Spontaneous growth morphology and symmetry selection of interfacial instabilities in anisotropic environments

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ABSTRACT

The displacement of a fluid by another less viscous one in a quasi-two dimensional geometry typically leads to complex fingering patterns. In an isotropic system, dense-branching growth arises, which is characterized by repeated tip-splitting of evolving fingers. When anisotropy is present in the interfacial dynamics, the growth morphology changes to dendritic growth characterized by regular structures. We introduce anisotropy by engraving a six-fold symmetric lattice of channels on a Hele-Shaw cell. We show that the morphology transition in miscible fluids depends not only on the previously reported degree of anisotropy set by the lattice topography, but also on the viscosity ratio between the two fluids, \(\eta_{in}/\eta_{out}\). Remarkably, \(\eta_{in}/\eta_{out}\) and the degree of anisotropy also govern the global features of the dendritic patterns, inducing a systematic change from six-fold towards twelve-fold symmetric dendrites. Varying either control parameter provides a new method to tune the symmetry of complex patterns, which may also have relevance for analogous phenomena of gradient-driven interfacial dynamics, such as directional solidification or electrodeposition.

Pattern growth is ubiquitous in nature and leads to the formation of complex structures\(^1\)\(^-\)\(^4\). Many interfacial patterns can be grouped into two ‘essential shapes’ or morphologies, which are repeatedly observed in different physical systems in a wide range of length scales: isotropic dense-branching growth and anisotropic dendritic growth. Dense-branching growth arises from repeated tip-splitting of the structures and leads to a ramified pattern with many branches\(^5\),\(^6\), controlled by the gradient-driven transport of mass, heat or charge to the interface. Veins of leaves, neuronal networks and blood vessels are all examples of dense-branching growth\(^7\)-\(^12\), as are diffusion-limited fractal electrodeposits (which are also commonly referred to as ‘dendrites’)\(^13\)-\(^15\). In contrast, anisotropic dendritic growth is characterized by stable protrusions and leads to more regular patterns with global symmetries\(^16\),\(^17\). Snowflakes or solidified alloys are examples of anisotropic dendritic structures with preferred directions set by orientation-dependent surface tension\(^17\)-\(^21\). Here, we show that similar dendritic patterns – with tunable symmetry – can arise when the growth occurs in anisotropic environments.

The phenomenon of viscous fingering has played an important role in elucidating the basic principles of these two types of growth\(^22\)-\(^25\), as well as methods to control the resulting patterns\(^26\)-\(^29\). Viscous fingers result from the Saffman-Taylor instability, when one fluid is displaced by another less viscous one in the quasi-two dimensional geometry of a Hele-Shaw cell\(^30\),\(^31\). It has been shown that dendritic growth requires anisotropy in the interfacial dynamics\(^17\),\(^32\)-\(^34\). In its absence, dense branching is instead the generic mode of growth\(^23\). Anisotropy fixes the tip of an advancing interface into a stable parabolic shape that prevents it from splitting\(^17\),\(^35\)-\(^37\) and introduces global symmetries along preferred growth directions, which are also seen in discrete models of diffusion-limited aggregation on crystal lattices\(^38\)-\(^40\). Experimentally, anisotropy can be introduced either externally in the growth environment or internally in one of the fluids. External anisotropy can be imposed by engraving ordered channels on one of the plates of a Hele-Shaw cell, by using channels confined with elastic membranes or by placing air bubbles at the tips of growing fingers\(^32\),\(^35\),\(^41\)-\(^45\). Internal anisotropy can be induced by replacing one of the fluids with a liquid crystal in the nematic phase\(^46\).
Previous studies have considered a particular limit of the viscous-fingering instability; the limit where the viscosity ratio between the less-viscous inner fluid and the more-viscous outer fluid, \( \eta_{in}/\eta_{out} \), is very low, which is typically the case when air or water displace a viscous liquid. The patterns are then characterized by one single growing length scale, the finger length. Under these conditions, experiments using a Hele-Shaw cell with engraved ordered channels have identified the degree of anisotropy, defined as the ratio between the channel height \( h \) and the plate spacing \( b, h/b \), as a control parameter for the morphology transition from dense-branching to dendritic growth. Dendritic structures form beyond a critical value of \( h/b \). When the two fluids are miscible, the degree of anisotropy is the only control parameter for the morphology transition. In the case of two immiscible fluids, the capillary number sets the critical \( h/b \) for the transition. For miscible fluids and for immiscible fluids at high capillary number, the dendrites directly reflect the underlying symmetry of the lattice; four-fold symmetric dendrites grow in a four-fold symmetric lattice, six-fold symmetric dendrites grow in a six-fold symmetric lattice. Dendrites grow in the direction of the channels, which are the regions of largest effective plate spacing within which the flow velocity is highest (the flow velocity is proportional to the square of the plate spacing).

We here reveal how a previously overlooked control parameter, the ratio of the viscosities of the inner and the outer fluid, \( \eta_{in}/\eta_{out} \), modifies both the morphology transition and, remarkably, the symmetry of the dendritic structures in miscible fluids. Recent studies in isotropic environments have identified the viscosity ratio as an important control parameter that governs the global features of the patterns and that in particular introduces a second length scale, the radius of a central region of complete outer-fluid displacement that grows concomitantly with the fingers. This central region becomes increasingly larger, and therefore the relative length of the fingers increasingly smaller, as the viscosity ratio between the two fluids increases. Here we show that a morphology transition from dense-branching to dendritic growth can occur over a large range of viscosity ratios. We engrave channels creating a six-fold symmetric lattice on one of the glass plates and show that the critical degree of anisotropy, \( h/b \), required for the transition to dendritic growth depends on the viscosity ratio between the two liquids. Remarkably, the dendrites can adopt a rich variety of emergent structures: they exhibit six-fold symmetric growth far from the morphology boundary and systematically transition towards twelve-fold symmetric structures as the boundary is approached. Our study reveals novel ways to tune both the morphology transition and the symmetry of dendritic patterns by either controlling the viscosity ratio between the two fluids or the geometric features of the growth environment.

**Results**

**Morphology transitions of miscible viscous fingering in an anisotropic Hele-Shaw cell.** For our experiments we use a radial Hele-Shaw cell of diameter 280 mm and with plate spacings \( b \) varying from 125 \( \mu \)m to 1000 \( \mu \)m. We engrave channels of height \( h \) on an acrylic plate that is placed on the bottom plate of the Hele-Shaw cell creating six-fold symmetric lattices, as shown in Fig. 1a. The ratio between the height of the channel and the plate spacing, \( h/b \), defines the degree of anisotropy. The fluids are injected through a 2 mm diameter hole in the center of one of the plates at a precise volumetric flow rate set by a syringe pump (Harvard PHD 2000). We investigate the growth of patterns in this anisotropic environment by using pairs of miscible fluids with different ratios of viscosities between the less-viscous inner fluid and the more-viscous outer fluid, \( \eta_{in}/\eta_{out} \). The use of miscible fluids allows us to investigate the role of viscosity ratio without concurrently varying the capillary number. Our experiments are performed at large Péclet numbers, ranging between 350 – 7540, where advection dominates over diffusion and the inter-diffusion of the fluids is negligible so that the fluids remain separated by a well-defined interface. We complement the experiments with high-resolution numerical simulations using the finite element software COMSOL Multiphysics (v5.4), which allows us to access the pressure distribution in the fluids. Our model closely replicates the viscosity ratios as well as the geometry of the Hele-Shaw cell in terms of the cell diameter, the inlet hole size, and the six-fold lattice dimensions and plate spacings. We couple the convection-diffusion mass-transport equation from the Transport of Diluted Species Module with the continuity equation for the single-phase, incompressible flow velocity from the Darcy’s Law Module.

In agreement with previous studies at very low viscosity ratios and high capillary numbers, we find that the
Figure 1. Control parameters for growth morphology transition. a, Schematic representation of the modified Hele-Shaw cell. Top image: top view of the bottom plate of the Hele-Shaw cell with an engraved six-fold symmetric lattice. Bottom image: side view of the modified Hele-Shaw cell, denoting the plate spacing $b$ and the channel height $h$. b, Examples of dendritic growth (top, for $h/b = 0.5$) and dense-branching growth (bottom, for $h/b = 0.04$) at low viscosity ratio $\eta_{in}/\eta_{out} = 0.0013$. The scale bar is 1 cm. c, Morphology diagram controlled by the viscosity ratio $\eta_{in}/\eta_{out}$ and the degree of anisotropy $h/b$. Blue symbols denote dense-branching growth, black symbols denote dendritic growth. Experiments are performed with engraved plates with different channel heights $h$ and plate spacings $b$ and at different volumetric flow rates $q$. $(\ast) h = 28 \, \mu m, \, q = 1ml/min; (\square) h = 28 \, \mu m, \, q = 10 \, ml/min; (\circ) h = 50 \, \mu m, \, q = 1 \, ml/min; (\bigtriangleup) h = 50 \, \mu m, \, q = 10 \, ml/min; (\bigcirc) h = 250 \, \mu m, \, q = 1ml/min; (+) h = 250 \, \mu m, \, q = 10 \, ml/min. The solid line denotes a fit to $(h/b - C) / (\eta_{in}/\eta_{out}) = A$ ($A = 3$ and $C = 0.04$ are best-fit parameters).

The morphology transition from dense-branching growth to dendritic growth occurs above a value of $h/b \approx 0.05^{32,47}$. Below this value, fingers grow by repeated tip-splitting which results in dense-branching growth, and above this value, the fingertip is stabilized which results in dendritic growth, as shown in Fig. 1b. Remarkably though, this critical $h/b$ depends strongly on the viscosity ratio: as $\eta_{in}/\eta_{out}$ increases, a higher $h/b$ is needed to transition from dense-branching growth to dendritic growth, as shown in Fig. 1c. We find that neither the absolute values of the channel height $h$ and the plate spacing $b$, nor the volumetric flow rate are control parameters for the morphology transition, as shown by the different symbols used in Fig. 1c which denote experiments performed with plates of various channel heights $h$ ranging from 28 $\mu m$ to 250 $\mu m$, various plate spacings $b$ ranging from 125 $\mu m$ to 1000 $\mu m$, and at two different volumetric flow rates of 1 ml/min and 10 ml/min. The degree of anisotropy and the viscosity ratio are the only parameters governing the morphology transition.

Dendritic growth adopts different symmetries. The viscosity ratio $\eta_{in}/\eta_{out}$ and the degree of anisotropy $h/b$ not only determine the morphology boundary, but have a more dramatic effect on the pattern growth in the dendritic regime. For a fixed $h/b$, an increase in the viscosity ratio $\eta_{in}/\eta_{out}$ leads to a systematic change in the pattern symmetry. Remarkably, the imposed six-fold symmetry of the engraved plate leads to six-fold symmetric growth only at the lowest viscosity ratio. At higher viscosity ratios, the pattern instead transitions towards a twelve-fold symmetry; in addition to the six main dendrites evolving along the straight channels, additional six sub dendrites emerge at a $30^\circ$ angle to the preferred growth direction, as shown in Fig. 2a. The length of the sub dendrites becomes larger with increasing viscosity ratio and eventually comparable to that of the main dendrites. The same trend is recovered in our simulations, as shown in Fig. 2b. A similar transition from six- to twelve-fold symmetry also occurs.
Figure 2. Systematic change from six- towards twelve-fold symmetric dendrites. a, Dendritic patterns for different viscosity ratios obtained at $h/b = 0.49$ (from left to right): $\eta_{\text{in}}/\eta_{\text{out}} = 0.001$, $\eta_{\text{in}}/\eta_{\text{out}} = 0.01$, $\eta_{\text{in}}/\eta_{\text{out}} = 0.05$. As $\eta_{\text{in}}/\eta_{\text{out}}$ increases, the additional generation of sub dendrites grows progressively larger. The scale bar is 1 cm. b, Snapshots of the simulations at $h/b = 0.49$ (from left to right): $\eta_{\text{in}}/\eta_{\text{out}} = 0.01$, $\eta_{\text{in}}/\eta_{\text{out}} = 0.05$, $\eta_{\text{in}}/\eta_{\text{out}} = 0.074$.

for a fixed $\eta_{\text{in}}/\eta_{\text{out}}$ with decreasing $h/b$. Previous studies in the limit of low viscosity ratios have seen hints towards the onset of these additional sub dendrites\textsuperscript{52–54}. Here we show their systematic growth and that they can become comparable in size to the main dendrites within a certain range of viscosity ratio and $h/b$.

To quantify the change from six- to twelve-fold symmetry, we define the length of the main dendrites, $R_m$, corresponding to the structures growing in the direction of the six straight channels, and the length of the sub dendrites, $R_s$, corresponding to the structures growing at an angle of $30^\circ$ with respect to the six straight channels, as shown in the inset of Fig. 3a. For a fixed $h/b$, the ratio $R_s/R_m$ monotonically increases with viscosity ratio. In addition, a decrease in $h/b$ leads to an increase in $R_s/R_m$. We can rescale all data by normalizing the viscosity ratio with $(h/b - 0.04)$, as shown in Fig. 3b. The factor of 0.04 will become evident in the discussion of the morphology boundary. The numerical results are in good qualitative agreement with the experiments and exhibit the same scaling with $h/b$, but yield slightly lower values of $R_s/R_m$ compared to the experimental results. We note that the experimental data has larger error bars at low viscosity ratios. This is a consequence of the limited space available for dendrites to grow because of the small radius of the central region of complete outer-fluid displacement at low $\eta_{\text{in}}/\eta_{\text{out}}$. As a result, not all dendrites grow, which leads to a larger variation in the average magnitude of $R_s/R_m$.

Discussion

The observation that both $\eta_{\text{in}}/\eta_{\text{out}}$ and $h/b$ allow one to systematically tune the symmetry of the patterns reveals a novel aspect of dendritic growth. Remarkably, the change in symmetry is also directly linked to the morphology transition to dense-branching growth. Indeed, when $R_s/R_m$ reaches $\sim 0.85$, corresponding to patterns with twelve dendrites of almost equal size, a further decrease in $h/b$ or a further increase in $\eta_{\text{in}}/\eta_{\text{out}}$ induces the transition to dense-branching growth. The morphology transition can therefore be described by the same functional form used to
Figure 3. Dependence of $R_s/R_m$ on viscosity ratio and degree of anisotropy. a, The ratio $R_s/R_m$ versus $\eta_{in}/\eta_{out}$ for $h/b = 1$ (△), $h/b = 0.49$ (○), $h/b = 0.328$ (★), $h/b = 0.2$ (□), $h/b = 0.1$ (★). The gray symbols denote experimental data, the orange symbols denote numerical data. $R_s/R_m$ is measured when $R_m = 40$ mm, but we note that $R_s/R_m$ is almost constant throughout the growth of the pattern. b, Scaled master curve of $R_s/R_m$ versus $(\eta_{in}/\eta_{out}) / (h/b - 0.04)$. The monotonic increase in $R_s/R_m$ denotes the systematic change from six-fold towards twelve-fold symmetric dendritic patterns.

normalize the data in Fig. 3a; the morphology boundary denoted by a solid line in Fig. 1c corresponds to

$$\frac{h}{b} = A \frac{\eta_{in}}{\eta_{out}} + C$$

(1)

where $A = 3$ and $C = 0.04$ are best-fit parameters. The value of 0.04 denotes the critical $h/b$ for the morphology transition in the limit of very low viscosity ratio.

Why do six-fold dendritic patterns only form far from the morphology boundary, and what leads to the growth of an additional generation of dendrites as we approach the boundary? That the viscosity ratio and the degree of anisotropy are important control parameters for determining $R_s/R_m$ can be seen in a simplified analysis taking into account the effective permeability at different locations corresponding to the growth of sub dendrites or main dendrites (see Methods). Note that the effective permeability in our system is isotropic and lacks a macroscopic preferred direction for single-phase flow. In general, the permeability tensor must be symmetric (by Onsager reciprocity for Stokes flow) and positive definite (by the Second Law of Thermodynamics) and thus represented by an orthogonal matrix, so its eigenvectors, corresponding to the fastest and slowest directions, must be mutually perpendicular. This orthogonality is incompatible with triangular symmetry, so the permeability eigenvalues in our textured Hele-Shaw cell must be degenerate, implying isotropic single-phase flow.

For two-phase flow, however, the gradient of viscosity at the interface between the two fluids can locally break the symmetry and induce an anisotropic effective permeability near the interface. Using concepts derived for the hydrodynamics of slippage on textured surfaces for two-phase flows over hydrophobic surfaces, we consider that the more-viscous outer fluid is partially trapped in the texture as the tip of the less-viscous fluid passes over the texture in the middle of the channel along the “path of least resistance”. For small textures, the trapped fluid leads to a local effective slip length tensor, $b_{slip}$, which causes the effective permeability tensor to become anisotropic and orthogonal in the vicinity of the interface, leading to the appearance of sub dendrites that impart this square symmetry to the pattern. In the limit of “weak anisotropy” in the slip tensor, $\text{Tr}(b_{slip}) << b$, as is the case for our
Figure 4. Formation and growth of sub dendrites. a, Pressure field for patterns at $\eta_{\text{in}}/\eta_{\text{out}} = 0.01$ (top) and $\eta_{\text{in}}/\eta_{\text{out}} = 0.05$ (bottom) at two different times. The lines indicate pressure contours. b, Schematic representation of the path followed by the main dendrites $R_m$ and the sub dendrites $R_s$. At a lattice junction (indicated by the red dot in the dotted circle), the flow predominantly selects the direction along the red arrow, which leads to the growth of the sub dendrites along the $30^\circ$ direction, as observed in both experiment (top image) and simulation (bottom image). c, Zoomed schematics of the lattice junction. The combination of the global pressure distribution from the main dendrites and the local pressure distribution from the tip of the sub dendrites leads to flow into channel 1 along the direction of the red arrow.

experiments, we find that the interface velocities of the sub dendrites and the main dendrites, and therefore $R_s/R_m$, are indeed governed by $\eta_{\text{in}}/\eta_{\text{out}}$ and $h/b$.

To get further insight into the growth of the dendrites, we consider their macroscopic path selection. The main dendrites $R_m$ grow along the six straight channels. The sub dendrites $R_s$, however, select a path at a $30^\circ$ angle from these straight channels. At early stage, two fingers form between each pair of neighboring main dendrites on each side of the $30^\circ$ direction, due to the anisotropy of the lattice. This is observed at any viscosity ratio, as shown in Fig. 4a. Whether these fingers will merge towards each other and grow into a sub dendrite or merge with the main dendrites resulting in a six-fold symmetric pattern depends on the pressure distribution imposed both globally by the main dendrites and locally at the tip of the sub dendrites. At low $\eta_{\text{in}}/\eta_{\text{out}}$ and high $h/b$, the rapid growth of the main dendrites sets up a large pressure gradient at their tip which in turn induces a small pressure gradient in the $30^\circ$ direction, as shown in Fig. 4a, which prevents the sub dendrites from growing. With increasing $\eta_{\text{in}}/\eta_{\text{out}}$ and decreasing $h/b$, however, the sub dendrites themselves build locally a high pressure gradient at their tips which amplifies their growth. We provide more details about the early stage of growth in Fig. S1 of the Supplementary Material.

Once the sub dendrites have emerged, they continue to grow along the $30^\circ$ direction following a zig-zag path, as illustrated in Fig. 4b. As the tip of the sub dendrite reaches a lattice junction, indicated by a red dot, the path towards the $30^\circ$ direction (red arrow) is selected, rather than the straight path (blue arrow). Both experiments and simulations confirm that this occurs. This selection results from the pressure profile induced in the outer fluid by the main dendrites, which effectively shields the sub dendrites from growing towards the main dendrites and pushes
they towards the $30^\circ$ direction. Indeed, when the tip of a sub dendrite reaches the entrance of a lattice junction, as schematically shown in the zoomed-in region in Fig. 4c, it does not grow straight towards channel 2, but is deviated towards the $30^\circ$ direction as a result of the global pressure distribution built up by the neighboring main dendrites. The local pressure distribution at the tip of the sub dendrite then induces a maximum pressure gradient towards channel 1, and most of the flow goes into channel 1. We report additional details of this path selection in Fig. S2 of the Supplementary Material. It is this combination of the global pressure distribution from the main dendrites and the local pressure distribution from the tip of the sub dendrites that leads to the rich pattern selection in dendritic growth.

These different paths selected by the main dendrites and sub dendrites also reveal the origin of the maximum value of $R_s/R_m \approx 0.85$. It reflects the condition where the velocity of the main and sub dendrites becomes approximately equal. As the path selected by the sub dendrites deviates from the radial direction at each junction, the total path is $2/\sqrt{3}$ times longer that of the main dendrites in the straight radial channels. The length of the main dendrite, $R_m$, is therefore $(2/\sqrt{3})R_s$, i.e., $R_s/R_m \sim \sqrt{3}/2 = 0.866$. Interestingly, once this condition is reached, a further increase in $\eta_{in}/\eta_{out}$ or decrease in $h/b$ induces the morphology transition to dense-branching growth. This suggests that the morphology transition occurs when the difference between the pressure gradient in the straight channels and the $30^\circ$ direction becomes negligible, and therefore the role of anisotropy becomes negligible, such that the parabolic tips can no longer be stabilized allowing for tip-splitting to occur.

In conclusion, our results reveal a rich morphology of patterns created by pairs of miscible fluids in anisotropic systems. They demonstrate the important role of the viscosity ratio between the two fluids, which, together with the degree of anisotropy, governs both the morphology transition from dense-branching to dendritic growth and the selected symmetry of the dendrites. Upon approaching the morphology boundary, the dendritic patterns systematically transition from six-fold towards twelve-fold symmetry, in the parameter regime where interfacial flow is governed by an effective slip tensor, whose orthogonality imparts square symmetry to the original pattern.

This diversity of different dendritic patterns provides novel opportunities for tuning the growth of complex structures, not only in viscous fingering, but perhaps also in other cases of interfacial motion limited by gradient-driven transport processes, which lie in the same universality class\textsuperscript{61}. In general, we expect that dendritic growth following the preferred directions of an anisotropic environment will tend to acquire orthogonal symmetry for “weak anisotropy”, whenever transport near the interface is governed by a local effective conductance tensor, which must be orthogonal like the effective slip tensor in a weakly textured Hele-Shaw cell\textsuperscript{56}. For example, in template-assisted directional solidification\textsuperscript{62,63}, a similar morphological transition may arise, controlled by the ratio of thermal diffusivities (analogous to the ratio of inverse viscosities here), whenever the pattern is controlled by the conduction of latent heat away from the interface in the liquid phase. Similarly, in template-assisted electrodeposition\textsuperscript{15,64–66}, it may be possible to tune the symmetry of dendritic patterns by varying the strength of diffusion anisotropy in the electrolyte domain. Active control of anisotropic dendritic growth may also be achieved, for example, by applying electric fields to control viscous fingering\textsuperscript{28,29} over patterned, charged surfaces\textsuperscript{67} having anisotropic electro-osmotic slip tensors\textsuperscript{68}.

**Methods**

**Experiments.** Our experiments are performed in a radial Hele-Shaw cell consisting of two 19 mm thick circular glass plates of diameter 280 mm. Six-fold symmetric lattices of diameter 145 mm are engraved on acrylic sheets with a laser cutter (Universal Laser Systems) and placed on the bottom glass plate of the Hele-Shaw cell. The width of the lattice channels is 800 $\mu$m, and the distance between the edges of two channels is 850 $\mu$m. Three channel depths $h$ of 28 $\mu$m, 50 $\mu$m, and 250 $\mu$m are used. The plate spacing between the engraved acrylic plate and the top glass plate, $b$, is maintained by six spacers around the perimeter. The liquids are injected through a 2 mm hole in the center of the top glass plate with a syringe pump (Harvard PHD 2000). The miscible fluids used in our study are glycerol and water. We tune the viscosity of the inner fluid by mixing glycerol and water in different proportions and we use pure glycerol as the outer fluid. The patterns are recorded with either a Point Grey camera (Grasshopper 3 GS3-U3-91S6M) at frame rates up to 9 f.p.s. or a LUMIX GH5 camera at frame rates up to 60 f.p.s.
Simulations. The governing equations are:

\[ \frac{\partial c}{\partial t} + \nabla \cdot \left(-D \nabla c + cu\right) = 0 \]

\[ u = -\frac{k}{\eta} \nabla p \]

\[ \nabla \cdot u = 0 \]

where \( c \) is the concentration of the inner fluid and \( D \) the molecular diffusivity of the inner fluid. The molecular diffusivity is chosen as negligible \( (D \sim 10^{-14} \text{ m}^2/\text{s}) \) given the high Péclet numbers of the experiments. \( \nabla \) is the in-plane gradient operator, \( u \) is the Darcy velocity set by the pressure gradient \( \nabla p \) and \( k \) and \( \eta \) are the hydraulic permeability and viscosity of the fluids, respectively. The flow in a Hele-Shaw cell can be approximated as quasi-two-dimensional as the plate spacing, \( b \), is much smaller than the radial dimension. The gap-averaged velocity of the fluids is then \( u = -\frac{h^2}{12b^2} \nabla p \) with \( k = b^2/12 \). Note that for miscible fluids, both the pressure and the normal velocity are continuous at the interface. The ratio between the permeability of the channels and that of the surrounding triangles within each lattice cell is \( (h + b)^2/b^2 \). We use an exponential mixing rule for the mixture viscosity and \( \eta = \eta_{\text{out}} e^{-Rc} \), where \( \eta_{\text{out}} \) is the viscosity of outer fluid and \( \eta_{\text{in}}/\eta_{\text{out}} = e^{-R} \). A normal inflow velocity for flow and a Dirichlet boundary condition \((c = 1)\) for transport are applied at the inlet around the cell center, and atmospheric pressure (open-flow) condition for flow and an outflow condition \((u \cdot D\nabla c = 0)\) for transport are imposed on the outer cell boundary. The initial conditions in the entire domain are \( c = 0 \) and \( p = 0 \). We solve for pressure and concentration fields in a fully coupled approach using the PARDISO solver and Newton’s method with dynamic damping for highly nonlinear systems.

A simplified model to account for the effect of \( h/b \) and the viscosity ratio. Our experiments and simulations reveal that the degree of anisotropy \( h/b \) and the viscosity ratio \( \eta_{\text{in}}/\eta_{\text{out}} \) govern the growth of the sub and main dendrites in an anisotropic environment. We simplify our topology by considering flow in two directions with respect to the direction of parallel engraved channels, to capture the role of \( h/b \) and \( \eta_{\text{in}}/\eta_{\text{out}} \) in governing the pattern growth: (a) the pressure gradient \( \nabla p \) is parallel to the channels, corresponding to the growth of the main dendrites and (b) the pressure gradient \( \nabla p \) is perpendicular to the channels, representing the growth of the sub dendrites, as shown in Figs. 5a and 5b. We note that such a texture, as well as that of our more complex six-fold symmetric lattice, is isotropic for a single-phase flow. For a two-phase flow, however, the presence of the interface leading to a gradient of viscosity in the flow direction can locally break the symmetry and lead to an anisotropic two-phase permeability of the interface region. To account for this, we use concepts derived for the hydrodynamics of slippage on textured surfaces for two-phase flows over hydrophobic surfaces\(^{57,58}\). In analogy to these concepts, and to account for the local asymmetry, we consider that the more-viscous outer fluid can get partially trapped in the channels as the less-viscous fluid flows above the texture following the path of least resistance, as schematically shown in Fig. 5c. This conceptualization allows us to introduce a local effective slip length to model the interface region\(^{57,59,60,69,70}\). We denote the height of the layer of trapped outer fluid as \( \delta = \alpha h \), where \( \alpha \) denotes a direction-dependent coefficient. The local effective slip length felt by the inner fluid over a valley of height \( h \), measured from the no-slip boundary at the surface of the channel-free region (dashed line in Fig. 5c), scales as

\[ b_{\text{slip}} = h - \delta \left(1 - \frac{\eta_{\text{in}}}{\eta_{\text{out}}}\right) = h - \alpha h \left(1 - \frac{\eta_{\text{in}}}{\eta_{\text{out}}}\right) \]

\[ (2) \]

The enhancement of permeability due to \( b_{\text{slip}} \) scales as

\[ \frac{b_{\text{slip}}}{b} = \frac{h}{b} - \frac{h}{b} \alpha \left(1 - \frac{\eta_{\text{in}}}{\eta_{\text{out}}}\right) \]

\[ (3) \]

For single-phase flow, where \( \eta_{\text{in}}/\eta_{\text{out}} = 1 \), \( b_{\text{slip}} = h \) and the permeability above the channel is proportional to \( h + b \). For two-phase flow, however, \( b_{\text{slip}} \) decreases with decreasing viscosity ratio leading to a smaller enhancement of the
Figure 5. Schematic representation of the simplified channel texture. a. The main dendrites grow along channels parallel to the flow direction. b. The sub dendrites grow along channels perpendicular to the flow direction. c. The effective slip length $b_{\text{slip}}$ at the interface between the two fluids modifies the local permeability as the inner fluid flows above the channels. The light blue region represents the less-viscous inner fluid, the white region represents the more-viscous outer fluid within the channel.

The decrease of $b_{\text{slip}}$ depends on the direction of the channels with respect to the flow, leading to a local symmetry breaking and the rich anisotropic pattern selection.

When $\text{Tr}(b_{\text{slip}})/b \gg 1$, the effect of the channels dominates and an analysis in terms of an effective slip tensor is not applicable. The case of interest here is when $\text{Tr}(b_{\text{slip}})/b \ll 1$, where the texture can be analyzed locally in terms of an effective slip tensor. This effective slip tensor is positive definite and $90^\circ$ symmetric between the fast and slow directions:

$$b_{\text{slip}} = S_{\theta} \begin{pmatrix} b_{\text{slip},||} & 0 \\ 0 & b_{\text{slip},\perp} \end{pmatrix} S_{-\theta}$$

(4)

where $S_{\theta} = \begin{pmatrix} \cos \theta & \sin \theta \\ -\sin \theta & \cos \theta \end{pmatrix}$ and $\theta$ is the angle between the pressure gradient and the texture. $b_{\text{slip},||}$, $b_{\text{slip},\perp}$ are two eigenvalues of $b_{\text{slip}}$. The subscripts $||, \perp$ denote the fast direction and the slow direction, respectively. Note that here, the fast direction of the effective slip tensor corresponds to the direction of the main dendrites, the slow direction corresponds to the direction of the sub dendrites. The two corresponding eigenvectors have a $90^\circ$ symmetry, corresponding to the formation of, respectively, the main dendrites at $0^\circ$, $60^\circ$, $120^\circ$ and the sub dendrites at $30^\circ$, $90^\circ$, $150^\circ$.

From equation (2), we have

$$b_{\text{slip},||} = h \left( 1 - \alpha_{||} + \alpha_{||} \frac{\eta_{\text{in}}}{\eta_{\text{out}}} \right)$$

(5)

and

$$b_{\text{slip},\perp} = h \left( 1 - \alpha_{\perp} + \alpha_{\perp} \frac{\eta_{\text{in}}}{\eta_{\text{out}}} \right)$$

(6)
where $\alpha_{||,\perp}$ denotes the coefficient for the fast and slow direction, respectively, related to the effective slip tensor. As $b_{\text{slip},||} > b_{\text{slip},\perp}$, we have $\alpha_{\perp} > \alpha_{||}$.

The dimensionless effective permeability tensor scale to its value without slip is expressed as

$$K = I + 3A_p$$

(7)

where $A_p$ is a dimensionless matrix describing a slip-driven plug flow in the $A_p \nabla p$ direction. We have

$$A_p = S_\theta \left( \begin{array}{cc} A_\parallel(b_\parallel) & 0 \\ 0 & A_\perp(b_\perp) \end{array} \right) S_{-\theta}$$

(8)

Substituting (5) and (6) into (9) yields

$$K = \left( \begin{array}{cc} K_{xx} & K_{xy} \\ K_{yx} & K_{yy} \end{array} \right) = \left( \begin{array}{cc} 1 + 3 \frac{1}{\eta_{\text{slip},||} + 1} & 0 \\ 0 & 1 + 3 \frac{1}{\eta_{\text{slip},\perp} + 1} \end{array} \right)$$

(9)

For our case where $b_{\text{slip}}/b \ll 1$, the effective permeability can be expressed as

$$\kappa \approx \frac{b^2}{12\eta_{\text{in}}} \left( 1 + \frac{3b}{\eta_{\text{in}}(1 - \alpha_|| + \alpha_{||} \frac{\eta_{\text{in}}}{\eta_{\text{out}}})} \right) \left( \begin{array}{cc} 1 + 3 \frac{1}{\eta_{\text{slip},||} + 1} & 0 \\ 0 & 1 + 3 \frac{1}{\eta_{\text{slip},\perp} + 1} \end{array} \right)$$

(10)

For the simplified topology in Fig. 5, the main dendrites form along the fast direction of the effective permeability and the sub dendrites form along the slow direction of the effective permeability:

$$\kappa_{\text{in}} = \frac{b^2}{12\eta_{\text{in}}} K_{xx} \approx \frac{b^2}{12\eta_{\text{in}}} \left( 1 + \frac{3h}{\eta_{\text{in}}(1 - \alpha_|| + \alpha_{||} \frac{\eta_{\text{in}}}{\eta_{\text{out}}})} \right)$$

(12)

$$\kappa_s = \frac{b^2}{12\eta_{\text{in}}} K_{yy} \approx \frac{b^2}{12\eta_{\text{in}}} \left( 1 + \frac{3h}{\eta_{\text{in}}(1 - \alpha_\perp + \alpha_{\perp} \frac{\eta_{\text{in}}}{\eta_{\text{out}}})} \right)$$

(13)

This shows that as $\eta_{\text{in}}/\eta_{\text{out}}$ increases, the effective permeabilities in the main and sub channels, $\kappa_{\text{in}}$ and $\kappa_s$, increase. Accordingly, the interface velocities in the main and sub channels, $u_{\text{in}} = \kappa_{\text{in}} \nabla p$ and $u_s = \kappa_s \nabla p$, increase. Let us now discuss the role of the viscosity ratio $\eta_{\text{in}}/\eta_{\text{out}}$ for the increase in permeability for both the main dendrites and the sub dendrites. The ratio of the derivatives of the permeability for sub dendrites and main dendrites is

$$\frac{\partial \kappa_s}{\partial \kappa_{\text{in}}} \frac{\partial (\eta_{\text{in}}/\eta_{\text{out}})}{\partial \eta_{\text{in}}} \approx \frac{\alpha_{\perp}}{\alpha_{||}} > 1$$

(14)

Therefore, for increasing viscosity ratio, the increase in permeability along the sub dendrites (slow direction) is larger than the increase in permeability along the main dendrites (fast direction).

The ratio of the interface velocities between the sub dendrites and the main dendrites scales as

$$\frac{u_s}{u_{\text{in}}} \approx \frac{\kappa_s}{\kappa_{\text{in}}} \approx 1 - \frac{3}{\frac{b}{\kappa} + 3} \left( \frac{\eta_{\text{in}}}{\eta_{\text{out}}} \right) \left( \alpha_{\perp} - \alpha_{||} \right)$$

(15)
where $\alpha_\perp - \alpha_\parallel \geq 0$.

This analysis shows that for the case of $b_{\text{slip}}/b \ll 1$, $u_s/u_m$ increases with an increase in the viscosity ratio $\eta_{\text{in}}/\eta_{\text{out}}$ or a decrease in the degree of anisotropy $h/b$. When the viscosity ratio $\eta_{\text{in}}/\eta_{\text{out}}$ approaches 1, $u_s/u_m$ will be close to 1. When $h/b$ approaches zero, $u_s/u_m$ approaches 1. Clearly, this description is oversimplified but it does capture the essential features of how the viscosity ratio $\eta_{\text{in}}/\eta_{\text{out}}$ and the degree of anisotropy $h/b$ affect the interface velocities of the main and sub dendrites, and therefore the growth of $R_m$ and $R_s$.

Data availability

The data supporting the findings of this study are available within the paper and its Supplementary Material and are available from the corresponding author upon request.

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**Author contributions**

Q.Z. and I.B. designed the research; Q.Z. performed the experiments; M.A.A. developed the computational models; M.A.A. and Q.Z. conducted the simulations; Q.Z., M.A.A., M.Z.B. and I.B. analyzed the experimental and numerical data; Q.Z., M.A.A., M.Z.B. and I.B. wrote the paper.

**Competing interests**

The authors declare no competing financial interests.