Published in final edited form as:

Water Air Soil Pollut. 2017 December ; 228: .

Optimizing the Gas Absorption/Chemical reaction Method for Measuring Air–water Interfacial Area in Porous Media

Ying Lyu^{1,2,3,4} and Mark L. Brusseau^{4,*}

¹Key Lab of Groundwater Resources and Environment, Ministry of Education, Jilin University, Changchun, 130026, P.R. China

²College of Environment and Resources, Jilin University, Changchun 130021, P.R. China

³Institute of Water Resources and Environmental, Jilin University, Changchun 130026, P.R. China

⁴Department of Soil, Water and Environmental Science and Department of Hydrology and Atmospheric Sciences, School of Earth and Environmental Sciences, University of Arizona, Tucson, AZ, USA

Abstract

The gas-absorption/chemical-reaction (GACR) method developed in Chemical Engineering to measure gas-fluid interface in reactor systems is adapted for natural porous geologic media. Several series of column experiments were conducted using model glass beads and a natural sand to determine optimal operational conditions for measuring air-water interfacial area with the adapted method. The impacts of operational variables were investigated, including liquid and gas volumetric flow rates, solution concentration, and temperature. The results show that the magnitude of the measured air-water interfacial area is dependent upon all of these variables to greater or lesser degrees. Larger fluid flow rates promote distribution and mixing of the fluids, enhancing absorption and reaction. Increasing the concentration of NaOH in solution reduced the relative utilization of NaOH, promoting pseudo first-order reaction conditions. The results elucidate the optimal operational conditions for application of the method to geomedia systems.

Keywords

gas absorption; chemical reaction; air-water interfacial area; fluid-fluid interface

1. Introduction

The significance of fluid-fluid interfacial area for numerous applications in environmental, geologic, and hydrologic systems has driven the development of several measurement methods. These include mass-balance tracer methods [Anwar et al., 2000; Anwar, 2001; Araujo et al., 2015; Schaefer et al., 2000], aqueous and gas-phase interfacial partitioning tracer tests [Brusseau et al., 1997, 2006, 2007, 2008, 2015; Cho and Annable, 2005; Costanza and Brusseau, 2002; Dobson et al., 2006; Kim et al., 1997, 1999; McDonald et al.,

*Corresponding author address: Department of Soil, Water and Environmental Science, School of Earth and Environmental Sciences, University of Arizona, Tucson, AZ, USA, Phone: (520) 621-3244, brusseau@email.arizona.edu.

2016; Narter and Brusseau, 2010; Peng and Brusseau, 2005; Saripalli et al., 1997; Zhong et al., 2016], and pore-scale imaging methods [Al-Raoush and Willson, 2005; Brusseau et al., 2006, 2007, 2008; Costanza-Robinson et al., 2008; Culligan et al., 2004; McDonald et al., 2016; Porter et al., 2010; Schnaar and Brusseau, 2005]. Each of the methods has certain constraints that can complicate and limit their use. For example, high-resolution pore-scale imaging requires specialized equipment generally only available at national user facilities, limiting access. The tracer-test methods are relatively time-consuming, and can be subject to the impact of experimental artifacts in the absence of thorough controls.

Recently, the gas-absorption/chemical-reaction (GACR) method developed in Chemical Engineering to measure gas-fluid interface in reactor systems was adapted for use in environmental systems (Lyu et al., 2016). This method has been applied to numerous types of reactors, including rotating bed [Luo et al., 2012; Tsai and Chen, 2015; Yang et al., 2011], packed column [Kolev et al., 2006; Pubanik and Vogelpohl, 1974, stirred cell [van Woezik and Westerterp, 2000], and bubble column [Maceiras et al., 2010]. These systems generally have extremely high porosities ($n > 90\%$) due to the use of structured packing media such as stainless steel wire mesh, Rachel rings, Pall rings, etc that are designed to provide large surface area for gas-liquid contact. Effective use of the GACR method for the relatively low porosities typical of geomedia requires significant changes in operating conditions compared to those employed for standard reactor systems.

The objective of this study is to determine optimal operational conditions for measuring air-water interfacial area in geomedia with the GACR method. Several sets of column experiments are conducted using model glass beads and a natural sand to investigate the impacts of operational variables on measured air-water interfacial area. The parameters investigated include liquid and gas volumetric flow rates, column diameter, solution concentration, and temperature.

2. Theory

The NaOH–CO₂ system was used because it is one of the most common systems employed for reactor systems [Svoboda and Rylek, 1979]. When the NaOH solution is in excess, the reaction can be expressed as:

Reaction (1) can be treated as a pseudo first-order reaction when the following inequality is satisfied [Sharma and Danckwerts, 1970]:

$$\sqrt{\frac{Dk_2c_{OH}}{k_L^2}} \leq 1 + \frac{c_{OH}}{2C_i} \quad (2)$$

where D is the diffusivity of CO₂ in solution (m²/s), k_2 is the second-order rate constant between CO₂ and OH[−] (m³/kmol·s), c_{OH} is concentration of OH[−] in solution (kmol/m³), k_L

is the liquid side mass transfer coefficient (m/s), and c_i is the concentration of CO_2 at the gas–liquid interface (kmol/m^3).

The absorption rate, Ra (kmol/s) can be expressed as equation (3) according to Danckwerts' mass transfer model [Sharma and Danckwerts, 1970]:

$$Ra = Ac_i \sqrt{Dk_2 c_{OH} + k_L^2} \quad (3)$$

where A is the total interfacial area in the packed column (m^2). The absorption rate Ra can be determined from the formation rate of Na_2CO_3 [e.g., Kasturi and Stepanek, 1974; Yang et al., 2011], with the concentration of Na_2CO_3 in the solution measured by titration. When the inequality (4) is satisfied,

$$\sqrt{\frac{Dk_2 c_{OH}}{k_L^2}} \geq 3 \quad (4)$$

the chemical reaction is sufficiently rapid such that it is not sensitive to k_L [Mohanty et al., 2007], and the absorption rate can be expressed as:

$$Ra = Ac_i \sqrt{Dk_1} \quad (5)$$

introducing the pseudo first-order rate coefficient, $k_1 (= k_2 c_{OH})$.

The procedure for measuring A is based on determining Ra from the GACR experiment, and then calculating A from eq 5, with values for D , k_1 , and c_i determined as follows. The diffusion coefficient D of CO_2 in water reported in the literature was corrected for the presence of high concentrations of NaOH , using the following equation [Bhat et al., 2000]:

$$D = 2 \times 10^{-9} \times (1 - 0.129c_{OH^-} - 0.261c_{CO_3^{2-}}) \quad (6)$$

The calculated values range from 1.4 to $1.7 \times 10^{-9} \text{ m}^2/\text{s}$, which are slightly lower than and consistent with the measured value for pure water ($2 \times 10^{-9} \text{ m}^2/\text{s}$) [Zeebe, 2011].

The k_1 value can be calculated via its relationship to k_2 and accounting for the temperature and electrolyte concentration dependency of k_2 (equation 7 [Roberts and Danckwerts, 1962])

$$\log k_2 = 0.1987 I - 0.012 I^2 + 11.895 - \frac{2382}{T} \quad (7)$$

where I is the ionic strength (kmol/m^3) and T is temperature (K).

The concentration of CO₂ at the gas-liquid interface can be determined by applying Henry's Law for mass transfer across a liquid film:

$$c_i = SP_{CO_2} \quad (8)$$

where P_{CO_2} is partial pressure of CO₂ in the gas mixture and S is the solubility coefficient of CO₂ in the electrolyte solution (kmol/m³ MPa), which can be estimated as follows [Yang et al., 2011]:

$$\log S - \log S_0 = -(c_{NaOH} \times 0.138 + 3c_{Na_2CO_3} \times 0.093) \quad (9)$$

$$S_0 = \frac{\rho_w}{M_w} \times \frac{1}{H} \quad (10)$$

where S_0 is solubility coefficient of CO₂ in pure water (kmol/m³MPa), H is Henry coefficient of CO₂ in pure water (kpa), and ρ_w (kg/m³) and M_w are the density and relative atomic mass of water, respectively. The partial pressure of CO₂ can be written as:

$$P_{CO_2} = \frac{y_{CO_2}^i - y_{CO_2}^o}{\ln(y_{CO_2}^i / y_{CO_2}^o)} \times P_{total} \quad (11)$$

where $y_{CO_2}^i$ and $y_{CO_2}^o$ are the inlet and outlet mole ratios of CO₂.

The effective volume-normalized specific gas-liquid interfacial area in the system can be expressed as:

$$A_{ia} = \frac{A}{V} \quad (12)$$

where V is the volume of the packing (m³), and A is obtained from Equation (5).

3. Experimental

3.1 System

Two columns were used for the experiments, one with inner diameter of 2.6 cm and the other with 1.4 cm. Both columns were constructed of polypropylene. The columns were oriented vertically for all experiments. Flow distributors were placed in contact with the

porous media on the top and at the bottom of the column to help promote uniform fluid distribution and to support the media. The experimental set-up is shown in Fig.1. A separator was placed at the bottom of the column to facilitate separation of the gas and solution. Effluent gas samples were collected from the outlet into a sealed conical flask, and liquid effluent samples were collected into 50 mL vials.

Two porous media were used for the experiments. The first is an ideal glass-bead medium ranging in size from 0.94 mm to 1.4 mm with mean diameter 1.16 mm. The second is a 45/50 mesh natural quartz sand whose mean diameter was 0.35 mm. The specifications of the column and porous media used in this study are presented in Table 1.

3.2 Experimental procedure

The column was dry packed, after which it was weighed to determine bulk density. The pack was then saturated with water to determine porosity gravimetrically. Gas was then injected briefly at a high flow rate to drain the pack to the target water saturation, which were 0.43 for the glass beads and 0.57 for the sand. These values were selected to represent moderately saturated conditions. After these initial preparations, NaOH solution was injected into the top of the vertically oriented column at a steady flow rate to initiate the GACR test. Concentrations of NaOH in the injected solution were constant for each experiment, and ranged from 1 to 2 M. Concurrently, gas containing 4.6% CO₂ (in a balance of nitrogen) was also injected into the top of the column. Concurrent as opposed to countercurrent liquid and gas flow was used due to the low porosity of the porous media.

The concentration and temperature of the inlet NaOH solution and the concentration and relative pressure of inlet CO₂ gas were measured prior to entry of the fluids into the column. Liquid and gas effluent samples were collected every 5 minutes to determine the concentrations of NaOH and Na₂CO₃ in solution and the concentration of CO₂ in the gas. Samples were collected until the effluent concentrations of all constituents were steady. The packed column was flushed with pure water after each experiment until constituent concentrations in the effluent solution were below detection.

3.3 Analysis methods

The concentration of CO₂ in the gas was measured using a CO₂ analyzer (Smart Bell plus, UEI Test Instrument Company, USA). Gas pressure was measured with a static pressure meter (EM201SPKIT, UEI Test Instrument Company, USA). The gas flow rate was measured with a direct-read rotameter (FLDA3412C, UEI Test Instrument Company, USA) placed between the gas cylinder and the column. The concentration of NaOH and Na₂CO₃ in solution were measured by titration with 0.5 M HCl solution. All of the chemicals used in this study were analytical grade and all solutions were prepared with ultra-pure water.

4. Results and discussions

4.1 Effect of column diameter

The impact of column diameter was investigated for the glass beads by comparing the results of experiments conducted with the two columns. The results in Fig. 2 show that

larger interfacial areas were obtained for the smaller-diameter column. Dye tests were conducted to characterize solution distribution under the operative conditions (data not shown). The results showed that the solution did not distribute completely across the column cross-section for the larger-diameter column. This explains the smaller interfacial areas obtained for tests conducted with that column. Hence, the smaller-diameter column was used for the remaining experiments.

4.2 Effect of gas flow rate

The gas flow rate was increased incrementally over a range from 50 to 1000 mL/min to investigate its impact on the measured interfacial area. CO₂ utilization was complete, with gas effluent concentrations below detection, at gas flow rates less than 200 mL/min. Determining interfacial areas is problematic under this condition. Conversely, CO₂ utilization was incomplete at the higher flow rates due to the reduced residence time, and interfacial areas could thus be determined. As seen in Fig. 2, increasing the gas flow rate initially led to an increase of the air-water interfacial area for a fixed liquid flow rate. Further increasing gas flow rate led to no further increase in interfacial area for the sand and a slight decrease for the glass beads.

The water saturation of the porous medium was measured gravimetrically before and after each experiment. The saturation exhibited minimal variation across all gas flow rates (coefficient of variation, COV, of 3%) for the glass beads for a given liquid flow rate. The saturations were constant (within measurement error) for the sand, with the exception of the highest gas flow rate, wherein water saturation decreased by 12%. The essentially constant water saturations indicate that water displacement did not change significantly over the range of gas flow rates tested. From these results, it is clear that the changes in air-water interfacial area exhibited in Fig. 2 were not caused by changes in water saturation.

The increase in interfacial area observed upon the increase in gas flow rate is likely a result of increased distribution and mixing of the gas and liquid at higher gas flow rates, which would enhance absorption and reaction. Similar results have been observed for applications of the GACR method to various chemical reactor systems [Piche et al., 2002; Yang et al., 2011], and have been attributed to enhanced distribution and mixing achieved with higher gas flow rates. The discontinuation of the increase in interfacial area most likely reflects that the system attained maximum effective distribution and mixing for the operative conditions. The slight decrease in interfacial area values observed for the glass beads in the smaller-diameter column may be a result of channeling induced at higher gas flow rates.

4.3 Effect of liquid flow rate

The results presented in Fig. 3 and Fig. 4 show that air-water interfacial area increased significantly when the liquid flow rate was increased from 5 mL/min to 12 mL/min. The water saturation changed minimally (COVs of 4%) for both glass-bead data sets and was constant for the sand for all solution flow rates for a given gas flow rate. Thus, the observed significant increases in interfacial area were not caused by a change in water saturation. The increase is likely due in part to enhanced solution distribution and mixing with the gas. In addition, as discussed in the following section, conditions for treating the reaction process as

pseudo first-order were not met at the lowest liquid flow rates. The rate of increase was lower at the highest water flow rates tested, particularly for the sand, which likely reflects that solution distribution was tending toward maximum, as well as that conditions for pseudo first-order reaction kinetics were fully satisfied.

4.4 Effect of solution concentration

Based on the assumption of pseudo first-order chemical reaction, the concentration of NaOH in solution should remain in excess during the experiment. NaOH in solution was monitored for all experiments to evaluate NaOH utilization and to test for satisfaction of equation 2. NaOH utilization was 45% for the glass beads system and 35% for the sand system at a solution flow rate of 5 mL/min and a gas flow rate of 1000 mL/min. This result indicates that the conditions for equation 2 were not sufficiently met at the lower liquid flow rates. Conversely, NaOH utilization decreased to 13% for the glass beads and 16% for the sand for liquid flow rates greater than 13 mL/min and 15 mL/min, respectively. This decrease in relative utilization is due to the reduction in hydraulic residence time associated with the increased liquid velocities.

Additional experiments were conducted to examine the impact of NaOH initial concentration on NaOH utilization and interfacial areas. By increasing the solution concentration, the NaOH utilization was reduced to only 7% for the glass beads and 11% for the sand. As shown in Figs. 5 and Fig. 6, increasing NaOH concentration resulted in measurably larger interfacial areas for both the glass beads and sand. However, further increase of the solution concentration caused an increase in liquid viscosity, which apparently reduced effective distribution and led to a slight decrease of interfacial area for the sand.

4.5 Effect of solution temperature

The effect of temperature on air-water interfacial area was also investigated. The GACR method involves both absorption of CO₂ into liquid and reaction of CO₂ with NaOH in solution. Hence, the impact of changing temperature may be two-fold, potentially influencing mass transfer and reaction. Inspection of Fig. 7 shows that temperature had no measurable impact on interfacial area over the range of 18 to 26 °C. Conversely, interfacial area decreased slightly above 26 °C. This suggests that the impact of higher temperature on reduced gas absorption outweighed the impact on increased reaction rates.

5. Conclusion

The GACR method from chemical engineering, which is based on pseudo first-order reaction of CO₂ absorbed into NaOH solution, was adapted to measure air-water interfacial area for geomeia systems. Experiment results indicate that the air-water interfacial area was dependent upon the gas flow rate, liquid flow rate, solution concentration, and solution temperature. The set of optimal conditions varied only slightly between the two porous media. However, greater differences may be observed for more heterogeneous media.

The apparatus employs standard components, and measurements can be completed within tens of minutes. Both of which are advantages compared to the other methods available. In

summary, the GACR method appears to be a viable method for measurement of robust air-water interfacial areas for geomechanics systems. The results of this work help to define the optimal set of conditions for employing the GACR method for these systems.

Acknowledgments

This research was supported by the NIEHS Superfund Research Program (P42 ES04940). The first author was supported by a scholarship from the China Scholarship Council during their stay at the University of Arizona. We thank Asma El Ouni for her assistance.

References

- Al-Raoush RI, Willson CS. A pore-scale investigation of a multiphase porous media system. *J Contam Hydrol.* 2005; 77:67–89. [PubMed: 15722173]
- Anwar AHMF. Experimental determination of air–water interfacial area in unsaturated sand medium. *New Approaches Characterizing Groundwater Flow.* In: Proceedings. of XXXI IAH Congress, Munich, Germany, September 10–14, Vol. 2001; 2:821–825.
- Anwar AHMF, Bettahar M, Matsubayashi U. A method for determining air–water interfacial area in variably saturated porous media. *J Contam Hydrol.* 2000; 43:129–146.
- Araujo JB, Mainhagu J, Brusseau ML. Measuring air–water interfacial area for soils using the mass balance surfactant-tracer method. *Chemosphere.* 2015; 134:199–202. [PubMed: 25950136]
- Bhat RDV, Kuipers JAM, Versteeg GF. Mass transfer with complex chemical reaction in gas–liquid systems: two step reversible reactions with unit stoichiometric and kinetic orders. *Chem Eng J.* 2000; 76:127–152.
- Brusseau ML, Popovicova J, Silva JAK. Characterizing gas-water interfacial and bulk-water partitioning for gas-phase transport of organic contaminants in unsaturated porous media. *Environ Sci Technol.* 1997; 31:1645–1649.
- Brusseau ML, Peng S, Schaar G, Costanza-Robinson MS. Relationships among air-water interfacial area, capillary pressure, and water saturation for a sandy porous medium. *Water Resour Res.* 2006; 42 article W03501.
- Brusseau ML, Peng S, Schnaar G, Murao A. Measuring air-water interfacial areas with X-ray microtomography and interfacial partitioning tracer tests. *Environ Sci Technol.* 2007; 41:1956–1961. [PubMed: 17410790]
- Brusseau ML, Janousek H, Murao A, Schnaar G. Synchrotron X-ray microtomography and interfacial partitioning tracer test measurements of NAPL-water interfacial areas. *Water Resour Res.* 2008; 44 article W01411.
- Brusseau ML, El Ouni A, Araujo A, Zhong H. Novel methods for measuring air–water interfacial area in unsaturated porous media. *Chemosphere.* 2015; 127:208–213. [PubMed: 25732632]
- Cho J, Annable MD. Characterization of pore scale NAPL morphology in homogeneous sands as a function of grain size and NAPL dissolution. *Chemosphere.* 2005; 61:899–908. [PubMed: 15950262]
- Costanza MS, Brusseau ML. Contaminant vapor adsorption at the gas-water interface in soils. *Environ Sci Technol.* 2000; 34:1–11.
- Costanza-Robinson MS, Brusseau ML. Air-water interfacial areas in unsaturated soils: Evaluation of interfacial domains. *Water Resour Res.* 2002; 38:13-11–13-17.
- Costanza-Robinson MS, Harrold KH, Lieb-Lappen RM. X-ray microtomography determination of air-water interfacial area-water saturation relationships in sandy porous media. *Environ Sci Technol.* 2008; 42(8):2949–2956. [PubMed: 18497149]
- Culligan KA, Wildenschild D, Christensen BSB, Gray WG, Rivers ML, Tompson AFB. Interfacial area measurements for unsaturated flow through a porous medium. *Water Resour Res.* 2004; 40:12413.
- Dobson R, Schroth MH, Oostrom M, Zeyer J. Determination of NAPL-water interfacial areas in well-characterized porous media. *Environ Sci Technol.* 2006; 40:815–822. [PubMed: 16509323]

- Kasturi G, Stepanek JB. Two phase flow-III. Interfacial area in cocurrent gas-liquid flow. *Chem Eng Sci.* 1974; 29:713–719.
- Kim H, Rao PSC, Annable MD. Determination of effective air-water interfacial area in partially saturated porous media using surfactant adsorption. *Water Resour Res.* 1997; 33:2705–2711.
- Kim H, Rao PSC, Annable MD. Gaseous tracer technique for estimating air-water interfacial areas and interface mobility. *Soil Sci Soc Am J.* 1999; 63:1554–1560.
- Kolev N, Nakov S, Ljutzkanov L, Kolev D. Effective area of a highly efficient random packing. *Chem Eng Process.* 2006; 45:429–436.
- Luo Y, Chu GW, Zou HK, Zhao ZQ, Dudukovic MP, Chen JF. Gas-liquid effective interfacial area in a rotating packed bed. *Ind Eng Chem Res.* 2012; 51:16320–16325.
- Lyu, Y., Brusseau, ML., Lin, X. Measuring gas-liquid interfacial area in natural porous media with the gas absorption/chemical-reaction method. Presented at the Annual meeting of the Soil Science Society of America; Phoenix, AZ. November 2016; 2016.
- Maceiras R, Alvarez E, Cancela MA. Experimental interfacial area measurements in a bubble column. *Chem Eng J.* 2010; 163:331–336.
- McDonald K, Carroll KC, Brusseau ML. Comparison of fluid-fluid interfacial areas measured with X-ray microtomography and interfacial partitioning tracer tests for the same samples. *Water Resour Res.* 2016; 52:5393–5399. [PubMed: 28936003]
- Mohanty K, Das D, Biswas MN. Mass transfer characteristics of a novel multi-stage external loop airlift reactor. *Chem Eng J.* 2007; 133:257–264.
- Narter M, Brusseau ML. Comparison of interfacial partitioning tracer test and high-resolution microtomography measurements of fluid-fluid interfacial areas for an ideal porous medium. *Water Resour Res.* 2010; 46:W08602.
- Peng S, Brusseau ML. Impact of soil texture on air-water interfacial areas in unsaturated sandy porous media. *Water Resour Res.* 2005; 41 article W03021.
- Porter ML, Wildenschild D, Grant G, Gerhard JL. Measurement and prediction of the relationship between capillary pressure, saturation, and interfacial area in a NAPL-water-glass bead system. *Water Resour Res.* 2010; 46 article W08512.
- Pubanik SS, Vogelpohl A. Effective interfacial area in irrigated packed columns. *Chem Eng Sci.* 1974; 29:501–507.
- Roberts D, Danckwerts PV. Kinetics of CO₂ absorption in alkaline solutions—I Transient absorption rates and catalysis by arsenite. *Chem Eng Sci.* 1962; 17:961–969.
- Saripalli KP, Kim H, Rao PSC, Annable MD. Measurement of specific fluid-fluid interfacial areas of immiscible fluids in porous media. *Environ Sci Technol.* 1997; 31:932–936.
- Schaefer CE, DiCarlo DA, Blunt MJ. Experimental measurement of air-water interfacial area during gravity drainage and secondary imbibition in porous media. *Water Resour Res.* 2000; 36:885–890.
- Schnaar G, Brusseau ML. Pore-scale characterization of organic immiscible-liquid morphology in natural porous media using synchrotron X-ray microtomography. *Environ Sci Technol.* 2005; 39:8403–8410. [PubMed: 16294880]
- Sharma MM, Danckwerts PV. Chemical methods of measuring interfacial areas and mass transfer coefficients in two fluid systems. *Br Chem Eng.* 1970; 15:522–528.
- Svoboda K, Rylek M. Measurement of interfacial area and mass transfer coefficients by chemical method of CO₂ absorption into aqueous solutions of NaOH in a model column with expanded metal sheet packing. *Chem Eng Commun.* 1979; 3:399–411.
- Tsai CY, Chen YS. Effective interfacial area and liquid-side mass transfer coefficients in a rotating bed equipped with baffles. *Sep Purif Technol.* 2015; 144:139–145.
- van Woezik BAA, Westerterp KR. Measurement of interfacial areas with the chemical method for a system with alternating dispersed phases. *Chem Eng Process.* 2000; 39:299–314.
- Wang M, Gao B, Tang D. Review of key factors controlling engineered nanoparticle transport in porous media. *J Haz, Mater.* 2016; 318:233–246.
- Yang K, Chu GW, Zou HK, Sun BC, Shao L, Chen JF. Determination of the effective interfacial area in rotating packed bed. *Chem Eng J.* 2011; 168:1377–1382.

- Zeebe RE. On the molecular diffusion coefficients of dissolved gases, and their dependence on isotopic mass. *Geochim Cosmochim Acta*. 2011; 75:2483–2498.
- Zevi Y, Gao B, Zhang W, Morales VL, Cakmak ME, Medrano EA, Sang WJ, Steenhuis TS. Colloid retention at the meniscus-wall contact line in an open microchannel. *Water Res*. 2012; 46:295–306. [PubMed: 22130000]
- Zhong H, El Ouni A, Lin D, Wang B, Brusseau ML. The two-phase flow IPTT method for measurement of nonwetting-wetting liquid interfacial areas at higher nonwetting saturations in natural porous media. *Water Resour Res*. 2016; 52:5506–5515. [PubMed: 28959079]

Fig. 1. Schematic of the experimental setup

1. Gas cylinder; 2. Gas flow meter; 3. Packed column; 4. Gas inlet; 5. NaOH solution reservoir; 6. Water pump; 7. Solution inlet; 8. Gas-liquid separation device; 9. Gas outlet; 10. Solution outlet; 11. Outlet gas collection device; 12. Solution sample collection device.

Fig. 2. Air-water interfacial area vs. gas flow rate for glass beads and 45/50 sand; liquid flow rate $Q_L = 5$ ml/min, $T = 22.6 \pm 0.1$ °C. Error bars represent 95% confidence interval. The error bars shown in this figure are representative of all data presented in later figures.

Fig. 3.
Air-water interfacial area vs. liquid flow rate for glass beads; $T = 22.6 \pm 0.1$ $^{\circ}\text{C}$

Fig. 4. Air-water interfacial area vs. liquid flow rate for 45/50 sand; Column $d = 1.4 \text{ mm}$, $T = 22.6 \pm 0.1 \text{ }^\circ\text{C}$

Fig. 5.
Air-water interfacial area vs. NaOH solution concentration for glass beads; column $d = 1.4$ mm, $T = 22.6 \pm 0.1$ °C

Fig. 6.
Air-water interfacial area vs. NaOH solution concentration for 45/50 sand; column $d = 1.4$ mm, $T = 22.6 \pm 0.1$ °C

Fig. 7.
Air-water interfacial area vs. solution temperature for glass beads.

Table 1

Specifications of the column and porous media used in this study.

Item	Value	
Inner radius of the column, cm	1.3 and 0.7	0.7
Height of the packing, cm	5	2.6
Packing media	Glass beads	45/50 sand
Diameter of porous medium, mm	1.16	0.35
Porosity of packed porous medium	0.38	0.35
Particle density of porous medium, g/cm ³	2.20	2.64
Bulk density of packed porous medium, g/cm ³	1.41	1.71

Bulk density ρ_b = Total packing mass / Total packing volume