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Size dependence of Nano-/Microparticles flowing through fracture structures

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ABSTRACT

Nano- and microparticle tracers are expected to be used for evaluating structures in geological developments, however their transport behavior is not well characterized. This study investigated nanoparticle transport in a fractured medium through a microscope. A single fracture and rock matrix (grain and pore space) were fabricated on a silicon wafer, which is called a micromodel. Water and particles were injected into the micromodel, and the temporal change of particle concentration was measured by Tunable Resistive Pulse Sensing (TRPS). When the particles flowed through the matrix, no size dependence was observed. In contrast, when a fracture existed in the matrix, the larger particles were observed only at early time, while the smaller particles were detected over a wider range of time. This indicates that particles with different sizes transport through fractured media differently. In the cases where preferential paths exist, particles tracers may be able to suggest the existence of preferential paths.

1. INTRODUCTION

Understanding mechanisms of fluid flow in fractured porous media is essential for geoscientific research. Because the fluid flow in fractured rocks is controlled by complex fractures, the fracture structures need to be characterized. Tracer testing has been conducted to evaluate flow properties, such as connectivity, flow velocity, and dispersivity. However, spatial heterogeneities of fracture distributions cannot be obtained from the response of conventional solute tracers. Fluid with nano- and microsized particles have been investigated in several fields (e.g., medicine, optics, and electronics) (Taylor et al., 2013). Geological development also expects to use nano- and microparticles as tracers, which may provide more effective information for reservoir characterization and improve the efficiency of energy resources development (Burtman et al., 2015).

Nano- and microparticles embedding specific materials may provide new methodologies to estimate spatial heterogeneities of fracture distributions. Burtman et al. (2015) conducted the measurements of electrical conductivity of brine with nanoparticles (Fe_3O_4, Fe_2O_3, NiO, and Al_2O_3) and showed the possibility to observe significant difference of conductivity with nanoparticles in rocks. Angayarkanni and Philip (2014) showed a 4366 % increase in electrical conductivity for SiO_2 water-based nanofluids due to the effect of the double electric layer surrounding each particle and particle size effect. If we use highly conductive nanofluid for Electrical Resistivity Tomography (ERT), time histories of three-dimensional spatial distributions of tracer migration in fracture networks may be obtained.

A DNA (deoxyribonucleic acid)-based nanotracer can be produced by attaching synthetic DNA molecules to the surface of a silica nanoparticle seed and adding a protective outer silica layer. DNA protected by silica nanoparticles has been proven to withstand temperature as high as 200°C (Paunescu et al., 2013), which potentially makes it applicable to high temperature regions in geothermal fields. A DNA molecule is composed of four types of nucleotides, and the different arrangements of nucleotides that compose a long DNA double helix result in DNA molecules with almost infinite possible sequences. The infinite number of DNA sequences leads to an unlimited number of uniquely identifiable nanoparticle tracers which could be applied to multiple wells or thousands of flow paths for tagging purposes without ambiguity (Zhang et al., 2017).

The ability to control size and shape is also an advantage of using nano-/microparticles as tracers. Most geothermal reservoirs are thought to have fluid passing through fractures with apertures of a few nano- and micrometers. If particles with different sizes are injected, only particles smaller than the largest aperture in the flow path are able to pass. Thus, it can be estimated that the size of the observed largest particle is the minimum aperture in the flow path. Nishiyama and Yokoyama (2017) investigated pore sizes and flow properties in porous rocks and found that the narrowest pore throat diameter has an influence on the bulk flow velocity through the flow path. They introduced a critical pore radius corresponding to a radius of the largest sphere that can freely pass through the porous medium. We aim at using nano- and microparticle tracers to identify the minimum fracture aperture in a flow path, which is called the critical aperture, and to evaluate their flow characteristics.

As mentioned, particle tracers embedded with specific material or fabricated with specific size may provide new methodologies to estimate spatial heterogeneities. The use of nanoparticles as tracers is expected to improve the effectiveness of subsurface evaluation, by allowing more accurate, more precise, and better controlled measurements from tracer responses. Some experimental investigations have been conducted to assess nanoparticle mobility in water-saturated glass-beads packs or sandpacks (e.g., Zhang et al., 2015). The transport of nano-/microparticles shows different behaviors from conventional solute tracers. However, the cause is not understood clearly yet.

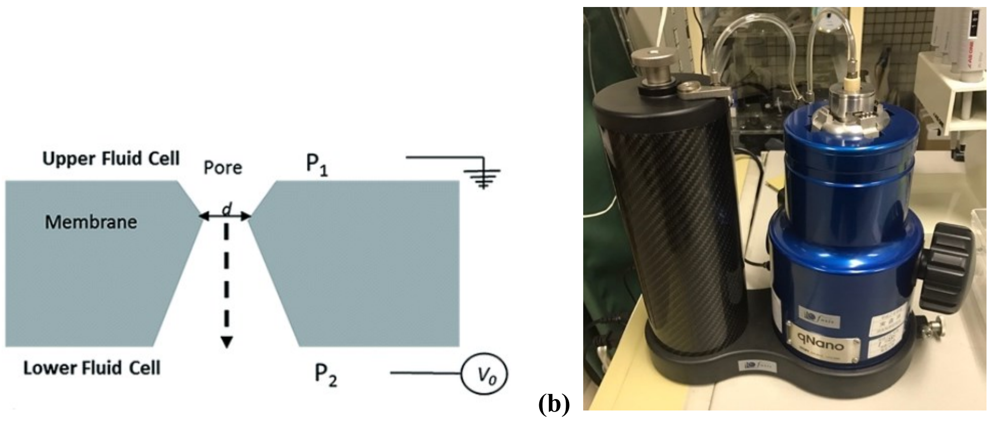
We aimed at developing a method to evaluate rock structures in fractured porous media. Alaskar et al. (2013) created a micromodel including a single fracture and matrix patterns (grain and pore spaces) obtained from real rocks. Suzuki et al. (2020) used the Alaskar et al. (2013)'s micromodel to visualize particle transport in fractured porous media. The tracer response curve was analyzed by SEM imaging and Tunable Resistive Pulse Sensing (TRPS). In the case of concentration measurement for counting the number of particles using TRPS, they succeeded in capturing the size information. In this study, we extended their experiments to further our understanding by flowing particles of different sizes into different structures.

2. METHODOLOGY

2.1 Nanoparticles

Suzuki et al. (2019) used fluorescent silica particles. The density of the particles was approximately 2.0 g/cm3. To avoid the effect of different density between water and particles, we changed the material of particle. In addition, only one size of particle (500nm) was used in Suzuki et al. (2019). We used 500 nm red and 3 µm blue polystyrene particles (micromod Partikeltechnologie GmbH Inc., Rostock, Germany). The density of both particles was 1.03 g/cm3. The original concentration of the 500nm polystyrene was 3.7×1011 particles/ml, and the original concentration of 3 µm polystyrene was 1.7×109 particles/ml. Both of particles were stable in aqueous buffers.

The particle size was measured by the qNano (Izon Science Inc.) using Tunable Resistive Pulse Sensing (TRPS) (Platt et al., 2012). A schematic of the TRPS system and an image of the qNano are shown in Figure 1. The qNano consists of a tunable pore sandwiched between the upper and lower fluid cells. At the beginning, the cells were filled with conductive liquid. A voltage was applied between two electrodes on the upper and lower cells, which controls the flow of electrical current through the tunable pore. When individual particles pass through the pore, they briefly increase the electrical resistance and create a resistive pulse, which is precisely proportional to particle volume. The measurement of particle size by the qNano is shown in Figure 2. The particle diameters were from 500 nm to 1500 nm, which is consistent with the observation of the SEM images.



(a) (b)

Figure 1: (a) System of Tunable Resistive Pulse Sensing (TRPS) and (b) image of qNano apparatus.

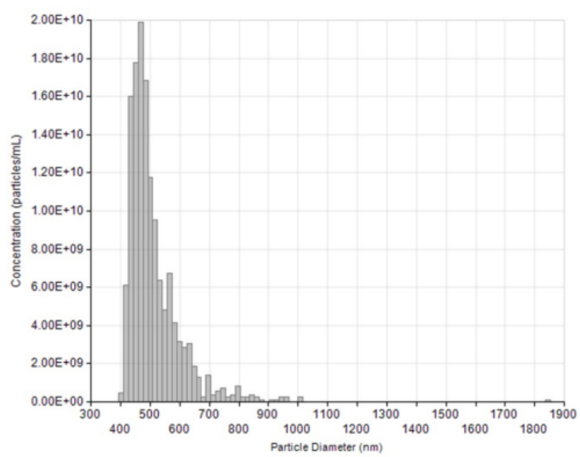


Figure 2: Frequency of polystyrene particle diameters measured by TRPS.

2.2 Micromodel

The micromodel used in this study was the same design as presented in Alaskar et al. (2013) and Suzuki et al. (2019). The model was fabricated at the Micro/Nano-Machining Research and Education Center (MNC) at Tohoku University http://www.mnc.mech.tohoku.ac.jp/index-e.html. The recipe of producing the micromodel was reconsidered.

In this study, two types of structures were used. One is a matrix structure, which is called "Matrix model". The pore network in the matrix was designed by using a thin section of real sandstone. The average pore size in the micromodel was about 9.53 µm. The grain sizes of the micromodel ranged from 3.7 to 133.2 µm, and the average was 60.9 µm. The depth of the pores and/or fracture channel was 50 µm. The porosity was calculated as the ratio of pore void area to the total area using image analysis and was found to be 31.5%. The other structure is "Fracture-Matrix model". This consisted of a single fracture of 50 µm aperture surrounded by matrix on both sides.

The micromodel was made of 4-inch silicon wafers. A schematic of the micromodel of "Fracture-Matrix model" is shown in Figure 3. The black areas in Figure 3(c)(d) (the flow paths) were dry-etched using an inductive charged plasma deep reactive ion etcher. The etched silicon wafer was covered with a transparent glass by anodic bonding. The glass had two holes, which were connected to tubing as the inlet and the outlet.

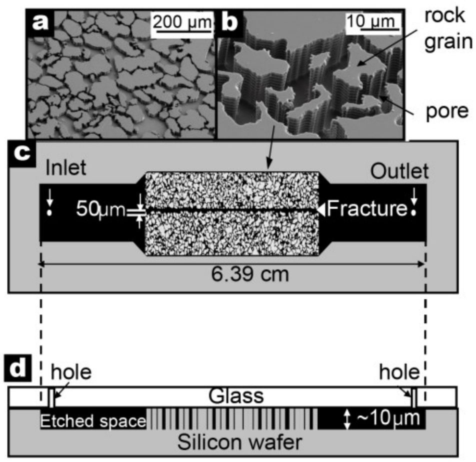


Figure 3: Schematic of the micromodel of "Fracture-Matrix model". (a)(b) SEM images of matrix in the micromodel, (c) view from the top, and (d) view from the side. The gray area is silicon wafer, and the black area is void spaces to be filled with water (the flow paths).

2.3 Flow Experiment

At the beginning of the flow experiment, Water-saturated micromodel should be prepared. The micromodels were saturated by flowing CO_2 and vacuumed pure water until full saturation was achieved. Water was injected using a syringe pump at room temperature. 1:40 dilution of 0.05 ml tracer was injected at the inlet. Then, water is injected by using a syringe pump with flow rate of 0.002 ml/min. For the "Matrix model", a mixture of 500nm and 3 µm (1:9) polystyrene particles was used, while for the "Fracture-Matrix model" a mixture of 500nm and 3 µm (1:1) polystyrene particles was used. Figure 4 shows the experimental setup for flow experiment.

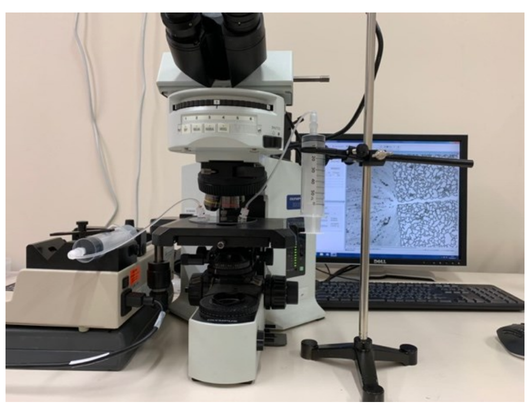


Figure 4: Experimental setup for flow experiment.

3. Results and Discussion

We sampled nine droplets at the outlet from the "Matrix model", and 21 droplets from the "Fracture-Matrix model", respectively. Each droplet was analyzed by TRPS to measure the frequency distributions of particle diameters. In the same way, nine droplets were analyzed and examined the number of particles observed at each time. Using the number of particles as the concentration, we obtained the tracer response.

Tracer responses for "Matrix model" and "Fracture-Matrix model" are shown in Figure 5. The number of particles was divided by a threshold of a certain size (1000nm, 2000nm, 2500nm). As shown in Figure 5(a)(c)(e), the tracer responses for "Matrix model" shows long tails. The long tails can be observed for both small and large particles. The same trend can also be observed by changing the threshold value from 1000nm, 2000nm, to 2500nm. This indicates that both large and small particles flow fast and slow and that the flow behavior does not change depending on the size of the particles.

In contrast, the tracer responses for "Fracture-Matrix model" show different trends as shown in Figure 5(b)(d)(f). If we set the threshold to 1000 nm, both large and small particles show long tails. When increasing the threshold to 2500 nm, although the small particles show the long tail, large particles only early response. The large particles were not observed at the late time. This indicates that the large particles only flowed fast. In the "Fracture-Matrix model", it is thought that the faster flow is formed in the fracture with larger aperture, which became a preferential flow path. Since larger particles has larger surface areas, they are more likely to be subjected to water pressure at the surface. If there is a preferential flow path, the larger particles are likely to flow through the preferential path with greater flow force than the smaller particles.

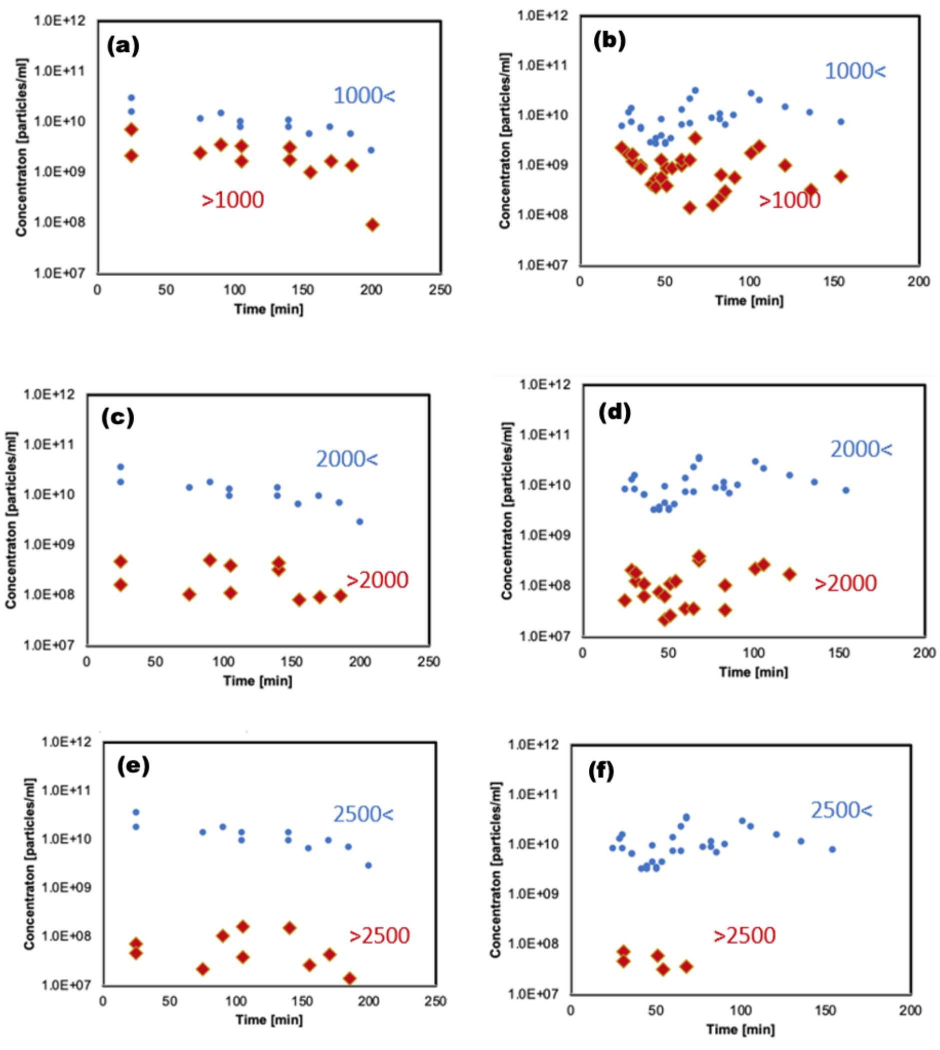


Figure 5: Tracer response for particle tracers from (a)(c)(e) "Matrix model" and from (b)(d)(f) "Fracture-Matrix model". (a)(b) Particles larger than (a)(b) 1000 nm, (c)(d) 2000 nm, and (e)(f) 2500 nm, are plotted as red diamonds. (a)(b) Particles smaller than (a)(b) 1000 nm, (c)(d) 2000 nm, and (e)(f) 2500 nm, are plotted as blue dots.

4. DISCUSSION AND CONCLUSIONS

We conducted flow experiments with micromodel to observe particle migration directly. In previous study (Suzuki et al., 2018), tracer response was obtained from the images of the SEM observation for silica particles, the response curve showed an early peak and a late tailing. In this study the same samples were analyzed by using TRPS. Only counting the number of particles did not give a tracer response as obtained from the SEM images. Instead, when the number of particles was divided into particle sizes and plotted, the response curve for larger particles showed a peak at early time. This result suggested larger particles were likely to flow through preferential paths (i.e., fracture). When changing the range of particle sizes, same behaviors could be observed in the micromodel with fracture and matrix. However, particles injected into the micromodel with matrix only, large particles tended to appear at early time and late time. This is considered there are not only one preferential path in the matrix but multiple flow paths.

Although two sizes of particles (500 nm and 3 μm) were injected at the same time, which was supposed to have a discontinuous size distribution, the size distribution analyzed at the outlet became continuous. This suggests that the particle cannot maintain their defined size by aggregating while flowing. Therefore, in future research, it will be necessary to consider aggregation of particles. In addition, according to observation of the micromodel, particles were clogged inside the matrix. This accumulation phenomenon not only makes the particle recovery difficult, but also makes the flow path blocked, which may affect reduction of the heat exchange in geothermal development. Considering the practical applications, it is necessary to develop particles that have reduced the effects of aggregation between particles and have solved the problems of accumulation and clogging in reservoirs.

In this study, we succeeded for the first time in acquiring tracer response curves according to particle sizes. In addition, by dividing by particle sizes, it was possible to discover differences in flow due to differences in particle size and in structures. These results would help to develop particle tracers and to understand flow behaviors of particles. We also found several difficulties to use nano-/microparticles as tracers. Based on these findings, we expect further research to progress.

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