ScienceDirect

International Journal of Hydrogen Energy
Volume 50, Part D, 2 January 2024, Pages 879-892

H_2, CH_4 and CO_2 adsorption on Cameo coal: Insights into the role of cushion gas in hydrogen geological storage

Xiaoqiang Li, Xun Sun, Clifford C. Walters, Tongwei Zhang

Show more

Outline | Share | Cite

https://doi.org/10.1016/j.ijhydene.2023.08.185
Get rights and content

Highlights

- Adsorbed amounts of H_2 are much smaller than CH_4 and CO_2.
- Weak affinity of H_2 with coals is quantitatively evaluated.
- H_2 loss by adsorption during geological storage can be significantly reduced by

the injection of CH_4 or CO_2 as cushion gas.

Abstract

Large-scale hydrogen underground storage is widely considered as an important solution for energy transition to mitigate global warming, however, our understanding on the various chemical and physical processes leading to H_2 loss during geo-storage remains largely unknown. We conducted adsorption isotherms measurements of H_2, CH_4 and CO_2 on Cretaceous Cameo coal at 35, 50 and 65 °C, to explore adsorption mechanisms of H_2 and deciphering the effect of cushion gas (e.g., CH_4, CO_2) on H_2 loss in the subsurface. The adsorbed amounts of H_2 are just 12% and 6% of CH_4 and CO_2 at temperature of 35 °C and pressure of 2.3 MPa, respectively, and H_2 adsorption behavior on Cameo coal can be described well by Langmuir monolayer theory. The extremely high Langmuir pressures (35–49 MPa) for H_2 indicate very weak affinity with coal, compared to CH_4 and CO_2. Differences of Langmuir adsorption capacities suggest that the gas adsorption of H_2 and CH_4 are mainly by physical interactions while both physical and chemical bonding with coal structures occurs for CO_2. The affinity with solid materials for the three gases can be quantitatively evaluated by the heat of adsorption, which was determined to be ~9.2 kJ/mol for H_2. Extrapolation of our experimental results to natural coal seams indicates that H_2 presents much smaller adsorption amounts at depth up to 2000 m, corresponding to temperatures lower than 65 °C and pressure less than 30 MPa, geological conditions that are suitable for hydrogen underground storage. This study confirms that the injection of CH_4 or CO_2 as cushion gas can largely reduce the H_2 loss by adsorption in the subsurface. Empirical calculation suggests H_2 adsorption will be insignificant with chemical composition of CH_4 above 8% or CO_2 above 2% at the storage sites (e.g., abandoned mines, depleted coal seams). Our study lays a foundation on the understanding of the risk of H_2 loss in large-scale hydrogen underground storage.

Previous

Next

Keywords

Energy transition; Hydrogen underground storage; Competitive adsorption; Hydrogen loss

1. Introduction

Energy transition from conventional carbon-releasing fossils to clean renewables is a key step to reach net-zero CO_2 emissions and mitigate the climate change [[1], [2], [3], [4]]. Hydrogen (H_2) is widely considered as a future zero-carbon energy carrier to provide a reliable, sustainable and renewable energy supply with large-scale storage in the subsurface [[5], [6], [7], [8]]. Depleted oil/gas reservoirs, artificial salt caverns, aquifers, hard rock caverns, and abandoned mines are regarded as suitable sites for H_2 underground storage [[9], [10], [11], [12], [13], [14]]. Various interactions among gas-fluid-rock system have been evaluated for the characterization of H_2 flow and risk assessment in the hydrogen geological storage, for example interfacial tension [[15], [16], [17], [18]], wettability [[19], [20], [21]], diffusion [22,23], and contact angle [17,24,25]. Recovery of H_2 stored in the subsurface is impacted by chemical and biological consumption, diffusive loss through sealing rocks and adsorption [1,[26], [27], [28]]. The latter is an important geological factor that needs to be investigated for a better understanding of the adsorption mechanisms that occur in the subsurface storage of H_2 compared to other gases.

Several studies have investigated the adsorption of H_2 in different solid media for use as a transportable fuel [29,30]. Carbon nanomaterials, metal organic frameworks and activated carbons have been proposed as suitable matrices for H_2 storage [[31], [32], [33], [34]]. These studies indicated that the amounts of adsorbed H_2 positively correlate with micropore volume and specific surface area of the sample materials. H_2 storage using synthetic materials or in surface tanks is impractical for the large volumes required for a zero-carbon economy, shifting attention to subsurface reservoirs (e.g., depleted oil/gas reservoirs, salt caverns) [1,6]. Currently, limited studies have reported H_2 adsorption on geological samples with relatively high specific surface areas, such as coals at different maturities [23,35,36], oil shale [37] and montmorillonite-types clays [38]. The results on coal samples indicate that maturity is likely an important geological