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Pore-scale capillary pressure analysis using multi-scale X-ray micromotography

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ABSTRACT

A multi-scale synchrotron-based X-ray microtomographic dataset of residually trapped air after gravity-driven brine imbibition was acquired for three samples with differing pore topologies and morphologies; image volumes were reconstructed with voxel sizes from 4.44 µm down to 0.64 µm. Capillary pressure distributions among the population of trapped ganglia were investigated by calculating interfacial curvature in order to assess the potential for remobilization of residually-trapped non-wetting ganglia due to differences in capillary pressure presented by neighbor ganglia. For each sample, sintered glass beads, Boise sandstone and Fontainebleau sandstone, sub-volumes with different voxel sizes were analyzed to quantify air/brine interfaces and interfacial curvatures and investigate the effect of image resolution on both fluid phase identification and curvature estimates. Results show that the method developed for interfacial curvature estimation leads to reliable capillary pressure estimates for gas ganglia. Higher resolution images increase confidence in curvature calculations, especially for the sandstone samples that display smaller gas-brine interfaces which are then represented by a higher number of voxels when imaged with a micron or sub-micron voxels size. The analysis of sub-volumes from the Boise and Fontainebleau dataset highlights the presence of a residually-trapped gas phase consisting of ganglia located in one or few pores and presenting significantly different capillary pressures, especially in the case of Fontainebleau sandstone. As a result, Ostwald ripening could occur, leading to gas transfer from ganglia with higher capillary pressure to surrounding ganglia with lower capillary pressures. More generally, at the pore-scale, most gas ganglia do present similar capillary pressures and Ostwald ripening would then not represent a major mechanism for residually-trapped gas transfer and remobilization.

1. Introduction

A critical aspect of geological carbon sequestration is the estimation of CO_2 plume mobility and extent after injection has ceased. This estimation is particularly important in the context of open storage systems or systems in close proximity to possible migration pathways e.g. permeable faults. After an initial drainage phase during injection, models typically posit that substantial $scCO_2$ volumes are trapped as the plume moves under buoyant forces during brine imbibition (Kumar et al., 2005; Juanes et al., 2006; Hesse et al., 2008; Celia and Nordbotten, 2009). Conventional multi-phase flow models assume that residually trapped portions of the plume do not remobilize and remain stationary until they dissolve into the brine phase (Goodman et al., 2011), as under reservoir flow conditions, viscous forces are usually not sufficient for mobilization. However, multiple physiochemical mechanisms exist which could potentially invalidate this assumption including $scCO_2$-induced transitions in wetting behavior (Kim et al., 2012; Wan et al., 2014; Wang et al., 2016), dissolution-induced ganglia size reduction (DePaolo and Cole, 2013), and inter-ganglia gas transfer via Ostwald ripening (Epstein and Plesset, 1950). In addition, recent findings from fast synchrotron-based X-ray computed microtomography (Armstrong et al., 2014; Rücker et al., 2015, Armstrong et al., 2016) and micromodel studies (Armstrong and berg, 2013) show that non-local cooperative displacement processes during water flooding have a large impact on pore scale fluid distributions and ganglion dynamics and could potentially affect the stability of a trapped non-wetting phase.

In this study, we seek to develop experimental methods for improving the understanding of one of these mechanisms, the potential for Ostwald ripening of the residually trapped CO_2 phase in porous media. Ostwald Ripening of gas bubbles in aqueous solutions has been extensively studied in homogeneous bulk media

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where dispersed particles of a second phase exist in a saturated solution, and where the bigger bubbles tend to grow at the expense of the smaller ones (Epstein and Plesset, 1950; Greenwood, 1956; Lifshitz and Slyozov, 1961; Voorhees, 1985; Schmelzer and Schweitzer, 1987; Möller et al., 1998). In porous media, the presence of walls changes the process significantly, mainly because capillary pressure, the driver for ripening, is no longer only a function of the particle radius but also of pore topology and size. Few attempts have been made to extend the study of Ostwald ripening process to porous media. Most often, the process is either neglected (Andrew et al., 2014a), studied in a context where other mechanisms like a constant pressure decline in the liquid play a disproportionate role (Li and Yortsos, 1991; 1995; Dominguez et al., 2000), or studied in a context where cluster growth stops or is strongly inhibited once there is significant interaction with the porous matrix walls, for viscous polymers and crystals growing in a solid matrix for instance (Schmelzer et al., 1995; Möller et al., 1998). Goldobin and Brilliantov (2011) investigated the diffusion of gas in porous media using mean thermodynamic properties, but did not account for pore-scale effects.

Ostwald ripening will be driven by differences in capillary pressure between gas ganglia, and subsequent diffusion of dissolved gas through the aqueous phase. Capillary pressure, which denotes the difference in pressure between a non-wetting and a wetting fluid (respectively CO_2 and brine in the case of interest), is expressed by the Young-Laplace equation $P_c = P_{nw} - P_w = 2\sigma\kappa$, where σ is the interfacial tension and κ is the mean curvature of the interface between the two fluids. In a bulk liquid medium, a bubble of gas is typically spherical and its curvature is therefore directly linked to the bubble size. Porous media, however, exhibit a variety of complex geometries for which no simple analytical solution exists to describe the interface between two immiscible fluids. As capillary pressures inside individual non-wetting phase ganglia strongly depend on pore geometry and topology (Bear, 1972), pore-scale investigations are required in order to understand and measure the distribution of capillary pressure in disconnected trapped ganglia of non-wetting phase, which is a critical question to assess Ostwald ripening mechanism.

Due to significant improvements over the last decades and its non-destructive characteristic, X-ray microtomography has become the foremost imaging technique for the visualization and quantification of pore-scale structures and processes as well as providing input to pore-scale modeling (Blunt et al., 2013; Cnudde and Boone, 2013; Wildenschild and Sheppard, 2013, Schlüter et al., 2014). Previous authors investigated residual trapping at the pore-scale using X-ray microtomography (Prodanovic et al., 2007; Kumar et al., 2010; Iglauer et al., 2011; Tanino and Blunt, 2012; Georgiadis et al., 2013; Chaudhary et al., 2013; Andrew et al., 2014a; Rücker et al., 2015; Herring et al., 2016, Schlüter et al., 2016). In particular, all of these studies show that the residually-trapped non-wetting phase takes the form of ganglia with various sizes and complicated irregular shapes, highlighting the complexity of the non-wetting and wetting phase interface geometry, as well as of the angle both fluids form with the solid surface.

Armstrong et al. (2012a) proposed a first method to calculate interfacial curvature using synchrotron-based X-ray microtomography images of water and oil in a glass beads packed column with a voxel size of 13 µm. They compared macro-scale capillary pressures measured via pressure transducer to micro-scale capillary pressures measured via interfacial curvatures calculated on the images using Avizo Software. They concluded, in particular, that higher image resolution was required as significant curvature errors were resulting from inadequate segmentation of small features. Following this work, Armstrong et al. (2012b) evaluated the limitations and advantages of measuring interfacial curvatures from X-ray microtomography data by comparing two approaches applied to idea spheres: the first approach measures curvature on triangular surfaces generated at interfacial regions, as described in Armstrong et al. (2012a), the second approach measures curvature directly from an image intensity gradient. The results showed that there is an optimal amount of smoothing with either approach and, in general, the surface-based approach gave the best results. Andrew et al. (2014b) then applied the method described by Armstrong et al., (2012a) to calculate interfacial curvature of water and supercritical CO_2 ($scCO_2$) (50 °C and 10 MPa) in a Ketton limestone sample using lab-based microtomography images with a pixel size of 3.5 µm. The authors reported that curvature distributions for a single ganglion present a single well-defined peak, representing a single capillary pressure across the entire CO_2-brine interface. They also related capillary pressure measurements to local pore space topography and showed that capillary pressure was inversely proportional to the radius of the largest restriction surrounding the ganglion. In a recent publication aiming at identifying individual dynamic drainage events at the pore-scale using fast synchrotron-based microtomography images of $scCO_2$/brine in the same Ketton limestone with a voxel size of 3.64 µm, Andrew et al. (2015) extended the pore-scale curvature measurement technique in order to be able to measure the smaller curvatures resulting from higher drainage capillary pressures. In particular they presented a method to select terminal menisci from the entire CO_2 ganglia interfaces using curvature anisotropy and compute the interfacial curvature using only these selected regions.

The Ostwald ripening process is driven by differences in capillary pressure, and consequently interfacial curvature, between ganglia. The study of the potential for Ostwald Ripening therefore requires accurate methods for measuring the interfacial curvature of an individual ganglia and its associated uncertainty in a variety of relevant rock types. The aim of the present study is to develop reliable methods for estimating the pore-scale capillary pressure of individual residually-trapped gas ganglia for different rock types, especially sandstones, and using higher resolution images in comparison to the previous studies by Armstrong et al. (2102a, 2012b) and Andrew et al. (2014b, 2015). Measurements are made on air ganglia trapped during gravitationally induced imbibition. Particular attention is given to the effect of image resolution on the precision of interfacial curvature calculations. A multi-scale synchrotron-based X-ray microtomography dataset of residually-trapped gas was therefore acquired in sintered glass beads and two sandstone samples (Boise and Fontainebleau) with voxel sizes varying from 0.64 to 4.44 µm. A method to estimate the interfacial curvature was developed based on this dataset and is presented, together with a sensitivity analysis of the method to parameters such as the degree of smoothing or the number of neighbor voxels used for the calculation. For each acquired image the non-wetting (air) and wetting (brine) phases were identified, as well as the interfaces they share both with each other and also with the solid surface, and the connectivity and size of the trapped gas were quantified. The curvature method was then applied to the different sub-volumes chosen for analysis. The distribution of curvature values, the mean value, and associated uncertainty is presented for each disconnected air ganglion. Sub-volumes of the rock are then analyzed to determine the distribution of capillary pressures among the trapped ganglia; in particular, whether there is a statistically significant difference in the capillary pressures of the ganglia trapped in the sub-volume. A detailed analysis of the impact of image resolution and interface identification on curvature estimates is also reported.

The key point to assess the potential for Ostwald ripening in the investigated pore structures is to determine if neighbor gas ganglia have significantly different capillary pressures or not. Therefore a part of this work also consists in investigating the accuracy of the