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Optimum geological storage depths for structural H₂ geo-storage

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ABSTRACT

H₂ geo-storage has been suggested as a key technology with which large quantities of H₂ can be stored and withdrawn again rapidly. One option which is currently explored is H₂ storage in sedimentary geologic formations which are geographically widespread and potentially provide large storage space. The mechanism which keeps the buoyant H₂ in the subsurface is structural trapping where a caprock prevents the H₂ from rising by capillary forces. It is therefore important to assess how much H₂ can be stored via structural trapping under given geo-thermal conditions. This structural trapping capacity is thus assessed here, and it is demonstrated that an optimum storage depth for H₂ exists at a depth of 1100 m, at which a maximum amount of H₂ can be stored. This work therefore aids in the industrial-scale implementation of a hydrogen economy.

1. Introduction

Hydrogen geo-storage (UHS) has been identified as a potential solution to store large quantities of hydrogen, which is currently the main barrier to implementing an industrial-scale hydrogen economy (Lord et al., 2014; Zhang et al., 2016; Flesch et al., 2018; Heinemann et al., 2018; Tarkowski and Czapowski, 2018; ShiJessen and Tsotsis, 2020; Pan et al., 2021a). In UHS, H₂ is injected into subsurface geologic formations, and it can be withdrawn again at any time. One target formation currently investigated are sedimentary reservoirs, which are abundant and can have large storage capacities – such sedimentary reservoirs are also considered for CO₂ geo-sequestration (CGS, e.g. see IPCC 2005; Lackner, 2003), and they can classically also hold natural gas reserves, which are exploited industrially for a long time now (e.g. Dake, 1978). The buoyant gases (H₂, CH₄, CO₂) are stored by an impermeable seal layer, termed caprock by geologists (e.g. Dake, 1978; IPCC 2005; Wollenweber et al., 2010; Busch et al., 2008). Technically a caprock is also a sedimentary rock and it is also porous, but it has a very low permeability (e.g. Nelson, 2009; Sondergeld et al. 2010). The reason why the buoyant gases cannot percolate into the caprock is the high capillary entry pressure (Pₑ,c) of the caprock, which is again related to the very small pores in the caprock (Nelson, 2009).

However, if the buoyancy pressure exceeds Pₑ,c, then the buoyancy forces overcome the counter-acting capillary forces, and gas will indeed migrate upwards through the caprock (e.g. Iglauer et al., 2015a,b). This can be quantified by equation (1) – a buoyancy force – capillary force balance:

\[ h = \frac{2\gamma \cos \theta}{\rho g \Delta \rho} \]  

(equation 1)

where h is the column height of gas which can be permanently immobilized beneath the caprock, g is the gravitational constant, Δρ is the difference between water density (ρₜ) and gas density (ρ₂), γ is the gas-water interfacial tension and θ is the water-rock-gas contact angle (e.g. Arif et al., 2016). Now it has been previously demonstrated that in the context of CGS, h varies with storage depth as Δρ, γ and θ all vary significantly with depth (Iglauer, 2018). Consequently, a lower depth barrier exists below which CO₂ cannot be permanently stored by structural trapping (although note that interestingly below 15,000 m CO₂ is heavier than formation brine, and sinks spontaneously (due to gravitational forces) deep into the reservoir (Span and Wagner, 1996; Iglauer, 2018)). An optimum CO₂ storage depth also exists where a maximum amount of CO₂ can be stored. It is now hypothesized here that UHS also has an optimum storage depth (in terms of storing the largest mass of H₂), albeit H₂ will always be buoyant due to its high volatility in a geologic reservoir (Leachman et al., 2009). This hypothesis is examined in detail in the below section, and indeed the conclusion is reached that an optimum (technical) UHS depth exists, where a maximum amount of H₂ can be stored.

2. Methodology

It is clear that three parameters in equation (1) are affected by pressure and temperature, and thus depth, namely γ, Δρ, and θ. To
conduct the analysis, a hydrostatic gradient of 10 MPa/km and a geothermal gradient of 30 K/km are assumed—which reflect common subsurface conditions (Dake, 1978; Meckel, 2010). Each parameter is therefore discussed in detail below.

2.1. The H$_2$-water density difference $\Delta \rho$

H$_2$ is an extremely highly compressible gas, a consequence is that the temperature and pressure ranges relevant in UHS (300–360 K and 0.1–20 MPa) only have a relatively insignificant effect on H$_2$ density ($\rho_{\text{H}_2}$), Fig. 1. Compare this with CO$_2$, which undergoes a drastic change in density with increasing pressure and thus depth (Iglauer, 2018)–also $\rho_{\text{H}_2}$ is generally very low (e.g. $\rho_{\text{H}_2}$ is 3.5747 kg/m$^3$, while $\rho_{\text{CO}_2}$ is 100.22 kg/m$^3$ and $\rho_{\text{CH}_4}$ 30.897 kg/m$^3$ at 5 MPa and 330 K; Span and Wagner, 1996; Leachman et al., 2009). Furthermore, brine density ($\rho_{\text{brine}}$; note that formation water is typically saline, e.g. Mc Cain, 1990) slightly decreases with depth, Fig. 1 (assuming a constant salinity versus depth; Reveillere, 2013). Consequently, $\Delta \rho$ ($\rho_{\text{brine}} - \rho_{\text{H}_2}$) is only slightly affected by depth and only slightly decreases with depth.

2.2. The H$_2$-water interfacial tension $\gamma$

The H$_2$-water interfacial tension $\gamma$ as a function of depth has not been evaluated previously. However, it has been demonstrated by laboratory experiments, that $\gamma$ decreases very slightly with increasing pressure, but decreases strongly with increasing temperature (Chow et al., 2018). The $\gamma$-versus-depth profile has thus been inferred from these datasets (Chow et al., 2018), Fig. 1. Effectively, $\gamma$ decreases linearly with depth, Fig. 1. For example, at a depth of 500 m, $\gamma$ is 69.5 mN/m, and $\gamma$ decreases slightly to 68.3 mN/m at 1000 m depth. However, $\gamma$ always clearly remains relatively high and positive, and H$_2$ and brine are thus always immiscible at reservoir conditions (except the small amount of H$_2$ which chemically dissolves in the brine and the small amount of water which evaporates into the H$_2$ gas).

2.3. The H$_2$-water-rock contact angle $\theta$

Importantly, $\theta$ increases relatively strongly with depth (mostly due to the increasing pressure, compare Iglauer et al. (2021)–note that here a rock surface which was exposed to 10$^{-2}$ M stearic acid is assumed—such organic content is most realistic in the subsurface due to the prevailing reducing atmosphere, e.g. Stalker et al., 2013; Ali et al., 2019).

Fig. 1. Parameters required for structural trapping capacity assessment; (a) H$_2$ density, (b) brine density, (c) H$_2$-brine density difference $\Delta \rho$, (d) H$_2$-brine interfacial tension $\gamma$, (e) H$_2$-brine-caprock contact angle $\theta$, (f) cos($\theta$) – all parameters are plotted against depth to illustrate the inherent dependencies.
Essentially a linear increase in $\theta$ with depth is predicted, compare Fig. 1 and Table 1 – precisely, $\theta$ increases from 44° at 0 m depth (= the surface) to 92° at 4000 m depth. This is a drastic increase, which sets a limit on $H_2$ storage capacities, see discussion below. This increase is caused by the increased $H_2$ density, which results in stronger $H_2$-rock intermolecular interactions, which again increase the affinity of the rock towards the $H_2$, e.g. Al-Yaseri et al. (2016, 2021). $\theta = 90^\circ$ (neutral-wettability) is passed at 3700 m depth. Note that $\theta_{\text{H}_2}$ is generally significantly smaller than the equivalent contact angle for $CO_2$ ($\theta_{\text{CO}_2}$), due to the lower $H_2$ density and the resulting lower $H_2$-rock intermolecular interactions (Iglauer, 2017; Iglauer et al., 2021; Ali et al., 2021; Pan et al., 2021). Consequently, $\cos \theta$ decreases linearly with storage depth, and importantly, $\cos \theta$ intersects the x-axis also at 3700 m, Fig. 1.

### 2.4. $H_2$ column height $h$

$H_2$ column height $h$ decreases monotonically with increasing depth, Fig. 2 (assuming a typical pore radius $r$ of 50 nm, Nelson (2009); and $g$ is 9.81 m/s$^2$). For instance, at 300 m storage depth, the $H_2$ column height is 183 m, which is reduced to 171 m at 500 m depth and 140 m at 1000 m depth. Importantly, $h$ reaches a zero value at 3700 m depth, and storage below this threshold depth would result in $H_2$ percolating through the caprock as $H_2$ turned into the wetting phase at this point ($\theta > 90^\circ$; $\cos \theta < 0$; see $\theta$ and $\cos \theta$ discussion above).

Compare this with equivalent storage heights of $CO_2$ (in the context of $CO_2$ geo-sequestration); $H_{\text{CO}_2}$ is significantly lower (due to the higher wettability of $CO_2$ towards the rock surface; compare also Iglauer et al., 2021) and consequently the threshold depth for $CO_2$ is much more shallow (2400 m); Iglauer (2018).

### 2.5. The mass of $H_2$ which can be stored by structural trapping

Importantly, and indeed analogue to $CO_2$ storage, more relevant than $h$ is the actual mass of $H_2$ ($m_{\text{H}_2}$) which can be stored (Firoozabadi and Cheng, 2010). $m_{\text{H}_2}$ can be predicted via equation (2),

$$m_{\text{H}_2} = \rho_{\text{H}_2} hA$$  \hspace{1cm} (2)

$A$ is the averaged lateral area swept by $H_2$, and $\phi$ is the porosity of the storage rock (not the caprock) – here $\phi = 0.2$ is assumed (which is a typical value for sandstone). A cannot be predicted easily as it also depends on reservoir geology, and a full-scale reservoir simulation is required to determine this parameter (e.g. see Lubon and Tarkowski, 2020; Al-Khdheewawi et al., 2017). $A$ is assumed here to be $A = 100 \times 100 = 10^4$ m$^2$ – although this may vary drastically based on the geology of the specific reservoir. Using these parameters and the parameters discussed above, the $m_{\text{H}_2}$ versus depth profile can be predicted, Fig. 2.

$m_{\text{H}_2}$ initially rapidly increases with depth, and reaches a maximum at 1100 m depth, Fig. 2. At higher depth $m_{\text{H}_2}$ decreases again, and reaches a zero value at 3700 m (as the $H_2$ column height is zero at this depth, see discussion above). At 300 m storage depth $m_{\text{H}_2}$ is 0.895 Mt, which increases to 1.33 Mt at 500 m and eventually 2.01 Mt at 1100 m.

This profile is somewhat similar to the $m_{\text{CO}_2}$ versus depth profile (for CGS, Iglauer, 2018), which also goes through a maximum, but at a slightly higher depth (1300 m). However, in absolute terms, $m_{\text{CO}_2}$ is generally much higher than $m_{\text{H}_2}$ ($m_{\text{CO}_2}$ is in the order of 100 Mt; while $m_{\text{H}_2}$ is only in the order of 1 Mt). Therefore, much less $H_2$ can be stored via this route when compared with $CO_2$. However, a storage capacity of 1 Mt $H_2$ is a high number generally, when compared to other current storage options (Zhang et al., 2016).

### 2.6. Correlations

Empirical correlations were calculated for each parameter ($\theta$, $\gamma$, $\rho$, $h$, $m_{\text{H}_2}$), these are listed in Table 1. Using these correlations initial UHS storage capacities can be predicted.
3. Conclusions and implications

H2 geo-storage is currently explored as a feasible and economic solution to enable large-scale, widespread storage of large quantities of H2. The main proposed storage mechanism for the buoyant H2 in the subsurface is structural trapping, where a caprock provides a geologic seal through which H2 cannot flow due to the high capillary entry pressure (of H2) into the caprock. However, it is clear that H2 can migrate upwards if buoyancy forces exceed the capillary forces—which again depend on the quantity (precisely the vertical H2 column height h) of H2 stored. This aspect is analysed here in more detail, and it is demonstrated that h is not a constant but declines monotonically with depth and reaches a zero value at 3700 m depth. Long term H2 storage below this threshold depth is thus not advised as H2 would percolate upwards through the caprock layer, due to wettability reversal at this depth. Importantly, the mass of H2 which can be stored (mH2) — which is the ultimate property of interest—goes through a maximum at 1100 m depth. There therefore exists a storage depth at which a maximum of H2 can be stored. This work thus provides a fundamental assessment of H2 structural storage efficiency, and aids in the implementation of a large-scale H2 economy.

Declaration of competing interest

The author declares that he has no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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<tbody>
<tr>
<td>p</td>
<td>pressure [Pa]</td>
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<tr>
<td>p0</td>
<td>buoyancy pressure [Pa]</td>
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<tr>
<td>pc</td>
<td>capillary pressure = pressure between wetting and non-wetting phase [Pa], i.e. pressure between H2 and brine phase</td>
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<tr>
<td>ρ</td>
<td>density [kg/m^3]</td>
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<tr>
<td>ρwater</td>
<td>density of water [kg/m^3]</td>
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<tr>
<td>ρH2</td>
<td>H2 density [kg/m^3]</td>
</tr>
<tr>
<td>Δρ</td>
<td>density difference between water and H2 [kg/m^3]</td>
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<tr>
<td>h</td>
<td>H2 plume height permanently immobilized by structural trapping [m]</td>
</tr>
<tr>
<td>d</td>
<td>depth [m]</td>
</tr>
<tr>
<td>θ</td>
<td>water contact angle [°]</td>
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<tr>
<td>γ</td>
<td>interfacial tension [N/m]</td>
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<td>r</td>
<td>pore radius [m]</td>
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<td>temperature [°C]</td>
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<td>dihydrogen</td>
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<tr>
<td>A</td>
<td>surface area of 3D H2 plume projected onto earth’s surface [m^2]</td>
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<tr>
<td>mCO2</td>
<td>mass of CO2 stored by structural trapping [kg]</td>
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<tr>
<td>R²</td>
<td>Pearson’s coefficient</td>
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References


