

μ -CT-Study of CO₂-Gas Exsolution caused by Hydrophobic Nucleation Sites

Bilal Zulfiqar¹ and Helmut Geistlinger¹

¹Helmholtz Centre for Environmental Research UFZ Halle

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Abstract

Background: Gas exsolution from supersaturated water injection (SWI) into porous media is of primary interest and has important applications (CCS, residual NAPL-remediation, CO₂-enhanced oil recovery). Bubble formation and growth kinetics are typically studied in 2D-micro-models (sandstones-analogues; Zuo et al., 2013; Zuo and Benson, 2013). The only 3D-study was conducted by Li et al., 2017, using the NMR-method, and indicated a strong dependency of the exsolution process on the initial CO₂-gas phase. However, the NMR-method was not able to attribute these different fluid-fluid-saturations to key parameters of the dissolution process such as gas cluster morphology and gas cluster size distribution. Methods: We conduct a series of column experiments to study the gas exsolution and gas cluster formation in 3D-porous media (natural sands, glass beads) using μ -CT. Based on this high-resolution non-invasive visualization method followed by image processing, we quantify (i) gas-cluster morphology, (ii) gas-cluster size distribution, (iii) correlation between pore structure and bubble formation, and (iv) the impact of surface roughness on exsolution efficiency. Results: We found that CO₂-saturated water equilibrated under ambient pressure, pCO₂ = 1.013 bar, (no supersaturation was measured and no pressure reduction was applied), already leads to gas exsolution of significant amount (about 10–12% gas saturation) in the presence of untreated SiO₂-surfaces (natural sands, glass beads) which exhibit heterogeneous wettability. To the best of our knowledge this exsolution phenomenon was not observed before, and we assume it to be caused by fluid-rock interactions, i.e. by hydrophobic nucleation sites at the siliceous surface. The heterogeneous wettability has a dramatic impact on capillary trapping efficiency and was experimentally observed in Glass-beads monolayer (Geistlinger and Ataei, 2015, Influence of the heterogeneous wettability on capillary trapping in glass-beads monolayers: Comparison between experiments and the invasion percolation theory, J. Colloid Interface Sci., 459, 230 - 240).

Objectives

- Injection of CO₂-Super-Saturated water and subsequent Gas Exsolution → Important applications [CCS, residual NAPL-remediation, CO₂-enhanced oil recovery]
- New Gas Exsolution Phenomena of CO₂-Equilibrated-water in Porous Media with ROUGH SURFACES
- μ -CT-Column Experiments to study Gas Exsolution and Gas Cluster Formation/Distribution in 3D-porous media (natural sand, glass beads, fine sand)
- New Result: CO₂-saturated water injection, already leads to Gas Exsolution of significant amount
- Heterogeneous Fluid-Rock Interactions, e.g. Hydrophobic nucleation sites and heterogeneous wettability has a dramatic impact on Capillary Trapping Efficiency
- RESULTS: Residual Gas saturation in Glass beads (1mm): 12-18%, Natural sand (1mm): 16 - 23%, Fine sand (0.17 mm):
- Key-parameter for GAS-cluster-dissolution: GAS-water-Interface $A_{g,w}$ (see Fig.5 and Table 1)
- Gas-water interface $A_{g,w}$ is about 60% of the total gas surface A_g

Methods

- pH electrode to control CO₂-concentration
- Gas-saturated water injection; hydraulic potential method (pre-defined Δh)
- Fluid-saturation measured; a) gravimetrically b) μ -CT

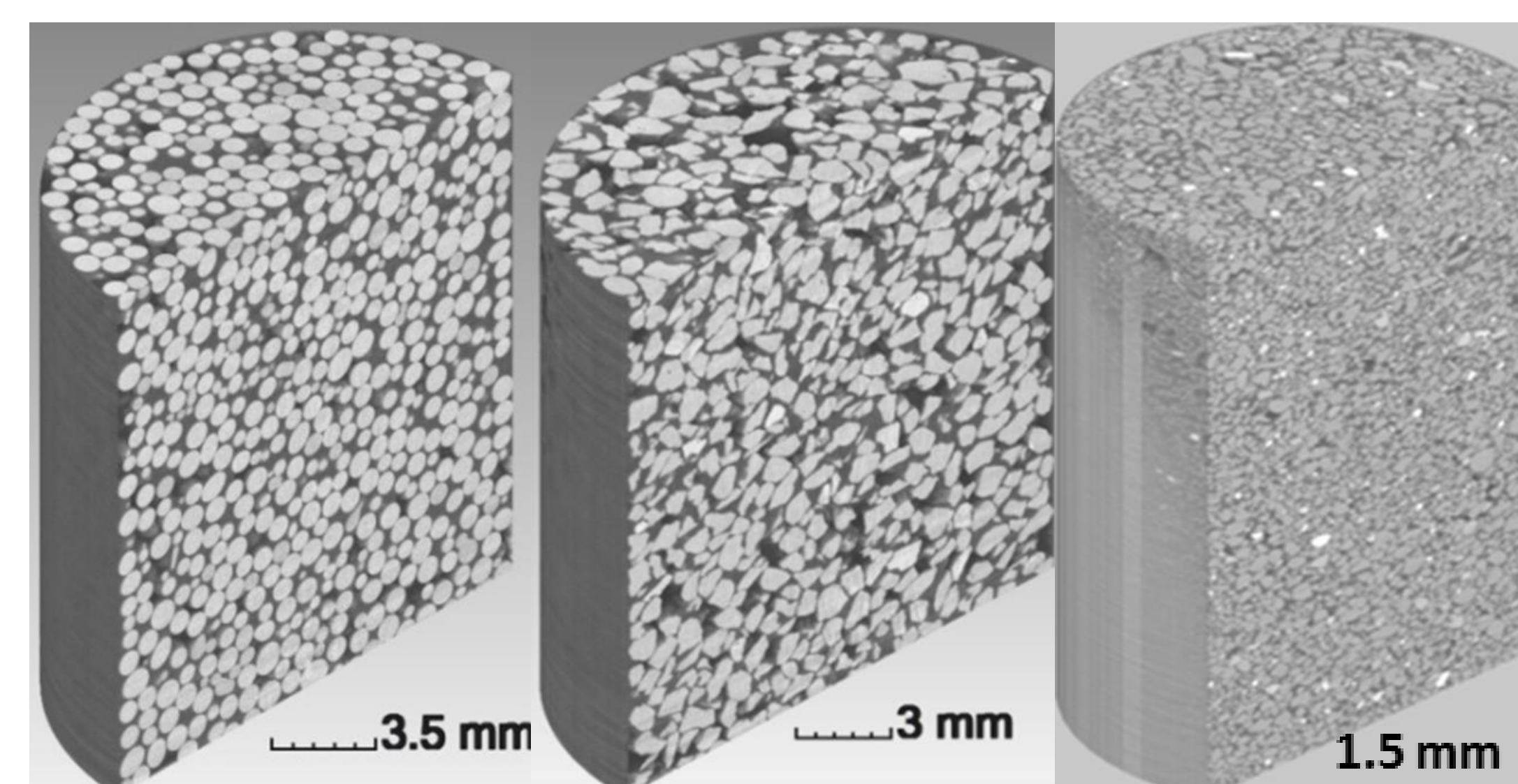


Fig 1. Raw CT images for 1mm-Natural sand, 1mm-GBS and fine sand

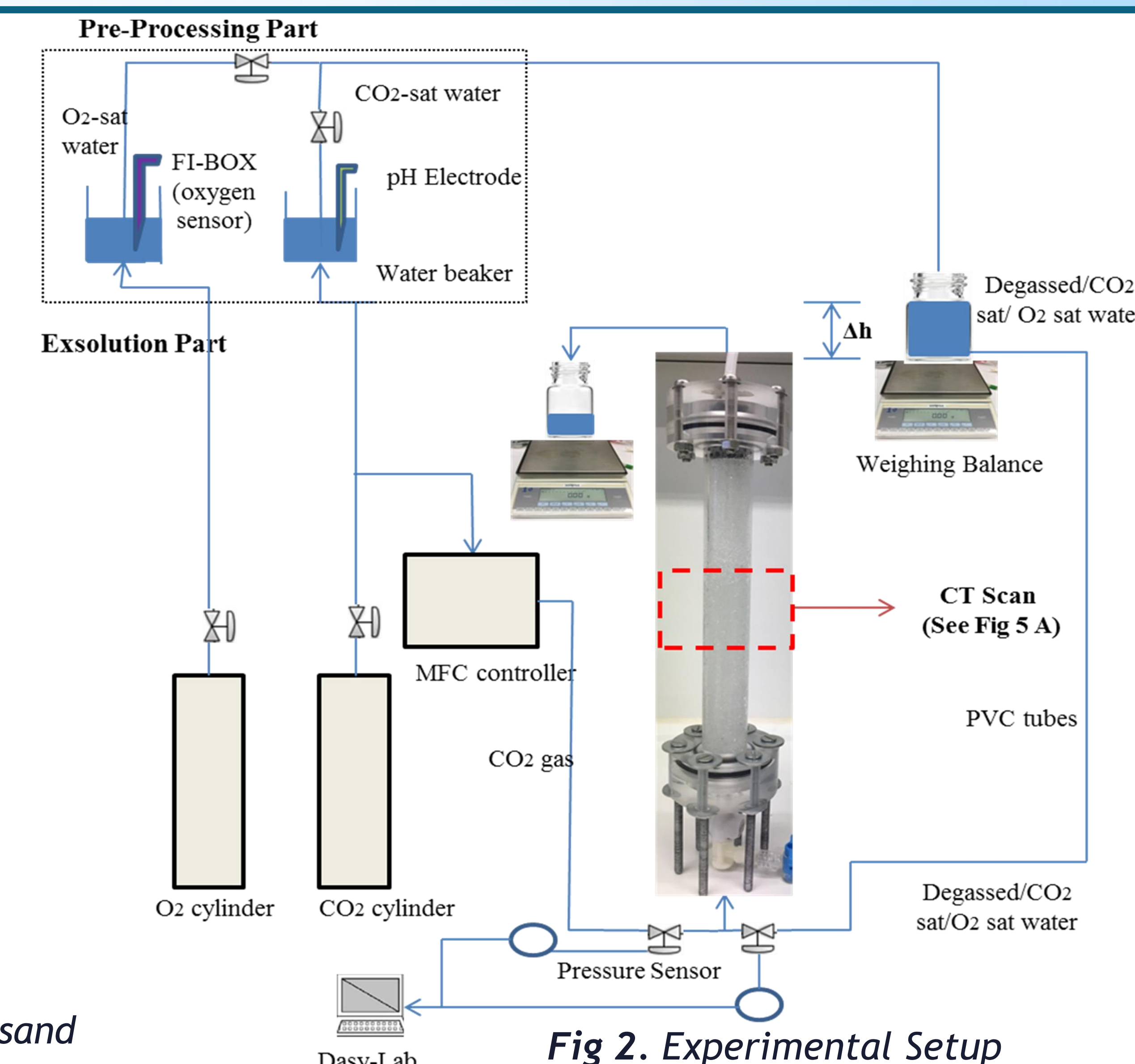
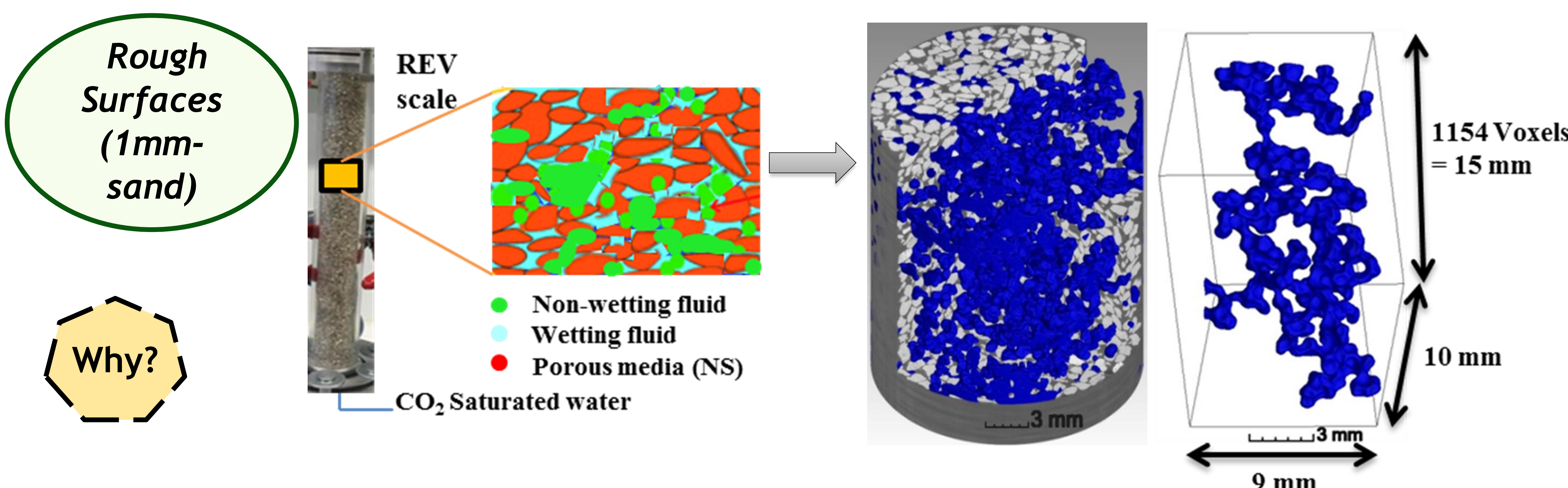
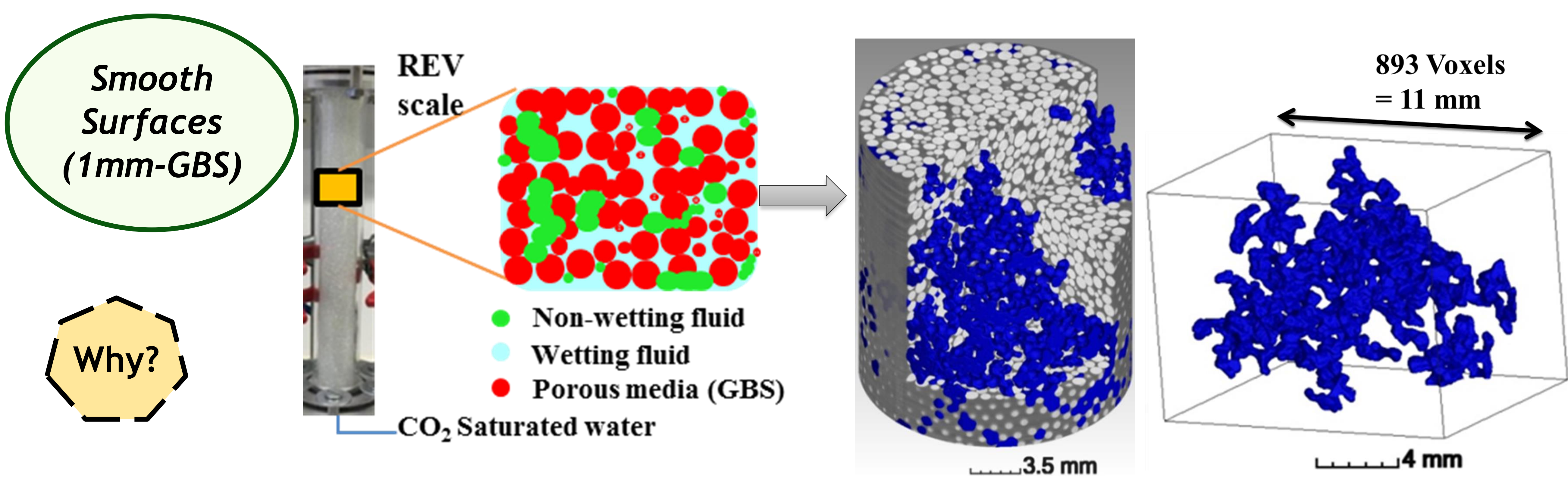


Fig 2. Experimental Setup

Results & Discussions: Gas Exsolution



- Rough surfaces exhibit **gas-filled cavities** and gas bubbles of different sizes which can be formed under certain thermodynamic conditions (see Fig.3)



- Chemisorbed molecules** (see Fig. 4)

Surface charge

Adsorbed dipole moment

Wetting behavior

Exsolution Process

A) Rough Surface

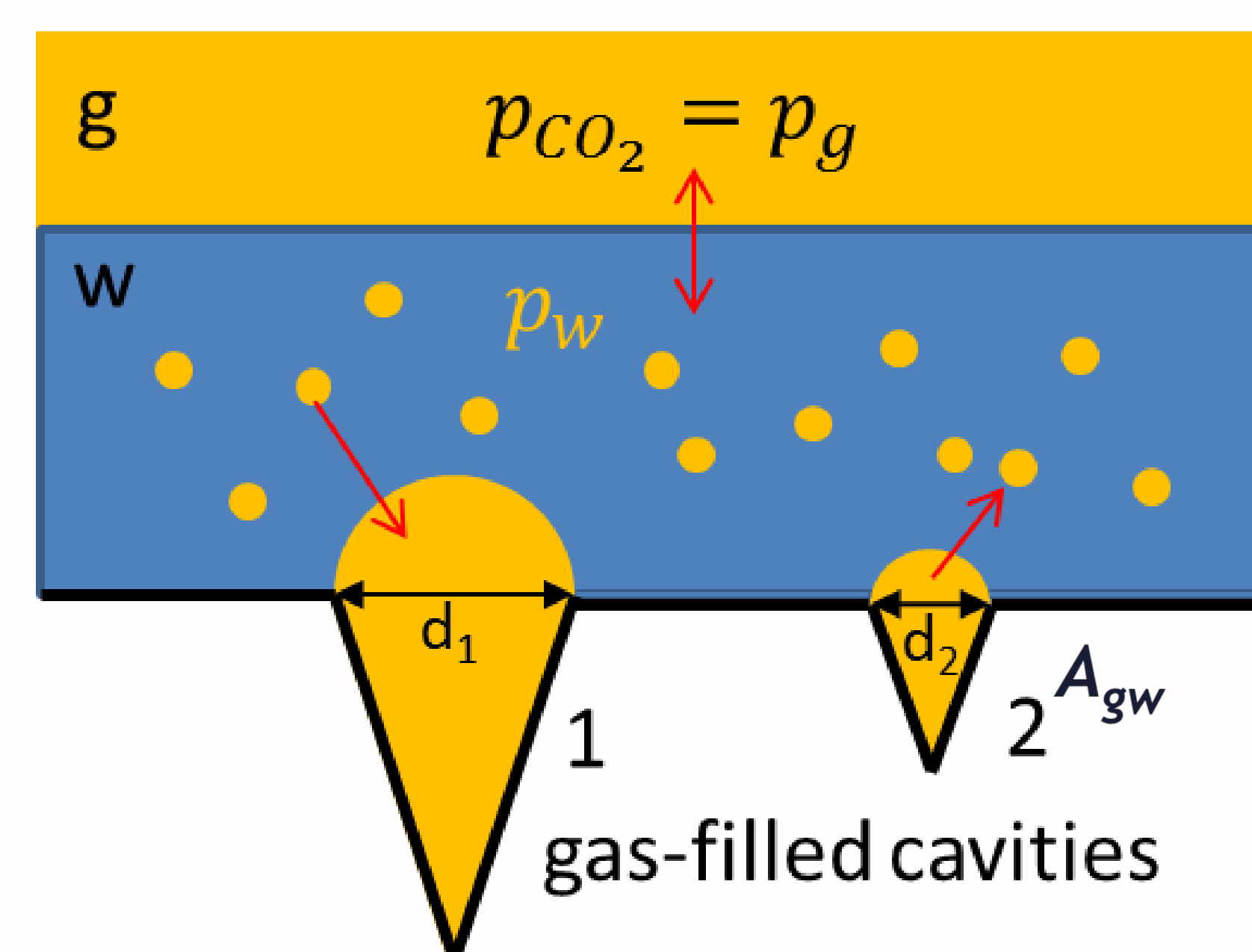


Fig 3. Bubble formation at two gas filled cavities of different size d_1 and d_2

B) Smooth Surface

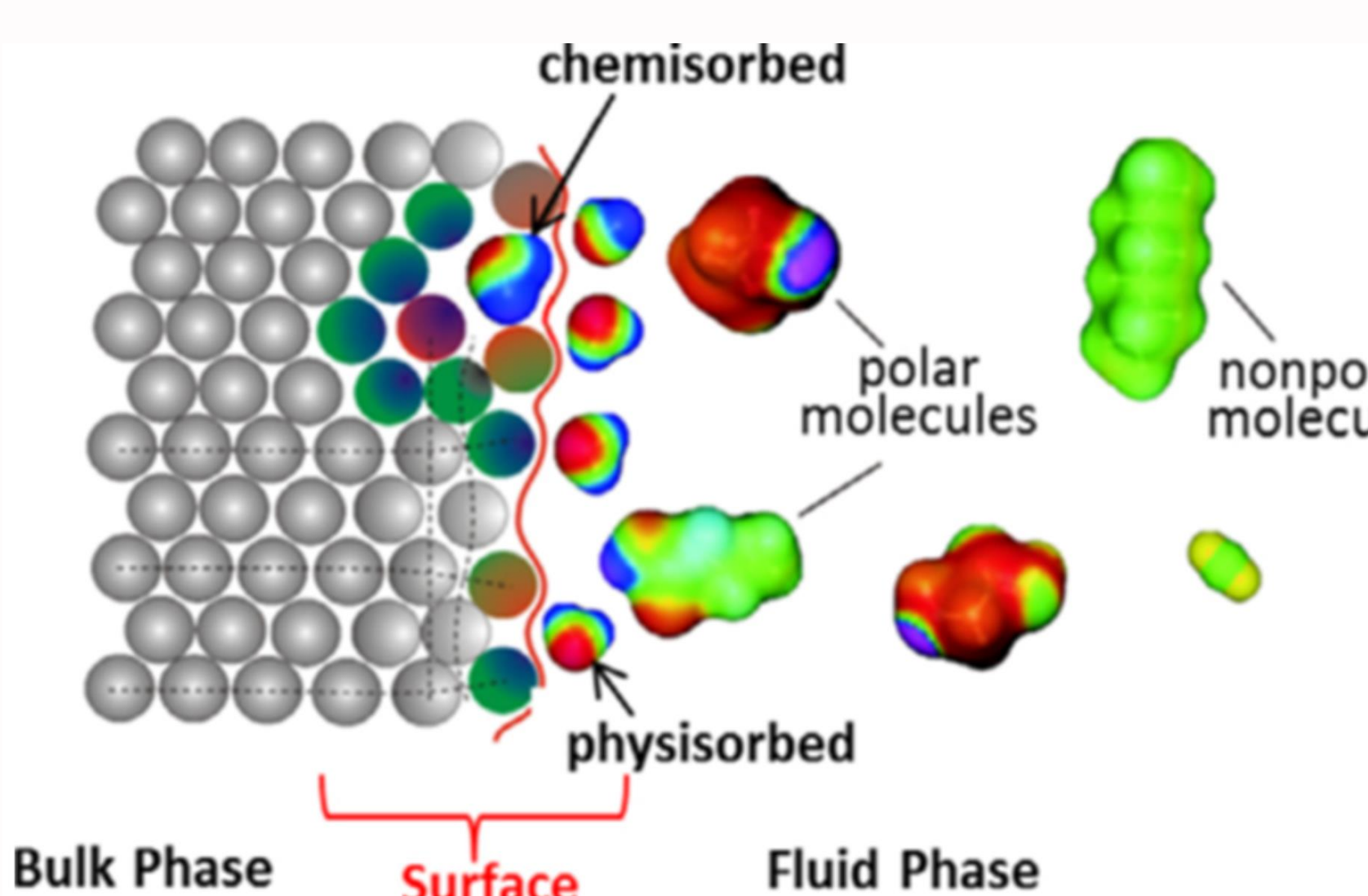
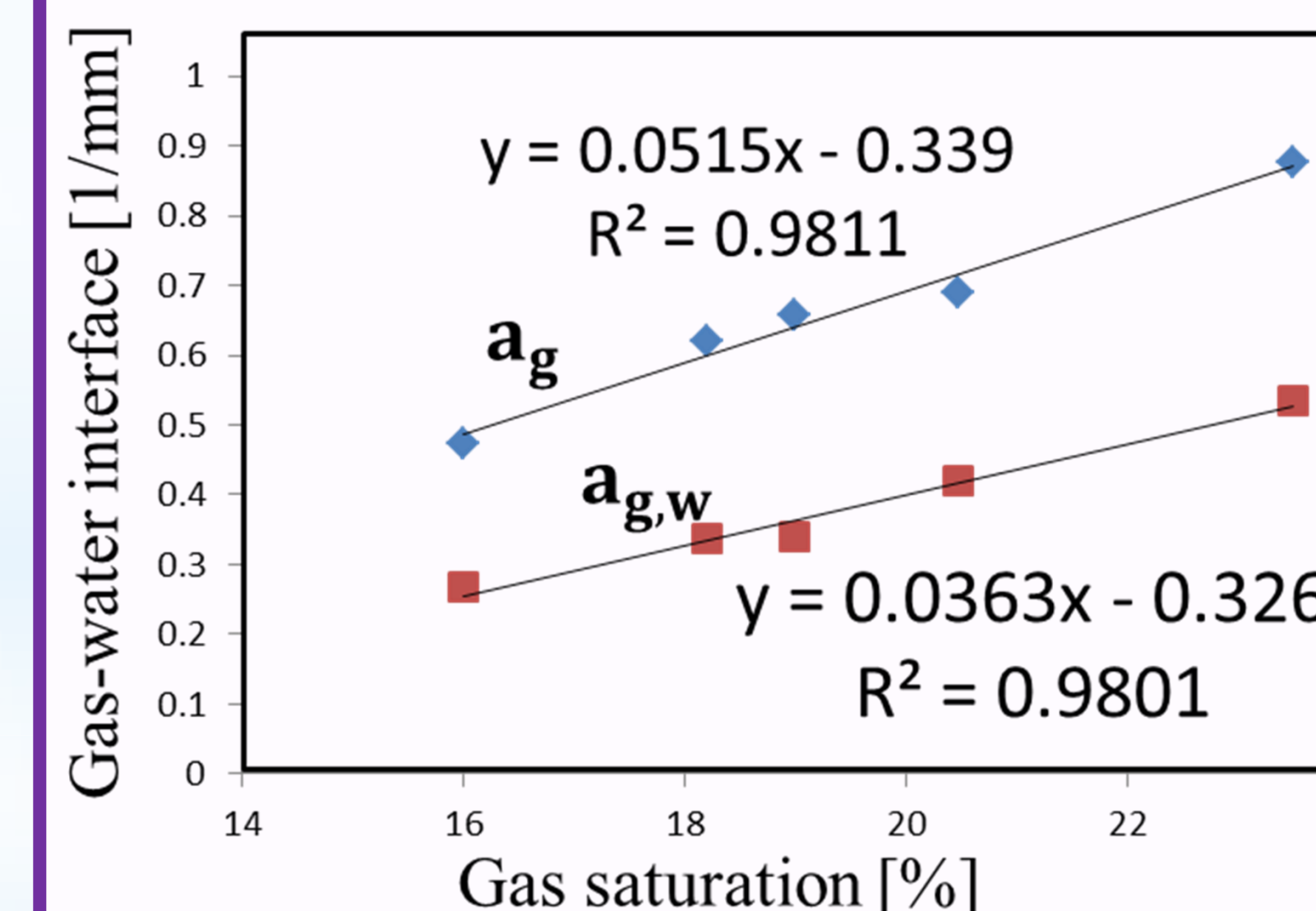


Fig 4. Real solid surface with faults and physisorbed & chemisorbed polar molecules present in the fluid phase

Hydrophobic nucleation sites ??

Quantification of the Gas-Water Interface

A) 1mm-Sand



B) GBS

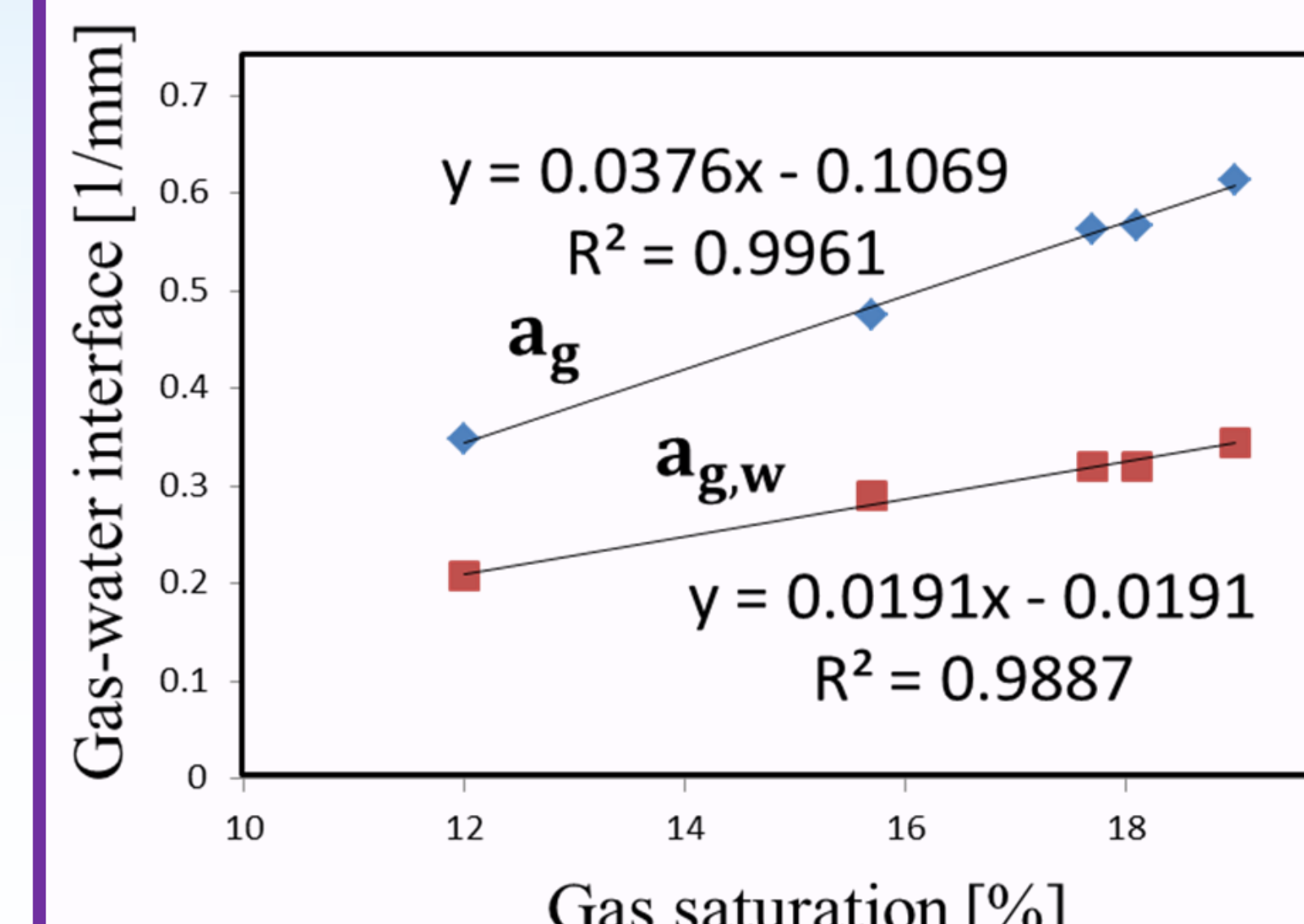


Fig 5. Gas-Water interface vs Gas saturation.

Exp	Porosity [-]	Gas Content [-]	A_g (mm ²)	$A_{g,w}$ (mm ²)
1	0.377	0.160	2765	1568
2	0.380	0.182	3597	1957
3	0.392	0.190	3877	2008
4	0.390	0.205	4140	2530
5	0.385	0.235	5208	3185

Exp	Porosity [-]	Gas Content [-]	A_g (mm ²)	$A_{g,w}$ (mm ²)
1	0.373	0.120	2078	1238
2	0.370	0.157	2886	1757
3	0.371	0.177	3376	1925
4	0.387	0.181	3405	1922
5	0.374	0.190	3584	2015

Table 1 Gas saturation & Gas-water interface.

References

- Geistlinger, H. et al., (2015), The impact of pore structure and surface roughness on capillary trapping for 2-D and 3-D porous media: Water Resour. Res., 51, 9094-9111
- Geistlinger, H et al., (2015), Influence of heterogeneous wettability on capillary trapping in glass-beads monolayers: Journal of Colloid & Inter Sci., 459, 230-240
- Li et al., (2017), Experimental Investigation on the Behavior of Supercritical CO₂ during Reservoir Depressurization. Environ. Sci. Technol., 51, 8869-8876.
- Zuo, L., and S. M. Benson (2014), Process-dependent residual trapping of CO₂ in sandstones, Geophys. Res. Lett., 41, 2820-2826

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www.testum-aquifer.de
Contact: bilal.zulfiqar@ufz.de