AMM-R2R - Applied Materials Genome Initiative - From Ideal Materials to Real-World Devices

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AMO Peer Review
Poster Presentation
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Overview

Timeline

- Project start date: January 2016
- Projected end date: (transitioned in FY 19)
  - Core Projects: September 2021
  - CRADAs: October 2019*
- Project completion for FY18
  - Core Projects:100%
  - CRADA Projects: 40%
- Project completion for FY19
  - Core Projects: 30%
  - CRADA Projects: ~80%

Barriers*

- Continuous processing
- Registration and alignment challenges
- Materials compatibility
- Stoichiometry control
- Availability of materials data

* AMO MYPP for FY 2017-2021, June 2017 draft, section 3.1.8

Budget

- FY 18
  - Core lab work: $1,257K per year
  - CRADA work: $1,150K plus $1,150K non-federal cost share
- FY19
  - Core lab work: $3,000K

Partners

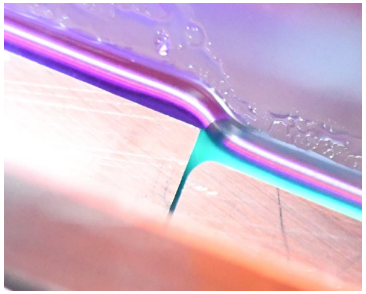
- ORNL, ANL, NREL, LBNL, SNL
- Navitas Systems
- Fisker, Inc.
- SolarWindow Technologies, Inc.
- Proton OnSite*

* Pending contract award

Project Objective(s)

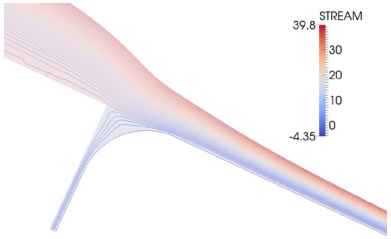
- This project focuses on connecting macroscopic properties (such as viscosity and drying time) of particle suspensions to interactions at the particle scale.
- Roll-to-Roll Processing, an Advanced Manufacturing Technology Area within the AMO MYPP, typically involves depositing particle suspensions on moving substrates.
- Reducing suspension processing energy (such as in film drying) and improving film consistency supports the AMO goal of improving productivity and energy efficiency in manufacturing.
- Process models of pre-metered, coating-flow-deposition, capillary-hydrodynamics have a proven track record of accelerating process development
- We are developing complementary experiments and models in order to understand fundamental influences on macroscopic suspension behavior. This understanding can then be used to engineer desired behavior by tuning component properties.

Objectives (cont.)

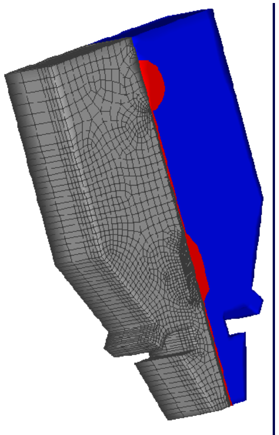


- Models of consortium's slot- and slide-deposition systems with accommodation for 3+ simultaneous miscible layers has no precedence (multiple layers of varying rheology)

- Modeling and simulation of the coating process, once validated, will enable process operating window optimization with respect to key parameters (layer flow rates, web-speed, slide/slot configuration, ink-design, etc.



Overall Objectives: (1) Influence NREL's and ORNL's process designs for multilayer slide- and slot-coating by Q4 FY20, (2) Mod/sim tools suitable for wide use by the energy coatings industry for multilayer coating



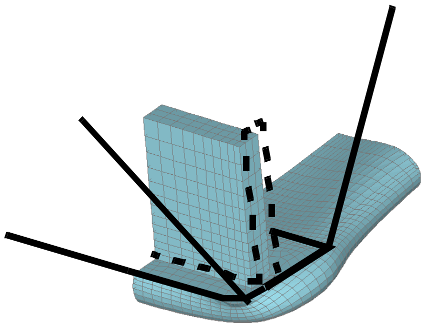
Technical Innovation

At present, suspension processing, coating, and drying procedures are typically developed through trial-and-error, making optimization costly.

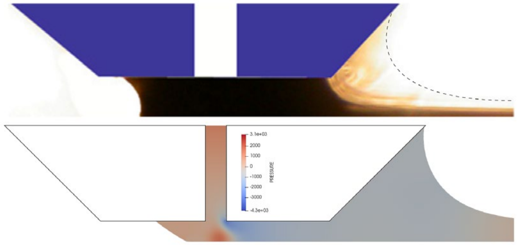
- Fundamental understanding and tools developed in this project could be used to engineer macroscopic suspension behavior to improve energy efficiency and productivity.
- Our new experimental setups will permit observation (by multiple techniques) of automated, consistent processing steps in tightly-controlled environments.
- Current commercial (*e.g*. COMSOL) and open source software tools (*e.g*. Goma 6.0) are available as foundational software, but no analyst workflows specific to multilayer coating exist. *Challenge*: Capillary hydrodynamics, complex rheology, and free and moving boundaries make the problem highly nonlinear

Technical Innovation (cont.)

- Our approach deploys Goma 6.0 as a software platform that has 20+ years development for coating and related manufacturing process flow modeling.
  - Model results, once validated allow for process window stability prediction, defect prediction, and factor-space effects of ink rheology, coating configuration, etc. *Reducing defects and widening the process windows for thinner films and faster rates translates to $$ savings*
  - Innovative ways of deploying robustness measures, including automatic meshing/remeshing, contact-line migration will greatly enhance the user-base.
- General, wide-user-base models will be distributable (with training and workflows) for process designers in the coating industry.



3D Slot-die coating edge bead



Validation of single-layer slot coating deposition (flow visualization in literature)

Technical Approach

Approach

- Develop complementary automated experiments and models to understand connection between macroscopic particle suspension behavior and particle-level component interactions.
- Build base models and workflows on Goma 6.0 (goma.github.io)
  - Finite element solution of the governing partial differential equations of capillary hydrodynamics with free and moving boundaries.
  - Integrate complex rheology constitutive equations representative of fuel-cell, battery coating inks/slurries
  - Permit free motion of contact lines across die faces, lands, and substrate, with remeshing/remapping

Team roles

- LBNL provides experience with suspension processing, suspension drying experiments, model development, mechanical design, will produce models of suspension behavior based on particle-level interactions
- SNL provides experience with high-resolution continuum simulation of non-Newtonian flows in complex geometries, will incorporate LBNL suspension models into flow simulations

Technical Approach

Risks, unknowns, and mitigation

- Unforeseen difficulties in measurement
  - Employ multiple measurement techniques with overlapping capabilities, use automation to improve reproducibility in repeated experiments
- Distribution and interaction of components in suspension is unknown
  - Early experiments will be designed to provide qualitative information about distribution to inform model framework; model parameter values will be determined in later experiments
- This is a first ever model development using moving-mesh approach of three-layer slide-coating flow. All coating flows are highly nonlinear, and these additional complexities will be managed with modern meshing/mapping tools.

Challenges

- Influence of disorder on suspension behavior will be difficult to capture with simple representations
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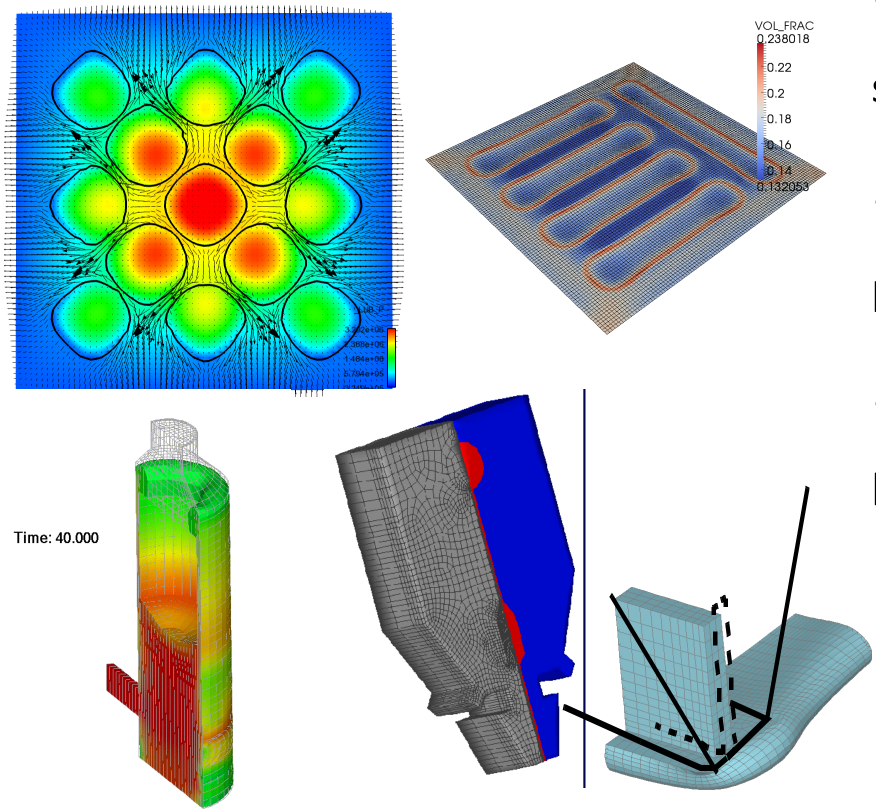
Technical Approach – Deposition modeling



- Goma 6.0 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 170+ differential equations
- *Open source*! Available at **http://goma.github.io**
- *Goma 6.0. training* is available on a regular basis

Goma has been used successfully in coating manufacturing for 2 decades

Results and Accomplishments

- FY19: Developed model connecting carbon black / PVDF / NMP suspension viscosity to particle-scale interactions, fulfilling initial project goal

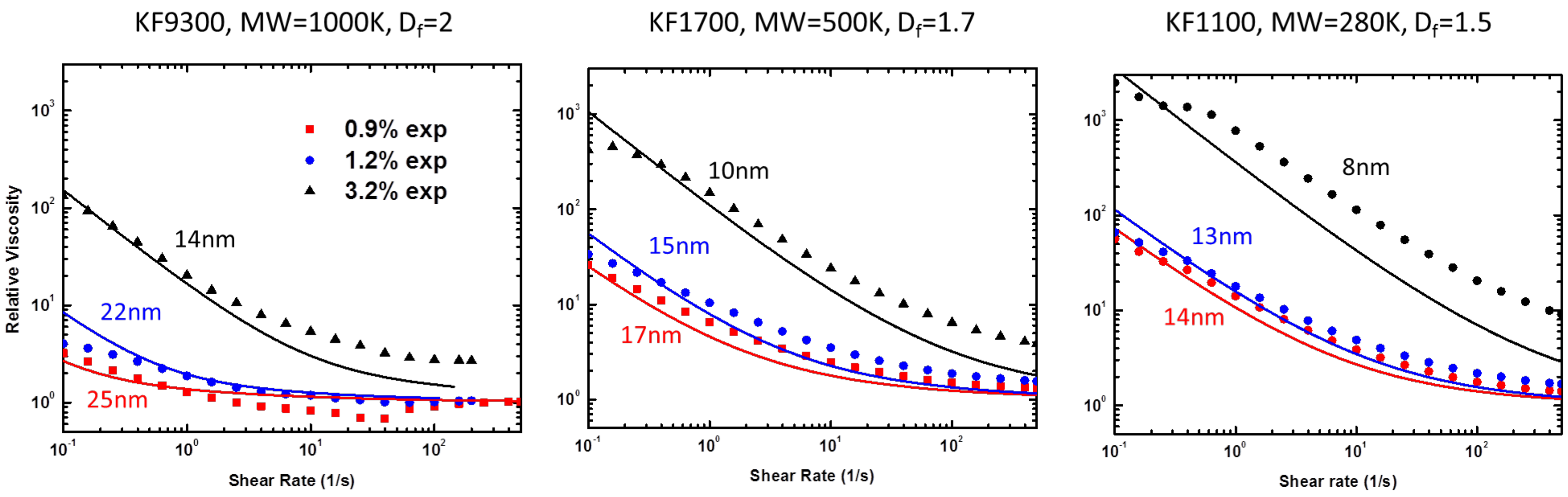
$$\eta_{total} = \eta_{hydr} + \eta_{struct}$$

Potanin, et al, J. Chem. Phys. 102(14) 1995

Kim et al, Polymer, 2015, 7(7), 1346-1378

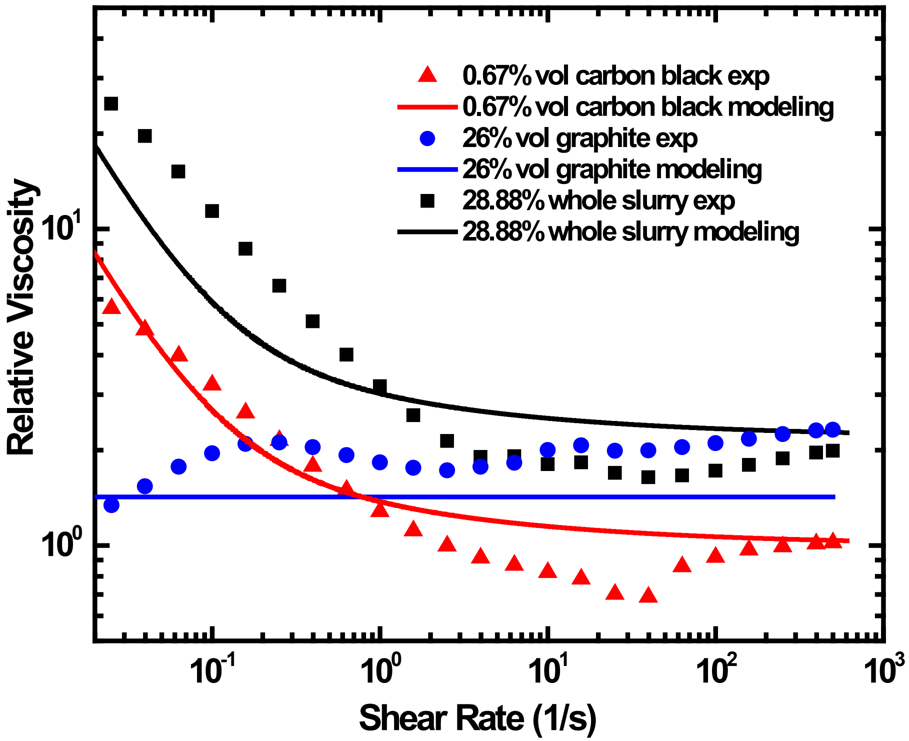
Krieger IM, Dougherty TJ. Trans Soc Rheol 1959;3:137.

- Fitting parameters
  - polymer coating length
  - fractal dimension of aggregates



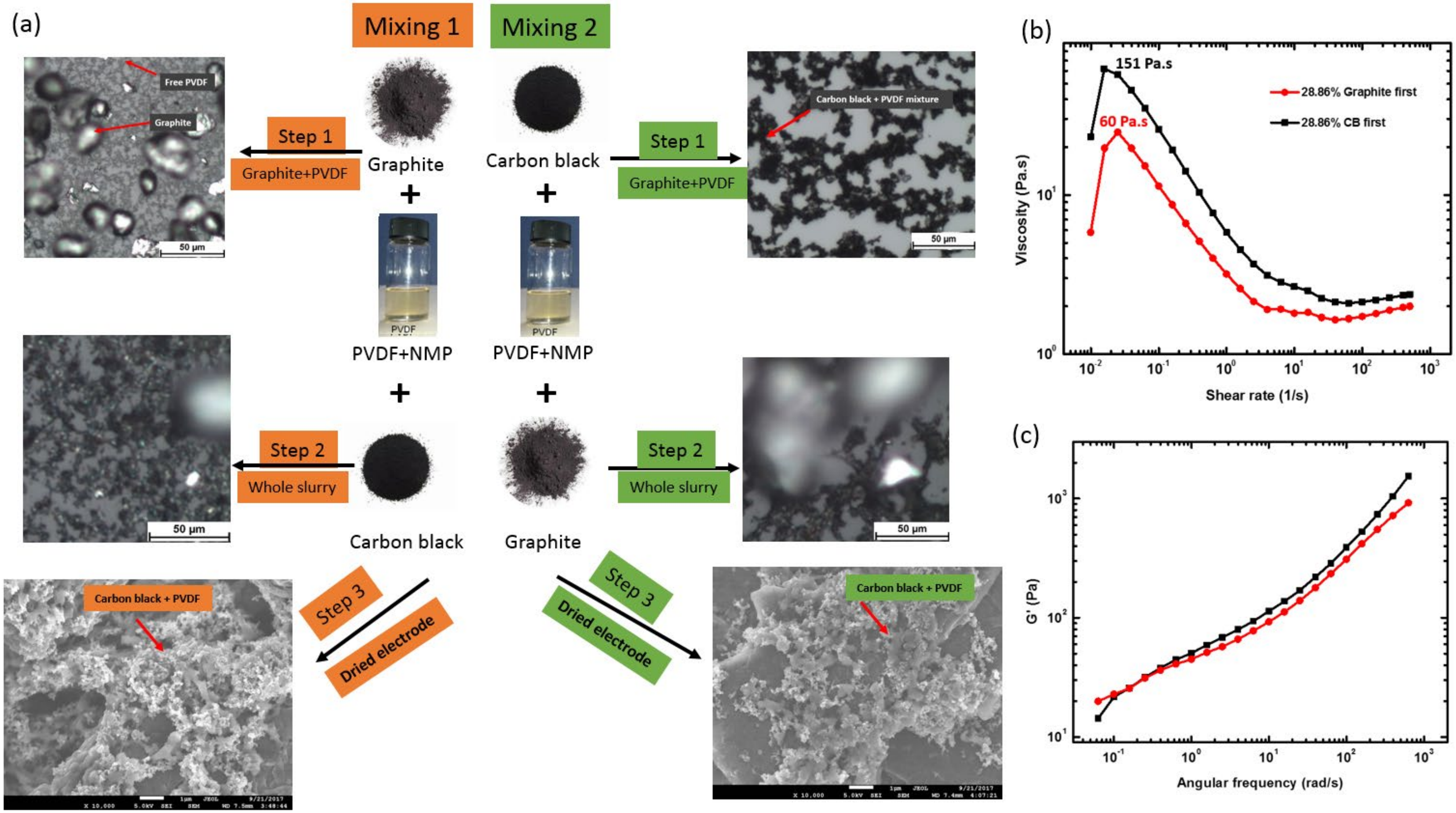
Results and Accomplishments

- FY19: Used Krieger-Dougherty approach to extend model to particle suspensions for graphite battery anodes (graphite particles, carbon black, PVDF, and NMP), obtaining reasonable experimental agreement



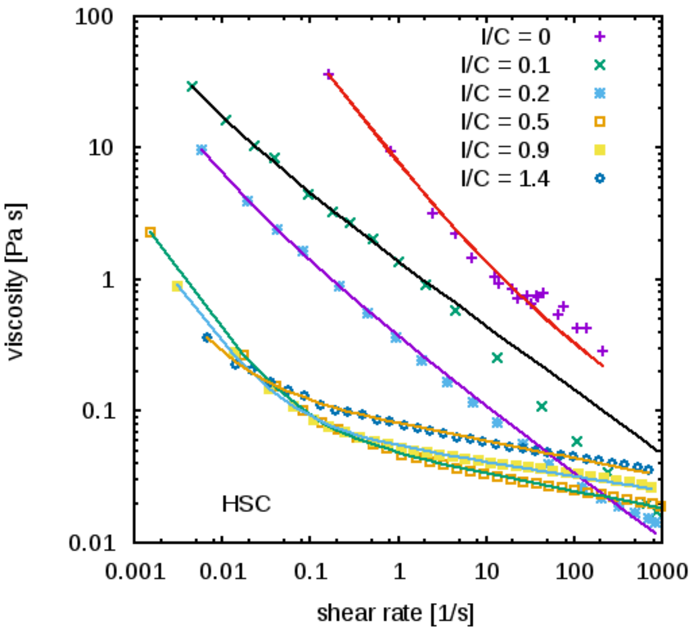
Results and Accomplishments

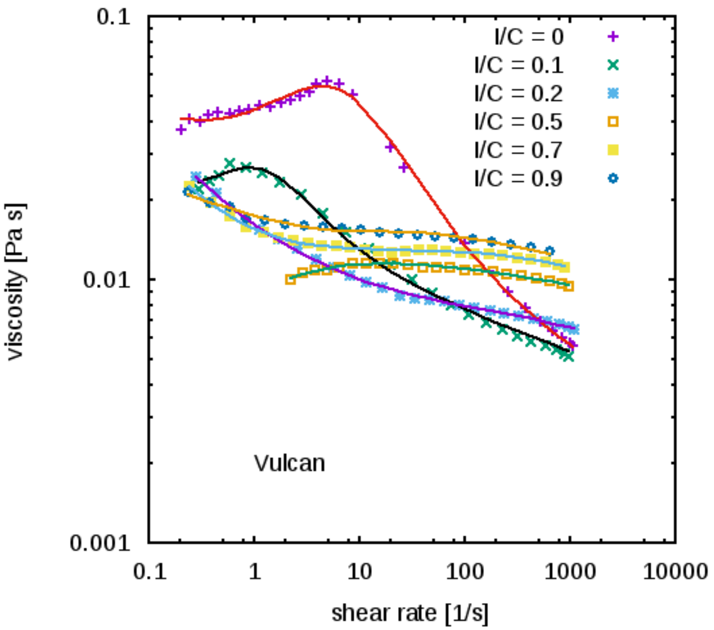
- FY19: Determined that mixing sequence affects distribution of PVDF in particle suspensions with graphite particles



Results and Accomplishments

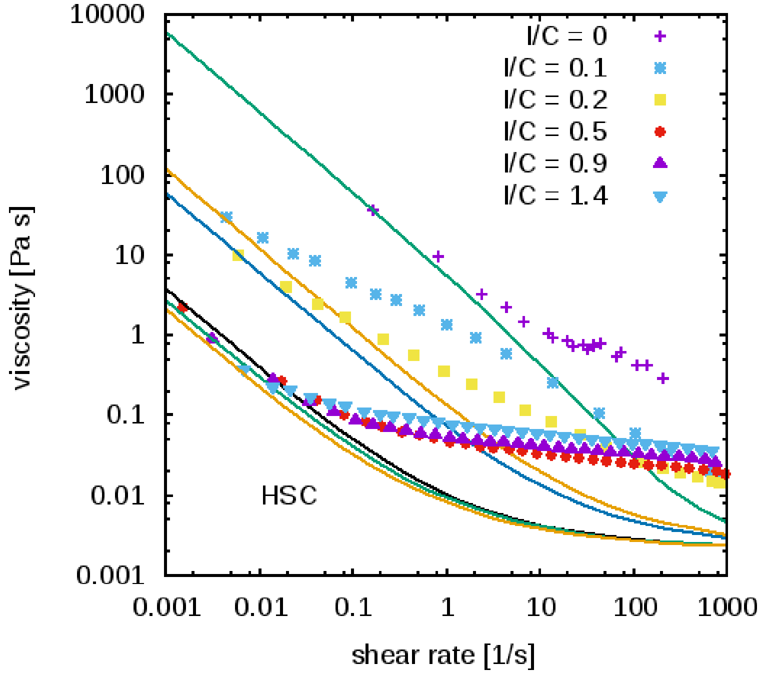
- FY19: Evaluated candidate mathematical forms for viscosity models describing NREL fuel cell inks
  - Found suitable forms that retain low shear-rate description from battery slurry model
  - HSC: $\eta = \frac{A}{\gamma} + \frac{B}{\gamma^C}$ Vulcan: $\eta = \frac{A}{\frac{D}{\gamma^E} + \gamma} + \frac{B}{\gamma^C}$





Results and Accomplishments

- FY19: Determined that high-shear term of micro-rheological model used as basis for CB/ PVDF / NMP suspension model cannot account for even the simpler NREL HSC suspension form $\eta = \frac{A}{\gamma} + \frac{B}{\gamma^C}$
  - Alternative sub-models must be considered



Results and Accomplishments

Planned for FY20 for rheology models (delayed funding start):

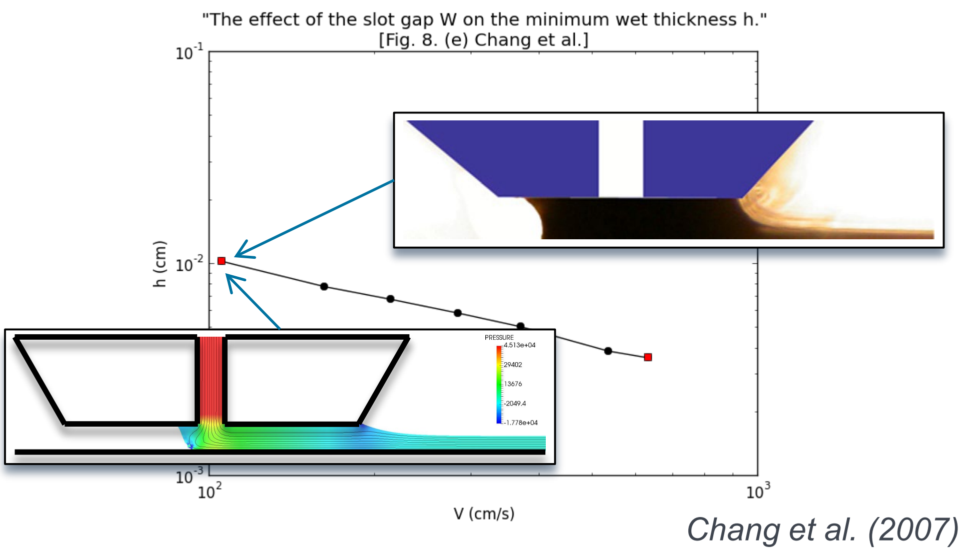
- Perform a few initial experiments to determine important qualitative aspects of suspension drying behavior to inform major decisions in model framework development
- Begin development of drying model for single film layer containing uniformly distributed particles
- Begin development of automated microscopic equipment for visualizing drying experiments

Results and Accomplishments-FY19

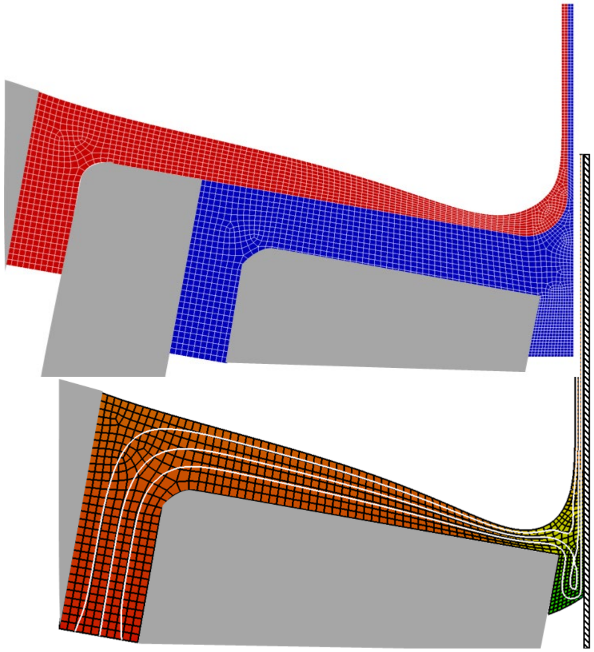
Progress to date of coating models:

- Build and test base models for multi-layer coating with relevant die configurations with Newtonian rheology
- Build and test base models for multi-layer drying/solidification
- Complete multilayer flow-process model validation for chosen test system
- Q4 SMART milestone; criteria: demonstrate 3-layer simultaneous coating, with the thickness of each layer from 5-20 µm, at a speed of at least 1 m/min)

Results so far:



<u>Single-layer slot-die deposition model validation: Pattern of streamlines (lower left) and published flow visualization</u>



<u>Two-layer and single-layer predicted pattern of flow and free surface shape</u>

Transition

- The Kodak R-2-R user facility will be used to demonstrate advances in casting and drying derived from the multiple aspects of the program.
- The experimental apparatuses developed in this project, along with the software tools containing mathematical models of slurry rheology, casting, and drying will be offered to industrial partners (still to be determined) to optimize their specific systems of interest.
- Next generation of *Goma* will be established and open to the public.