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Impact of gravity and inertia on stable displacements of DNAPL in highly permeable porous media

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Abstract

The flow of Dense Non-Aqueous Phase Liquid (DNAPL) in highly permeable porous media is characterized by a complex interplay between surface tension, viscous, gravity, and inertia forces. Gravitational effects in these systems have been particularly studied in the context of displacement instability, but little work has focused on the impact of gravitational and inertia forces on the stable displacement of DNAPL in highly permeable but non-fractured porous media. Here, we study the impact of the gravity and Forchheimer numbers on the stable displacement of DNAPL fronts in porous media. We first performed DNAPL injection experiments in bead packings of different sizes for different inlet flow rates and initial saturation. These experiments were accurately modeled using a Darcy-Forchheimer model combined with an Arbitrary Lagrangian-Eulerian tracking of the DNAPL front. Once validated against stable injections in glass beads, the model was then used to

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perform a broader parametric study than available with our experimental setup. We explored different injection and pumping scenarios over a range of dimensionless numbers. We found that gravity can significantly alter the fluid front in flows commonly found in contaminant hydrology. We estimate, however, that inertia will have a non-negligible impact upon the displacement of DNAPL only during active remediation techniques (e.g., free product pumping method) or pipe rupture events involving low-viscous DNAPL in highly-permeable porous media.

Keywords: Porous Media, Inertia, Gravity, Remediation, DNAPL

Nomenclature

Abbreviations

ALE Arbitrary Lagrangian-Eulerian

DNAPL Dense Non-Aqueous Phase Liquid

IBVP Initial Boundary Value Problem

TDR Time-Domain Reflectometer

Greek

η Passability (m)

η_{ri} Relative passability

μ Dynamic viscosity (Pa s)

ϕ Porosity

ρ Density (kg m^{-3})

σ Surface tension (N m^{-1})

θ Contact angle ($^\circ$)

ε Permittivity

Latin

\mathbf{F}_{ij} Inertial correction tensor

\mathbf{K}_{ij} Multiphase permeability tensor (m^2)

\mathbf{V} Filtration velocity (m s^{-1})

\mathbf{w} Macroscopic interface velocity (m s^{-1})

Bo Bond number

Ca Capillary number

d_b Beads' diameter (m)

e Eccentricity

F_o Forchheimer number

G_r Gravity number

K_0 Absolute permeability (m^2)

k_{ri} Relative permeability

P Pressure (Pa)

P_c Capillary pressure (Pa)

r_μ Viscosity ratio

r_ρ Density ratio

Re Reynolds number

S Saturation

S_{ei} Irreducible saturation in ethanol

S_{or} Residual saturation in oil

U Characteristic velocity (m s^{-1})

1. Introduction

The flow of two immiscible fluids such as Dense Non-Aqueous Phase Liquid (DNAPL) and water in soils is ubiquitous in contaminant hydrology (Fetter et al., 1999). Our ability to model two-phase flows is important to estimate the spatio-temporal evolution of a pollutant and to evaluate the effectiveness of treatment strategies. Comparison between models and experiments have been proposed for different DNAPL's and flow scenario, including mercury (D'Aniello et al., 2018), PCE infiltration (Zheng et al., 2015), coal tar pumping (Colombano et al., 2020) or flow of a dense fluid that mimics DNAPL such as hydrofluoroether (Engelmann et al., 2021), to cite the most recent. However, two-phase flows in soils have been mostly studied in the context of petroleum extraction (Christie et al., 2001) and this literature inspired models used for DNAPL displacements (e.g. Abriola and Pinder (1985)). However, multiphase flows in petroleum engineering are different from those encountered in soil pollution applications. Porous media in oil reservoirs have in most cases a low permeability and are consolidated, whereas polluted aquifers are mostly unconsolidated and often highly permeable. Apart from soil remediation, two-phase flows in highly permeable porous media are common in hydrodynamics of packed-bed chemical reactors (de Santos et al., 1991; Attou et al., 1999) or in debris bed cooling of nuclear reactors (Tung and Dhir, 1988; Clavier et al., 2017).

In systems with high permeability and therefore larger characteristic pore sizes, viscous, gravity and inertia forces may dominate over capillary forces at pore-scale. The relative influence between these forces is captured by the capillary, Bond and Reynolds numbers. Tab. 1 gives typical values for

26 these dimensionless numbers, either for two-phase flows in consolidated low
27 permeability porous media (e.g. shale reservoir) or unconsolidated highly
28 permeable porous media commonly encountered in aquifers (e.g. sandy soils)
29 or chemical engineering (e.g. packed-bed reactors).

30 Porous media encountered in soil remediation include a wide range of
31 permeability from about 1 darcy unit for fine sand to 10^6 darcy units for
32 fractured rocks (1 darcy unit = 0.98×10^{-12} m²). Here, we are interested
33 in homogeneous unconsolidated porous media of absolute permeability be-
34 tween 300 and 3000 darcy units, which roughly corresponds to silty sand to
35 fine gravels (Freeze and Cherry, 1979). Specifically, such values are often en-
36 countered in fluvio-glacial deposits (Käser and Hunkeler, 2016; Vaudan et al.,
37 2005; Nofal et al., 2019; Theel et al., 2020), and have also been studied in
38 laboratory experiments involving DNAPL contamination (Yoon et al., 2009;
39 Wilking et al., 2013). The effect of inertia on the displacement of DNAPL
40 has been studied for flows in fractured rocks. For example, Ji et al. (2008)
41 found a nonlinear relation between pressure gradient and filtration velocity
42 starting at $Re \approx 10$ and a non-negligible impact on the flow. However, there
43 is very little work on the effect of inertia on the displacement of DNAPL in
44 highly permeable porous media. The issue is of interest because active reme-
45 diation techniques (e.g., pumping) induce velocities that can be very large,
46 especially in highly permeable media.

47 Finally, the interplay of different forces in highly permeable media has a
48 direct impact on modeling. For stable flows (notion discussed in § 2.2), the
49 fluid front may be sharp enough to justify modeling strategies based on an
50 explicit description of the movement of a macroscopic front, rather than the

Table 1: Typical range of values for capillary, Bond and Reynolds numbers for different porous media. Modified from De Santos et al. (1991).

Porous media	$Ca = \frac{\text{viscosity}}{\text{capillary}}$	$Bo = \frac{\text{gravity}}{\text{capillary}}$	$Re = \frac{\text{inertia}}{\text{viscosity}}$
Consolidated	$10^{-7} - 10^{-3}$	$10^{-9} - 10^{-2}$	$10^{-9} - 10^{-2}$
Non-consolidated	$10^{-2} - 10$	$10^{-1} - 10$	$10^{-2} - 10^2$

51 traditional hyperbolic equation models that produce diffuse front solutions
52 due to non-linear mobilities and, therefore, need constitutive relations. The
53 interest of using a sharp front-tracking model is twofold: i) it is more rep-
54 resentative of the experimentally observed front when the front is a clearly
55 defined interface (which is the case in this study), ii) it only requires to know
56 the residual saturation of the displacement to be modeled, and thus saves
57 time compared to the drainage and imbibition cycles needed to find the pa-
58 rameters of the constitutive relations. This study develops such an explicit
59 front displacement model to reproduce laboratory-scale experiments. We
60 then use this model to study the effects of gravity and inertia upon the fluid
61 front and to evaluate whether these effects can be encountered in real-life
62 applications in contaminant hydrology. In addition, decimeter-scale experi-
63 ments of stable injection in glass beads packing were carried out in laboratory
64 to validate the mathematical model.

65 2. Theoretical background

66 Theoretical background for this work consists in two main part, namely
67 the equations that govern the two-phase flow displacement with inertia and

68 the stability of two-phase displacements. We summarily turn to both subjects
69 in that respective order in the two following sections.

70 *2.1. Governing equations for two-phase flows at Darcy-scale*

71 Several macro-scale models describing two-phase flows in porous media
72 including inertia effects have been proposed in the literature following ex-
73 tensions to Darcy's law proposed by Ergun (1952) or Forchheimer (1901).
74 Generally, the extension consists in correcting the linear relation between
75 the pressure gradient and the filtration velocity with a new term which takes
76 into account the supplementary resistance due to inertial flows. This term
77 scales as the filtration velocity magnitude raised to the power of n , where
78 $2 \leq n \leq 3$. The more general model was obtained from upscaling techniques.
79 Lasseux et al. (2008) used the volume-averaging technique to upscale the
80 Navier-Stokes equations that govern two-phase flows at pore-scale. The au-
81 thors extended the previous works on macroscopic creeping two-phase flows in
82 homogeneous (Marle, 1982; Whitaker, 1986; Auriault and Sanchez-Palencia,
83 1986; Lasseux et al., 1996) and heterogeneous porous media (Quintard and
84 Whitaker, 1988), and derived macroscopic equations for homogeneous media
85 taking inertia effects into account. The complete model, not recalled here,
86 involved primary and coupling multiphase permeability tensors, as well as
87 primary and coupling inertia correction tensors.

88 Clavier et al. (2017) recently conducted column two-phase flow experi-
89 ments and proposed constitutive relations for each relative permeability and
90 inertia correction term. As pointed out in Davit and Quintard (2019), de-
91 termining all the terms is very difficult and is still largely an ongoing work.
92 In this study, we found low irreducible saturation in water, partly because

93 of the very low interfacial tension between the chosen fluids. By considering
 94 the traditional curves for coupling permeabilities (Bacri et al., 1990; Roth-
 95 man, 1990; Dullien and Dong, 1996), we estimated that coupling between
 96 the fluids is negligible because these are significant only for intermediate to
 97 high saturation in wetting fluid ($0.7 \lesssim S_w$). Consequently, we used a simpler
 98 model that does not take into account coupling permeabilities and coupling
 99 inertia correction.

100 Furthermore, we assumed that the model apparent permeability obeys a
 101 simple form, namely the extension of the well known Ergun’s law (Ergun,
 102 1952) for two-phase flows (Fourar et al., 2001). Consequently, continuity and
 103 momentum transport equations read

$$\nabla \cdot \mathbf{V}_i = 0 \quad i = w, n, \quad (1)$$

104

$$\nabla P_i = -\mathbf{V}_i \left(\frac{\mu_i}{K_0 k_{ri}} + \frac{\rho_i}{\eta \eta_{ri}} \|\mathbf{V}_i\| \right) + \rho_i \mathbf{g} \quad i = w, n, \quad (2)$$

105 where k_{ri} and η_{ri} are the relative permeability and passability of phase i , re-
 106 spectively, and η is the passability. We considered a homogeneous isotropic
 107 porous medium, that allows us to write the primary multiphase permeability
 108 as the product between a scalar relative permeability and a scalar absolute
 109 permeability K_0 (Bear et al., 1987; Quintard and Whitaker, 1988). Lipin-
 110 ski (1982), Schulenberg and Müller (1987) and Hu and Theofanous (1991),
 111 among others, have proposed correlations for relative permeabilities and rela-
 112 tive passability, which depend on saturation and porosity. Ergun’s correlation
 113 (Ergun, 1952) for η is used in this work and reads

$$\eta = \frac{\phi^3 d_b}{1.75(1 - \phi)}. \quad (3)$$

114 We non-dimensionalized Eq. 2 with the following,

$$\mathbf{x} = L\mathbf{x}^*, \mathbf{V} = U\mathbf{V}^*, t = t^*(L/U), P = P^*\frac{\mu_n UL}{K_0}, \quad (4)$$

115 where the star exponent indicates dimensionless quantities. L and U are
 116 characteristic length and filtration velocity, respectively. The dimensionless
 117 equation, considering homogeneous and isotropic media, reads

$$\nabla^* P_i^* = -\mathbf{V}_i^* \left(\frac{r_{\mu i}}{k_{ri}} + r_{\rho i} F_{oi} \|\mathbf{V}_i^*\| \right) + r_{\rho i} G_r \mathbf{e}_z, \quad i = w, n, \quad (5)$$

118 where

$$G_r = \frac{\rho_n g K_0}{\mu_n U}, \quad (6)$$

119 is the gravity number, which compares the effect of gravity forces to the effect
 120 of the viscous forces at macroscopic scale,

$$F_{oi} = \frac{\rho_n U K_0}{\mu_n \eta_{ri} \eta} \quad i = w, n, \quad (7)$$

121 is the Forchheimer number (Ruth and Ma, 1992) for fluid i , which indicates
 122 the onset of significant macroscopic inertia effects (Ma and Ruth, 1993), and

123

$$r_\mu = \mu_w / \mu_n, \quad r_\rho = \rho_w / \rho_n, \quad (8)$$

124 are the viscosity and the density ratios, respectively.

125 2.2. Flow stability

126 Front displacement instabilities, by favoring the creation of preferential
 127 flow paths, also called fingers, make it very difficult to model the migration
 128 of a DNAPL (NAPL denser than water) from a source zone with Eqs. ??-??.
 129 We commonly refer to the different instabilities as gravity, viscous or capillary

130 fingerings, depending on the value of the dimensionless numbers and ratios
 131 of fluids viscosity and density (Saffman and Taylor, 1958; Lenormand et al.,
 132 1988). For DNAPL, gravity fingering has received a lot of attention (Schwille
 133 and Pankow, 1988; Glass and Nicholl, 1996; Nsir et al., 2012). Because the
 134 DNAPL is denser than water, its infiltration into an aquifer may lead to
 135 gravity fingering, depending on the magnitude and direction of DNAPL ve-
 136 locity. Among solutions to reproduce gravity fingering, Cueto-Felgueroso and
 137 Juanes (2009) introduced an additional fourth-order derivative of saturation
 138 in terms of the spatial coordinates. Saffman and Taylor (1958) and Chuoke
 139 et al. (1959) conducted a linear-stability analysis for a sharp invading front
 140 between two immiscible fluids in the absence of capillary forces. They gave
 141 a criterion for stability, which expresses the interplay between viscous and
 142 gravity forces. The Saffman-Taylor/Chuoke criterion is expressed in terms
 143 of V_c , the critical velocity of the fluid front between the invading fluid (*in*)
 144 and the displaced fluid (*d*),

$$|V_c| = \frac{\cos(\beta) (\rho_{in} - \rho_d) g K_0}{\phi(\mu_{in} - \mu_d)}, \quad (9)$$

145 where β is the angle formed by the gravitational vector and the direction of
 146 the flow. Tab. 2 lists the different stability scenarios. Among these scenarios,
 147 the intrusion of a DNAPL more viscous than water into a water-saturated soil
 148 is either conditionally stable for a downward flow or unconditionally stable for
 149 an upward flow. In practice, downward flows are observed when the DNAPL
 150 freely migrates from a source zone above the water table, whereas upward
 151 flows are observed in case of pressurized pipe ruptures below the water table,
 152 DNAPL pumping or groundwater upwelling. We only deal with stable flows
 153 in the following.

Table 2: Stability of two-phase flows, adapted from Glass and Nicholl (1996). V is the velocity of the invading front between the invading in and the displaced d fluids. The critical velocity may be either negative (downward flow) or positive (upward flow).

V_c	Conditionally stable		Unconditionally unstable	Unconditionally stable
< 0	$\mu_{in} < \mu_d,$ $\rho_{in} < \rho_d$	$\mu_{in} > \mu_d,$ $\rho_{in} > \rho_d$	$\mu_{in} < \mu_d,$ $\rho_{in} > \rho_d$	$\mu_{in} > \mu_d,$ $\rho_{in} < \rho_d$
	$ V < V_c $ $ V > V_c $			
> 0	$\mu_{in} < \mu_d,$ $\rho_{in} > \rho_d$	$\mu_{in} > \mu_d,$ $\rho_{in} < \rho_d$	$\mu_{in} < \mu_d,$ $\rho_{in} < \rho_d$	$\mu_{in} > \mu_d,$ $\rho_{in} > \rho_d$
	$ V < V_c $ $ V > V_c $			

154 **3. Experimental method**

155 The experimental setup and protocols were designed to produce a stable
156 displacement with an upward flow of DNAPL in a porous medium saturated
157 with the wetting fluid. The complete laboratory-scale (decimetric) setup is
158 described below and shown in Fig. 1.

159 *3.1. 2D tank and pumping system*

160 The porous medium was embedded in the main reservoir of a tank made
161 in polyvinylidene fluoride (PVDF). Two counter-channels positioned on both
162 lateral sides allowed to keep the pressure head constant. These counter-
163 channels were separated from the main reservoir by thin perforated metal
164 grids. The front part of the tank was made of glass in order to monitor
165 the two-phase displacement with photography. However, the back part was
166 made with PVDF to allow inserting a network of Time-Domain Reflectometer
167 (TDR) sensors (Decagon Devices 5TE 40567). The dimensions of the main
168 reservoir were: 50 cm long, 30 cm high and 7 cm thick. Five nozzles of
169 diameter 3.125 cm, at the bottom of the tank, allowed the connection of
170 tubes to inject the fluids. Given the tank depth and the size of the inlet,
171 small 3D effects near the nozzle may occur, but Philippe et al. (2021) have
172 shown that this effect remained negligible using the same apparatus and
173 fluids.

174 The injection system consisted of two peristaltic pumps (Watson Marlow
175 530U). One was dedicated to the injection of the oil by a bottom nozzle while
176 the other was connected to the two lateral tank reservoirs and maintained a
177 constant level of ethanol during the injection. During the injection, the in-

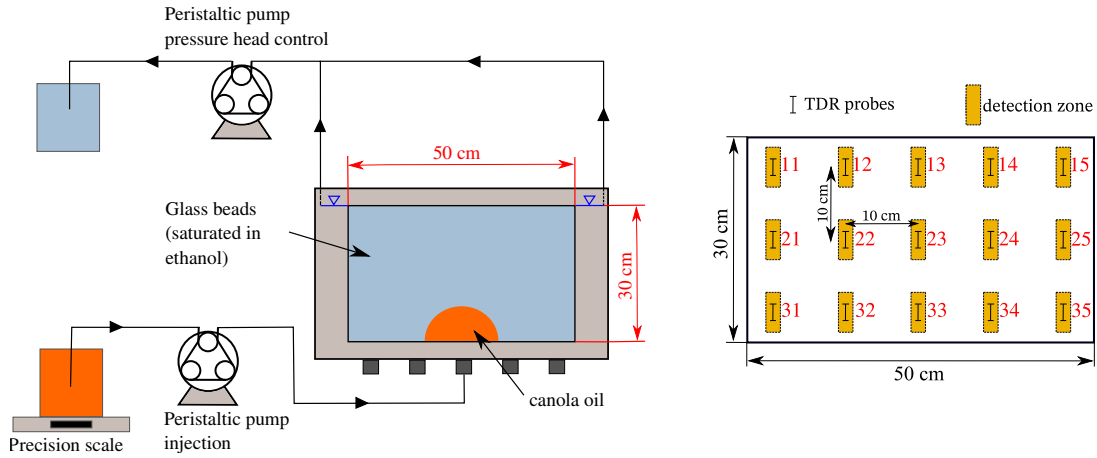


Figure 1: Left: schematic front view of the experimental 2D tank setup. One peristaltic pump is connected to the two lateral tank reservoirs and controls the pressure head, while a second pump injects the DNAPL in the main reservoir. This reservoir was filled with glass beads and initially saturated in wetting fluid. The precision scale recorded the injected mass of DNAPL. Right: arrangement of the 3×5 TDR probes in the main reservoir. The labeling is done following the row/column indice. The orange zone represents the detection area of the probes.

178 vading fluid was pumped from a beaker placed on a precision scale (Sartorius
 179 Cubis MSE ± 0.1 g). The mass of fluid injected was recorded every 300 ms
 180 and the inlet flow rate was calculated accordingly. Fig. 1 shows a sketch of
 181 the complete pumping setup.

182 3.2. Porous media and fluids

183 The unconsolidated porous medium was a packing of homogeneous glass
 184 beads of diameter d_b . We conducted different experiments using either 1
 185 mm or 2 mm diameter glass beads to study the impact of permeability and
 186 gravity forces during the injection. We measured the same porosity for each
 187 diameter ($\phi \approx 0.4$), and $K_0 = 3.5 \times 10^{-10}$ m² and $K_0 = 4.2 \times 10^{-9}$ m² for the

Table 3: Dynamic viscosity, density, surface tension and contact angle of the canola oil and ethanol used in the experiments (20°C) (Philippe et al., 2020). Viscosity ratio is $\mu_o/\mu_e = 61$ and density ratio is $\rho_o/\rho_e = 1.12$.

Properties	Canola oil (o)	Ethanol (e)
μ (Pa s)	1.18×10^{-1}	1.93×10^{-3}
ρ (kg/m ³)	911	808
$\sigma_{o/e}$ (N/m)	2.2×10^{-3}	
$\theta_{o/e}$	127°	

188 absolute permeability of the 1 mm and 2 mm glass beads, respectively.

189 We used ethanol 95% as the wetting invaded fluid and canola oil as the
 190 non-wetting invading fluid. We selected this fluid pair because the ratio
 191 between the different fluid properties is very close to a coal tar-water pair
 192 (Philippe et al., 2020). On the other hand, these fluids are non-toxic unlike
 193 the initial coal tar, which eased manipulation. Dynamic viscosity, density,
 194 surface tension, and contact angle at 20°C are given in Table. 3. The canola
 195 oil stood for the DNAPL, compared to ethanol, and the viscosity ratio en-
 196 sured an unconditionally stable displacement. These two fluids have very
 197 low solubility (Rao and Arnold, 1956), which allowed precise identification
 198 of the fluid front, considering the time scale of the experiments (less than 20
 199 minutes at a controlled temperature room of around 20°C).

200 3.3. Monitoring system

201 The monitoring of the fluid front was made in two ways:

- 202 1. By acquiring the averaged saturation around TDR probes that are dis-
 203 posed as an array of 3×5 probes. We show in Fig. 1 (right) the

204 arrangement of the probes and their labeling, following a row/column
 205 identification (tdr_{ij} refers to the row i /column j probe). The TDR
 206 probes measured the averaged relative permittivity in the area close
 207 (up to 2 mm away) to the probes. The measured averaged relative
 208 permittivity $\bar{\varepsilon}$, acquired every 30 s, corresponds to the relative permit-
 209 tivity of the mixture of the three phases. The saturation in ethanol was
 210 then obtained through the permittivity ε_o of the mixture {glass beads
 211 + oil} and ε_e for the mixture {glass beads + ethanol}, then considering
 212 a linear relationship between these two endpoints,

$$S_w = \frac{\bar{\varepsilon} - \varepsilon_o}{\varepsilon_e - \varepsilon_o}, \quad (10)$$

213 where ε_e and ε_o are specific to each TDR probe and for each diameter of
 214 glass beads. We suggested a linear relationship because the difference
 215 between the two endpoints (porous media saturated with oil or ethanol)
 216 relative permittivity is small ($\varepsilon_e \simeq 6$ and $\varepsilon_o \simeq 14$) and that it gives a
 217 good approximation of the commonly used power law model (Complex
 218 Refractive Index Model, CRIM) (Birchak et al., 1974). This last has
 219 been proved to be accurate for a DNAPL/mineral/water mixture (Ajo-
 220 Franklin et al., 2004; Colombano et al., 2021a).

221 2. By acquiring the fluid front after post-processing the photos. We used
 222 a digital camera Nikon D810 with NIKKOR LENS 105, and a setup
 223 including two floodlights and black and white reflectors, to avoid any re-
 224 flections and to optimize the contrast. To obtain a homogeneous light-
 225 ing, experiments were performed in a dark room and the only source
 226 of light came from the two floodlights. Photos were taken each 30 s as
 227 soon as the injection tube was filled with oil. All photos were acquired

228 in RAW Nikon format (.nef). The post-processing was made with Im-
229 ageJ, an open-source image processing program (Schneider et al., 2012).
230 The process of going from raw photos to the interface is as follows. We
231 first converted the raw photos to .tiff format with RawTherapee soft-
232 ware and then to 8-bit format with ImageJ. A gray scale on the side
233 of the tank ensures that the min and max values are set correctly for
234 linear scaling to 256 shades of gray. Next, a px to cm conversion scale
235 was provided to ImageJ based on the graduations on each side of the
236 tank. The position of the interface was finally determined manually
237 using the difference in shades of gray, as was done in (?), and recorded
238 as the 2D coordinates of a set of points.

239 3.4. *Experimental protocols*

240 Fig. 2 shows the flowchart of the experimental protocols. The first one
241 involved performing several injections of canola oil from the center bottom
242 hole in glass beads ($d_b=1$ and 2 mm) fully saturated with ethanol (as schemat-
243 ized in Fig. 1). The flow rate was also changed for glass beads $d_b=1$ mm.
244 Another experiment, performed with $d_b=2$ mm, consisted in injecting the oil
245 in the same way as before but in glass beads with an initial field of residual
246 oil saturation. Starting from glass beads saturated with ethanol, the residual
247 oil saturation was created by performing a prior drainage, followed by a final
248 imbibition.

249 For each experiments, a volume of glass beads of very small diameter (0.1
250 mm) was poured to fill the dead volume between the bottom of the tank
251 and the top of the injection nozzle (about 1.5 cm) to prevent the flow of
252 DNAPL below the injection point. Then, the packing was made by pouring

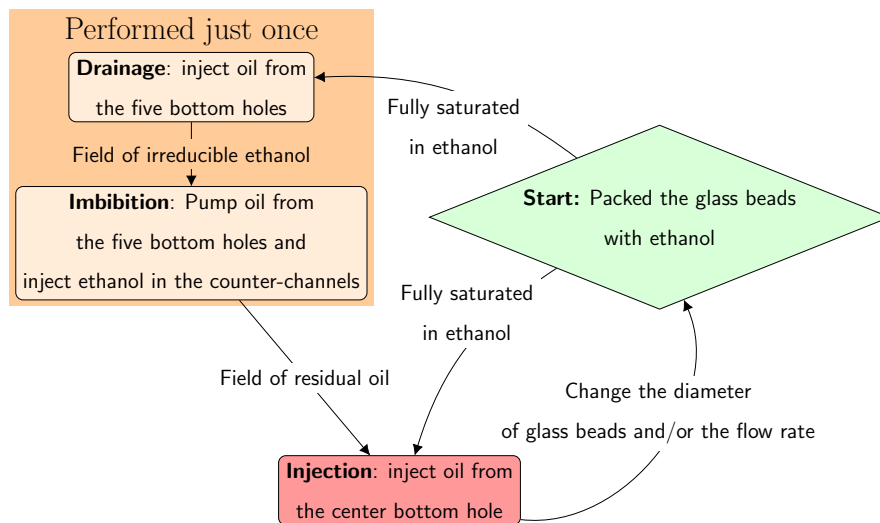


Figure 2: Flowchart of the protocols used in this study. One experiment was dedicated to injecting the oil in the glass beads with an initial residual saturation in oil. The other experiments involved injecting the oil in a fully saturated porous media for different size of glass bead and flow rate.

253 in several times a layer of beads into a volume of ethanol to obtain a more
 254 homogeneous packing.

255 4. Numerical method

256 The experimental results showed that the displacement front is sharp.
 257 Generalized Darcy's laws produce diffusive fronts, although shocks may be
 258 obtained if there is no capillary pressure and the diffusive front is due to
 259 mobility non-linearity. Effective properties in generalized Darcy's law may
 260 be tweaked to obtain a diffuse interface method as shown in ???. However,
 261 given the fact that the geometry remains very simple, experimental results
 262 suggest that a more direct Lagrangian method may be more effective. We

263 therefore adopted an Arbitrary Lagrangian-Eulerian method (ALE), which is
 264 described in the next section along with its implementation. For comparison
 265 we have also reproduced the experimental results with the generalized Darcy
 266 model.

267 *4.1. Equations*

268 ALE methods are commonly used to track interfaces between deforming
 269 domains, for example in fluid-structure interaction problems (Hu et al., 2001).
 270 The geometry and the mesh can change, given a set of constraints. In our
 271 case, the interface between the two fluid domains was deformed under the
 272 computed Darcy-velocity field and the displacement of the mesh nodes was
 273 computed using Laplace’s equation

$$\nabla^2 \mathbf{X} = 0, \quad (11)$$

274 where \mathbf{X} are the cartesian coordinates of the material frame. The differential
 275 operator is defined in respect to the Cartesian coordinates of the spatial
 276 frame.

277 We solved the continuity equation and a single-phase Darcy’s law with
 278 Forchheimer correction. For a homogeneous isotropic medium, the equations
 279 read

$$\nabla \cdot \mathbf{V} = 0, \quad \nabla P = -\mathbf{V} \left(\frac{\mu_i}{K_0} + \rho_i \eta^{-1} \|\mathbf{V}\| \right) + \rho_i g \mathbf{e}_z, \quad (12)$$

280 where the material properties μ and ρ are attached to a domain (fluid) and
 281 η is the passability, which depends only on the porous medium. As pointed
 282 out in the recent review in Davit and Quintard (2019), equation 12 has
 283 limitations. For instance, it does not take into account anisotropy for the
 284 inertia corrections (see e.g. Lasseux et al. (2011)). The quadratic correction is

285 also only an approximation and other inertia regimes exist (Mei and Auriault,
 286 1991; Wodie and Levy, 1991; Firdaouss et al., 1997; Lasseux et al., 2011).
 287 However, Ergun’s equation 12 remains a simple and widely used model that
 288 takes into account the additional drag from inertia with good accuracy.

289 These equations were solved using a finite element approach with a poly-
 290 nomial (quadratic) interpolation function for the pressure and time step-
 291 ping was governed by an implicit Backward Differentiation Formula (BDF)
 292 method. The whole numerical model was solved using Comsol Multiphysics®.

293 We also solved the generalized Darcy equations to compare with the ex-
 294 perimental results. The model reads

$$\phi \frac{\partial S_i}{\partial t} + \nabla \cdot \mathbf{V}_i = 0, \quad (13a)$$

$$\mathbf{V}_i = -\frac{k_{ri}(S_w, \dots) \mathbf{K}_0}{\mu_i} \cdot (\nabla P_i - \rho_i g \mathbf{e}_z), \quad (13b)$$

$$S_w + S_n = 1, \quad (13c)$$

$$P_w = P_n. \quad (13d)$$

298 Introducing Eq. 13b into Eq. 13a and using Eq. 13c and Eq. 13d, we
 299 eliminate two unknowns and choose as primary variables the pressure of the
 300 non-wetting fluid and the wetting fluid saturation. We obtain the following
 301 non-linear parabolic system,

$$\phi \frac{\partial S_w}{\partial t} = \nabla \cdot \left[-\frac{k_{rw}(S_w) \mathbf{K}_0}{\mu_w} \cdot (\nabla P_n - \rho_w g \mathbf{e}_z) \right], \quad (14a)$$

$$\phi \frac{\partial (1 - S_w)}{\partial t} = \nabla \cdot \left[-\frac{k_{rn}(S_w) \mathbf{K}_0}{\mu_n} \cdot (\nabla P_n - \rho_n g \mathbf{e}_z) \right], \quad (14b)$$

302 which is the starting point to solve two-phase flows in different multiphase
 303 flow solvers at Darcy-scale (Flemisch et al., 2011; Horgue et al., 2015).
 304 Here, we used the OpenFoam toolbox developed by Horgue et al. (2015),
 305 in which an IMPES algorithm is adopted. We used van Genuchten relations
 306 (Van Genuchten, 1980) for the relative permeabilities. The appropriate co-
 307 efficients for the pair of liquids used in this study in 1 mm glass beads were
 308 measured by Philippe et al. (2020). To be consistent with the experimental
 309 observations and ALE modeling, we neglected the capillary pressure as well
 310 as the coupling relative permeabilities.

311 4.2. Geometry and boundary conditions

312 Fig. 3 shows the geometry used in the numerical study. A pressure head
 313 condition was set on the left/right boundaries (pressure of ethanol). A null
 314 flux was set on the bottom boundary (i.e., the top of the layer of 0.1 mm
 315 glass beads), except at the inlet, where a given flux is imposed following the
 316 measured $U_{2D}(t)$ obtained previously. We also used a pressure condition at
 317 the inlet to model a pumping scenario, more suitable to emphasize the impact
 318 of inertia upon the shape of the oil front in that case. As the interface was
 319 handled explicitly, we imposed a boundary condition, derived in Appendix
 320 A, which reads

$$\mathbf{w} \cdot \mathbf{n} = \frac{1}{\phi(1 - S_{ei} - S_{or})} \mathbf{V} \cdot \mathbf{n}, \quad (15)$$

321 where \mathbf{w} is the velocity of the interface, \mathbf{n} the normal vector at the interface,
 322 and S_{ei} and S_{or} are those obtained from the TDR probes (Tab. 6). The
 323 initial fluid configuration was a half-ellipse corresponding to the interface
 324 measured at the initial time (2 min after the beginning of the injection) of
 325 each experiment.

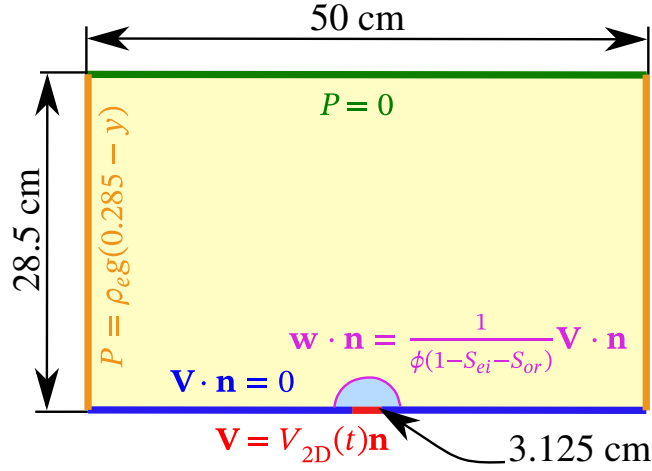


Figure 3: Geometry and label of the boundaries. Dirichlet conditions were set on the left and right boundaries (pressure head), and the top boundary (reference pressure). Neumann conditions were set on the inlet boundary (from the relation $U_{2D}(t)$) and on the bottom boundary (null flux). The condition at the fluid front depends on the Darcy-velocity field.

326 4.3. Mesh sensitivity analysis

327 The mesh sensitivity analysis is based on the position of the interface
 328 between the two fluids after 11 min of injection. The inlet velocity is constant
 329 in time, the residual and irreducible saturation are zero and the porosity is
 330 the same as for the actual porous media ($\phi = 0.4$). The absolute permeability
 331 is $K_0 = 5 \times 10^{-9} \text{ m}^2$ to test the mesh convergence with non-negligible gravity
 332 forces. An automatic remeshing procedure allowed us to keep the global mesh
 333 quality higher than a given threshold, which is mandatory to avoid element
 334 degeneration. Fig. 4 shows the position of the front at the final time for three
 335 different mesh sizes. The position of the front was identical regardless of the
 336 number of mesh elements. We chose the finest mesh, as computational time
 337 is low even for the finest mesh.

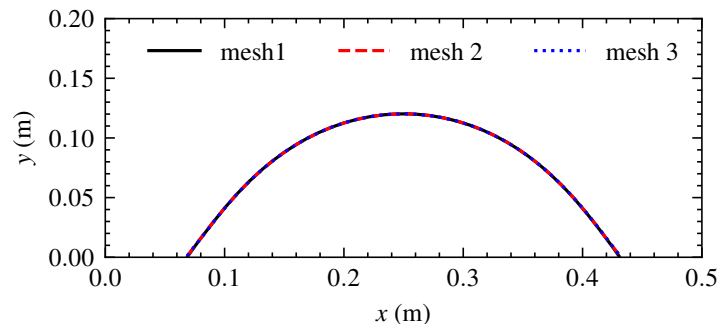


Figure 4: Comparison of the position of the interface for three different meshes. Mesh 1, 2, and 3 are made of 2,150, 23,600, and 52,400 free triangular mesh elements, respectively.

338 5. Experimental results: laboratory-scale injections

339 We first analyze the data from the precision scale, from which we can
 340 derive the inlet flow rate. From the flow rate and the fluid and porous media
 341 properties, we calculate the dimensionless numbers that characterized the
 342 two-phase displacements. Finally, post-processing of TDR data and pho-
 343 tos allow us to characterize the two-phase displacement, providing insight
 344 regarding which model is suitable to reproduced the experiments.

345 5.1. Mass balance analysis

346 We injected between 18% and 32% of the pore volume after twelve minutes
 347 of injection (based on porosity $\phi = 0.4$), depending on the flow rate. Two
 348 injections in 2 mm and 1 mm glass beads were similar, whereas a second
 349 injection in 1 mm was conducted with a higher flow rate. In the following,
 350 we refer to these 1 mm experiments as 1 mm Q_l and 1 mm Q_h , respectively.

351 The corresponding 2D velocity was calculated based on the flow rate,
 352 the tank's depth and the diameter of the inlet hole. Fig. 5 shows the 2D

353 velocity as a function of time. We obtained the relation $V_{2D}(t)$ by linear
354 regression over the time interval 120 s to 690 s. These relations were used
355 for the numerical modeling. V_{2D} slightly decreased with the injection time,
356 and the difference in injection velocity between 1 mm Q_h and 1 mm Q_l
357 experiments was about 15%. The decreasing trend can be imputed to the
358 increasing pressure head, as ethanol was replaced by oil, which is denser.
359 The y-intercept values V_{2D} were used to compute the relevant pore-scale and
360 Darcy-scale dimensionless numbers.

361 Pore-scale and Darcy-scale dimensionless numbers for each experiment
362 are listed in Tab. 5. Reynolds and capillary numbers were about the same
363 order of magnitude for each experiment and sufficiently low for considering
364 that the hypothesis of creeping flow with a low capillary number at pore-scale
365 was fulfilled. On the other hand, Bond number is up to 1.8 for 2 mm ex-
366 periments, so within the limits of the assumptions underlying the continuous
367 macroscale model used in the following. An order of magnitude separates the
368 gravity numbers for the 1 mm and 2 mm experiments, suggesting different
369 large-scale dynamics between these experiments. The Forchheimer number
370 was very low for each experiment, which suggests negligible macroscopic in-
371 ertia effects in our experiments. This is confirmed with previous studies that
372 found a negligible error on the pressure drop when calculated with Darcy's
373 law for Forchheimer numbers below 10^{-1} (Zeng and Grigg, 2006; Macini
374 et al., 2011).

375 *5.2. TDR analysis*

376 TDR probes were used to study the dynamic of the injection but also to
377 obtain the irreducible saturation in ethanol S_{ei} and the residual saturation in

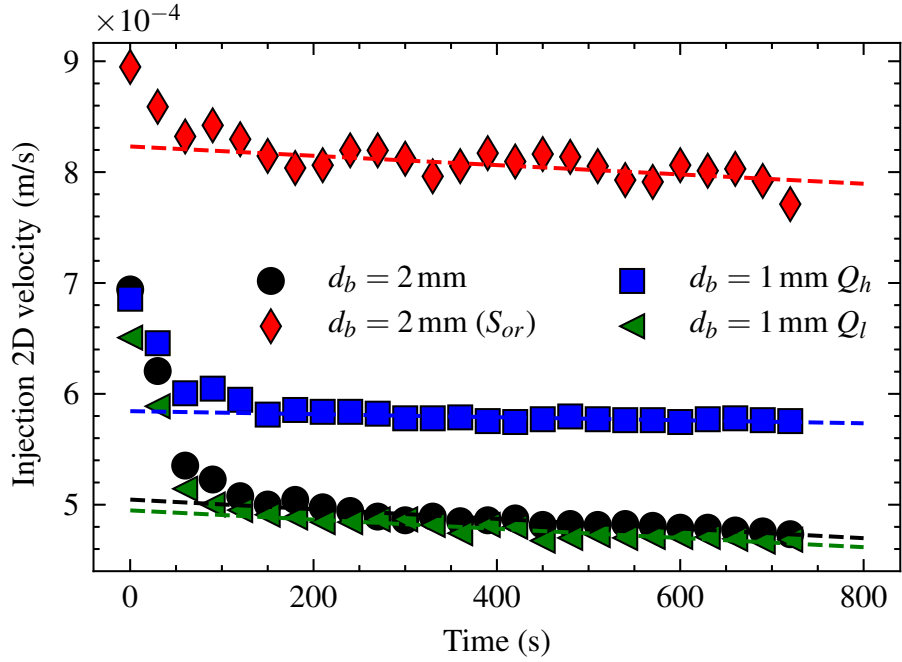


Figure 5: Injection 2D velocity V_{2D} as a function of time for each experiment. V_{2D} was calculated by dividing the injection flow rate by the tank's depth and the diameter of the inlet hole. The relations $V_{2D}(t)$ were obtained by linear regression over the time interval 120 s to 690 s, and were used as a boundary condition for the numerical model. The y-intercept value was used to calculate dimensionless numbers.

Table 4: Pore-scale dimensionless numbers for each experiment.

Experiment	$Re = \frac{V_{2D}\rho_o d_b}{\phi\mu_o}$	$Ca = \frac{V_{2D}\mu_o}{\phi\sigma}$	$Bo = \frac{\Delta\rho g d_b^2}{\sigma}$
2 mm	1.9×10^{-2}	2.7×10^{-2}	1.8
2 mm S_{or}	3.2×10^{-2}	4.4×10^{-2}	1.8
1 mm Q_h	0.9×10^{-2}	3.1×10^{-2}	0.4
1 mm Q_l	1.1×10^{-2}	2.6×10^{-2}	0.4

Table 5: Darcy-scale dimensionless numbers, as defined as in Eqs. 6-7, for each experiment. Here, $\eta_{ri} = 1$ in fluid i .

Experiment	G_r	F_o
2 mm	31.5×10^{-2}	6.7×10^{-5}
2 mm S_{or}	19.3×10^{-2}	10.9×10^{-5}
1 mm Q_h	4.5×10^{-2}	2.6×10^{-5}
1 mm Q_l	5.3×10^{-2}	2.2×10^{-5}

oil S_{or} . The irreducible saturation and the residual oil saturation were used for the modeling part. Fig. 6 shows the saturation given by the three TDR probes 32 to 34 (middle columns, lower row, see Fig. 1) for each experiment. Based on probes 32 and 34, we see that the evolution of the front was symmetrical on each side of the injection point, except for the injection in the 1mm Q_h glass beads for which the right side was slightly ahead.

Irreducible ethanol saturation and residual oil saturation were obtained by averaging the results from probes 32 to 34 at the final time. Tab. 6 summarizes all the results. We measured a lower irreducible saturation for a lower flow rate. The difference is noticeable, between 12% and 6%, for 1mm Q_h and 1mm Q_l , respectively, although the difference in inlet flow rate remained moderate for these two injections. For the same flow rate, the irreducible saturation was higher in the more permeable medium (2 mm beads). However, the small number of values and the dispersion between these values does not allow to be categorical on this point. Finally, we found that the irreducible ethanol saturation is lower for the experiment with 2 mm glass beads with an initial residual oil saturation (about 22% of residual oil)

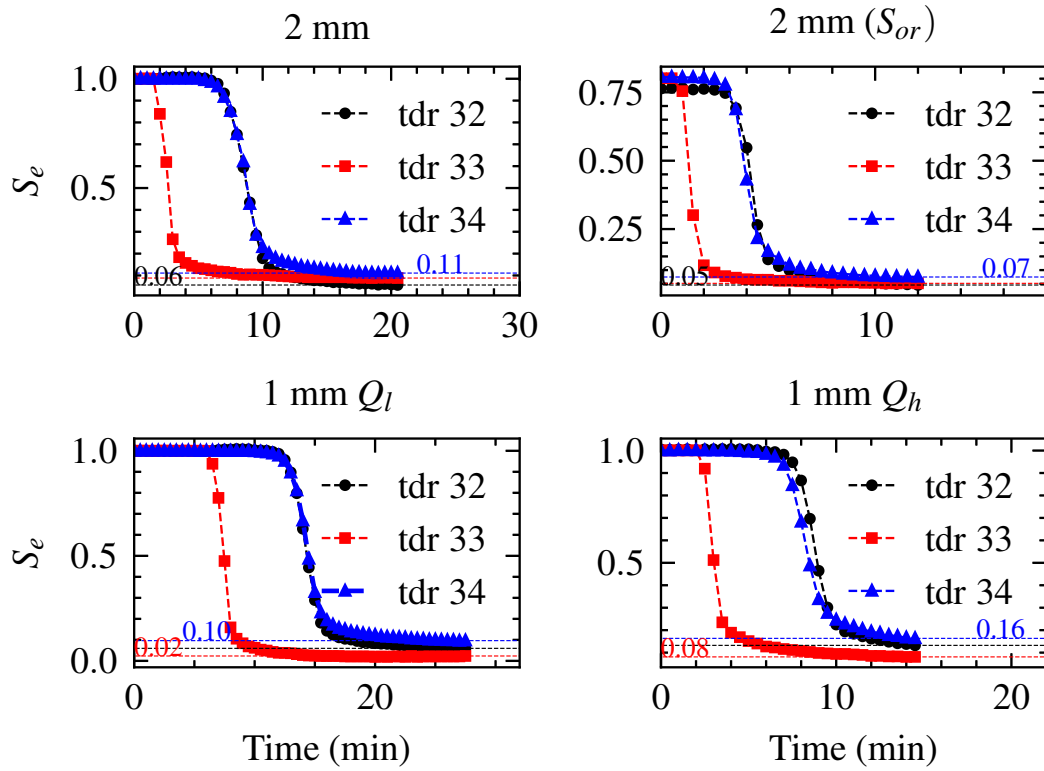


Figure 6: Evolution of the saturation given by the three TDR (center columns/bottom row). The final value indicates the irreducible ethanol saturation in the oil zone.

Table 6: Estimation of the irreducible saturation in ethanol for the different experiments. Results obtained from the means of the three TDR probes (32-33-34).

Experiment	S_{ei}	S_{or}
2 mm	$8.7\% \pm 2.0\%$	-
2 mm (S_{or})	$5.6\% \pm 0.9\%$	$22.3\% \pm 2.0\%$
1 mm Q_h	$12.3\% \pm 3.3\%$	-
1 mm Q_l	$6.0\% \pm 3.2\%$	-

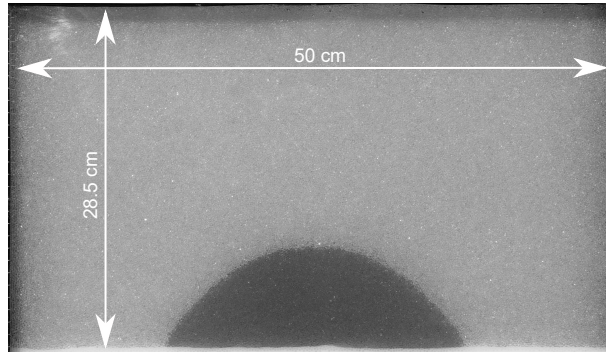
395 compared to the fully saturated case, despite the higher flow rate.

396 5.3. Photos analysis

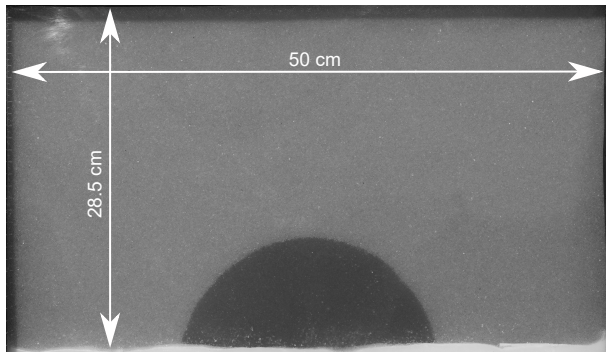
397 From the photos¹ (see Fig. 7a and Fig. 7b showing as an example the
 398 injected oil after 7 minutes, in 2 mm and 1 mm Q_l glass beads, respectively),
 399 we observed a flattened half-disk shape of the oil zone for 2 mm glass beads.
 400 This shape is due to the impact of gravitational forces on the injection pro-
 401 cess. In contrast, the shape of the oil zone looks like a half-disk in the case
 402 of injection in 1 mm glass beads (at both flow rates).

403 We also observed that the fluid front was very well-defined and sharp for
 404 each injection. The front spanned only over a few glass beads at most, and the
 405 displacements were stable due to the fluid properties and the experimental
 406 protocol. Consequently, all injections can be modeled by the advance of a
 407 macroscopic front. This is an important feature, which allowed us to use a

¹All the photos and the fluid front coordinates are available as supplementary material in the associated data repository: <https://data.mendeley.com/datasets/7njyh65w6/draft?a=ef3a9a24-ae72-4260-bb3b-62a81b210329>



(a) Saturation field at the time $t = 7$ min for 2 mm injection.



(b) Saturation field at the time $t = 7$ min for 1 mm Q_I injection.

Figure 7: Photos of oil saturation at the time $t = 7$ min. The thin layer of very low permeable beads (0.1 mm) allows to close the 1.5 cm space at the bottom of the tank so that the useful medium is at the level of the injection point.

408 numerical method that explicitly captures the front displacement. To this
409 end, photo analysis was used to extract the fluid interface, as described in
410 the method section, and compare them with the numerical model results.

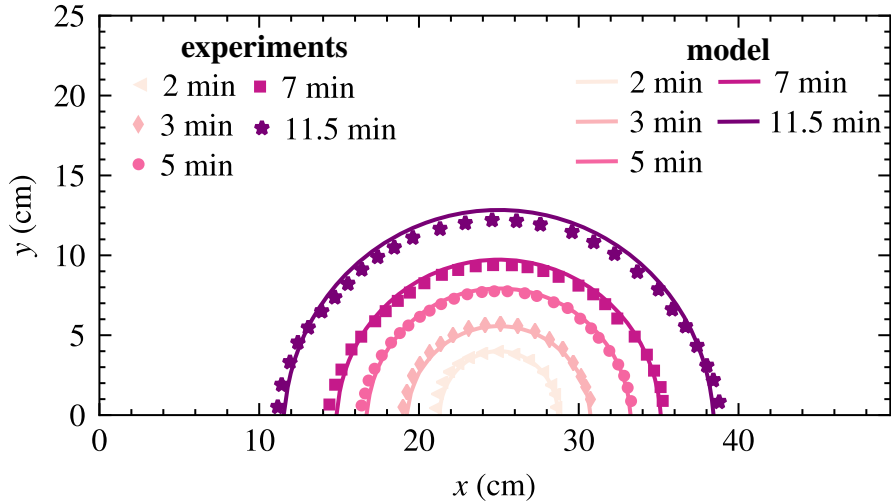


Figure 8: Comparison at various time of the position of the fluid front from numerical modeling (solid lines) and experimental measurements (markers). Results for injection in 1 mm Q_l glass beads. Here, $S_{ei} = 0.06$, $\eta = 0$, and $K_0 = 3.5 \times 10^{-10} \text{ m}^2$.

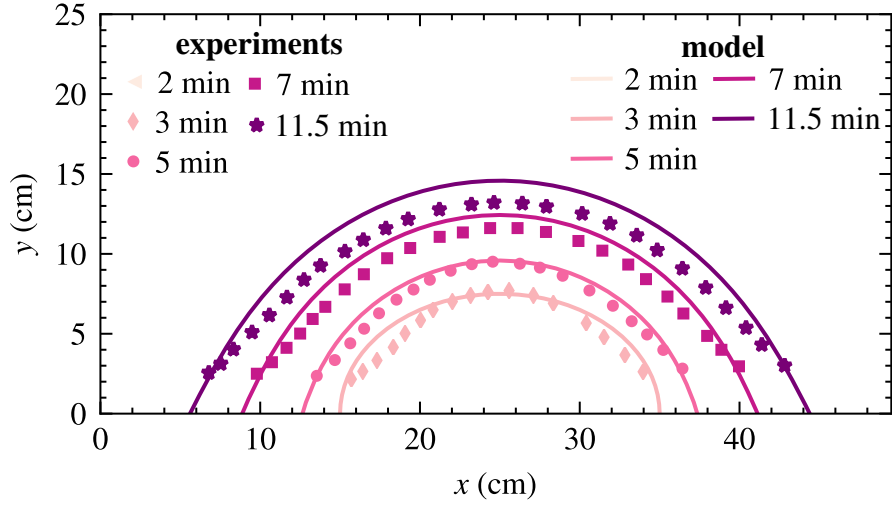
411 6. Numerical results: model validation

412 The ALE code was validated against the experimental injection in glass
 413 beads packing by reproducing the fluid front for different times and each ex-
 414 periment. Fig. 8 shows the numerical and experimental fluid fronts between
 415 2 and 11.5 minutes for 1 mm (Q_l) experiment. Results for the other 1 mm
 416 experiment are analogous. The proposed model predicted well the experi-
 417 mental results for the whole injection. As for the experiments, we observed
 418 the evolution of a semi-circular front. This shape of interface confirms that
 419 the displacement is mainly dominated by viscous forces and with very little
 420 influence from gravity forces. We noticed a small discrepancy ($\approx 6\%$) at the
 421 top of the oil dome after 11.5 minutes of injection, where the numerical front
 422 was slightly in advance compared to the measured front.

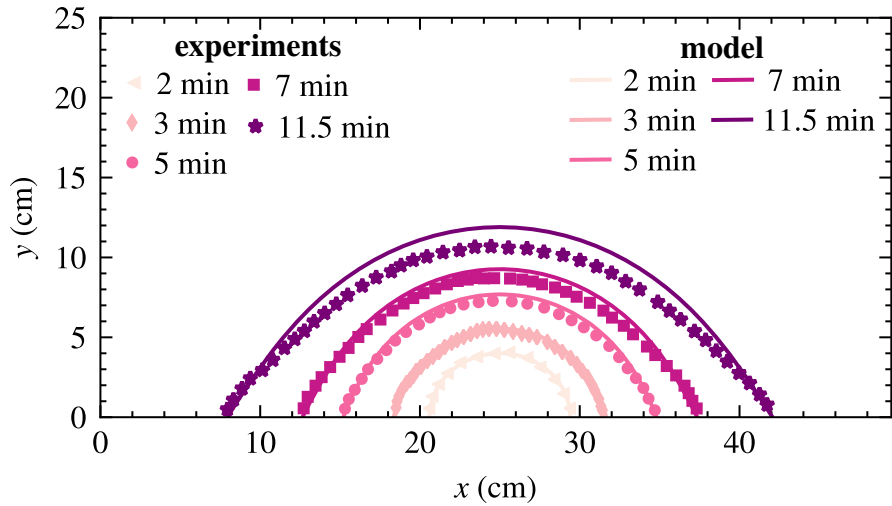
423 Fig. 9a and Fig. 9b show the numerical and experimental fluid front for
424 2 mm experiments with and without initial residual oil saturation, respec-
425 tively. Here again, the position of the fluid front during injection was well
426 reproduced, in particular the flattening of the front and the lateral exten-
427 sion of the oil under the action of gravity. We noticed a larger discrepancy
428 ($\approx 10\%$) between the model and experiments at the final time compared to
429 1 mm experiments. The numerical model predicted a less flattened oil zone
430 than what we measured experimentally. This may have happened because
431 of the uncertainty on the absolute permeability or a locally lower irreducible
432 saturation in ethanol. Indeed, we measured the absolute permeability in a
433 column apparatus so the packing may be different from the one in the tank.

434 Fig. 10 shows the saturation field from modeling and the experimental
435 front as a black line. The top row shows experiment vs. modeling results in
436 1 mm Q_h glass beads at time 7 min and 11.5 min. The bottom row shows
437 the same comparison for the experiment in 2 mm glass beads. In both cases,
438 the position of the front is well reproduced, as well as the lateral spreading
439 of the oil under gravity forces (experiments in 2 mm beads)

440 In conclusion, the proposed model can predict the evolution of the front
441 in versatile situations (flow rate, irreducible and residual saturation) with
442 good accuracy for 1 mm glass beads and 2 mm glass beads during the first
443 part of the injection. However, it fails to accurately reproduce the position of
444 the top of the oil front in the last part of the injection in 2 mm glass beads.
445 As shown, the generalized Darcy equations also fails to reproduce precisely
446 this part of the experimental results.



(a) Comparison for injection in 2 mm glass beads with initial residual saturation ($S_{ei} = 0.056$ and $S_{or} = 0.23$).



(b) Comparison for injection in 2 mm glass beads ($S_{ei} = 0.087$).

Figure 9: Comparison at various time of the position of the fluid front from numerical modeling (solid lines) and experimental measurements (markers). Results for $\eta = 0$, and $K_0 = 4.2 \times 10^{-9} \text{ m}^2$.

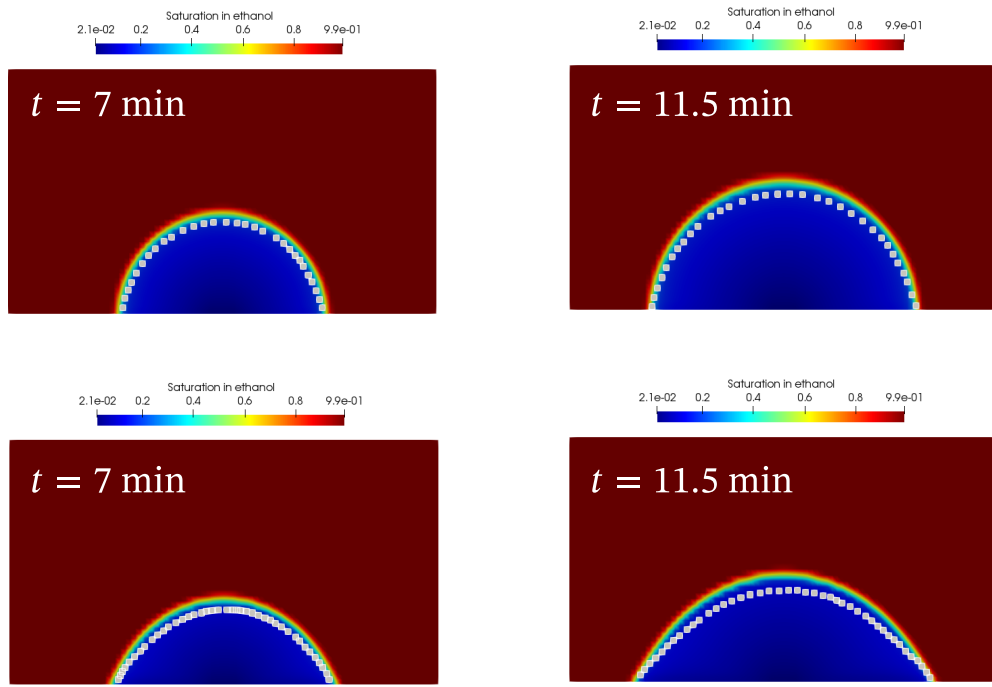


Figure 10: Comparison between experiments (black line) and modeling with generalized Darcy equations (field). Top row shows result for 1 mm Q_h glass beads and bottom row shows result for 2 mm glass beads experiment.

447 **7. Numerical results: parametric study of pumping and injection**
448 **scenarios**

449 Comparison against the experimental injection in glass beads packing
450 at different flow rate and different glass beads diameter have allowed us to
451 validate our modeling approach but are very limited in scope. To under-
452 stand further the impact of the displacement of DNAPL in highly permeable
453 porous media, we investigated two different numerical cases: (i) an injection
454 scenario similar to the experiments previously described, and (ii) a pumping
455 scenario very close to the experiments conducted in Philippe et al. (2021).
456 In the latter, the authors performed isothermal and non-isothermal pumping
457 experiments in the same 2D tank used in our study. They also used the same
458 apparatus and the same fluids. The major difference is that we considered a
459 viscosity ratio $r_{\mu e} = 1$ to avoid viscous fingering. A schematic representation
460 of both scenarios is given in Fig. 11. We used the same boundary and initial
461 conditions as already presented for the validation step, except for a symme-
462 try condition on the left boundary and an impermeable wall condition on the
463 right boundary for the pumping scenario.

464 Tab. 7 lists the values of parameters for both scenarios. We considered
465 that there was no residual oil saturation and used a default value of irre-
466 ducible ethanol saturation $S_{ei} = 5\%$. The viscosity and density ratios, as
467 well as the porosity, have the same values as for the experiments, except for
468 the pumping scenario for which we used matching viscosity for both fluids.
469 We investigated gravity number values between 0.02 and 2 (values for ex-
470 periments were between 0.05 and 0.3) without inertia correction ($F_o = 0$).
471 We also studied values of F_o between 0.02 and 2 for a fixed gravity number

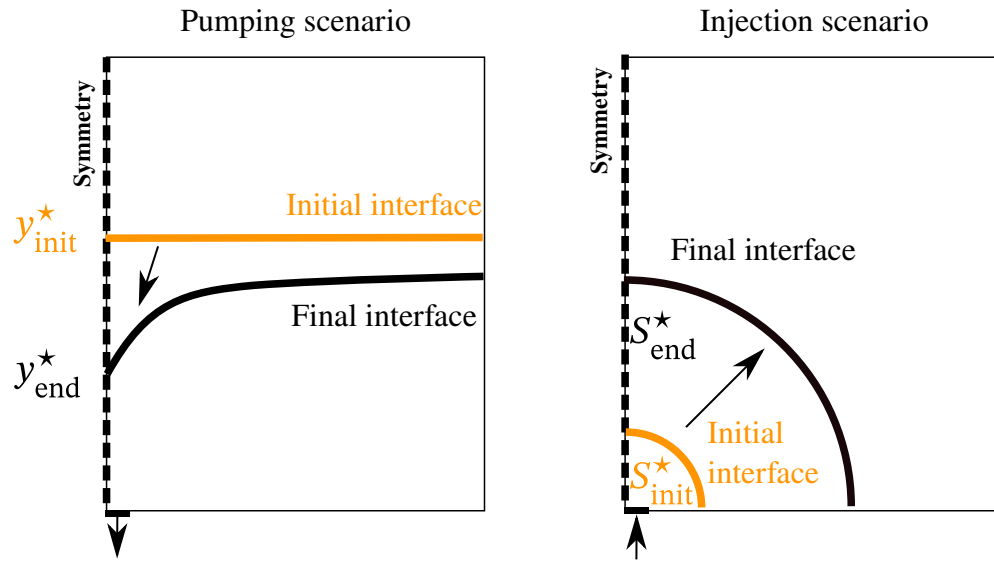


Figure 11: Schematics of the pumping (left) and injection (right) scenarios. y^* and S^* denote the dimensionless position along the y axis and the dimensionless surface occupied by the injected fluid, respectively. Only one half of the system was considered for the parametric studies. The boundary conditions are the same as those used for the validation (Fig. 3), except for the symmetry boundary condition on the left and an impermeable wall condition on the right boundary for the pumping scenario.

Table 7: Parameters used, range of values and default values for both scenarios. Particular values for the pumping scenario are indicated with †.

Parameter	Lower bound	Upper bound	Default value
G_r	0.02	2	0.5
F_o	0.02	2	0
ϕ			0.4
$r_{\rho e}$	-	-	0.887
$r_{\mu e}$	-	-	0.016 – 1 [†]
S_{ei}	-	-	0.05
S_{or}	-	-	0

472 ($G_r = 0.5$) for both scenarios. The range used for the Forchheimer number
 473 in this parametric study is much broader than for the experiments (Tab. 5),
 474 for which inertia forces were negligible. The ranges of values chosen for these
 475 two parameters allowed us to study the influence of gravitational and iner-
 476 tial forces in very different situations, for example for low (G_r and F_o small)
 477 or highly permeable soils (F_o and G_r high). As an example, and assuming
 478 that the characteristic velocity and fluid properties are the same than for
 479 the experiments, the range value for G_r roughly corresponds to an absolute
 480 permeability between 100 and 10,000 darcy units.

481 7.1. Injection scenario

482 Fig. 12 shows the fluid front at the time $t^* = 0.3$ for different values of
 483 the gravity numbers. We observed the same behavior as captured during
 484 the experiments, i.e., a perfect semi-circular front for low gravity numbers
 485 (viscosity-dominated displacement) and a flattened fluid front for a gravity

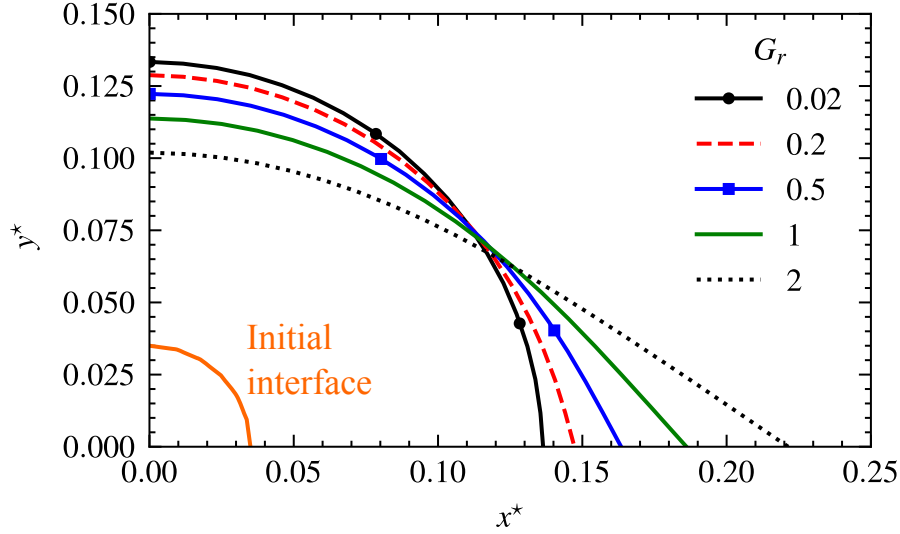


Figure 12: Fluid front at the dimensionless time $t^* = 0.3$ for different gravity numbers (injection scenario). x^* and y^* denote the dimensionless length along the x and y axis, respectively.

486 number equal or higher than 0.2. Fig. 13 further shows the evolution of the
 487 eccentricity parameter as a function of the gravity number. We observed
 488 no critical gravity number for which the front starts to flatten, although the
 489 slope is not constant and the dependence upon the gravity number is not
 490 linear.

491 To investigate the impact of the Forchheimer number, we changed the
 492 inlet boundary condition from a constant inlet velocity to a constant pressure.
 493 The reason is that, when the velocity is imposed, we observed no significant
 494 change of the fluid front but rather a modification of the pressure drop, see
 495 Eq. 5). The inlet pressure, $P^* = 0.15$, was chosen from the previous study
 496 on gravity numbers to be approximately equal to the pressure at the inlet
 497 when $G_r = 0.5$.

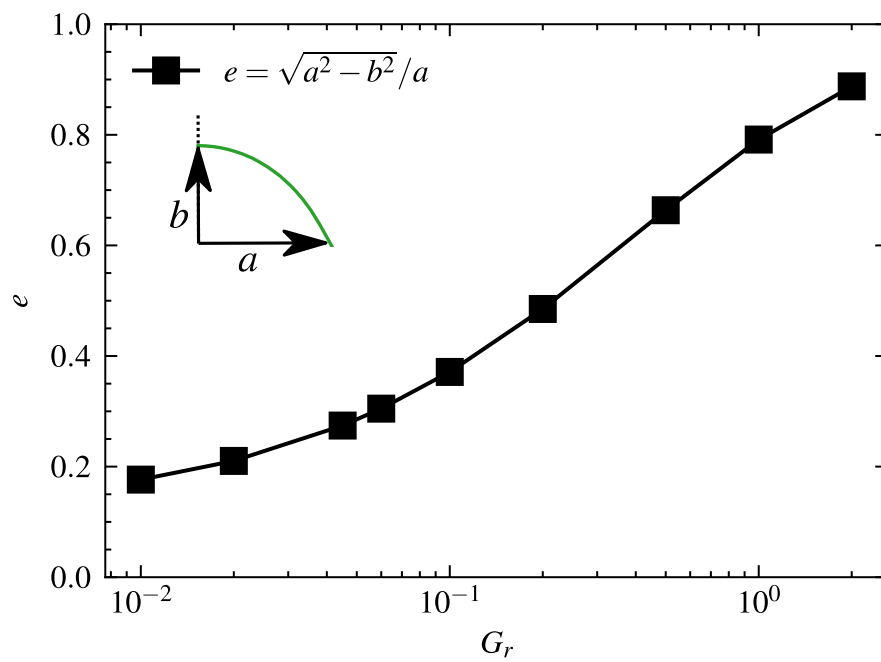


Figure 13: Eccentricity parameter characterizing the shape of the fluid front at the dimensionless time $t^* = 0.3$ for different gravity numbers (injection scenario).

498 Fig. 14 shows the fluid front at time $t^* = 0.3$ for different values of
 499 the Forchheimer numbers. The eccentricity parameter changes only slightly,
 500 rather the position changes with a slower front when the Forchheimer number
 501 increases due to the non-linear drag. Because of the inlet pressure condition,
 502 the injected volume at a given time evolves freely and is also determined by
 503 the drag. We plotted the surface occupied by the injected fluid at the time
 504 $t^* = 0.3$ for different Forchheimer numbers (Fig. 15). The surface of injected
 505 fluid starts to significantly decrease for $0.1 \lesssim F_o$, with a change in behavior
 506 that is more pronounced than the previous study on the gravity number. A
 507 decrease in the surface of injected fluid corresponds to a lower Darcy-velocity,
 508 i.e., a non-negligible part of the pressure gradient is now overcoming the non-
 509 linear drag.

510 *7.2. Pumping scenario*

511 Fig. 16 shows the fluid front at $t^* = 0.3$ for different values of the grav-
 512 ity numbers for the pumping scenario. The first observation is that larger
 513 gravity numbers correspond to a flatter front and to a longer time before the
 514 breakthrough. For $G_r = 2$, the fluid front is about 20% further from the
 515 pumping point than for $G_r = 0.02$, with respect to the initial distance.

516 Fig. 17 shows the fluid front at the time $t^* = 0.3$ for different values
 517 of the Forchheimer numbers. As for the injection scenario, we changed the
 518 inlet boundary condition from a constant inlet velocity to a constant inlet
 519 pressure. This pressure was set to $P^* = 10^{-2}$, which is low enough to pump
 520 the fluid when $G_r = 0.5$. The final volume of pumped fluid freely changes as
 521 a function of the supplementary drag due to inertia effects.

522 The breakthrough was almost reached for $F_o = 0.02$, for which the re-

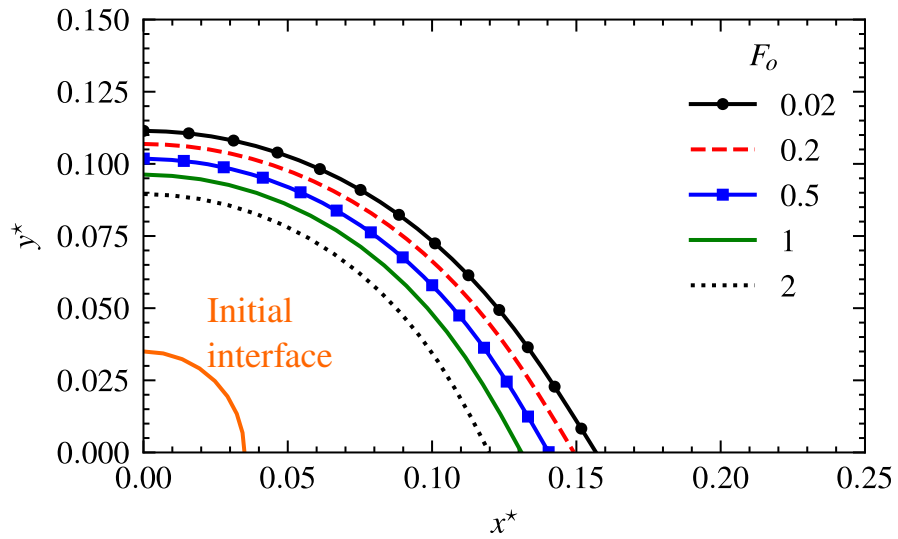


Figure 14: Fluid front at the dimensionless $t^* = 0.3$ as a function of the Forchheimer number for the injection scenario. Results obtained for $G_r = 0.5$. x^* and y^* denote the dimensionless length along the x and y axis, respectively. The dimensionless inlet pressure is $P^* = 0.15$.

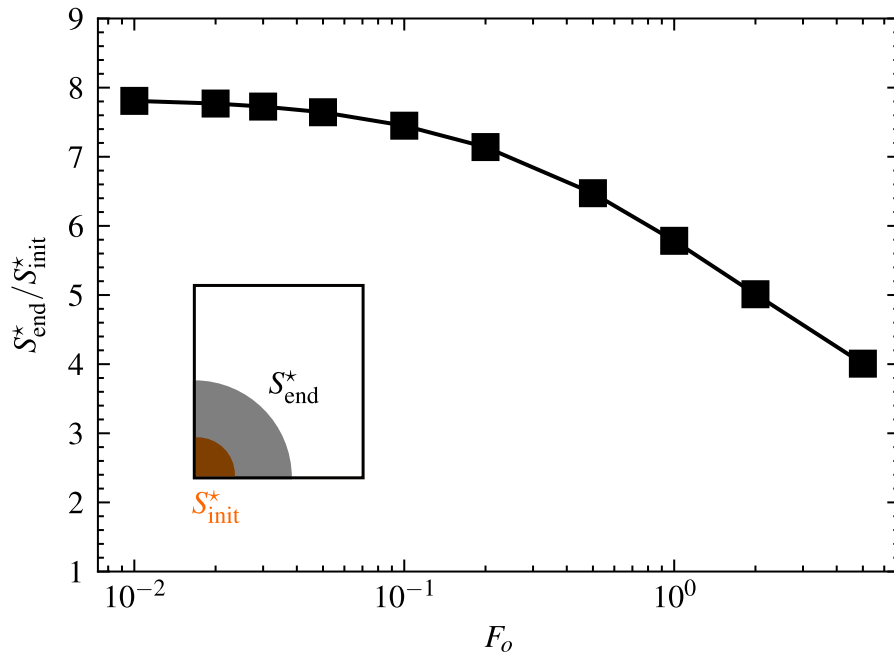


Figure 15: Dimensionless surface of injected fluid at the dimensionless time $t^* = 0.3$ for different Forchheimer numbers (injection scenario). S^* denotes the dimensionless surface occupied by the injected fluid. Results obtained for $G_r = 0.5$. The dimensionless inlet pressure is $P^* = 0.15$.

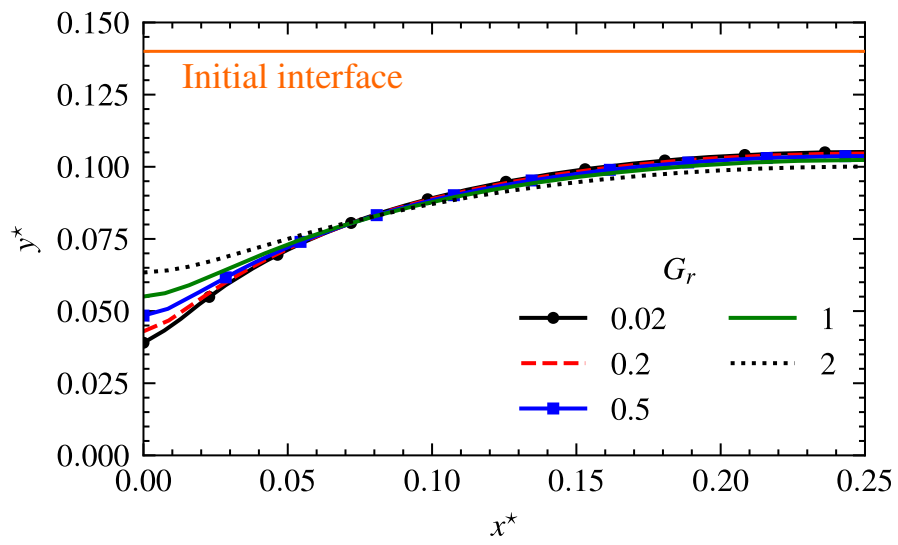


Figure 16: Fluid front at the dimensionless time $t^* = 0.3$ for different gravity numbers (pumping scenario). x^* and y^* denote the dimensionless length along the x and y axis, respectively.

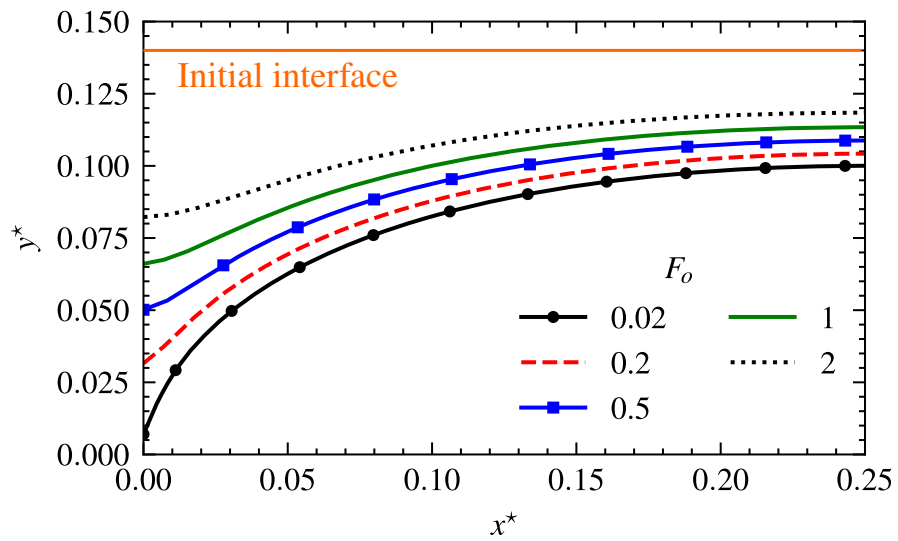


Figure 17: Fluid front at the dimensionless time $t^* = 0.12$ for different Forchheimer numbers (pumping scenario). Results obtained for $G_r = 0.5$. x^* and y^* denote the dimensionless length along the x and y axis, respectively. The dimensionless inlet pressure was $P^* = 10^{-2}$.

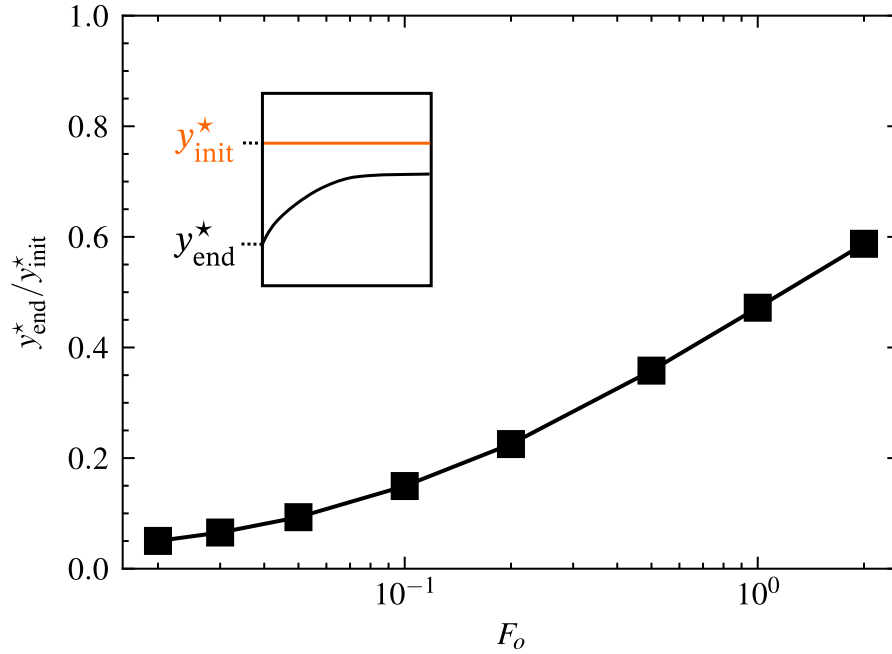


Figure 18: Distance between the fluid front and the pumping point for different Forchheimer numbers at the dimensionless time $t^* = 0.12$ (pumping scenario). y^* denotes the dimensionless length along the y axis. Results obtained for $G_r = 0.5$.

523 sistance due to inertia is very small and the magnitude of the velocity field
 524 was higher. In contrast, when the resistance due to inertia was higher, much
 525 less fluid was pumped and the fluid front was about 55% further away from
 526 the pumping point for $F_o = 2$ than for the case of $F_o = 0.02$ (Fig. 18). The
 527 impact of inertia was significant for $0.1 \lesssim F_o$ (about 0.2 for the injection
 528 scenario) with a front that was at least 15% further away from the pumping
 529 point.

530 8. Discussion

531 Our results for different problem classes indicate that the impact of inertia
532 and gravity on the fluid front are non-negligible. While we found no clear
533 critical value, we observed a mild threshold value for inertia, with significant
534 effects starting for $0.1 \lesssim F_o$. For gravity, the evolution is more progressive
535 without a threshold value.

536 Gravity numbers between 10^{-1} and 1 are commonly encountered in two-
537 phase flows in contaminant hydrology. For example, $G_r \approx 0.6$ for a con-
538 taminated site (chlorinated hydrocarbon, groundwater velocity 10 m/day,
539 and permeability between 10^{-11} to 10^{-9} m²) located in southeastern France
540 (Omirbekov, 2020). The "DNAPL" and ethanol couple used in this work is
541 similar to a coal tar/ water couple. Since coal tar is a highly viscous DNAPL
542 ($\mu_o \approx 0.12$ Pa s), gravity numbers $G_r \approx 1$ were observed only for the most
543 permeable media of 4,200 darcy units (2 mm glass beads). However, gravity
544 effects may be non-negligible for lower and more standard permeabilities,
545 since many DNAPLs are less viscous than the coal tar displacement we have
546 mimicked here with a canola oil and ethanol couple, e.g., chlorinated solvents
547 or elemental mercury.

548 Several works have discussed "threshold" values for F_o from which inertia
549 effects may be considered non-negligible (remembering, as discussed above,
550 that there is no such thing as a critical number). Zeng and Grigg (2006) found
551 that for $F_o = 0.11$, 10% of the pressure drop is due to inertia effects. Macini
552 et al. (2011) found the onset of non-negligible inertia effects for $0.22 < F_o <$
553 0.56 based on experiments in different sands for gas injection. Ghane et al.
554 (2014) proposed a similar range of values ($0.14 < F_o < 0.55$) with water

555 injection in denitrification beds. The threshold value for the Forchheimer
556 number that we have found is therefore consistent with the literature.

557 Such values are harder to reach in contaminant hydrology and less com-
558 mon than large gravity numbers. Previous work focuses either on gas flows,
559 which is much less viscous than DNAPL, at high pressure with superficial
560 velocities as high as tens of cm/s in less permeable media (i.e. the passability
561 is smaller) or on water flow in highly permeable media such as wood chips (up
562 to 10^{-8} m^2). Considering a DNAPL used recently by Colombano et al. (2020)
563 ($\mu_o = 5 \times 10^{-3} \text{ Pa s}$ and $\rho_o = 1661 \text{ kg m}^{-3}$), the superficial velocity required
564 to reach the threshold value for Forchheimer number in 2 mm glass beads
565 ($K_0 = 4200$ darcy units) is in the range of several cm/s. Thus, the impact of
566 inertial forces, as observed in the numerical parametric study, is less likely
567 to be observed in contaminant hydrology than gravity effects, and should be
568 non-negligible for specific active mechanical remediation techniques or pipe
569 rupture event in highly permeable porous media.

570 **9. Conclusion**

571 We studied experimentally the impact of gravitational and inertial forces
572 on the injection of a model DNAPL (canola oil) into a wetting fluid (ethanol)
573 in saturated highly permeable porous media. We conducted laboratory-scale
574 injections in packing of glass beads of diameter 1 and 2 mm, with different
575 injection flow rates and initial saturation (either fully saturated in ethanol or
576 with an initial residual oil saturation). These experimental results, provided
577 in the linked data repository, can be used as a simple benchmark for two-
578 phase models.

579 Due to the high viscosity of the oil, the low interfacial tension, and the
580 inlet condition, capillary effect can be considered as negligible and the dis-
581 placement was mainly driven by viscous and gravity forces. Experiments
582 showed (i) a gravity stabilized fluid front for each case, (ii) a strong impact
583 of gravity forces which flattened the fluid front and laterally spread the in-
584 jected oil, (iii) a small irreducible saturation in ethanol remaining in the oil
585 zone (between 6% and 12%, depending on flow rates, and (iv) a sharp front
586 displacement for each experiment.

587 This latter observation allowed us to use an arbitrary Lagrangian-Eulerian
588 method, coupled with single-phase Darcy-Forchheimer equations, to repro-
589 duce the experiments and further investigate the impact of gravity and inertia
590 forces on the flow process. We studied either an injection scenario similar to
591 the experiments or a pumping scenario resembling experiments from Philippe
592 et al. (2021). Based on the parametric studies, we found that the impact of
593 gravity was most important from $0.1 \lesssim G_r$. This threshold value is low
594 enough to be reached during most DNAPL flows encountered in contami-
595 nant hydrology. Gravity effects manifested themselves by flattening the fluid
596 front, which is particularly visible in the injection scenario. Moreover, the
597 observed behavior is consistent with the experiments, which further proves
598 the interest in using dimensionless numbers obtained at Darcy-scale to study
599 large-scale flow.

600 Macroscopic inertia effects (indicated with the Forchheimer number F_o)
601 were non-negligible and increased faster from $F_o \approx 0.1$. These effects were
602 mainly visible on the fluid front closer to the injection point, or further away
603 from the pumping point, depending on the scenario, due to the increasing

604 flow resistance from inertia. Such values are harder to reach in contaminant
605 hydrology, but may occur for active mechanical remediation techniques in
606 coarse sand, gravels, or other very coarse materials and involving low viscosity
607 DNAPL (e.g. chlorinated solvents). More broadly, this work can be used to
608 study the recovery of DNAPL as a pool in porous media (choice of DNAPL
609 pumping rates or non-Newtonian liquid injection rates to recover DNAPL).

610 The impact of inertia forces was better highlighted using a different Initial
611 Boundary Value Problem (IBVP) than for the impact of gravity. Whereas we
612 tested different configurations (pumping, injection, constant inlet pressure,
613 or constant inlet velocity), we have by no means exhausted all the possi-
614 bilities. We have also only considered stable displacements in this study.
615 Gravity or viscous instabilities are very common phenomena in the subsur-
616 face. For example, recent experiments by Philippe et al. (2020) have clearly
617 shown that viscous fingering is important during DNAPL pumping with ther-
618 mal enhancement. Consequently, further interesting and valuable work may
619 consist of studying the impact of inertia and gravity on instabilities.

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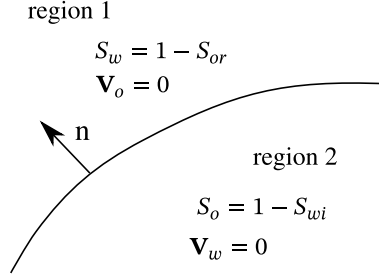


Figure A.19: Macroscopic front between the two fluids. Unit normal vector to the interface \mathbf{n} is oriented from fluid o toward fluid w .

889 Appendix A. ALE boundary condition at the fluid front

890 We write Darcy equations for each phase, distributed as in Fig. A.19, as
 891 a departure to derive the boundary condition at the fluid front for the ALE
 892 model.

893 In region 1:

$$0 = \frac{\partial \phi S_w \rho_w}{\partial t} + \nabla \cdot \rho_w \mathbf{V}_w, \quad (\text{A.1a})$$

$$\mathbf{V}_w = -\frac{\mathbf{K}_w}{\mu_w} \cdot (\nabla P_w - \rho_w \mathbf{g}). \quad (\text{A.1b})$$

894 In region 2:

$$0 = \frac{\partial \phi (1 - S_w) \rho_o}{\partial t} + \nabla \cdot \rho_o \mathbf{V}_o, \quad (\text{A.2a})$$

$$\mathbf{V}_o = -\frac{\mathbf{K}_o}{\mu_o} \cdot (\nabla P_o - \rho_o \mathbf{g}). \quad (\text{A.2b})$$

895 The next step is to write a unique macroscopic momentum balance equation,
 896 written in sense of distributions (Schwartz, 1961), as

$$\mathbf{V}_\alpha = -\frac{\mathbf{K}_\alpha}{\mu_\alpha} \cdot (\{\nabla P_\alpha\} + [P] - \rho_\alpha \mathbf{g}), \quad (\text{A.3})$$

897 where α refers to region 1 and 2, and $\{\nabla f\}$ indicates the gradient of func-
 898 tion f in the sense of functions (usual gradient) and $[\cdot]$ the jump across the
 899 discontinuity interface. Here, we have

$$[P] = P_w - P_o = 0. \quad (\text{A.4})$$

900 We treat the mass balance equation for each fluid w and o independently.

901

902 For fluid w :

$$\left\{ \frac{\partial}{\partial t} (\phi S_{w\alpha} \rho_w) \right\} + \{ \nabla \cdot (\rho_w \mathbf{V}_{w\alpha}) \} = 0. \quad (\text{A.5})$$

903 For fluid o :

$$\left\{ \frac{\partial}{\partial t} (\phi(1 - S_{w\alpha}) \rho_o) \right\} + \{ \nabla \cdot (\rho_o \mathbf{V}_{o\alpha}) \} = 0. \quad (\text{A.6})$$

904 The associated jump discontinuity reads

$$-\phi [S_{w\alpha} \rho_\alpha] \mathbf{n} \cdot \mathbf{w} + [\rho_w \mathbf{V}_{w\alpha}] \cdot \mathbf{n} = 0, \quad (\text{A.7})$$

905 for fluid w , and

$$-\phi [(1 - S_{w\alpha}) \rho_o] \mathbf{n} \cdot \mathbf{w} + [\rho_o \mathbf{V}_{o\alpha}] \cdot \mathbf{n} = 0, \quad (\text{A.8})$$

906 for fluid o and where \mathbf{w} is the front velocity. We use the time derivative of a
 907 function on a discontinuity interface, in the sense of distributions,

$$\frac{\partial f}{\partial t} = \left\{ \frac{\partial f}{\partial t} \right\} - [f] \mathbf{w} \cdot \mathbf{n}. \quad (\text{A.9})$$

908 Since $\mathbf{V}_{w2} = 0$, $\mathbf{V}_{o1} = 0$, $S_{w1} = 1 - S_{or}$ and $S_{o2} = 1 - S_{wi}$, we can write

$$\mathbf{V}_{w1} \cdot \mathbf{n} = \phi(1 - S_{wi} - S_{or}) \mathbf{n} \cdot \mathbf{w}, \quad (\text{A.10})$$

$$\mathbf{V}_{o2} \cdot \mathbf{n} = \phi(1 - S_{wi} - S_{or}) \mathbf{n} \cdot \mathbf{w}, \quad (\text{A.11})$$

909 As \mathbf{w} is unique, continuity of the velocity should be ensured, and the front
910 velocity is finally expressed as

$$\mathbf{w} \cdot \mathbf{n} = \frac{1}{\phi(1 - S_{wi} - S_{or})} \mathbf{V} \cdot \mathbf{n}. \quad (\text{A.12})$$