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Dynamic wetting of cellulosic substrates by model liquids

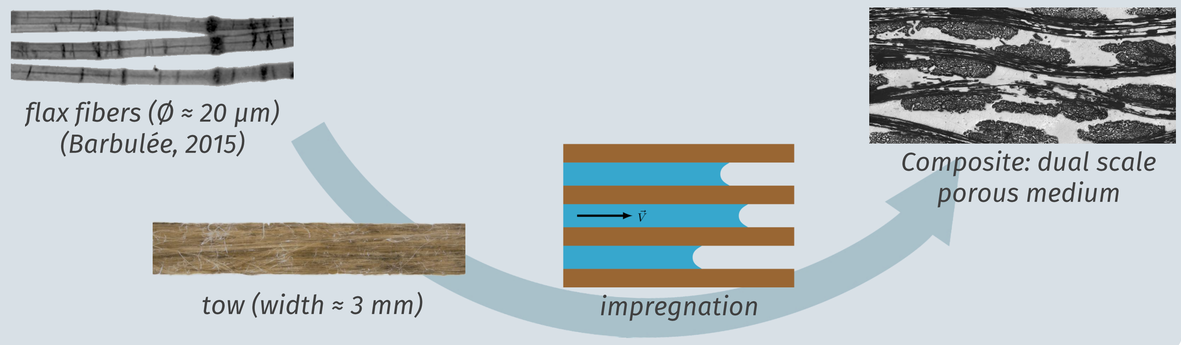
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Motivation

The processing of **flax-polymer composites** is a **multiscale** problem, requiring an accurate description of **capillary flows** occurring inside and between the tows. Since natural fibers exhibit complex structure, **model materials** are used to implement the processing mechanisms. The main goals are:

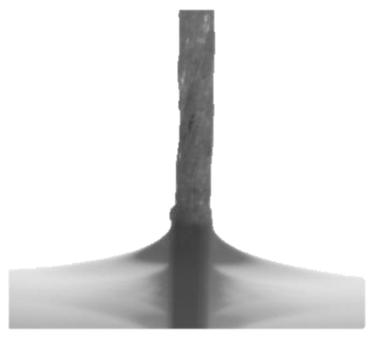
- modeling dynamic wetting phenomena for cellulose-polymer systems
- assessing the influence of physical parameters on dynamic wetting regimes
- evaluating the accuracy of existing models



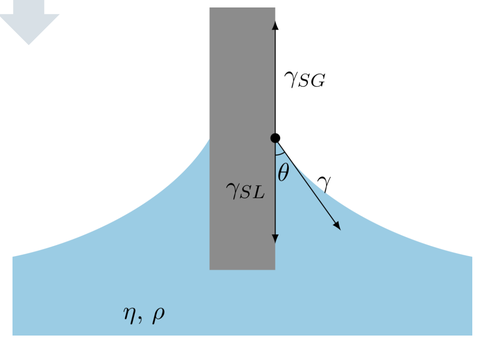
Dynamic wetting models

Characterized by the (dynamic) ***contact angle*** *θ ⇨ unbalanced capillary forces*

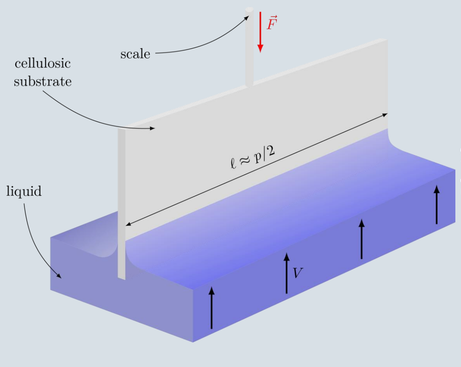
Model family	Hydrodynamic (HD)	Molecular kinetics (MK)	Combined (MK-HD)
Main parameters	viscosity	intermolecular forces	viscosity + intermolecular forces
Example	*Cox-Voinov* $\theta^3 - \theta_e^3 = 9\Gamma \mathrm{Ca}$	*Blake (small angles)* $\theta^2 - \theta_e^2 = \mathrm{Ca}\left(\frac{\upsilon N k_B T}{hK\lambda}\right)$	*Petrov* $\theta^3 = \theta_c^3(\mathrm{Ca}) + 9\mathrm{Ca}\,(\Gamma - C_V)$



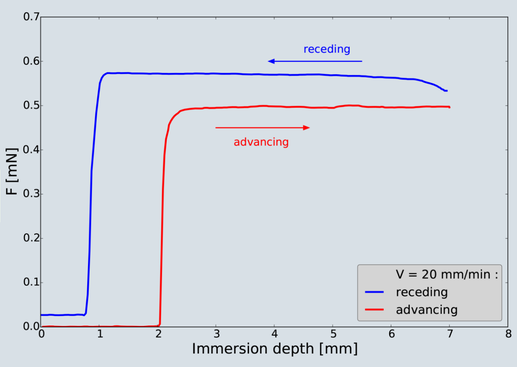
water meniscus on a flax tow (Pucci, 2016)



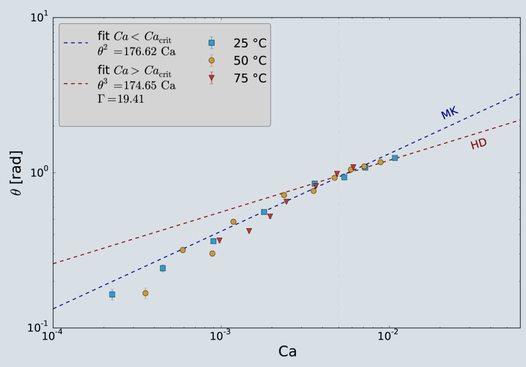
Preliminary study



Principle of the Wilhelmy plate experiment



Results : capillary force measurement



Evolution of θ with the contact line velocity of paraffin oil for different temperatures

- **Wilhelmy method** (force measurement) on a **cellulosic substrate** for:
  - Paraffin oils 'total wetting)
  - PEG 1450, 3350 and 10000 (partial wetting)
  - Different temperatures (25, 50, 75 °C)
  - Forced wetting: multiple bath velocities (10^{-4} < Ca < 10^{-1}, advancing)
- Materials **characterization**
  - Surface tension Y, viscosity η, density ρ (liquids)
  - Roughness, composition (substrates)

A microbalance measures the resulting **force** *F*, thus the **contact angle**:

$$F = \gamma p\,(\cos\theta - \cos\theta_e)$$

Velocities are expressed with the non dimensional **capillary number** $\mathrm{Ca} = \frac{\eta V}{\gamma}$ and plotted against θ for each temperature.

High Ca data is fitted with the **Cox-Voinov** law, giving an estimate for Γ.
We find a **master curve**. The slope change (½ to ⅓) around a critical value Ca = 5×10^{-3} shows a transition between MK and HD regimes.

Numerical study

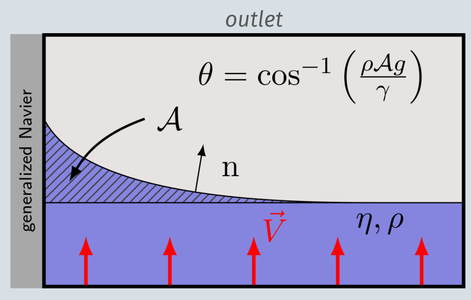
The Wilhelmy plate is reproduced in a **2D FEM** simulation using COMSOL Multiphysics:

- Mesh and parameters are validated (comparison with analytical results)
  - Washburn's law in capillary tubes
  - Spreading of a droplet
- Geometry is wide enough (7 to 8 capillary lengths)
- Materials properties are known (experimental work)
- **Level set** function ϕ (diffuse interface) separating the fluids
- Inlet velocity (Ca) and temperature sweep

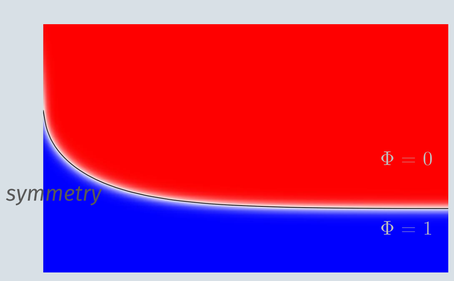
Boundary conditions:

- Symmetry (right), velocity (bottom), no normal stress (top)
- Contact line: generalized Navier slip (*interface formation model*): $n \cdot P\,(I - nn) + \frac{1}{2}\nabla\gamma = \beta\left(v_\parallel^L - v_\parallel^S\right)$

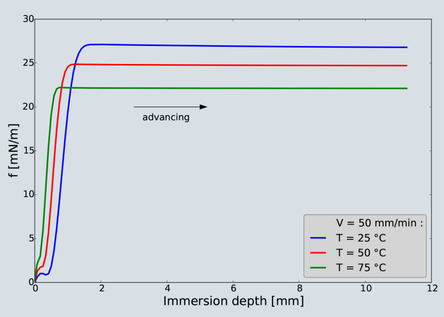
Meniscus area is measured (matching the experimental method) to compute θ.
Above-critical Ca values are fitted with the Cox-Voinov law (HD)
The accuracy of the numerical model is assessed by comparing $\Gamma = \ln\left(\frac{\ell_{max}}{\ell_{min}}\right)$ and its experimental value.



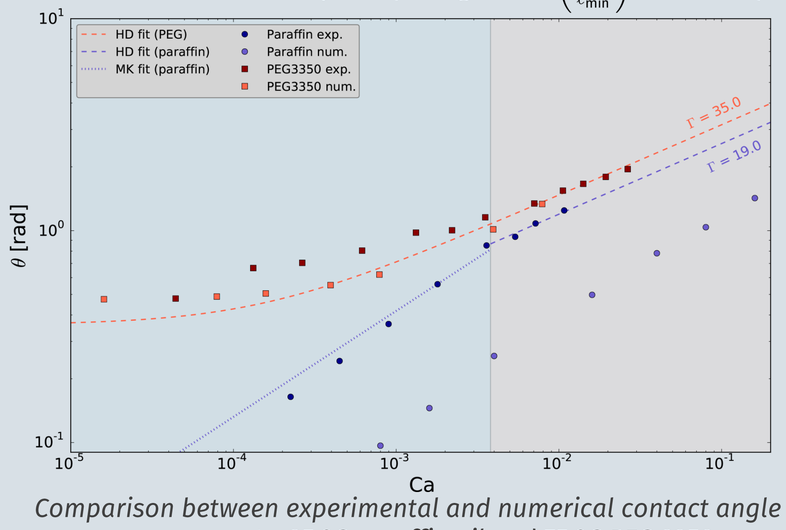
Numerical Wilhelmy plate model setup



Meniscus formed by a paraffin oil for T=25 °C and V = 50 mm/min after 10 s: level set function Φ



Advancing contact line: evolution of the meniscus weight per unit length



Comparison between experimental and numerical contact angle measurements: 25 °C paraffin oil and 75 °C PEG 3350

The wetting behavior is reproduced. The partially wetting PEG 3350 is in good accordance with the experimental data.
An important shift appears for the totally wetting paraffin oil: while the experimental value of Γ is 19, the numerical model yields a value of 1.9
Further experiments indicate that the model needs a finer mesh for an accurate simulation of totally wetting systems.

Acknowledgements

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Future work

- Finer mesh for total wetting
- Adaptive meshing
- Influence of molar mass
- Cylindric geometry (fibers)
- Larger scale (tow)
- Surface rugosity

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