PRF#59026-DNI9; Connecting Pore-Scale Dynamics and Oil Mobilization by Polymers in 3D Porous Media PI: Prof. Sujit S. Datta, *Chemical and Biological Engineering, Princeton University*

Polymer solutions are frequently used in enhanced oil recovery (EOR) to improve the recovery of trapped oil. However, applications are limited by an incomplete understanding of the flow in porous media. The tortuous pore structure imposes both shear and extension, which elongates polymers; moreover, the flow is often at large Weissenberg numbers, Wi, at which polymer elasticity in turn strongly alters the flow. This dynamic elongation can even produce flow instabilities with strong spatial and temporal fluctuations despite the low Reynolds number, Re. Unfortunately, macroscopic approaches are limited in their ability to characterize the pore-scale flow. This project seeks to address this gap in knowledge using imaging in microfluidic porous media. Specifically, our Aims are to:

- (1) Correlate spatial and temporal fluctuations in polymer solution flow to pore-scale geometry, and
- (2) Directly visualize how the flow of polymer solution dynamically changes the 3D conformations of trapped oil ganglia themselves.

The proposed work will thus provide new insights into the use of polymers for EOR by linking polymer properties, flow parameters, structural properties of porous media, and overall oil recovery.

Support from the ACS PRF DNI grant has been instrumental to realizing this work, which previously represented a completely new direction for the PI— PI's prior work was restricted to the study of Newtonian fluids, whereas this work studies polymer solutions and complex instabilities in their flow. In the past year, we have made considerable progress towards both Aims, particularly Aim 1. As described below, we have successfully developed new experimental tools to visualize polymer solution flow in 2D and 3D microfluidic porous media, at pore-scale resolution, and have already discovered new phenomena relating to how spatial and temporal fluctuations in polymer solution flow are correlated to pore geometry. This work, which has been led by a new graduate student in the PI's lab, with assistance from a postdoctoral researcher, has so far resulted in one paper that is currently under review in a leading fluid mechanics journal, and one review article on polymer solution flow in porous media that has been accepted in the leading technical journal *Small*.

To elucidate how the spatial and temporal characteristics of polymer solution flow depend on pore geometry, we developed a way to microfabricate model porous media using 3D printing. The media are made of 1D arrays of pore throats (Figure 1a), enabling us to directly test the relative importance of polymer memory on the flow by varying the spacing between pore throats. When the pore spacing is large, we find that unstable eddies form upstream of each pore throat, similar to observations of an isolated pore. By contrast, when the pore spacing is sufficiently small, we find that the flow exhibits a surprising bistability—which to our knowledge has never been reported for flow through porous media before. In each pore, the flow persists over long durations in one of two distinct flow states: an eddy-dominated state in which a pair of large unstable eddies forms in the corners of the pore body, and an eddy-free state in which strongly-fluctuating fluid pathlines fill the entire pore body and eddies do not form (Fig. 1b).

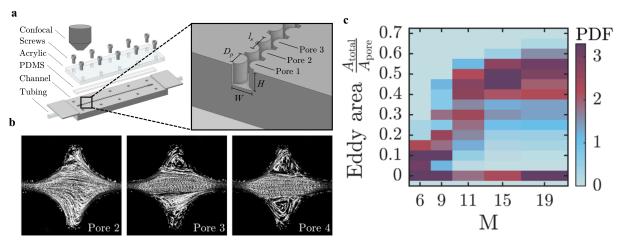


Figure 1. (a) Experimental setup with W=2 mm, H=2 mm, $D_p=1.6$ mm, $I_s=W$. The setup is inverted and videos are captured on a confocal microscope; left panel is vertically flipped for clarity. (b) Pathline images of polymer solution flow through a porous medium with 30 pore throats at M=11.4, where M is a non-dimensional parameter that increases with flow rate as first defined by Pakdel and Mckinley (1996). Pore 4 exemplifies the eddy-dominated state, with eddies that continually fluctuate both internally and at their boundaries, while pore 2 exemplifies the eddy-free state, with strongly-fluctuating pathlines that fill the entire pore body. (c) Probability density functions of measured eddy sizes in the different pores. The PDFs for M>9 are bimodal, showing two characteristic eddy areas, indicating a bistability in unstable flow states.

This bistability can be quantified by the PDFs of the time-averaged measurements of A_{total} , which describes the total eddy area measured in each pore combined for all pores in the medium. We parameterize the flow by the M parameter $M = (Wi \cdot De)^{1/2}$ as is conventionally done for unstable polymer solution flow; Wi is the Weissenberg number and De is the Deborah number. Below $M \approx 9$, $A_{\text{total}} \approx 10\%$ of the pore area A_{pore} , and eddies do not change in time; by contrast, above the onset of the flow instability at $M \approx 9$, we find that the PDFs become bimodal, reflecting the bistability in flow behavior (Fig. 1c). The eddy-dominated state is represented by the upper branch of the PDFs, in which A_{total} increases with M, eventually plateauing at $\approx 60\%$ of A_{pore} at the highest values of M tested. The eddy-free state is represented by the lower branch of the PDFs, in which $A_{\text{total}} \approx 0$ over all M. Importantly, this bistability does not arise when we test porous media with wider pore spacings: specifically, the lower branch of the PDFs does not appear. Thus, when the spacing between pore throats is sufficiently small—and thus, elongation of individual

polymers is more likely to persist across multiple pores—the flow abruptly becomes bistable, exhibiting two coexisting unstable flow states.

We therefore hypothesize that this unusual behavior arises from the interplay between flow-induced polymer elongation, which promotes eddy formation, and relaxation of polymers as they are advected between pores, which enables the eddy-free state to form. Consistent with this idea, we find that the flow state in a given pore persists for long times. In addition, we find that the instantaneous flow state is correlated between neighboring pores; however, these correlations do not persist long-range. By quantitatively characterizing this intriguing bistability in flow behaviors, our results thus help to elucidate the rich array of behaviors that can arise for polymer solution flow through porous media. In follow up work, we have been testing how ganglia of non-aqueous trapped within the pore space can be mobilized by the unstable polymer solution flow. In our preliminary work, we have found that the conformation of the ganglion interface demonstrates strong fluctuations that are correlated with spatial and temporal fluctuations in the surrounding polymer solution flow (Fig. 2). We hypothesize that these fluctuations can enable a trapped oil ganglion to be mobilized from a porous medium; we are currently testing this hypothesis using this experimental platform.

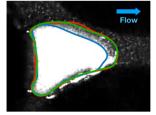


Figure Non-aqueous ganglion (white region) trapped within a pore body shows strong interfacial fluctuations when the surrounding polymer solution (seeded with tracer particles, shown as white dots) is unstable. Colors show interface at different times.

We have also succeeded in extending our flow visualization to disordered 3D porous media, which are inherently more complex but also provide a closer mimic to real-world reservoir rocks. Specifically, we have made model 3D media by lightly sintering densely-packed glass beads within square quartz capillaries. Importantly, we have formulated high-molecular weight polymer solutions that have the same refractive index as the glass; the opaque medium then becomes transparent, and the pore-scale flow can be visualized using confocal microscopy of fluorescent tracer particles. Using this approach, we have directly visualized unstable polymer solution flow within 3D porous media at pore scale resolution—which to our knowledge has never been done before. Consistent with our 2D measurements, we observe bistable flow, with pores switching between a state with unstable eddies (Fig. 3a) and a state with no eddies at all (Fig. 3b). These achievements now uniquely position us to be able quantify the spatial and temporal characteristics of the pore-scale flow (Fig. 3c-d) throughout the entire medium (Fig. 3e), and ultimately, link unstable polymer flow to mobilization of oil from 3D porous media.

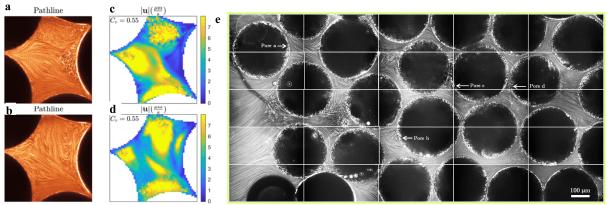


Figure 3. (a,b) Pathline images of pore-scale flow in a 3D medium at two different times, showing flow fluctuations. Particle Image Velocimetry of tracer particle trajectories enables us to characterize the flow field; examples of the instantaneous flow speeds corresponding to (a,b) are shown in (c,d). (e) Combined map of the unstable flow of a polymer solution in a 3D medium over a larger region, showing the flow in multiple pores. Image is created by combining pathline images across multiple pores, showing the strong spatial and temporal fluctuations in the flow under these conditions.