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**This highlight refers to the PhD Thesis by Thejas Hulikal Chakrapani (2022)**

# Mesoscale modelling of multiphase flow and wetting

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## Introduction

In the PhD thesis by Thejas Hulikal Chakrapani (2022) wetting and multi-phase flow problems pertinent to ink-penetration in paper are studied using computer simulations. Jetting ink is composed of water, glycerol and constituents such as colourants, surfactants and polymers; the physico-chemical interactions of these components renders ink complex. Paper is a complex porous solid manufactured by compressing cellulosic fibres, characterised by irregular pore shapes, a broad pore-size distribution and (de)swelling of fibres upon (de)hydration. The complex interaction between ink and paper gives rise to interesting problems, several of which we investigate by systematically increasing the complexity of the fluid, the complexity of the solid, or both, in highly idealised models.

## Simulation methodology

Many-body Dissipative Particle Dynamics (MDPD) is a coarse-grained particle-based simulation technique used to resolve flow at mesoscopic length scales from nanometer to micrometer. The particles, each representing a collection of fluid molecules, interact through short-ranged conservative forces, friction forces and fluctuating Brownian forces obeying the fluctuation-dissipation theorem. Thermodynamic properties such as viscosity, surface tension and (de)mixing of fluids emerge as natural consequences of these forces. Rigid bodies such as colloids and walls are implemented by introducing harmonic springs between particles or by restraining particles to fixed positions.

### A complex fluid in a simple geometry: the imbibition of fluid mixture into a pore

The capillary imbibition of cylindrical pores by fluids is investigated (see Hulikal Chakrapani and den Otter, 2020).  A seminal result describing this process is the Bell-Cameron-Lucas-Washburn (BCLW) theory: the imbibition depth increases with the square root of time. Quantitative agreement with BCLW theory is observed for pure fluids and fluid mixtures, provided the imbibing fluid has a small equilibrium contact angle and its viscosity is sufficiently high. The imbibition of partially wetting fluids and fluid mixtures is modestly enhanced by slip at the wall. Binary mixtures of fluids differing in their affinities to the wall show partial segregation during imbibition: the wall-philic component forms a monolayer covering the wall, while the wall-phobic component is over-represented in the centre of the pore. Since the flow velocity is higher in the centre, the wall-phobic component is carried – on average – further into the pore than the wall-philic component and consequently dominates at the imbibition front, as illustrated in Figure 1. This enrichment of the wall-phobic component at the front is of non-equilibrium origin, and gradually disappears after cessation of the flow; the enrichment of the wall-philic component at the walls is of thermodynamic origin and persists in the absence of flow.

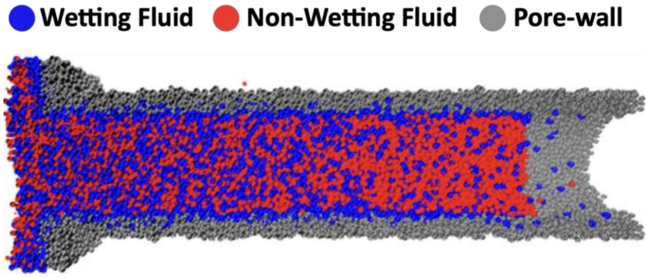

*Figure 1. Simulation snapshot of the spontaneous imbibition into a cylindrical nanopore of a binary liquid mixture, composed of equal concentrations of perfectly wetting and non-wetting fluids.*

**A simple fluid in a complex geometry: the permeability of micro-pillared channels**

Darcy's law describes the volumetric flux of a Newtonian fluid flowing through a bulk porous media as the product of the applied pressure difference, the fluid's viscosity and the medium's permeability. Brinkman extended Darcy's law with a viscous stress term, thereby enabling boundary conditions to the flow field at the boundaries of the medium. Our porous media are ordered and disordered arrays of parallel cylindrical pillars, bounded by flat walls, see Figure 2. The validity of Brinkman's term, and the value of its effective viscosity, have been heavily debated since their introduction nearly 75 years ago. We find that the simulated velocity profiles are well described by an expedient interpretation of Brinkman's theory that combines the permeability of ideal pillar arrays (devoid of boundaries, i.e. infinitely long pillars) with drag by the walls. Depending on the solid volume fraction and pillar arrangement, Brinkman’s effective viscosity varies between two and three times the fluid’s viscosity. The calculated effective permeabilities of the flow devices, by combining established expressions for the permeability of ideal pillar arrays with Brinkmans’ theory, agree well with our experimental data; see Hulikal Chakrapani et al., 2023). This approach enables fast and accurate estimates of the effective permeability of micropillared chips. Contrary to this simple picture, the force distributions extracted from the simulations indicate that the drag forces at the walls are still governed by the bulk viscosity, while the permeability of the pillar array varies with the distance to the wall.

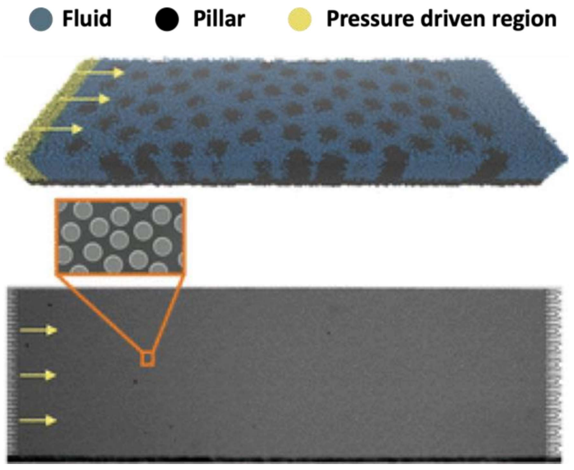

*Figure 2. Pressure-driven fluid flow through a microchannel containing a disordered array of cylindrical pillars. Identical pillar arrays are used in simulations (top) and silicon-etched micro-fluidic chips (bottom).*

**A complex fluid in a complex geometry: the imbibition of ink into a model paper**

The complexities of fluid and solid are combined to study the capillary-assisted penetration of ink droplets into disordered fibrous structures (Hulikal Chakrapani. and W.K. den Otter, 2023); see Figure 3. The toy-model substrate mimics the fibrous composition, fibre orientation and wide pore-size distribution of paper. The droplets are binary mixtures composed of carrier fluid particles and 0% to 20% dye fluid particles, with the latter having a higher affinity for the fibre particles than the former. The heterogeneity of the substrate is taken into account by averaging droplet penetration times and deposition patterns over multiple deposition points. Droplets containing dye penetrate quicker into the fibre assembly than pure carrier droplets. Irregular anisotropic patterns develop around the deposition points, caused by the irregular anisotropic stacking of the fibres. Interestingly, these patterns are unaffected by the dye concentration in the droplet. Though the heterogeneity of the substrate affects the details of the penetration process, the overall trends are robust.

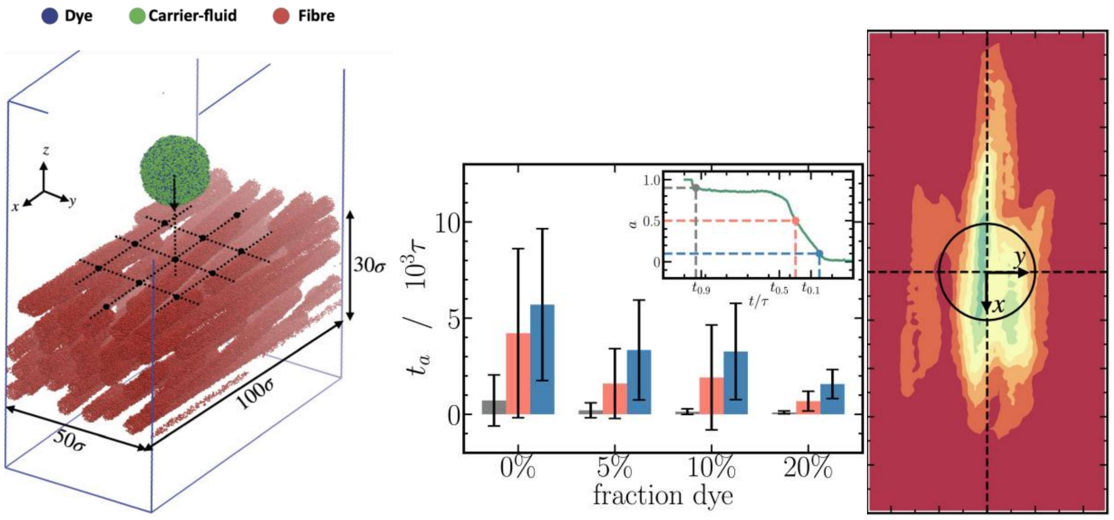

*Figure 3. (Left) Simulation snapshot of an ink droplet (green and blue) descending on a porous assembly of disordered fibres (red) of 52% porosity. The fibres are tilted by random small angles to the $x$ axis. The black dots mark deposition points, i.e. intersections of the top 'surface' of the fibre assembly and the lines of approach of the vertically descending droplets. (Center) Average residence times $t_a$ of fractions $a$ of the initial droplet mass remaining above the top surface, where $a$ = 90% (grey), 50% (orange) and 10% (blue), for droplets with dye-concentrations from 0% to 20%. The inset shows the typical evolution of the mass fraction remaining on the surface after impact, highlighting the three aforementioned fractions and the corresponding residence times. (Right) An example deposition pattern after imbibition into the substrate, demonstrating preferential spreading along the average direction of the fibres. The black lines indicate the droplet's initial size and impact point.*

## References

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