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Is Hydrogen Really Stored by Adsorption? Revisiting Quantification in Geological Materials

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Hydrogen adsorption on natural minerals influences hydrogen mobility in deep geological environments, including natural hydrogen systems, underground hydrogen storage (UHS), and nuclear waste repositories. Under reservoir-relevant conditions (>20 °C, hydrated pore space, moderate surface areas of 10–50 $m^2\ g^{-1}$), expected H_2 uptake is extremely small, typically below 10^{-2} wt.%. This often approaches the detection limits of standard volumetric methods. At such low signal levels, cumulative uncertainties from void-volume calibration, sensor resolution, and sequential expansion steps can generate apparent Langmuir-type adsorption trends and systematic overestimation of adsorbed quantities.

To address this limitation, we designed and benchmarked a dual-cell differential volumetric apparatus specifically engineered to resolve low H_2 adsorption signals up to 100 bar. The system employs symmetric plumbing, large-volume sample cells, high-precision differential pressure sensing, and complete immersion of the gas-accessible volume in a thermostated water bath to suppress thermal gradients and common-mode drift. A unified metrological framework integrates full uncertainty propagation with real-gas corrections using the multiparameter Helmholtz equation of state (REFPROP).

Building on this validated methodology, we investigated a suite of reference adsorbents, swelling and non-swelling clays (Laponite, Kunipia F, Sumecton SA), clay-rich caprocks (Callovo–Oxfordian claystone, Boom Clay), and selected geological materials under controlled dry and hydrated states. Across clay-rich materials, excess H_2 adsorption at ambient temperature remains at or below ~0.05 mmol g^{-1} (~0.01 wt.%), frequently approaching the statistically defined Lower Limit of Quantitation (LLOQ ≈ 0.03 mmol g^{-1} or ~0.006 wt.%). Under hydrated conditions, differential pressure evolution differs from the dry state, consistent with volumetric changes associated with swelling and modified gas accessibility. In swelling clays, the resulting excess uptake shifts toward or within the LLOQ envelope, limiting quantitative interpretation of adsorption changes.

These results provide experimentally constrained upper bounds for H_2 adsorption in natural rocks and demonstrate that, under realistic subsurface conditions, H_2 retention by mineral adsorption is limited and highly sensitive to water content. By explicitly defining metrological thresholds and hydration effects, this work establishes a quantitative framework for evaluating H_2 mobility, retention, and risk in geological system