

# Supporting Information: Multiscale experimental study of H<sub>2</sub>/brine multiphase flow in porous rock characterizing relative permeability hysteresis, hydrogen dissolution, and Ostwald ripening

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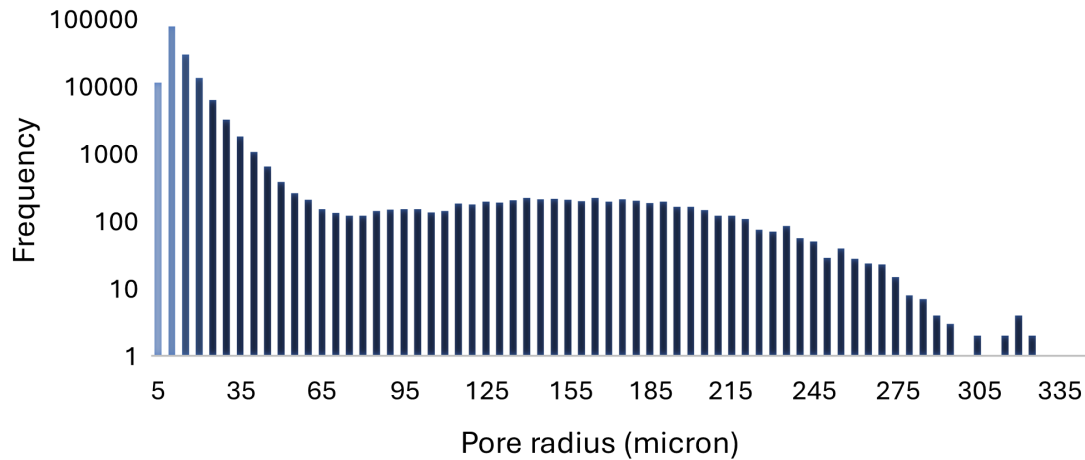
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## 1 Pore-size distribution

The pore-size distribution of the Berea sandstone used in the experiment can be seen in Supplementary figure 1. It was obtained from high resolution CT images (6.5  $\mu\text{m}$  voxel size) of a 1 cm long section in the middle of the core.



**Supplementary figure 1.** Pore size distribution of the Berea Sandstone rock sample obtained using high-resolution CT imaging of a 1 cm long section at the center of the core. The light blue shade indicates a larger possible error. These pore sizes are close to the voxel size (6.5  $\mu\text{m}$ ) and could, therefore, potentially be attributed to noise.

## 2 Overview of experimental studies

| Study                               | Lithology               | Core dimensions<br>[cm] (D) / [cm] (L) | Temperature<br>[°C] | Pressure<br>[bar] | Porosity<br>(%) | Permeability<br>[mD] |
|-------------------------------------|-------------------------|----------------------------------------|---------------------|-------------------|-----------------|----------------------|
| Yekta et al., 2018 <sup>1</sup>     | Vosges Sandstone        | 1.5 / 6.1                              | 20-45               | 55-100            | 19              | 44                   |
| Jha et al., 2021 <sup>2</sup>       | Gosford Sandstone       | 0.5 / 1.5                              | 20                  | 1                 | 18              | -                    |
| Al-Yaseri et al., 2022 <sup>3</sup> | Fontainebleau Sandstone | 3.9 / 5.5                              | 25                  | 4                 | 10              | 190                  |
| Rezaei et al., 2022 <sup>4</sup>    | Sandstones & Carbonate  | 3.8 / 8.9-9.5                          | 80                  | 1-206.8           | 10.5-16.4       | 3-34                 |
| Boon et al., 2022 <sup>5</sup>      | Berea Sandstone         | 3.8 / 17                               | 18                  | 100               | 20              | 203                  |
| Lysyy et al., 2022 <sup>6</sup>     | Berea Sandstone         | 3.8 / 27.6                             | 30                  | 30                | 18              | 107                  |
| Thaysen et al., 2023 <sup>7</sup>   | Clashach Sandstone      | 0.5 / 5.4-5.7                          | 20                  | 20-70             | 13-14           | -                    |
| Jangda et al., 2023 <sup>8</sup>    | Bentheimer Sandstone    | 0.6 / 2.7                              | 50                  | 100               | 23              | 2900                 |
| Zhang et al., 2023 <sup>9</sup>     | Bentheimer Sandstone    | 0.6 / 2.9                              | 25                  | 10                | -               | -                    |
| Goodarzi et al., 2024 <sup>10</sup> | Bentheimer Sandstone    | 1.3 / 6.0                              | 25                  | 10                | 22.6            | 1400                 |
| Higgs et al., 2024 <sup>11</sup>    | Bentheimer Sandstone    | 0.6 / 1.3                              | 25                  | 21.3              | 23              | 653                  |
| This study                          | Berea Sandstone         | 1.0 / 17                               | 25                  | 50                | 18.3            | 104                  |

**Supplementary table 1.** Rock type and experimental conditions for previous relative permeability and residual trapping studies. Empty fields indicate that the data was either not determined in the study or not provided by the authors. This table is based on data gathered by Higgs et al.<sup>11</sup> and has been modified and extended.

| Study                               | Capillary number<br>range [-]                               | Displacement<br>methodology | Flow<br>orientation | Imaging<br>technique |
|-------------------------------------|-------------------------------------------------------------|-----------------------------|---------------------|----------------------|
| Yekta et al., 2018 <sup>1</sup>     | $1.17-1.25 \times 10^{-8}$ <sup>(d)</sup>                   | Steady-state                | Vertical            | No                   |
| Jha et al., 2021 <sup>2</sup>       | $2.49 \times 10^{-10} - 3.68 \times 10^{-8}$ <sup>(i)</sup> | Unsteady-state              | Vertical            | Micro-CT             |
| Al-Yaseri et al., 2022 <sup>3</sup> | $3.54 \times 10^{-8}$ <sup>(d)</sup>                        | Unsteady-state              | Vertical            | NMR                  |
| Razaei et al., 2022 <sup>4</sup>    | -                                                           | Unsteady-state              | Vertical            | No                   |
| Boon et al., 2022 <sup>5</sup>      | $9.1 \times 10^{-9} - 1.1 \times 10^{-6}$ <sup>(d)</sup>    | Steady-state                | Horizontal          | Medical CT           |
| Lysyy et al., 2022 <sup>6</sup>     | $1.88 \times 10^{-9} - 1.74 \times 10^{-7}$ <sup>(d)</sup>  | Steady-state                | Vertical            | X-ray (unspecified)  |
| Thaysen et al., 2023 <sup>7</sup>   | $1.2 \times 10^{-8} - 9.5 \times 10^{-6}$ <sup>(i)</sup>    | Unsteady-state              | -                   | Micro-CT             |
| Jangda et al., 2023 <sup>8</sup>    | $4.2 \times 10^{-9} - 2.3 \times 10^{-6}$ <sup>(d)</sup>    | Unsteady-state              | Vertical            | Micro-CT             |
| Zhang et al., 2023 <sup>9</sup>     | $2.55 \times 10^{-9} - 2.62 \times 10^{-7}$                 | Unsteady-state              | Vertical            | Micro-CT             |
| Goodarzi et al., 2024 <sup>10</sup> | $1.93 \times 10^{-7} - 5.79 \times 10^{-9}$ <sup>(d)</sup>  | Unsteady-state              | Vertical            | Micro-CT             |
| Higgs et al., 2024 <sup>11</sup>    | $2.92 \times 10^{-7} - 2.92 \times 10^{-5}$ <sup>(d)</sup>  | Steady-state                | Vertical            | Micro-CT             |
| This study                          | $1.3 \times 10^{-8} - 1.3 \times 10^{-6}$ <sup>(d)</sup>    | Steady-state                | Vertical            | Micro-CT             |

<sup>(d)</sup> Determined with Darcy velocity <sup>(i)</sup> Determined with Interstitial velocity

**Supplementary table 2.** Experimental methods of previous relative permeability and residual trapping studies. Empty fields indicate that the data was either not determined in the study or not provided by the authors. This table is based on data gathered by Higgs et al.<sup>11</sup> and has been modified and extended.

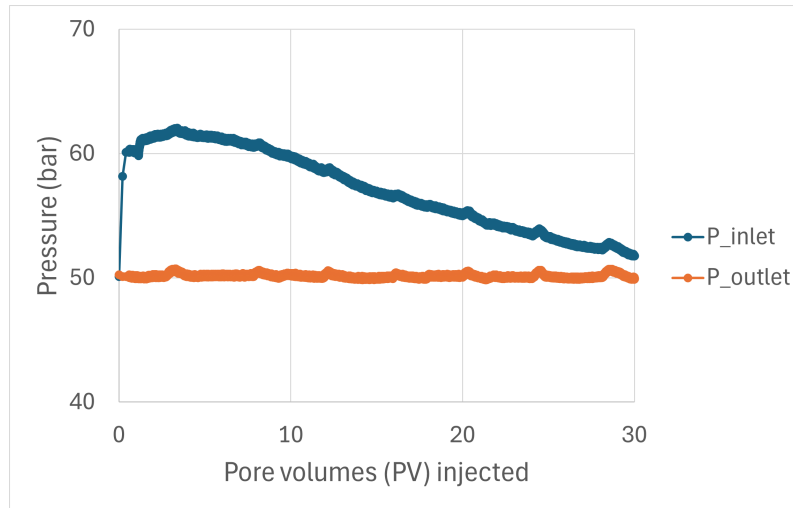
| Study                               | Initial gas<br>saturation [-]             | Residual gas<br>saturation [-]            | Hydrogen end-point<br>relative permeability [-] | Max. pressure<br>drop [bar]               |
|-------------------------------------|-------------------------------------------|-------------------------------------------|-------------------------------------------------|-------------------------------------------|
| Yekta et al., 2018 <sup>1</sup>     | 0.59-0.6                                  | -                                         | 0.035-0.044                                     | 0.9 <sup>(D)</sup> - 2.0 <sup>(D)</sup>   |
| Jha et al., 2021 <sup>2</sup>       | 0.65 <sup>(x)</sup>                       | 0.41 <sup>(x)</sup>                       | -                                               | -                                         |
| Al-Yaseri et al., 2022 <sup>3</sup> | 0.04 <sup>(x)</sup>                       | 0.02 <sup>(x)</sup>                       | -                                               | -                                         |
| Rezaei et al., 2022 <sup>4</sup>    | 0.22-0.28                                 | -                                         | 0.260-0.360                                     | -                                         |
| Boon et al., 2022 <sup>5</sup>      | 0.36 <sup>(y)</sup>                       | 0.24-0.17 <sup>(y)</sup>                  | 0.019                                           | 0.6 <sup>(D)</sup> - 4.9 <sup>(I)</sup>   |
| Lysyy et al., 2022 <sup>6</sup>     | 0.41 <sup>(x)</sup>                       | 0.36 <sup>(x)</sup>                       | 0.037-0.083                                     | 0.3 <sup>(D)</sup> - 4.2 <sup>(I)</sup>   |
| Thaysen et al., 2023 <sup>7</sup>   | 0.50-0.53                                 | 0.1-0.21                                  | -                                               | -                                         |
| Jangda et al., 2023 <sup>8</sup>    | 0.36 <sup>(x)</sup> - 0.36 <sup>(y)</sup> | 0.20 <sup>(x)</sup> - 0.25 <sup>(y)</sup> | -                                               | -                                         |
| Zhang et al., 2023 <sup>9</sup>     | 0.53 <sup>(x)</sup>                       | 0.07 <sup>(x)</sup>                       | -                                               | -                                         |
| Goodarzi et al., 2024 <sup>10</sup> | 0.30 <sup>(x)</sup>                       | 0.10 <sup>(x)</sup>                       | -                                               | -                                         |
| Higgs et al., 2024 <sup>11</sup>    | 0.76 <sup>(x)</sup>                       | 0.40 <sup>(x)</sup>                       | 0.049                                           | 0.1 <sup>(D)</sup> - 0.9 <sup>(I)</sup>   |
| This study                          | 0.44 <sup>(y)</sup>                       | 0.32 <sup>(y)</sup>                       | 0.043                                           | 18.0 <sup>(D)</sup> - 15.9 <sup>(I)</sup> |

<sup>(x)</sup> Determined with non-equilibrated aqueous phase <sup>(y)</sup> Determined with equilibrated aqueous phase <sup>(D)</sup> Drainage <sup>(I)</sup> Imbibition

**Supplementary table 3.** Results from previous relative permeability and residual trapping studies. Empty fields indicate that the data was either not determined in the study or not provided by the authors. This table is based on data gathered by Higgs et al.<sup>11</sup> and has been modified and extended.

### 3 Pressure profile - last step of the imbibition experiment

After the second no-flow period, 30 PV of equilibrated brine were injected into the core. The pressure evolution at the inlet and outlet can be seen in Supplementary figure 2.



**Supplementary figure 2.** Pressure versus pore volume graph for the last step of the imbibition experiment. 30 pore volumes (PV) of pre-equilibrated brine were injected after a no-flow period.

### References

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