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#298 Paper Review

SUMMARY ABSTRACT REVIEW PAPER REVIEW HISTORY

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Authors	Boby Royan Alfadlil, Gregory P Knowles, Muhammad P Parsa, Dirgarini P Subagyono, Alan P Chaffee, Daniel Daniel
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Carbon monolith from Victorian brown coal for hydrogen storage

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Abstract. The application of carbon monolith from VBC (Victorian Brown Coal) for H_2 storage has been studied. The storage capacities of the monolith were measured using an isothermal adsorption process at different temperatures and pressures. In this study the adsorption capacities of monolithic carbon and those of activated carbon from other literatures were compared. It was found that temperature and pressure affected the hydrogen adsorption capacity. It was also observed that density of the carbon monoliths also have significant effects on hydrogen storage capacity.

Keywords: carbon monolith, victorian brown coal, adsorption, hydrogen storage.

1. Introduction

The emission of carbon dioxide to the atmosphere increases the earth's greenhouse effect, leading to an increase of the global temperature. A possible solution is its capture and sequestration, extensive research has been carried out on alternative energy sources such as sun, geothermal, tides, windmills, biomass, nuclear energy, etc [1]. with the aim of energy applications (e.g. storage of H_2 and CH_4 gas for transportation technology) or environmental protection goals (e.g. the absorption of CO_2 gas to reduce the negative effects of the greenhouse effect) [2].

The storage of these gases is usually performed by compression in high pressure vessels. However, the storage capacities of such devices are restricted, because of the low density of these gases and pressure limitations due to safety reasons [3]. An alternative technology for gas storage is adsorption on porous materials [4]. Thereby, the density of the gas accumulated in the porous structure of these materials is increased, due to the attractive forces between its molecules and the material.

Given the massive scale of Victoria's brown-coal resource, there is potential for material economic benefits if it is developed and it is now recognized that with suitable up-grading, primarily drying, it has the potential to become the basis of the supply of energy in a variety of forms [5].

The interest in using monolithic structures for chemical conversion and adsorption processes is increasing. A relatively new type of monolith is based on carbon. The combined favorable properties of carbon and monolithic structures create a support with great potential in catalytic and adsorption processes [6]. Monolith is basically a uniform block, consisting of parallel channels that can be prepared via extrusion of an activated carbon into various shapes and sizes such as circular, square, triangular or hexagonal and many more. This study reports the application of carbon monolith from Victorian brown coal as a H_2 storage. The H_2 storage capacity of the materials were compared with that of several carbon materials.

2. Experimental section

The volumetric technique consists of introducing [dosing] a known amount of gas [adsorptive] into the chamber containing the sample to be analyzed. When the sample reaches equilibrium with the adsorbate gas, the final equilibrium pressure is recorded. These data are then used to calculate the quantity of gas adsorbed by the sample.

This process is repeated at given pressure intervals until the maximum preselected pressure is reached. Then the pressure can be decreased to provide a desorption isotherm. Each of the resulting equilibrium points [volume adsorbed and equilibrium pressure] is plotted to provide an isotherm.

The adsorption capacity of H_2 in carbon monolith samples was measured at 273, 293 and 313 K. In the isothermal adsorption process the temperature was kept constant by Cooloant was recirculated. thermal bath / dewar for optimum adsorption process. HPVA system. Each isotherm experiment took approximately 24 hours to complete.

2.1. Carbon Monolith preparation Materials

Commented [GK1]: And General Materials Physico-Chemical Characterisation?

The material used in this study was obtained from Monash University Chaffe Group Research which was prepared using the extrusion method of monolithic carbon. The carbon monolith was activated by CO_2 at 850 ºC for 1 hour. Carbon dioxide gas serves as the activating gas where the gas will react with carbon so as to open the pores which will increase surface area. Aside from that, carbon dioxide gas also acts to prevent the entry of oxygen gas into the activator. with chemical activation using NaOH. Before monolithic isothermal carbon adsorption testing, the degassing process needs to be done to remove all impurities and other volatile matter material to get out of the pore. the stage of the degassing process is carbon monolith which has been roughly crushed heated at a temperature of 105 degrees Celsius for approximately 6 hours under vacuum (0.01 mTorr).

2.2. Physico-Chemical Characterisation

Density, porosity, structure and surface morphology were all evaluated in the present study. The true density measurement was conducted using AccuPyc II 1340. The porosity parameters of the carbon monolith (i.e., Brunauer-Emmett-Teller (BET) surface area, micropore surface area, total pore volume and average pore diameter) were determined from the nitrogen adsorption data at 196 ºC using an accelerated surface area and porosimeter system (ASAP 2010, Micromeritics). The ultimate analysis were indentified by TruSpec Micro for delivers optimal performance in C, H and N determination in micro samples (1 to 10 mg) supported by Geoservices Coal Laboratory. The morphology structure of the carbon monolith were elucidated on a SEM SU3500 from Bandung Institute of Technology.

2.3. Apparatus

The HPVA II Series of adsorption analyzers from Particulate Systems uses the static volumetric method to obtain high-pressure adsorption and desorption isotherms, which is shown schematically in Fig. 1. The temperature was controlled by Colora Messatechnik GMBH refrigerated/heating circulator and the An Edwards RV3F vacuum pump was provided by Edward Corporation (UK).

This was via separate Micromeritics VacPrep 062 degasser for removing adsorbed contaminants. In addition to flowing gas, this sample preparation unit provides vacuum to prepare samples by heating and evacuation. The VacPrep offers the user a choice of vacuum or gas flow on each of the six degassing stations. Needle valves allow the user to introduce the flowing gas or vacuum slowly to prevent fluidization of samples.

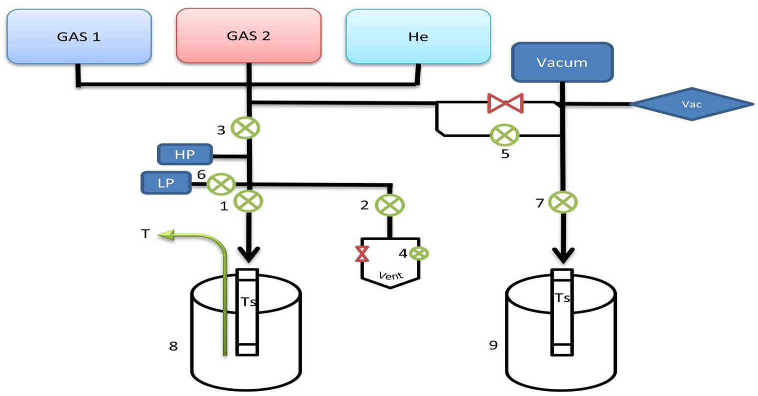

Figure 1. HPVA II-200. HP, high-pressure transducer; LP, 1000 torr pressure transducer; T, temperature probe; Ts, Tube sample; (1) analysis port valve; (2) vent valve; (3) manifold valve; (4) full vent valve; (5) full vacuum valve; (6) 1000 torr isolation valve; (7) degas port valve; (8) Analysis station bath; (9) Degas station bath.

In the HPVA II measurement procedure, there exists an exhaust process during each desorption step to control the pressure. The gas composition in the adsorption cell after achieving equilibrium was measured during each desorption step, which could be used to calculate the adsorbed amount at each desorption step to improve the accuracy of calculations. The total adsorbed amount at one pressure step (Δn_{ads}) can be calculated using:

$$\Delta n_{ads} = \Delta n_{dosed} - \Delta n_{Nads}$$

where Δn_{dosed} is the amount dosed from the manifold at that pressure step and Δn_{Nads} is the non-adsorbed amount at that pressure step. Likewise, the total adsorbed amount at the n^{th} pressure step (Δn_{Nads}) can be calculated using:

$$\Delta n_{adsn} = \Delta n_{dosedn} - \Delta n_{Nadsn} = n_{An} - n_{Bn} - n_{Aadsn} + n_{Nadsn}$$

where n_{An} is the number of moles of gas in the manifold before dosing at the n^{th} pressure step, n_{Bn} is the number of moles of gas in the manifold after dosing at the n^{th} pressure step and $n_{Nadsn-1}$ is the number of moles of gas not adsorbed by the sample at the n^{1s} pressure step.

3. Results and discussion

3.1. Material characterisation

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The physical and chemical characteristic of carbon monolith from Victorian Brown Coal are presented in Table 1. Where in this study carbon monolith was used with a surface area of 973 m^2/g consisting of 82.5% carbon, 2.7% hydrogen and 0.5% nitrogen. The conductivity value that is owned is 160.5 $\Omega^{-1}cm^{-1}$. The value of conductivity is influenced by the amount of carbon in the honeycomb structure, with density 1.82 g/cm^3. SEM analysis on one image for one sample given in Figure 2. Showed that the monolithic carbon possessed macropore structures which are interconnected, other than the macroporous channels the materials is understood to be microporous. Microporous adsorbents show the highest adsorption activity towards various gases, including hydrogen this is due to the mechanism of volume filling of pores inherent in these materials. The key parameter determining the efficiency of an adsorption storage system is the amount of the target substance (hydrogen gas) stored in the system under particular thermodynamic conditions determined by the pressure (P) and temperature (T).

Table 1. Characteristic of Carbon Monolith VBC

Parameter	Value
Surface area (m^2/g)	973
True density (g/cm^3)	1.82
Conductivity ($\Omega^{-1}cm^{-1}$)	160.5
Carbon (%)	82.5
Hydrogen (%)	2.7
Nitrogen (%)	0.5

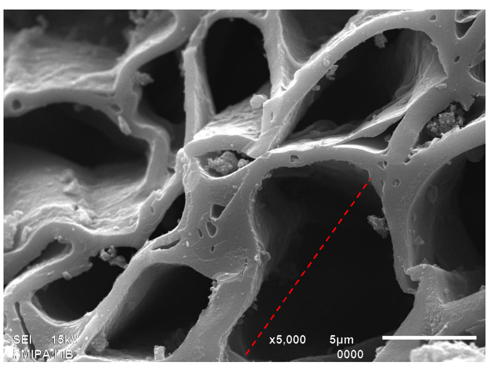

Figure 2. SEM image characterization of carbon monolith with 5000 magnification (Red line is Macropore channel).

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3.2. Adsorption study

The adsorption capacity of hydrogen with carbon monolith from Victorian Brown Coal was measured at three different temperatures (Figure 3). The best adsorption capacity was obtained at 273 K (1.646 mmol/g) because the temperature was the closest to the critical temperature of hydrogen (33 K). In other words, the higher the temperature used, the lower amount of hydrogen gas that can be stored in the carbon monolith. The increase of adsorption temperature to 293 K decreased adsorption capacity to 1,282 mmol / g and this value continued to decline at higher temperature (313 K) with a storage capacity of only 0.922 mmol /g.

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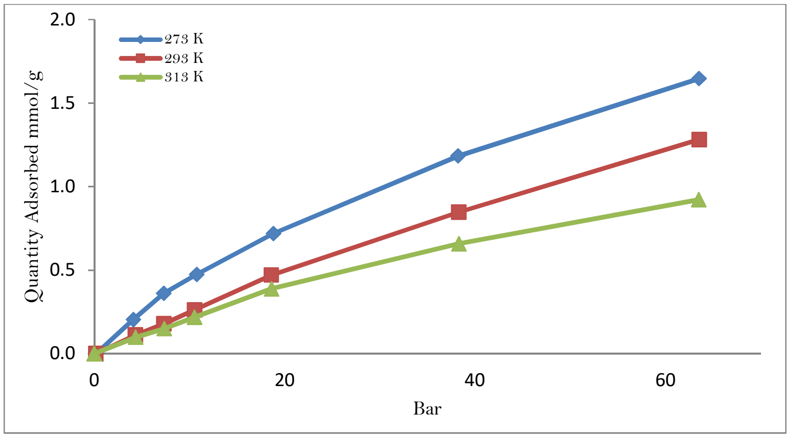

Figure 3. H_2 adsorption isotherm with carbon monolith from Victorian brown coal at different temperatures.

Comparison of hydrogen gas storage capacity at 293 - 298 K of several carbon materials was carried out to determine the monolithic carbon quality used in this study. This temperature range was chosen as the temperature which was most widely used in hydrogen storage applications using volumetric methods. For this purpose, the hydrogen storage capacities of some carbon materials made from various sources and different activation methods (some of which were in the form of carbon monolith, powder and granular) were presented in Table 2.

Table 2 and Figure 4 clearly showed that VBC carbon monolith material is superior to hydrogen storage compared to other carbon materials. This maybe due to the higher material density than that of other materials (surface area 900 – 1500 m^2/g) with similar shapes (monolith form). The result clearly indicate that high surface area is not the only determining factor for materials to possess high adsorption capacity, but material density is also one of the factors that can increase adsorption capacity.

Table 2. Hydrogen storage capacity of carbon materials.

Sample	Surface area (m^2g^{-1})	Density (g cm^{-3})	H_2 Storage on Volumetric basis (mmol/g)	Refference
M-A1	928	1.00	1.9344	[1][4][9]
M-A336	1367	0.87	2.0833	
M3M	2610	0.42	1.1746	
MOF-210	6240	0.25	0.4960	
D10 PACK	2259	0.62	3.7301	
D10 TAP	2259	0.36	2.9761	
D7 PACK	2364	0.63	3.2241	

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Note the near order of magnitude difference, are the calculations correct?

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Commented [GK15]: This is SA (VBC-HM with much less than other materials in table), not density.

Commented [GK16]: Monoliths, powders?

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D7 TAP	2364	0.34	2.8769	
M-ACF15	1127	0.69	2.8521	
ACF15 PACK	1193	0.94	2.8422	
ACF15 TAP	1193	0.24	2.3690	
M-ACF20	2068	0.54	2.9265	
M-ACF25	1838	0.54	2.5793	
CM-VBC	973	1.82	1.2823	This study

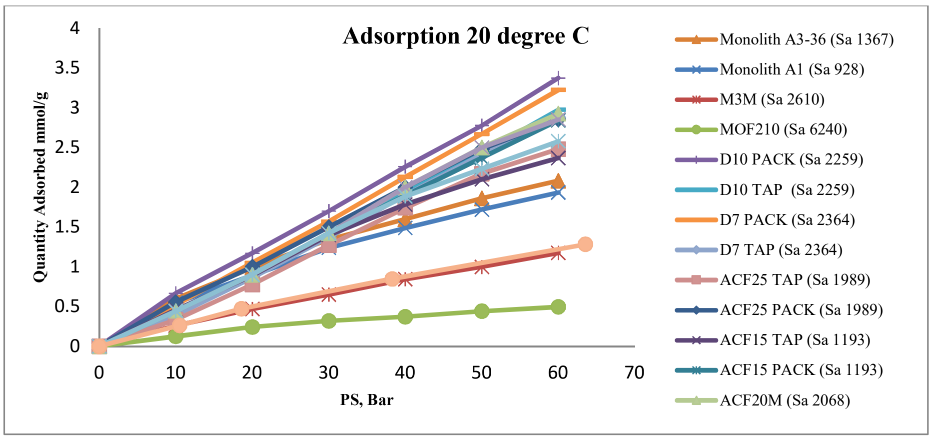

Figure 4. Comparison of H_2 adsorption isotherms of several carbon materials at 298 K.

4. Conclusion

Carbon Monolith from Victorian Brown Coal is potential as material for hydrogen storage. The maximum adsorption capacity of hydrogen gas was 1,646 mmol/g which was obtained at 273 K. It was found that material density was one key factor for hydrogen storage capacity.

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Acknowledgements

We would like to acknowledge valuable contribution of …..

Commented [GK20]: Were Monash MCEM used for SEM then there is a standard stamen required for inclusion. They would also provide accurate statement as to how the image was collected (method) as is missing.

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