

# Similarity and dissimilarity of factors controlling methane and hydrogen gas adsorption in geologic formations

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Natural reservoirs and underground storage of two major energy-bearing gases, methane (CH<sub>4</sub>) and molecular hydrogen (H<sub>2</sub>), require the presence of ultra-low permeability rocks, either forming the gas reservoir (usually a shale) or acting as a barrier cap rock (shale, bentonite) in porous reservoirs. Clay minerals and organic matter (OM - kerogen, solid bitumen) are the compounds responsible for forming a complex, tortuous, nm-scale pore network.

A significant portion of CH<sub>4</sub> and H<sub>2</sub> gas can be physisorbed in a rock's micro- and mesopores. Due to the different effective diameters of CH<sub>4</sub> and H<sub>2</sub>, and different adsorption enthalpies, the gases can enter different adsorption sites.

In OM-poor rocks and sediments, clay minerals exclusively form the microporosity available for gases to adsorb, whereas the mesopore (>2 nm diameter) surface area and volume do not play an important role. The CH<sub>4</sub> and H<sub>2</sub> adsorption capacity is primarily controlled by the texture of the clay particles represented by the crystallites' planar dimensions and the way they aggregate. The adsorption sites are located mainly outside the interlayer galleries of non-expandable and expandable clays unless they are open wide enough (e.g. pillared by organic cations or incompletely dried cations of high hydration enthalpy). The CH<sub>4</sub> adsorption volume in OM-poor rocks is approximated by N<sub>2</sub>-accessible (at 77K) micropore volume, however, H<sub>2</sub> can enter narrower interlayers even unavailable for CH<sub>4</sub>, resulting in increased H<sub>2</sub> over CH<sub>4</sub> selectivity in shales and bentonites.

In OM-rich shales, the CH<sub>4</sub> and H<sub>2</sub> adsorption is predominantly controlled by the OM micropore texture. While both H<sub>2</sub> and CH<sub>4</sub> occupy the same micropore volume, H<sub>2</sub> seems to propagate into even finer sites. That adsorption volume can be linearly approximated by the micropore volume available for CO<sub>2</sub> adsorption at ~273K.

Because of the density of the adsorbed phase, the total gas content in a pore system can be greater than that based on the temperature-pressure relationship of the free phase. A quantitative model of adsorbed H<sub>2</sub> versus CH<sub>4</sub> in geologic formations is presented in respect to depth and geothermal gradients.