PRF# 57496-DNI9

Understanding Transport of Soft Units through Porous Media by Correlating Pore-scale Dynamics and Macroscale Properties

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The goal of the project supported by this ACS PRF DNI grant is to understand the pore-scale dynamics of the transport of soft units, such as microgel particles and drops, through porous media and to correlate this pore-scale information with changes in macroscale permeability that result from the presence of soft units in the media. We proposed multi-scale experiments and mechanistic analysis to address our research objectives.

Microgel fabrication and characterization

After training and practice, the graduate students who are supported by this grant have mastered the capillary-based microfluidic technology to fabricate hydrogel micro-spheres, or microgels, with a variety of materials, such as polyethylene glycol diacrylate (PEGDA), polyacrylamide (PAM), and gelatin methacryloyl (GelMA). At this stage we've been focusing on PAM microgels with monomer concentrations of 10% and the monomer-crosslinker mole ratio of 37.5:1 for the subsequent flow experiments in this project. The inner phase in microfluidic fabrication is aqueous solution containing acrylamide, bis-acrylamide, and ammonium persulfate. Outer phase is hexadecane with Span 80 and EM 90 as surfactants. Drops are collected in hexadecane with surfactant and TEMED to crosslink and form the microgels. We fabricated microgels with various sizes ranging from 150 µm to 250 µm, to fit different sized channels and to realize different gel-pore size ratio in the subsequent experiments. The resultant monodisperse PAM microgels is shown by the micrograph in **Figure 1**. We measured the Young's modulus of the microgels using low-frequency tapping mode with atomic force microscope (AFM). The measured Young's modulus of our 10% PAM microgels is around 10 kPa.

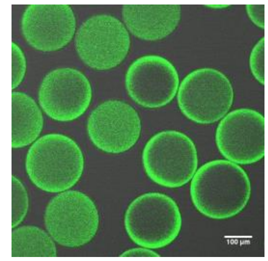

Figure 1. Confocal micrograph of PAM microgels.

Examine flow dynamics of microgels through a constrictive capillary

We examine the flow of microgels through a constrictive capillary (**Figure 2a**) to obtain the constitutive relationship between the drop in pressure due to gel blocking and changes in flow conditions. The constrictive capillary is used to mimic a single-pore geometry in porous media. A gel blocked is exposed to hydrodynamic pressure force driving the gel to pass through, compression force (normal stress) from the channel wall resisting the gel to move, and friction force from the wall resisting the gel to move. By balancing the forces on the axial direction, we can derive the minimum pressure that can push the gel through the constriction. To do so we need the information of friction coefficient between the wall and the gel. We thus study the microgel passing a straight channel to obtain the friction coefficient (**Figure 2b**).

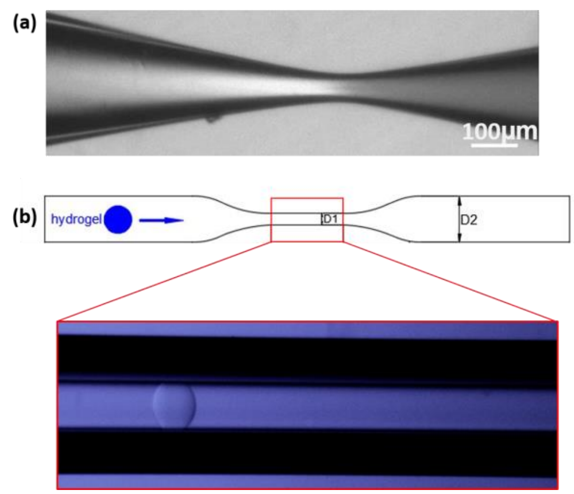

Figure 2. (a) Micrograph of constrictive channel; (b) schematic and micrograph of constrictive channel with a straight section.

When the gel moves in a circular straight channel at a constant velocity, the friction from the wall balances the hydrostatic pressure. We thus derive the friction coefficient by measuring the pressure at constant gel moving speed. **Figure 3** demonstrates two typical measurements in terms of pressure and velocity. We adjust the flow rate setting of the syringe pump to realize constant velocity of gel, as indicated by the orange dots. The radial stress of confined gel can be estimated by

$$\sigma_r = K\,(2\varepsilon_r + \varepsilon_z) + \frac{2}{3}G\,(\varepsilon_r - \varepsilon_z) \qquad (1)$$

where ε_r and ε_z are radial and axial strain measured from the image. *K* and *G* are bulk and shear modulus of the microgel. Based on the Young's modulus measured from AFM, and the typical negative Poisson ratio of hydrogel, we can derive both *K* and *G*, and subsequently σ_r. The friction coefficient *f* is then estimated by balancing pressure force and friction force $F=f\cdot\sigma_r\cdot A$, where *A* is the contacting area. The estimated friction coefficient is ~14. This is an apparent friction coefficient that also considers the friction between water and the porous polymer network.

We also simulate the process of gel passing through a constrictive channel with COMSOL Multiphysics. The same physical, geometric, and flow parameters are used in the simulation. The channel geometry and a few snapshots of the gel passage process are shown in **Figure 4**. The pressure data obtained from simulation agree well with our analysis and experiments, which confirm that our mechanistic analysis is reliable.

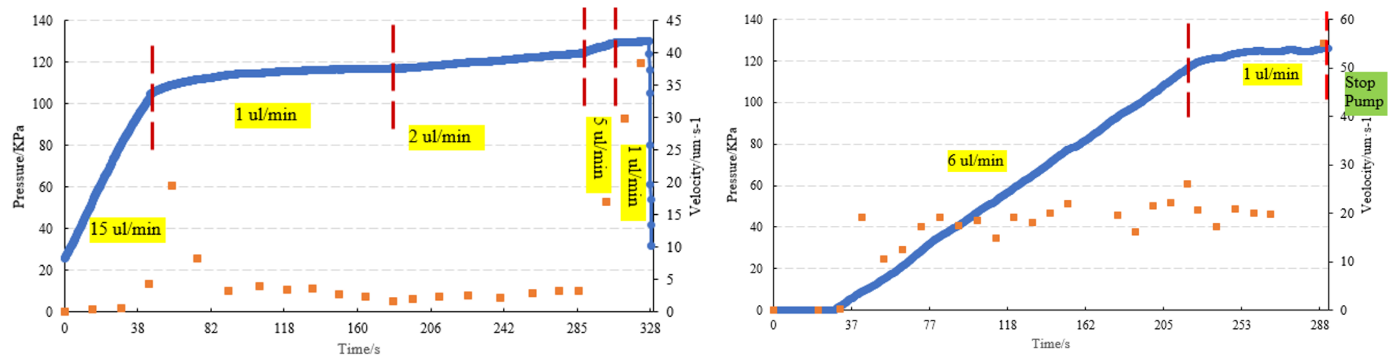

Figure 3. Variation of pressure and velocity for single microgel moving through a straight channel.

Research plan for the 2nd year

In the second year of the project, we will conduct experiments, analysis, and simulation in the constrictive channel as shown in Figure 2(a). In the analysis we will use the friction coefficient that we obtained from our study on the straight channel case. Through this systematic study we expect to get the correlation between the pressure and the flow, geometric, and material properties at the pore scale. In the next step, we will develop a scheme to extend this pore-scale information to porous medium, and estimate the permeability change of the porous medium due to the presence of microgels in the medium. We will also conduct experiments in micromodels to verify the effectiveness of our model. The resultant findings would benefit a variety of enhanced oil recovery processes such as gel treatment and emulsion flooding.

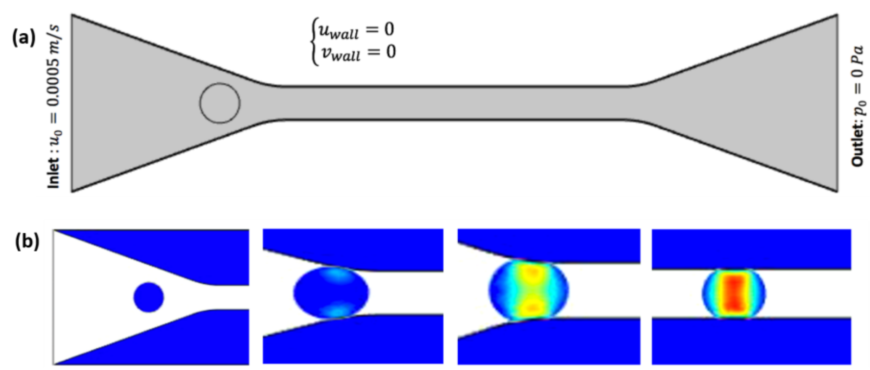

Figure 4. (a) Geometry of the microfluidic constriction channel and boundary conditions in the simulation by COMSOL; (b) a few snapshots of the gel passage process with the color map indicating the stress distribution.

Career and student training impacts

This grant has had substantial positive impact on the careers of the PI and participating students. One PhD student, Shuaijun Li, and one master student, Hung-Ta Chien, have been financially supported by this grant. Hung-Ta Chien just graduated from CCNY and is currently a PhD student at Texas A&M. Shuaijun Li will continue his study on this project in the next year. The two students presented their preliminary findings at the 91st ACS Colloid & Surface Science Symposium held at CCNY. Another PhD student of the PI and 4 summer intern students, including 1 visiting student from Polytech Montpellier at France, have also contributed to the project, both in simulation and experiments, although they were supported by other funding sources. In addition, we have published a review paper on microfluidic model porous media (micromodels) in *Small*. This is not only the first comprehensive review on this topic and therefore will benefit researchers in a wide range of areas, but also enables the contributing students to develop a deep understanding on micromodels, which have positive impact on their subsequent study for this project using micromodels.

We will soon present our current findings at IMECE 2019, and in the process of preparing a manuscript as journal publication. We expect to have at least two journal publications regarding our research findings by the end of the 2nd year. In the meantime the PI will prepare a proposal to seek for grant funding from NSF.