Pattern selection in radial Hele-Shaw flow: effects of interfacial tension and aging fluid viscoelasticity

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Abstract

Intricate fluid displacement patterns, arising from the unstable growth of interfacial perturbations, can be driven by gradients in viscosity, elasticity and surface tension. Using video imaging, we explore the miscible and immiscible displacements of a soft glassy colloidal suspension by a Newtonian fluid in a quasi-two-dimensional radial geometry. We report the emergence of a rich variety of interfacial pattern morphologies: dense viscous, dendritic, viscoelastic fracture, flower and jagged. We distinguish each pattern morphology by the natural logarithm of its areal ratio, defined as the area of the fully developed pattern normalized by the area of the smallest circle enclosing it. We segregate all the distinct patterns in a three-dimensional nonequilibrium phase diagram spanned by the aging time of the displaced suspension, the flow rate of the displacing fluid and the interfacial tension. Besides demonstrating the range of patterns generated using a Newtonian-non-Newtonian fluid pair, our results are important in understanding and predicting growing interfaces involving mud displacements.

Keywords: Colloidal clay suspensions, Radial Hele-Shaw flows; Viscoelastic fluids; Interfacial patterns; Viscoelastic fractures; Newtonian-non-Newtonian interfaces; Miscible displacements; Immiscible displacements.

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1 Introduction

The intrusion of a less viscous fluid into a more viscous one generates a rich array of interfacial patterns that depend on factors such as surface tension \([1,2]\), fluid viscoelasticity \([3,8]\) and driving pressure \([9]\). Unstable interfaces arise from the inherent nonlinearities in the system and have several implications in the technological realm, for example, in flows through porous media \([10]\), microfluidics \([11]\), sugar refining \([12]\), enhanced oil recovery \([13]\) and geophysics \([14,15]\). A Hele-Shaw cell, a setup comprising two parallel plates typically separated by a gap of a few hundred micrometers, is often used to study confined flows \([16]\). Such confined flows are governed by Darcy’s law, where flow velocity depends on the pressure gradient, fluid viscosity and the gap between the plates. Interfacial patterns resulting from the displacement of one Newtonian fluid by another are sensitive to the viscosity contrast between the fluid pair \([17]\), the interfacial tension \([18]\) and the pressure gradient \([9]\).

However, instabilities that arise due to the displacement of a non-Newtonian fluid by a Newtonian fluid result in interfacial patterns that are quite distinct from those involving Newtonian-Newtonian displacements \([3,7]\). Numerical simulations revealed that the displacement of shear-thinning fluids by air resulted in the narrowing of fingers, multiple side branches and a suppression in finger-tip splitting \([3]\). When foam, a yield stress fluid, was displaced by air at increasing shear rates, a transition from jagged to smooth interfacial patterns, believed to arise from the distortions and rearrangements of foam bubbles at high shear rates, was observed experimentally \([4]\). Recent work on the displacement of dense granular cornstarch suspensions by glycerol-water mixtures reported a significant suppression of interfacial instabilities by either increasing the viscosity ratio of the fluid pair or the concentration-dependent elasticity of the displaced viscoelastic suspension \([5]\). The radial displacement of Bentonite clay suspensions by water resulted in viscoelastic fracturing when the stored elastic energy of the displaced suspension exceeded the energy required for fracture onset \([6]\). The displacement of clay suspensions occurs in a broad range of scenarios ranging from the formation of river networks \([15]\) to drilling applications \([19]\).

Synthetic clay particles of Laponite® are disk-shaped with diameters 25-30 nm and thickness \(\approx 1\) nm. Dry Laponite powder exists in the form of one-dimensional stacks called tactoids with Na\(^+\) ions residing in the intergallery spaces of the clay platelets \([20]\). In aqueous suspensions of pH < 11, the rims of the Laponite disks bear positive charges \([21]\). The Laponite faces acquire negative charges due to the diffusion of Na\(^+\) ions into the aqueous medium. These Na\(^+\) counterions contribute to the
formation of diffuse electric double layers around the Laponite disks. Osmotic pressure differences within the suspension cause tactoid swelling and the eventual exfoliation of Laponite platelets from the ends of the tactoids, leading to a time-dependent interparticle electrostatic potential $[20,22]$. The heterogeneously charged Laponite disks gradually self-assemble to form fragile microstructures composed of overlapping coins and house of cards (HoC) aggregates $[23]$. In a phenomenon identified as physical aging, percolation of these microstructures throughout the system and the continuous particle rearrangements within these fragile structures contribute to a spontaneous increase in sample rigidity with time $[24]$. It was reported that the aging dynamics of aqueous Laponite suspensions can effectively be controlled by changing clay concentration and suspension temperature, and by incorporating various additives in the dispersion medium $[22,25–27]$. The inclusion of dissociating additives modifies the interparticle electrostatic interactions, thereby altering the aging dynamics of clay suspensions. Non-dissociating additives, in contrast, alter the structure of the dispersion medium. The resultant changes in intermolecular solvation forces have a non-trivial influence on sample aging.

Fig. 1: (a) Evolution of elastic modulus $G'$ (●) and viscous modulus $G''$ (○) for a 3.25% w/v Laponite suspension with increasing aging time $t_w$. The inset on the top-left shows a photograph of a freshly prepared aqueous clay suspension in a glass vial and a schematic illustration of well dispersed Laponite clay particles. The inset on the bottom-right shows a photograph of the same sample, acquired after $t_w = 24$ h, and a schematic representation of the house of cards suspension microstructures formed by self-assembly of Laponite clay particles. (b) Viscosity vs. shear rate ($\dot{\gamma}$) flow curves for 3.25% w/v Laponite suspensions at aging times $t_w = 1$ h (●), 5 h (○) and 24 h (●) show shear-thinning rheology. (c) Viscosity vs. shear rate ($\dot{\gamma}$) curves for water (■) and mineral oil (○) display Newtonian flow.
Rheological measurements such as in Fig. 1(a) can be employed to observe the spontaneous evolution of the mechanical moduli of viscoelastic clay suspensions with aging time \( t_w \); here \( t_w \) is the time duration between sample loading and measurement. Additional details about rheological measurements are provided in Supplementary Information section ST1. We see from the photos in the insets of Fig. 1(a) that while a clay suspension at an early age shows liquid-like rheology (top-left inset), the same suspension, when aged considerably, behaves like a solid and supports its own weight against gravity (bottom-right inset). Schematic illustrations of the arrangement of Laponite particles in the aqueous suspension medium are also displayed in the insets. While the platelets are well-dispersed in the medium at low suspension ages, the clay particles self-assemble to form fragile percolating microstructures at higher ages. Investigations into the phase behavior of Laponite suspensions have reported phase separation below a concentration of 1 % w/v \([28]\), structural buildup and aging, presumably through Laponite edge to face (HoC) associations, between 1 % w/v and 2 % w/v \([29,30]\) and formation of a repulsive colloidal glassy phase above 2 % w/v \([31]\).

We report interfacial patterns that result when an aging viscoelastic Laponite clay suspension is displaced by a Newtonian fluid in a radial Hele-Shaw cell. We study miscible and immiscible displacements by using water and mineral oil as the displacing Newtonian fluids. As seen in Fig. 1(b), Laponite suspensions are strongly shear-thinning. In contrast, as shown in Fig. 1(c), water and mineral oil show Newtonian flow and their viscosities are independent of the imposed shear rates. We report three distinct interfacial pattern morphologies: dense viscous patterns (DVP), dendritic patterns (DP) and viscoelastic fractures (VEF) when aqueous clay suspensions, characterized by increasing \( t_w \) and therefore having progressively larger elasticities, are displaced by water, a miscible solvent. In contrast, flower patterns (FP) transform to irregularly-shaped jagged patterns (JP) when an aqueous clay suspension of increasing \( t_w \) is displaced by immiscible mineral oil. We note that the observed pattern morphologies are determined by the spontaneous aging and shear-thinning rheology of the displaced clay suspension, and the interfacial tension between the fluid pair. We quantify all the observed interfacial patterns by computing their average branch frequencies and average branch widths. Finally, we propose a new parameter, an areal ratio \( A_p/A \), which we define as the area of the fully developed interfacial pattern, \( A_p \), divided by the area of the smallest circle enclosing it, \( A \). Estimation of \( \ln(A_p/A) \) allows us to segregate all the distinct morphologies in a three-dimensional nonequilibrium phase diagram spanning the aging time \( t_w \) of the displaced clay suspensions, the flow rate \( q \) of the displacing Newtonian fluid and the interfacial tension \( \sigma \).
Fig. 2: (a) Schematic diagram showing the side view of a radial Hele-Shaw cell. (b) A representative grayscale displacement pattern of water (black) invading an aging aqueous clay suspension (gray). $R$ is the longest finger length of the pattern such that a circle of radius $R$ encloses the entire pattern. The scale bar is 5 cm.

2 Materials and Methods

Dried Laponite XLG (BYK additives Inc.) powder was vigorously stirred in Milli-Q water (Millipore Corp., resistivity 18.2 MΩ·cm) to prepare 3.25% w/v Laponite suspensions. The freshly prepared Laponite clay suspension was first loaded in a radial Hele-Shaw cell, comprising two circular glass plates each of radius 30 cm and gap 170 μm, with a syringe pump (NE-8000, New Era Pump Systems, USA) through a 3 mm central hole drilled on the top plate (Fig. 2(a)). The aging time $t_w = 0$ corresponds to the time when the loading of the clay suspension was completed. Milli-Q water and mineral oil (Acros Organics) were used as the miscible and immiscible displacing fluids respectively in two separate sets of experiments. These Newtonian fluids were injected with flow rate $q$ through the same central hole of the Hele-Shaw cell after allowing the clay suspension to age to a pre-determined aging time $t_w$. The displacement of clay suspensions by the Newtonian fluids resulted in interfacial patterns whose evolutions were recorded. The stack of images obtained from the videos was converted to grayscale format (Fig. 2(b)) and analyzed using the MATLAB®2018 image processing toolbox. A stress-controlled rheometer (Anton Paar, MCR 501) was used to perform rheological measurements in a cone and plate geometry. All the experiments were performed at room temperature ($25^\circ$C). Additional experimental details are provided in Supplementary Information section ST1. Raw images of representative interfacial patterns are shown in Supplementary Fig. S1. The procedure for binarization of raw images is provided in Supplementary Fig. S2.
3 Results and Discussion

3.1 Aging time $t_w$ of the displaced suspension determines interfacial pattern morphology

Figures 3 (a1-g1, a2-g2) show representative interfacial patterns formed during the radial displacement of aqueous clay suspensions of increasing ages, $t_w$, by both miscible and immiscible Newtonian displac-

![Graph showing areal ratio $A_p/A$ vs. $t_w$ for miscible ($σ = 0$) and immiscible ($σ ≠ 0$) displacements.](image)

Fig. 3: Miscible (interfacial tension $σ = 0$) displacement patterns in grayscale obtained by the displacement of 3.25% w/v aging clay suspensions by water at constant flow rate $q = 5$ ml/min and for different suspension aging times $t_w$: (a1) dense viscous patterns (DVP), (b1-c1) dendritic patterns (DP) and (d1-g1) viscoelastic fractures (VEF). Immiscible ($σ ≠ 0$) displacement patterns obtained by the displacement of aging clay suspensions at identical $t_w$ and $q$: (a2-c2) flower patterns (FP). Yellow dotted circles illustrate the central dark stable region. (d2-g2) jagged patterns (JP). All the patterns correspond to a fixed longest finger length $R = 7.01 ± 0.01$ cm. The scale bar is 4 cm. (h) The areal ratio $A_p/A$ of the patterns vs. $t_w$ for miscible (■) and immiscible (●) displacements. Here, $A_p$ is the area of the fully developed pattern and $A = πR^2$ is the area of the smallest circle enclosing the entire pattern. (i) Average branch frequency ($f$, □) and average width ($w$, △) of the fingers as a function of $t_w$ for miscible ($σ = 0$) displacement patterns. (j) $f$ (○) and $w$ (▼) vs. $t_w$ for immiscible ($σ ≠ 0$) displacement patterns. Error bars in $f$ and $w$ represent the standard deviations in measurements of multiple branches. The vertical color bar on the right maps the distinct morphological patterns to the natural logarithms of their areal ratios: $\ln(A_p/A)$. 

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ing fluids at a constant flow rate $q = 5 \text{ ml/min}$. These interfacial patterns correspond to a fixed longest finger length $R = 7.01 \pm 0.01 \text{ cm}$ ($R$ is defined in Fig. 2(b)). We note three distinct morphologies for miscible displacements by water (interfacial tension $\sigma = 0$) as the elasticity of the displaced clay suspensions increases due to aging (Fig. 3(a1-g1)). For a clay suspension of low age ($t_w = 1 \text{ h}$), we observe the frequent merging of neighboring fingers during pattern evolution, resulting in a pattern without well-defined boundaries (Fig. 3(a1)). We designate this pattern, characterized by narrow fingers and side branches, as a dense viscous pattern (DVP). At intermediate $t_w$, we observe dendritic patterns (DP, shown in Figs. 3(b1,c1)) with considerable side-branching and well-defined interfacial boundaries. For highly aged suspensions undergoing miscible displacement, viscoelastic fractures (VEF) emerge (Figs. 3(d1-g1)) and propagate rapidly. The VEF patterns display sharp finger-tips and perpendicular offshoots from the primary cracks, and are morphologically very dissimilar to DVP and DP.

The observed side-branching in DVP and DP occurs due to flow-induced anisotropies \[32\] as a result of shear-thinning of the increasingly elastic displaced clay suspensions. Interestingly, simulations involving the displacement of viscoelastic fluids \[3\] reported that a competition between enhanced finger-tip velocity due to shear-thinning of the displaced fluid and simultaneous tip broadening due to imposed driving pressures can result in the formation of side branches. Here, we estimate the finger-tip velocity of each pattern by recording the evolution of the longest finger of length $R$ with time. With the transition of the patterns from DVP to VEF, the time-averaged values of finger-tip velocities ($\bar{U}$, Supplementary Fig. S3(a)) increase. These velocity values are used to calculate the shear rates ($\dot{\gamma} = 2\bar{U}/b$ where $b$ is the Hele-Shaw cell gap \[33\]) imposed by the displacing fluid. The estimated values of shear rates at the finger-tips ($36.47 \text{ s}^{-1} \leq \dot{\gamma} \leq 552.94 \text{ s}^{-1}$) increase due to enhanced shear-thinning of the displaced clay suspension when its age, $t_w$, increases. This leads to the increased shedding of side branches. However, for highly aged clay suspensions characterized by the largest $t_w$ values, elastic effects dominate over shear-thinning, which results in the emergence of VEF patterns. Viscoelastic fractures were reported in earlier experiments involving miscible displacements of elastic Bentonite clay suspensions \[6\]. Such patterns were also seen when highly elastic agar gel suspensions \[34\] and shear-jammed cornstarch suspensions \[7\] were displaced by immiscible air under large driving pressures. In our experiments at high $t_w$, percolating microstructures in the aged clay suspension cause the buildup of elastic stresses. Local yielding events in the jammed clay suspension during its displacement by water lead to the rapid release of elastic stresses and result in the emergence of viscoelastic fractures (VEF).
Since the formation of sharp tips costs more energy when the interfacial tension is non-zero, immiscible displacement patterns preferentially preserve smooth and rounded finger-tips. The stabilizing effect of interfacial tension results in considerably lower finger-tip velocities $\bar{U}$ (Supplementary Fig. S3(b)) and shear rates ($12.94 \, \text{s}^{-1} \leq \dot{\gamma} \leq 16.47 \, \text{s}^{-1}$) than in miscible displacement experiments (Supplementary Fig. S3(a)). Furthermore, for immiscible displacements, $\bar{U}$ does not show significant dependence on the age of the displaced clay suspension. We note that immiscible displacement patterns (Figs. 3(a2-g2)) have very distinct morphologies when compared to their miscible counterparts at identical suspension ages and displacing fluid flow rates. Flower patterns (FP) with round finger-tips and a dark central region (yellow dotted circles in Figs. 3(a2-c2)) are observed when suspensions of low to intermediate ages are displaced by mineral oil. This dark central region results from a delay in onset of instability at non-zero interfacial tension and resembles the observations in previous experiments involving Newtonian fluid pairs \cite{2,35}. Displacement of the highly elastic clay suspensions at the highest ages $t_w$ requires disruption of the fragile suspension microstructures by a process that costs substantial energy and leads to a transition to more ramified jagged patterns (JP, shown in Figs. 3(d2-g2)). It is to be noted that the observed jagged patterns closely resemble those seen during the immiscible displacement of shaving foam, a non-zero yield stress material, by air \cite{4}.

We quantify the morphologies of the interfacial patterns by estimating their areal ratios ($A_p/A$), average branch frequencies ($f$) and average branch widths ($w$). Figure 3(h) shows the areal ratios, defined as the area of the fully developed pattern ($A_p$) normalized by the area of the smallest circle enclosing it ($A = \pi R^2$, Fig. 2(b)), as a function of aging time $t_w$. We see that the areal ratios $A_p/A$ for immiscible patterns are always greater than for the miscible ones at the same suspension age due to the wider branching morphologies in the former case. The measurements of average branch frequencies and average branch widths are performed by analyzing a narrow annular region close to the outermost boundary of the fully developed interfacial patterns, as displayed in Supplementary Fig. S4. The average branch frequency is defined as $f = < N/2\pi r >_r$, where $N$ is the number of branches intersecting a circle of radius $r$ and $< >_r$ denotes the average over circles of different radii in the annulus of interest. As seen in Fig. 3(i), viscoelastic fractures (VEF) display smaller branch frequencies when compared to dendritic patterns (DP) and dense viscous patterns (DVP). The fingers formed in immiscible displacement experiments are thick and of uniform widths (Fig. 3(j)) and point to the critical role that surface tension plays in stabilizing flow instabilities. To conclude, we note that the interfacial tension between the fluid pair and the aging time of the displaced suspension are reliable
control parameters in determining elasticity-induced changes in the morphological features of miscible and immiscible displacement patterns.

3.2 Pattern morphology can be tuned by controlling the shear-thinning rheology of the displaced clay suspension

The viscous modulus of a 3.25% w/v Laponite clay suspension stabilizes at $t_w \approx 24$ h whereas its elastic modulus evolves continuously (Fig. 1(a)). We next explore the radial displacement of clay suspensions

![Images of displacement patterns](image)

Fig. 4: Miscible displacement patterns in grayscale, obtained by the displacement of 3.25% w/v clay suspensions of $t_w = 24$ h by water at various flow rates $q$: (a1-c1) viscoelastic fractures (VEF) and (d1-f1) dendritic patterns (DP). Immiscible displacement patterns for identical flow rates as in (a1-f1): (a2-c2) jagged patterns (JP) and (d2-f2) flower patterns (FP). All the patterns have fixed longest finger length $R = 7.01 \pm 0.01$ cm. The scale bar is 4 cm. (g) $A_p/A$ as a function of flow rates for miscible ($\sigma = 0$) and immiscible ($\sigma \neq 0$) displacements. (h) Average branch frequency ($f$, □) and average width ($w$, △) of the fingers as a function of $q$ for miscible ($\sigma = 0$) displacement patterns. (i) $f$ (○) and $w$ (▽) vs. $q$ for immiscible ($\sigma \neq 0$) displacement patterns. The distinct morphological regimes are mapped on a color bar using the natural logarithmic values of the areal ratios: $\ln(A_p/A)$.
of age $t_w = 24$ h in the Hele-Shaw cell while injecting Newtonian displacing fluids at different flow rates $q$. Viscoelastic fractures (VEF) with perpendicular offshoots are observed when water displaces the highly elastic clay suspensions at low flow rates (Figs. 4(a1-c1)). The low driving pressures cause minimal shear-thinning of the displaced clay suspension such that fractures propagate rapidly due to the release of elastic stresses. On increasing the flow rate, VEF are replaced by more ramified dendritic patterns (DP) with pointed tips displaying multiple splitting events during their growth (Figs. 4(d1-f1)). The average finger-tip velocities ($\bar{U}$) increase with increasing flow rate of the displacing fluid (Supplementary Fig. S5(a)). The shear rates ($158.82 \text{ s}^{-1} \leq \dot{\gamma} \leq 1907.05 \text{ s}^{-1}$) imposed at the finger-tip by the displacing fluid, estimated using $2\bar{U}/b$ [33] as discussed earlier, therefore also increase with increasing flow rates, resulting in considerable shear-thinning of the displaced clay suspensions. At the highest flow rates of the displacing fluid, the fragile microstructures formed by the self-assembling Laponite disks rupture irreversibly. This reduces suspension elasticity and results in the observed change in interfacial pattern morphology from VEF to DP.

The immiscible displacement of clay suspensions with mineral oil leads to a transition from jagged patterns (JP) to flower patterns (FP) as the flow rate of the displacing mineral oil is increased (Figs. 4(a2-f2)). The finger-tip velocities ($\bar{U}$) and the corresponding shear rates ($4.70 \text{ s}^{-1} \leq \dot{\gamma} \leq 82.35 \text{ s}^{-1}$) imposed by the mineral oil increase with increasing flow rate but are always less than those noted in miscible displacement experiments (Supplementary Fig. S5(a,b)). In order to understand the observed flow stabilization against viscous fingering instabilities at increasing flow rates, we estimate the viscosity ratios of the fluid pair $\eta_{in}/\eta_{out}$, where $\eta_{in}$ is the viscosity of the displacing Newtonian fluid and $\eta_{out}$ is the shear-dependent viscosity of the displaced clay suspension at different flow rates $q$. The shear-thinning of the displaced clay suspensions at high shear rates leads to an increase in the viscosity ratio of the fluid pair (Supplementary Information section ST2). This reduces the destabilizing action of viscous forces and results in increasingly stable interfaces at high injection flow rates. Such suppression of viscous instabilities has been noted in previous work involving Newtonian-Newtonian [17,36] and Newtonian-non-Newtonian displacements [5]. Since mineral oil is more viscous than water (Fig. 1(c)), the increase in viscosity ratio with increasing $q$ is more pronounced in immiscible displacements when compared to the miscible cases. This causes the observed flow stabilization and the transition from JP to FP when $q$ is increased (Fig. 4(a2-f2)). We conclude from the data presented in Fig. 4 that shear-thinning effects dominate as the injection flow rate increases, and the flow stabilizes due to two factors, viz., an increase in the viscosity ratio of the fluid pair and the presence of a non-zero interfa-
cial tension. Pattern morphologies at the radial quasi-two-dimensional interface can therefore also be effectively tuned by controlling the flow rate of the displacing fluid.

As in section 3.1, we quantify the morphologies of the observed interfacial patterns in terms of their areal ratios ($A_p/A$), average branch frequencies ($f$) and average branch widths ($w$). Similar to the

![Graph](image)

**Fig. 5:** (a) $A_p/A$ vs. $fw$ for miscible ($\triangle$ for variable $t_w$, $\Box$ for variable $q$ ) and immiscible ($\triangledown$ for variable $t_w$, $\circ$ for variable $q$) displacements. (b) Phase diagram relating the flow rate $q$, the aging time $t_w$ and the interfacial tension $\sigma$: bottom plane shows miscible ($\sigma = 0$) displacements with three different pattern morphologies: dense viscous patterns (DVP), dendritic patterns (DP) and viscoelastic fractures (VEF). Top plane shows immiscible ($\sigma \neq 0$) displacement patterns with three different morphologies: flower patterns (FP), jagged patterns (JP) and stable patterns (SP). The color bar represents the different patterns in terms of the natural logarithms of their areal ratios: $\ln(A_p/A)$. The colors of the solid circles in the phase diagram have a one-to-one mapping with the distinct interfacial patterns.
earlier experiments at different suspension aging times, $t_w$ (Fig. 3(h)), $A_P/A$ is larger for immiscible displacement patterns when compared to the miscible ones at a fixed flow rate of the displacing fluid (Fig. 4(g)). Dendritic patterns (DP) obtained at high $q$ during miscible displacements display larger branch frequencies when compared to viscoelastic fractures (VEF). The presence of non-zero interfacial tension results in larger average branch widths $w$ in the immiscible displacement patterns (Fig. 4(i)) when compared to their miscible counterparts (Fig. 4(h)). The interfacial patterns obtained at different $t_w$ of the displaced clay suspensions (displayed in Figs. 3(a1-g1,a2-g2)) can therefore be recovered by keeping $t_w$ fixed while appropriately changing the flow rate $q$ of the displacing fluids (Figs. 4(a1-f2,a2-f2)). Interestingly, shear-thinning of the displaced clay suspensions at increasing flow rates results in the same sequence of pattern morphologies that were observed while decreasing $t_w$ of the displaced suspension at a fixed injection flow rate of the displacing fluid.

Regardless of whether the interfacial patterns were generated while changing the flow rate of the displacing fluids or the aging time of the displaced clay suspensions, we see from Fig. 5(a) that their areal ratios ($A_p/A$) are linearly correlated with the products of their average branch frequencies ($f$) and widths ($w$). Furthermore, we observe a stable pattern (SP) with a circularly growing interface at all times during the immiscible displacement of a clay suspension of low age ($t_w = 1$ h) at a high flow rate ($q = 50$ ml/min, Supplementary Fig. S6). This stable circular interface arises due to the large viscosity ratio (Supplementary Fig. S5(c)) of the fluid pair which causes a suppression of viscous instability. Since it is not feasible to estimate the branch frequency and width of the stable pattern, we compute the natural logarithms of the areal ratios, ln($A_p/A$), of all the patterns and segregate them in a three-dimensional nonequilibrium phase diagram (Fig. 5(b)) spanning the aging time ($t_w$) of the displaced clay suspension, the flow rate ($q$) of the displacing fluid and the interfacial tension ($\sigma$). All the interfacial patterns obtained in our experiments are represented on the phase diagram in Fig. 5(b) by distinct colors based on their respective ln($A_p/A$) values, as defined in the color bar. We therefore identify ln($A_p/A$) as a single parameter that can be used to effectively segregate the wide range of quasi-two-dimensional interfacial patterns formed in Newtonian-non-Newtonian displacement experiments.
4 Conclusions

We observe a wide variety of interfacial morphologies during the miscible and immiscible displacements of an aging aqueous Laponite clay suspension in a radial Hele-Shaw cell. These interfacial morphologies strongly depend on the elasticity of the displaced suspension, parameterized in terms of the suspension aging time, the injection flow rate of the displaced Newtonian fluid, and the miscibility of the fluid pair. For both miscible and immiscible displacements, we understand the transitions between the observed interfacial patterns by considering the competition between interfacial, elastic and viscous forces in the aging and shear-thinning clay suspensions. Our work supplements the existing experimental literature involving displacements of charged clay suspensions \cite{6,37,38} by establishing the suspension aging time $t_w$ as an additional parameter that reliably determines interfacial pattern morphologies. This has important implications in understanding geophysical phenomena such as river delta formation, and in controlling the displacements of viscoelastic materials such as mud and cement slurries.

The control of instabilities at the interface between a pair of fluids is crucial in material transport. Interfacial instabilities are essential in the design and optimization of several processes such as filtration and flow in porous media \cite{10}. However, they are undesirable in certain scenarios such as in the formation of dendrites at the anode-electrolyte interface in rechargeable lithium batteries \cite{39} and the emergence of viscous fingers at the water-oil interface in enhanced oil recovery \cite{40}. Our demonstration that the natural logarithm of the areal ratio of an interfacial pattern can effectively segregate distinct morphologies on a three-dimensional phase diagram can have far-reaching ramifications in controlling and predicting pattern morphologies between a fluid pair with well-characterized physicochemical properties.

Interparticle electrostatic interactions, and therefore the aging dynamics of a charged colloidal clay suspension, can be altered by varying clay concentration, temperature, and by incorporating dissociating additives such as salts and acids \cite{25,27}. It would be of interest to systematically study the roles of diverse physicochemical conditions in the formation of interfacial patterns during the displacement of a clay suspension by miscible and immiscible Newtonian fluids. Another possible extension of this work could involve the addition of non-dissociating molecules such as glucose, which induces structure in the aqueous suspension medium, or N,N-Dimethylformamide, which disrupts water structure \cite{22}. The concomitant changes in the aging dynamics and rheology of the suspension will strongly influence the morphologies of the patterns. While this paper focuses only on the distinct
morphological features of fully developed interfacial patterns, we note that the time-evolution of the
interface is quite different in each case. A complete understanding of the patterns that form at the
interface between an aging clay suspension and a Newtonian fluid therefore requires high-speed video
imaging of pattern onset and growth. Such work is ongoing.

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Supplementary Information

Pattern selection in radial Hele-Shaw flow: effects of interfacial tension and aging fluid viscoelasticity

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ST1 Additional experimental details

ST1.1 Sample preparation

Hygroscopic Laponite® XLG powder (BYK additives Inc.) was baked in an oven at 120°C for 18-20 hours prior to sample preparation. We prepared an aqueous suspension of concentration 3.25% w/v by adding 1.625 g of dried Laponite powder to 50 ml double distilled water (Millipore Corp., resistivity 18.2 MΩ.cm). We stirred the sample continuously and vigorously for 45 minutes at room temperature (25°C), and the resulting suspension was filtered through a syringe filter (Millex®, Sigma Aldrich) of pore size = 0.45 µm.

ST1.2 Hele-Shaw Cell

A radial Hele-Shaw cell (Fig. 2(a) in the main paper) comprising two circular glass plates, each of radius 30 cm and thickness 10 mm, was used to study interfacial patterns. Teflon spacers were used to ensure a constant gap of 170 µm between the plates. An aqueous clay suspension, the displaced fluid in our experiments, was first filled in this quasi two-dimensional geometry with a syringe pump (NE-8000, New Era Pump Systems, USA) at a constant volumetric flow rate of 20 ml/min through a 3 mm hole drilled in the top plate. Water and mineral oil were used as the displacing fluids in two separate sets of experiments and were injected through the same hole after a pre-determined time $t_w$.

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at flow rates \((q)\) ranging from 1 ml/min to 50 ml/min. We used water and mineral oil for the miscible (interfacial tension \(\sigma = 0\)) and immiscible \((\sigma \neq 0)\) displacement experiments, respectively. Water and mineral oil were respectively dyed with Rhodamine B \((\geq 95\% \text{ (HPLC)})\) and Oil Red O, procured from Sigma Aldrich, to ensure adequate contrast at the interface. We recorded the growth of the interfacial patterns with a DSLR camera (D5200, Nikon, Japan) that was set up below the Hele-Shaw cell with a spatial resolution of 1920×1080 pixels (one pixel area = \(2.2 \times 10^{-3} \text{ cm}^2\)) and a frame rate of 30 fps. An area enclosed by a circle of radius 1.18 cm is excluded to avoid experimental artifacts in image analysis.

**ST1.3 Rheology**

The sample preparation protocol for the rheological measurements was identical to that for the Hele-Shaw experiments. The sample temperature was maintained at room temperature \((25^\circ \text{C})\) using a Peltier temperature control device (C/PPTD 180/MD). We loaded the freshly prepared Laponite suspension in the cone and plate geometry and shear melted it at a high shear rate of 500 s\(^{-1}\) to remove any memory of the sample loading process. The time at which shear application was stopped and the sample was allowed to evolve spontaneously in the rheological experiments was noted as aging time \(t_w = 0\). The elastic and viscous moduli \((G'\text{ and } G'')\) of Laponite suspensions as a function of \(t_w\) were recorded at an applied strain amplitude 0.5 % and angular frequency 1 rad/s.
Supplementary Fig. S1: Raw RGB images illustrating distinct morphologies of interfacial patterns obtained during the miscible and immiscible displacements of aging Laponite® suspensions at constant flow rate $q = 5 \text{ ml/min}$. (a1-a3) Temporal evolution of dense viscous pattern (DVP) at $t_w = 1 \text{ h}$ for an interfacial tension $\sigma = 0$. (b1-b3) Temporal evolution of dendritic pattern (DP) for $t_w = 3 \text{ h}$ and $\sigma = 0$. (c1-c3) Temporal evolution of viscoelastic fracture (VEF) at $t_w = 24 \text{ h}$ and $\sigma = 0$. (d1-d3) Temporal evolution of flower pattern (FP) at $t_w = 1 \text{ h}$ and $\sigma \neq 0$. (e1-e3) Temporal evolution of jagged pattern (JP) at $t_w = 24 \text{ h}$ and $\sigma \neq 0$. 
Supplementary Fig. S2: Flow chart illustrating the binarization of RGB images.
Supplementary Fig. S3: The time-averaged finger-tip velocities vs. aging time $t_w$ for (a) miscible (■) and (b) immiscible (●) displacements of Laponite suspensions by the displacing Newtonian fluids (water and oil respectively) at constant flow rate ($q = 5$ ml/min). DVP, DP, VEF, FP and JP refer to the experimentally observed dense viscous patterns, dendritic patterns, viscoelastic fractures, flower patterns and jagged patterns, respectively.

Supplementary Fig. S4: A representative interfacial pattern showing the annular region of width 1.5 cm, used in the calculations of average branch frequencies ($f$) and branch widths ($w$). The average branch frequency, $f$, is estimated as $f = \frac{1}{m} \sum_{j=1}^{m} \frac{N_j}{2\pi r_j}$ over $m = 32$ circles within the annular region. Here, $N_j$ is the number of branches intersecting a circle of radius $r_j$, such that $5.2 \text{ cm} \leq r_j \leq 6.7 \text{ cm}$. The average branch width, $w$, of an interfacial patterns is estimated as $w = \frac{1}{m} \sum_{j=1}^{m} <W >_{r_j}$, where $<W >_{r_j}$ denotes the average width of the branches intersecting a circle of radius $r_j$ in the same annular region of interest.
ST2  Calculations of viscosity ratios

The values of the time-averaged finger-tip velocity $\bar{U}$ and shear rates $\dot{\gamma}$ are estimated using the protocols mentioned in section 3.1 in the main paper. For increase in the injection flow rate, $q$, of the displacing fluid, the estimated values of $\bar{U}$ vary in the range of $1.35 - 16.21$ cm/s for miscible (Supplementary Fig. S5(a)) and $0.04 - 0.70$ cm/s for immiscible displacements (Supplementary Fig. S5(b)). The shear-dependent viscosities $\eta_{\text{out}}$ of the displaced Laponite suspensions are extracted at shear rates $(2\bar{U}/b)$ from the data in Fig. 1(b). The viscosities $\eta_{\text{in}}$ of the displacing Newtonian fluids are extracted at shear rate $\dot{\gamma} \to 0$ in Fig. 1(c). The viscosity ratios $\eta_{\text{in}}/\eta_{\text{out}}$ of the fluid pairs are then evaluated and plotted in Fig. S5(c).

Supplementary Fig. S5: The time-averaged finger-tip velocities $\bar{U}$ vs. $q$ for (a) miscible (■) and (b) immiscible (●) displacements of Laponite suspensions at age ($t_w = 24$ hrs). (c) Viscosity ratios ($\eta_{\text{in}}/\eta_{\text{out}}$) vs. $q$ where $\eta_{\text{in}}$ is the viscosity of the displacing Newtonian fluids and $\eta_{\text{out}}$ is the shear-dependent viscosity of the displaced Laponite suspension. The viscosity ratio corresponding to the stable pattern observed during displacement of a Laponite suspension of $t_w = 1$ h by mineral oil at $q = 50$ ml/min (●) is also included.

Supplementary Fig. S6: Temporal evolution of stable patterns (SP) at $t_w = 1$ h, $q = 50$ ml/min and $\sigma \neq 0$ in RGB format.