension
- Stiff flexure

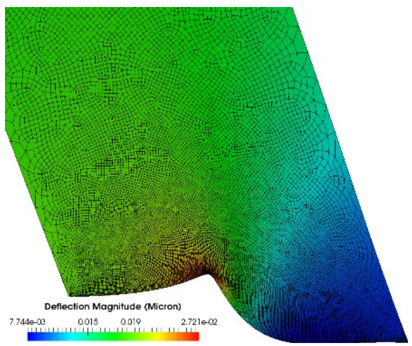
- ***Disparate length scales*** → device/machine scale model needs to be reduced-order
- ***Surface effects***, i.e. nanotopology, forces dominate

Simulation Tool: Goma 6.0

2014 R&D 100 Award Winner



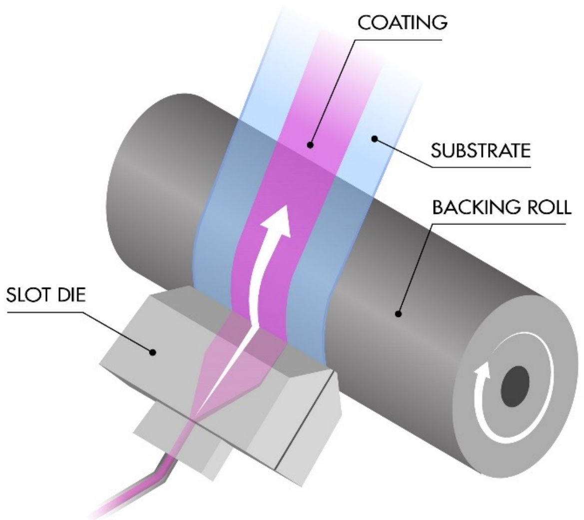


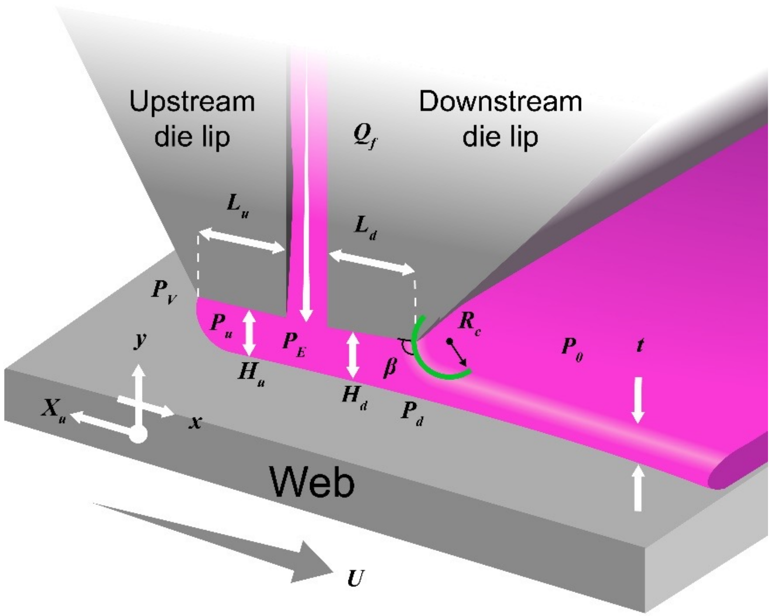


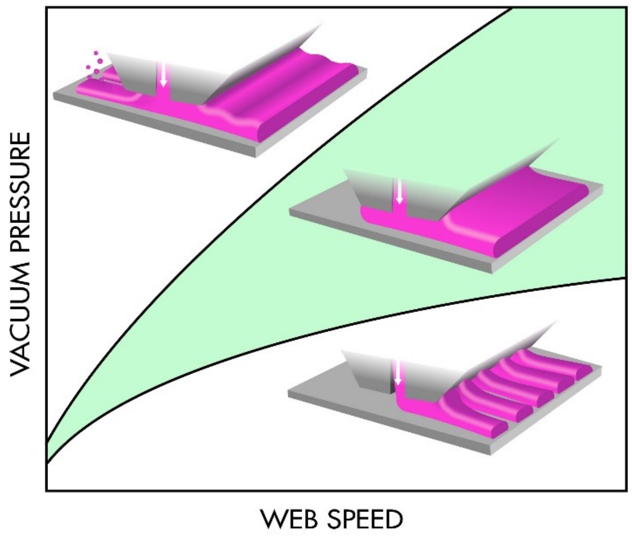
- Multiphysics ***finite element*** code, suitable for both ***research*** and ***production***
- Fully-coupled ***free*** and moving ***boundary*** parameterization – ALE, Level Set, etc.
- Modular code; ***easy to add equations*** – currently has 180+ differential equations
- ***Open source***! Available at http://goma.github.io
- ***Goma 6.0. training*** is available!

Goma has been used successfully in coating manufacturing for 3 decades!

Slot-Die Coating

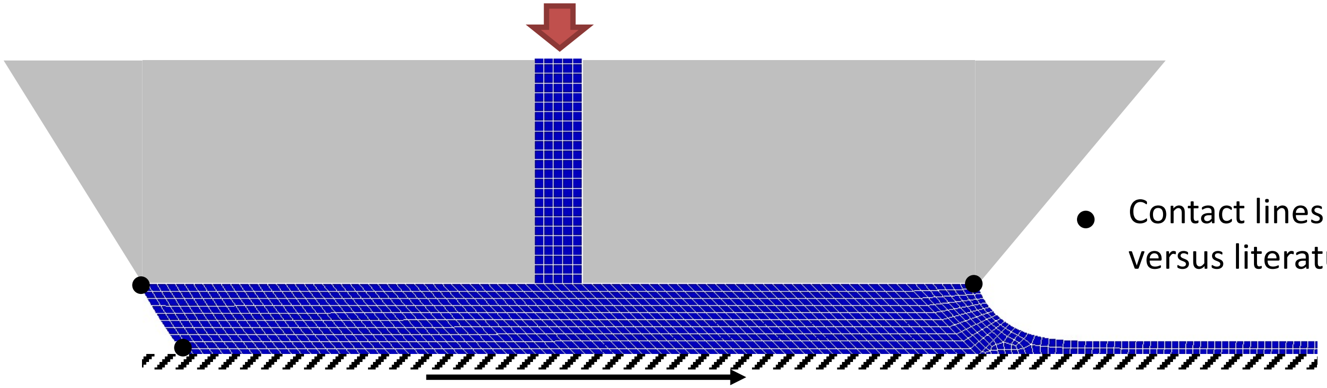






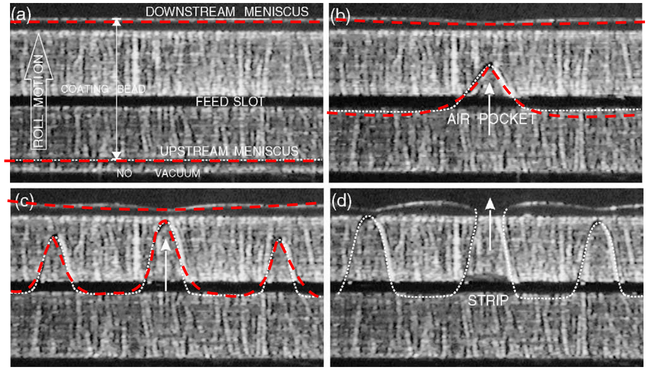
- A method ideal for ***precisely coating single or two layers***
- Thickness is set solely by flow rate and coating speed: $h = Q_f / U$ → ***premetered method***
- Coating quality, i.e. uniformity, depends on liquid properties and operating conditions – ***coating window***
- Goal: ***Predict coating window*** to guide process development

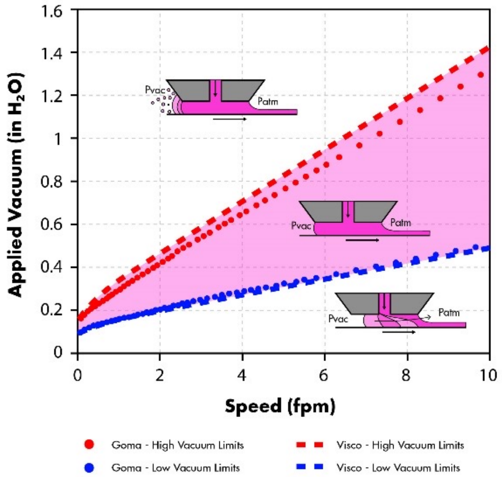
Modeling Approach



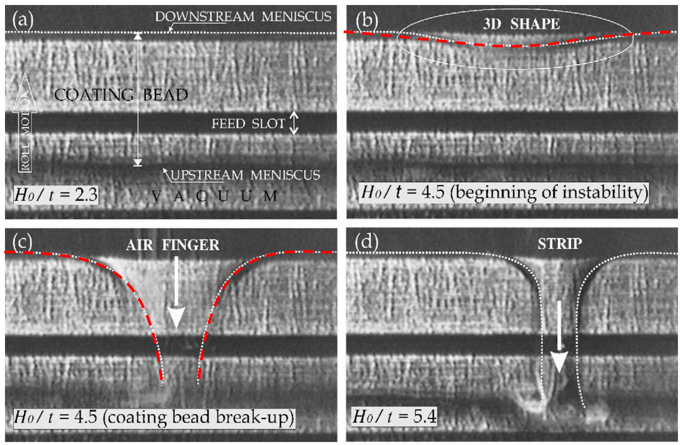
- Contact lines are not pinned; unique aspect versus literature

Low Vacuum Limit





Low Flow Limit

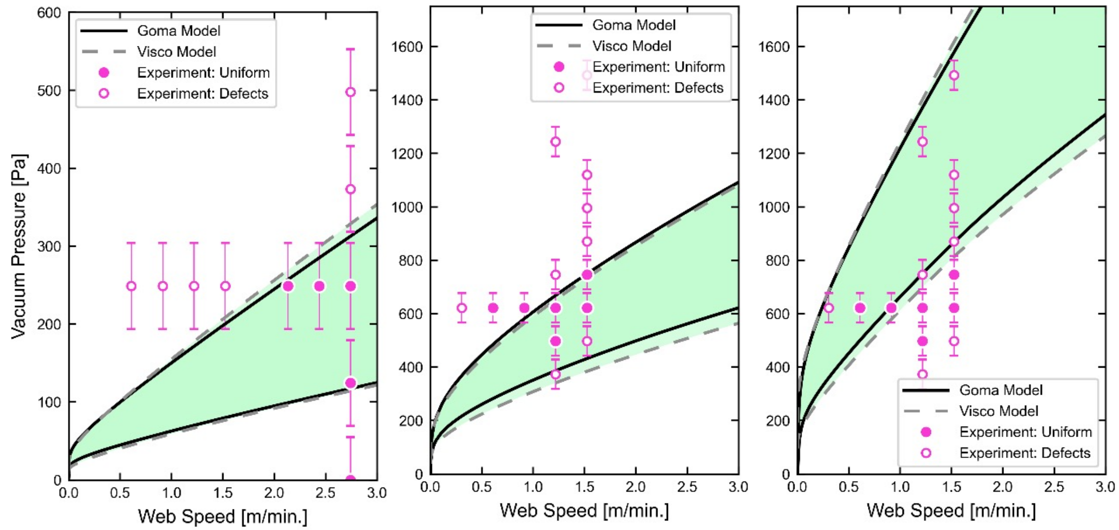


Romero et al. *Journal of Non-Newtonian Fluid Mechanics* 118.2-3 (2004): 137-156.

- 2-D steady state ***Navier-Stokes*** with ***arbitrary Lagrangian Eulerian*** (ALE) method to deform the mesh
- Predict coating window limits based on the ***contact lines positions***

Outcome: Coating Window Prediction

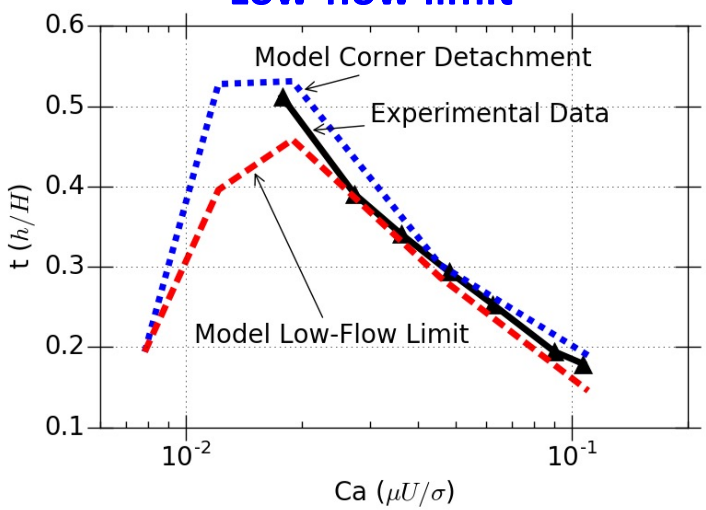
Vacuum limits



Creel et al. *Journal of Colloid and Interface Science* (2021).

- Predicted vacuum limits of slot-coated fuel cell ink (Pt in black carbon support) – shear thinning rheology
- Prediction agrees with experimental observation

Low-flow limit



Malakhov et al. *AIChE Journal* 65.6 (2019): e16593.

- Predicted low flow limits agrees with experimental works reported in literature
- Uncover physical mechanisms of low flow limits with no vacuum
- How to coat thinner: Smaller gap, but limited by roll runout and surface imperfection
- Another approach: Replace backing roll with tensioned web – ongoing research

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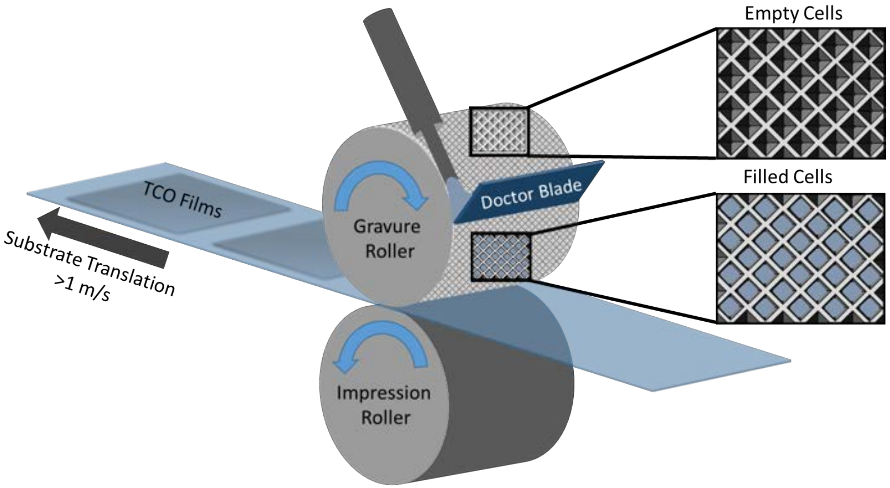
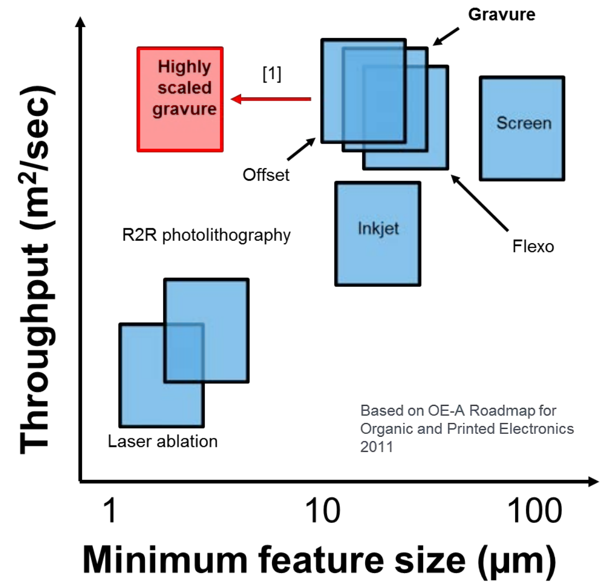
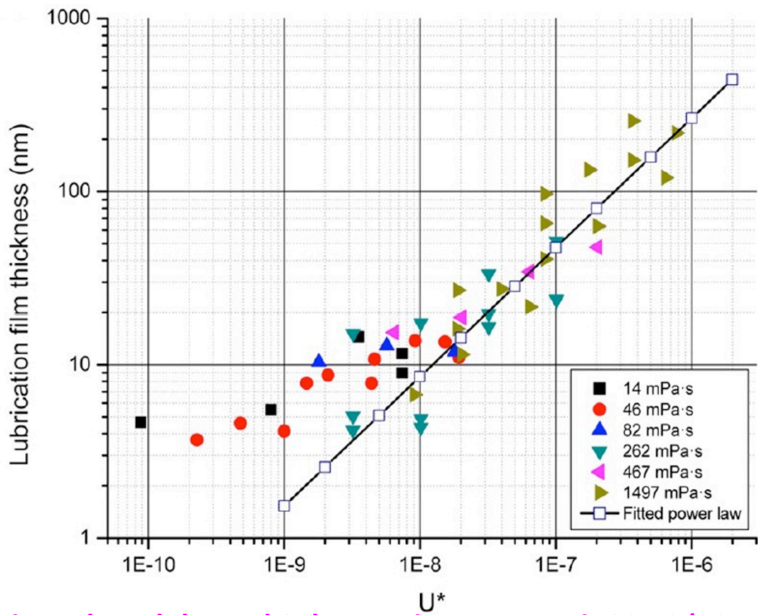
Gravure Printing



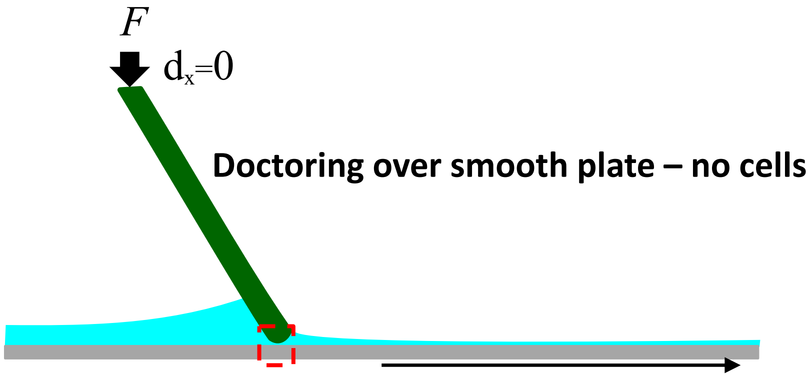


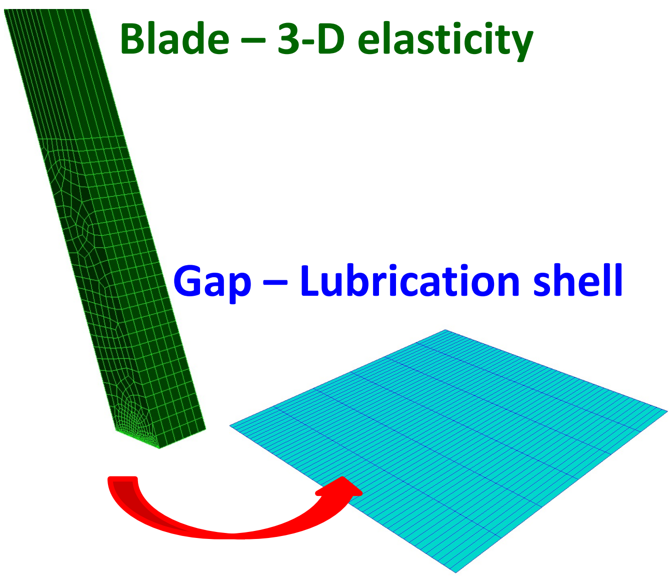


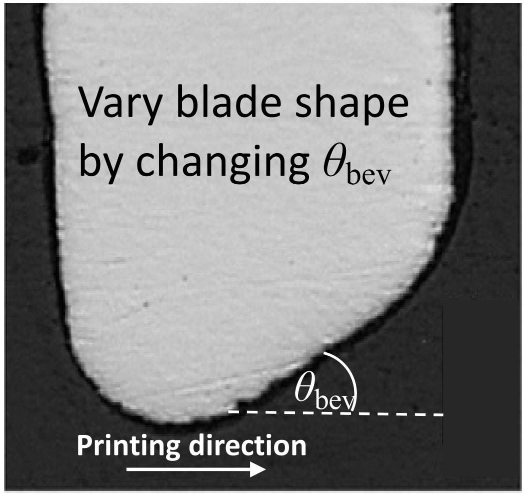
- Suitable for ***high-speed roll-to-roll*** system.
- Challenge: Smaller feature scale, overlay registration.
- Approach: Improve ***doctoring step*** to reduce residual film thickness – ***elastohydrodynamic lubrication*** regime
- Goal: Predict residual film thickness given operating condition as well as liquid and blade properties.

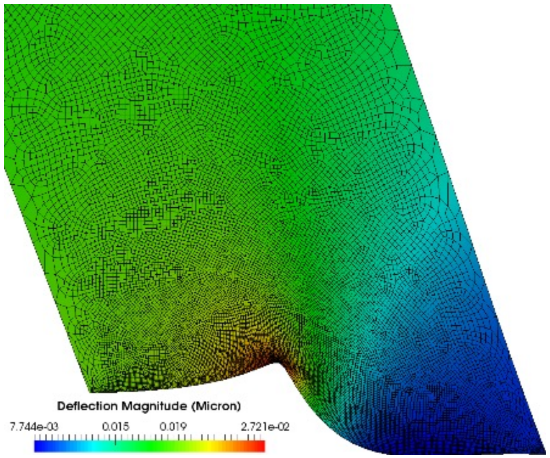
Kitsomboonloha and Subramanian. *Langmuir 30.12* (2014): 3612-3624
Hariprasad et al. *Journal of Applied Physics* 119.13 (2016): 135303.

Modeling Approach



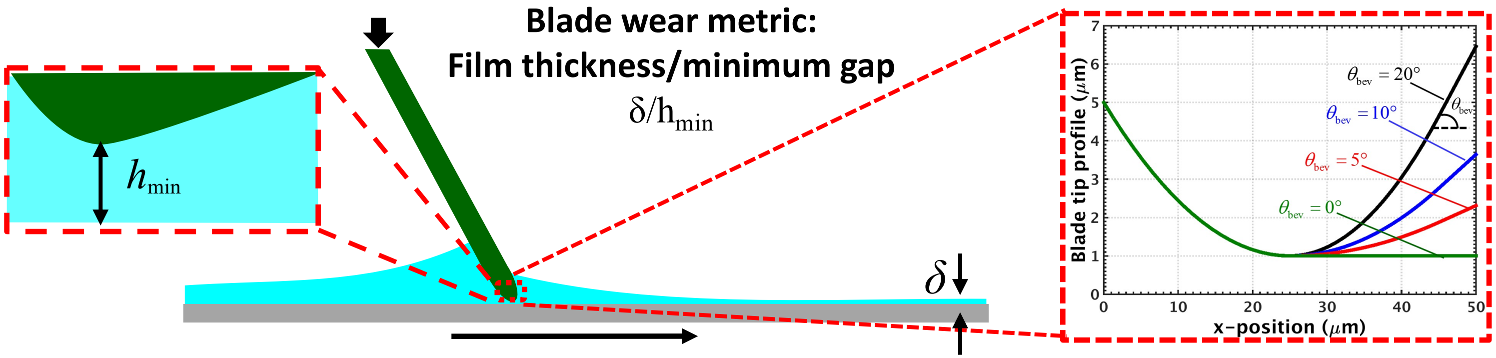


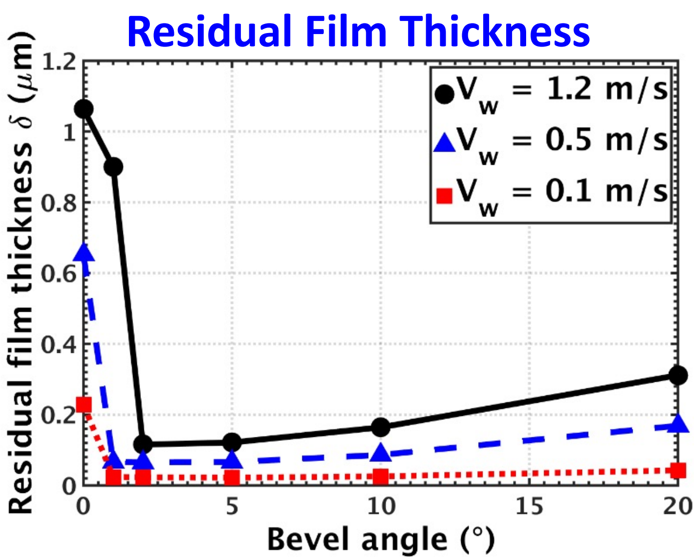


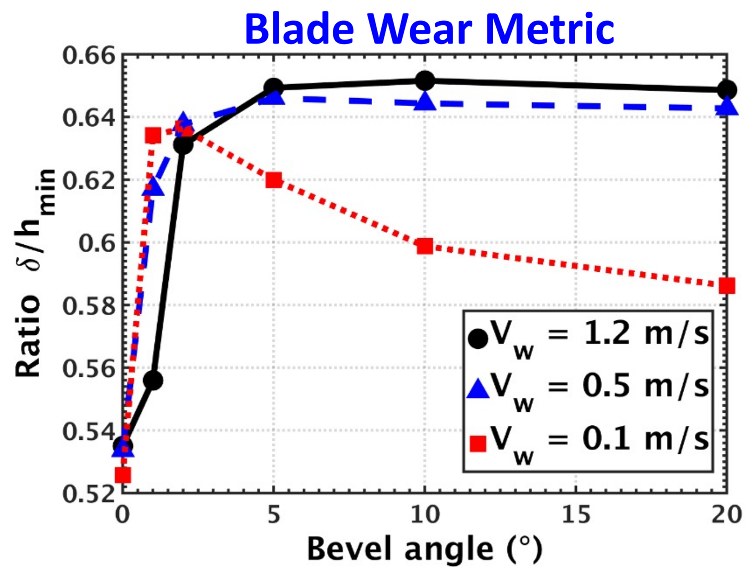


- Fully coupled ***3-D elastic blade*** and ***lubrication flow*** underneath
- Predict residual ***film thickness*** and likelihood of ***blade wear*** as function of printing speed, loading force, and blade shape (θ_{bev}).

Outcome: Predicted Residual Film Thickness and Blade Wear



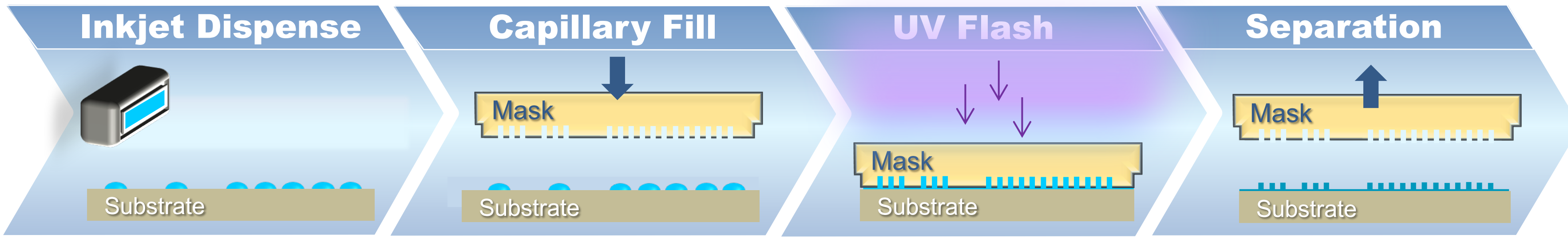




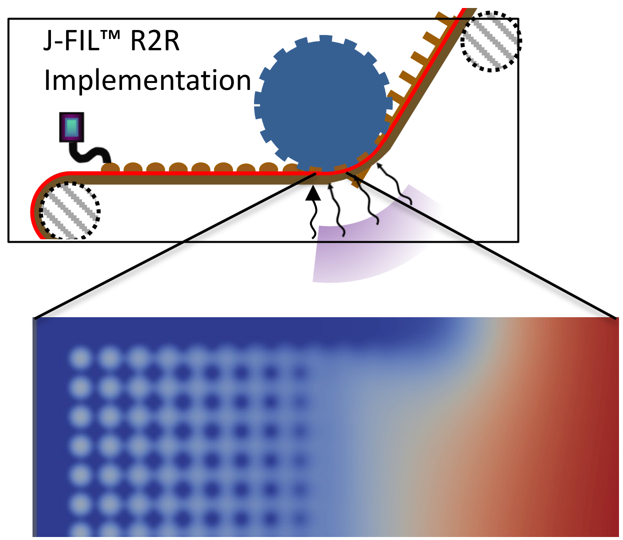
Tjiptowidjojo et al. *Journal of Coatings Technology and Research* 15.5 (2018): 983-992.

- Higher doctoring speed leads to thicker residual film – ***elastohydrodynamic lubrication regime***
- Blade shape ***dictates pressure field → residual film thickness***
- Need to consider blade wear in selecting blade shape ***→ optimum blade shape***

Nanoimprint Lithography



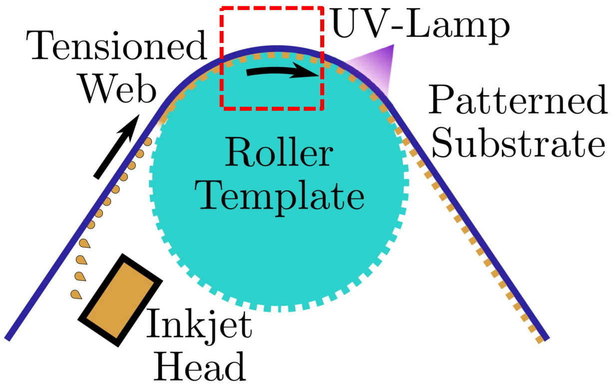
- Dispense Photo-Polymer (Resist) via Ink-Jet – reduces resist volume and residual layer thickness
- Imprint the resist layer with template pattern – drop merging and capillary-driven filling
- UV cure the imprint pattern – photopolymerization
- Separate patterned resist from template – traction separation law



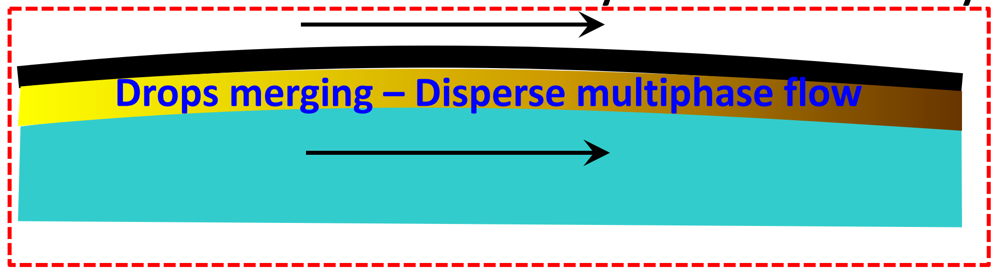
Challenges in drop merging stage:

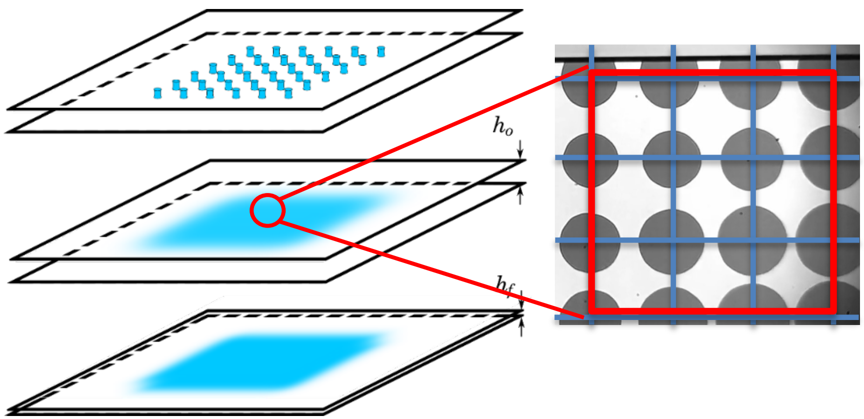
- ➢ Gas trapping (and dissolution) → incomplete filling, voids, longer processing time
- ➢ Substrate and template deformation → ***non-uniform residual layer thickness***

Modeling Approach



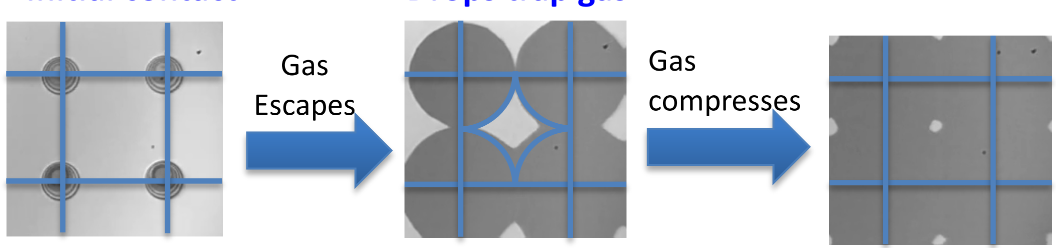
Tensioned web – inextensible cylindrical shell theory



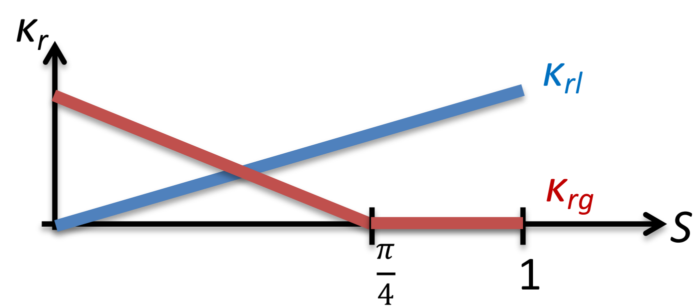


- Representative volume element is a ***drops lattice***
- Flow dynamics is governed by ***relative permeability*** of each phase – presence of one phase impedes flow of the other phase
- Surface tension forces applied via ***effective stress principle***

Initial contact | **Drops trap gas** | **Gas dissolves**

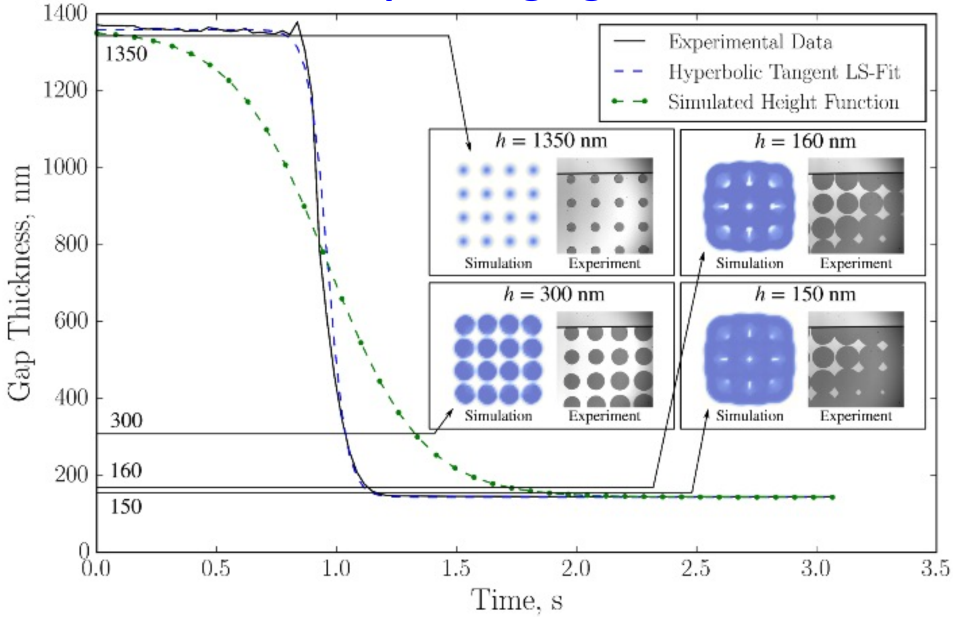


$h_o \rightarrow S < \frac{\pi}{4}$ | $h_c \rightarrow S = \frac{\pi}{4}$



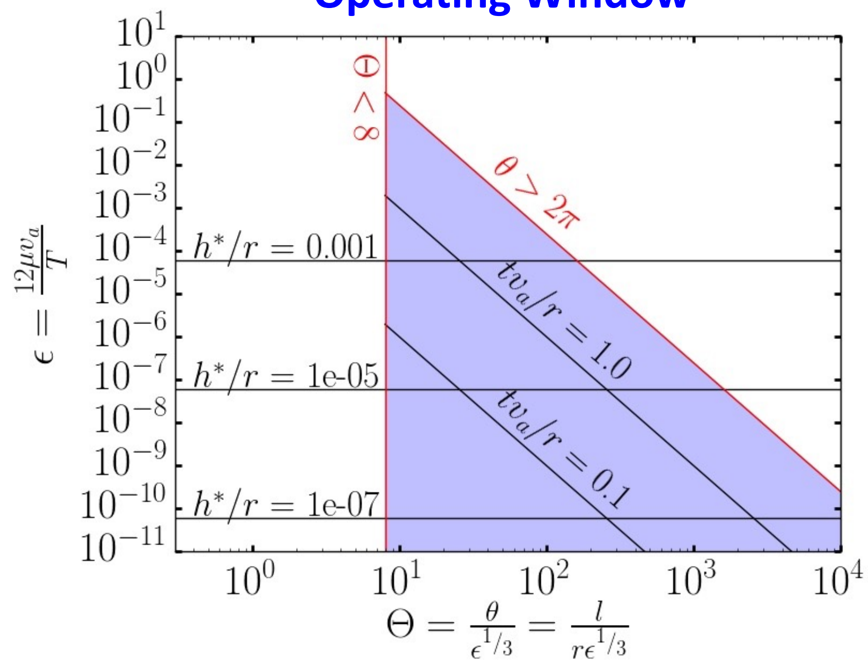
Outcomes: Drops Merging Rate and Operating Window

Drops Merging Rate



Cochrane et al. *International Journal of Multiphase Flow* 104 (2018): 9-19.

Operating Window



Cochrane et al. *Industrial & Engineering Chemistry Research* 58.37 (2019): 17424-17432.

- Predict ***rate of drops merging*** as well as gas trapping (and dissolution) as a function of roll closure speed and fluids (liquid and gas) properties
- Predict ***minimum and maximum wrapping angles*** required to maintain uniform residual layer thickness – needed to estimate UV-cure residence time.
- Next step: Incorporate flow and structural mechanics at the template

Conclusions

- Modeling tools developed under NASCENT has produced simulators for sheet-to-sheet or R2R gravure printing, precision slot die coating, and R2R imprint lithography.
  - *Operability limits can be determined for slot-die and imprint litho*
  - *Models were experimentally validated for slot die coating and gravure wiping*
  - *Models were qualitatively (through visualization) validated for imprint lithography*

- Continuum models for R2R nano-manufacturing challenged by

  -Large aspect ratios in full scale process simulations

  -Multi-physics (structural, capillary/surface, fluid mechanical)

  -Wide range of time scales (ms from capillary to seconds at process scale
- Reduced order models essential for thin structures/regions
- Multiphase flow models essential for printing and imprinting (at scale)

- Future work: Include template features in imprint litho models; 3D web-mechanics capabilities; particle effects