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Liquid Fragmentation Induced by Particle Aggregation During Two-Phase Flow in 3D Porous Media

Key Points:

- We experimentally study particle transport and aggregation during two-phase flow in porous media
- We identify a previously overlooked pore-scale event: wetting fluid fragmentation induced by particle aggregation
- Particle aggregation strongly influences fluid connectivity and relative permeability in the porous media

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Supporting Information:

Supporting Information may be found in the online version of this article.

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Abstract Natural or engineered microparticles are often encountered in subsurface multiphase flow systems. This introduces a complex flow scenario with a variety of applications. However, the influence of particles on multiphase flow dynamics and the underlying mechanism remain elusive. Here, we investigate particle transport behavior and fluid phase distribution within 3D porous media through direct visualization utilizing laser scanning confocal microscopy. The mechanisms and factors governing particle aggregation during two-phase flow are elucidated. We identify a previously overlooked pore-scale phenomenon: fragmentation of wetting liquid induced by spontaneous particle aggregation in localized regions. This process dramatically increases the number of wetting clusters while reducing the fluid connectivity, resulting in a change in the relative permeability of fluids. These findings reveal the dynamic coupling mechanism between pore-scale particle aggregation and fluid flow and transport properties at larger scales, providing critical insights for predicting and controlling particle mediated geophysical flow processes.

Plain Language Summary Particle transport during two-phase flow is frequently involved in various natural processes and engineering applications, ranging from water infiltration to oil recovery. Therefore, understanding particle transport behaviors and their impacts on two-phase flow in porous media is essential. Here, by using confocal microscopy we directly visualize particle transport and aggregation at the pore scale under various particle sizes, grain sizes, and flow rates. The results show that particles tend to aggregate at the wetting-nonwetting interfaces or the solid-wetting-nonwetting triple point at high flow rates and smaller pore sizes. Furthermore, we observe that particle aggregation in the narrow regions of the wetting phase can facilitate the breakup of liquid clusters into smaller ones, leading to a dramatic increase in the total number of wetting liquid clusters. This indicates that particle aggregation in porous media can reduce the wetting phase connectivity by inducing liquid breakup, which subsequently reduces the relative permeability of fluids. These findings improve our understanding of the coupled processes of two-phase flow and particle transport and thus have broad implications for both geological and engineering systems.

1. Introduction

Two-phase flow is a fundamental process relevant to various natural and subsurface engineering applications, including water infiltration (Mohanty et al., 2015), geological CO₂ sequestration (Vinogradov & Jackson, 2011), and pollutant remediation (Pak et al., 2020; Wang et al., 2022). In this context, nano- and micro-particles such as microsized silt and clay minerals, as important components of geological formations, play a significant role in influencing medium properties and regulating fluid flow dynamics (Drummond et al., 2019). Previous studies on single-phase flow have demonstrated that particle transport affects medium permeability in two opposing ways: the erosion of particles can enhance permeability, while their accumulation and clogging at pore throats can reduce it (Jung et al., 2012). However, in two-phase flow systems, the effects of particle transport and aggregation on flow dynamics remain poorly understood, which poses a challenge to predicting and controlling particle-related engineering and environmental processes such as contaminant transport (Bhattacharjee & Datta, 2019; Paswan & Sharma, 2023; Sen, 2011; Xian et al., 2022) and petroleum recovery (H. Zhang et al., 2014).

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Particle behaviors in porous media essentially depend on their interactions with the solid matrix, fluid, and surrounding particles, and are affected by flow velocity, particle concentration, chemical properties, temperature, as well as pore and particle size (Bradford et al., 2012; Chequer et al., 2019; Gerber et al., 2018; Liang et al., 2022; Sainers & Lenhart, 2003; Wu et al., 2024; You et al., 2015). While the transport and retention of particles in porous media are generally controlled by the relative size of the pores and particles (Han et al., 2020), multiphase flow conditions introduce additional complexities due to the presence of liquid-liquid or air-liquid interfaces (Han et al., 2020). During two-phase flow, particles can be retained in porous media through a variety of mechanisms, such as attachment at interfaces (Zevi et al., 2005), film straining (He et al., 2023; Wan & Tokunaga, 1997; Wu et al., 2021), bridging (Lin et al., 2021a), and filtration (Auset & Keller, 2006; Johnson & Hilpert, 2013). Furthermore, the presence of an immiscible fluid can induce particle agglomeration via adhesive forces, facilitating pore clogging (R. Zhang et al., 2023). While previous works have demonstrated that clogging of particles in pores can alter porous medium homogeneity and permeability (Han & Kwon, 2023), the impact of particle aggregation during two-phase flow on the fluid topology and flow dynamics remains elusive. Understanding fluid topology at the pore scale is crucial, as it largely determines transport and transformation behaviors at larger scales (Armstrong et al., 2016; Mathiesen et al., 2023; Rücker et al., 2015). For example, the trapping efficiency of a fluid is directly linked to the shape of the displacement front (Hu et al., 2018; Lan et al., 2020; Schlüter et al., 2016; Zhao et al., 2019), and the complex morphology of the liquid-liquid interface may affect the adsorption and desorption behaviors of pollutants (Binks, 2017; Flury & Aramrak, 2017; Lazouskaya et al., 2013; Mishurov et al., 2008; Oettel & Dietrich, 2008; Thompson et al., 1998). Given the critical role of particle aggregation in these processes, it is essential to elucidate its effects on flow dynamics and fluid topology during two-phase flow.

Systematic investigation of particle transport and aggregation in 3D porous media by direct imaging remains challenging due to the opacity of natural porous media. Previous efforts to explore the particle transport and retention processes in porous media have mainly focused on column scale experiments which rely on the analysis of breakthrough curves (Tong et al., 2008; Torkzaban et al., 2008). While these studies provided insight into particle transport and deposition mechanisms, they did not quantify the distribution characteristics of particles and fluids in 3D porous media. Recent works have extended these investigations to 3D porous media and provided additional information on the spatial distribution of particles in pores (Bizmark et al., 2020; Gerber et al., 2018, 2019). However, these works only considered the particle behaviors under saturated conditions. A fundamental understanding of the coupling between two-phase flow and particle transport and aggregation is still lacking, despite its significant relevance to a number of important engineering and environmental applications.

In this work, we systematically study the particle distribution and the structures of fluid phases during two-phase flow in 3D porous media by using a sophisticated 3D imaging and visualization technique. The pore-scale, high-resolution visualization allows us to explore particle aggregation and its effect on the wetting fluid cluster size distribution. We find that particle aggregation in porous media can significantly facilitate the liquid cluster breakup. Particle aggregation during two-phase flow also has an impact on the relative permeabilities of fluid by changing its spatial distribution, which can in turn affect the transport of particles. We further discuss the implications of these findings for relevant applications in subsurface engineering settings.

2. Methods and Materials

In this study, we conducted two-phase flow experiments within a porous medium created by randomly packing borosilicate glass beads in a quartz capillary tube (refer to Figure S1 in Supporting Information S1). The glass beads, with an average diameter $D = 69 \mu\text{m}$ ($63 \mu\text{m} < D < 75 \mu\text{m}$), $137.5 \mu\text{m}$ ($125 \mu\text{m} < D < 150 \mu\text{m}$), and $196 \mu\text{m}$ ($180 \mu\text{m} < D < 212 \mu\text{m}$), were selected to simulate a relatively homogeneous pore structure representative of fine-grained soils. The particles are fluorescent polystyrene particles (Thermo Fisher Scientific, R0100) with a mean diameter of $d = 1 \mu\text{m}$ or $2 \mu\text{m}$, corresponding to the size range of colloidal particles commonly found in soils, such as clay particles, microplastics, and mineral debris. To minimize light scattering during 3D imaging, the wetting fluid was comprised of a mixture of dimethyl sulfoxide (DMSO) and deionized water at 91.4% and 8.6% by weight, respectively. The nonwetting fluid is a hydrocarbon oil mixture (Cargille 5040) with the same refractive index as the wetting fluid and glass beads. The densities of the wetting and nonwetting fluids are $\rho_w = 1.1 \text{ g/cm}^3$ and $\rho_{nw} = 0.83 \text{ g/cm}^3$, respectively, and the interfacial tension is $\gamma = 13.0 \text{ mN/m}$. The viscosities of the wetting and nonwetting fluids are $\mu_w = 2.14$ and $\mu_{nw} = 19.849 \text{ mPa s}$, respectively. The three-phase contact is $\theta \approx 34.5^\circ$, as measured from the pore-scale images (Figure S2 in Supporting Information S1).

The experiment involved the injection of the nonwetting phase into a porous medium saturated with the particle suspension at a concentration of 0.1 wt% ($\sim 10^9$ particles mL^{-1}). Initially, we visualized the pore and particle distribution by a confocal microscope (Olympus, FV3000) and confirmed that the particles were well-dispersed in the pore space before drainage (Figure S3 in Supporting Information S1). Then, 10 pore volumes (PVs) of the nonwetting liquid were injected into the medium at a fixed flow rate ($Q = 0.1\text{--}50$ $\mu\text{L}/\text{min}$), corresponding to a capillary number $Ca = \mu_{\text{nw}}U/\gamma$ between 6.36×10^{-6} and 1.91×10^{-2} . This range of flow rates, spanning three orders of magnitude, encompasses a wide range of flow conditions commonly encountered in geological and engineered systems and allows us to capture the variability in particle transport behavior across different hydrogeological scenarios. After injection, the flow cell was scanned by using confocal imaging under static conditions. The image acquisition was restricted to the middle section of the flow cell to minimize inlet and outlet effects, and the resolution was set as 1.591×1.591 $\mu\text{m}/\text{pixel}$ for each horizontal slice. The 3D structure with approximately 400 μm in depth (z -direction) was constructed from 100 optical slices in the x - y plane, spaced by 4 μm from each other along the z -direction (Figure S1 in Supporting Information S1).

3. Results and Discussion

3.1. Particle Aggregation During Two-Phase Flow

To investigate the pore-scale dynamics of particle transport and multiphase flow in 3D porous media, we first examine the distribution characteristics of particles and wetting fluid within the porous media at varying flow rates, as shown in Figures 1a–1c. At a low flow rate (e.g., $Q = 0.1$ $\mu\text{L}/\text{min}$), the wetting liquid occupies most of the pore space, with particles relatively uniformly dispersed within the remaining wetting liquid (see Figure 1a). With the increase in flow rate, the remaining wetting liquid occupies a smaller volume and mainly exists in the form of pendular rings. In this case, particles of both sizes (1 and 2 μm) tend to aggregate near the wetting–nonwetting interface (WNI) or the solid–wetting–nonwetting triple point (Figure 1d). Two main mechanisms contribute to particle retention at these locations: one is the retention at triple points (sites 1, 2, 4) due to the flow funneling and the vortices of low velocity near those wedge-shaped pore spaces (W. Zhang et al., 2010). The other is film straining (site 3), where particles are trapped in a thin film with a thickness smaller than the particle diameter (Babakhani et al., 2017; Bradford et al., 2007; Bradford & Torkzaban, 2008).

Figure 1e shows the proportion of particle clusters ($V > V_0$) in the total number of particle aggregates (N_0) detected in the sample after drainage. V_0 ($V_0 = 120$ μm^3) is a characteristic volume to minimize the influence of resolution limit on the experimental results, where particles within a sufficiently short distance may be identified as a cluster due to the resolution of image acquisition, more details can be found in Figure S4 of Supporting Information S1. Figure 1e suggests that the small particles ($d = 1$ μm) can aggregate to form clusters in the small-grain packings ($D = 69$ μm) at large flow rates ($Q > 5$ $\mu\text{L}/\text{min}$), as evidenced by the increase of $N_{V > V_0}/N_0$ from 0.08 to 0.5 (red dotted line), while the aggregation is negligible in the case of larger packing grain sizes ($D = 137.5$ and 196 μm) for all flow rates (blue and purple dotted lines). In contrast, particles with larger diameters ($d = 2$ μm) exhibit a stronger tendency to aggregate during two-phase flow. Figure 1e shows that aggregation of large particles can be observed under all experimental conditions, with the degree of aggregation initially increasing with the flow rate and then reaching a plateau. The cumulative size distribution (Figure S5 in Supporting Information S1) of particle clusters after drainage further confirms this observation. The effect of increasing flow rate can be attributed to the decrease in the wetting phase saturation, which allows more particles to be transported through pore space regions where straining processes may occur. In addition, the results also show that decreasing the grain/pore size can facilitate particle aggregation, especially for large particles. This effect can be attributed to the changes in the morphology of liquid bridges and their connecting bands (Wu et al., 2023), where their height is in proportion to the packing grain size.

3.2. Wetting Cluster Breakup Induced by Particle Aggregation

During the displacement process, we observe a notable phenomenon: particles aggregate in localized regions of the liquid cluster, resulting in the breakup of larger liquid clusters into smaller ones. As illustrated in Figure 2a, the wetting liquid is initially well-connected, spanning multiple pores and throats (Figure 2, left column). However, with continuous injection of the nonwetting liquid, particles gradually aggregate at the connections between pendular rings of the wetting liquid. This aggregation destabilizes the fluid–fluid interface within the throat, leading to the breakup of the wetting liquid near the aggregated particles, and the number of wetting clusters (N_w)

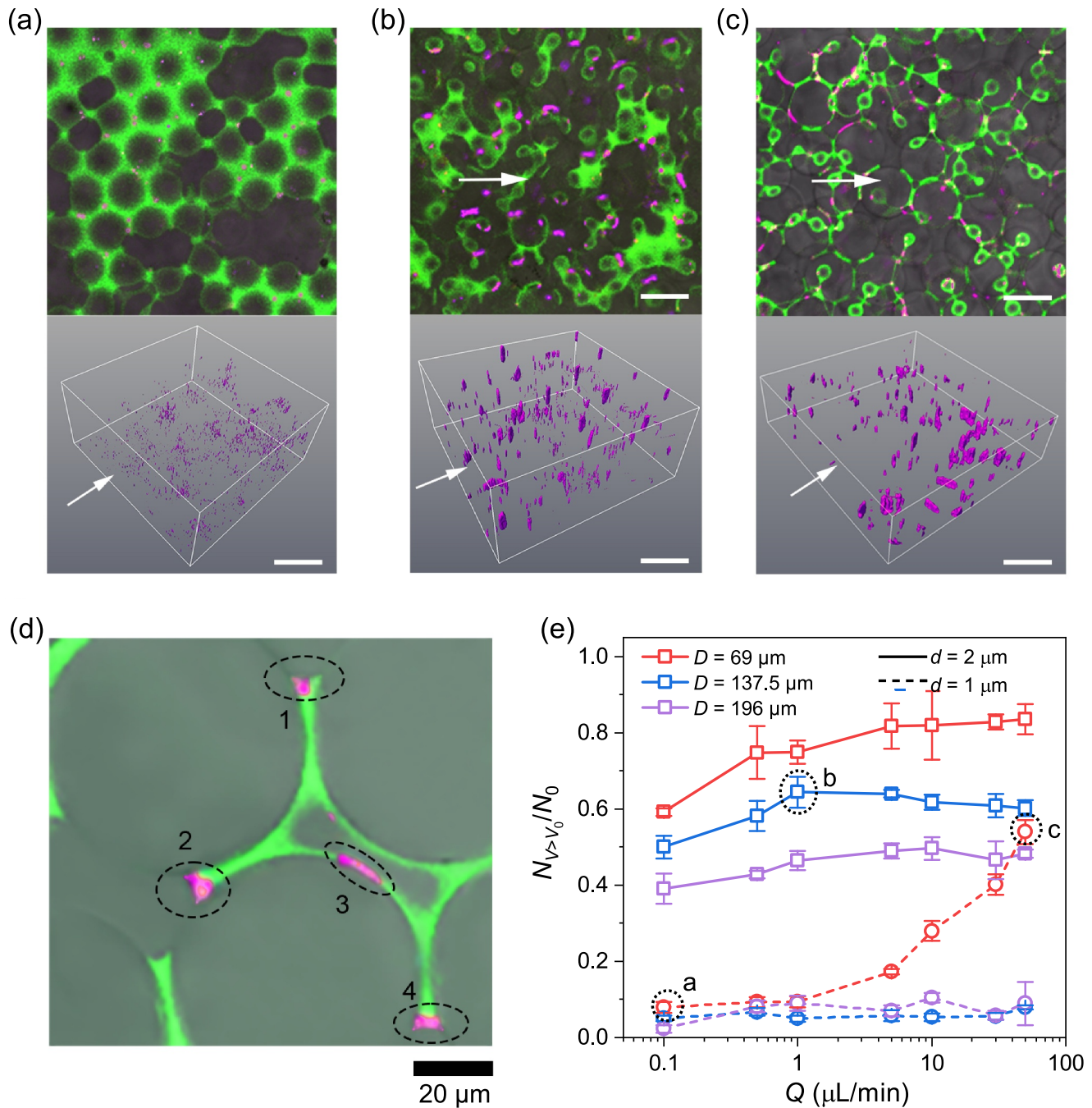


Figure 1. (a–c) Upper row: images showing the spatial distribution of wetting fluid (green) and particles (purple) in a porous medium ($D = 69 \mu\text{m}$) after drainage. The experimental parameters are (a) flow rate $Q = 0.1 \mu\text{L}/\text{min}$, particle diameter $d = 1 \mu\text{m}$; (b) $Q = 1 \mu\text{L}/\text{min}$, $d = 2 \mu\text{m}$; (c) $Q = 50 \mu\text{L}/\text{min}$, $d = 1 \mu\text{m}$. The bottom row is the corresponding 3D reconstructed image showing particle distribution, and the scale bars represent $100 \mu\text{m}$. The arrows indicate the flow direction. (d) Observed retention sites of particles at 1: solid–wetting–nonwetting triple point, 2 and 4: WNI and the solid–wetting–nonwetting triple point, 3: thin film (film straining). (e) Normalized number of particle aggregates as a function of flow rate Q in different porous media with $D = 69 \mu\text{m}$ (red), $137.5 \mu\text{m}$ (blue), and $196 \mu\text{m}$ (purple). The solid and dotted lines represent the cases with $d = 2 \mu\text{m}$ and $d = 1 \mu\text{m}$, respectively. The three data points marked by circles correspond to the images in panels (a–c). The error bars denote the standard deviation.

becomes 4, as shown in Figure 2a. Additional images for similar phenomena are presented in Figure S6 of Supporting Information S1. Following these breakup events, the wetting liquid reconfigures its flow paths until the next breakup event occurs. Notably, the breakup of wetting clusters preferentially occurs at the locations of particle aggregation. This effect can be attributed to the alteration of local flow structures caused by particle

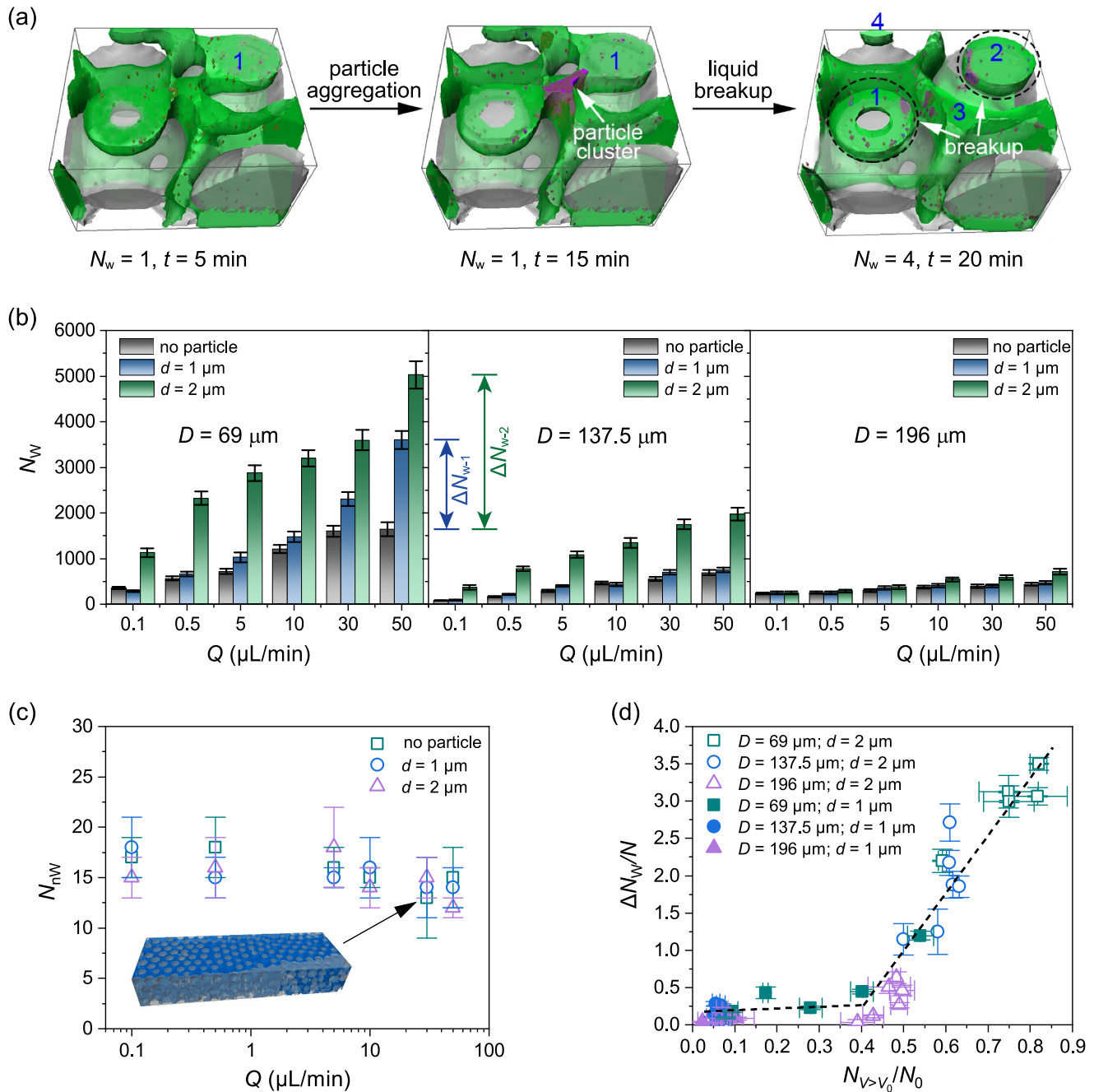


Figure 2. (a) Particle aggregation and wetting liquid breakup for the case of grain size $D = 69 \mu\text{m}$, $Q = 5 \mu\text{L}/\text{min}$. The scale bar is $50 \mu\text{m}$. (b) Total number of wetting clusters for different flow rates, particle sizes, and grain diameters. (c) Total number of nonwetting clusters (N_{nw}) for different flow rates, with $D = 69 \mu\text{m}$. The inset shows the typical distribution of the nonwetting fluid after experiments. Panel (d) shows the relationship between the fragmentation effect and the degree of particle aggregation, where ΔN_w represents the difference in the number of wetting clusters between experiments with and without particles, and N is the number of wetting clusters without particles. The dashed line is trend line for experimental data.

aggregation, which increases the viscous stress (Bizmark et al., 2020; Schneider et al., 2021). The enhanced viscous force directly promotes the breakup of liquid clusters during two-phase flow (Datta et al., 2014). Furthermore, liquid cluster breakup will in turn affect particle transport in porous media due to a reduction of connectivity. Therefore, this coupling between pore fluid flow and particle transport in two-phase flow is expected to strongly influence solute transport dynamics when the solute transport is accompanied by particle influx in a porous matrix (Chen et al., 2009).

To further quantify the influence of particles on the fluid distribution during two-phase flow, we count the total number of wetting phase clusters after drainage, as shown in Figure 2b. The results indicate that particle affects the number of residual wetting phase clusters, depending on the flow rate, as well as the size of the glass beads and particles. Specifically, for suspension with particles of $d = 2 \mu\text{m}$, the total number of liquid clusters is significantly higher than that in the experiments with particles of $d = 1 \mu\text{m}$ and no particle when the grain sizes are $D = 69$ and $137.5 \mu\text{m}$. In contrast, in large-grain packings ($D = 196 \mu\text{m}$), large particles only slightly facilitate cluster formation when $Q > 5 \mu\text{L}/\text{min}$. Additionally, the results also show that the small particles ($d = 1 \mu\text{m}$) affect the wetting cluster distribution only under conditions of $D = 69 \mu\text{m}$ and $Q > 5 \mu\text{L}/\text{min}$. These results indicate that particle size, hydrodynamic condition, and grain size synergistically control the distribution of the wetting liquid in porous media. Notably, particle aggregation exerts minimal impact on the nonwetting phase distribution, as shown in Figure 2c and Figure S7 of Supporting Information S1. Consequently, this study primarily focuses on the influence of particle aggregation on the structure of wetting clusters.

By analyzing the fragmentation-induced increase in wetting cluster number ΔN_w (normalized by the wetting cluster number in the case of no particle, N), we find that the fragmentation is predominantly controlled by the extent of particle aggregation, independent of grain/pore size, as evidenced by the fact that experimental data across varying grain sizes align to the same trend line (Figure 2d). Furthermore, two distinct trends concerning the relationship between $N_{V > V_0}/N_0$ and $\Delta N_w/N$ can be observed. Specifically, when $N_{V > V_0}/N_0 < \sim 0.4$, the impact of particle aggregation on wetting phase fragmentation remains negligible. However, when $N_{V > V_0}/N_0 > \sim 0.4$, the fragmentation is markedly enhanced with increasing particle clusters. This suggests a critical threshold of 0.4 for $N_{V > V_0}/N_0$, above which the fragmentation effect becomes substantial and cannot be overlooked. Importantly, this critical value demonstrates consistency across varying sizes of particles and grains/pores within our experimental systems. These findings further underscore the critical role of particle aggregation in promoting the breakup of wetting clusters during two-phase flow.

3.3. Changes in the Fluid Hydraulic Connectivity

As discussed above, particle aggregation can facilitate liquid cluster breakup during two-phase flow, which can have a marked impact on the two-phase hydraulic and transport properties. Here we analyze the cluster size distribution of the residual wetting liquid. Figures 3a–3c shows the 3D renderings of the wetting liquid after drainage with $D = 69 \mu\text{m}$. Additional experimental images for $D = 137.5$ and $196 \mu\text{m}$ are provided in Figures S8 and S9 of Supporting Information S1. In these visualizations, different liquid clusters are randomly colored, with uniform coloring indicating fewer clusters. It is evident that the connectivity of the wetting liquid decreases with the flow rate. For example, for $Q = 0.1 \mu\text{L}/\text{min}$ ($Ca = 6.36 \times 10^{-6}$), the largest wetting cluster forms a percolating network that spans the entire sample (blue cluster in Figures 3a and 3b). In contrast, for $Q = 50 \mu\text{L}/\text{min}$ ($Ca = 1.91 \times 10^{-2}$), numerous smaller clusters form after drainage, leading to a marked increase in the total number of clusters. This is because increasing flow rate (or Ca) can shift the flow regime from capillary force-dominated to viscous force-dominated, facilitating the breakup of large clusters into smaller ones. Note that different conclusions can be drawn from various definitions of capillary number (Guo et al., 2022). For example, when accounting for the size of fluid clusters (Armstrong et al., 2014), the capillary number at the end of the experiment is likely to be smaller than at the beginning, as the fluid cluster size decreases over time. A comparison of Figures 3a and 3b reveals that particles can further reduce the connectivity of the residual wetting phase, particularly at large flow rates. This effect is corroborated by the changes in the volume of the largest wetting cluster (V_{max}), as illustrated in Figure 3d, which shows that as particle size increases, the largest wetting cluster size decreases under the same flow conditions. This reduction is linked to the liquid cluster breakup induced by particle aggregation within pores.

With the aim of quantifying the influence of particle size and flow rate on cluster formation, we analyze the cumulative size distribution (the number of clusters with sizes larger than or equal to a certain volume) of the wetting liquid clusters for various experimental conditions, as shown in Figure 3e. Clusters smaller than $200 \mu\text{m}^3$ (about 20 voxels) are excluded to eliminate the influence of noise in the raw data. The results show that increasing both flow rate and particle size leads to a substantial increase in the number of wetting clusters, especially small clusters (similar trends can be found in Figures S8 and S9 of Supporting Information S1). Additionally, with the decrease in flow rate, the influence of particles on the number of wetting clusters becomes less pronounced, likely due to a decrease in particle aggregation. This is evidenced by the fact that the cluster size distribution curves for

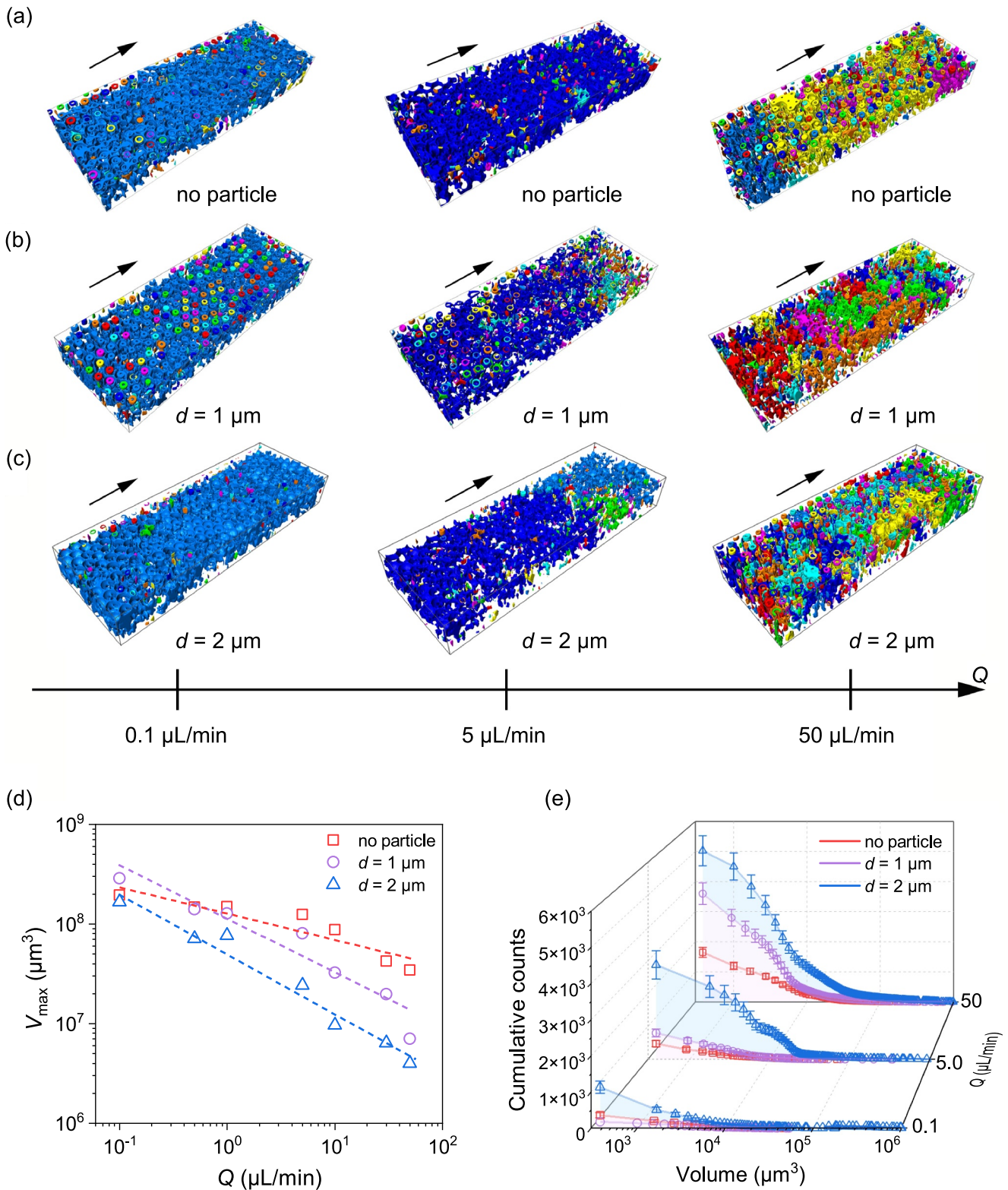


Figure 3. 3D rendering of the wetting liquid clusters after drainage with (a) no particle, (b) $d = 1 \mu\text{m}$, and (c) $d = 2 \mu\text{m}$. The packing bead diameter for the porous medium is $69 \mu\text{m}$. Individual clusters are randomly rendered in different colors, that is, the more uniform the colors, the lesser the number of clusters. The arrows indicate the flow direction. (d) The volume of the largest wetting cluster as a function of Q and particle size. The dashed lines are fitted according to the experimental data. (e) Cumulative cluster size distribution of the wetting liquid.

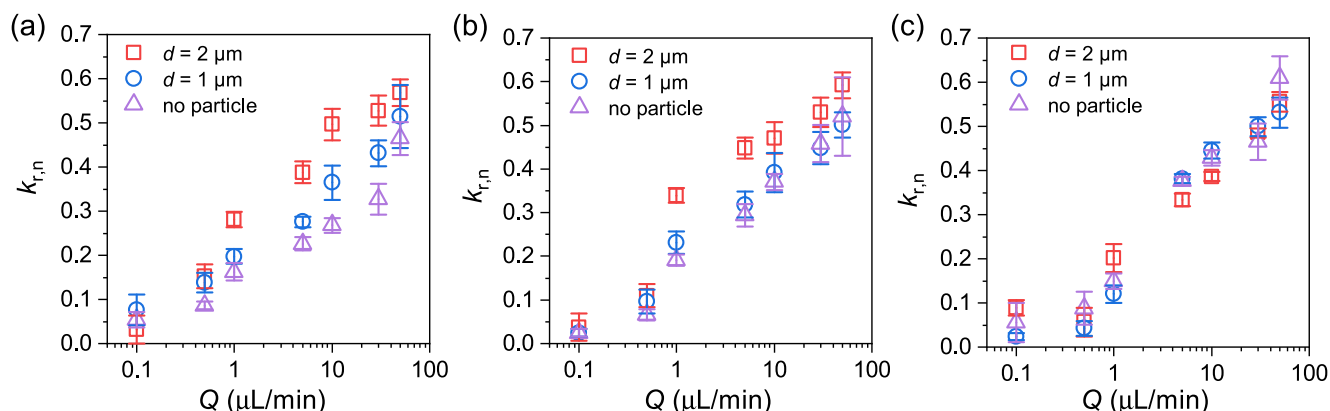


Figure 4. Relative permeability of the nonwetting phase. From left to right, the packing grain diameters are (a) $D = 69 \mu\text{m}$, (b) $D = 137.5 \mu\text{m}$, and (c) $D = 196 \mu\text{m}$, respectively.

the cases with no particle and particle of $d = 1 \mu\text{m}$ almost coincide when $Q = 0.1 \mu\text{L}/\text{min}$ (Figure 3e), since the particle with $d = 1 \mu\text{m}$ does not aggregate in these experiments and therefore has a negligible effect on fluid distribution.

Our pore-scale visualization of particle distribution reveals that the extent of wetting liquid fragmentation is strongly affected by particle aggregation. This means the aggregation of particles is expected to affect the hydraulic connectivity of both the wetting and nonwetting phases in two-phase flow systems. Here, we quantify this effect by estimating the relative permeability of the nonwetting phase based on measured pressure drops and flow rates after drainage reaches a steady state. As shown in Figure 4, in the small-grain packing ($D = 69 \mu\text{m}$), the presence of particles of both sizes (1 and $2 \mu\text{m}$) can lead to an increase in the nonwetting phase relative permeability, with larger particles having a more pronounced effect. Additionally, this effect weakens with the increase of grain/pore size, becoming negligible when $D = 196 \mu\text{m}$ (Figure 4c). These trends are closely tied to the extent of particle aggregation, suggesting that particle aggregation can alter the connectivity of fluid phases and strongly influence the dynamics of multiphase flow in porous media. Considering that changes in permeability affect not only particle transport but also the spatial variability in pore fluid flow (Chen et al., 2009), this finding has significance for further understanding contaminant transport in general multiphase systems and in many groundwater remediation technologies involving nano- and micro-particles and multiple fluids.

4. Discussion

Although previous studies have shown that particle deposition and aggregation can observably modify the internal structure of porous media (Agbangla et al., 2014; Chen et al., 2008; Sendekie & Bacchin, 2016), the role of particle aggregation in multiphase flow remains unclear. This work highlights the significant impact of particle transport and aggregation on the distribution and connectivity of fluid phases in porous media and thus has broad practical implications. By 3D imaging, we directly visualize the particle transport and distribution during two-phase flow, which contributes to an improved understanding of the pore-scale mechanisms of particles (including microorganisms, microplastics, nanoparticles) transport in complex environments (e.g., in the vadose zone where air-water two-phase flow occurs). We discover that particles tend to aggregate in key flow paths, depending on the flow conditions and grain sizes. This behavior can hinder, for example, the transport of solutes and possibly the delivery of remediation agents such as surfactants, chemical oxidants, and microorganisms to the contaminant source zones. On the other hand, this work demonstrates that particle aggregation can facilitate fragmentation of the liquid phase in porous media, which can increase the interface area between different immiscible fluids (Pak et al., 2015). More importantly, the increase in the liquid-liquid interface area can potentially improve the effectiveness of surfactant addition and accelerate microbial degradation (McCray et al., 2001), which may facilitate contaminant remediation in soils and aquifers (Valletti et al., 2023; M. Zhang et al., 2019).

Our results also show that particle aggregation in the pore space can change the relative permeabilities of the liquid phase, suggesting that particle behaviors at the pore scale can have macroscale implications. More

importantly, changing the relative permeability in turn can affect the further transport of particles and solutes in porous media (Molnar et al., 2020), yielding strongly coupled dynamics between the particle transport and multiphase flow. However, traditional advection-dispersion equations describing particle transport in porous media often ignore the influence of particle behavior on hydrodynamic conditions, which likely results in reduced accuracy in the model's prediction (Babakhani et al., 2017; Tong & Johnson, 2006). Therefore, incorporating the particle aggregation behavior at the pore scale into transport models is critical for quantifying and controlling the particle fate in soil and groundwater, which merits further investigations. Moreover, our experiments are carried out in a homogenous porous media. However, natural porous media (such as soils and rocks) are typically heterogeneous with preferential flow pathways (Lin et al., 2021b). How the particle aggregation will influence or be influenced by the preferential flow patterns remains to be explored in future studies.

5. Conclusions

The coupled process of particle transport and multiphase flow is a challenging issue due to the complex transport behavior and intricate morphology of fluid phases. In this study, we investigate the aggregation behavior of particles and their impacts on fluid phase connectivity during two-phase flow in porous media. By using confocal microscopy and 3D imaging, we directly visualize the particle transport and aggregation at the pore scale under different particle and grain sizes, as well as hydrodynamic conditions. The results show that particle transport and distribution in porous media are highly sensitive to its size, grain/pore size, and flow rate. Small particles ($d = 1 \mu\text{m}$) aggregate only under conditions of $D = 69 \mu\text{m}$ and $Q > 5 \mu\text{L}/\text{min}$, while larger particles ($d = 2 \mu\text{m}$) aggregate strongly across all flow rates, with aggregation increasing at higher flow rates and smaller grain sizes. Remarkably, our experimental results suggest that particle aggregation can lead to liquid fragmentation, thereby changing the spatial distribution morphology of the fluid and affecting its relative permeability, and these changes in turn can influence the particle transport during two-phase flow. Additionally, a critical threshold of 0.4 for the proportion of particle clusters is found, above which the fragmentation effect of wetting clusters becomes significant. Taken together, these findings underscore the profound effects of particle behaviors on two-phase flow dynamics, which have significant implications for the prediction of particle transport and fate in subsurface environments and can potentially be exploited to enhance the efficiency of contaminant remediation and energy recovery.

Data Availability Statement

Experimental images used to generate the 3D reconstruction figures and data sets, including the size distribution of wetting fluid clusters, the size distribution of particle clusters, as well as relative permeability of the nonwetting phase are available at the Zenodo repository (Wu & Yang, 2024).

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