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# Local instabilities during capillary-dominated immiscible displacement in porous media

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**Abstract:**
Fully understanding the mechanism of pore-scale immiscible displacement dominated by capillary forces, especially local instabilities and their influence on flow patterns, is essential for various industrial and environmental applications such as enhanced oil recovery, $CO_2$ geo-sequestration and remediation of contaminated aquifers. It is well known that such immiscible displacement is extremely sensitive to the fluid properties and pore structure, especially the wetting properties of the porous medium which affect not only local interfacial instabilities at the micro-scale, but also displacement patterns at the macro-scale. In this review, local interfacial instabilities under three typical wetting conditions, namely Haines jump events during weakly-wetting drainage, snap-off events during strongly-wetting imbibition, and the co-existence of concave and convex interfaces under intermediate-wet condition, are reviewed to help understand the microscale physics and macroscopic consequences resulting in natural porous media.

## 1. Introduction

Capillary-dominated immiscible displacement in porous media is of great importance in many environmental and industrial applications such as $CO_2$ geological sequestration, enhanced oil recovery, and remediation of contaminated aquifers (Michael et al., 2010; Zhang et al., 2011; Chen et al., 2019). Although significant achievements have been made by pioneers through various means, such as microfluidic and synchroton X-ray imaging experiments, as well as different numerical models, the full understanding of pore-scale immiscible displacement mechanisms dominated by capillary force, especially local instabilities and their influences on flow patterns, is still a great challenge. However, it is well known that during capillary-dominated immiscible displacements, at the pore scale phase interfaces do not advance in a smooth and continuous way, but as a sequence of local pore-scale filling events, such as snap-off, piston-like displacement, corner flow, cooperative pore filling, Haines jump, or droplet fragmentation (Moebius and Or, 2012; Pak et al., 2015; Rücker et al., 2015; Hu et al., 2017; Singh et al., 2017). In addition, displacement patterns can range from a stable and compact flood front to a highly ramified front with preferential flow paths (fingers). Both displacement patterns and local pore-scale filling events in capillary-dominated immiscible displacement are strongly affected by the wettability of the pore surfaces (Rücker et al., 2019).Wettability indicates the relative tendency of immiscible fluids to adhere to a solid surface, as well as directly controls the displacement patterns and the dynamics of displacement events (Cottin et al., 2011; Holtzman and Segre, 2015; Iglauer, 2017; AlRatrout et al., 2018; Hu et al., 2018; Avendano et al., 2019). Generally, the wetting conditions of a solid surface can be classified into strongly-wet, intermediately-wet, and weakly-wet according to the contact angle between two fluids at a solid surface (Iglauer et al., 2015; Rabbani

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et al., 2017). Based on the wettability of the invading fluid, immiscible displacements in porous media can be classified into imbibition, in which a wetting fluid invades an initially non-wetting fluid filled porous matrix, or drainage, where a non-wetting invading fluid displaces a wetting fluid from the pore space (Blunt, 2017).

Thus, how to describe the effect of wettability on the displacement patterns in porous media and how to link local pore-scale filling events to the regional cooperative and/or competitive filling behavior are still poorly understood at present (Hu et al., 2018). In this work several breakthroughs made by various researchers in this area are reviewed to help understand the microscale physics and macroscopic consequences resulting in natural porous media.

## 2. Local interfacial instabilities during imbibition: Experiments

Since immiscible displacement is influenced by many factors, such as fluid properties, pore structures, and physical or chemical interactions between fluids and solids, many laboratory experiments have been carried out using various techniques, aimed at improving the understanding of basic pore-level physics and to thus identify key parameters. Those experiments can be divided into two-dimensional (2D) microfluidics experiments and three-dimensional (3D) X-ray tomography experiments.

Quasi 2D micromodel experiments have become a powerful tool for studying the fundamental physics of immiscible displacement in porous media, because they allow for dynamic and high resolution observations. In addition, porosity, pore size and connectivity of the micromodels can be readily manipulated during the fabrication process, thereby allowing for systematic studies of the influence of different pore structures (Karadimitriou and Hassanizadeh, 2012; Anbari et al., 2018). Therefore, it is particularly suitable for studying various pore filling events under strongly-wetting conditions in the imbibition process. In a recent article (Zhao et al., 2016), the powerful control of wettability on multiphase flow in porous media has been demonstrated through systematically varying the wettability of the flow cell over a wide range of contact angle values in a patterned microfluidic. Specifically, when the invading fluid perfectly wets the medium, both, pore-scale displacement mechanisms and macroscopic displacement patterns are significantly influenced by the macroscopic capillary numbers (which characterize the importance of viscous forces relative to capillary forces, also see Eq. (1) ). Thus the leading films flow along the solid surfaces at high capillary numbers ($2.9 \times 10^{-1}$), while corner flow prevails at low capillary numbers ($2.9 \times 10^{-3}$) due to capillary suction in the corners.

Furthermore, due to significant technical improvements over the last decades and its non-destructive characteristic, X-ray micro-tomography has become the foremost imaging technique for the visualization and quantification of porous structures (Blunt et al., 2013; Iglauer and Lebedev, 2018). In particular, the latest in-situ X-ray micro-tomography setup can provide 4D (i.e., transient 3D) noninvasive imaging of fluid distribution in rocks under reservoir conditions without disturbing the flow. By utilizing this visualization technique, the two most important pore-scale processes, i.e. piston-like displacement and snap-off events, under strongly-wetting conditions during capillary-dominated immiscible displacement can be observed directly and dynamically in real natural rock samples (Berg et al., 2013; Herring et al., 2014; Andrew et al., 2015; Bultreys et al., 2015; Rücker et al., 2015; Singh et al., 2017). Thus some new insights into these two processes were won as a function of (three) different local pore-space geometries and fuid configurations; this was achieved by conducting a time-resolved pore-by-pore analysis of the local curvature and capillary pressure (Singh et al., 2017). Mechanistically, snap-off depended on the geometric and topological properties of the pore system, as well as the initial location of the interfaces; it was also shown how brine layers swell - leading to oil snap-off in a three-dimensional porous medium and which time scale is associated with that (Singh et al., 2017). Considering a scenario where snap-off occurred at a junction between three pores for example, initially the brine-oil interface on the side of the disconnected oil cluster proceeded in a piston-like displacement without oil trapping (Fig. 1a). When the interface reached the pore, the brine wetting layers in the junctions (marked by blue arrows in Fig. 1b) started to swell (Fig. 1c). Once the brine layer grew to a critical point, the oil in the throat could no longer remain stable and disconnected to create an isolated oil cluster (Fig. 1d). The local capillary pressure of the disconnected oil cluster and of the continuously connected oil phase during the whole pore-filling process as a function of normalized oil saturation is plotted in Fig. 1f.

## 3. Local interfacial instabilities during drainage: Numerical simulation

With the improvement of simulation algorithms and computational power, pore-scale numerical approaches for immiscible displacement in porous media studies have been widely developed to further investigate the displacement mechanisms that can be difficult to observe with existing research equipment. Generally, these numerical approaches can be divided into two broad categories, namely direct numerical simulation methods solving Navier-Stokes equation directly based on discretized pore structures, and indirect numerical simulation methods based on simplified geometries and governing equations (Bultreys et al., 2016). Direct numerical simulation (DNS) methods include the volume of fluid (VOF), phase-field (PF), level set (LS), or smoothed particle hydrodynamics (SPH) models, as well as the Lattice Boltzmann Method (LBM) (Inamuro, 2006; Raeini et al., 2012; Blunt et al., 2013; Jettestuen et al., 2013; Sivanesapillai et al., 2016; Chen et al., 2018; Prokopev et al., 2019; Yin et al., 2019). LBM is a pseudo-molecular method based on particle distribution functions, which determine the fluid-fluid interface dynamics through the kinetic approach (where diffusion interfaces related to the distribution of ideal particles are formed due to microscopic interaction). For example, the formation of an interface between immiscible fluids is determined by the color gradient force at the interface in the color model or

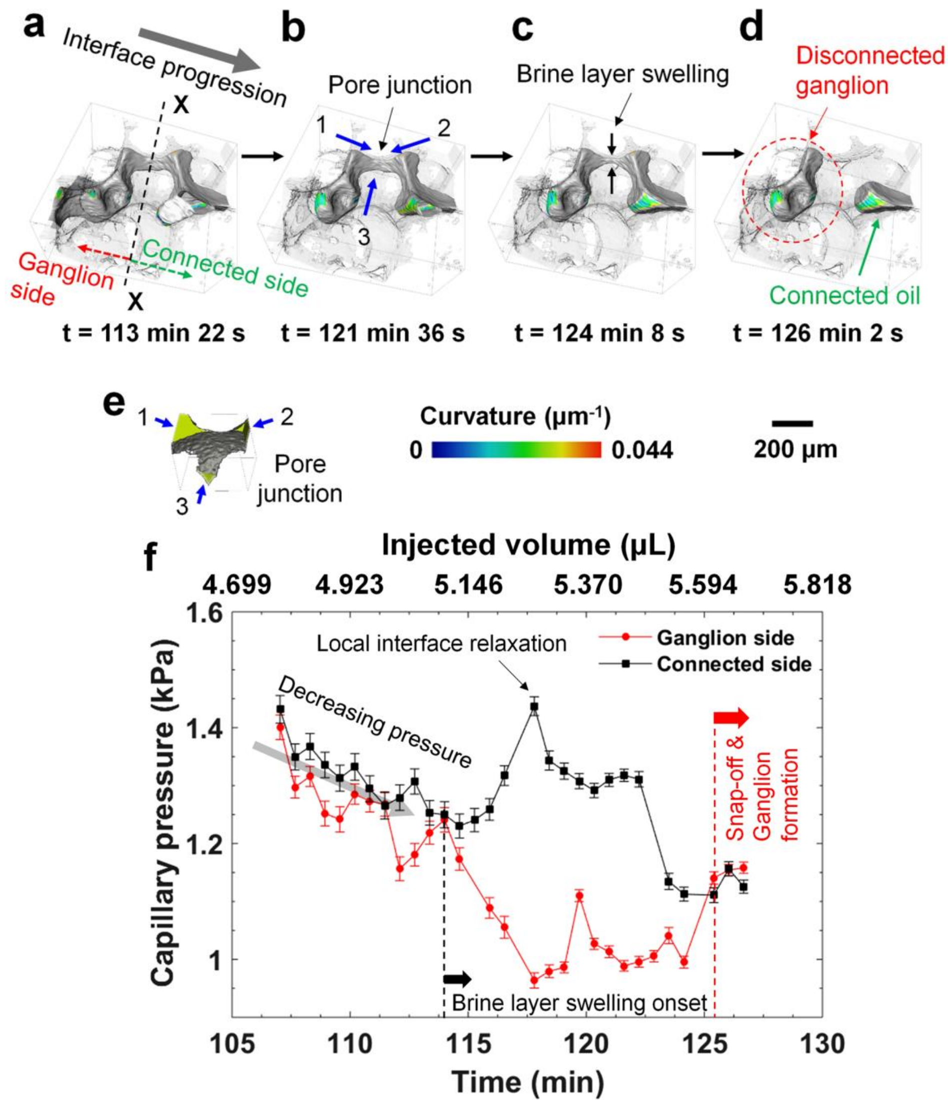


**Fig. 1.** Snap-off during imbibition at a pore junction. (a-d) Various time steps during brine injection showing displacement of oil and ganglion trapping. (e) Location of the pore junction where the snap-off occurred. (f) Capillary pressure in the disconnected and continuously connected oil clusters is plotted against time and injected volume (Singh et al., 2017).

the non-local force in the Shan-Chen model (Huang et al., 2015). Therefore, this powerful method provides a way to obtain a great number of pore space properties, such as relative permeability and capillary pressure curves, which can be applied to the highly complex geometric structure of natural porous media through straightforward implementation (Huang et al., 2011). Because these numerical simulation methods, especially the LBM, are based on a large number of discrete mesh elements or lattice nodes, they are very computationally demanding and require a large amount of computational power and can greatly benefit from high-performance parallel computations. Simplified geometric structures and underlying physical laws enable indirect numerical simulation methods (e.g. pore network models), which have computational advantages, but cannot capture flow details (Al-Kharusi and Blunt, 2008).

These advanced numerical simulation techniques provide convenience for the systematic study of different displacement mechanisms in the drainage process. Drainage pore filling events previously observed in 2D experiments, such as Haines jump, snap-off, ganglion dynamics, cooperative pore filling and droplet fragmentation, can now be easily analyzed in a 3D system via such numerical simulation techniques. For instance, the fluid redistribution associated with Haines jumps and its effects on the displacement process in capillary fingering regimes are investigated by using multi-GPU free energy lattice Boltzmann simulations (Zacharoudiou et al., 2018). Their work demonstrated three main typical features associated with the Haines jumps: i) local (single pore) sharp increase in the non-wetting phase velocity, ii) localized sharp pressure drops and iii) extensive fluid redistribution. Their study also revealed that Haines jumps can potentially decrease the displacement efficiency of the injected phase through cooperative distal snap-off events, as shown in Fig. 2. Thus a Haines jump (indicated in red cycle in Fig. 2a2) fills many pores in a cascade-like manner, accompanied by a sudden drop in the local pressure of the non-wetting phase. At the same time snap-off events occur at throats far (i.e. multiple pore spacings away) from where the Haines jump takes place, which results in the trapping of the non-wetting phase.

## 4. Local interfacial instabilities under intermediate-wet conditions: Numerical simulations

Up to now, pore-scale mechanisms of immiscible displacements have been extensively studied under two wetting conditions, namely strongly-wetting conditions for imbibition and strongly non-wetting conditions for drainage. However, different immiscible displacement dynamics are expected for intermediate-wet conditions. Intermediate-wet conditions prevail extensively in many natural formations (Alyafei and Blunt, 2016), and only a few studies have focused on this issue (Iglauer et al., 2016). For instance, the co-existence of concave and convex interfaces occurring in intermediate-wet rock was demonstrated by using a 2D VOF simulation (Rabbani et al., 2017), which emanate from the interplay between the wetting characteristics and pore geometry (as illustrated in Fig. 3a). This phenomenon stems from the increasing dependence of interface curvature on the angularity of the pores (i.e. the angle at which a pore converges or diverges) as wettability changes from strongly-wet to intermediate-wet conditions. The appearance of positive and negative curved interfaces is directly controlled by the capillary pressure, which is determined by the fluid-fluid interfacial tension and the complex interplay between contact angle and pore angularity. Interestingly, the co-existence of concave and convex interfaces promotes the occurrence of other local instabilities, such as pinning of convex interfaces (Fig. 3c), pore-level reverse displacement (Fig. 3c), and interface instability (Fig. 3d), that further influence the pore-scale displacement dynamics and immiscible displacement pattern.

## 5. Discussion

Local instabilities and the various displacement patterns mentioned above are essentially the results of unbalanced forces acting on the immiscible fluids in the pore space. Thus immiscible displacements within the pore space of a random permeable medium are controlled by three main forces, namely viscous, capillary, and gravity forces (Zhang et al., 2011). The relative magnitude of these three forces and their contribution to the displacement behavior depend not only on the properties of fluid and pore structure, as well as the physical or chemical interplay between fluid and pore space wall, but also on the physical scale, the flow state and spatial configuration of the immiscible fluids (Al-Housseiny et al., 2012). For example, the magnitude of the capillary force is determined by the intermolecular forces between the different fluids and the rock (e.g. Liang et al., 2017; Abramov et al., 2019). Thus the capillary pressure $P_c$, a macroscopic expression of the balance of capillary forces, is a function of the interfacial tension (IFT), pore space geometries and the pore surface's wettability (Iglauer, 2017). At equilibrium Pc can be quantified as the difference in pressure across the interface between the fluids. Importantly, the capillary force can be a driving or resistive force, depending on the flow process (imbibition versus drainage). In contrast, the viscous force always acts as a resistive force and its magnitude depends on the applied pressure gradient and fluid viscosities (Bandara et al., 2011). The last force, gravitational force, directly relates to the difference in density between the immiscible fluids and their characteristic length (Bandara et al., 2011). Therefore, strict separation and quantitative characterization of these forces at pore scale are generally very difficult. To simplify the situation, two dimensionless numbers were introduced to quantify the relative significance of these three forces. Capillary number (Eq. (1)) indicates the relative importance of capillary force versus viscous force, while the Bond number (Eq. (2)) indicates the relative importance of the gravity versus capillary force;

$$Ca = \frac{v_i \mu_i}{\sigma} \tag{1}$$

$$Bo = \frac{\Delta\rho g \delta^2}{\sigma} \tag{2}$$

where $\sigma$ is the fluid-fluid interfacial tension, $v_i$ and $\mu_i$ are the the flux and the viscosity of the invading fluid, respectively. $\delta$ is the average pore size, $\rho_i$ and $\rho_d$ in $\Delta\rho = (\rho_i - \rho_d)$ are the densities of invading and defensing fluids, respectively.

Using these two dimensionless numbers plus the viscosity ratio of the immiscible fluids, flow diagrams can be constructed which show the different flow regimes, i.e. where viscous fingering, capillary fingering, stable displacement and transition occur. These regimes are established under various conditions and indicate the different displacement patterns and associated local interface instabilities (Chen et al., 2018).

## 6. Implications

Darcy's law extended to multi-phase flow is often used in hydrological sciences and petroleum engineering communities to predict multiphase flow in reservoirs and aquifers, thus guiding actual production (Tomin and Lunati, 2016). However, due to the continuity nature of Darcy's law, detailed pore-scale multiphase flow phenomena described above cannot be considered (Tang et al., 2019). These insufficiencies in Darcy's (multi-phase) law result in a large uncertainty, and therefore it is of great significance to study the key factors of the abovementioned unstable filling events and their effects on macro-scale flow for secure and efficient industrial and environmental applications. For example, by designing $CO_2$ injection schemes that increase the number of snap-off events, more $CO_2$ can be trapped in pore space (Al-Khdheeawi et al., 2017). In this way, large amounts of greenhouse gases in atmosphere can be sequestrated in deep geological formations (Bandara et al., 2011). However, during oil production, the occurrence of snap-off events should be avoided to maximize

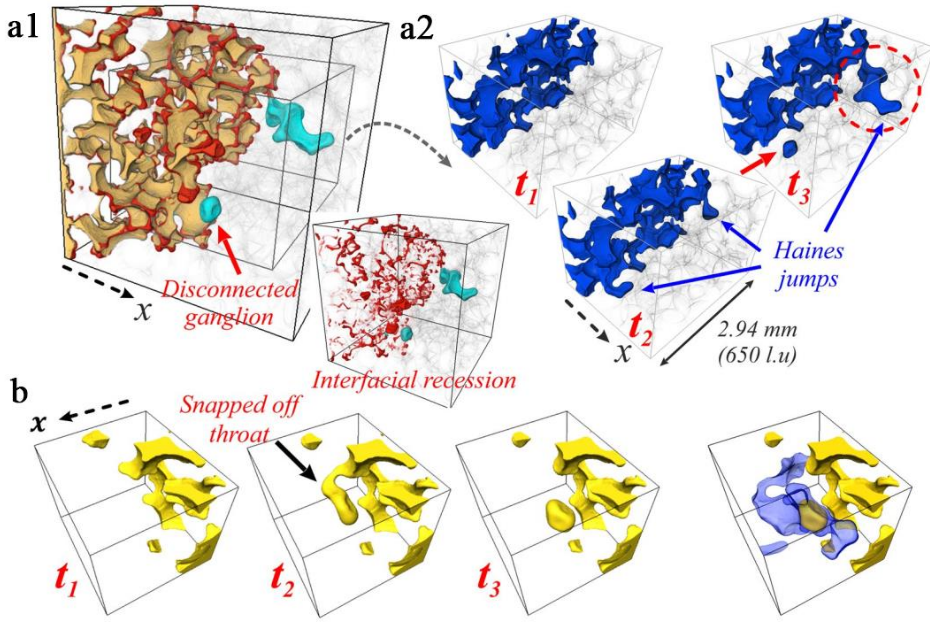


**Fig. 2.** Distal snap-off and Haines jump events during drainage. (a1) Fluid rearrangement associated with the jump events. (a2) Multiple jump events lead to distal snap-off (red arrow). (b) Same distal snap-off event from a different viewing angle. The region occupied by the non-wetting phase that remains unchanged during the event is shown in yellow, while the draining pore and locations of interfacial recession are shown in light blue and red respectively (Zacharoudiou et al., 2018).

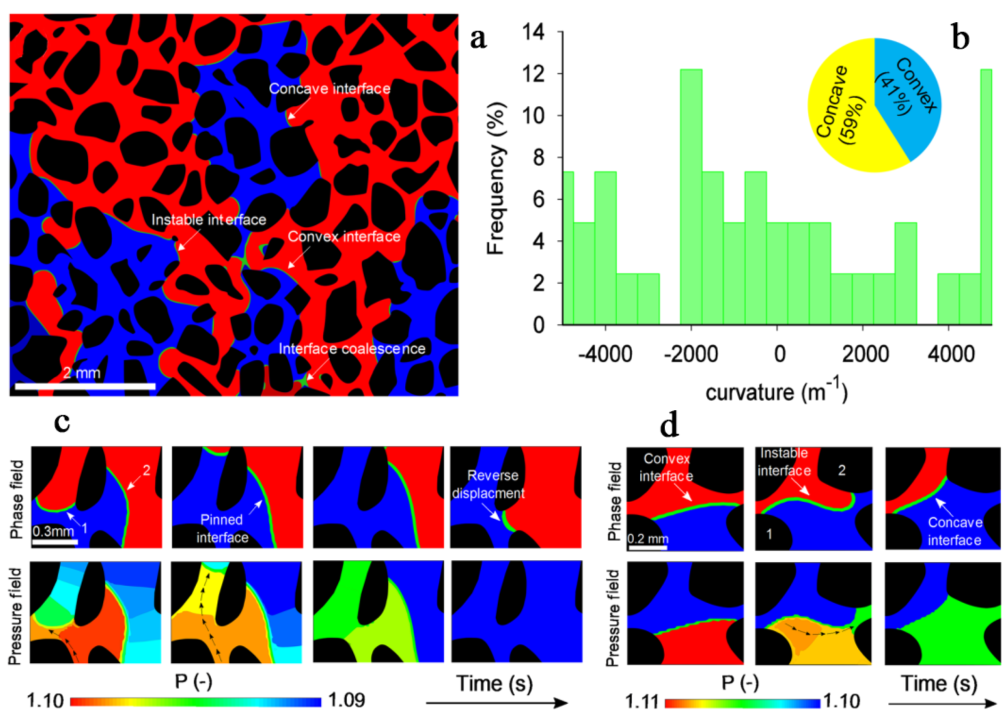


**Fig. 3.** (a) the main interfacial features observed during 2D VOF modelling of immiscible two-phase flow in an intermediate-wet porous medium ($\theta = 60°$). (b) Curvature distribution of interfaces shown in Fig. 1(a). (c) Dynamics of concave (labelled as "1") and convex (labelled as "2") interfaces during displacement. (d) Interface instability in a single pore. In the phase distribution shown in Fig. 1(a,c,d), red, blue and green represents defending fluid, invading fluid and the fluid-fluid interface, respectively. The pressure field shown in Fig. 1(c-d) indicates the pressure values normalized with respect to the outlet pressure. The direction of injection in all images is from bottom to top (Rabbani et al., 2017).

oil/gas recovery. Thus, despite the efforts made and new insights won as outlined above, more in-depth research is still needed in future.

## 7. Conclusions and future outlook

Here we presented a review of the latest developments in pore-scale dynamics of capillary-dominated immiscible displacement in porous media. The focus was given to local instability events under three typical wetting conditions, including Haines jump events during drainage under weakly-wetting conditions, snap-off events during imbibition under strongly-wetting conditions, as well as the co-existence of concave and convex interfaces under intermediate-wet conditions. These pore-scale instabilities affect not only the distribution of fluids in the pore space, but also the displacement efficiency of the various fluids. However, current research achievements on local instabilities are mainly based on two-dimensional or three-dimensional idealized porous media such as dense random arrangements of circular disks, monolayer spherical beads in 2D microfluidic experiments, monodisperse or polydisperse arrangement of glass beads in X-ray CT imaging experiments, or real porous media with relatively large pore spaces (Singh et al., 2019). In future, with the improvement of space-time resolution of CT scanning, the dynamical pore-scale flow mechanisms in real geological porous media with smaller pores and more complex structures will be studied. This is of particular importance as more comprehensive information on local instabilities in porous media with more complex pore structures, such as tight oil reservoir rocks or shales for different fluid properties and wetting conditions is needed. Only by fully understanding the occurrence conditions of these pore filling events can we formulate development plans to maximize the development of future oil and gas reservoirs and improve $CO_2$ geo-storage security.

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## Conflict of interest

The authors declare no competing interest.



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