Impact of Pore Morphology on Colloid Migration at Variable Saturation Levels of Natural Porous Media

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Abstract

The migration of colloids within porous media has a critical impact on many important industrial processes such as oil production and groundwater recharge. Colloids can clog the pore space and hence impair the permeability of fluids which adversely impacts the efficiency of fluids movement through such media. Therefore, understanding the mechanisms of pore clogging at the pore-scale is critical to develop efficient remediation methodologies for permeability reduction at different physio-chemical conditions. To study pore clogging at a pore-scale, microfluidic chips were fabricated to mimic geometries of natural porous media extracted from tomographic scans of sand packs. A colloidal suspension was injected in three phases into the system. The phases consisted of an initial imbibition of the suspension, followed by drainage of the suspension from the system, and finally, a second imbibition. During each phase, a series of images are taken of a section of the porous media. Findings reveal that pore-clogging considerably impairs saturation levels of the porous media through blocking the flow from reaching the gas phase within the system. Considerably increasing the time the gas is trapped in the pore-space, which in turn develops higher irreducible water saturation. This was also observed in the case of drainage of the colloidal suspension from the pore-space where colloids blocked pathways of the gas phase and prevented its migration through the pore space. In contrast, the migration of colloids was also impacted by the presence of the gas phase. Gas provided a clogging surface while forcing colloids to migrate through the pore space and accumulate at other pores. This implies that gas phase presence within a low porosity system can increase pore clogging at a significant rate. This is also supported by the short period between the clogging of two pores and the clogging of a dozen pores within the observed system.

Keywords: Colloid; Pore-scale; Pore-clogging; Saturation; Multiphase

1 Introduction

A multitude of highly impactful industries are impacted by colloid migration in porous media. These industries include CO_2 injection and sequestration (Yusof et al., 2020, 2021; Yusof et al., 2022), groundwater recharge (Akhtar et al., 2021; Reddi et al., 2000)(Akhtar et al., 2021; Reddi et al., 2000), and oil production (Khilar & Fogler, 1983; Liu et al., 2019). This is because colloids can clog the pore throats of the media by accumulating at those throats and thereby reduce the permeability of the media itself (Yusof et al., 2022). Permeability reduction as a consequence of pore clogging was also observed by (Hannun et al., 2022) and (Prempeh et al., 2020), where the latter study additionally reported non-monotonic permeability reduction relative to clay content due to the residual gas phase blocking flow paths. While (Han et al., 2020) observed pore clogging at velocities beginning from over 25 mm/s and 5 mm/s for single and two-phase flows respectively using a semi-circular cylinder to examine colloid transportation under both single and two-phase flows. The studies by Prempeh et al. and Han were not visually observed at a pore-scale, which may have yielded more information on how the mechanics of pore clogging interact with colloid migration in a multiphase system. On the other hand, (Cao et al., 2019) performed experiments using a PDMS micromodel chip with homogeneous circular grains with pore throat widths ranging from 20 to 100 µm, on clogging behaviour of sediment fines that were sieved to be under 75 µm. The study found that the presence of a gas phase along the model enhances clogging of pores irrespective of particle types or solution salinity. (Bate et al. (2022) also made use of PDMS microchips to investigate the flow of Montmorillonite and Kaolinite particles in a multiphase system. The PDMS chips were contained homogeneous micro-pillars to simulate grains in porous media. Clogging was found to occur more easily at the throats within a multiphase flow. Additionally, (Jung et al., 2018) performed pore-scale experiments using a micromodel with homogeneous circular grains under both saturated and unsaturated conditions to investigate clogging due to effect of AWI. The throat width was ranged from 50, 100, 300, and 700 µm. the colloids used in the experiment were PS particles with an average diameter of 19 µm. And from the obtained results, they concluded that bridging and clogging occurred due to accumulation of fines on the gas interface, which is then followed by depositing the colloids onto pore throats upon the gas phase passing through that throat. This differs from the conclusions by (Prempeh et al., 2020) possibly due to the differentiation between a multiphase system and residual phases. While the previous studies were visually observed at a pore-scale, the model of the microchips does not accurately simulate the heterogeneity of actual porous media as they used homogeneous micro-pillars instead of modelled grains.

The structure of the porous media itself plays an important role in colloid migration, and this was confirmed by (Li & Prigiobbe, 2018) in their numerical simulation of the migration of 4 particles within actually simulated porous media. And they found that the media structure plays a dominant role in determining colloid migration. (Rod et al., 2018) examines the impact of media heterogeneity using glass bead columns in place of actual media. Where the heterogeneity was obtained through mixing different sized beads into a column, which would be compared with a homogeneous column filled with same-sized glass beads. The study found that heterogeneity would retard transport of colloids more than homogeneity. Though the study does not allow for visual analysis on how the retardation occurred. To that end, (Kokubun et al., 2019) used a numerical model to simulate transport of colloids in both hetero and homogeneous porous media in single-phase conditions. The media itself was made up of circular pillars, where heterogeneity was achieved through changing the diameter of some of the pillars along the flow path. The results show that heterogeneous media accumulated at low and high velocities, while homogeneous media only showed accumulation for low velocities. Furthermore, the study found that accumulation at high velocity regions leads to clogs forming. This shows that heterogeneous media has a higher likelihood of pore-clogging. Other studies have examined specific features of pore throats such as the angle of convergence (Mondal et al., 2016)and the shape of the throat itself (Dersoir et al., 2015). And while studying such specifics can help in developing future equations and formulae, observing how these effects impact pore clogging on a larger scale can lead to other discoveries. One such discovery is how clogged pores can affect the clogging of other pores around them in a phenomenon called dependent clogging. Liot et al. (2018) studied this phenomenon using 10 adjacent micro-channels with a width, length, and depth of 5, 50, and 0.83 µm, respectively. And injecting polystyrene particles with a diameter of 250 nm. The results from the experiments showed an increase of clog growth rate as the number of clogged channels increased. (Liu et al., 2019) expanded on this concept using homogeneous porous media simulated through a PDMS micromodel under single-phase convergent radial flow conditions. Where dependent clogging was attributed to any clogs that occur adjacent to an already clogged pore throat. The study then concluded that dependent clogging is more effective in lowering permeability than independent clogging. Though considering adjacency alone in determining dependence can be inaccurate. This is because an important factor to consider in dependent clogging is tortuosity. (Shen & Ni, 2017) studied the relation between clogging and tortuosity using column experiments with glass beads as the media and concluded that clogging increases tortuosity and causes particles to travel in transverse paths for longer periods. In addition to that, the remaining open pores experience an increase in pressure as the number of clogged pores increases. Parvan et al. (2020) confirms Shen and Ni’s conclusion regarding how clogging increases tortuosity through numerically investigating colloid flow within models based on actual porous media. Bacchin et al. (2014) shows how this relation is reciprocal in that they found an increase in clogged pores in more tortuous paths. The conclusion reached from the examination of the previous papers is the accumulative effect of dependent clogging in terms of increased tortuosity, which in turn promotes pore clogging. Therefore this paper suggests that tortuosity can be used to examine dependent pore clogging on a scale that is not confined with adjacency.

With an increased interest in examining the reciprocal relationship between pore clogging and saturation degradation, this paper presents the following objectives:

1- Visually investigate how residual gas phases impact colloid migration and pore clogging.

2- Visually investigate how colloid-clogged pores impact desaturation on porous media.

3- Visualize the link between dependent clogging and tortuosity and flow paths.

2 Methodology

2.1 The Microchip

A hydrophobic PDMS microchip was with a surface area of **6.7x5 mm²** and having a depth of 20 µm (Micronit Micro Technologies) was etched with a geometry based on actual porous media. The geometry was extracted using synchrotron x-ray micro-computed tomographic images from sand packs. These images were taken at the Argonne National Laboratory (ANL) synchrotron facility. Using global threshold, the tomographic images were binarized. Additionally, grain boundaries were determined through the construction of line segments with a non-zero gradient value between adjacent pixels. The characteristics of the geometry used in the experiment are presented in table, with the symbols explained in table.

Table 1: Geometry characteristics

Φ	R_{Throat} (µm)	R_{Pore} (µm)	$R_{Pore/}\ R_{Throat}$	Ψ_{Grains}	θ_{Grains} (Degree)	τ	K (µm^2)
0.526	22.6	53.9	2.38	0.586	43.87	1.32	5.99

Table 2: meaning of geometry characteristics symbols

Φ: Porosity	R_{Throat}: Median Throats Radius
R_{Pore}: Median Pore Radius	$R_{Pore/}$ R_{Throat}: Pore Aspect Ratio
Ψ_{Grains}: Median Grains Sphericity	θ_{Grains}: Median Grains Orientation
τ: Tortuosity	k: Permeability

2.2 Colloid Suspension

Two 10% stock colloid solutions were used to construct the suspension employed during the experiment. The two stock solutions were hydrophobic Polystyrene (PS) and hydrophilic Carboxylate Modified Polystyrene (CMPS). With both having a mean diameter of 5µm and a density of 1.05 g/cc. The suspension was made through mixing 50 µl of the CMPS solution and 50 µl of the PS solution together inside a beaker and using deionized water to fill the beaker up to 10 ml. Prior to commencing the experiment, the colloid suspension was sonicated with an ultrasonic processor (SONICS, Vibra cell) for one minute in a cold bath to achieve adequate monodispersing of the colloids.

2.3 Experimental Setup

The setup for the experiments included a microscope stage (Leica Z6 APO) (Nishad & Al-Raoush, 2021), where the microchips would be placed, and fastened using tape during each experimental run. The microscope stage is equipped with a precise moving stage that can move the micromodel with an accuracy of 1 µm. The micromodel is connected to a precision syringe pump (Kats Scientific, NE-1010) at the inlet for the sake of injecting the colloid suspensions into the model. The connection is via a tube with an outer diameter of approximately 1.5875 mm, and an inner diameter of 0.25 mm. While another tube was connected to the outlet and left hanging over an empty beaker to simulate atmospheric pressure. The Leica camera was used for image and video acquisition purposes during each injection phase with no shading and robust sharpening. The settings on the camera for exposure, gain, gamma, saturation, and resolution were 16.5 ms, 2.0x, 0.80, 103.00, and 2592x1944 respectively.

2.4 Procedure

The chips are cleaned with ethanol, followed by deionized water by manually injecting them into the media using syringes for several pore volumes. Afterwards, a syringe filled with air is inserted into the system and air is injected into the media until saturation reaches or is close to 0% of the void space.

The experiment consists of three distinct injection phases, the first imbibition (IMB1), drainage (DRN), and the second imbibition (IMB2). Where in (IMB1), the colloidal suspension is injected into the microchip at a velocity of 3 µl/min using the syringe pumps. This is done until the gas phase within the media is completely gone or the saturation reaches at least 75% of the total void space pictured within the chosen area to be investigated. During the IMB1 phase. In the DRN phase, the geometry is drained by setting the pump to the withdraw function at the velocity of 3 µl/min. The third stage involves a second imbibition IMB2 of the suspension into the geometry once again at the same velocity. The outlet is connected to open air using a tube hanging over an empty beaker to collect the suspension during the experimental run. During each phase, images are acquired continuously at a zoom level of 2.5 on the middle left portion of the geometry, which is facing the inlet.

3 Results

3.1 IMB1 Phase

Figure 1 demonstrates the accumulative effect of pore clogging in the IMB1 phase. As it shows clogging events developed over a small period of time. Some of the clogging events documented in the figure can be taken as a result of residual gas bubbles dissolving, resulting in opening more pathways for the colloids to migrate through. The impact of clogged pores on the clogging of surrounding pores remains apparent, especially going from Figure [1, c] to [1, d]. From examining figures [1, c] and [1, d], the change in the tortuosity of the colloid pathways becomes clear. Wherein the colloids are forced into more tortuous paths that lead to a higher amount of clogged pores. This is exemplified in the 23rd and 24th clogging events documented in Figure [1, d], as prior to the lower clogging events, the upper middle area of the recorded geometry was not under much pressure from the migration of colloids compared to its latest state. The dependency of these clogging events becomes more apparent when considering that clogging events 5, 6, and 9 in blue, and 18, 19, and 20 in red block the direct pathways towards clogging events 23 and 24. This proves that the lower clogging events caused alterations in the flow paths of the colloids and moved them upwards.

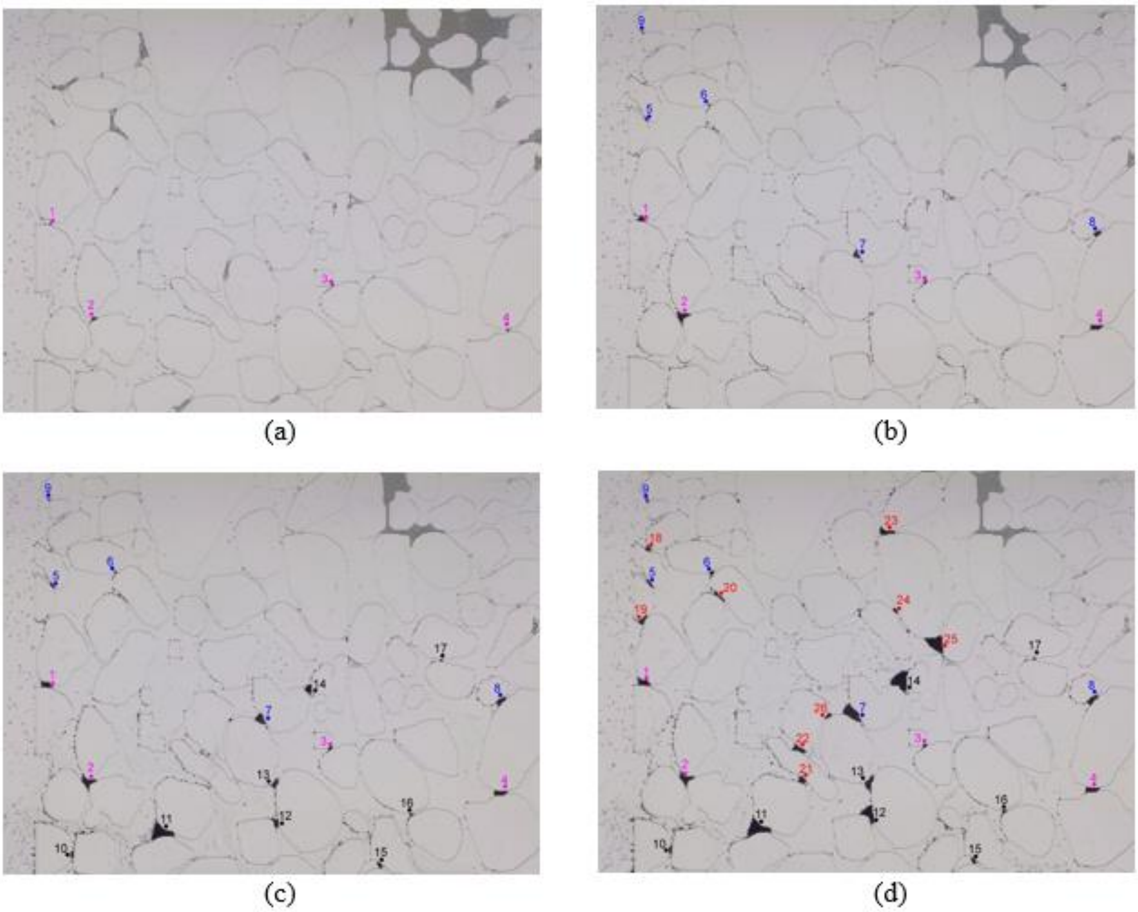

Fig. 1: Accumulative clogging through developing pathways. (a) was acquired at 10:46, (b) was acquired at 10:51, (c) was acquired at 10:53, and (d) was acquired at 10:54. The colors indicate when each clogging event occurred. The colours of purple, blue, black, and red indicate clogging developments for images (a) through (d)chronologically, respectively.

Figure 2, on the other hand, shows the blocking effect of residual gas bubbles within the media on colloid migration and pore clogging. In that in the five highlighted clogging events, colloids do not even approach nor attach around residual gas bubbles that block pore throats. This is indicative of the effect that residual gas has on colloid migration and pore clogging through lowering the number of throats that colloids can pass through. Thereby forcing them to accumulate instead of branching out in multiple pathways. This visually confirms the findings of (Prempeh et al., 2020) with regard to residual gas lowering permeability.

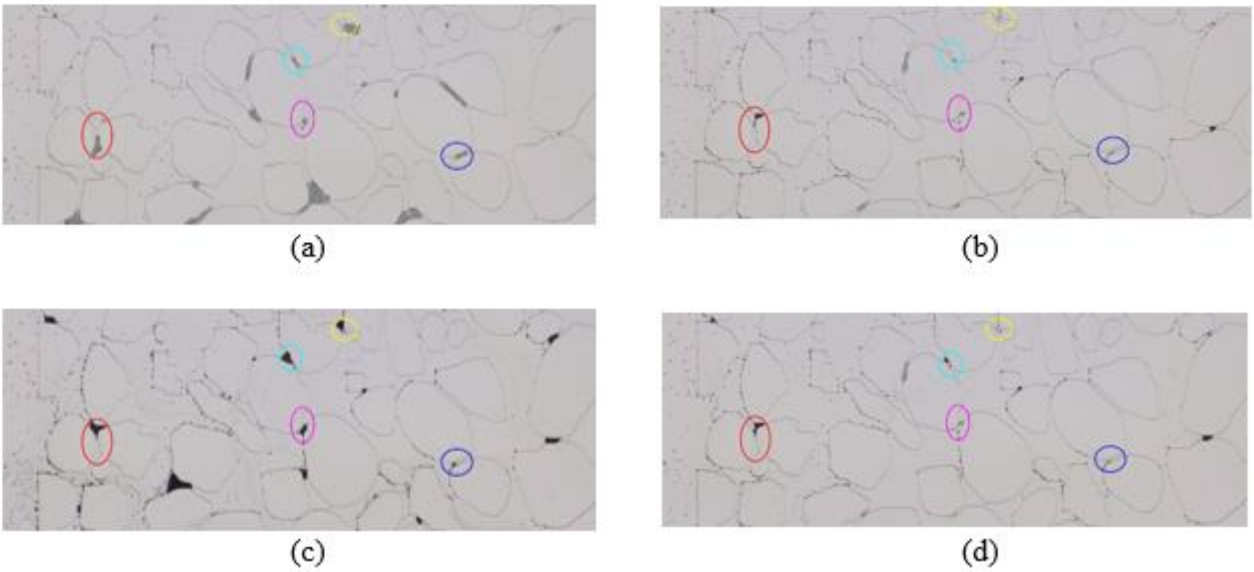

Fig. 2: Residual gas bubbles blocking colloid attachment and pore clogging prior to dissolving. Images **(a)** through **(d)** are ordered chronologically, with **(a)** being acquired first, and **(d)** being acquired last

3.2 DRN Phase

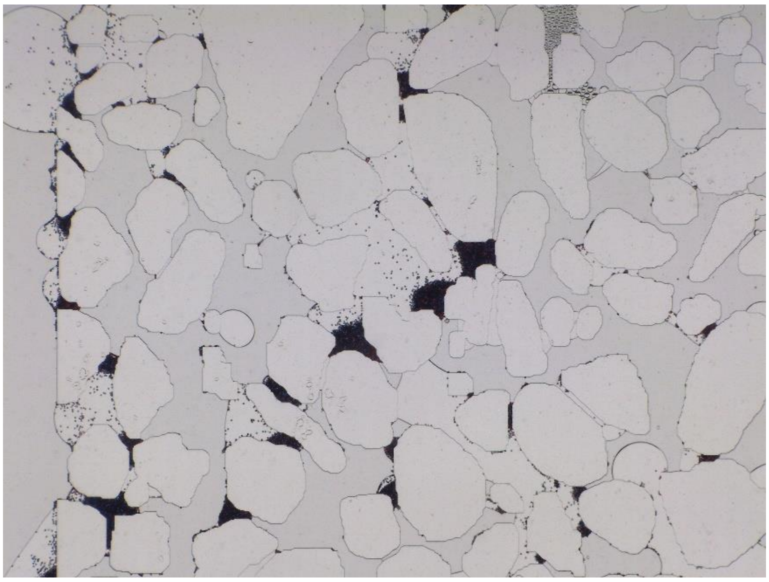

Fig. 3: Clogged pores successfully blocking gas phase from expanding

Figure 3 from the drainage phase clearly displays how the pores that were clogged by the accumulation of colloids successfully block the gas phase in the recorded region from escaping or expanding further. The loose gas phase on the right side of the image is most likely a result of unclogged pores from above and below the recorded area. And while there was some dispersion of the clogging colloids during the beginnings of the DRN phase, most of the colloids remained clogging the pore throats.

3.3 IMB2 Phase

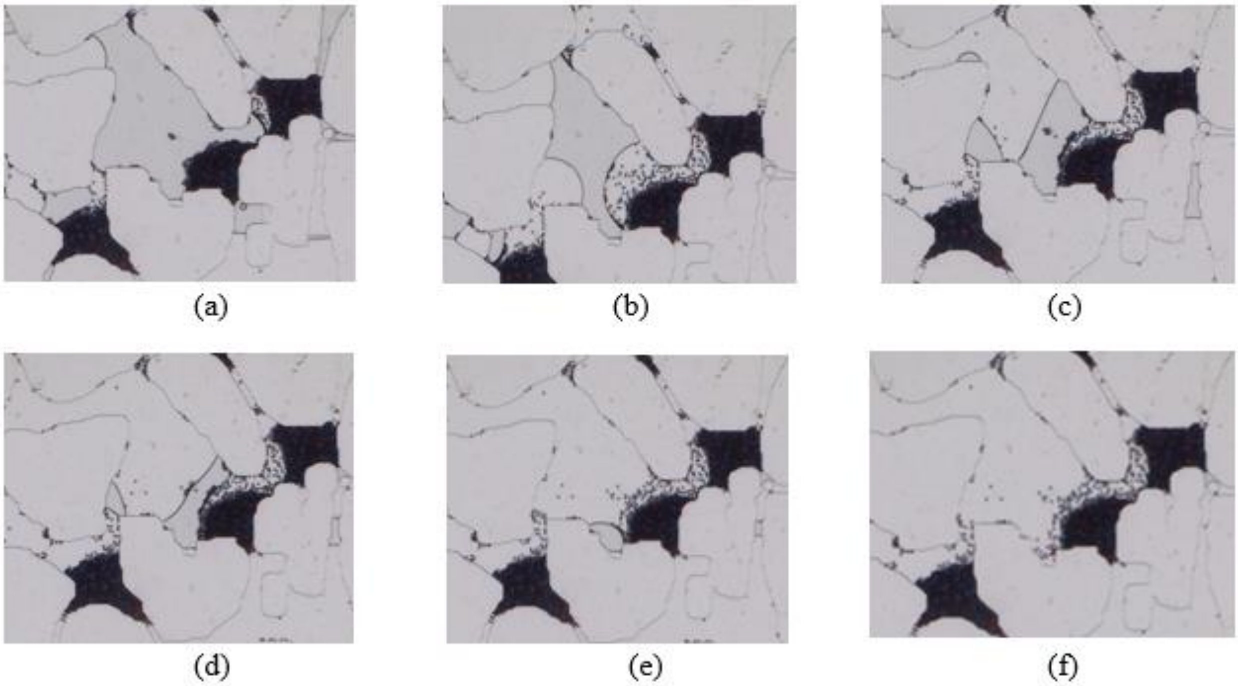

Fig. 4: Dissolved bubble blocked from protruding into geometry.
The images are ordered chronologically from **(a)** to **(f)**

The series of images from Figure 4 show that the clogged pores also block gas bubbles from moving throughout the geometry, and trap them in place. This, along with the injection of the colloidal solution during this phase leaves no option for the trapped gas bubbles adjust to the applied pressure other than dissolving. It is also notable that these trapped gas bubbles cannot carry colloids with them throughout the system like the case mentioned in (Nishad & Al-Raoush, 2021). This can be safely attributed to the lower porosity and smaller throat sizes of this geometry compared to the one used in that study.

4 Conclusion

This study has managed to visualize the relation between dependent clogging and flow paths within a geometry modelled after actual porous media. This is an important observation when studying dependent clogging, as it was mostly studied based on adjacency alone as done previously by (Liot et al., 2018; Liu et al., 2019). The paper also shows the concurrent impact that the residual gas phase and colloids have on each other. As clearly shown with the case of the residual gas bubbles blocking colloids from approaching certain throats that they occupy. And with the case of colloid-clogged throats trapping the gas bubbles within them. Preventing them from expanding and migrating throughout the system, and forcing them to dissolve.

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References

Akhtar, M. S., Nakashima, Y. & Nishigaki, M. (2021). Clogging mechanisms and preventive measures in artificial recharge systems. *Journal of Groundwater Science and Engineering*, *9*(3), 181–201. https://doi.org/10.19637/j.cnki.2305-7068.2021.03.002

Bacchin, et al. (2014). Clogging of microporous channels networks: role of connectivity and tortuosity. *Microfluidics and Nanofluidics*, *17*(1), 85–96. https://doi.org/10.1007/s10404-013-1288-4

Bate, et al. (2022). The Migration and Deposition Behaviors of Montmorillonite and Kaolinite Particles in a Two-Dimensional Micromodel. *Materials*, *15*(3), 855. https://doi.org/10.3390/ma15030855

Cao, et al. (2019). 2D micromodel study of clogging behaviour of fine-grained particles associated with gas hydrate production in NGHP-02 gas hydrate reservoir sediments. *Marine and Petroleum Geology*, *108*, 714–730. https://doi.org/10.1016/j.marpetgeo.2018.09.010

Dersoir, et al. (2015). Clogging of a single pore by colloidal particles. *Microfluidics and Nanofluidics*, *19*(4), 953–961. https://doi.org/10.1007/s10404-015-1624-y

Kokubun, et al. (2019). A pore-scale study of transport of inertial particles by water in porous media. *Chemical Engineering Science*, *207*, 397–409. https://doi.org/10.1016/j.ces.2019.06.036

Han, et al. (2020). Fines migration and pore clogging induced by single- and two-phase fluid flows in porous media: From the perspectives of particle detachment and particle-level forces. *Geomechanics for Energy and the Environment*, *23*, 100131. https://doi.org/10.1016/j.gete.2019.100131

Hannun, et al. (2022). Fines effect on gas flow in sandy sediments using µCT and pore networks. *Journal of Natural Gas Science and Engineering*, *108*, 104834. https://doi.org/10.1016/j.jngse.2022.104834

Jung, et al. (2018). A microfluidic pore model to study the migration of fine particles in single-phase and multi-phase flows in porous media. *Microsystem Technologies*, *24*(2), 1071–1080. https://doi.org/10.1007/s00542-017-3462-1

Khilar, K. C. & Fogler, H. S. (1983). Water Sensitivity of Sandstones. *Society of Petroleum Engineers Journal*, *23*(01), 55–64. https://doi.org/10.2118/10103-PA

Li, Q. & Prigiobbe, V. (2018). Numerical Simulations of the Migration of Fine Particles Through Porous Media. *Transport in Porous Media*, *122*(3), 745–759. https://doi.org/10.1007/s11242-018-1024-3

Liot, et al. (2018). Pore cross-talk in colloidal filtration. *Scientific Reports*, *8*(1). https://doi.org/10.1038/s41598-018-30389-7

Liu, Q., Zhao, B. & Santamarina, J. C. (2019). Particle Migration and Clogging in Porous Media: A Convergent Flow Microfluidics Study. *Journal of Geophysical Research: Solid Earth*, *124*(9), 9495–9504. https://doi.org/10.1029/2019JB017813

Mondal, S., Wu, C. H., & Sharma, M. M. (2016). Coupled CFD-DEM simulation of hydrodynamic bridging at constrictions. *International Journal of Multiphase Flow*, *84*, 245–263. https://doi.org/10.1016/j.ijmultiphaseflow.2016.05.001

Nishad, S. & Al-Raoush, R. I. (2021). Colloid retention and mobilization mechanisms under different physicochemical conditions in porous media: A micromodel study. *Powder Technology*, *377*, 163–173. https://doi.org/10.1016/j.powtec.2020.08.086

Parvan, et al. (2020). Insight into particle retention and clogging in porous media; a pore scale study using lattice Boltzmann method. *Advances in Water Resources*, *138*, 103530. https://doi.org/10.1016/j.advwatres.2020.103530

Prempeh, et al. (2020). Effects of the capillary-entrapped phase on fines migration in porous media. *Journal of Natural Gas Science and Engineering*, *73*, 103047. https://doi.org/10.1016/j.jngse.2019.103047

Reddi, et al. (2000). Permeability Reduction of Soil Filters due to Physical Clogging. *Journal of Geotechnical and Geoenvironmental Engineering*, *126*(3), 236–246. https://doi.org/10.1061/(ASCE)1090-0241(2000)126:3(236)

Rod, et al. (2018). Effect of chemical and physical heterogeneities on colloid-facilitated cesium transport. *Journal of Contaminant Hydrology*, *213*, 22–27. https://doi.org/10.1016/j.jconhyd.2018.03.012

Shen, J. & Ni, R. (2017). Experimental investigation of clogging dynamics in homogeneous porous medium. *Water Resources Research*, *53*(3), 1879–1890. https://doi.org/10.1002/2016WR019421

Yusof, et al. (2020). Petrophysical Changes of Sandstone Due to Salt Precipitation and Fines Migration During Carbon Dioxide Injection. *Journal of Computational and Theoretical Nanoscience*, *17*(2), 1207–1213. https://doi.org/10.1166/jctn.2020.8791

___. (2021). Influence of Brine–Rock Parameters on Rock Physical Changes during CO2 Sequestration in Saline Aquifer. *Arabian Journal for Science and Engineering*. https://doi.org/10.1007/s13369-021-06110-8

___. (2022). Experimental study of CO2 injectivity impairment in sandstone due to salt precipitation and fines migration. *Journal of Petroleum Exploration and Production Technology*. https://doi.org/10.1007/s13202-022-01453-w

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