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Pore-scale Diffusive Mixing Between Hydrogen and Carbon Dioxide: Implications for Underground Hydrogen Storage

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Underground hydrogen storage (UHS) is recognized as a viable solution for storing significant amounts of hydrogen (H₂) in the advancement of a low carbon energy system. The diffusive mixing between H₂ and cushion gas, which leads to a reduction in the purity of produced H₂, persists through the entire storage period. Therefore, it is crucial to understand the diffusive mass transfer and its impact on the migration of H₂. Due to experimental challenges, there is limited research dedicated to quantifying the diffusion between H₂ and cushion gas under reservoir conditions. For the first time, we measured the diffusive process between H₂ and CO₂ (acting as the cushion gas) in a high-pressure capillary cell in situ. The time-dependent Raman spectroscopy was used to monitor the diffusive mass transfer, and the diffusion coefficient was determined based on the measured concentration profiles. We showed that the diffusive process was adequately captured by the Fick's second law with a constant diffusion coefficient. The diffusion coefficient scales linearly with the reciprocal viscosity of CO₂ across the pressure and temperature range associated with subsurface storage conditions. Based on the measured diffusion coefficient, we quantified the diffusion distance for the mixing occurring at field-scale. Results indicate that the mixing between H₂ and CO₂ can reach tens of meters over a one-year period. This suggests that the dispersive mixing plays a role in the purity of produced H₂.

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