# Retention of Hydrophobic Colloids in Unsaturated Porous Media using Microfluidics

**Jamal Hannun**
PhD Candidate, Department of Civil and Architectural Engineering, Qatar University, Doha, Qatar
jh1300283@qu.edu.qa

**Riyadh Al-Raoush**
Professor, Department of Civil and Architectural Engineering, Qatar University, Doha, Qatar
riyadh@qu.edu.qa

## Abstract

Water recharge wells can provide a solution for 3.5 billion people, living in regions suffering from water scarcity. Due to fines migration, freshwater wells that are used to recharge aquifers, often experience expedited deterioration. Colloidal clay fine particles can be mobilized from within aquifers due to hydrodynamic forces or the sweeping of gas-water interface (GWI). The released colloids concentration increases then starts to retain and clog at the pores within the aquifer formation. Although fines migration is responsible for decommissioning many recharge wells, yet there is a lack of pore scale observations that uncover clogging mechanisms within porous media. Thus, this study utilizes wide-field optical macroscopy and microfluidic models with pore morphology of sandstone, to investigate the clogging mechanisms of hydrophobic colloids. The aim is to discover how interfacial surfaces within porous media retain colloids. Hence imbibition and drainage of colloidal suspension were carried to vary water saturation. Flow experiments were imaged at a resolution of 1µm/pixel, while colloids diameter was 5 µm. Images were segmented into solid, water, gas and colloids. Then the amount of colloids retained on each interface was quantified. Findings revealed that hydrophobic colloids retained mainly on the GWI. For colloids suspension in deionized water, affinity of colloids to GWI was high enough to cause bubble stabilization. In both hydrophobic and hydrophilic porous media, colloids disconnected the gas phase to create larger GWI surface. More than 90% of hydrophobic colloids were cleaned from the media after drainage, uncovering an efficient remediation technique for water aquifer.

**Keywords:** Fines clogging; Unsaturated Porous Media; Microfluidic; Hydrophobicity; Colloid retention

## 1 Introduction

Managed or artificial water recharge is the practice of using groundwater wells to pump surplus water into aquifers, to be used during water shortages, or to support overexploited aquifers ("Ground Water Recharge Using Waters of Impaired Quality," 1994). Nearly two thirds of earth population live in regions suffering from severe water scarcity during drought periods (Mekonnen & Hoekstra, 2016). The recharge of water aquifers can provide a cost effective and a sustainable solution for this problem. Kim et al. (2021) assessed that there is a growth in the practice of collecting rainwater in order to recharge groundwater aquifers. Specially in arid regions, where rainwater is the primary renewable water source (Edmunds, 2003). This is because naturally trivial segment of rainwater reaches to groundwater aquifers (Baalousha et al., 2018). Thus, artificial geological water recharge (AGWR) is an environmental, sustainable and an economic solution, for many water-stressed populations around the globe. AGWR can raise the recovery ratio of rainwater by 1000% (Mohieldeen et al., 2021).

However, due to fines migration, freshwater wells that are used to recharge aquifers, often experience expedited deterioration. Fines colloidal clay particles are released from within the porous media of sediments that compose the structure of aquifers, this is due to either the hydrodynamic forces, or the sweeping of gas-water interface (GWI) between the pores (Hannun et al., 2022). As the concentration of the released colloids increases within the pores, the colloids then start to retain and clog within the formation of the aquifer (Hannun et al., 2020), radially outward of an injection well, around the well formation (Jarrar et al., 2020). Even though fines migration and the subsequent clogging are responsible for the early decommissioning of many recharge wells, yet there is a lack of pore scale observations that reveal clogging mechanisms within porous media, especially during multiphase flow of gas and water. Other than groundwater recharge, progress in numerous environmental, hydrological and industrial applications is restricted, due to a knowledge gap on how the hydrophilicity of the porous media, affects retention and clogging of colloids (Wang et al., 2020).

The target of this paper is to explore the influence of hydrophilicity of porous media on colloids retention at the different interfaces within a porous media, and to connect colloids retention with different flow conditions such as drainage and imbibition. To capture the release and retention of colloids at each interface. Hence developing the ability to use microscopic images of micromodels to quantify water saturation, volume of colloids in pores and at the interfaces. With the aim to discover how interfacial surfaces within porous media retain colloids, and how the hydrophilicity of the porous media influences the distribution of colloids on interfaces.

## 2 Methodology

### 2.1 Materials Experimental Setup

Table 1 details the parameters of the microfluidic flow experiments. The two glass porous media with pore geometry of sandstone were used to compare how the media hydrophilicity will affect colloids retention on the solid collector surfaces. One chip was made of normal glass which is hydrophilic, while the other was made with glass that is coated with a hydrophobic material. The contact angle of a water droplet under atmospheric pressure was 37° for the hydrophilic chip, and 75° for the hydrophobic chip, measured from solid to air through water. The colloids used were water unfavourable polystyrene latex microspheres, which is naturally hydrophobic. Both imbibition and drainage of the colloidal suspension were carried out to vary water saturation and test how changing flow condition alters the distribution of colloids at the interfaces. The macroscope was used to collect coloured images, where the water saturation was varied to capture four steps per flow condition.

**Table 1:** Experimental test matrix

| Property | Parameters | |
|---|---|---|
| Collector hydrophilicity | Hydrophobic (75°) | Hydrophilic (37°) |
| Colloids water favorability | Water unfavorable | |
| Flow condition | Drainage | Imbibition |
| Saturation steps | 4 steps at each flow condition, a total of 8 | |

## 2.2 Experimental Setup

The experiment employed a widefield macroscope with microfluidic chips that resemble the pore morphology of sandstone rock samples. To study, the mechanisms of hydrophobic colloids retention and subsequent clogging (Hannun et al., 2019). The microfluidic flow experiments were captured using 5 MP images at a spatial resolution of 1µm/pixel. The sandstone media have a well-connected porous media, with pore and throat sizes ranging between 500 µm to 50 µm.

The chips are cleaned by pumping multiple pore volumes of deionized water and ethanol as per the procedure of Nishad & Al-Raoush (2021). Then a water-colloid suspension is prepared by mixing the polystyrene latex microspheres with deionized water (0.5% colloids and 99.5% water). The suspension is then sonicated to uniformly disperse the colloids after that placed into a syringe. The microspheres have a diameter of 5 µm, ten times smaller than the narrowest throat. The syringe is placed in a syringe pump and connected to one inlet of the microfluidic chip. The chip has an outlet that is open to the atmosphere. The experiment starts with the micromodel fully saturated with water-colloid suspension, then the pump actuates the syringe to force drainage and imbibition. The chips are imaged during the flow experiments to collect coloured microscopic observations.

## 2.3 Analysis

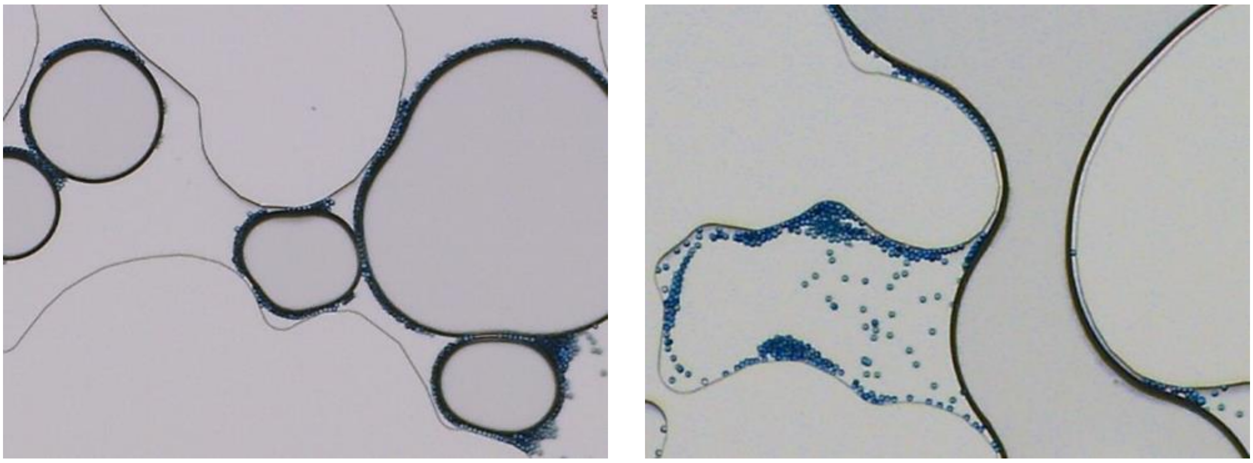
Images were segmented into gas, water, solid and colloids using computer vision algorithms (Al-Raoush et al., 2019), allowing the quantification of porosity, water saturation and colloidal fines content (Jarrar et al., 2021). Next edge detection was used to extract the interfaces of GWI, Gas-Solid interface and Water-Solid interface; permitting the measurements of each interfacial area. Then the amount of colloids retained on each interface was quantified, to study the location and quantity of colloids retention at the interfaces.

# 3 Results

In the hydrophilic micromodel that resembles a sandstone porous media, drainage was carried from a fully saturated media to an irreducible water saturation of 25%. Gas entered the micromodel mobilizing the colloids retained within the pores and at the solid interface, of the collector media, GWI started to sweep and retain the colloids. While the gas front progressed through the media, the colloids attached to the GWI started to disconnect the front, leaving detached bubbles throughout the media. Figure 1 (A) shows how the colloids are responsible for bubble stabilization.

In the hydrophobic micromodel, drainage was carried from full saturation to an irreducible water saturation of 18%. As gas invaded the media, colloids started to retain within the pores, at the water-solid interface and at the GWI, as shown in Figure 1 (B). Compared to hydrophilic media, colloids in hydrophobic media are more distributed over the different interfaces during drainage.

Figure 2 (I) displays a microscopic image of hydrophilic porous media during drainage, at a water saturation of 58%; the image was segmented into labels that were quantified to measure the amount of colloids retained on each interface. Figure 2 (II) demonstrates that the ratio of colloidal fines retained on the GWI to the media volume, increases with drainage up to 58% water saturation. This increase is caused by the separation of the gas front into disconnected bubbles, where the colloids expand the GWI surface area to allow for more retention. This reveals that water unfavourable colloids can be swept out of a hydrophilic porous media under drainage.

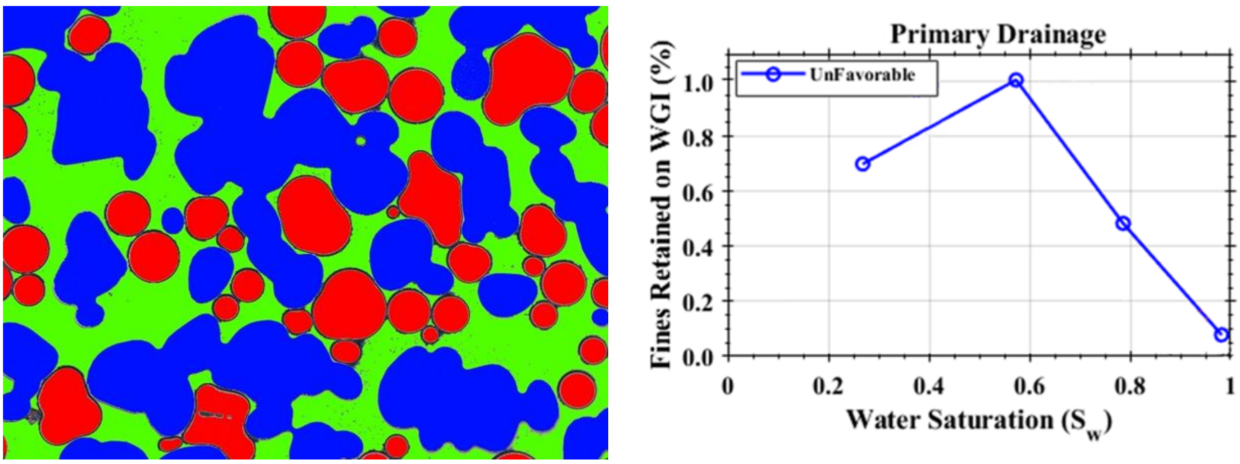


**(a)** **(b)**

**Fig. 1:** Microscopic images showing the first flow condition of air drainage, mobilizing the colloids-water suspension from the porous media, **(a)** hydrophilic collector model with water unfavourable colloids. Similarly **(b)** hydrophobic collector model with water unfavourable colloids

The second part of the experiment test the flow condition of imbibition, colloids were distributed more evenly within the pores and over the interfaces of the porous media, compared to drainage. This shows that the flow condition is responsible for generating hydrodynamic forces and an invading pattern that can change the distribution of colloids retention locations and quantities.

The breakthrough of this work is connecting quantitative measurements of colloidal fines with microscopic visual observations, of colloids retention at the interfaces within a porous media. Visual and quantitative evidence demonstrates that unfavourable colloids undergoing drainage in a hydrophilic micromodel, mainly retain on GWI causing a disconnected flow regime due to bubble stabilization.




**(I)** **(II)**

**Fig. 2:** (I) shows a segmented microscopic image of hydrophilic porous media with water unfavourable colloids, phases of gas, water, solid and colloids, are labelled as red, green, blue and grey, respectively. (II) is the measured ratio of colloidal fines retained on GWI to the volume of the porous media, versus water saturation during drainage.

## 4 Conclusion

Findings revealed that:

1- Hydrophobic colloids (5µm diameter latex spheres), have higher retention at the GWI, compared to other interfaces or the pores within the porous media.

2- In a deionized water medium, a suspension of hydrophobic colloids can cause bubble stabilization within the porous media, because of the high affinity of colloids toward GWI.

3- Retention of colloids at the GWI, disconnected the invading gas phase during drainage, increasing the GWI surface area, noticeably more in hydrophilic compared to hydrophobic porous media.

4- Drainage removed over 90% of hydrophobic colloids from the porous media, uncovering an efficient method for the remediation of water aquifers, which can utilize the drainage of water and air bubbles to clean a contaminated porous media within an aquifer.

5- During water imbibition, the hydrophobic colloids are distributed over all the interfaces in the porous media, unlike drainage where the majority is swept by the GWI.

## References


Al-Raoush, et al. (2019). Impact of fines type on gas flow using 3D micro-computed tomography. *Society of Petroleum Engineers - SPE Kuwait Oil and Gas Show and Conference 2019, KOGS 2019*. https://doi.org/10.2118/198106-ms

Baalousha, et al. (2018). Groundwater recharge estimation and its spatial distribution in arid regions using GIS: a case study from Qatar karst aquifer. *Modeling Earth Systems and Environment*, *4*(4), 1319–1329. https://doi.org/10.1007/S40808-018-0503-4/TABLES/8

Edmunds, W. M. (2003). Renewable and non-renewable groundwater in semi-arid and arid regions. *Developments in Water Science*, *50*(C), 265–280. https://doi.org/10.1016/S0167-5648(03)80023-0

Ground Water Recharge Using Waters of Impaired Quality (1994). *Ground Water Recharge Using Waters of Impaired Quality*. https://doi.org/10.17226/4780

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

Hannun, et al. (2019). *Pore Networks to Characterize Formation Damage Due to Fines at Varied Confinement and Sand Shape*. https://doi.org/10.3997/2214-4609.201903116

Hannun, et al. (2020). *Use of 3D Images to Evaluate Formation Damage Induced by Montmorillonite Fines in Porous Media Systems*. 644–648. https://doi.org/10.29117/cic.2020.0082

Jarrar, et al. (2021). New model for estimating geometric tortuosity of variably saturated porous media using 3D synchrotron microcomputed tomography imaging. *Soil Science Society of America Journal*, *85*(6), 1867–1879. https://doi.org/10.1002/saj2.20289

Jarrar, et al. (2020). 3D synchrotron computed tomography study on the influence of fines on gas driven fractures in Sandy Sediments. *Geomechanics for Energy and the Environment*, *23*, 100105. https://doi.org/10.1016/J.GETE.2018.11.001

Kim, et al. (2021). Assessment of the need and potential for groundwater artificial recharge based on the water supply, water demand, and aquifer properties in a water shortage region of South Korea. *Environmental Earth Sciences 2021 80:3*, *80*(3), 1–15. https://doi.org/10.1007/S12665-021-09445-5

Mekonnen, M. M. & Hoekstra, A. Y. (2016). Sustainability: Four billion people facing severe water scarcity. *Science Advances*, *2*(2). https://doi.org/10.1126/SCIADV.1500323/SUPPL_FILE/1500323_SM.PDF

Mohieldeen, Y. E., Elobaid, E. A. & Abdalla, R. (2021). GIS-based framework for artificial aquifer recharge to secure sustainable strategic water reserves in Qatar arid environment peninsula. *Scientific Reports 2021 11:1*, *11*(1), 1–13. https://doi.org/10.1038/s41598-021-97593-w

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

Wang, et al. (2020). Colloid transport through soil and other porous media under transient flow conditions—A review. *Wiley Interdisciplinary Reviews: Water*, *7*(4), e1439. https://doi.org/10.1002/WAT2.1439

**Cite as**: Hannun J., Al-Raoush R., "Retention of Hydrophobic Colloids in Unsaturated Porous Media using Microfluidics", *The 2nd International Conference on Civil Infrastructure and Construction (CIC 2023)*, Doha, Qatar, 5-8 February 2023, DOI: https://doi.org/10.29117/cic.2023.0177