

Pore-Scale Imaging to Quantify the Evolution and Reduction in Trapped CO₂ due to Ostwald Ripening

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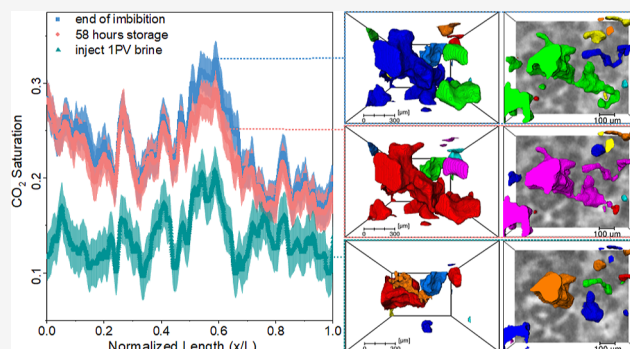
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Supporting Information

ABSTRACT: Geological carbon storage is a key strategy for mitigating climate change, but the long-term stability of trapped CO₂ remains uncertain. Transport of dissolved CO₂ in the aqueous phase can cause the rearrangement of capillary-trapped CO₂ in the pore space, which is called Ostwald ripening. Using high-resolution three-dimensional X-ray imaging, we visualized the in situ evolution of CO₂ ganglia in reservoir sandstone during storage and quantified its impact on trapped CO₂ saturation. Pore-scale imaging showed the concurrent shrinkage and growth of CO₂ ganglia, reduced morphological complexity, and enhanced connectivity, resulting from Ostwald ripening. Ganglia exhibited a size-dependent response: small ganglia dissolved and disappeared, intermediate ones shrank or grew, and large ganglia stabilized with occasional fragmentation. After waiting for 58 h with no flow, originally residual CO₂ reconnected, and subsequent brine injection led to a decrease in saturation from 22.8% to 15.6%, consistent with previous estimates based on pore-scale modeling. This work suggests that measurements that ignore the effect of Ostwald ripening overestimate the residual saturation by a factor of approximately a third.

KEYWORDS: geological CO₂ storage, pore-scale imaging, reservoir sandstone, Ostwald ripening, ganglia rearrangement, trapped saturation reduction



1. INTRODUCTION

Geological CO₂ storage is a vital technology for mitigating greenhouse gas emissions and achieving global net-zero targets.^{1–3} Successful implementation of CO₂ sequestration and long-term storage stability critically depends on a thorough understanding of CO₂ flow and trapping mechanisms. In recent years, significant progress has been made in characterizing CO₂ multiphase flow dynamics,^{1,4} as well as in elucidating the impacts of reservoir heterogeneity⁵ and geochemical reactions.^{6,7} However, the dynamics of stored CO₂ following injection, particularly during trapping, remain poorly understood. This limitation largely stems from the fact that much of the current knowledge of multiphase flow in porous media is derived from experience with hydrocarbon recovery,⁸ where it is assumed that oil and water are completely immiscible, and hence once oil is surrounded by brine in the pore space, it remains trapped.^{9,10} This capillary trapping process has also been observed for CO₂,^{11,12} however, in early studies, the rearrangement and reconnection of trapped ganglia mediated by transport of dissolved CO₂ in the aqueous phase, called Ostwald ripening, was ignored.

Multiple experiments^{13–19} and numerical modeling^{20–23} have recently investigated Ostwald ripening, demonstrating

that it can drive substantial mass redistribution in porous media. X-ray computed tomography (CT) imaging studies by Garing et al.,¹³ Zhang et al.,¹⁵ Goodarzi et al.,^{14,16} and Boon et al.,¹⁷ along with microfluidic observations by Xu et al.,²⁴ have revealed that gas ganglia can enlarge and become more connected. Pore-scale modeling by Singh et al.^{21,22} further showed that larger ganglia become increasingly ramified at the expense of smaller, spherical ones. Similar phenomena, including shrinkage and growth of trapped bubbles or foams, were observed in micromodel experiments by Huang et al.,²⁵ Xu et al.,¹⁸ Wang et al.,²⁶ and Feng et al.¹⁹ Joewondo et al.²⁷ combined microfluidic experiments with pore-network modeling to show that larger bubbles exhibited continuous growth, while smaller bubbles initially grew but eventually dissolved. Adebimpe et al.²³ used a percolation-based pore-network model to predict the impact of Ostwald ripening on residual

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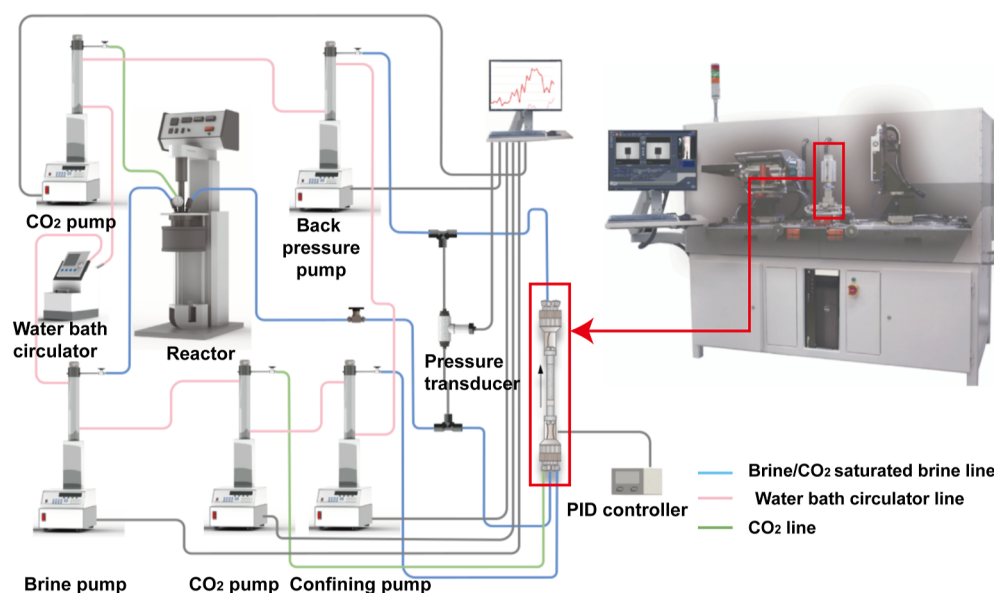


Figure 1. Schematic diagram of the experimental apparatus.

gas trapping. Their work assumed that local capillary equilibrium is achieved at the scale of a few pores, which typically takes on the order of days to weeks for most gases.²⁸ Once equilibrium is established, further displacement of the initially trapped gas becomes possible, ultimately leading to lower residual saturation. Although previous studies suggest that Ostwald ripening rearranges trapped ganglia and may reduce gas trapping, direct experimental confirmation is lacking. Moreover, existing investigations have primarily focused on ganglia size evolution, typically reporting only statistical trends, while the detailed dynamics of ganglion transformation remains poorly understood. In addition, most prior experiments were conducted in micromodels or homogeneous quarry sandstones, which do not capture the geological complexity of actual or planned CO₂ storage reservoirs.

This study visualizes the *in situ* Ostwald ripening of CO₂ ganglia in reservoir sandstone and quantitatively evaluates its impact on residual saturation using high-resolution three-dimensional X-ray imaging during controlled displacement experiments. Uniquely, we tracked the individual evolution of ganglia over time, providing a detailed pore-scale characterization of the ganglion dynamics during Ostwald ripening. Our observations enable refinement of multiphase flow properties used in field-scale modeling, ensuring that the long-term impacts of Ostwald ripening are properly incorporated into CO₂ storage predictions.

2. MATERIALS AND METHODS

2.1. Materials. A cylindrical sandstone sample, obtained from a depleted gas reservoir that is a proposed site for CO₂ storage, was used for the experiment. The sample measured 6.0 ± 0.1 mm in diameter and 23.7 ± 0.1 mm in length with a porosity of 25.4% and an absolute permeability of 200 ± 10 mD. Its mineralogical composition consisted of 73.6% quartz, 16.6% plagioclase, 2.8% calcite, 0.2% siderite, and 6.8% clay. To enhance X-ray attenuation contrast among the brine, CO₂, and rock phases during micro-CT scanning and facilitate subsequent image processing and analysis, a 25 wt % potassium iodide (KI)-doped brine^{7,29,30} was used in all the experiments.

Although the addition of KI may influence physicochemical properties, including CO₂ solubility and mass transfer rates, the effect on the patterns of Ostwald ripening of CO₂ ganglia, the primary focus of this work, is unlikely to be significant.

2.2. Methods. Figure 1 illustrates the experimental apparatus. The system comprised five pumps—for brine equilibration and injection, CO₂ injection, confining pressure, and back pressure regulation—along with a high-pressure reactor, a core holder, and a pressure transducer, all interconnected via polyether-ether-ketone (PEEK) tubing. The core holder's temperature was regulated by a proportional-integral-derivative (PID) controller that drove a heater jacket with thermocouple feedback, while the temperatures of the pumps and tubing were maintained using a recirculating water bath. X-ray micro-CT images were acquired by using a Zeiss Xradia 510 Versa X-ray microscope.

Residual sandstone, remaining after drilling the cylindrical sample, was fragmented into small pieces and placed in a high-pressure reactor. 600 mL of 25 wt % KI-doped brine was added, and the reactor was sealed, heated to 50 °C, and pressurized with CO₂ to 8 MPa for 2 weeks to achieve thermodynamic equilibrium between the brine, CO₂, and rock phases, yielding CO₂-saturated brine and brine-saturated CO₂. The cylindrical sample was dried in an oven at 60 °C for 2 days and then enclosed in a Viton sleeve and mounted into a core holder. The assembled core holder was then installed vertically in the CT scanner and connected to the fluid delivery and receiving system, the pressure transducer, and other experimental components. For the small sample used, gravitational effects are likely to be negligible, as confirmed by detailed calculations in Text S1 of the Supporting Information. A confining pressure of 2 MPa was applied to ensure proper sealing and eliminate the bypass flow between the sample and sleeve. CO₂ was injected for 30 min to flush residual fluids and fines, followed by 12 h of vacuuming to evacuate remaining gases. A dry scan of the sample was first performed. Brine was injected at a flow rate of 0.20 mL/min while the system was gradually brought to reservoir conditions (8 MPa and 50 °C), followed by a scan after the system pressure had stabilized for over 2 h. Subsequently, CO₂ was injected at the same flow rate

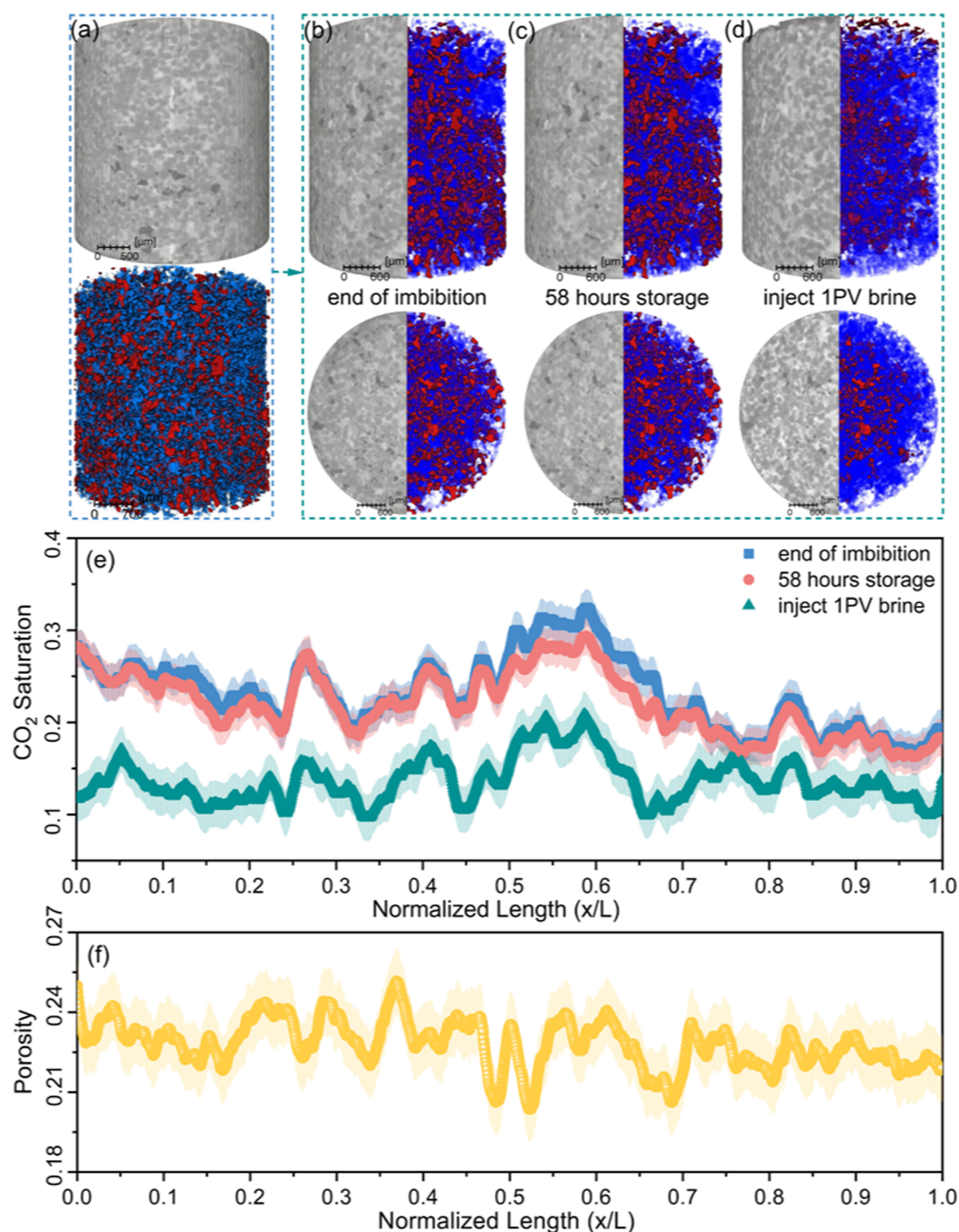


Figure 2. Effect of Ostwald ripening on trapped CO₂ saturation. (a) Schematic illustration of the procedure used to generate three-dimensional visualizations and cross-sectional images of both raw (bright: brine; gray: sandstone; dark: CO₂), (b) at the end of imbibition, (c) after 58 h of storage, and (d) after injecting one pore volume of brine. (e) CO₂ saturation profile along the sample. (f) Porosity distribution along the sample. The shaded regions in (e) and (f) represent uncertainties associated with image segmentation.

until the pressure restabilized and remained steady for at least 2 h. Following this drainage step, brine was reinjected at 0.20 mL/min, and a scan was acquired after pressure stabilization for 2 h. To investigate Ostwald ripening, the system was then held under static conditions (constant pressure, no flow) for 58 h to allow trapped CO₂ ganglia to equilibrate via diffusion-driven processes, followed by another scan. The waiting period was based on estimates of the time for Ostwald ripening to have a significant effect throughout the sample, as detailed in

Text S2 of the [Supporting Information](#). While changes in pressure as the system reaches equilibrium could change the balance of dissolved and free-phase CO₂, this effect is likely to be small. Finally, 1 pore volume of brine was injected to displace any mobilized CO₂ resulting from Ostwald ripening, and a final scan was performed. For each stage, a scan of the whole sample (voxel size: 4.5 μm) was conducted to quantify CO₂ saturation, accompanied by a higher-resolution, zoomed-in image (voxel size: 1.82 μm) focusing on the central region

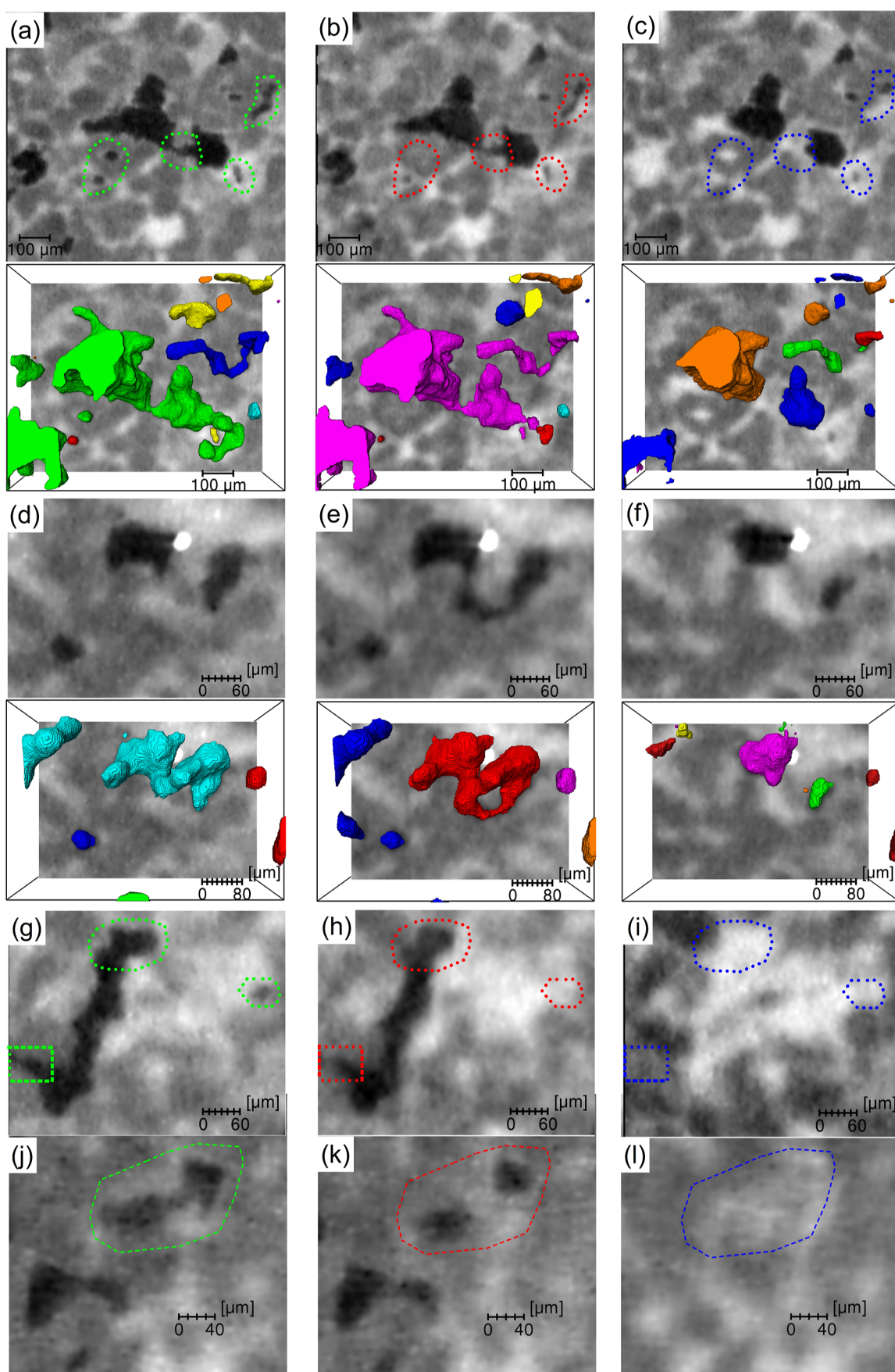


Figure 3. continued

Figure 3. Visualization of CO₂ ganglia evolution at different stages. Two-dimensional images (black represents CO₂, gray denotes rock, and white indicates brine) and the corresponding three-dimensional renderings (with individual CO₂ ganglia shown in distinct colors) acquired at the end of imbibition (a,d,g,j), after 58 h of storage (b,e,h,k), and following the injection of one pore volume of brine (c,f,i,l).

of the sample to characterize the size, morphology, and connectivity of trapped ganglia. The capillary number was 6.6×10^{-8} , based on the definition $Ca = \frac{\mu q}{\sigma}$,^{7,8} where μ is the viscosity of the CO₂, q is the Darcy velocity, and σ is the interfacial tension between the two phases; this value remained in the capillary-dominated regime, similar to reservoir conditions.

Imaging was conducted at 90 keV X-ray energy and 8 W power. Each scan consisted of 3001 projections with a 2 s exposure time and 1× binning. Images were reconstructed using Zeiss Reconstructor software with corrections applied for center shift and beam hardening. Following reconstruction, the image processing and analysis were performed using commercial image analysis software, Avizo. Specifically, images were denoised using a nonlocal means filter, normalized to grayscale, and spatially registered to the dry scan using Lanczos resampling. Phase segmentation was conducted by combining differential imaging with a marker-based watershed method.⁷ In this process, the multiphase image was subtracted from the fully brine-saturated image to highlight CO₂, while the subtraction of the dry scan from the multiphase image enhanced the brine phase. Both CO₂ and brine were then segmented using the watershed algorithm, and the remaining voxels were classified as sandstone minerals. Finally, quantitative analysis of CO₂ saturation and CO₂ ganglia properties, including volume, surface area, and Euler characteristics, was performed.

Additionally, we tracked the temporal evolution of individual CO₂ ganglion volumes to characterize Ostwald ripening at the pore scale. First, all discrete CO₂ regions were identified in each three-dimensional image using connected-component labeling with each ganglion assigned a unique identifier. Ganglia smaller than 27 voxels were excluded from further analysis since they may be segmentation artifacts. To track ganglia over time, spatial correspondence between consecutive images was established by evaluating voxel-wise overlaps. A ganglion was classified as complete dissolution if it had no overlap in the subsequent image. If a ganglion mapped to a single ganglion in the next image and the mapping was mutual, it was considered stable if its volume changed by less than 1% or was categorized as growth or shrinkage depending on whether the volume increased or decreased. If a ganglion split into multiple parts, the event was classified as fragmentation; if multiple ganglia merged into one, it was classified as coalescence. Ganglia that newly appeared without any overlap in the previous image were defined as nucleation growth.

3. RESULTS AND DISCUSSION

3.1. Reduction in Trapped CO₂ Saturation. The raw and segmented images at different stages, along with the CO₂ saturation profile and porosity distribution, are shown in Figure 2. As illustrated in Figure 2a–d, after several hundred pore volumes of brine flushing, CO₂, as a nonwetting phase in this reservoir sandstone,⁷ was immobilized as discrete ganglia within relatively large pores. The average trapped CO₂

saturation was 22.8%, as depicted in Figure 2e. This residual trapping results from capillary forces that induce snap-off at restrictions in the pore space, called throats, disconnecting and immobilizing the nonwetting CO₂ in pore bodies^{11,31,32} and preventing its further mobilization under typical reservoir conditions. Furthermore, CO₂ saturation qualitatively correlated with local porosity, with higher-porosity regions generally retaining greater amounts of CO₂ (Figures 2e and 2f). This correlation stems from capillary pressure scaling inversely with throat radius:⁸ larger throats enable easier CO₂ invasion into associated larger pores during drainage, followed by preferential residual trapping during subsequent imbibition.³³

After 58 h of storage, the trapped CO₂ saturation remained stable, suggesting that Ostwald ripening has a minimal impact on overall saturation. This diffusion-driven process redistributes mass via curvature-driven solubility gradients while conserving total volume.^{14,16,18} Following the subsequent injection of up to 1 pore volume of brine, the trapped CO₂ saturation decreased significantly to 15.6%. This reduction is likely due to the rearrangement of the CO₂ ganglia during storage—i.e., Ostwald ripening that reconnects a fraction of the initially trapped CO₂. Similar effects of Ostwald ripening in enhancing H₂ withdrawal have been reported by Goodarzi et al.^{14,16} and Zhang et al.¹⁵ The reduction in residual saturation is consistent with pore-scale modeling that predicted a 20–25% reduction in residual saturation due to Ostwald ripening;²³ we see a larger reduction of 32%, which may be caused by some dissolution of CO₂ during the final brine injection.

3.2. Qualitative Characterization of CO₂ Ganglia Evolution. Figure 3 presents two- and three-dimensional visualizations of the CO₂ ganglia at different stages. At the end of imbibition (Figures 3a, 3d, 3g, and 3j), the trapped CO₂ ganglia exhibit a wide range of sizes and morphologies, consistent with observations by Andrew et al.¹¹ and Iglaue et al.³² During Ostwald ripening, significant changes occur in ganglia size, morphology, and connectivity. Ganglia with high interfacial curvature—particularly small, isolated ones—tend to shrink, while neighboring ganglia with lower curvature expand, often accompanied by coalescence and enhanced overall connectivity (Figures 3b and 3e). However, such expansion and coalescence in the complex pore networks are not unrestricted. As ganglia grow, they invade adjacent throats, increasing local interfacial curvature and triggering localized dissolution at these constrictions, followed by mass transfer to nearby ganglia (Figures 3g and 3h). Additionally, ganglia fragmentation is also observed, likely due to mass-transfer-induced snap-off, which reduces the ganglion volume and weakens connectivity (Figures 3j and 3k). Following 1 pore volume brine injection, the CO₂ ganglia—initially trapped by capillary forces, then rearranged and reconnected via Ostwald ripening—are ultimately displaced, as shown in Figures 3c, 3f, 3i, and 3l.

The ganglia rearranged by mass transport of CO₂ dissolved in the brine.^{28,34} This process aims to make the local capillary pressure constant by promoting the dissolution of high-

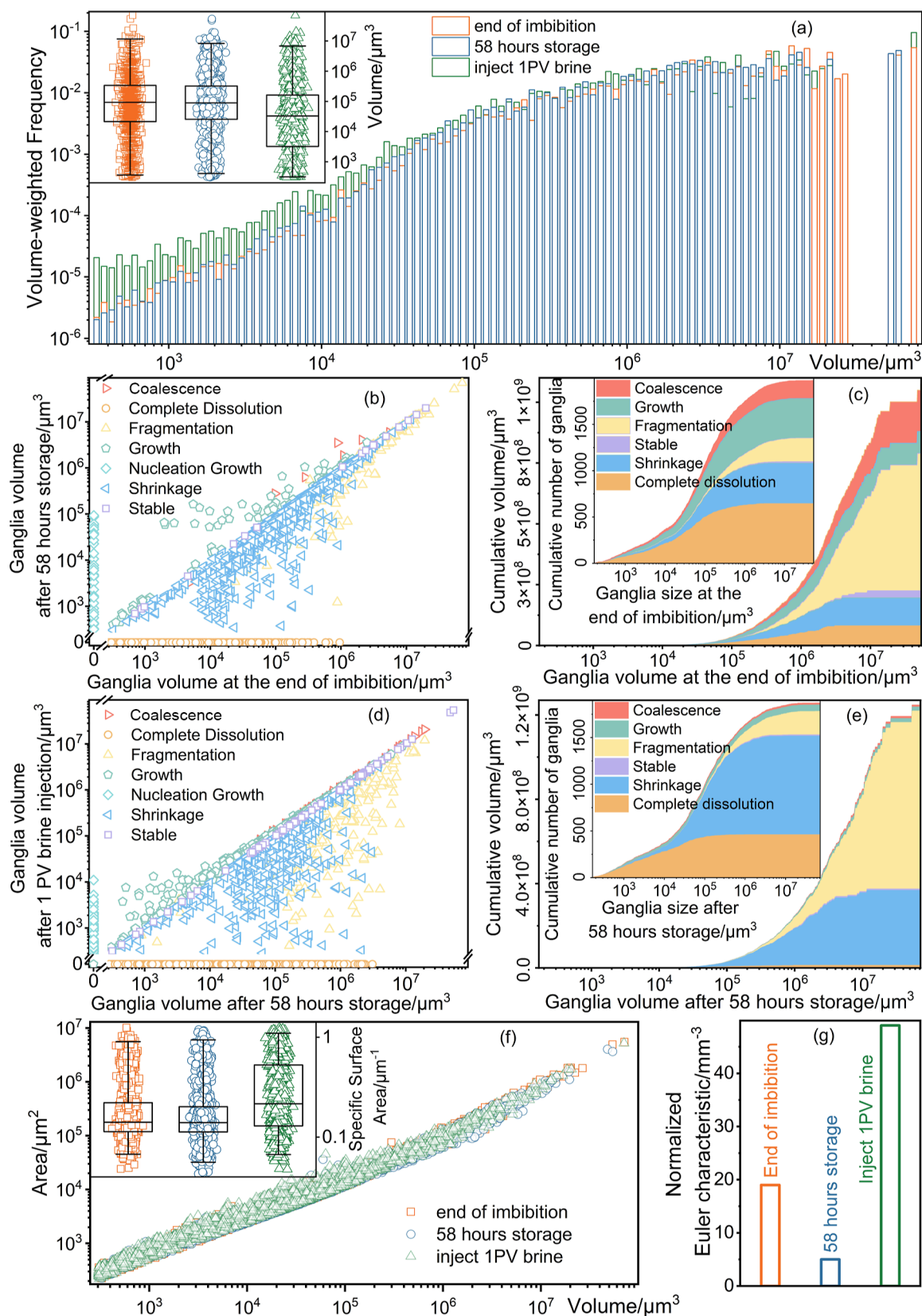


Figure 4. Statistical analysis of CO₂ ganglia evolution at different stages. (a) Statistical analysis of CO₂ ganglia volume at the end of imbibition, after 58 h of storage, and following the injection of one pore volume of brine. (b,d) Evolution of ganglion volumes between stages and (c,e) corresponding statistical analyses of cumulative volume and ganglion count relative to the end of imbibition and post-storage, respectively, excluding nucleation growth. (f) Specific surface area of CO₂ ganglia at different stages. (g) Normalized Euler characteristic^{14,16} of CO₂ ganglia at different stages.

curvature ganglia and the growth of low-curvature ones.³⁵ Higher interfacial curvature elevates CO₂ chemical potential via increased Gibbs surface excess energy from the Laplace capillary pressure,³⁶ thereby enhancing equilibrium solubility and local dissolved CO₂ concentrations in the brine; lower curvature has the opposite effects. This disparity creates concentration gradients that drive diffusive mass transfer through the aqueous phase from high- to low-curvature regions, ultimately resulting in ganglion shrinkage, expansion, coalescence, and fragmentation as the system evolves toward equilibrium.

3.3. Quantification of CO₂ Ganglia Evolution. Figure 4 quantifies the evolution of the CO₂ ganglia at different stages. As shown in Figure 4a, at the end of imbibition, ganglia exhibit a broad size distribution, with the largest volume reaching $7.17 \times 10^7 \mu\text{m}^3$. Over 58 h of storage, corresponding to the Ostwald ripening process, the proportion of ganglia with volumes less than $2.2 \times 10^3 \mu\text{m}^3$ and greater than $8.5 \times 10^6 \mu\text{m}^3$ decreases, while the fraction within this intermediate range increases. Specifically, a clear size-dependent behavior is observed during Ostwald ripening in Figures 4b and 4c: small ganglia tend to dissolve and disappear; intermediate ganglia either shrink or grow; and large ganglia remain relatively stable, though occasional fragmentation occurs. Subsequent injection of 1 pore volume of brine mobilizes and displaces the rearranged, large, and connected ganglia, while small isolated ones remain trapped and occupy a growing proportion of residual CO₂. As shown in Figures 4d and 4e, brine displacement not only induced complete dissolution, growth, shrinkage, and coalescence of CO₂ ganglia but also significantly enhanced the fragmentation of large ganglia, likely driven by flow-induced snap-off.^{37,38}

The specific surface area, defined as the ratio of a ganglion's surface area to its volume, serves as a quantitative descriptor of ganglion morphology. Lower values correspond to more regular, spherical shapes with reduced morphological complexity.³⁹ As shown in Figure 4f, the average specific surface area of ganglia decreases during Ostwald ripening. This trend reflects the progressive elimination of morphologically complex ganglia and a transition toward more spherical, energetically favorable configurations resulting from ganglion coarsening and rearrangement. During subsequent brine injection, ganglia with more regular shapes are preferentially mobilized and displaced. In contrast, those with higher specific surface areas—indicative of irregular or elongated morphologies—are more likely to remain trapped within the pore space.

The Euler characteristic^{26,40} quantifies fluid connectivity in porous media, with lower values indicating enhanced connectivity. As indicated in Figure 4g, initially, the normalized Euler characteristic of the CO₂ ganglia was measured at 19 mm^{-3} , indicating trapped ganglia. Following Ostwald ripening, this value decreased to 5 mm^{-3} , signifying better ganglion connectivity over time. This transition is likely driven by the dissolution and disappearance of small, isolated ganglia as well as the coalescence of neighboring ganglia through the transport of dissolved CO₂, as illustrated in Figure 3. A comparable increase in connectivity has been observed in H₂ systems undergoing Ostwald ripening.^{14–16} During subsequent brine injection, these interconnected ganglia were preferentially displaced, leaving behind isolated ganglia with a high normalized Euler characteristic of 35 mm^{-3} .

Transport of dissolved CO₂ in the aqueous phase, Ostwald ripening, can drive the rearrangement of capillary-trapped CO₂

in the pore space and potentially reduce residual saturation. High-resolution 3D X-ray imaging was employed to investigate this in situ process at the pore scale. The experiments demonstrated that the rearrangement of the CO₂ ganglia during Ostwald ripening reconnects previously trapped CO₂, reducing its saturation from 22.8% to 15.6%. The morphological evolution of CO₂ ganglia is driven by simultaneous growth and dissolution, accompanied by reductions in morphological complexity, increases in sphericity, and enhanced connectivity. Ganglion size-dependent behavior is evident: small ganglia dissolve and disappear; intermediate ones exhibit either growth or shrinkage, while large ganglia remain largely stable, with occasional fragmentation. Our study provides the first quantitative analysis of Ostwald ripening's effect on CO₂ trapping efficiency, offering in situ insights into the evolution of CO₂ ganglia. These findings align with those of Xu et al.¹⁸ and Feng et al.,¹⁹ which emphasize the critical role of Ostwald ripening in porous media. While our imaging and quantitative analysis captured these Ostwald ripening dynamics, we did not identify a definitive threshold for interganglion spacing that universally triggers or accelerates ripening. This limitation arises from the complexity of natural porous media, where ripening transcends simple distance metrics and is influenced by several interdependent factors: heterogeneous pore structures that create tortuous diffusion pathways, variable capillary barriers that modulate brine connectivity, and diverse ganglion sizes that shape local concentration gradients. Consequently, Ostwald ripening proceeded continuously across the observed separations, with even remotely positioned small ganglia dissolving via viable pathways, underscoring the process's inherently probabilistic and system-dependent nature rather than a simplistic binary threshold.

While this work was conducted at the scale of a few millimeters, it does have implications for field-scale displacement. Since Ostwald ripening is a diffusive process, reaching equilibrium through transport in the aqueous phase alone at meter scales or larger is extremely slow,^{20,35} taking thousands to many millions of years. However, what our work shows is that over a few days, trapped gas ganglia can reconnect.^{14,15} This means that gas can be transported in its own phase through advective flow, which is potentially much more rapid.^{28,41} Our work suggests that initially trapped gas can begin to flow and that there can be further displacement with field-scale consequences. In large-scale modeling, the end result is simply a reduction in local residual saturation: in our work, we see a reduction of approximately one-third compared to measured values with immiscible fluids or from experiments done quickly so that Ostwald ripening has not had sufficient time to reconnect some of the gas. In addition, there is a need to correct relative permeabilities and capillary pressures measured too rapidly to account for Ostwald ripening to better represent field conditions where equilibrium is likely to be reached, at least over length scales sufficient to affect advective transport. Incorporating Ostwald ripening through longer experimental durations is an option. In addition, the approach to use could follow a simple rescaling of the saturation dependence, using the adjusted lowered residual saturation as suggested by pore-scale modeling.²³

Building on this research on the Ostwald ripening mechanisms, future research will examine the effects of temperature, pressure, salinity, and heterogeneity on the ripening rates in porous media. Temperature effects may be

competing: higher temperatures could enhance molecular diffusion rates and accelerate ripening, yet reduce CO₂ solubility,⁴² diminishing chemical potential gradients and potentially slowing kinetics, with net outcomes dependent on the exact conditions. Similarly, elevated pressures could enhance CO₂ solubility via higher partial pressures, amplifying concentration gradients and diffusive fluxes, but could concurrently suppress diffusion rates and retard the process. Salinity gradients, increasing with depth in formations, may reduce CO₂ solubility via the salting-out effect,⁴³ weakening mass transfer gradients and slowing ripening. Heterogeneity might accelerate ripening in high-connectivity zones while slowing it in low-permeability regions, yielding uneven CO₂ distributions; additionally, capillary entry pressure may hinder interblock mass transfer, and end effects could enhance local trapping at block boundaries, influencing dissolution and growth dynamics. Moreover, in larger-scale heterogeneous media, a likely scenario is that CO₂ ganglia reconnect via Ostwald ripening, enabling advective flow in their own phase—a process far faster than mass diffusion.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.est.5c06424>.

Sample orientation and length effects on Ostwald ripening: quantitative analysis of gravitational forces and comparison with existing literature; and determination of experimental time scales: time scale of Ostwald ripening and justification of shut-in time (PDF)

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Notes

The authors declare no competing financial interest.

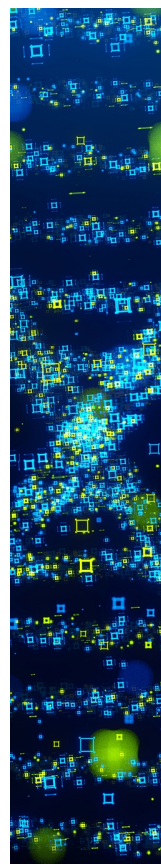
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■ REFERENCES

- (1) Shen, T.; Miller, Q. R. S.; Lahiri, N.; Terry, O.; Owen, A. T.; Kou, Z.; Schaef, H. T.; Kelly, S. A. Multiphase flow regime controls carbonate precipitation morphologies during CO₂ injection in subsurface basalts. *Environ. Sci. Technol.* **2025**, *59*, 20345–20356.
- (2) Shukla, P. R.; Skea, J.; Slade, R.; Al Khourdajie, A.; van Diemen, R.; McCollum, D.; Pathak, M.; Some, S.; Vyas, P.; Fradera, R.; Belkacemi, M.; Hasija, A.; Lisboa, G.; Luz, S.; Malley, J. *Climate Change 2022: Mitigation of Climate Change. Contribution of Working Group III to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change*; Cambridge University Press: Cambridge, UK and New York, NY, USA, 2022.
- (3) Shokri, J.; Ruf, M.; Lee, D.; Mohammadrezaei, S.; Steeb, H.; Niasar, V. Exploring carbonate rock dissolution dynamics and the influence of rock mineralogy in CO₂ injection. *Environ. Sci. Technol.* **2024**, *58*, 2728–2738.
- (4) Ni, H.; Li, B.; Darraj, N.; Ren, B.; Harris, C.; Krishnamurthy, P. G.; Bakar, I.; Berg, S.; Snippe, J.; Ringrose, P.; Meckel, T.; Krevor, S.; Benson, S. The impact of capillary heterogeneity on CO₂ flow and trapping across scales. *Earth-Sci. Rev.* **2025**, *270*, 105257.
- (5) Reynolds, C. A.; Blunt, M. J.; Krevor, S. Multiphase flow characteristics of heterogeneous rocks from CO₂ storage reservoirs in the United Kingdom. *Water Resour. Res.* **2018**, *54*, 729–745.
- (6) Seyyedi, M.; Giwelli, A.; White, C.; Esteban, L.; Verrall, M.; Clennell, B. Effects of geochemical reactions on multi-phase flow in porous media during CO₂ injection. *Fuel* **2020**, *269*, 117421.
- (7) Chai, R.; Ma, Q.; Goodarzi, S.; Yow, F. Y.; Bijeljic, B.; Blunt, M. J. Multiphase reactive flow during CO₂ storage in sandstone. *Engineering* **2025**, *48*, 81–91.
- (8) Blunt, M. J. *Multiphase Flow in Permeable Media: A Pore-Scale Perspective*; Cambridge University Press, 2017.
- (9) AlRatrou, A.; Blunt, M. J.; Bijeljic, B. Wettability in complex porous materials, the mixed-wet state, and its relationship to surface roughness. *Proc. Natl. Acad. Sci. U.S.A.* **2018**, *115*, 8901–8906.
- (10) Chai, X.; Tian, L.; Zhu, Y.; Ding, H.; Zhang, G. Dynamic diffusion processes of miscible CO₂ geological utilization with complex microscopic pore-throat structures. *Sep. Purif. Technol.* **2025**, *362*, 131578.
- (11) Andrew, M.; Bijeljic, B.; Blunt, M. J. Pore-scale imaging of trapped supercritical carbon dioxide in sandstones and carbonates. *Int. J. Greenhouse Gas Control* **2014**, *22*, 1–14.
- (12) Kimbrel, E. J. H.; Wildenschild, D.; Herring, A. L.; Armstrong, R. T. The effect of original and initial saturation on residual nonwetting phase capillary trapping efficiency. *Int. J. Greenhouse Gas Control* **2022**, *120*, 103758.
- (13) Garing, C.; de Chalendar, J. A.; Voltolini, M.; Ajo-Franklin, J. B.; Benson, S. M. Pore-scale capillary pressure analysis using multi-scale X-ray micromotography. *Adv. Water Resour.* **2017**, *104*, 223–241.
- (14) Goodarzi, S.; Zhang, Y.; Foroughi, S.; Bijeljic, B.; Blunt, M. J. Trapping, hysteresis and Ostwald ripening in hydrogen storage: A pore-scale imaging study. *Int. J. Hydrogen Energy* **2024**, *56*, 1139–1151.
- (15) Zhang, Y.; Bijeljic, B.; Gao, Y.; Goodarzi, S.; Foroughi, S.; Blunt, M. J. Pore-scale observations of hydrogen trapping and migration in porous rock: Demonstrating the effect of Ostwald ripening. *Geophys. Res. Lett.* **2023**, *50*, No. e2022GL102383.
- (16) Goodarzi, S.; Zhang, G.; Bijeljic, B.; Blunt, M. J. Ostwald ripening leads to less hysteresis during hydrogen injection and withdrawal: A pore-scale imaging study. *Int. J. Hydrogen Energy* **2025**, *114*, 475–485.

- (17) Boon, M.; Rademaker, T.; Winardhi, C. W.; Hajibeygi, H. Multiscale experimental study of H_2 /brine multiphase flow in porous rock characterizing relative permeability hysteresis, hydrogen dissolution, and Ostwald ripening. *Sci. Rep.* **2024**, *14*, 30170.
- (18) Xu, K.; Bonnacaze, R.; Balhoff, M. Egalitarianism among bubbles in porous media: An Ostwald ripening derived anticoarsening phenomenon. *Phys. Rev. Lett.* **2017**, *119*, 264502.
- (19) Feng, Z.; Zhang, M.; Wang, J.; Zhan, M.; Xu, X.; Xu, P. Ripening of capillary-trapped ganglia in porous media. *Exp. Therm. Fluid Sci.* **2025**, *163*, 111364.
- (20) Li, Y.; Garing, C.; Benson, S. M. A continuum-scale representation of Ostwald ripening in heterogeneous porous media. *J. Fluid Mech.* **2020**, *889*, A14.
- (21) Singh, D.; Friis, H. A.; Jettstuen, E.; Helland, J. O. A level set approach to Ostwald ripening of trapped gas bubbles in porous media. *Transp. Porous Media* **2022**, *145*, 441–474.
- (22) Singh, D.; Friis, H. A.; Jettstuen, E.; Helland, J. O. Ostwald ripening of gas bubbles in porous media: Impact of pore geometry and spatial bubble distribution. *Adv. Water Resour.* **2024**, *187*, 104688.
- (23) Adebimpe, A. I.; Foroughi, S.; Bijeljic, B.; Blunt, M. J. Percolation without trapping: How Ostwald ripening during two-phase displacement in porous media alters capillary pressure and relative permeability. *Phys. Rev. E* **2024**, *110*, 035105.
- (24) Xu, R.; Li, R.; Huang, F.; Jiang, P. Pore-scale visualization on a depressurization-induced CO_2 exsolution. *Sci. Bull.* **2017**, *62*, 795–803.
- (25) Huang, Z.; Su, M.; Yang, Q.; Li, Z.; Chen, S.; Li, Y.; Zhou, X.; Li, F.; Song, Y. A general patterning approach by manipulating the evolution of two-dimensional liquid foams. *Nat. Commun.* **2017**, *8*, 14110.
- (26) Wang, C.; Mehmani, Y.; Xu, K. Capillary equilibrium of bubbles in porous media. *Proc. Natl. Acad. Sci. U.S.A.* **2021**, *118*, No. e2024069118.
- (27) Joewondo, N.; Garbin, V.; Pini, R. Experimental evidence of the effect of solute concentration on the collective evolution of bubbles in a regular pore-network. *Chem. Eng. Res. Des.* **2023**, *192*, 82–90.
- (28) Blunt, M. J. Ostwald ripening and gravitational equilibrium: Implications for long-term subsurface gas storage. *Phys. Rev. E* **2022**, *106*, 045103.
- (29) Lin, Q.; Bijeljic, B.; Raeini, A. Q.; Rieke, H.; Blunt, M. J. Drainage capillary pressure distribution and fluid displacement in a heterogeneous laminated sandstone. *Geophys. Res. Lett.* **2021**, *48*, No. e2021GL093604.
- (30) Wang, S.; Spurin, C.; Bultreys, T. Pore-scale imaging of multiphase flow fluctuations in continuum-scale samples. *Water Resour. Res.* **2023**, *59*, No. e2023WR034720.
- (31) Lv, P.; Chang, Y.; Liu, F.; Liu, Y.; Wang, B.; Guo, X.; Song, Y. CO_2 -brine mass transfer patterns and interface dynamics under geological storage conditions. *Int. J. Heat Mass Transfer* **2024**, *222*, 125184.
- (32) Iglauer, S.; Paluszny, A.; Pentland, C. H.; Blunt, M. J. Residual CO_2 imaged with X-ray micro-tomography. *Geophys. Res. Lett.* **2011**, *38*, L21403.
- (33) Pentland, C. H.; El-Maghraby, R.; Iglauer, S.; Blunt, M. J. Measurements of the capillary trapping of super-critical carbon dioxide in Berea sandstone. *Geophys. Res. Lett.* **2011**, *38*, L06401.
- (34) Johnson, M.; Peakall, J.; Fairweather, M.; Barnes, M.; Davison, S.; Jia, X.; Clare, M. A.; Harbottle, D.; Hunter, T. N. Sediment microstructure and the establishment of gas migration pathways during bubble growth. *Environ. Sci. Technol.* **2019**, *53*, 12882–12892.
- (35) de Chalendar, J. A.; Garing, C.; Benson, S. M. Pore-scale modelling of Ostwald ripening. *J. Fluid Mech.* **2018**, *835*, 363–392.
- (36) Voorhees, P. W. The theory of Ostwald ripening. *J. Stat. Phys.* **1985**, *38*, 231–252.
- (37) Andrew, M.; Bijeljic, B.; Blunt, M. J. Pore-by-pore capillary pressure measurements using X-ray microtomography at reservoir conditions: Curvature, snap-off, and remobilization of residual CO_2 . *Water Resour. Res.* **2014**, *50*, 8760–8774.
- (38) Armstrong, R. T.; McClure, J. E.; Berrill, M. A.; Rücker, M.; Schlüter, S.; Berg, S. Beyond Darcy's law: The role of phase topology and ganglion dynamics for two-fluid flow. *Phys. Rev. E* **2016**, *94*, 043113.
- (39) Maroof, M. A.; Mahboubi, A.; Noorzad, A. A new method to determine specific surface area and shape coefficient of a cohesionless granular medium. *Adv. Powder Technol.* **2020**, *31*, 3038–3049.
- (40) Herring, A. L.; Robins, V.; Sheppard, A. P. Topological persistence for relating microstructure and capillary fluid trapping in sandstones. *Water Resour. Res.* **2019**, *55*, 555–573.
- (41) Xu, K.; Mehmani, Y.; Shang, L.; Xiong, Q. Gravity-induced bubble ripening in porous media and its impact on capillary trapping stability. *Geophys. Res. Lett.* **2019**, *46*, 13804–13813.
- (42) Lara Cruz, J.; Neyrolles, E.; Contamine, F.; Cézac, P. Experimental study of carbon dioxide solubility in sodium chloride and calcium chloride brines at 333.15 and 453.15 K for pressures up to 40 MPa. *J. Chem. Eng. Data* **2021**, *66*, 249–261.
- (43) Liu, Y.; Hou, M.; Yang, G.; Han, B. Solubility of CO_2 in aqueous solutions of NaCl, KCl, $CaCl_2$ and their mixed salts at different temperatures and pressures. *J. Supercrit. Fluids* **2011**, *56*, 125–129.



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