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[10.1029/2021GL092976](https://doi.org/10.1029/2021GL092976)

Iglauer, S., Abid, H., Al-Yaseri, A., & Keshavarz, A. (2021). Hydrogen adsorption on sub-bituminous coal: Implications for hydrogen geo-storage. *Geophysical Research Letters*, 48(10), article e2021GL092976. <https://doi.org/10.1029/2021GL092976>

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# Geophysical Research Letters



## RESEARCH LETTER

10.1029/2021GL092976

### Key Points:

- Sub-bituminous coal adsorbs a significant amount of hydrogen gas
- Hydrogen adsorption in coal increases significantly with pressure
- Hydrogen adsorption decreases with temperature only very slightly

### Supporting Information:

Supporting Information may be found in the online version of this article.

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### Citation:

Iglauer, S., Abid, H., Al-Yaseri, A., & Keshavarz, A. (2021). Hydrogen adsorption on sub-bituminous coal: Implications for hydrogen geo-storage. *Geophysical Research Letters*, 48, e2021GL092976. <https://doi.org/10.1029/2021GL092976>

Received 15 FEB 2021

Accepted 25 MAR 2021

## Hydrogen Adsorption on Sub-Bituminous Coal: Implications for Hydrogen Geo-Storage

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**Abstract** Hydrogen is a clean fuel that can potentially revolutionize the energy supply chain and decarbonize fuel consumption. However, a key hurdle that needs to be overcome before a full-scale hydrogen economy can be established is hydrogen storage which is currently the main limitation. Here, we propose that hydrogen gas can be stored in underground coal seams, where it adsorbs on the coal surface. However, currently, no hard data related to such a procedure exists. We, therefore, demonstrate experimentally that significant amounts of hydrogen gas can be stored via this route. Hydrogen adsorption capacities reached 0.6 moles H<sub>2</sub>/kg of coal at 14.3 MPa, and adsorption capacity initially increased strongly with pressure (up to ~4 MPa) and then plateaued out, while temperature only had a very minor influence. This study provides fundamental data for hydrogen storage in coal seams, and thus aids in the industrial implementation of a hydrogen supply chain.

**Plain Language Summary** Hydrogen is a clean fuel which can potentially revolutionize the energy supply chain and decarbonise fuel consumption. However, a key hurdle which needs to be overcome before a full-scale hydrogen economy can be established is hydrogen storage which is currently the main limitation. Here, we propose that hydrogen gas can be stored in underground coal seams, where it adsorbs on the coal surface. We demonstrate experimentally that significant amounts of hydrogen gas can be stored via this route. This work provides fundamental data for hydrogen storage in coal seams, and thus aids in the industrial implementation of a hydrogen supply chain.

## 1. Introduction

Hydrogen is a clean fuel that can potentially revolutionize the energy supply chain and decarbonize fuel consumption (Hanley et al., 2018; Tarkowski, 2019). However, hydrogen storage is currently the key hurdle that prevents the establishment of an industrial-scale hydrogen economy (Berta et al., 2018; Flesch et al., 2018; Heinemann et al., 2018; Iglauer et al., 2021; Lord et al., 2014; Shi et al., 2020; Tarkowski & Czapowski, 2018; Yekta et al., 2018; Zhang et al., 2016). It is, thus, necessary to develop better hydrogen-storage solutions (Zhang et al., 2016). One new concept is the storage of hydrogen gas in underground coal seams, where hydrogen adsorbs on the coal surface and can be withdrawn again at any time when required. This concept is analog to methane storage in such geologic underground coal seams. Methane is adsorbed on the coal surface over geologic times, and it is now a resource produced via coal bed methane (CBM) production (e.g., Moore, 2012; Seidle, 2011). Connected with that is also enhanced CBM production, where CO<sub>2</sub> is injected into a deep coal seam; the CO<sub>2</sub> displaces more methane while it is itself adsorbed on the coal surface (Chaturvedi & Sharma, 2020; Keshavarz et al., 2017).

However, while CH<sub>4</sub> and CO<sub>2</sub> adsorption has been extensively studied to optimize CBM, no data are available for hydrogen adsorption on coal. We, therefore, measured H<sub>2</sub> adsorption on a typical sub-bituminous coal sample, and demonstrate that substantial amounts of H<sub>2</sub> can be stored via this approach, even at moderate pressures. This study will thus aid in the larger-scale implementation of a hydrogen economy.

## 2. Experimental Procedure

Sub-bituminous coal (Pan Upper, supplied by Premier Coal located in Collie, Western Australia; maximum vitrinite reflectance: 0.38%) was selected and thoroughly analyzed for its essential properties (Table 1). In addition, the coal was characterized via Thermal Gravimetric Analysis (TGA) and the coal surface was characterized via Fourier-Transformed Infrared Spectroscopy (FTIR) and energy dispersive spectroscopy, the

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**Table 1**  
*Essential Analysis Properties of Pan Upper Raw Coal*

Proximate analysis (wt %)			
Inherent moisture	Ash	Volatile matter	Fixed carbon
21.7	2.5	28.0	47.8
Ultimate analysis (wt%)			
Carbon	Hydrogen	Nitrogen	Relative density (%)
58.1	2.98	1.15	1.39
Maceral composition (volumetric, %)			
Vitrinite	Liptinite (Exinite)	Inertinite	Mineral matter
33.3	11.3	43.9	11.5

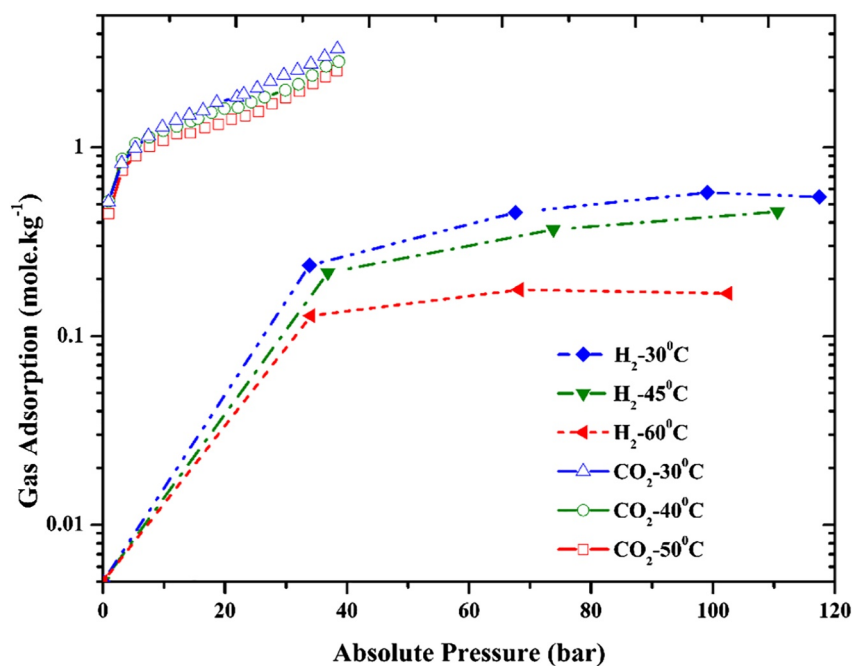
detailed results can be found in the Supporting Information. The coal was then crushed with a blade grinder and sieved to a homogeneous particle size ( $<250 \mu\text{m}$ ). Low-pressure  $\text{N}_2$  (99.999 vol%) BET adsorption and desorption curves and the zeta potential of the coal powder were then measured to further characterize the coal surface. Note that the zeta potential was measured with a Zetasizer Nano ZS in deionized water at pH = 8.7, 0.1 MPa, and 296 K. For more details regarding the TGA, FTIR, and  $\text{N}_2$  BET measurements, please see the Supporting Information.

Subsequently, the adsorption profiles of  $\text{CO}_2$  (purity = 99.99 mol%) and  $\text{H}_2$  (purity = 99.995 mol%) were measured at elevated temperatures and a wide pressure range (0.1–14.3 MPa) using a PCTpro adsorption analyzer (Setaram Instrumentation). The average standard deviation of the high-pressure adsorption experiments was estimated as  $\pm 7.3\%$  ( $\text{H}_2$ ) and  $8.0\%$  ( $\text{CO}_2$ ), based on replicate measurements.

### 3. Results and Discussion

Clearly,  $\text{H}_2$  adsorption increased dramatically with increasing pressure, Figure 1. Initially, this increase was very steep, but the adsorption curve plateaued out at  $\sim 4$  MPa. Thus, at low pressure (0.1 MPa), only small amounts of  $\text{H}_2$  could be adsorbed (0.005 moles/kg coal), while at 3.38 MPa, 0.23709 moles  $\text{H}_2$ /kg coal were adsorbed (both measured at 303 K), a 47,418-fold increase. The  $\text{H}_2$  amount adsorbed increased further to 0.54686 moles  $\text{H}_2$ /kg coal at 11.74 MPa (at 303 K) and 0.60183 moles  $\text{H}_2$ /kg coal at 14.3207 MPa (at 318 K), a 120,000-fold increase. Such a type I adsorption curve indicates the presence of micropores (which is expected in coal; Okolo et al., 2015) and that  $\text{H}_2$  was adsorbed as a mono-layer (Singh et al., 1985). This is in contrast to the low-pressure  $\text{N}_2$ -BET adsorption profiles which indicated a type II adsorption system (compare the Supporting Information), thus  $\text{N}_2$  was adsorbed in multiple molecular layers on the coal surface at low pressure. Note that the type II  $\text{N}_2$ -isotherm was caused by  $\text{N}_2$  condensing in narrow pore throats, as  $\text{N}_2$  boiling temperature (at atmospheric pressure) is 77 K (and the experiment was performed at 77 K) (Flores, 2014). Furthermore, changes in temperature (from 303 to 333 K) only slightly influenced the adsorption profile (a marginally smaller amount of  $\text{H}_2$  was adsorbed at the higher temperature). We conclude that substantial amounts of  $\text{H}_2$  can be stored in deep coal seams where high pressures and elevated temperatures prevail.

However, approximately 10 times the amount of  $\text{CO}_2$  could be adsorbed (e.g., 2.64 moles of  $\text{CO}_2$ /kg coal at 3.302 MPa vs. 0.23709 moles of  $\text{H}_2$ /kg coal at 3.382 MPa, both measured at 303 K). This is mainly related to the very low zeta potential of the coal ( $-70$  mV measured at pH 8.7, 296 K, and 0.1 MPa in deionized water). Such a high negative surface charge will strongly interact with a molecule that has a high quadrupole moment (the  $\text{CO}_2$  quadrupole moment is  $-4.3 \times 10^{-26}$  esu.cm<sup>2</sup>; Buckingham et al., 1968). Note that  $\text{CO}_2$  can also undergo Lewis acid-base reactions because of this high quadrupole moment, and form hydrogen bonds with alcohol or carbonyl groups (Raveendran et al., 2005; Fujii et al., 2002). As coal is a very complex organic material (Hatcher & Clifford, 1997), the coal tested also contained alcohol and carbonyl groups (compare the infrared spectra in the Supporting Information); these groups further strengthened the affinity (and thus adsorption) between  $\text{CO}_2$  and coal. This is in contrast to  $\text{H}_2$  which also has no external electric dipole moment and a quadrupole moment which is one magnitude smaller ( $+0.651 \times 10^{-26}$  esu.cm<sup>2</sup>; Buckingham



**Figure 1.** Hydrogen adsorption on sub-bituminous coal as a function of pressure and temperature. Data for CO<sub>2</sub> have been added for comparison.

et al., 1968). H<sub>2</sub> adsorption onto the coal surface is therefore driven by dispersive (non-polar–non-polar interactions, that is, induced dipole moment – induced dipole moment interactions) Van der Waals forces only, which in total are substantially weaker than the total interaction forces between CO<sub>2</sub> and the coal surface. This weaker interaction between the H<sub>2</sub> and the coal surface results in reduced H<sub>2</sub> adsorption capacity.

#### 4. Conclusions

Hydrogen storage is currently a key barrier to the implementation of an industrial-scale hydrogen economy. We, therefore, examined here the potential of hydrogen storage in underground coal seams, where H<sub>2</sub> could be stored as an adsorbed phase on the coal surface. Indeed, this is possible from a fundamental technical perspective as we demonstrated via laboratory adsorption experiments. Substantial amounts of H<sub>2</sub> (0.23709 moles H<sub>2</sub>/kg coal) could be stored at moderate pressure and temperature (303 K and 3.3 MPa, note that these thermo-physical conditions prevail at a depth of approximately 300 m; Dake, 1978); this adsorption capacity could be further enhanced at higher pressures (14.3 MPa) to 0.60183 moles H<sub>2</sub>/kg coal (at 318 K). We conclude that hydrogen storage in deep coal seams is potentially possible, this study, therefore, supports the large-scale set-up of a hydrogen supply chain.

#### Conflict of Interests

The authors declare that there are no conflict of interests.

#### Data Availability Statement

Adsorption data for H<sub>2</sub> and CO<sub>2</sub> as functions of pressure and temperature are available at <https://ro.ecu.edu.au/datasets/56/>.

### Acknowledgments

The authors would like to acknowledge Edith Cowan University for providing the required infrastructure for this work.

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