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PAPER

Permeability heterogeneity and bulk linear elasticity of displaced clay suspensions determine interfacial pattern morphologies in Hele–Shaw experiments

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E-mail: ranjini@rri.res.in**Keywords:** viscous fingering instability, clay, interfacial patterns, skewering, zig–zag propagationSupplementary material for this article is available [online](#)

Abstract

When a less viscous Newtonian fluid displaces an aging aqueous clay suspension in a confined space, a rich array of interfacial patterns emerges due to a predominantly viscous instability. Earlier studies on clay displacement identified macroscopic morphologies such as fingering and fracturing, but the local finger propagation modes did not receive much attention. For the present work, we controlled the mechanical properties of clay suspensions by incorporating additives and observed a range of local finger propagation modes when the suspensions were radially displaced by water in a Hele–Shaw cell. When the elasticity of clay was low, the interfacial dynamics exhibited features of nonlinear viscous fingering in heterogeneous media. By tuning the nature and content of additives that delay clay aging, we uncovered two novel finger propagation mechanisms: skewering and zig–zag growth, both characterised by tortuous trajectories and thick finger profiles. These patterns have hitherto never been observed in experiments with colloidal systems. For moderate clay elasticities, we report that shear-thinning-induced flow anisotropy leads to the formation of dendrites with dominant side branches. As clay elasticity increases due to the incorporation of salts, the energy required to create fractures becomes smaller than that for system-wide yielding. This scenario is characterised by the emergence of viscoelastic fractures. Our work demonstrates that incorporating additives in the suspension medium significantly modifies clay elasticity and permeability and is an effective strategy to manipulate the growth and morphologies of instabilities at clay–water interfaces.

1. Introduction

Studies of fluid flow in porous media are crucial for modelling material transport in industrial and natural processes [1, 2]. When a less viscous fluid displaces a more viscous fluid in a quasi-two-dimensional geometry, the interface becomes unstable and intricate fingering patterns form due to a viscous fingering instability [3–6]. Understanding such instabilities is important in various fields, such as in enhanced oil recovery processes [7], carbon dioxide sequestration [8] and enhanced mixing at low Reynolds numbers [9]. In such displacement experiments, finger propagation, fingertip splitting and pattern morphology depend on several factors, such as viscosity contrast and interfacial tension between the fluid pair [3, 6, 10], the injection flow rate or injection pressure of the less viscous displacing fluid [11], and the gap of the Hele–Shaw (HS) cell [12]. The interfacial patterns that form during miscible displacement experiments (characterised by interfacial tension between the fluid pair tending to zero) are more complex than those formed by immiscible displacement [13]. In miscible Newtonian displacements, linear stability analysis of the moving interface shows that the wavelength of unstable fingers is affected by dispersion [14]. The emergence of complex patterns can also be driven by local heterogeneities of the displaced medium. In sharp contrast to reports on interfacial pattern formation in homogeneous

media, numerical simulations of viscous fingering in heterogeneous media [15, 16] have uncovered a novel channelling regime, in which a finger advances by preferentially choosing high-permeability paths.

The complexity of interfacial patterns increases dramatically when either or both fluids are non-Newtonian [17]. Non-Laplacian pressure fields and non-linear mode coupling lead to deviation from Darcy's law and result in side branching and dendritic growth [18–20]. The shear-thinning rheology [21, 22], non-zero yield stress [23, 24] and elasticity [25, 26] of a non-Newtonian fluid in a displacement experiment can significantly alter interfacial pattern morphologies [17, 27–30]. When a shear-thinning fluid is displaced, the velocity of the propagating finger is maximum at the fingertip [22]. The resistance to finger growth is minimal in the forward direction, which leads to the formation of narrow fingers and delays the onset of tip-splitting (TS). Given the anisotropic flow conditions, the lower viscosity in the flow direction when compared to the lateral direction results in the formation of side branches and dendritic patterns (DPs) [22, 30]. When an yield stress fluid is displaced, fractures develop due to an elastic response below their yielding threshold, while viscous fingers appear above the yielding point [23, 24]. The elasticity of a displaced viscoelastic fluid can shift the most unstable wavelength and drive a transition from viscous fingers to fracture-like patterns with bi-Laplacian growth [31, 32]. Earlier experiments on displacement of Bentonite clay by water reported a transition from fractal viscous fingering (TS at low Deborah numbers [33]) to viscoelastic fracturing (at high Deborah numbers) by increasing clay concentration or by decreasing HS cell gap [26, 34].

In this report, we describe experiments in which aging aqueous Laponite® clay suspensions were displaced by miscible water in a confined, radial HS geometry. In an aqueous medium, Laponite clay particles carry negative charges on their faces and positive charges on the rims below their isoelectric point at $\text{pH} < 11$ [35–37]. The mechanical properties of aqueous clay suspensions in the concentration range between 1.0% w/v and 4.0% w/v evolve over time in a physical aging process that is driven by the gradual self-assembly of the clay particles into system-spanning networks of interconnected particles [37–39]. We had reported in earlier studies that miscible displacements of clay suspensions of progressively higher ages resulted in interfacial patterns that transformed from dense viscous (dominated by TS) to DPs (dominated by side branching) and eventually to viscoelastic fractures (VEFs) [30, 40].

It has been shown in recent studies that the incorporation of different additives, such as salts [41, 42], acids [43] and non-dissociative molecules [42] in the suspension medium can accelerate or delay the aging dynamics of clay. It should therefore be possible to generate an even broader range of novel interfacial patterns during miscible HS displacements of aqueous clay by incorporating appropriate additives to the aqueous medium in which the clay was prepared. In contrast to previous studies of confined displacements of clays that focussed mainly on the global pattern morphologies, the present work focuses on local finger propagation modes across a broader elasticity range. This was achieved at a fixed clay concentration (3% w/v) and a fixed clay age (5 h) by utilising additives to tune the physical aging of the clay sample. The growth of interfacial patterns due to the miscible displacement of clay that was prepared in pure water without any additive was primarily dominated by (i) TS, where the tip of a finger divided into two distinct fingers, (ii) finger coalescence, where a slower trailing finger merged with a faster neighbouring finger, and (iii) trailing lobe detachment (TLD), where coalescence events resulted in detachment of small portions of the displaced clay from the main body. When additives that delay clay aging, such as dimethylformamide (DMF) and tetrasodium pyrophosphate (TSPP) [42, 44], were introduced in the medium while preparing aqueous clay, we observed multiple nonlinear viscous fingering mechanisms like skewering, dense-branching, channelling and zig-zag finger propagation, with the finger propagation profile seen to be highly sensitive to the nature of the additive used. In contrast, when additives like sodium chloride (NaCl), potassium chloride (KCl) and glucose were added to the suspension medium, clay aging and sample elasticity were seen to enhance rapidly and the interfacial pattern morphologies were dominated by side branches and fractures.

The patterns in these experiments were sensitive only to the concentration of the additive rather than to its chemical composition. The observed variations in finger propagation modes arise from the contrasting effects of various additives on clay suspension elasticity and microstructural development. We characterised the distinct interfacial pattern morphologies by computing time-dependent areal ratios, A_p/A [30, 40], a measure of pattern compactness in two dimensions, the instantaneous velocity of the longest fingertip, U , and the total number of fingertips, N . In addition to these global pattern-level metrics, we estimated single-finger-scale metrics, such as finger-path tortuosity to quantify finger waviness, and finger-width distributions to quantitatively distinguish between various local finger propagation modes. To conclude, a rich array of interfacial patterns was generated during the confined miscible displacements of clay samples of a fixed concentration and age. This was achieved by changing the elasticity and microstructural development of the displaced clay suspension via the controlled incorporation of additives (those that delay aging, such as DMF and TSPP, and those that accelerate it, such as NaCl, KCl,

and glucose) in the suspension medium. The present work also reports the first experimental observations of zigzag and skewering propagation modes in colloidal systems.

2. Material and Methods

2.1. Preparation and characterisation of aqueous Laponite clay

Moisture from Laponite[®] XLG powder (purchased from BYK Additives Inc.) was removed by baking at 120°C for 18–20 h in an oven. Aqueous media were prepared by mixing desired concentrations of additives with deionised water (Millipore Corp., resistivity 18.2 MΩ cm⁻¹). NaCl (LABORT Fine Chem Pvt. Ltd), KCl (Sigma-Aldrich), glucose (Sigma-Aldrich), (DMF, SDFCL Fine Chem Pvt. Ltd) and (TSPP, E. Merck (India) Ltd) were used as received without further purification. Laponite clay suspensions were freshly prepared by weighing predetermined amounts of dried Laponite powder, which were gradually added to an aqueous medium while stirring vigorously to prepare pure clay samples of concentration 3.0% w/v. Clay was added to solutions prepared with additives at predetermined concentrations to produce additional batches of 3.0% clay suspensions. After 45 min of vigorous stirring, the transparent clay samples were filtered through syringe filters of pore size 0.45 μm (Millex[®], Sigma Aldrich) and immediately loaded into experimental geometries for rheological and displacement experiments. The pH values of all the clay samples were measured using an Eutech 2700 pH meter.

Laponite clay is composed of disk-shaped nanoplatelets with an average diameter of 25–30 nm and a thickness of 1 nm. Debye screening of electrostatic interactions results in short-range repulsion between like-charged surfaces (face–face) and short-range attraction between oppositely charged surfaces (edge–face). These interactions govern the gradual self-assembly of the clay particles into system-spanning networks of interconnected particles. As a result, the mechanical properties of aqueous clay suspensions evolve over time in a physical aging process [37–39]. The presence of NaCl, KCl, and glucose in the suspension medium promotes the formation of microstructures and accelerates the physical aging of clay [42]. In contrast, DMF and TSPP delay or inhibit clay microstructure formation [42, 44] and delay the physical aging process. The additives used by us in this work and their influence on clay aging are summarised in table 1. We see that the aging and rheology of clay suspensions are strongly dependent on the additive incorporated in the aqueous medium during preparation. It is commonly believed that clusters of clay particles exfoliate into individual platelets when dispersed in water. However, several studies have shown that clay particles can exist as rigid aggregates called tactoids that consist of multiple platelets arranged in one dimensional stacks [45, 46].

2.2. Radial HS cell used for displacement experiments

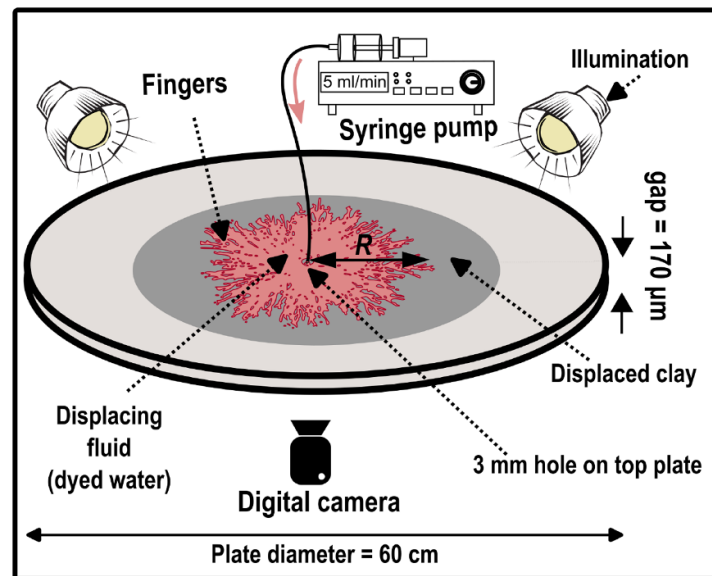
Displacement experiments were performed in a quasi-two-dimensional radial HS cell [47] consisting of two horizontally arranged parallel glass plates, each of radius 30 cm and thickness 1 cm, as shown in figure 1. The gap between the two circular glass plates of the HS cell was maintained at 170 μm with Teflon spacers. A freshly prepared clay sample was injected into the HS cell through the 3 mm hole at the centre of the top plate. The time at which suspension loading was completed was noted as the aging time, $t_w = 0$ h. The clay sample was left undisturbed inside the HS cell and allowed to age spontaneously for $t_w = 5$ h. At this instant, the displacing fluid was injected through the same central hole at a constant injection flow rate $q = 5$ ml min⁻¹ using a syringe pump (NE-8000, New Era Pump Systems, USA). Deionised water from Millipore Corp., used as the displacing Newtonian fluid in all our experiments, was dyed by dissolving a small amount of Rhodamine B (Sigma-Aldrich) to enhance interfacial contrast. The evolution of interfacial patterns was recorded at 29.97 frames per second at 1920 × 1080 pixel resolution using a Nikon D5200 DSLR camera. The image sequences of the observed interfacial patterns were analysed using the MATLAB Image Processing Toolbox. Spatial resolution of the acquired images was 0.472 mm pixel⁻¹. All experiments were performed at room temperature. The Peclet number, $Pe = UL/D$, which controls interface blurriness during imaging, was computed with finger velocity $U \sim 1$ cm s⁻¹, relevant pattern lengthscale $L = 10$ cm, and diffusion coefficient of dye $D = 10^{-6}$ cm² s⁻¹, and estimated to be $\sim 10^7$ for our flow experiments. Simultaneously, using $\eta_{\text{displaced}}$ values for clay from flow curve measurements in figure 2 and $\eta_{\text{displacing}} = 10^{-3}$ Pa for water, we estimate that the viscosity ratios, $\eta_{\text{displaced}}/\eta_{\text{displacing}}$ in our experiments range between $\sim 10^3$ and 10^6 . All our experiments were therefore performed in viscously unstable and advection-dominated regimes, where interfacial pattern selection is governed by viscous and elastic forces rather than diffusive processes.

2.3. Rheological experiments to measure elastic and viscous moduli of aqueous clay

An Anton Paar MCR 702 rheometer was used to perform rheological measurements. A freshly prepared clay suspension was loaded in the measuring geometry and shear melted at a high shear rate $\dot{\gamma}$

Table 1. Descriptions of additives used in this work and the mechanism by which clay aging is modified in the presence of each additive.

Additive name	Additive nature	Concentration used	Mechanism by which clay aging is modified	Influence on clay aging
Tetrasodium pyrophosphate (TSPP)	Dissociative and kosmotropic	1–2 mM	Inhibits edge-face bonding by adsorbing on clay rims	Delays/inhibits clay aging
Dimethylformamide (DMF)	Nondissociative and chaotropic	130–260 mM	Disrupts medium structure	Delays clay aging
Sodium chloride (NaCl)	Dissociative and kosmotropic	1–2 mM	Screens electrostatic repulsion	Accelerates clay aging
Potassium chloride (KCl)	Dissociative and chaotropic	1–2 mM	Screens electrostatic repulsion	Accelerates clay aging
Glucose	Nondissociative and kosmotropic	130–260 mM	Promotes medium structure	Accelerates clay aging

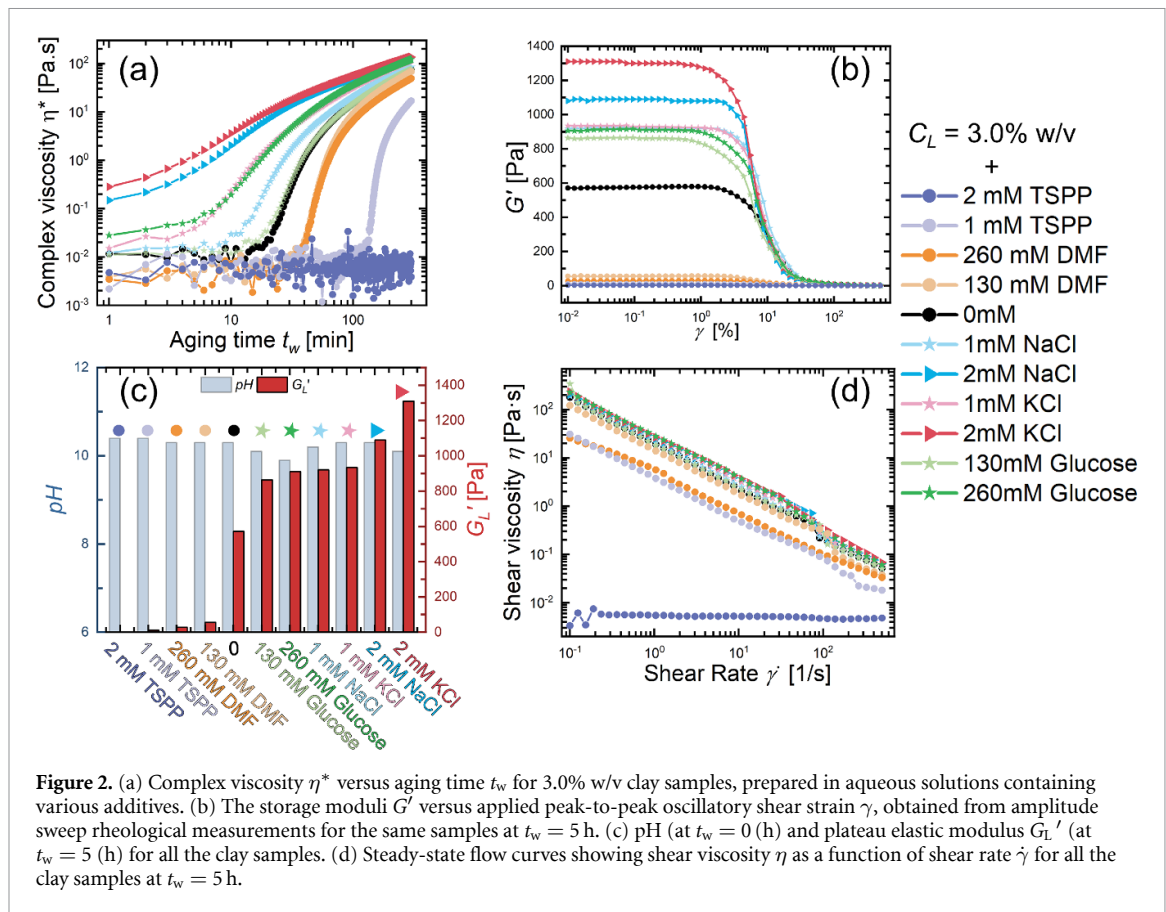
**Figure 1.** Schematic illustration of the radial Hele-Shaw cell geometry with plate separation $b = 170 \mu\text{m}$ used in displacement experiments. Here, the grey region represents the displaced clay, while the pink region represents the displacing fluid (water). A small amount of dye was added to the displacing fluid to visualise the interface clearly. The length of the longest propagating finger is denoted by R .

$= 500 \text{ s}^{-1}$ for 2 min to ensure identical initial conditions in all rheological experiments. The time at which shear melting was stopped was noted as aging time $t_w = 0$ h. A parallel plate (PP50) measuring geometry was used to perform time sweep and flow curve measurements, while a double gap geometry (DG26.7) was used for amplitude sweep measurements. Time sweep measurements were initiated immediately after shear melting was stopped. For the amplitude sweep and flow curve experiments, the clay sample was left undisturbed in the measuring geometry and allowed to age to $t_w = 5$ h. The suspension temperature was maintained at 22°C throughout the experimental duration using a Peltier device and a water circulation system (Viscotherm VT2) in parallel plate and double gap geometries, respectively. Silicone oil of viscosity five cSt was used as a solvent trap to prevent water evaporation during the measurements. The data presented in figure 2 was from a single trial for each sample and exhibits the same trends as already reported in the literature [42]. For each rheological condition shown in figure 2, multiple displacement experiments were subsequently performed, yielding reproducible interfacial pattern morphologies.

3. Results and discussion

3.1. Controlling the aging and rheology of Laponite clay by incorporating additives

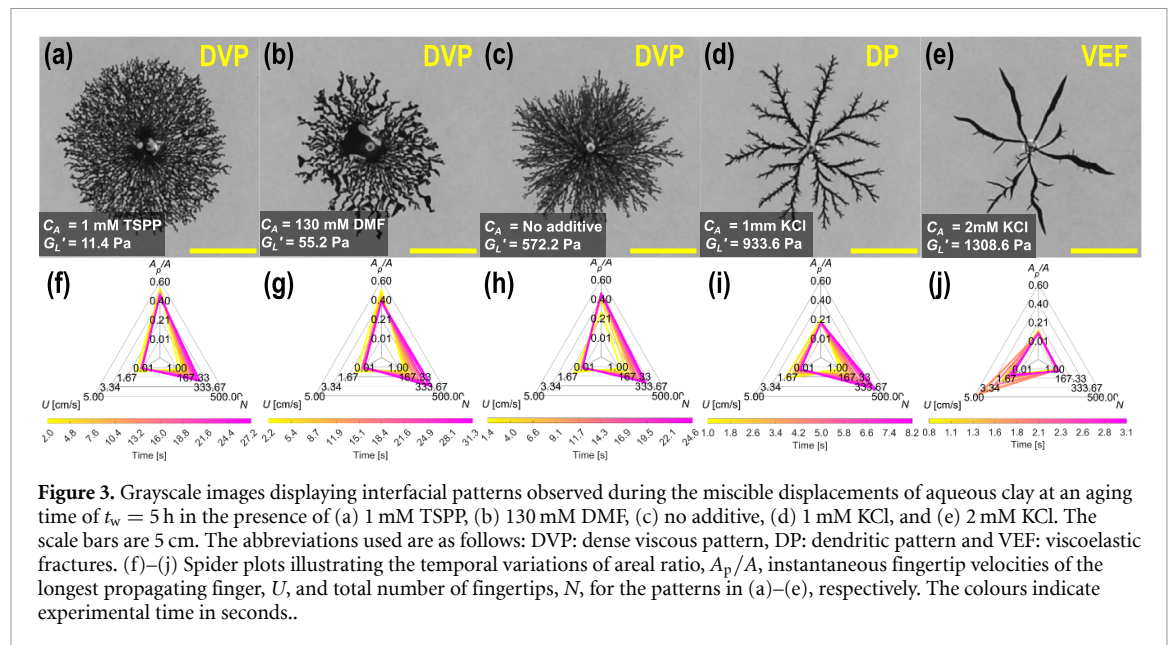
We measured the rheological responses of the pure clay sample and the clay samples with additives, viz., DMF, TSPP, sodium chloride (NaCl), potassium chloride (KCl) and glucose, incorporated in the aqueous suspension medium. Figure 2(a) shows the time evolution of the complex viscosity, defined



as $\eta^* = \sqrt{G'^2 + G''^2}/\omega$, for each clay sample studied in this work. In the expression for η^* , G' and G'' are the elastic and viscous moduli of the samples, respectively, measured in oscillatory time-sweep experiments at an angular frequency, ω , of 6 rad s^{-1} and shear strain amplitude, γ , of 0.1%. Figure 2(b) presents the storage modulus G' as a function of strain amplitude for all the samples at an aging time of $t_w = 5$ h. The corresponding loss moduli G'' obtained from these amplitude-sweep experiments are provided in supplementary figure S1. The linear elastic moduli G'_L , which represent the elastic response of the sample and are extracted from the linear rheological data as described in figure S2 of the supplementary information, are shown in figure 2(c). Figure 2(c) also displays the pH values of all the suspensions. We note that the measured pH values all lie between 9.9 and 10.4. Since the Laponite isoelectric point is 11, this observation indicates that the Laponite disks in all the clay samples are negatively charged while their rims are positively charged [35]. It is therefore expected that the clay particles self-assemble in the aqueous medium via overlapping coins or house-of-card particle associations. Finally, figure 2(d) shows the steady-state shear viscosity η as a function of shear rate $\dot{\gamma}$ for all the clay suspensions, as estimated from flow curves measured in rotational rheology experiments.

The addition of both DMF and TSPP hinders the development of clay microstructures and delays physical aging of the suspension. DMF disrupts the hydrogen-bonding network in water and limits the swelling of clay tactoids, which accelerates particle dynamics and delays suspension aging [42]. TSPP, on the other hand, neutralises the positive rim charges on the clay particles via the attachment of pyrophosphate ($\text{P}_2\text{O}_7^{4-}$) groups. This inhibits edge-face associations and delays network formation [36, 44]. Therefore, even though the mechanisms are quite distinct, DMF and TSPP both have a similar effect on clay aging. The extremely low steady-shear viscosity and the absence of shear-thinning behaviour in the 2 mM TSPP sample (blue circles in figure 2(d)) both indicate a lack of microstructure development. The low elastic moduli of clay at $t_w = 5$ h with DMF and TSPP added to the medium (figure 2(b)) are consistent with their delayed gelation behaviour.

In contrast, the addition of NaCl, KCl and glucose significantly accelerates the aging process by accelerating the buildup of self-assembled suspension microstructures [41, 42]. Monovalent salts like NaCl and KCl reduce the Debye screening lengths around the clay platelets, promote edge-face interactions between the particles and facilitate the formation of space-spanning particulate networks [41, 42, 48].



Glucose accelerates clay aging via a non-DLVO mechanism, wherein hydrogen bonds form between glucose and water molecules, which effectively reduces the amount of free water available to hydrate the clay [42, 49]. As a consequence, clay samples with glucose exhibit moderately high complex viscosities (green stars in figure 2(a)) and storage moduli (figure 2(b)), and shear-thinning behaviour (figure 2(d)). The rheological signatures in figures 2(a)–(c) are all consistent with the hypothesis that NaCl, KCl and glucose speed up clay aging by accelerating microstructural buildup. In contrast, the rheological signatures for DMF and TSPP are indicative of delayed suspension aging. Finally, we note that while clay elasticities are vastly different due to the incorporation of additives, they do not change appreciably due to physical aging during our displacement experiments which last between 3 and 40 seconds.

3.2. Fully developed interfacial pattern morphologies: classification and quantification

In agreement with earlier reports on displacement of clay by miscible water [26, 30, 34, 40], we identified three distinct interfacial patterns, *viz.* dense viscous patterns (DVPs), DPs, and VEFs by inspecting their global appearances. Images of the distinct interfacial patterns are presented in figures 3(a)–(e). Figures 3(a)–(c) show representative images of DVPs, which were generated during the displacement of clay samples prepared in pure water and in additive solutions containing DMF and TSPP that delay sample aging. Additional DVP images are provided in supplementary figure S3. DVP growth is predominantly driven by viscously unstable flow in homogeneous or heterogeneous media. These patterns were therefore observed only when the displaced clay had a lower rigidity, $G_L' \leq 573$ Pa, and did not exhibit strong nonlinear rheology. DPs (DP, figure 3(d)), characterised by dominant side branching, formed when clay samples with comparatively larger rigidities ($800 \text{ Pa} \leq G_L' \leq 950 \text{ Pa}$), achieved by adding moderate concentrations of polar salts (KCl and NaCl) and glucose, were displaced. In contrast to DVP, DP growth is expected to be governed by the shear-thinning rheology of the displaced clay. More images of DP in the presence of NaCl, KCl and glucose are presented in supplementary figure S4. VEF, figure 3(e) were observed for the most elastic clay samples ($G_L' \geq 1089$ Pa), prepared by using very high concentrations of polar salts and glucose in the suspension medium. These interfacial instabilities closely resemble the fracture patterns observed in brittle materials and are characterised by a very small number of primary fingers, fast fingertip propagation and the frequent shedding of side branches in a direction orthogonal to the primary fingers.

We next characterised the patterns by estimating the temporal evolution of certain metrics: (i) pattern areal ratios, A_p/A , where A_p is the areal coverage of the interfacial pattern and A is the area of the smallest circle enclosing the entire pattern at that instant. This parameter was proposed in earlier work [30, 40] to quantify the extent of the spatial coverage of interfacial patterns. We have also plotted (ii) the total number of fingertips, N , indicating the combined effects of TS, side branching and coalescence of the pattern studied here, and (iii) the instantaneous fingertip velocity, U , of the longest propagating finger, which parametrises the speed of the interfacial dynamics.

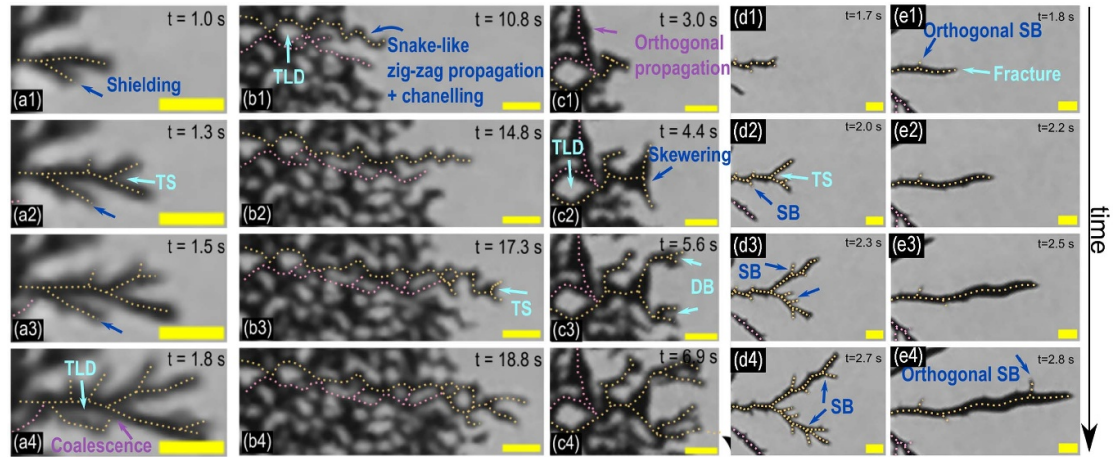


Figure 4. Cropped grayscale images displaying the temporal propagation profiles of fingers formed during the miscible displacements of 3.0% w/v clay at $t_w = 5$ h. The clay samples in these experiments were prepared (a1–a4) without additives, with (b1–b4) 2 mM TSPP, (c1–c4) 260 mM DMF, (d1–d4) 1 mM NaCl and (e1–e4) 2 mM NaCl. Dotted lines have been drawn to highlight the propagation mechanisms of the interfacial fingers. The scale bar in each image is 0.5 cm. The abbreviations used are as follows: TS: tip-splitting, TLD: trailing lobe detachment, DB: dense branching and SB: side branching.

Details of measurement protocols are provided in supplementary Information section ST2 and the temporal evolutions of all three metrics (A_p/A , N and U) for all samples are shown separately in supplementary figure S6. To simultaneously visualise the variations of all three parameters and classify different pattern morphologies, we constructed spider plots with A_p/A , N and U as three independent axes (figures 3(f)–(j)). The colour bars in figures 3(f)–(j) indicate experimental time in seconds. It is evident from these plots that the observed DVP, DP and VEF patterns display unique and distinct morphological features and growth kinetics. We see that DVPs are the most compact/ space-filling and propagate at the slowest speeds. DP, in contrast, are relatively less compact, as indicated by the intermediate values of A_p/A ratios in figure 3(i). We further note that the number of fingertips, N , shown in figure 3(i), increases more rapidly for DP than for DVP. The propagation velocities, U , for DP, shown in figure 3(i), are faster than for DVP at initial times and eventually slow down due to the shedding of multiple side branches. VEFs are the least compact of the three morphologies, as indicated by their lowest A_p/A values in figure 3(j). Since TS and side branching are minimal during the growth of VEF patterns, their morphologies exhibit a very small number of fingertips, N . Lastly, we note that VEF propagate rapidly during the initial stages, but pattern growth is substantially inhibited at later times due to the formation of orthogonal side branches. The propagation velocity of the longest finger in a VEF therefore shows a non-monotonic propagation velocity, U . Interestingly, one of the DPs studied by us, marked by a red arrow in supplementary information figure S6(c), exhibits a non-monotonic velocity profile, suggesting that this pattern lies in a crossover zone between DP and VEF morphologies.

Interestingly, prior studies have shown that transitions between dense viscous, dendritic, and VEF patterns can be achieved either by increasing the clay concentration or sample age, or by reducing the injection flow rate or cell gap [26, 34, 40]. Our results show that transitions between interfacial pattern can also be achieved by systematically varying the additive type and concentration. We note that all the protocols proposed earlier [26, 30, 40] and in the present work effectively tune the bulk elasticity (G_L') of clay, the primary control parameter that determines interfacial morphology. Supplementary section ST3 presents transitions between DVP to DP patterns, achieved by increasing clay concentration.

3.3. Finger propagation profile during miscible displacement of pure clay

We followed individual fingers to identify local propagation modes across a broad range of clay elasticities. Figures 4(a1)–(a4) show the sequential evolutions of finger propagation profiles, with yellow and pink dashed lines highlighting the propagation of two neighbouring fingers, when a clay sample of concentration $C_L = 3\%$ w/v and elasticity $G_L' = 572.2$ Pa, prepared in pure water, was displaced by dyed water. These images were cropped during the growth of the pattern shown in figure 3(c), and the movie from which these snapshots were extracted is provided as supplementary movie 1. The interface between the water and the clay is poorly defined due to fluid mixing. We observed finger shielding (figures 4(a1)–(a3)), with one advancing finger retarding the growth of a neighbouring finger. Additionally, as displayed in figure 4(a2), the fingers underwent consecutive TS events, with the

fastest propagating fingers splitting at their tips to form two distinct fingers. We also identified finger coalescence events (figure 4(a4)), wherein the tip of the trailing finger bent and merged with a neighbouring finger. Multiple coalescence events eventually led to the detachment of small portions of the displaced clay from the main body, a phenomenon known as TLD [50–52], marked in figure 4(a4). Coalescence and the formation of trailing lobes are intricately linked to each other. It is well known that a propagating finger that is shielded by a neighbouring finger witnesses a loss of momentum, merges with the latter and results in the formation of trailing lobes that eventually detach from the main body. We observed that the trailing lobes constituted by the displaced clay diffuse into the displacing fluid over time.

Non-linear finger interactions such as shielding, TS, coalescence, and TLD have been widely reported in simulations of Newtonian fluid pairs characterised by a high mobility ratio [50], experiments involving the displacement of glycerol by water [53] and during the displacements of non-Newtonian fluids in confined geometries [54]. We note from our experiments that when an aqueous clay without any salts/additives is displaced by water, the growth of interfacial patterns is governed by the Saffman-Taylor instability [3] and is driven by the viscosity contrast between clay and water.

3.4. Finger propagation profiles during miscible displacements of clay samples of low elasticities

As already outlined in table 1, the addition of TSPP and DMF in the aqueous medium while preparing clay suspensions delays clay aging by retarding the self-assembly of clay particles. Figures 4(b1)–(b4) and (c1)–(c4) illustrate the sequential evolutions of finger propagation profiles for clay samples of concentration $C_L = 3\%$ w/v prepared in 2 mM TSPP and 260 mM DMF solutions respectively. The movies from which these snapshots were extracted are provided as supplementary movies 2 and 3, respectively. Inspection of figure 2(c) shows that the displaced clay in these experiments were characterised by low plateau elastic moduli, $2.0 \leq G_L' \leq 55.4$ Pa.

Images of interfacial instabilities with clay samples containing different concentrations of TSPP and DMF are provided in figure S3 of the supplementary information. The global morphology of the interfacial patterns observed in the presence of TSPP and DMF can be categorised as a DVP. Figures 4(b1)–(b4) display magnified views of the propagating fingers with 2 mM TSPP and reveal that the fingers propagate in a snake-like zig-zag manner, as shown by pink dashed lines, with relatively fewer TS events. The finger growth profile, in this case, is reminiscent of channelling [15, 16], wherein interfacial fingers propagate through preferential flow pathways in the displaced suspension. Channelling has been reported for displacements in heterogeneous porous media in which flow occurs through regions of high permeability [15, 16].

Interestingly, the images acquired in the experiments with DMF (figures 4(c1)–(c4)) reveal a very different mode of finger propagation. In these displacement experiments, we observed that a growing finger suddenly stopped propagating radially outward. Next, the tip widened (figure 4(c2)) in a mechanism that closely resembled a skewering mechanism proposed earlier [50, 55]. Eventually, we noted that multiple new fingers grew at the broadened fingertip. These new fingers propagated in a non-linear fashion, while interacting with one another to form trailing lobes. We identified this mechanism as dense branching (DB). We note that such skewering propagation was reported in miscible and immiscible displacement experiments involving non-Newtonian fluids and in numerical simulations of miscible viscous fingering involving a Newtonian fluid pair of high mobility ratio. This finger propagation mode was reported to arise from interface-destabilising velocity fluctuations between high-mobility ratio fluid pairs [50, 55]. In contrast, we believe that the skewering mechanism in our work arises due to the presence of areas characterised by stochastic permeability heterogeneity within the displaced clay suspension. For finger propagation dominated by viscous forces, the presence of unbroken tactoids and disconnected strands of clay particles presents a heterogeneous environment to the growing finger. The resultant stochastic variations in permeability lead to the emergence of preferential flow pathways that allow the advancing fingers to propagate with high mobility. We conclude therefore that channelling flow due to the heterogeneous permeability of the displaced clay drives the observed snake-like zig-zag finger propagation profile, finger growth via skewering and the orthogonal movement of fingers. The skewering mechanism is seen to coexist with another novel finger propagation mechanism, wherein a few fingers propagate orthogonally to the flow direction for a small period of time (indicated by pink dotted line in figures 4(c1)–(c4)). We note that the zigzag and skewering propagation modes reported here were observed during the entire duration of pattern growth.

We quantified the distinct finger propagation mechanisms at the clay-water interface during the displacements of pure clay and clay prepared in DMF and TSPP solutions. The data presented in figures 4(a)–(c) was analysed to compute some standard metrics. Finger coalescence events were quantified by estimating the total number of trailing lobes and their average areas as a function of time elapsed

since the displacing fluid was injected into the HS cell. This data is plotted in supplementary information figures S8(a) and (b) and shows that the number of trailing lobes grows monotonically with time and is highest when the displaced clay was prepared in TSPP solution. We attribute this to frequent mergers of the snake-like zig-zag finger propagation profiles. Simultaneously, the average areas of the trailing lobes, plotted in figure S8(b), are seen to be highest for clay with added DMF due to the skewering of fingertips before they split and grew.

3.5. Finger propagation profiles during miscible displacements of aqueous clay samples of moderate and high elasticities

The presence of NaCl, KCl and glucose in the aqueous medium enhances the elasticity of the clay suspension by accelerating its physical aging via the rapid self-assembly of clay particles. Figures 4(d1)–(d4) and (e1)–(e4) display the evolution of interfacial fingers when highly elastic clays, prepared by adding NaCl of concentrations $C_A = 1$ mM and $C_A = 2$ mM to the suspension medium, respectively, were displaced by injecting water in the HS cell. The movies from which these snapshots were extracted are provided as supplementary movies 4 and 5, respectively. For these displacement experiments, the fluid-suspension interface is comparatively sharp, the spacings between the neighbouring fingers increased significantly when compared to DVPs, and no coalescence events were noted.

For the clay samples with $C_A = 1$ mM NaCl, we see from figures 4(d1)–(d4) that finger propagation is dominated by side branching, though occasional TS events are also noted. Side branching, which refers to the development of smaller, secondary fingers behind the tip of the main finger, was reported when a shear-thinning fluid was displaced by a Newtonian fluid [22]. As in earlier reports [5, 22, 56], we note that the displacement of shear-thinning clay samples results in large flow anisotropy in our experiments, with the tip of the propagating finger experiencing lower viscosity along the flow direction when compared to the lateral direction. This flow anisotropy couples with the broadening of the finger due to the applied injection pressure and results in the shedding of multiple side branches. We therefore conclude that when the displaced clay is characterised by moderately high plateau elastic moduli ranging between $800 \text{ Pa} \leq G_L' \leq 950 \text{ Pa}$ (figure 2(c)), achieved by the introduction of additives such as NaCl, KCl and glucose in the suspension medium to enhance clay aging, pattern growth is dominated by significant side branching and occasional TS events.

When a clay sample containing the highest concentration of polar salt, 2 mM NaCl, was displaced by dyed water (figures 4(e1)–(e4)), we observed comparatively thicker interfacial fingers with pointed tips. These fingers displayed side branching with branching angles close to 90° , as marked in figures 4(e1)–(e4). Such orthogonal branching, a typical characteristic of fracture propagation, is noted by us only in those displacement experiments involving clay samples of elastic moduli $G_L' > 10^3 \text{ Pa}$. Our results are in agreement with an earlier report on the displacement of Bentonite clay [26], wherein the emergence of fractures was noted for Deborah numbers $De \gg 1$, which is indicative of the viscoelastic solid-like nature of the displaced clay sample. For samples prepared with very high amounts of polar additives like NaCl and KCl, large elastic stresses are expected to develop below the clay suspension yielding point of the sample. The observed fractures emerge due to the rapid release of these elastic stresses. Since the linear elastic moduli of samples with glucose are much smaller in comparison to clays with added polar salts (figure 2(c)), we did not observe any fracture patterns in these samples (supplementary figures S4(a) and (b)). We note that it may still be possible to generate fracture patterns in the presence of glucose by allowing the clay to age for a longer time or by enhancing the glucose content in the suspension medium. An inspection of figure S4(b), which displays the interfacial pattern generated during the displacement of a clay sample with a large glucose content, reveals an instability that closely resembles a fracture pattern.

3.6. Characterisation of finger propagation modes at clay-water interfaces

To quantitatively distinguish between the finger propagation mechanisms uncovered in our experiments, we introduced two metrics to characterise the local propagation modes: (i) finger-path tortuosity to characterise the waviness of the finger path, and (ii) finger-width distribution as an indirect measure of heterogeneous permeability of the displaced clay suspension. Tortuosity was computed as the ratio of the cumulative arc-length of the fingertip path to the radial distance from base to tip [57]. Finger width was calculated at all times by measuring the lateral distance across the finger near its tip along a line perpendicular to the growth direction. Tortuosity and width measurements, computed from seven distinct fingers, are displayed in figures 5(a), (b) respectively. Fingers propagating by zig-zag and skewering modes exhibit the highest tortuosity, which indicates that their paths are the most convoluted (figure 5(a)). The violin plots in figure 5(b) show that these two mechanisms produce fingers with greater mean widths

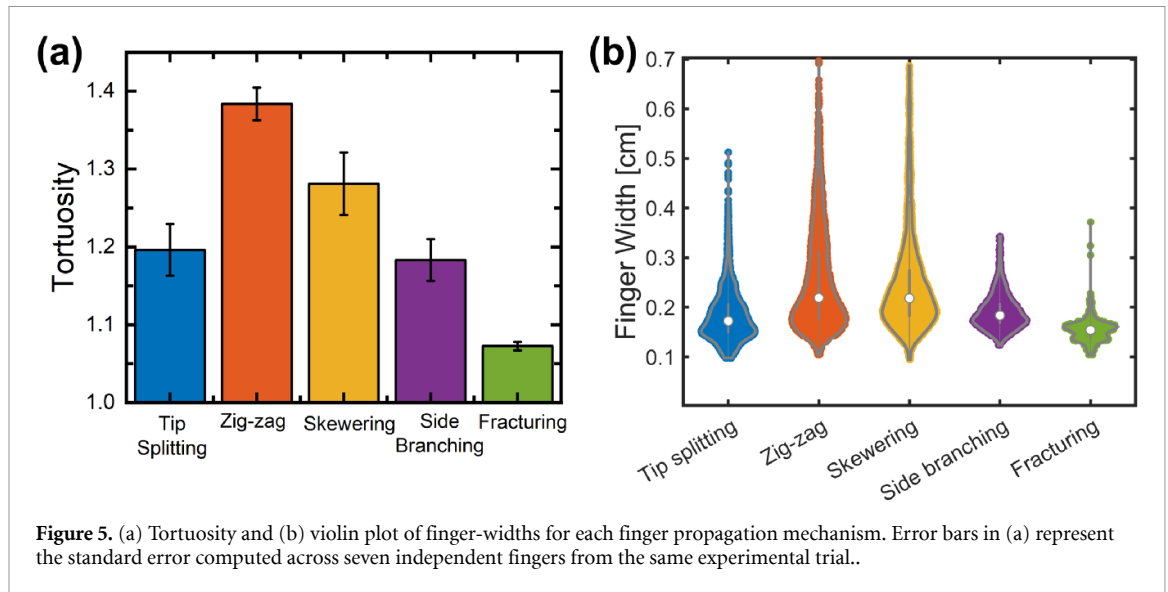


Figure 5. (a) Tortuosity and (b) violin plot of finger-widths for each finger propagation mechanism. Error bars in (a) represent the standard error computed across seven independent fingers from the same experimental trial..

Table 2. Summary of additives added to the clay suspension medium, elasticities of the clay suspensions, finger propagation modes, and the observed interfacial patterns..

Additive name	Additive concentration	Elasticity [Pa]	Mode of finger propagation	Global interfacial pattern
TSPP	2 mM	2	Zig-zag	Dense viscous pattern (DVP)
	1 mM	11.4	Zig-zag	Dense viscous pattern (DVP)
DMF	260 mM	28.2	Skewering	Dense viscous pattern (DVP)
	130 mM	55.5	Skewering	Dense viscous pattern (DVP)
No additive	0 mM	572.2	Tip-splitting	Dense viscous pattern (DVP)
Glucose	130 mM	863.1	side branching	Dendritic pattern (DP)
	260 mM	910.4	Side branching	Dendritic pattern (DP)
NaCl	1 mM	920.9	Side branching	Dendritic pattern (DP)
	2 mM	1089.4	Fracturing	Viscoelastic fractures (VEF)
KCl	1 mM	933.6	Side branching	Dendritic pattern (DP)
	2 mM	1308.6	Fracturing	Viscoelastic fractures (VEF)

and broader width distributions, which suggests the inherent stochasticity of propagation through heterogeneous permeability fields. This is in agreement with a study of nonlinear viscous fingering in heterogeneous media [15]. Patterns that grow via tip splitting and side branching yield intermediate tortuosity values due to their branched morphologies (figure 5(a)). These patterns are characterised by narrower finger width distributions and lower median widths, as illustrated in figure 5(b). Since fracturing produces fingers that are straight and the least branched, these fingers display the smallest tortuosity (figure 5(a)), the narrowest width distribution and the smallest median widths (figure 5(b)). We note that these new metrics quantitatively distinguish between novel propagation modes like zig-zag and skewering growth and previously reported mechanisms such as TS, side branching and fracturing. While these parameters alone cannot distinguish between zig-zag and skewering modes, they are useful in classifying the propagation modes based on finger trajectories and morphologies.

The interfacial patterns reported in the present work and the corresponding finger propagation modes are tabulated in table 2. Clay suspensions are very complex thixotropic materials. In the present work, we demonstrated that a rich array of patterns, *viz.* TS, skewering, channelling, zig-zag finger propagation, side branching, and fracturing, can be generated by the miscible displacement of 3%w/v clay suspensions prepared in aqueous additive solutions. The novelty of our experiments lies in the several distinct finger propagation modes that we uncovered by displacing clay samples of a fixed particle concentration, the physical aging behaviour of which we tuned by incorporating common additives in the suspension medium. A substantial body of theoretical and computational studies on the viscous fingering instability is available in the literature [14, 18–20, 31, 32]. However, given the complexity of

clay rheology, we do not believe that a stand-alone theory can explain all the different interfacial pattern morphologies and local propagation modes reported in this work.

4. Conclusions

The incorporation of additives to the aqueous medium during the preparation of clay suspensions allowed us to sensitively control the aging dynamics and, therefore, the evolution of the bulk elastic moduli of the samples [42]. We have demonstrated here that clay elastic modulus and permeability heterogeneity strongly influence interfacial pattern growth during the confined miscible displacements of clay samples in a radial HS geometry. We established that modifications in inter-clay interactions and disruption of hydrogen bonds in the aqueous suspension medium play crucial roles in determining local finger propagation profiles and global pattern morphologies.

When the elasticity of the displaced clay suspension was small, our study revealed novel local modes of finger propagation, such as zig-zag propagation and skewering, during the growth of DVPs. We characterised the images using single-finger-scale metrics like finger-path tortuosity and finger-width distributions. We concluded that these novel propagation modes arise from the coupling of interfacial viscous instabilities with heterogeneities in the permeability of the displaced clay. While zig-zag finger propagation was predicted theoretically in heterogeneous porous media [15], we report this propagation mechanism in a colloidal suspension for the first time. Skewering patterns have been previously reported during the displacement of Newtonian and polymeric fluids [50, 55]. We demonstrated here that skewers (figure 3(c)) can also form due to stochastic heterogeneity in the permeability of the displaced aqueous colloidal clay. Finally, increasing clay elasticity by the incorporation of polar additives such as NaCl, KCl and nonpolar glucose resulted in a smooth transition from dendritic to fracture patterns. These patterns were characterised by lower values of tortuosities, finger widths and finger width distributions.

While multiple theoretical frameworks explain the individual observations reported here, we note that the complexity of pattern formation in aging clay suspensions precludes the use of a single theory to capture all the propagation modes uncovered in our study. The direct quantification of local permeability fields remains a subject for future work. Since the clay samples did not age substantially during the displacement experiments reported here, the stochastic permeability heterogeneity profile was assumed to be stationary as water invaded clay. We note that if the injection speed of the displacing water is reduced substantially, such that the clay sample ages during the displacement experiment, an even wider range of novel pattern morphologies and pattern growth mechanisms can be obtained. In future experiments, it would be interesting to control the heterogeneous permeability resulting from the unbroken clay tactoids and disconnected strands of clay particles. The development of clay elasticity and permeability heterogeneity on the one hand, and the finger propagation profiles during displacement experiments on the other, must be correlated. It would be interesting to study these correlations in systematic experiments. An even larger array of interfacial patterns can be generated simply by tuning the miscibility of the fluid pair. We conclude by noting that the potential to control interfacial displacement patterns can have far-reaching implications in materials processing applications. The suppression of instabilities during the transport of clay slurries, for example, can maximise transport efficiency while reducing energy costs.

Data availability statement

The data that support the findings of this study will be openly available following an embargo at the following URL/DOI: [10.5281/zenodo.17062524](https://doi.org/10.5281/zenodo.17062524) [58]. Data will be available from 2026 March 05.

Supplementary Material available at <https://doi.org/10.1088/1367-2630/ae27ec/data1>.

Finger growth in pure clay available at <https://doi.org/10.1088/1367-2630/ae27ec/data2>.

Zig-zag finger propagation available at <https://doi.org/10.1088/1367-2630/ae27ec/data3>.

Skewering fingers available at <https://doi.org/10.1088/1367-2630/ae27ec/data4>.

Side-branching in clay available at <https://doi.org/10.1088/1367-2630/ae27ec/data5>.

Viscoelastic fractures available at <https://doi.org/10.1088/1367-2630/ae27ec/data6>.

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