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LETTER

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Methane storage in a commercial Activated Carbon

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A commercial activated carbon was examined for possible methane storage application. The structural and surface properties of the carbon were characterized by Nitrogen adsorption isotherm at 77 °K. It was found that the carbon is largely microporous with a surface area of approximately 860 m^2/g. Adsorption test shows the carbon is able to achieve a methane storage capacity of approximately 70/cc.

Natural gas (NG) is attracting more and more attentions as a fuel because of its clean nature and abundance. Methane is the main component of NG, which presents a superior octane number (~ 120) in conventional internal combustion engines. However, methane is a gas at room temperature, which exerts constraints on its applications. The automobile industry urgently requires a safe and efficient technology for NG storage. On the other hand, methane is also an important greenhouse gases, a good methane adsorption/storage technology can help to reduce the methane emission (for example, for bio-gas) to the environment.

Activated carbons (ACs) are excellent adsorbents for NG storage in the form of adsorbed natural gas (ANG). Many researches have been conducted to develop ACs for NG storage [1 - 4]. The deliverable storage capacities achieved are approximately 70 - 140 v/v (at 25 ºC and 3 - 4 MPa), which are getting close to the practical standard (approximately 150 v/v at ambient temperature and a pressure of 3.5MPa) [5]. For the optimal storage of NG, an AC should possess large surface area and uniform micropores with the size (d) of approximately 0.8 - 1 nm [6-8]. Large pores (d > 2 nm for mesopores and d > 50 nm for macropores by IUPAC), although not efficient for the storage capacity, are necessary for the charging and discharge kinetics.

Economical production of carbon sorbents for NG storage is another important issue [9] which is greatly in favor of commercial ACs. This study will investigate the feasibility of NG storage in a commercial Norit® AC, which was produced via steam activation.

The commercial Norit AC was provided by Esco Singapore Pte. Ltd. in the form of cylindrical extruders with the diameter of approximately 5 mm. The adsorption rig uses the volumetric technology and is capable of attaining the sorption pressure up to 50 atm. About 0.5 grams of AC sample was placed in the sample cell and subsequently degassed under high vacuum (with a molecular turbo-pump) and under the temperature of 250 ºC for overnight. Methane gas (purity > 99 %) was supplied by SOXIAL Singapore. During the sorption measurements, the cell was submerged in a water bath of controlled temperature.

The structure of the AC sample was analyzed with the pore and surface analyzer (Quantachrome, Autosorb-1c). Nitrogen sorption isotherms were measured on the sample at 77 °K. Prior to the measurements, the sample was degassed at 250 ºC overnight under high vacuum.

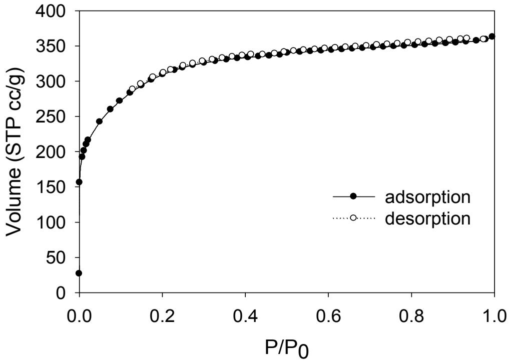

Figure (1) : N_2 isotherm on Norit AC at 77 °K.

Figure (1) shows N_2 isotherms on the AC sample at 77 °K. It can be seen that the AC is primarily microporous with a very small hysterisis in the desorption isotherm. The BET (Brunauer-Emmett-Teller) surface area of this carbon is found to be 860 m^2/g, which suggests that the AC is a reasonably good sorbent for NG storage.

Table (1) : Surface parameters of Norit AC (V_t = total volume, V_{mic} = micropore volume).

BET area (m^2/g)	V_t (cc/g)	V_{mic} (cc/g)	V_{me} (cc/g)
860	0.55	0.48	0.07

Figure (2) shows the pore size distribution (PSD) of the AC derived from N_2 isotherm using the non-local density functional theory (NLDFT). It is seen that the Norit AC presents a narrow micropore peak around 11 Å with little mesoporosity. This PSD further suggests that the AC is a good candidate for NG storage, as the pore size is close to the optimal pore size of 8 – 10 Å for NG storage. The surface properties of the AC sample are listed in Table (1).

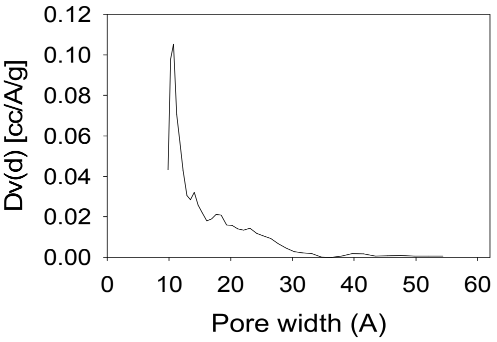

Figure (2) : The PSD of the Norit AC derived from N_2 isotherm at 77 ^oK using NLDFT.

Methane adsorption isotherms were measured on the AC sample at 273 ^oK and 303 ^oK, respectively with the pressure up to 50 bar. Figure (3) presents the sorption isotherms of methane as symbols. A Toth isotherm was employed to fit the isotherm data, which take the form of :

$$C_{\mu} = C_{\mu s} \frac{b \times P}{\left[1 + (b \times P)^t\right]^{1/t}} \qquad (1)$$

where b is the sorption affinity and t is the heterogeneity parameter of the adsorbent. The fitting results are shown in Figure (3) as lines while the optimally obtained fitting parameters were listed in Table (2). It is seen that the Toth equation fits the isotherm data very well. The heterogeneous parameter t = 0.33 means that the carbon surface is very heterogeneous towards methane molecules, probably due to that the narrow micropores induce strong variation in adsorption potential with the progressive fillings in micropores.

Table (2) : Isotherm parameters of methane on AC sample.

Temperature (^oK)	$C_{\mu s}$ (mmol/g)	b (1/bar)	t
273	20.65	1.58×10^{-2}	0.333
300	11.31	7.78×10^{-3}	0.493

The volumetric storage capacity of methane is then calculated based on the methane isotherm and the bulk density of the AC. The standard ASTM (American Society for Testing and Materials) method was used to determine the density of the AC sample, in which the sample was crushed into fine powder and put in a measuring cylinder for its volume determination. The storage capacity was found to be approximately 70 v/v which compares favorably in terms of cost/performance against other designated AC samples reported in the literature. For example, the capacity of 80 and 86 v/v were achieved by [4] and [6], in which the ACs were produced via the expensive and complicated template synthesis or the combination of physical plus chemical activation processes. Methane storage in other two commercial carbons (Picazine ans Sigma) also reported favorable results of 77 and 71 v/v, respectively [10].

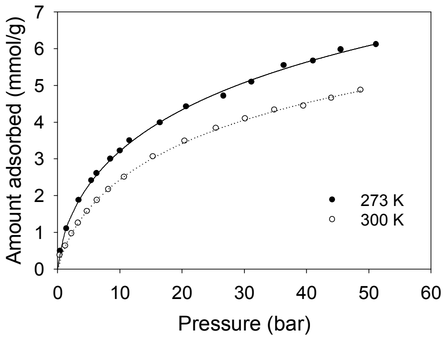

Figure (3) : Methane isotherm on Norit AC.

In conclusion, commercial AC samples may present good performance for methane storage, provided that the sample presents a narrow pore size distribution around 8-11 Å and a surface area ≥ $900 m^2/g$. Such commercial products have the advantage on economical side. What is more, the structure of such a commercial AC may be further tailored to improve the methane storage capacity, for example, via the chemical/physical activations to open more micropores, or via the chemical vapor deposition to reduce the size of large pores [11]. Both techniques are well developed and worthwhile to try in the future.

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