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## Slip transition in dynamic wetting for a generalized Navier boundary condition

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### Abstract

*Hypothesis:* Computer fluid dynamics simulations of dynamic wetting are often performed using a slip model on the substrate. In previous studies, the generalized Navier boundary condition (GNBC) has shown promising results and could help clear the gap between molecular and continuum scales, but lacks quantitative comparisons to experiments. We seek to investigate the dependence between the contact-line velocity and the slip length in a GNBC, by confronting numerical simulations to experimental data.

*Experiments:* The physical properties of a molten polymer (polyethylene glycol) were assessed thoroughly. Its dynamic contact angle on a cellulosic substrate was measured carefully using the Wilhelmy method. The experiment was reproduced in a finite elements model using a GNBC. It was repeated for capillary numbers between  $10^{-6}$  and  $10^{-1}$ , and slip lengths ranging from  $1\ \mu\text{m}$  to  $1\ \text{mm}$ .

*Findings:* A realistic value of the slip length was selected by matching the dynamic contact angles issued from numerical simulations and their exper-

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imental counterparts. The slip length behavior as a function of contact line velocity displayed a clear transition. The model also reproduced a dynamic wetting transition between frictional and viscous dissipations, which seems to be linked to an increasing difference between microscopic and macroscopic contact angles.

*Keywords:* Dynamic wetting, transition, slip length, generalized Navier boundary condition, polymer

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## 1. Introduction

The modeling of multiphase flow is still an active research problem. In particular, wetting dynamics, which deals with time evolution of moving contact lines on solid surfaces, also plays a role in a variety of natural phenomena, from groundwater flows to the famous example of water striders. It is also present in a number of industrial applications such as oil recovery, polymer processing, paint coating, composites manufacturing and many more.

The current study was developed in the context of liquid composite molding (LCM) processes for composites manufacturing. This family of processes involves phenomena occurring at different scales, and in particular capillary phenomena represent the smallest scale that is usually considered by studies. The void content, which should be minimized to improve the mechanical properties of a part, is influenced by the interplay between capillarity and viscosity. The evolution of voids is also heavily dependent on the velocity of liquid-gas interfaces [1, 66]. The role of capillarity was nevertheless ignored for a long time, and is still being neglected in many applications where production rate is preferred, engendering defective parts. In this context dy-

19 namic wetting is also of particular interest, since the quality of fiber-matrix  
20 adhesion is known to depend on fiber wettability [17].

21     Considering those facts, we will focus on a liquid displacing a gas on  
22 a solid surface, idealized as a smooth and rigid phase. This case, while  
23 simplified by the absence of defects when compared to real surfaces [3, 4],  
24 still encompasses multiple challenges. First, we will briefly present some  
25 theoretical models tackling this situation, as well as numerical approaches  
26 developed to simulate a moving contact line. Next, we will use numerical  
27 simulations to model the problem, the results of which will be compared  
28 with experimental data. It turns out that this procedure reveals various  
29 transitions and mechanisms, some of which may be linked to the numerical  
30 model, and others presumably to more fundamental mechanisms.

## 31 **2. Modeling multiphase flow**

32     Even though the first pioneer models in wetting dynamics were formu-  
33 lated more than 50 years ago, the variety of phenomena involved makes the  
34 establishment of a complete theory a formidable task. The extreme care  
35 needed in experimental characterization of dynamic wetting and the diffi-  
36 culty to reproduce ideal systems makes this even more difficult. This might  
37 explain why, to this date, even a model dealing with the ideal case of a  
38 smooth solid, a simple Newtonian liquid and an inviscid gas, is yet to be  
39 established and accepted by the scientific community. The already existing  
40 models can generally be grouped by families, according to the framework in  
41 which they were established [13, 22, 36, 59]. They usually give a relationship  
42 between the macroscopic dynamic contact angle  $\theta$ , which is the angle ob-  
43 tained by direct experimental measurements, and the contact-line velocity

44  $V$ . The latter is generally represented by the dimensionless capillary number  
 45  $\text{Ca} = \eta V / \sigma$ , where  $\sigma$  is the liquid-gas surface tension and  $\eta$  is the dynamic  
 46 viscosity of the liquid.

### 47 2.1. Theoretical models

48 Some of the earlier models involve statistical physics and deal with molec-  
 49 ular displacements at the contact line. The molecular kinetic theory (MKT),  
 50 initially proposed by Blake and Haynes in 1969 [10], was revisited and refined  
 51 later to include different sources of free energy (viscosity [5] and adhesion  
 52 [8]). These models generally express the contact angle as:

$$\cos \theta = \cos \theta_e - \Lambda \sinh^{-1} \left( \frac{\text{Ca}}{\text{C}_B} \right) \quad (1)$$

53 where  $\theta_e$  is the equilibrium contact angle as defined by Young's law,  
 54  $\Lambda = 2k_B T / (\sigma \lambda^2)$  is a dimensionless thermocapillary ratio,  $\lambda$  is the average  
 55 distance between reaction sites on the substrate for the adsorption of liquid  
 56 molecules,  $T$  the temperature and  $k_B$  the Boltzmann constant.  $\text{C}_B$  was  
 57 nicknamed the "Blake constant" by Petrov et al. [45], and depends on the  
 58 flavor of MKT that is being used. In its second development, which takes the  
 59 fluid viscosity into account,  $\text{C}_B = 2h\kappa_s\lambda / (\sigma v_m)$  where  $\kappa_s$  is the frequency of  
 60 molecular displacements,  $v_m$  is the molecular flow volume and  $h$  the Planck  
 61 constant.

62 Another family of models describes the problem using continuum me-  
 63 chanics. The so-called hydrodynamic theories (HD) deal with a macroscopic  
 64 flow, and usually split the flow domain into multiple scales, matching each  
 65 solution to obtain a global picture and extract a relationship between the  
 66 macroscopic contact angle  $\theta$  and the contact-line velocity  $V$ . This process  
 67 also requires to deal with a contact line paradox: the viscous stresses diverge

68 when approaching the contact line with the usual no-slip boundary condi-  
 69 tion [31]. Some of the most known models in this family were formulated  
 70 by Voinov [64] and Cox [20], often approximated as the Cox-Voinov law:

$$\theta^3 = \theta_\mu^3 + 9\Gamma\text{Ca} \quad (2)$$

71 Here  $\Gamma = \ln(x_{\max}/x_{\min})$  is used to close the contact line paradox, where  
 72  $x_{\max}$  and  $x_{\min}$  are macroscopic and microscopic cut-off lengths. They are  
 73 usually considered to be the lengths between which capillarity is significant,  
 74 and taken as  $x_{\max} = \ell_c$  where  $\ell_c = \sqrt{\sigma/(\rho g)}$  is the capillary length, and  
 75  $x_{\min} = a$  is the size of the liquid molecules, at which hydrodynamic laws  
 76 are no longer valid.  $\theta_\mu$  is a microscopic contact angle, viewed as a boundary  
 77 condition for the interface shape. Many authors tend to use the equilibrium  
 78 contact angle  $\theta_e$  instead, which is not necessarily correct, but often stems  
 79 from a lack of experimental or theoretical data on the microscopic contact  
 80 angle. In his original paper, Cox [20] postulates that  $\theta_\mu$  should also depend  
 81 on contact-line velocity, but keeps it constant in his approach.

82 Other models combine the two previous views, and consider the roles  
 83 of friction and viscous dissipation occurring simultaneously. In particular,  
 84 Petrov and Petrov's model [45, 46] uses the Cox-Voinov law, in which they  
 85 replace the constant microscopic contact angle  $\theta_\mu$  by a dynamic contact  
 86 angle computed with the MKT:

$$\theta^3 = \left[ \cos^{-1} \left( \cos \theta_e - \Lambda \sinh^{-1} \left( \frac{\text{Ca}}{\text{C}_B} \right) \right) \right]^3 + 9\Gamma\text{Ca} \quad (3)$$

87 This model thus takes into account not only two dissipation channels, but  
 88 also two scales of the wetting process. It has proven to be quite successful  
 89 to fit different kinds of experimental data, but can yield a poor agreement

90 between the values of parameters involved and their physical meaning.

91 Another promising theory, sometimes called the interface formation model  
92 (IFM), was established by Y. Shikhmurzaev over more than a decade of var-  
93 ious publications [11, 35, 38, 54–57, 61]. This model, formulated in the  
94 frame of hydrodynamics and non-equilibrium thermodynamics, states that  
95 wetting a solid with a moving liquid causes the creation and destruction of  
96 interfaces. In this context, an elementary volume of fluid moves between a  
97 liquid-solid and a liquid-fluid interface in a finite amount of time. This leads  
98 to the apparition of a surface tension gradient around the contact line as  
99 the surface tension varies continuously, because of a local change in density.  
100 A generalized Navier boundary condition (GNBC) is used to describe the  
101 fluid-solid boundary. The GNBC provides a regularization to the contact-  
102 line paradox by taking into account the added stress caused by a surface  
103 tension gradient around the triple line. When moving further away from  
104 the interface, it reduces to the classical Navier boundary condition. The  
105 IFM does not enforce the contact angle, which rather arises as a response  
106 to the overall flow from the model. Although some authors questioned its  
107 physical meaning [25] or practicality [58] (the model involves a large number  
108 of parameters), it was also implemented in numerical simulations [42, 60, 61]  
109 with promising results.

## 110 *2.2. Numerical models*

111 As a powerful predictive tool for testing models and predicting the be-  
112 havior of complex systems, numerical simulation has been used extensively  
113 to describe a moving contact line. A variety of boundary conditions were  
114 tested in the literature, with different numerical methods and domains of  
115 validity.

116

117 Continuum CFD models, solving the Navier-Stokes equation in the fluid  
118 phases, represent a common way of treating interfaces. Allowing the de-  
119 scription of a wide variety of scales in the flow, they are still by nature  
120 unable to reproduce microscopic phenomena, and need to include them as  
121 phenomenological constants or boundary conditions. They decompose the  
122 fluid domain as a grid, made of elementary cells or elements of variable  
123 sizes, over which the fields and variables are discretized and evaluated. An  
124 additional method is then used to discriminate the immiscible fluids in the  
125 computational domain.

126 Popular frameworks in CFD include the Volume of Fluid (VoF) ap-  
127 proach, using the PLIC method to reconstruct the interface [70]. It can  
128 also handle interfaces [47] and contact lines [30] using penalty terms in the  
129 boundary conditions.

130 The Finite Elements Method (FEM), which is probably the most used  
131 in scientific computing, is a good candidate for multiphysics problems. This  
132 potential for coupling makes it an interesting framework to simulate moving  
133 contact lines, since they are often influenced by other phenomena such as  
134 heat transfer or substrate deformation. Interface tracking can be achieved  
135 by additional layers such as the level set method. It is an originally non-  
136 conservative approach, deducing the interface from a signed distance func-  
137 tion, then advected by the velocity field. A conservative form was formulated  
138 afterwards [43, 44], improving the method's accuracy for the description of  
139 multiphase flow.

140 A boundary condition on the solid surface is then required for wetting.  
141 The simplest approach of imposing the contact angle to be its equilibrium  
142 value  $\theta_e$  as a boundary condition works for static contact lines, but is non-

143 physical when they move [68]. Some different approaches include setting  
144 the dynamic contact angle from a theoretical model [27], using a Navier  
145 slip model, or imposing an exhaustive mechanical equilibrium that includes  
146 capillary effects [16, 19]. This last approach can be linked to the use of a gen-  
147 eralized Navier boundary condition [51, 68, 69], which other studies found  
148 to adequately model dynamic wetting, but also introduces new parameters  
149 which, to the best of our knowledge, haven't been studied so far.

150 Sharp interface models are also commonly used for free-surface flows. In  
151 this case, an arbitrary Lagrangian-Eulerian (ALE) formulation in which the  
152 computational domain follows the deformations of the free surface is often  
153 preferred. While this method provides a more precise description of inter-  
154 faces, it prevents topological changes, which could have been an interesting  
155 feature to investigate different mechanisms such as the apparition of bub-  
156 bles. The GNBC [26] and the interface formation model [42, 61] were also  
157 implemented successfully in this framework.

158

159 Simulations at the microscopic scale are generally tackled with molecu-  
160 lar dynamics (MD). Using classical interatomic potentials (usually Lennard-  
161 Jones potentials), simple systems such as droplets are left to evolve, and may  
162 then be described at a macroscopic scale with thermodynamic quantities by  
163 averaging procedures. While these models are supposed to yield significant  
164 results due to the use of relatively fundamental mechanisms, they are limited  
165 by the computing power that is required to run a simulation. The systems  
166 typically involve up to a million of particles, and evolve for a short amount  
167 of time (nanoseconds to microseconds) [2]. Multiphase flow can be modeled  
168 by MD simulations, for which each phase is given a possibly different poten-  
169 tial. This allows to describe the evolution of a contact line, and thus study

170 dynamic wetting. Authors often rely on MD to explore nanometric scales  
171 for which no experimental observation is available yet.

172 MD is also a powerful pathway for upscaling, by comparing behaviors at  
173 the molecular and continuum scales. It was recently used to demonstrate  
174 how the inclusion of mass exchange across a wall can help regularize slip  
175 models [37]. MD studies confirmed the validity of MKT at microscopic  
176 scales for simple liquids [21], with Wang et al. suggesting that the modeling  
177 of both microscopic and macroscopic scales is needed when larger systems  
178 are being described [65].

179 In 2003, Qian et al. modeled the dynamics of immiscible two-phase Cou-  
180 ette and Poiseuille flows using molecular dynamics [50]. They showed that  
181 the whole solid-liquid boundary follows the GNBC, and that a continuum  
182 hydrodynamics formulation of the same system with a GNBC yields similar  
183 results. This important result motivates the use of this boundary condition  
184 to describe the time evolution of a contact line.

### 185 **3. Experimental data**

186 An experimental study of polyethylene glycol (PEG) with molecular  
187 weights  $M_n = 1450$  to  $20\,000$  g/mol spreading on a cellulosic substrate was  
188 carried out (for more information on the substrates, see the supplementary  
189 material). The substrates were attached to a tensiometer, plunged at differ-  
190 ent constant velocities in the melted polymer with a controlled temperature,  
191 while the advancing contact angle was being measured by the Wilhelmy  
192 method.

193 The relevant properties of PEGs were also measured in the  $75$  to  $120^\circ\text{C}$   
194 range. This allowed the definition of material properties for surface ten-

$\rho$	$\eta$	$\sigma$	$\theta_e$
kg/m <sup>3</sup>	Pa · s	mN/m	rad
1088	0.16	41.1	0.88

Table 1: Relevant material properties of the PEG 3350 used in this study, for a temperature of 75 °C.

$\lambda$	$\kappa_s$	$\Gamma$	$\chi^2$
nm	GHz		
$1.8 \pm 0.1$	$13 \pm 7$	13.9	0.0009

Table 2: Parameters for the best fit to experimental PEG 3350 data by Petrov and Petrov’s model. The molecular flow volume was computed as  $v_m = M_n/(\rho N_A)$ .

195 sion  $\sigma(T)$ , density  $\rho(T)$ , and viscosity  $\eta(T)$  in those particular conditions.  
196 For  $M_n = 3500$  g/mol, surface tension and density were found to be lin-  
197 ear functions of temperature with  $\sigma = 47.14 - 0.08T$  mN/m and  $\rho =$   
198  $1148.0 - 0.8T$  kg/m<sup>3</sup>, with  $T$  in Celsius. Viscosity followed an Arrhenius  
199 law  $\eta = 1.14 \cdot 10^{-5} \exp(2.83 \cdot 10^4/(RT))$  Pa · s, where  $R$  is the gas constant  
200 and  $T$  is in Kelvin. The liquid was found to be Newtonian in the range of  
201 temperature and shear rates involved. The static contact angles  $\theta_e(T)$  on  
202 a cellulosic substrate were also measured. In this paper, we will focus on  
203 modeling the behavior of a PEG with  $M_n = 3350$  g/mol (Table 1).

204 The contact angle versus capillary number data was fitted with differ-  
205 ent models. Petrov and Petrov’s model was deemed the most accurate to  
206 interpolate the data, and the resulting parameters are given in Table 2 for  
207 the PEG 3350. They will be used later in this study to assess and check  
208 numerical simulations.

209 **4. Numerical study setup**

210 The numerical model aims at reproducing the experiments described  
 211 in the previous section. The experimental data will then allow to assess,  
 212 compare and correct the numerical results.

213 The experiment is modeled as a monolithic fluid flow and the problem is  
 214 solved by the finite elements method with COMSOL Multiphysics<sup>®</sup>. A gas  
 215 (air) and a liquid (PEG 3350) are distinguished by using a level set function  
 216  $\Phi$  which tracks the liquid-gas interface. It is defined as a scalar function  
 217 of the signed distance from the interface, and varies between 0 (gas) and 1  
 218 (liquid) within the domain.

219 Its implementation in COMSOL is initialized as a function of the distance  
 220 from the interface  $d_0$  in the whole domain as:

$$\Phi = \frac{1}{1 + e^{\pm d_0/\varepsilon}} \quad (4)$$

221 where  $\varepsilon$  represents the characteristic interfacial width.  $\Phi$  can be seen as a  
 222 smeared Heaviside function, allowing the computation of a smooth gradient.

223 The fluid density  $\rho$  and dynamic viscosity  $\eta$  then depend on the value of  
 224  $\Phi$ , giving the two phases their properties that vary smoothly at the interface:

$$\begin{cases} \rho = \rho_g + (\rho_l - \rho_g)\Phi \\ \eta = \eta_g + (\eta_l - \eta_g)\Phi \end{cases} \quad (5)$$

225 The subscripts  $l$  and  $g$  represent the liquid and gas phases respectively.

226 The level set function is advected by a velocity field  $\mathbf{u} = ue_x + ve_y$  and  
 227 its evolution is governed by a convection-diffusion equation:

$$\frac{\partial \Phi}{\partial t} + \mathbf{u} \cdot \nabla \Phi = \gamma_{LS} \nabla \cdot \left[ \varepsilon \nabla \Phi - \Phi(1 - \Phi) \frac{\nabla \Phi}{|\nabla \Phi|} \right] \quad (6)$$

228 Here  $\gamma_{LS}$  is a reinitialization parameter, controlling the amount of reinitial-  
 229 ization of the level set function. Reinitialization steps are used to enforce  
 230 the preservation of the signed distance function and to retain the regularity  
 231 of  $\Phi$  [18, 28, 40].

232

233

234 The fluid flow is governed by the Stokes equation for an incompressible  
 235 liquid:

$$\begin{cases} \rho \frac{\partial \mathbf{u}}{\partial t} = \nabla \cdot [-p\mathbf{I} + \eta(\nabla \mathbf{u} + (\nabla \mathbf{u})^T)] + \rho \mathbf{g} + \mathbf{F}_{st} \\ \nabla \cdot \mathbf{u} = 0 \end{cases} \quad (7)$$

236 where  $p$  is the pressure,  $\mathbf{g} = -ge_y$  is the acceleration of gravity, and  $\mathbf{F}_{st}$   
 237 is a surface tension force which stems from the pressure jump at the curved  
 238 interface according to Laplace's law. This surface tension force is evaluated  
 239 by using the continuum surface force (CSF) method [15] as:

$$\mathbf{F}_{st} = \sigma \delta \kappa \mathbf{n} + \delta \nabla_s \sigma \quad (8)$$

240 where  $\mathbf{n} = \nabla \Phi / |\nabla \Phi|$  is the unit vector normal to the interface,  $\nabla_s =$   
 241  $(\mathbf{I} - \mathbf{nn}^T)\nabla$  is the gradient operator along the free surface,  $\kappa = -\nabla \cdot \mathbf{n}$  is  
 242 the local interface curvature and  $\delta = 6|\nabla \Phi| |\Phi(1 - \Phi)|$  is a smoothed Dirac  
 243 function centered on the diffuse interface.

244 The second term in Equation 8 represents a Marangoni-induced force  
 245 which may originate from a temperature gradient along the interface, or an-  
 246 other source of surface tension gradient. One can also relate it to Shikhmurzaev's  
 247 interface formation model, which assumes that the surface tension varies  
 248 continuously from its liquid-gas to its liquid-solid equilibrium value across  
 249 the triple line. In his model however, this variation is supposed to mainly

250 stem from the global flow shearing the interface, which is not what happens  
251 in the present setup.

252 A consistent streamline and crosswind artificial diffusion scheme is used  
253 for stabilization. The level set and Stokes equations are solved on quadratic  
254 and P2/P1 elements respectively.

#### 255 *4.1. Geometry and boundary conditions*

##### 256 *4.1.1. Geometry*

257 The flow on one side of the Wilhelmy plate is reproduced ([Figure 1](#)). The  
258 flow occurring between the bottom of the vessel and the plate is considered  
259 negligible, and one takes advantage of the configuration to consider only one  
260 side of the plate. Then, neglecting the influence of the plate borders allows  
261 to use a 2D model. The surface of the substrate is represented by the left-  
262 hand side boundary. At the initial state, each of the two fluids occupies one  
263 half of the domain (top or bottom), and the air-liquid interface is horizontal  
264 – forming a  $90^\circ$  angle with the solid. More details on the choices motivating  
265 the dimensions of the model are given in the supporting material.

##### 266 *4.1.2. Generalized Navier boundary condition (GNBC)*

267 As shown by the literature [[31](#)], setting the classical no-slip boundary  
268 condition on a liquid-solid boundary is not physically acceptable, since it re-  
269 sults in a stress singularity at the contact line and prevents its displacement.  
270 A boundary condition allowing the fluid to slip on the wall is thus neces-  
271 sary. This means that the velocity of the wall and the tangent velocity in  
272 the liquid near the wall may differ. The difference between these velocities,  
273 called slip velocity  $v_{\text{slip}}$ , depends on the slip model applied on the boundary.

274 Various slip models have been studied to relieve the singularity caused by  
275 a no-slip condition. Among them, the classical Navier boundary condition

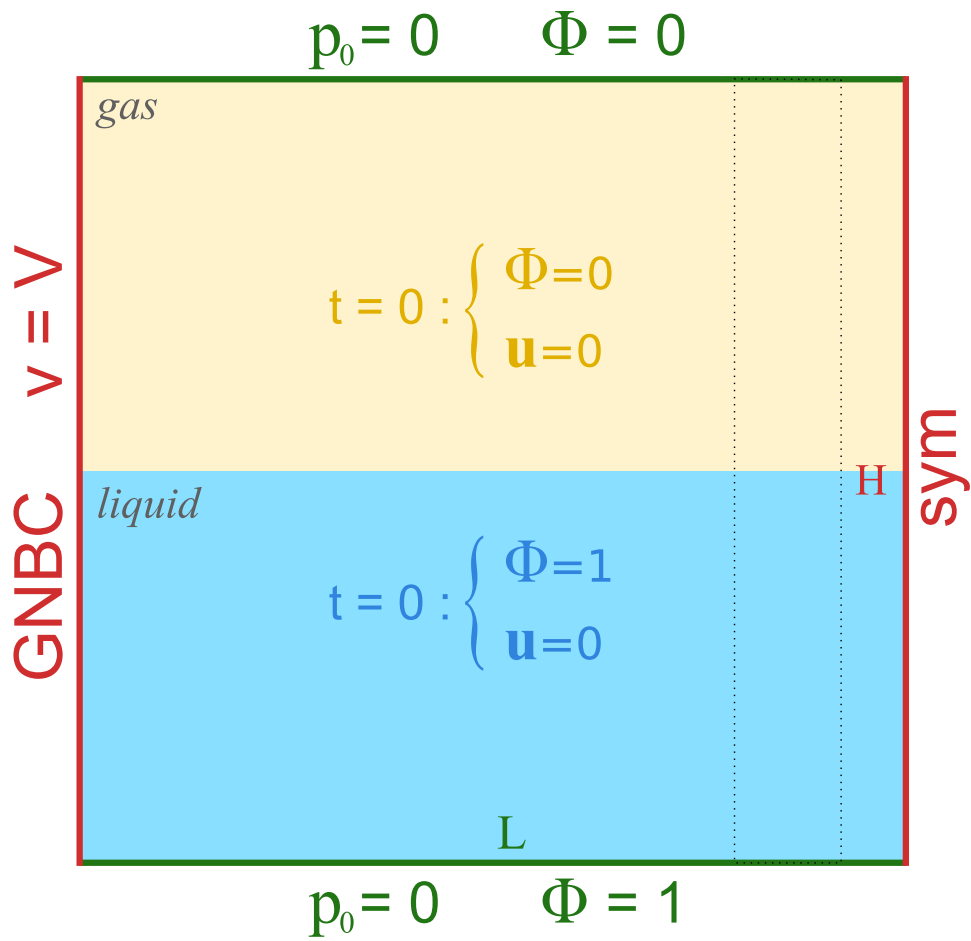


Figure 1: Geometry and boundary conditions of the Wilhelmy plate simulation at the initial state. The subdomain located between the dashed lines will be used later to compute a macroscopic contact angle.

276 (NBC) allows the liquid to slip on the solid surface, at a magnitude that is  
 277 proportional to the exerted viscous stress. However, this approach requires  
 278 the addition of an extra condition on the contact angle to close the prob-  
 279 lem for multiphase flow, making it system-dependent [14, 53]. Considering  
 280 the lack of a globally accepted contact angle model, the Navier boundary  
 281 condition is not retained in this work.

282 A generalization of the Navier boundary condition has already been pro-  
 283 posed and used by various authors for the modeling of a moving contact  
 284 line [12, 39, 49, 51, 53, 54, 57, 60, 68]. The GNBC states that the amount  
 285 of slip on the solid boundary is proportional to the total stress level, in-  
 286 cluding the viscous stress  $\tau_{\text{visc}}$  like the NBC, but also the uncompensated  
 287 (or unbalanced) Young’s stress  $\tau_Y = \sigma(\cos \theta - \cos \theta_e)$ . This is necessary to  
 288 correctly describe the presence of an interface because surface tension gives  
 289 rise to this additional stress, and ultimately relieves the pressure singularity  
 290 caused by viscous forces:

$$\frac{\eta}{\beta} v_{\text{slip}} = \tau_{\text{visc}} + \tau_Y \quad (9)$$

291 Here  $\beta$  represents a slip length, which may be interpreted as the extrap-  
 292 olated length inside the substrate for which the fluid velocity would vanish.  
 293 The slip length controls the total amount of dissipation at the boundary,  
 294 caused by solid-fluid friction. It is often expressed as a slip coefficient  
 295  $\mathcal{B} = \eta/\beta$ . The viscous stress in the  $y$ -direction can be decomposed into  
 296 its normal and tangential contributions as  $\tau_{\text{visc}} = \tau_{yy} + \tau_{yx}$ .

297 Shikhmurzaev’s interface formation model uses a similar condition to de-  
 298 scribe an *apparent* slip in the interfacial region. Slippage reflects a velocity  
 299 difference between the solid and liquid layers facing the interface, induced

300 by a surface tension gradient. An important difference with the current  
 301 model lies in the mechanism responsible for the fluid motion. The inter-  
 302 face formation model implies that the unbalanced Young's force originates  
 303 from a local variation of the surface tension around the triple line, which is  
 304 primarily caused by the flow field. Applying Young's law, a dynamic value  
 305 of the contact angle appears naturally. Here, the unbalanced Young's force  
 306 stems from a deviation of the microscopic contact angle from its equilibrium  
 307 value. The IFM could thus be seen as a more fundamental description of the  
 308 sheared interfacial layer, with the present model only extracting the resulting  
 309 apparent slip and uncompensated Young's stress as boundary conditions.

310 In the present model, the GNBC is applied on the solid boundary (Fig-  
 311 ure 2), and is expressed as:

$$\sigma\delta(\mathbf{n} \cdot \mathbf{n}_w - \cos\theta_e)\mathbf{n} + \tau_{\text{visc}} = -\frac{\eta}{\beta}(\mathbf{u} - \mathbf{U}) \quad (10)$$

312 Here  $\mathbf{U}$  is the prescribed wall velocity, and  $\mathbf{n}_w$  is a unit vector normal to  
 313 the boundary. In this expression, the microscopic contact angle is evaluated  
 314 as  $\cos\theta_\mu = \mathbf{n} \cdot \mathbf{n}_w$ , and allows the calculation of the unbalanced Young's  
 315 stress represented by the first term. This stress component is localized  
 316 around the interface via the  $\delta$  function. Since the fluid is Newtonian, the  
 317 viscous stress  $\tau_{\text{visc}}$  is expressed as  $2\eta\dot{\gamma}$ , where  $\dot{\gamma} = \frac{1}{2}(\nabla\mathbf{u} + (\nabla\mathbf{u})^T)$  is the  
 318 shear rate.

319 A non-penetration condition  $\mathbf{u} \cdot \mathbf{n}_w = 0$  is also enforced. This condition  
 320 sets the horizontal component of velocity  $v$  to zero, simplifying the expres-  
 321 sion for the shear rate  $\dot{\gamma}$  since  $\partial_y u = 0$  on the boundary.

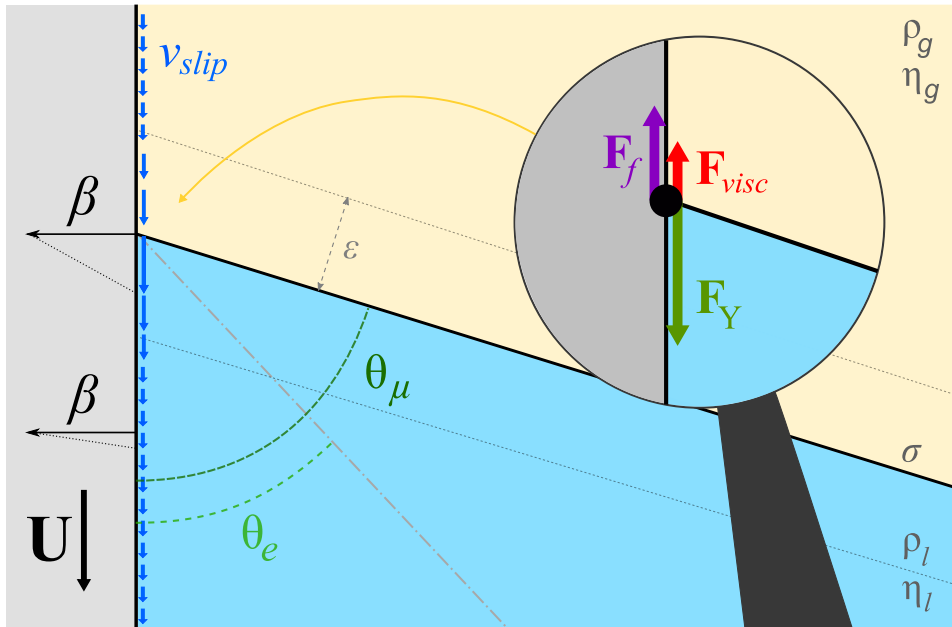


Figure 2: Diagram of the flow near a contact line according to the GNBC. An important slippage occurs on the solid boundary, and is reduced further away. The liquid-gas surface tension is constant, and the unbalanced Young force arises from a deviation of the microscopic contact angle. It is balanced by viscous drag and slippage-induced friction on the whole boundary.

322 *4.1.3. Other boundary conditions and initial values*

323 The level set function is set to  $\Phi = 0$  and  $\Phi = 1$  on the upper and  
 324 lower boundaries respectively, forcing the presence of the gaseous and liquid  
 325 phases. The pressure is imposed from a no tangential stress condition:

$$\mathbf{n}_w^T \left[ -p\mathbf{I} + \eta \left( \nabla \mathbf{u} + (\nabla \mathbf{u})^T \right) \right] \mathbf{n}_w = p_{\text{atm}} + \rho_l g h \quad (11)$$

326 Here  $p_{\text{atm}} = 1$  bar is a constant atmospheric pressure. The boundary also  
 327 enforces a normal flow via  $\mathbf{u} \cdot \mathbf{t} = \mathbf{0}$  (where  $\mathbf{t}$  is a tangential unit vector).

328 On the left-hand side boundary (representing the substrate), along with  
 329 the GNBC, the boundary condition for the level set function is set to:

$$\begin{cases} \mathbf{n}_w \cdot \left[ \varepsilon \nabla \Phi - \Phi(1 - \Phi) \frac{\nabla \Phi}{|\nabla \Phi|} \right] = 0 \\ \mathbf{n}_w \cdot \mathbf{u} = 0 \end{cases} \quad (12)$$

330 *4.2. Mesh and parameters*

331 The geometry is discretized with a rather coarse structured mesh first,  
 332 with elements of size  $h_0 = 160 \mu\text{m}$ . A series of adaptive mesh refinements  
 333 is then performed, in order to reach a final element size  $h \leq 40 \mu\text{m}$  in the  
 334 areas of interest (details and illustration in the supplementary material).

335 The liquid properties are given in [Table 1](#). Since this numerical study  
 336 involves comparisons with experiments performed at  $75^\circ\text{C}$ , this temperature  
 337 will also be chosen for the current simulations. The air viscosity and density  
 338 are set to  $\eta = 0.021 \text{ mPa} \cdot \text{s}$  and  $\rho = 1.014 \text{ kg/m}^3$ . It should be noted  
 339 that the effect of temperature on dynamic wetting has only been explored  
 340 by a few researchers as of today [[6](#), [48](#)]. Since the current model takes the  
 341 temperature dependence of material properties into account, it could be used  
 342 as a basis for those studies. Another interesting feature of this model, the  
 343 Marangoni effect described by the last term of [Equation 8](#), is also known to

344 originate from temperature gradients along interfaces, and could be studied  
345 using this approach as well.

346 In a first approach, the slip length is set to be  $\beta = h_0$ . This parameter  
347 will be studied in more detail in [section 5](#).

### 348 4.3. Dynamic contact angles

349 As the literature and our experiments point out, the measured value of  
350 a contact angle depends on the length scale at which it is evaluated. For  
351 a given situation, we will thus distinguish between a *macroscopic* contact  
352 angle  $\theta$  and a *microscopic* contact angle  $\theta_\mu$ . The former can be measured  
353 directly by experiments, with length scales ranging from micrometers to  
354 the capillary length (a few millimeters). The latter is the angle formed by  
355 the liquid in the close vicinity of the solid at the molecular scale, which is  
356 usually nanometric. The microscopic contact angle is used as a boundary  
357 condition in hydrodynamic theories, which is also the case in the present  
358 study. It cannot be observed experimentally with conventional techniques,  
359 and therefore has to be computed from macroscopic data following a model.  
360 This brings the question of how the macroscopic and microscopic dynamic  
361 contact angles are being evaluated by the numerical model.

#### 362 4.3.1. Macroscopic contact angles

363 Since the experimental data is measured using a tensiometer, a similar  
364 approach is chosen here: the macroscopic contact angles are derived from the  
365 total weight of the meniscus. This is achieved by measuring the difference  
366 between the mass of the liquid in the whole domain, and the mass of an  
367 equivalent system at rest with:

$$m = \int_{\Omega} \rho(\Phi) d\Omega - \frac{L}{L_0} \int_{\Omega_0} \rho(\Phi) d\Omega_0 \quad (13)$$

368 Here  $\Omega_0$  is a portion of the whole domain  $\Omega$  in which the interface curva-  
 369 ture is low. This subdomain of width  $L_0 = 2$  mm can be spotted in [Figure 1](#)  
 370 (between the dashed lines) and in the mesh (see the supporting material).  
 371 Since this is a 2D model,  $m$  has the dimension of a mass per unit length. The  
 372 Wilhelmy equation is then used to compute the corresponding macroscopic  
 373 contact angle:

$$\theta = \cos^{-1} \left( \frac{mg}{\sigma} \right) \quad (14)$$

#### 374 4.3.2. Microscopic contact angles

375 A microscopic contact angle can also be evaluated numerically. The  
 376 evaluation of this quantity is less straightforward, since in this model it is  
 377 defined on the whole solid boundary ([Equation 10](#)), and may vary greatly  
 378 away from the interface. It has no physical meaning outside of the contact  
 379 line however, so its value will only be estimated near the center of the  
 380 interface ( $\Phi = 0.5$ ), by using the  $\delta$  function. A weighted averaging across  
 381 the whole boundary is then performed:

$$\cos \theta_\mu = \frac{\int_w (\mathbf{n} \cdot \mathbf{n}_w) \delta}{\int_w \delta} \quad (15)$$

382 Here  $w$  denotes the left-hand boundary (the solid-fluid interface), on  
 383 which the GNBC is applied. The microscopic contact angle also depends on  
 384 the wall velocity, and is given by the model according to the GNBC. Close  
 385 to equilibrium, the microscopic and macroscopic contact angles take similar  
 386 values. We will show in [section 5](#) that for a specific set of conditions, this is  
 387 no longer the case.

388 [Yamamoto et al.](#) suggested another approach that reduces the influence  
 389 of the mesh resolution, which consists in computing the microscopic contact  
 390 angle using the Cox-Voinov relation. This in turn would modify the evalu-

391 ation of the Young’s force, but still requires the definition of a microscopic  
392 length, the selection of which still appears unclear.

#### 393 *4.4. Preliminary results*

394 The simulation consists of a first step in which the meniscus evolves  
395 towards its equilibrium position for 2 seconds, by setting  $\mathbf{U} = \mathbf{0}$ . In a second  
396 step, starting from the previous near-equilibrium configuration, the plate  
397 velocity is imposed at a constant value  $\mathbf{U} = -V\mathbf{e}_y$ .

398 A first qualitative inspection yields satisfactory results. The meniscus  
399 height is dependent on the velocity at the boundary, and the dynamic con-  
400 tact angle varies monotonously from the equilibrium value. The flow in the  
401 liquid follows a rolling motion (Figure 3), which was observed experimen-  
402 tally by many authors [24, 52]. Similarly to previous studies [49, 51], it  
403 appears that the GNBC produces a large slip in the vicinity of the contact  
404 line, and retains the same characteristics as those found by previous studies.

405 A closer quantitative inspection is however needed in order to assess the  
406 physicality of these simulations. The resulting macroscopic contact angles  
407 can be compared to their experimental counterparts. While their veloc-  
408 ity dependence are relatively similar, the values extracted from numerical  
409 simulations are smaller and the differences with experimental measurements  
410 increase when Ca is increased. We find indeed that modifying the slip length  
411 has a major influence on the contact angles. The wide range of values that  
412 can be displayed by the model for a given velocity is unphysical, since ex-  
413 periments yield a unique and repeatable value in each case. This cannot be  
414 ignored for further analyses: determining the slip length remains a topical  
415 issue. It is indeed the only parameter which may be tweaked easily, contrary  
416 to the interface width  $\varepsilon$  that would require refining the mesh.

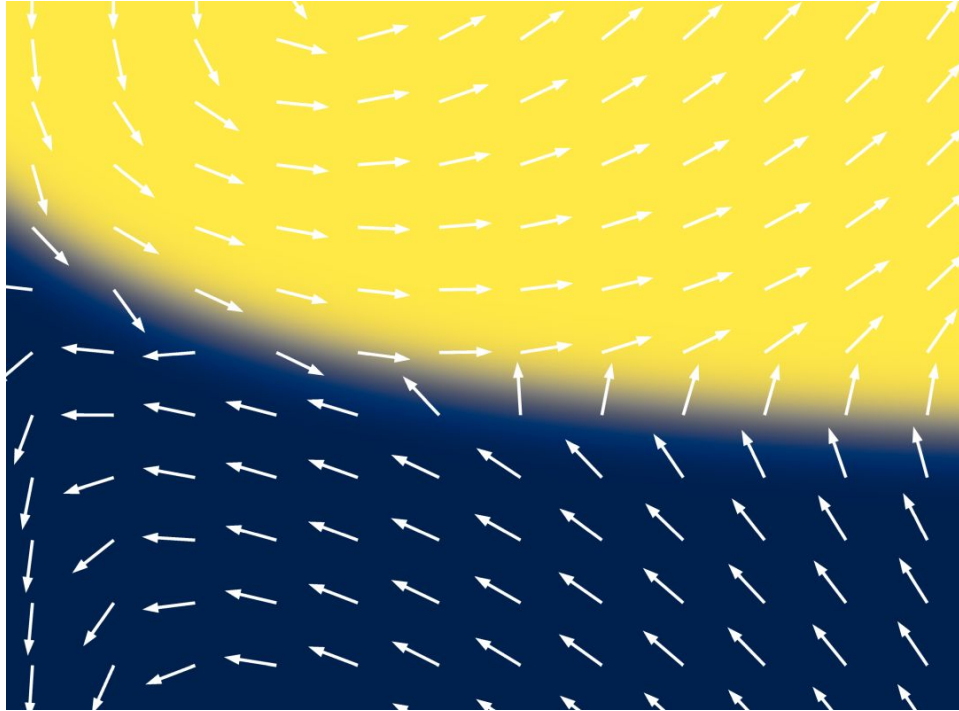


Figure 3: Zoomed-in view of the meniscus formed by PEG 3350 for a high wall velocity ( $Ca = 4.6 \cdot 10^{-3}$ ,  $V = 70$  mm/min). The area shown here is approximately 7 mm by 4 mm. The velocity field shown here is not proportional.

## 417 5. Influence of the slip length

418 The slip length  $\beta$  expressed in the GNBC governs the amount of dissipa-  
 419 tion at the triple line. This friction mechanism, balancing the viscous and  
 420 Young's stresses, provides a relation for the dynamic equilibrium of the con-  
 421 tact line. The stresses depend on the overall flow profile, which influences  
 422 the contact angle. This feature, which is well documented in the literature  
 423 [7, 38, 67], is encouraging regarding the physicality of the GNBC. While the  
 424 viscous stress is controlled by liquid properties, the Young's stress depends  
 425 on surface parameters – usually lumped in the surface tension terms. The

426 slip length is then the only parameter that can modify the force balance,  
427 influencing the flow and thus modifying the contact angle.

### 428 *5.1. Parametric study*

429 To evaluate a realistic value for  $\beta$ , a series of parametric studies were  
430 performed. For a given liquid, starting from a meniscus at rest, the velocity  
431 of the solid boundary is imposed. The simulation is then run for 13 s, and  $\beta$   
432 is varied as an exponentially decaying function of time from  $10^{-3}$  to  $10^{-6}$  m.

### 433 *5.2. Force balance*

434 Decreasing the slip length increases the macroscopic contact angle. The  
435 resulting increase in friction is balanced by the Young's and viscous stresses  
436 according to [Equation 10](#) as shown in [Figure 4b](#). The strong dependence  
437 between the microscopic contact angle and friction is shown in [Figure 5b](#).  
438 For low velocities, the relation can be considered as linear. This indicates  
439 that in this regime, friction is mostly compensated by the deviation of the  
440 microscopic contact angle. At higher velocities, a different behavior appears,  
441 most probably caused by a large increase of viscous dissipation. This results  
442 in a transition discussed in [subsection 5.3](#), where the two scales start to  
443 dissociate. Another consequence is that the velocity corresponding to the  
444 microscopic to viscous transition can be fine-tuned by the value of the slip  
445 length. A high amount of friction (corresponding to a small slip length)  
446 impedes the movement of the contact line.

447 This competition between frictional, viscous and Young's stresses is gov-  
448 erned by the GNBC. It can be shown by the force balance in the  $y$  direction  
449 by integrating [Equation 10](#) on the wall, in the vicinity of the liquid-gas

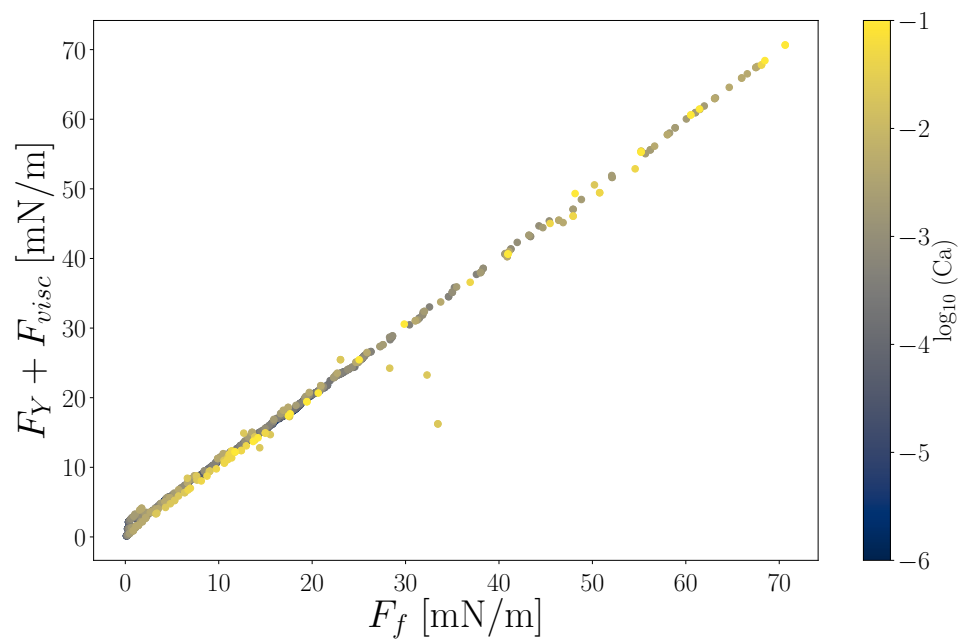


Figure 4: Sum of Young's and viscous forces plotted against the frictional force. Taking viscosity into account brings the high-velocity points back to the main curve compared to [Figure 5b](#).

450 interface:

$$\begin{aligned} & \sigma \int_w \delta(\cos \theta_\mu - \cos \theta_e) dw + \int_w \eta \left( 2 \frac{\partial v}{\partial y} + \frac{\partial v}{\partial x} + \frac{\partial u}{\partial x} \right) dw \\ & = -\frac{1}{\beta} \int_w \eta(v - V) dw \end{aligned} \quad (16)$$

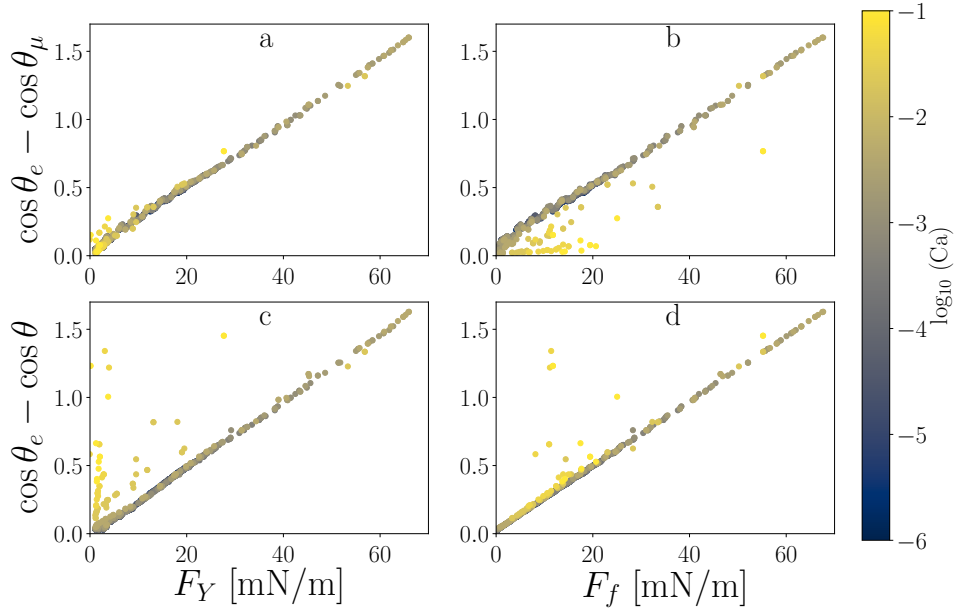


Figure 5: Deviation of the microscopic and macroscopic contact angle from the static value, plotted against friction and unbalanced Young's force. Each point corresponds to a different quasi-static situation controlled by the value of  $\beta$ . When comparing microscopic and macroscopic contributions, the microscopic origin of  $F_Y$  appears clearly, while the influence of viscous forces on the overall dissipation is also noticeable.

451 *5.2.1. Uncompensated Young's force*

452 Here, the uncompensated Young's force  $F_Y = \sigma \int_w \delta(\cos \theta_\mu - \cos \theta_e) dw$   
 453 is viewed as a consequence of the microscopic dissipation. Consequently,  
 454 the macroscopic contact angle is not a good indicator of the microscopic  
 455 processes, and a discrepancy between  $F_Y$  and the force associated with

456  $\cos \theta - \cos \theta_e$  starts to appear at higher velocities (Figure 5 a and c). Since  
457 the GNBC takes the viscous forces into account unlike the Navier boundary  
458 condition, this difference is justified. It enables the evaluation of a velocity  
459 for which the microscopic dissipation no longer dominates, and for which  
460 viscous forces can no longer be neglected.

### 461 5.2.2. Viscous force

462 The viscous term is simplified by assuming that  $u$  vanishes on the bound-  
463 ary (no-penetration BC). This is verified in the simulation results. The vis-  
464 cous stress is however still difficult to predict because of its dependence on  
465 the velocity field, closely related to the contact angle itself. We can notice  
466 indeed that for values around  $\theta = 90^\circ$ , the tangential component of the  
467 viscous stress  $\tau_{yx}$  becomes small (Figure 6).

468 Without a model for the viscous stress exerted on a wall by an inter-  
469 face, it is not possible to predict the macroscopic contact angle that will  
470 be yielded by the GNBC, especially for the highest contact line velocities.  
471 Shikhmurzaev suggests using the solution given by Moffatt [41] for a flow in  
472 a corner.

### 473 5.2.3. Frictional force

474 In most cases, the viscous component is negligible in the vicinity of the  
475 contact line (Figure 5), where the Young's stress dominates by an order  
476 of magnitude. Assuming the microscopic contact angle is constant in the  
477 whole contact line area, Equation 10 could be approximated by a simpler  
478 form, yielding an analytic expression for the microscopic contact angle such  
479 that  $\cos \theta_\mu = \cos \theta_e - \frac{Ca}{\delta\beta}$ . However, we face another issue in this case. The  
480 capillary number used here is derived from the wall velocity  $V$ , which is the

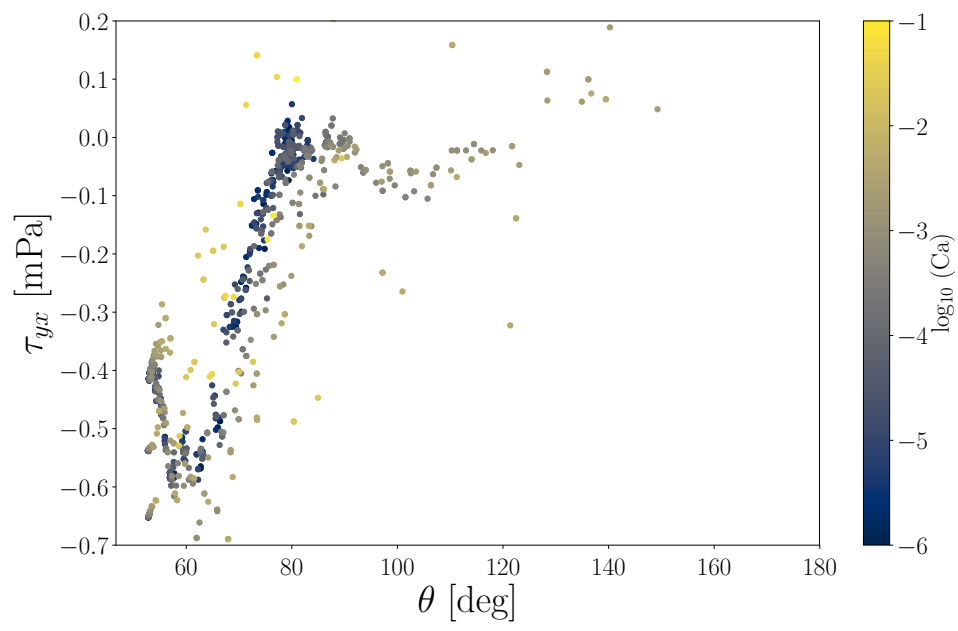


Figure 6: Dependence of the tangential component of the viscous stress on the macroscopic contact angle.

481 only one that is known. Computing the actual force balance would involve  
482 using the slip velocity  $v - V$  at the wall instead. This quantity is however *a*  
483 *priori* unknown and is part of the solution to the weak problem. Estimations  
484 in the numerical simulations indicate an important difference between the  
485 slip velocity and the wall velocity, which makes this approach unsuitable.

486 This is where theoretical models could probably provide some additional  
487 information, because they provide a more fundamental explanation on the  
488 origin of slippage. The IFM describes how the apparent slip constitutes the  
489 response of the interface to an external constraint, generally imposed by the  
490 flow (an intuitive and more detailed explanation can be found in [11]). The  
491 MKT links the slip velocity to molecular processes at the triple line, and  
492 can yield a relation between the slip length and the local shear rate [9].

### 493 *5.3. Dynamic wetting regime transition*

494 When dissipation increases (high velocities and small slip lengths), the  
495 microscopic and macroscopic contact angles diverge rapidly with a sharp  
496 transition point. Their difference remains small for low contact line velocities  
497 and large slip lengths.

498 The threshold of very rapid growth of  $\theta - \theta_\mu$  seems to originate from  
499 the apparition of viscous bending. As presented in subsection 5.2, when the  
500 velocity increases, the viscous stresses become significant in the overall dis-  
501 sipation. The microscopic contact angle  $\theta_\mu$  and the Young's stress increase  
502 slowly, and the morphology of the bulk flow is mostly influenced by viscous  
503 bending. The macroscopic contact angle  $\theta$  then starts to increase rapidly,  
504 in agreement with the experimental data.

505 When fitting the experimental data with the Petrov and Petrov model,  
506 one can distinguish between the microscopic and macroscopic contact angles

507 by separating the MKT and HD contributions. In previous studies, this  
 508 model was being fitted by letting  $\Gamma$ ,  $\lambda$ ,  $\kappa_s$  as free parameters. While the  
 509 agreement between the model and data is generally excellent, the values  
 510 obtained for the best fits tend to display unexpected values. This behavior  
 511 is also observed with the Cox-Voinov model, for which unphysically high  
 512 values of  $\Gamma$  are routinely found. If Petrov and Petrov’s model is appropriate,  
 513 we observe that the respective contributions of microscopic and macroscopic  
 514 dissipations can take multiple values that would fit the data well. Meanwhile,  
 515 for a given set of experimental conditions and a resulting contact angle,  
 516 those contributions are uniquely defined. Since the microscopic dissipation  
 517 is hardly measurable, an estimation of the viscous term is needed in order  
 518 to find a unique solution.

519 Knowing this, following the approach of de Gennes [23], we set a fixed  
 520 value of  $\Gamma$  by using the capillary length  $\ell_c = \sqrt{\sigma/(\rho g)}$  for the macroscopic  
 521 cut-off length, and the molecular size computed as  $a = (M_n/(\rho N_A))^{1/3}$  for  
 522 the microscopic cut-off, where  $N_A$  is the Avogadro constant. This value is  
 523 then used for a fit of Petrov and Petrov’s model, letting only  $\lambda$  and  $\kappa_s$  as  
 524 adjustable parameters. The best fits for each liquid yield values of  $\kappa_s =$   
 525 13 GHz and  $\lambda = 1.8$  nm (Table 2). Those are acceptable values regarding  
 526 their physical interpretation, as given by the MKT in Equation 1: they  
 527 respectively represent an average of the molecular movements’ frequency  
 528 and distance.

529 Starting from the data for macroscopic contact angle  $\theta$  fitted by the  
 530 Petrov-Petrov model, the contribution of  $\theta_\mu$  can be extracted from Equa-  
 531 tion 3 as the first term of the right-hand side. While most models of dynamic  
 532 wetting do not distinguish between these scales, this approach also allows  
 533 to compute how the microscopic and macroscopic contact angles deviate

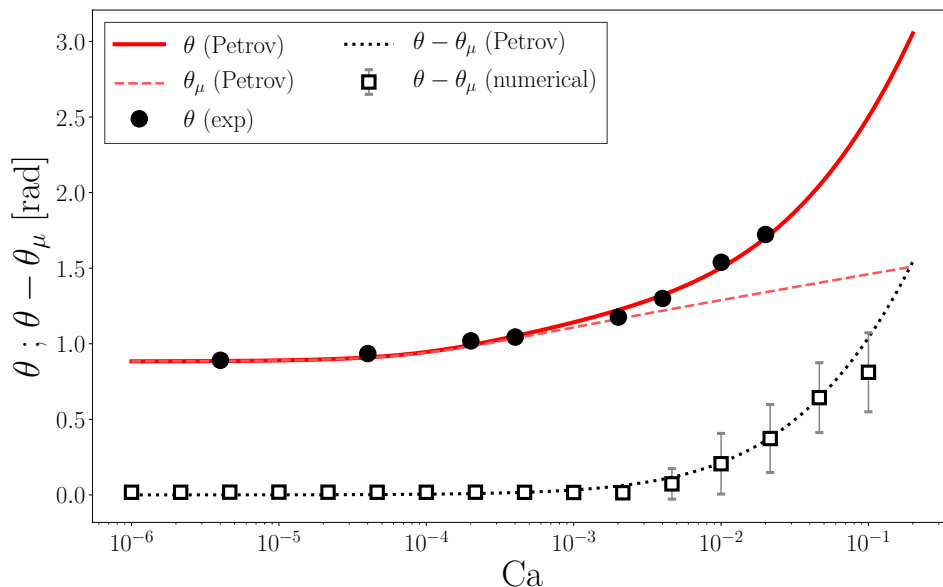


Figure 7: Petrov and Petrov fit on experimental data (full circles) for PEG 3350. The best values for this fit were  $\lambda = 1.8$  nm,  $\kappa = 13$  GHz.  $\Gamma$  was fixed to 13.9 by taking the capillary length and molecular size of PEG 3350. The macroscopic (full line) and microscopic (dashed line) contributions are represented, as well as their difference  $\theta - \theta_\mu$  (dotted line). The latter starts to diverge around  $Ca \approx 10^{-3}$ . The same divergence is observed in the numerical simulations (empty squares), for which contact angles were averaged over 77 different slip lengths.

534 depending on the contact line velocity (Figure 7). The contribution of  $\theta_\mu$   
535 keeps a relatively low value even at high  $Ca$ . The macroscopic contact angle  
536 stays very similar to  $\theta_\mu$  up to  $Ca \approx 10^{-3}$ . Then at higher velocities, viscous  
537 bending comes into play, and  $\theta$  and  $\theta_\mu$  start to diverge.

538 This mechanism is well reproduced by the numerical simulations. Aver-  
539 aging  $\theta - \theta_\mu$  over each slip length, it can be plotted for different contact-line  
540 velocities, and turns out to agree well with the best fit of Petrov and Petrov's  
541 model on experimental data. The viscous origin of this transition can be

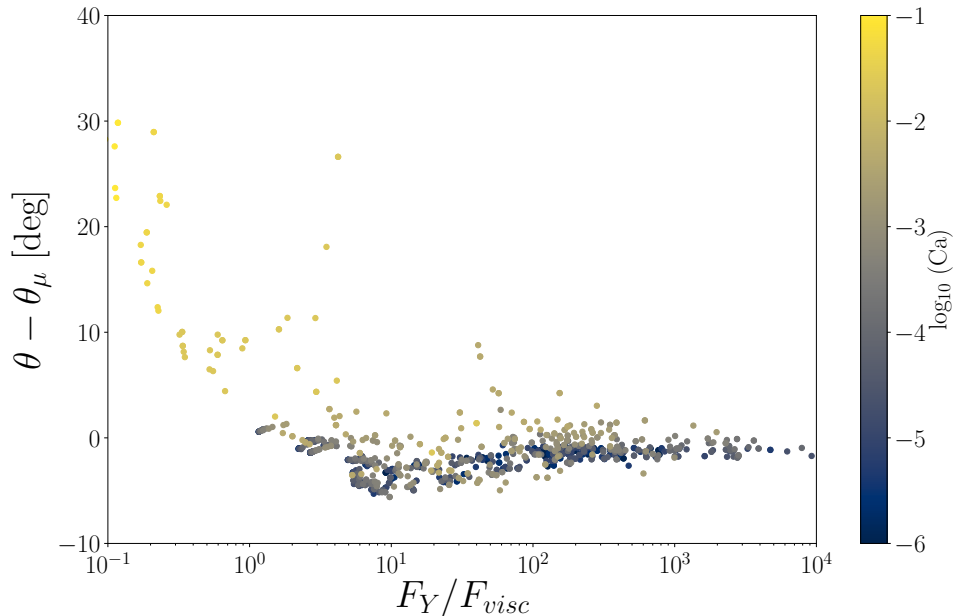


Figure 8: Evidence of the transition’s viscous origin: the difference between the macroscopic and the microscopic contact angles is plotted against the ratio of the Young’s force  $F_Y$  and viscous force  $F_{visc}$ . A transition occurs when  $F_Y \approx 10F_{visc}$ .

542 shown by studying how the two scales separate. When the Young’s force is  
 543 dominant,  $\theta_\mu$  and  $\theta$  are similar. Once the viscous force exerted in the con-  
 544 tact line area reaches the same order of magnitude as the Young’s force, the  
 545 microscopic and macroscopic contact angles start to dissociate (Figure 8),  
 546 and viscosity dominates the deformation of the free surface.

547 This interpretation can be linked to a recent study showing a very sudden  
 548 appearance of viscous behavior in dynamic wetting [71]. The two domains  
 549 are well described either by the MKT (low viscosities) or HD theories (high  
 550 viscosities). Petrov and Petrov’s approach constitutes an interesting combi-  
 551 nation of the two. Other studies started to show a divergence between the  
 552 microscopic and macroscopic contact angles caused by an increase in contact

553 line velocity [33, 34]. While the scale dependence of contact angles is widely  
554 accepted, these two studies are, to the best of our knowledge, the only ones  
555 showing the influence of velocity on the separation between microscopic and  
556 macroscopic contact angles. The curves showing the velocity dependence  
557 are relatively similar to the present work and involve polymers as liquids  
558 or coating on the substrates, but were obtained using a different analysis.  
559 Interestingly, the critical capillary number at which the transition occurs  
560 was already identified in [33], but also in different contexts. The minimiza-  
561 tion of void content in LCM processes was indeed reached for this particular  
562 condition [29], and being a phenomenon driven by the competition between  
563 viscous and capillary forces, the overall picture seems consistent.

#### 564 *5.4. Velocity-dependence*

565 The parametric study described in subsection 5.1 was performed for a  
566 range of plate velocities. This also allows to assess how the slip length  $\beta$   
567 evolves with Ca in the present framework. Different  $\theta$  versus  $\beta$  curves are  
568 obtained for each velocity. A velocity-dependent slip length was already  
569 observed and modeled for solid-liquid interfaces [32, 62, 63], and could thus  
570 constitute a reasonable hypothesis for a slip model applied to a moving  
571 contact line.

572 A realistic value of the slip length may then be selected by comparing  
573 the simulation results to experimental data. For a given plate velocity,  
574 the fit according to Petrov and Petrov's model  $\theta_{\text{fit}}$  of the experimentally  
575 measured macroscopic contact angles is used to interpolate the experimental  
576 data. This in turn allows to select a physically acceptable value of the  
577 slip length for any imposed plate velocity, denoted  $\beta_0$ , for which the model  
578 matches experiments. The value of  $\beta_0$  is identified by a linear fit of numerical

579 contact angles  $\theta$  close to  $\theta_{\text{fit}}$  for the same plate velocity: we find  $\beta_0$  such  
 580 that  $\theta(\beta_0) \simeq \theta_{\text{fit}}$ .

581 Using this procedure, a value of  $\beta_0$  is then extracted for each value of  
 582 Ca. The evolution of  $\beta_0$  may then be plotted against contact line velocity  
 583 (Figure 9). In the resulting curve, the slip length decreases exponentially  
 584 until the system reaches a critical capillary number  $\text{Ca}_c$ , corresponding to a  
 585 macroscopic contact angle  $\theta = \pi/2$ . The slip length then increases strongly  
 586 when the velocity is increased, apparently following a power law. Overall,  
 587 an empirical fit over each domain yields a velocity-dependence for  $\beta_0$  such  
 588 that:

$$\begin{cases} \text{Ca} < \text{Ca}_c : \ln \beta_0 = \frac{-1}{a_1 \text{Ca} + b_1} \\ \text{Ca} > \text{Ca}_c : \beta_0 = a_2 \text{Ca}^{3/2} + b_2 \end{cases} \quad (17)$$

589 Such a fit would yield  $a_1 = 4.7 \cdot 10^{-6}$ ,  $b_1 = 7.9 \cdot 10^{-2}$  and  $a_2 = 1.7 \cdot 10^{-3}$ ,  
 590  $b_2 = 5.6 \cdot 10^{-7}$ . It was not plotted here due to the lack of a satisfying inter-  
 591 pretation for those values, but some observations can still be made.

592 The transition displayed by  $\beta_0$  as a function of contact-line velocity could  
 593 stem from the increasing importance of viscous forces, which come with  
 594 the augmentation of the capillary number. At low contact line velocities,  
 595 the viscous stresses in the interfacial area are negligible compared to the  
 596 Young's stress. The frictional dissipation is dominated by the slight increase  
 597 of the microscopic contact angle, as stated by the GNBC. This either means  
 598 increasing the slip velocity, or decreasing the slip length. Once the contact  
 599 line velocity becomes important, an increase in the slip length moderates the  
 600 increase in frictional dissipation. This means that in the interfacial area,  
 601 the Young's stress can still be a dominant mechanism, without becoming

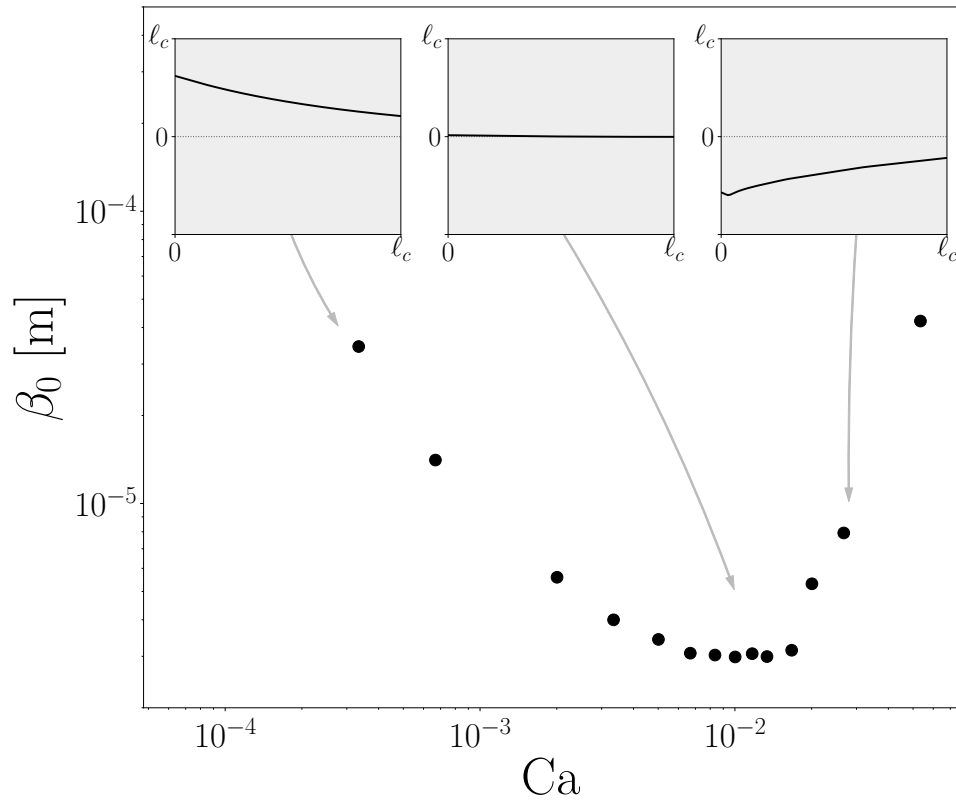


Figure 9: Velocity dependence of the slip length, as identified from the numerical and experimental data. Some close-ups of the corresponding interfaces are plotted in the insets for three regimes.

602 negligible compared to the viscous stress.

603 This transition could also be interpreted as a shift from a partially wet-  
604 ting to a non-wetting system, because here the sharp change in the slip  
605 behavior identified from the experiments corresponds to a contact angle  
606  $\theta = \pi/2$ . In non-wetting conditions, dewetting becomes an energetically  
607 favorable mechanism. In our case, increasing the dynamic contact angle  
608 reduces the total solid-liquid interactions, and thus diminishes the total  
609 (frictional) dissipation. According to the GNBC, this can be achieved by  
610 increasing the slip length, which is what is observed in our results.

## 611 **6. Conclusion**

612 The dynamic equilibrium of a contact line was reproduced with the finite  
613 elements method using a diffuse interface model and a generalized Navier  
614 boundary condition (GNBC) on the solid interface. The simulations aimed  
615 at reproducing experimental data both qualitatively and quantitatively, to  
616 assess the evolution of a correct slip length  $\beta_0$  in the GNBC. An exper-  
617 imentally observed transition between microscopic and macroscopic (vis-  
618 cous) dissipation could be reproduced [34, 48, 59], and seems to occur at  
619 similar velocities. The generalization of the Navier boundary condition,  
620 which does not directly enforce any model for the dynamic contact angle,  
621 seems to capture all of the characteristics that are observed experimentally  
622 (velocity dependence, rolling motion), in accordance with previous studies  
623 [49, 51, 68, 69].

624 A quantitative evaluation showed that the slip length  $\beta_0$  has to vary with  
625 the contact line velocity to reproduce the contact angles measured experi-  
626 mentally. In previous studies however, the GNBC was implemented using

627 a constant value for the slip length. This parameter, while being seemingly  
628 of purely numerical nature, also appears in Shikhmurzaev’s interface forma-  
629 tion model [57], which uses a GNBC as well. One could interpret it as a  
630 characteristic of how the interface responds to shearing. The variation of  $\beta_0$   
631 required to obtain realistic contact angles displays a sharp transition with  
632 the contact line velocity. This transition seems to be linked to a mitiga-  
633 tion of the large increase in viscous forces in the interfacial area induced  
634 by the high contact line velocity. It can also be interpreted as a switch in  
635 wettability, which would modify the energetic landscape.

636 The role of viscosity in this problem should be studied further in future  
637 works. While the situation that was modeled here involved a gas (air) of  
638 very low viscosity compared to the liquid phase, using two fluids of closer  
639 viscosities could yield interesting results, and modify the role of viscous  
640 stresses in the overall dissipation. This could be analyzed by expanding  
641 Petrov and Petrov’s model [46] using the original formulation by Cox [20],  
642 which takes the ratio of viscosities between the two phases into account.  
643 This could again be compared to numerical simulations, assessing the role  
644 of the viscosity ratio on  $\beta_0$ . Other aspects of the liquid properties should be  
645 explored as well. Most models in wetting dynamics deal with *simple* liquids,  
646 and thus do not take the morphology of polymers into account. Meanwhile,  
647 polymers represent a family of materials that are used in many industrial  
648 processes involving contact lines. For instance, the effect of varying the  
649 molecular weight on dynamic wetting is still not known to this day.

650 Finally, models in wetting dynamics that involve non-Newtonian liquids  
651 are at a very early stage. Most of them deal with particular situations  
652 such as droplets spreading, total wetting or particular rheological models  
653 [36]. Knowing the importance of the viscous stresses in interfacial flows

654 and the common use of non-Newtonian polymers, the development of a  
655 model suitable for any liquid rheology would constitute a major achievement  
656 for wetting dynamics. While the theoretical development of such a model  
657 appears to be a formidable task, the GNBC's consistency encourages the use  
658 of a numerical study which, by comparison with experimental data, would  
659 constitute a first approach.

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# Slip transition in dynamic wetting for a generalized Navier boundary condition: supplementary information

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## A. Experimental data

### a. Liquid properties

The relevant physical properties of the polymer used in this study, a polyethylene glycol with a molecular weight  $M_n = 3350$  g/mol, were carefully assessed. They were studied for temperatures ranging from  $T = 75$  to  $120$  °C.

#### **Viscosity $\eta$ .**

The polymer viscosity was measured using a Malvern Kinexus Pro cone-plate rheometer. Its dependence on shear rate was measured for fixed temperatures of 65, 80, 100, 120, 150 and 180 °C. The experimental data was fitted by a Carreau-Yasuda model for the dependence on shear rate  $\dot{\gamma}$ , and by an Arrhenius model for the dependence on temperature  $T$ :

$$\eta(\dot{\gamma}) = 0.122 + 0.024 \left[ 1 + (\dot{\gamma}/161)^{1.57} \right]^{-1/1.57} \text{ Pa} \cdot \text{s} \quad (\text{A.1})$$

In the experimental conditions of the dynamic contact angles measurements, the PEG was considered to be in the Newtonian plateau, and could thus be treated as a Newtonian liquid. The shear rate in the contact line area resulting from the numerical simulations is plotted and compared to the liquid rheology in [Figure I](#).

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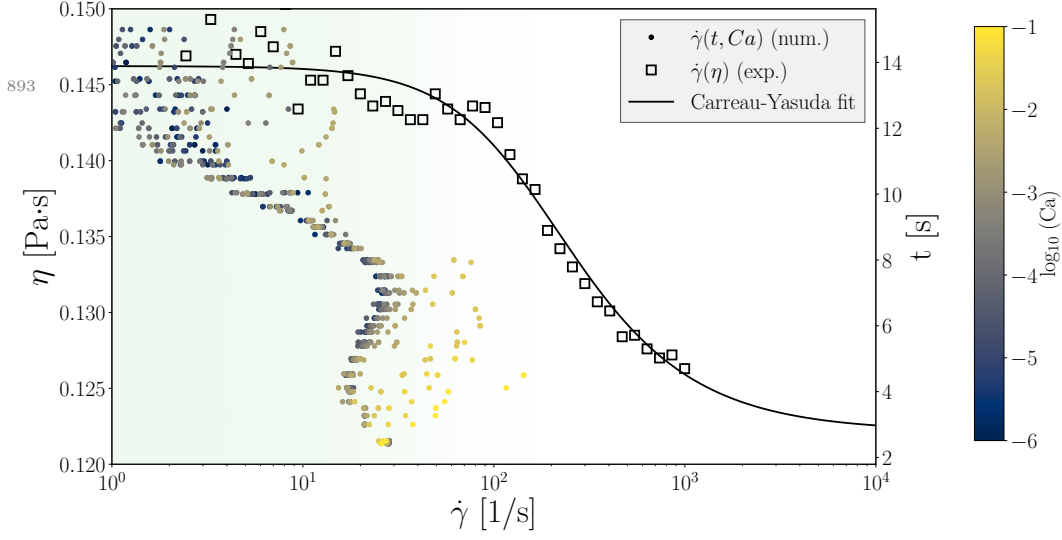


Figure I: Evolution of PEG3350 viscosity with the shear rate, compared with the numerical estimations of shear rate in the contact line area.

The dependence on temperature  $T$  was measured at a fixed shear rate of  $100 \text{ s}^{-1}$ . The temperature was increased from  $60$  to  $200 \text{ }^\circ\text{C}$  with a rate of  $1 \text{ }^\circ\text{C} \cdot \text{min}^{-1}$ , and the data was fitted by an Arrhenius model:

$$\eta(T) = 1.14 \cdot 10^{-5} \exp\left(\frac{2.83 \cdot 10^4}{RT}\right) \text{ Pa} \cdot \text{s} \quad (\text{A.2})$$

where  $R$  is the gas constant, and  $T$  is the temperature in Kelvin. A good correspondance was found with the data yielded by the study of shear rate (Figure II).

### **Density $\rho$ .**

The density was measured with a Dataphysics DCAT25 microbalance, equipped with a calibrated density probe. For each temperature, 3 measurements of buoyancy were averaged, and the temperature-dependence of density could be fitted by a linear relation (Figure III):

$$\rho(T) = 1148.0 - 0.8T \text{ kg/m}^3 \quad (\text{A.3})$$

where  $T$  is the temperature in Celsius, measured with a thermocouple on the precise location at which the probe is immersed in the liquid bath.

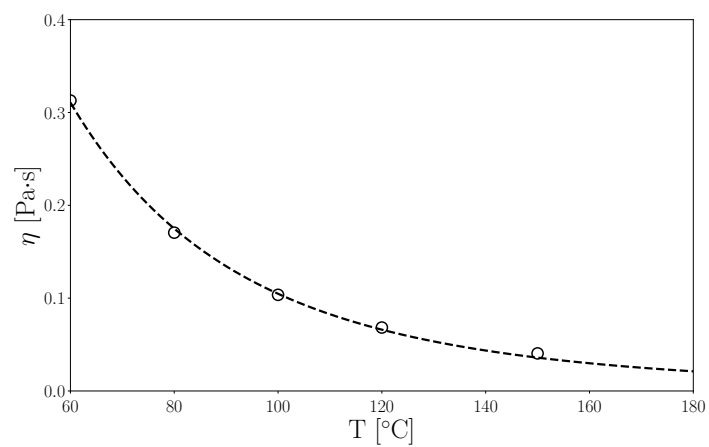


Figure II: Evolution of viscosity with temperature: comparison between the Arrhenius fit and the data yielded by the dependance on shear rate (empty circles).

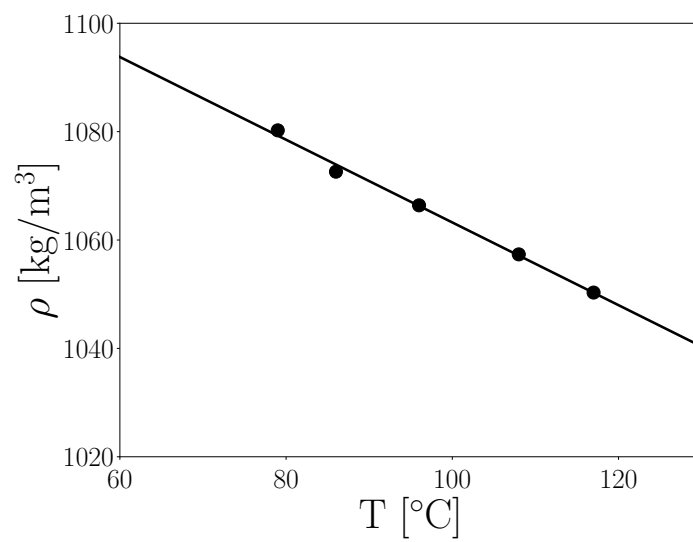


Figure III: Evolution of the density of PEG 3350 with temperature: experimental data and its corresponding linear fit.

### *Surface tension $\sigma$ .*

The <sup>895</sup>surface tension was measured with a Krüss DSA100 drop shape analyzer, using the pendant drop method. Temperatures ranging from 80 to 120 °C with steps of 10 °C were explored, and each value was averaged over 10 droplets. The surface tension followed the Eötvös rule and could thus be fitted by a linear relation with respect to the temperature  $T$  in °C (Figure IV):

$$\sigma(T) = 47.14 - 0.08T \quad \text{mN/m} \quad (\text{A.4})$$

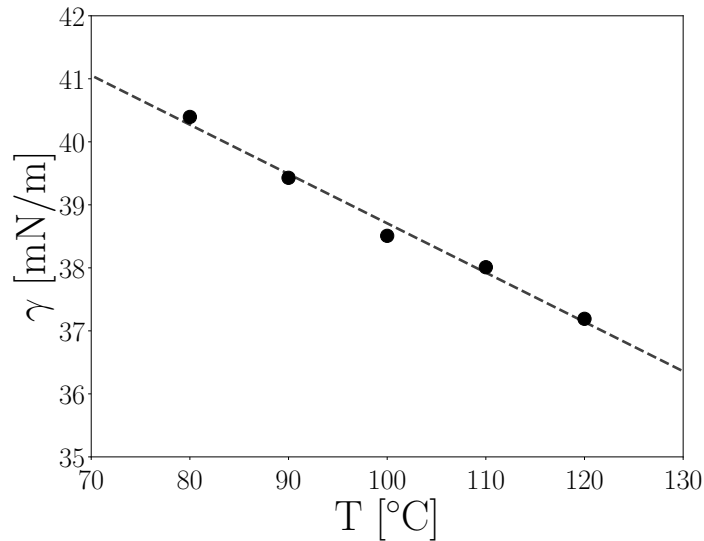


Figure IV: Evolution of the PEG 3350 surface tension with temperature: measurements and linear fit.

### *b. Substrates*

The substrates used in this study were 23  $\mu\text{m}$  thick NatureFlex<sup>TM</sup> NP cellulosic films provided by Innovia Films. They were bathed in ethanol for 1 h to remove the plasticizing glycerol on the surface, air-dried and cut with a scalpel to approximately  $1 \times 2 \text{ cm}^2$  rectangles. The films presented oriented ridges when observed through a light source with the naked eye. They were cut so that the contact line would end up perpendicular to those ridges. The ridges were however not visible through a scanning electron or optical microscope. An earlier study on those same films [3] showed that they could be considered smooth compared to the scales at which the Cassie-Baxter and Wenzel models apply.

*c. Dynamic contact angles*

The dynamic contact angle formed by PEG on the cellulosic substrate was measured by the Wilhelmy method with a Dataphysics DCAT25 microbalance. The PEG was poured in the microbalance vessel and heated to 75 °C. A thermocouple was used to check the temperature at the center of the vessel.

The substrate was then attached to the microbalance, with the grooves perpendicular to the free surface. The vessel was moved upwards at a constant velocity  $V$ , until the substrate was plunged into the liquid with an immersion depth  $h = 7$  mm. At this point, the vessel was left to rest for 2 min (Figure V), and then lowered at the same velocity  $V$ .



Figure V: Snapshot of the meniscus formed by PEG 3350 on a static cellulosic substrate, captured at an angle above the liquid surface. The substrate is attached to a microbalance, measuring the force exerted by the contact line. This allows to evaluate the macroscopic contact angle.

After detaching the substrate from the microbalance, its width  $w$  was measured carefully with an optical microscope to determine the length of the contact line  $2w$ . The thickness of the substrate was considered negligible.

The contact angle  $\theta$  was then determined using the Wilhelmy equation, using a linear fit the on force - position relation to eliminate the effects of buoyancy and viscosity:

$$\cos \theta = \frac{F}{2\sigma w} \quad (\text{A.5})$$

where  $F$  is the force exerted by the contact line, derived from the mass  $m$  yielded by the microbalance<sup>897</sup>. The regression (Figure VI) enables the elimination of buoyancy and viscous drag on the substrate, assumed to be proportional to the depth of immersion  $h$  [2], by using the extrapolated value at the very beginning of immersion ( $h = 0$ ).

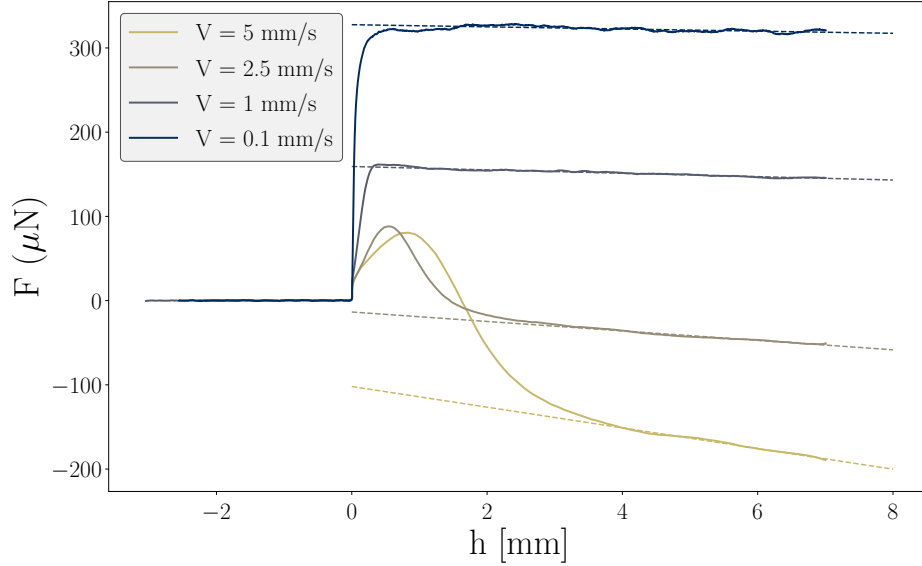


Figure VI: Tensiometric curves used to determine the dynamic contact angle for different velocities.

The equilibrium contact angle  $\theta_e$  was determined using the last points of the resting phase, with the lowest velocities  $V$  used in this study. All of the low-velocity experiments yielded a similar value, so that this measurement was considered reliable. This was confirmed by further experiments, in which a substrate was immersed and the meniscus was left at rest, spreading for 5 h, yielding a value of  $\theta_e$  within  $1^\circ$  of those measured by the previous method (Figure VII).

The dynamic contact angles of the advancing liquid were determined for a set of 8 different velocities. Velocities higher than  $5 \text{ mm} \cdot \text{s}^{-1}$  were not used: the corresponding menisci do not seem to reach a quasi-static equilibrium before approaching the bottom of the vessel. The dynamic contact angle  $\theta$  versus capillary number  $\text{Ca}$  data was fitted with different models. Petrov and Petrov's model was deemed the most appropriate to interpolate the data (Figure VIII), and the resulting parameters are given in Table 2 for the PEG 3350.

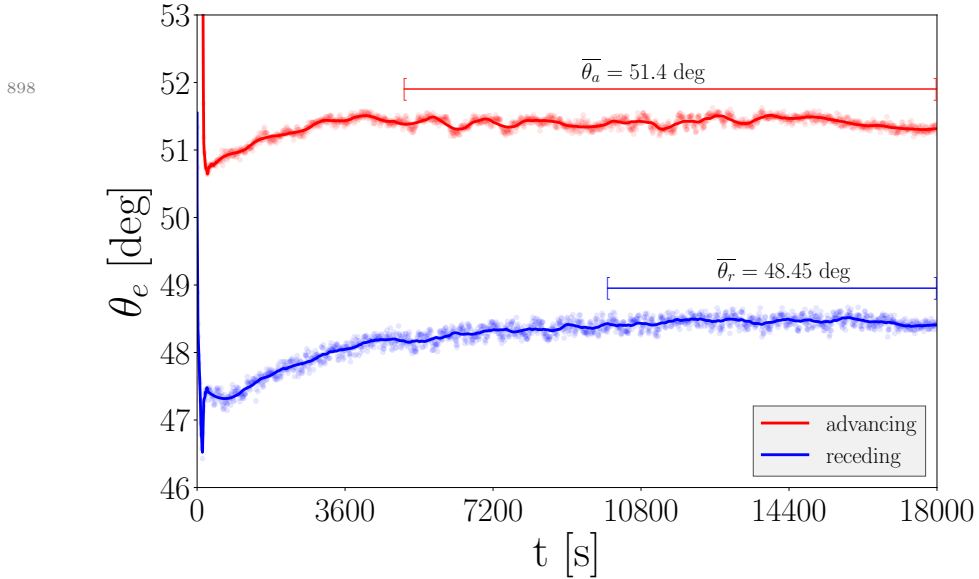


Figure VII: Static advancing and receding contact angles measured in a timespan of 5 h.

## B. Numerical model

### a. Geometry

The dimensions of the 2D domain were chosen to reproduce the experimental conditions. Considering the capillary length  $\ell_c \approx 2$  mm in our experimental conditions, the domain requires a horizontal width  $L$  long enough so that far from the contact line, the interface stays unperturbed. Since the elevation of a meniscus decays exponentially as a function of the distance  $x$  to the surface of the plate [1], we will consider that it is at rest for  $x > 5\ell_c$ , and thus impose  $L = 13$  mm to keep an extra margin for contact angle measurements. The symmetry boundary condition may then be used at the right-hand side of the geometry to obtain an unperturbed interface and flow.

The height of the domain  $H$  is also selected by considering the rise of a meniscus on a vertical plate. In a steady state, this height is  $\ell_c \sqrt{2(1 - \sin \theta_e)}$ , which would amount to approximately 2.8 mm for total wetting in the present case. However, an advancing contact angle cannot be smaller than its equilibrium value, and our simulations deal with partially wetting liquids. With the equilibrium contact angles considered here, this height reduces

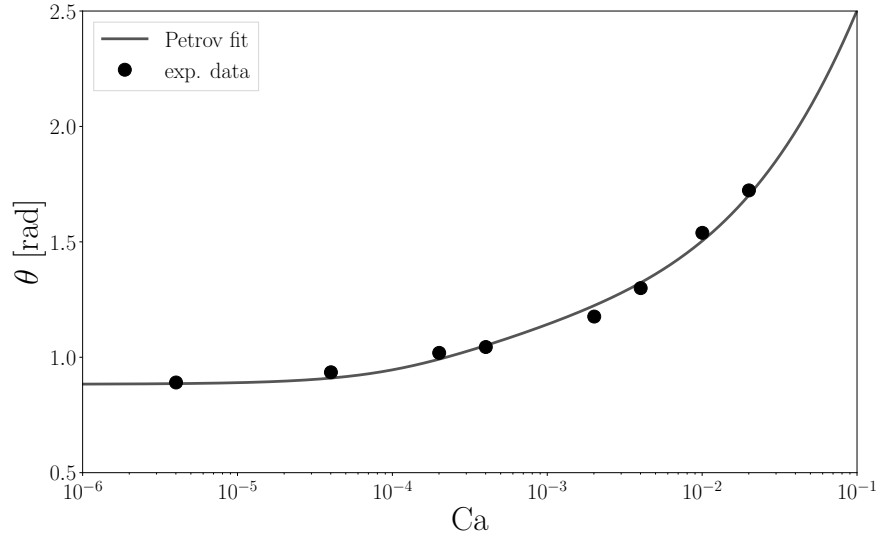


Figure VIII: Experimental measurements of dynamic contact angles formed by PEG 3350 on a cellulosic substrate, and their corresponding best Petrov-Petrov fit.

to less than 2 mm. Consequently, the domain height is chosen to be  $H = 10$  mm, to keep a safety margin like we did for the domain width, also allowing the flow to develop further from the interface.

*b. Mesh, materials and parameters*

The choice of an element size  $h \leq 40 \mu\text{m}$  was made by performing simulations on regular meshes of decreasing elements size, until reaching a stable macroscopic contact angle (Figure IX).

Since a refinement splits each element in two right triangles, with quadrangular initial elements, a convergence is reached after 4 iterations on the coarse mesh. The refinements are applied following the indicator  $\sqrt{(\partial_x \Phi)^2 + (\partial_y \Phi)^2} / (x + L/4)$ . This allows to increase the spatial resolution across the interface and around the contact line (Figure X), while keeping it coarser on the overall flow.

The characteristic interface width  $\varepsilon$  is set to be  $h_0/2 = 80 \mu\text{m}$ . Initially, the reinitialization parameter  $\gamma_{LS}$  used in Equation 6 is chosen so that the mesh Péclet number  $\text{Pe} = uh/(2\gamma_{LS}\varepsilon) = u/\gamma_{LS} = 1$ . The maximum possible velocity is  $\sigma/\eta$ . But with this value

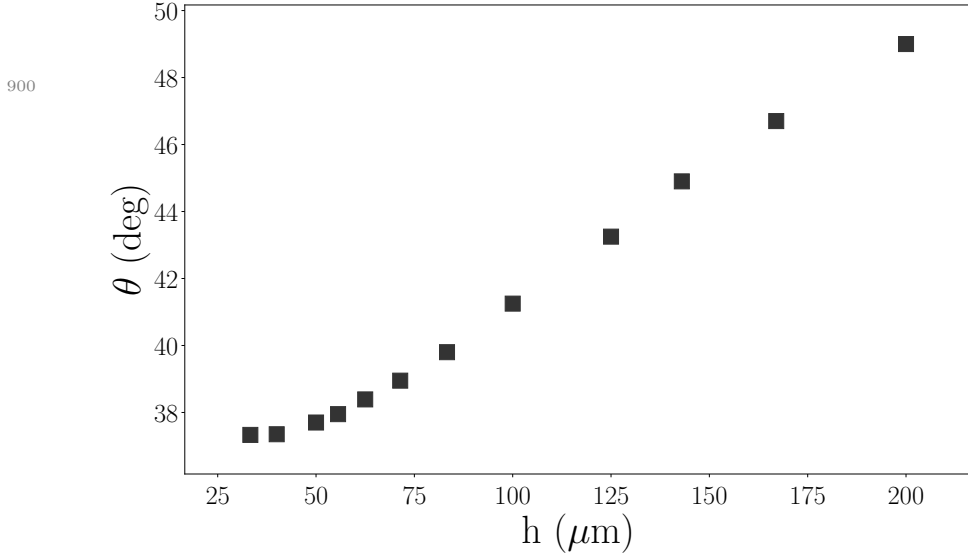


Figure IX: Mesh convergence study assessed by the evolution of the macroscopic contact angle as a function of element size  $h$ . A stable value is reached for  $h \approx 40 \mu\text{m}$ .

the initial calculation conditions would be too severe and this would considerably lengthen the simulation time. A more reasonable value of  $\gamma_{LS} = \sigma/(10\eta)$  was therefore chosen, which corresponds to a velocity associated with  $\text{Ca} = 0.1$ . This value is also a limit for the validity of the Cox model, and is thus seen as suitable. An insufficient rate of reinitialization is however known to cause the apparition of spurious currents. The set of parameters used here aims at avoiding them and usually succeeds. At large contact line velocities however, they sometimes become important. Unless noted otherwise, the sets of results displaying a noticeable amount of spurious currents are not presented or analyzed.

*c. Slip profile*

In [Figure XI](#), the slip velocity inside the liquid  $\Phi v_{slip}$  is plotted along the solid boundary for different plate velocities. Similarly to previous studies [\[4, 5\]](#), it appears that the GNBC produces a large slip in the vicinity of the contact line, and retains the same characteristics as those found by other authors.

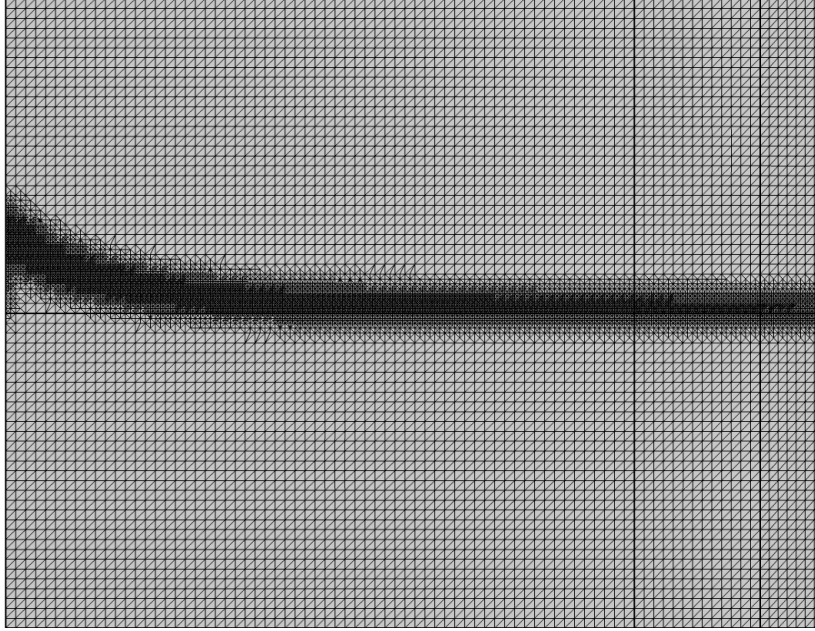


Figure X: Refined mesh obtained with a low plate velocity in a PEG 3350. The mesh displayed here contains 28563 elements, with sizes ranging from 40 to 160  $\mu\text{m}$ . The PEG 3350 occupies the bottom part of the domain.

*d. Preliminary results: contact angles*

The macroscopic contact angles obtained for a constant slip length  $\beta = h_0$  are compared to their experimental counterparts in [Figure XII](#). The obvious discrepancy between numerical and experimental contact angles, along with the dependence of  $\theta$  on  $\beta$ , lead to consider the use of a velocity-dependent slip length.

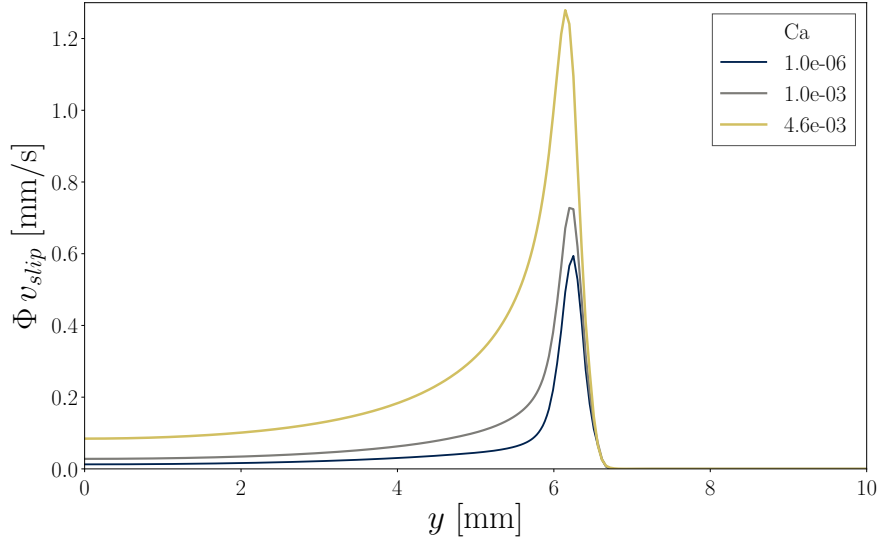


Figure XI: Slip velocity on the solid boundary for three different wall velocities. An important slip occurs across the contact line.

### C. Influence of the slip length

#### a. Slip velocity and slip length

Estimations in the numerical simulations indicate an important difference between the slip velocity and the wall velocity (Figure XIII), especially at higher wall velocities. This means that the entrainment of liquid by the solid is losing efficiency as the velocity increases, and has a number of implications, such as interface shearing. It could also mean that the capillary number associated to high velocities is overestimated.

#### b. Dynamic wetting regime transition

The separation between microscopic and viscous dissipation can be controlled by the slip length, which sets the total dissipation. Here, we plot the difference between  $\theta$  and  $\theta_\mu$  at various constant wall velocities, with relation to a continuously decreasing slip length (Figure XIV). The corresponding curves show the sudden separation between the main sources of dissipation.

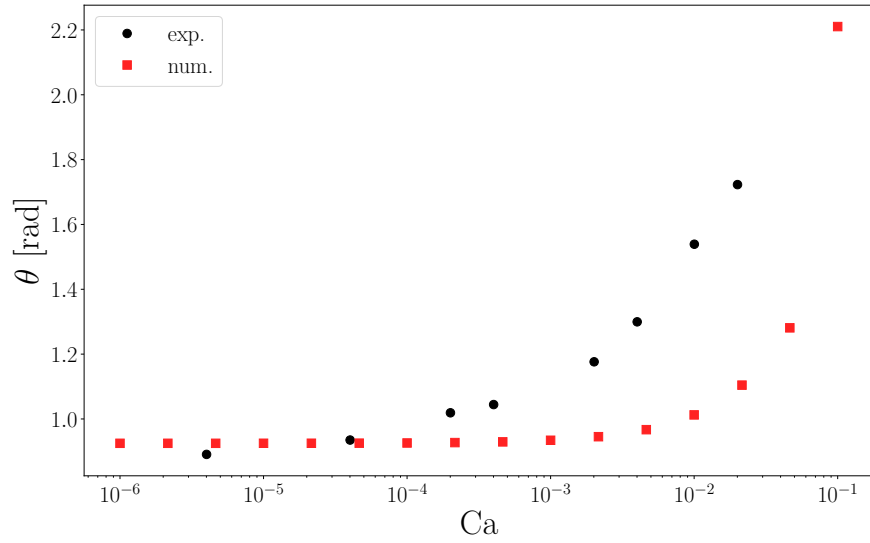


Figure XII: Dynamic macroscopic contact angle: comparison between the experimental values and the first numerical model ( $\beta = h_0$ ).

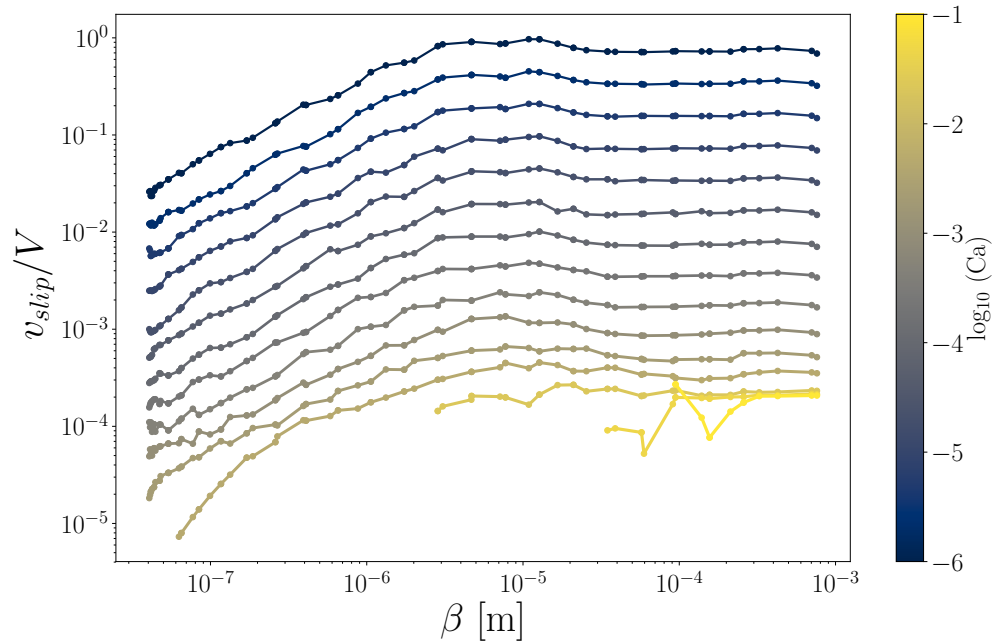


Figure XIII: Dependence of the relative slip velocity on the slip length.

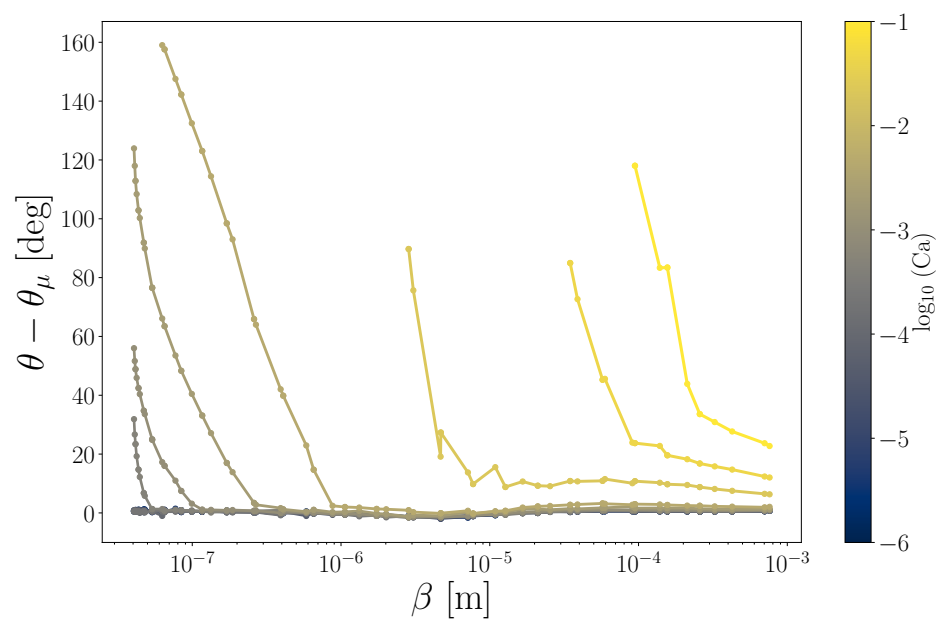


Figure XIV: Difference between the macroscopic and microscopic contact angle as a function of slip length, for different velocities.

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