



Contribution ID: 396

Type: Oral Presentation

## Long-term redistribution of residual gas due to non-convective transport

Wednesday, 2 June 2021 19:20 (15 minutes)

Global climate change due to high dependence on fossil fuel has necessitated the need to deploy decarbonization technologies. Carbon capture and storage (CCS), a technology that stores CO<sub>2</sub> permanently in the subsurface, is largely seen as necessary to reduce CO<sub>2</sub> emissions from large-scale industrial sources. As CCS technology has become commercially viable, recent years have witnessed a large expansion of CCS projects driven by innovative incentive mechanisms and sustainable government support. Although mitigating global warming is a long-term goal, current engineering methods for CCS projects are focused on short-term operational parameters, typically 20-30 years. While CO<sub>2</sub> is expected to remain securely trapped in the subsurface, the long-term fate of injected CO<sub>2</sub> over thousands of years has yet to be investigated.

An important trapping mechanism for injected CO<sub>2</sub> is residual trapping, where a large fraction of injected CO<sub>2</sub> is disconnected into ganglia trapped in the pore spaces by capillary forces. A widely accepted theory is that residual trapping is responsible for permanent entrapment, as the trapped CO<sub>2</sub> ganglia is assumed to remain immobile until they eventually dissolve into the reservoir brine. However, field observation that supports this theory is hardly available due to long time scales of dissolution. In previous studies, we have shown that the residually trapped CO<sub>2</sub> is thermodynamically unstable due to capillary heterogeneity. This instability induces Ostwald ripening among residually trapped CO<sub>2</sub>, thereby redistributing the gas phase across the system despite the gas phase remaining immobile. This redistribution causes residually trapped CO<sub>2</sub> to re-aggregate and potentially remobilize, undermining the long-term security of geological CO<sub>2</sub> storage.

In this study, we evaluate the thermodynamic stability of residually trapped CO<sub>2</sub> from a fundamental perspective. By analyzing the chemical potential of the residually trapped CO<sub>2</sub>, we identify that in addition to capillary heterogeneity, both hydrostatic pressure and geothermal gradients lead to thermodynamic instability of residually trapped CO<sub>2</sub>. These driving forces induce non-convective transport of dissolved CO<sub>2</sub> in the aqueous phase, thereby redistributing the residually trapped CO<sub>2</sub> throughout the system in different directions. The resulting non-convective transport is a combination of multiple mass transfer mechanisms, including molecular diffusion, sedimentation of solutes and the Soret effect.

Furthermore, we study the characteristics of the non-convective transport and the resulting redistribution of residually trapped CO<sub>2</sub> through numerical simulation. Results indicate that hydrostatic pressure dominates the redistribution of residually trapped CO<sub>2</sub> by inducing molecular diffusion of dissolved CO<sub>2</sub>. This diffusive flux is upward such that the gas phase depletes in the bottom and accumulates at the top under the seal. Geothermal gradients can somewhat mitigate this accumulation by reducing the CO<sub>2</sub> solubility gradient. This diffusive flux depletes residually trapped gas in a rate of  $10^5$  years / m approximately. Although the non-convective transport is slow, the residually trapped CO<sub>2</sub> accumulates under the seal inevitably in a shorter period. Re-aggregation and remobilization of residually trapped CO<sub>2</sub> under the seal is highly likely to occur, which will be characterized in our future study.

### Time Block Preference

Time Block C (18:00-21:00 CET)

### References

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**Session Classification:** MS6-A

**Track Classification:** (MS6-A) Physics of multi-phase flow in diverse porous media