

Adsorption Isotherms of CH₄ on Activated Carbon from Indonesian Low Grade Coal

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ABSTRACT: This article presents an experimental approach for the determination of the adsorption isotherms of methane on activated carbon that is essential for methane storage purposes. The experiments incorporated a constant-volume–variable-pressure (CVVP) apparatus, and two types of activated carbon have been investigated, namely, activated carbon derived from the low rank coal of the East of Kalimantan, Indonesia, and a Carbotech activated carbon. The isotherm results which cover temperatures from (300 to 318) K and pressures up to 3.5 MPa are analyzed using the Langmuir, Tóth, and Dubinin–Astakhov (D-A) isotherm models. The heat of adsorption for the single component methane-activated carbon system, which is concentration- and temperature-dependent, is determined from the measured isotherm data.

■ INTRODUCTION

Gas adsorption phenomena are widely applied in the field of gas separation and gas purification.¹ One significant commercial application of gas adsorption phenomena is the adsorption of methane gas for storage purposes. The conventional method of natural gas supply is through pipelines. If there are populated areas remote from the gas wells, an alternate method has to be investigated. In recent years, adsorbed natural gas (ANG) storage systems have attracted much attention as an alternative to compressed natural gas (CNG) for energy storage and transportation purposes. This is because ANG storage systems are designed to have a higher energy density yet operate at a lower pressure than the CNG method.^{2–10} Effective natural gas adsorption storage technology by activated carbon is essential for the utilization of natural gas.^{11,12} The developments of adsorption-based storage systems require a basic understanding of the isotherms over a wide range of pressure and temperatures for various types of adsorbents. These data are essential for designing the ANG system at a given gas feed, product specifications, and environmental conditions.

This article reports the ongoing effort toward generating experimental adsorption isotherm data for the adsorption of methane gas on activated carbons. The adsorption apparatus is based on a volumetric method, and the experiments were conducted at temperatures ranging from (300 to 318) K and pressures up to 3.5 MPa. Two types of activated carbon, (i) a low rank coal from the East of Kalimantan, Indonesia, which is produced in the laboratory (KT) and (ii) a commercially available activated carbon (Carbotech), were used in the experiments. The constant-volume–variable-pressure (CVVP) method is employed for the investigation of the CH₄ uptake. The experimental data were regressed using the Langmuir, Tóth, and Dubinin–Astakhov (D-A) models, which are suitable for capturing the heterogeneous behavior of activated carbon adsorbents.¹⁴

Table 1. Composition of Raw Coal from the East of Kalimantan, Indonesia

proximate analysis	
total moisture	19.74 %
moisture	13.56 %
ash	4.80 %
volatile matter	40.83 %
fixed carbon	40.81 %
ultimate analysis	
moisture	13.56 %
ash	4.80 %
carbon	56.47 %
hydrogen	3.96 %
nitrogen	0.38 %
total sulfur	2.41 %
oxygen	18.43 %

■ EXPERIMENTS

Materials. Methane with a purity of 99.9995 % was used in the experiment as the adsorbate. Two types of activated carbon have been investigated, namely, Carbotech activated carbon (particles of 0.063 mm) and KT activated carbon. The fresh coal from the East of Kalimantan, Indonesia was crushed and sieved to particles of 0.10 mm before undergoing physical activation. The physical activation of the KT activated carbon was completed in a vertical autoclave activation reactor placed inside a furnace. The production processes include the carbonization/oxidation and activation. The fresh coal was first heat treated by

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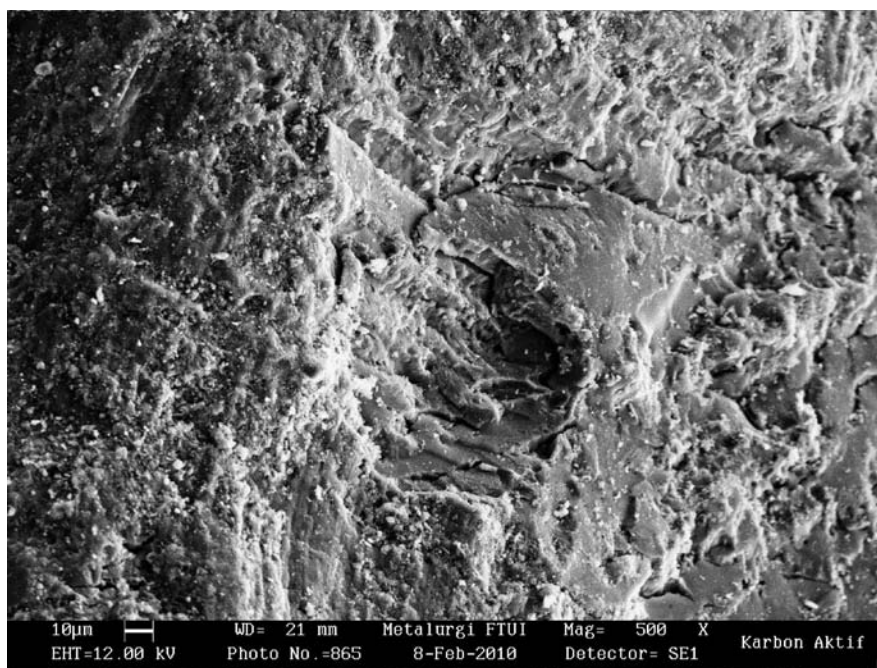


Figure 1. Scanning electron micrographs (SEM) photo of KT at 500× magnification.

Table 2. Thermophysical Properties of Activated Carbon Derived from the East of Kalimantan, Indonesia and Carbotech Activated Carbon Determined Using the Quantachrome Gas Sorption Instrument (Autosorb-1)

physical properties	Carbotech	KT
micropore volume/ $\text{m}^3 \cdot \text{kg}^{-1}$	$0.514 \cdot 10^{-3}$	$0.470 \cdot 10^{-3}$
density/ $\text{kg} \cdot \text{m}^{-3}$		2200
surface area/ $\text{m}^2 \cdot \text{kg}^{-1}$	$0.885 \cdot 10^6$	$0.668 \cdot 10^6$

flowing O_2 at $100 \text{ mL} \cdot \text{min}^{-1}$ at 573 K for 6 h; the composition of the low rank coal is listed in Table 1. Following the carbonization process, the char samples were activated in a stream of CO_2 at $150 \text{ mL} \cdot \text{min}^{-1}$. The samples were activated from room temperature to maximum heat treatment temperatures of 1223 K for 6 h. A scanning electron micrograph (SEM) of the KT activated carbon is shown in Figure 1 with 500× magnification. Table 2 gives the physical properties (BET surface area, pore volume, and skeletal density) for both the KT and the Carbotech activated carbons. These structural characteristics are determined using a Quantachrome gas sorption instrument (Autosorb-1). The surface area was calculated by the Brunauer–Emmett–Teller (BET) method from N_2 adsorption isotherm data, which has been performed at 77.3 K. The pore size distribution (PSD) was obtained by the nonlocal density functional theory (NLDFT) method.

Apparatus and Procedures. Figure 2 shows a schematic diagram of the CVVP test rig. The experimental apparatus consists of a measuring/adsorption cell and a charging cell with internal volumes of (84.02 and 1150.98) mL, respectively, made of stainless steel (SS 304). The measuring cell and charging cells are connected through a capillary tube. The apparatus is immersed in a constant-temperature water bath (Hüber) which is controlled to preselected temperatures with a precision of ± 0.2 K. The pressure was measured using an absolute pressure transmitter (Druck, PTX 1400), with a pressure range of (0 to 40)

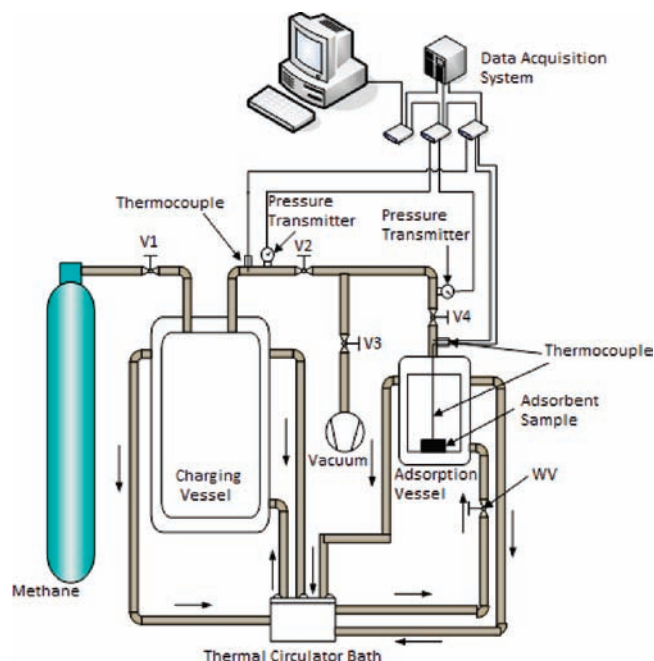


Figure 2. Schematic diagram of the experimental apparatus.

bar and ± 0.15 % full scale uncertainty. Class-A type K thermocouples were used for temperature measurement. A thermocouple was installed in the measuring cell keeping in contact with the activated carbon. This enables a direct temperature measurement of the adsorbent to be obtained. All temperature and pressure readings were monitored with a data logging system using a sampling interval of 10 s.

Prior to the isotherm test, the entire assembly was evacuated for 8 h using an Aruki single stage vacuum pump to a pressure level of 0.01 mbar. During the evacuation, the adsorption cell temperature was maintained at (403 to 413) K to desorb any

Table 3. Experimental Uptake Data for Adsorption of CH₄ on KT Activated Carbon from the East of Kalimantan, Indonesia

<i>P</i>	<i>C</i>	<i>P</i>	<i>C</i>	<i>P</i>	<i>C</i>
kPa	kg·kg ⁻¹	kPa	kg·kg ⁻¹	kPa	kg·kg ⁻¹
T = 300 K		T = 308 K		T = 318 K	
58.81	0.010	61.89	0.009	57.95	0.007
141.37	0.018	139.18	0.016	137.05	0.014
246.66	0.024	250.58	0.022	238.15	0.019
466.09	0.031	500.64	0.031	471.03	0.027
716.40	0.037	733.43	0.036	716.58	0.033
987.29	0.041	977.72	0.039	1005.36	0.037
1444.82	0.045	1481.74	0.044	1478.86	0.042
1962.04	0.048	1985.22	0.047	1990.81	0.044
2445.67	0.049	2453.72	0.048	2471.63	0.045
2951.47	0.050	3012.58	0.049	2964.92	0.047
3495.75	0.051				

residual gas in the cell. Helium gas was injected into the system during desorption to improve the evacuation. After the degassing process, the CVVP apparatus is ready for the adsorption isotherm experiments. Similar descriptions on the experimental procedures are reported elsewhere by Loh et al.¹³

DATA REDUCTION

The amount of gas adsorbed is required to obtain the isotherm parameters of the single-component adsorbate and adsorbent system. It is calculated using the density, which is a function of temperature and pressure of the charging cell. In the initial stage of the experiment, the charging cell was pressurized with adsorbate (methane gas). The mass of the adsorbate in a particular state is calculated using eq 1:

$$m_i = \rho_{ab_i}(P, T_{\text{charging}})V_{\text{charging_cell}} \quad (1)$$

where ρ_{ab_i} is the initial adsorbate density at charging cell temperature and volume, that is, T_{charging} and $V_{\text{charging_cell}}$, respectively.

Assuming that the adsorption occurs in the micropores, thus the void volume, V_{void} in the adsorption cell is calculated as:

$$V_{\text{void}} = V_{\text{ad_cell}} - \frac{m_{\text{ac}}}{\rho_{\text{solid}}} - v_{\mu}m_{\text{ac}} \quad (2)$$

Here $V_{\text{ad_cell}}$ is the adsorption cell volume, m_{ac} is the mass of activated carbon, ρ_{solid} is the solid density of activated carbon, and v_{μ} is the micropore volume of the activated carbon. The vapor space volume in the adsorption cell is accounted for in the calculation of the void volume. Therefore, the mass of adsorbate in the void volume is calculated as $m_{\text{void}} = \rho_{\text{ab}}(P, T_{\text{ads}})V_{\text{void}}$ at T_{ads} , while ρ_{ab} denotes the density of gas (adsorbate) in the adsorption cell. The amount of adsorbate left at each isotherm is calculated using:

$$m_f = \rho_{ab_f}(P, T_{\text{ads}})V_{\text{total}} \quad (3)$$

where $V_{\text{total}} = V_{\text{charging_cell}} + V_{\text{void}}$. $V_{\text{charging_cell}}$ is the charging cell volume, and ρ_{ab_f} is the adsorbate density at the respective isotherm temperature. The density of the adsorbate, ρ_{ab} , is determined using the Setzmann and Wagner correlations.¹⁴ The amount of gas adsorbed, m_{ads} , can then be estimated from:

$$m_{\text{ads}}(P, T, C) = m_i(P, T) - m_f(P, T) \quad (4)$$

Table 4. Experimental Uptake Data for the Adsorption of CH₄ on Carbotech Commercial Activated Carbon

<i>P</i>	<i>C</i>	<i>P</i>	<i>C</i>	<i>P</i>	<i>C</i>
kPa	kg·kg ⁻¹	kPa	kg·kg ⁻¹	kPa	kg·kg ⁻¹
T = 300 K		T = 308 K		T = 318 K	
62.87	0.008	65.44	0.007	65.44	0.006
143.71	0.014	143.93	0.012	147.02	0.011
258.45	0.021	239.56	0.017	244.38	0.016
479.62	0.029	498.64	0.027	477.22	0.024
715.04	0.036	721.77	0.033	717.31	0.030
1017.96	0.043	995.84	0.038	971.50	0.035
1447.22	0.048	1463.85	0.045	1439.61	0.041
1921.46	0.054	1961.17	0.050	1936.57	0.045
2483.84	0.057	2467.09	0.053	2478.85	0.049
2971.65	0.058	2974.83	0.056	2949.98	0.052
		3519.64	0.059	3518.04	0.055

Here, m_{ads} is dependent on pressure, temperature, and specific uptake. Finally the specific uptake, C , is determined as

$$C = \frac{m_{\text{ads}}}{m_{\text{ac}}} \quad (5)$$

Three different adsorption isotherm models, namely, (i) Langmuir, (ii) Tóth, and (iii) D-A, were used to correlate the experimental data. The Langmuir model is the simplest model which describes monolayer type adsorption (type I isotherm of the IUPAC classification) in microporous solids, such as the adsorption of methane in activated carbon. The Langmuir model presumes a homogeneous surface of the adsorbents where the adsorption energy is constant over all sites. This model also assumes that the adsorption on the surface is localized and each site can accommodate only one molecule or atom.¹⁵ The Langmuir model is written as

$$\frac{C}{C_0} = \frac{k_0 \exp\left(\frac{h_{\text{st}}}{RT}\right)P}{1 + k_0 \exp\left(\frac{h_{\text{st}}}{RT}\right)P} \quad (6)$$

where C_0 is the saturated amount adsorbed, P is the equilibrium pressure, k_0 is the equilibrium constant, h_{st} is the isosteric heat of adsorption, and R is the gas constant. The Langmuir model has limitations to fit high pressure data and for material heterogeneity.

The Tóth model is commonly used for heterogeneous adsorbents such as activated carbon to predict the behavior at both low and high pressure conditions^{15,16} and is represented by,

$$\frac{C}{C_0} = \frac{k_0 \exp\left(\frac{h_{\text{st}}}{RT}\right)P}{\left\{1 + \left[k_0 \exp\left(\frac{h_{\text{st}}}{RT}\right)P\right]^t\right\}^{1/t}} \quad (7)$$

where C is the amount adsorbed, C_0 is the limiting adsorbed amount, P is the equilibrium pressure, k_0 is the equilibrium constant, h_{st} is the isosteric heat of adsorption, R stands for the gas

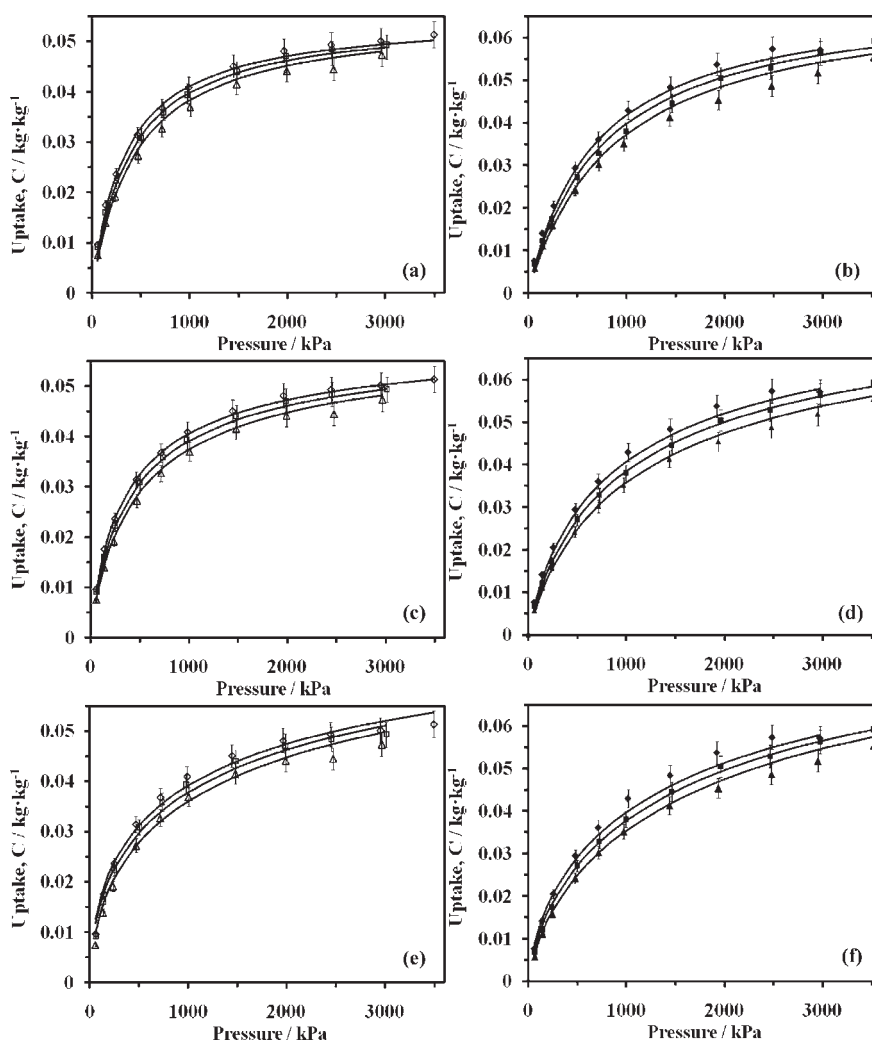


Figure 3. Adsorption isotherms of CH₄ on KT (◇, 300 K; □, 308 K; △, 318 K) and Carbotech (◆, 300 K; ■, 308 K; ▲, 318 K) activated carbons. Solid lines represent the Langmuir model for CH₄ on (a) KT and (b) Carbotech, Tóth model for CH₄ on (c) KT and (d) Carbotech, and D-A model for CH₄ on (e) KT and (f) Carbotech activated carbons.

constant, and t is the parameter that indicates the heterogeneity of the adsorbent. Similarly, the D-A equation is written as:

$$C = C_0 \exp \left[- \left(\frac{A}{E} \right)^n \right] \quad (8)$$

or

$$\ln C = \ln C_0 \left[- \left(\frac{A}{E} \right)^n \right] \quad (9)$$

where A is the adsorption potential and C is the adsorbed volume. C_0 is the limiting volume of adsorption space of the adsorbent, E is the characteristic energy of the adsorption system, and n is the structural heterogeneity parameter. The adsorption potential A is given by,

$$A = RT \ln \left(\frac{P_s}{P} \right) \quad (10)$$

where R is the gas constant, T is the equilibrium temperature, and P_s is the saturation pressure. The pseudovapor pressure, P_s , at a given isotherm temperature is calculated by the Dubinin's method¹⁷

and is widely used for methane adsorption.^{11,18,19}

$$P_s = \left(\frac{T}{T_c} \right)^2 P_c \quad (T > T_c) \quad (11)$$

where P_c and T_c are the critical pressure and the critical temperature of methane.

The isosteric heat of adsorption (h_{st}) is the evolution of heat during the adsorption process since the adsorbate molecules are more stabilized on the adsorbent surface than in the bulk phase. This parameter is useful for the thermal management of any adsorption system. The values of the heat of adsorption (h_{st}) have been extracted from the isotherm data using the Clausius–Clayperon equation along with the correction term for the non-ideality of the gaseous phase, recently derived by Chakraborty et al.,²⁰ which is as follows.

$$h_{st} = RT^2 \left[\left(\frac{\partial(\ln P)}{\partial T} \right)_C \right] + T v_g \frac{dP}{dT} (P, T) \quad (12)$$

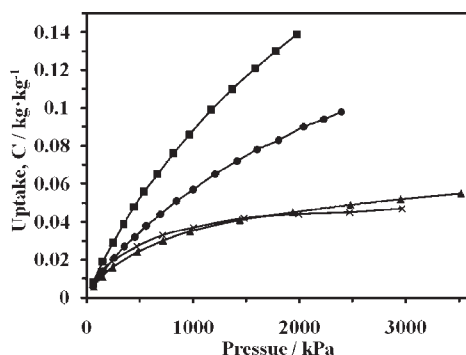
Here, the first term of the right-hand side is derived from the Clausius–Clayperon equation and can be expanded by using the

Table 5. Adsorption Parameters (C_0 , k_0 , h_{st} , and t) for Langmuir and Tóth Models

parameters	KT activated carbon		commercial activated carbon, Carbotech	
	Langmuir	Tóth	Langmuir	Tóth
$C_0/\text{kg}\cdot\text{kg}^{-1}$	0.055	0.061	0.070	0.084
$h_{st}/R/K$	1341.6	1471.4	1388.0	1508.9
k_0/kPa^{-1}	$3.38\cdot 10^{-5}$	$2.69\cdot 10^{-5}$	$1.43\cdot 10^{-5}$	$1.05\cdot 10^{-5}$
t	1	0.77	1	0.73
avg. regression err (%)	2.93	1.90	4.1	2.89

Table 6. Adsorption Parameters (C_0 , E , and n) for the D-A Isotherm Model

parameters	KT activated carbon	commercial activated carbon, Carbotech
$C_0/\text{kg}\cdot\text{kg}^{-1}$	0.062	0.072
$E/\text{J}\cdot\text{mol}^{-1}$	9835.8	8258.3
n	1.60	1.66
avg. regression err (%)	4.69	2.55

Figure 4. Adsorption isotherms for CH₄ on activated carbons Maxsorb III (■), ACF-A20 (●), Carbotech (▲), and KT (×) at 318 K.

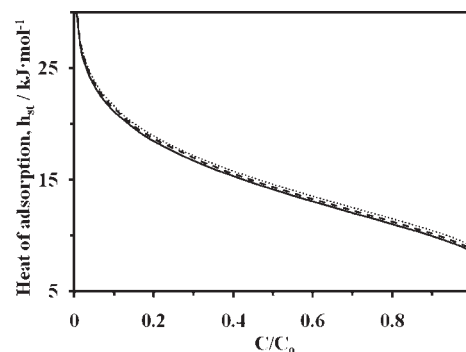
D-A isotherm model. A second term is introduced which defines the behavior of the adsorbed mass with respect to both the pressure and the temperature changes during an adsorbate uptake, which occur due to the nonideality of the gaseous phase. Ultimately, eq 12 becomes

$$h_{st} = 2RT + E \left(\ln \frac{C_0}{C} \right)^{1/n} + T v_g \frac{dP}{dT} (P, T) \quad (13)$$

where v_g is the specific volume of the vapor phase and dP/dT represents the gradient of the pressure with the temperature of the adsorbate.

RESULTS AND DISCUSSION

The measured uptake data for the adsorption of methane on activated carbon derived from the East of Kalimantan, Indonesia and Carbotech commercial activated carbon are furnished in Tables 3 and 4. The experiments cover the temperature ranging from (300 to 318) K and pressures up to 3.5 MPa. The isotherms data for the KT (empty symbols, ◇, 300 K; □, 308 K; △, 318 K) and Carbotech (filled symbols, ◆, 300 K; ■, 308 K; ▲, 318 K) activated carbons with methane are graphically presented on a

Figure 5. Isothermic heat of adsorption for CH₄ on KT activated carbon using a corrected correlation (eq 13) at 300 K (solid line), 308 K (dashed line), and 318 K (dotted line).

pressure–concentration–temperature (P – C_0 – T) plane in Figure 3. It is observed that the experimental uptake profiles are categorized as a Type 1 adsorption isotherm in the IUPAC classification. The specific uptake, C_0 , for each specimen is considerably different at any given set of conditions.

The regressed isotherms with the Langmuir and Tóth models are superimposed onto the isotherms data for KT + CH₄ and Carbotech + CH₄ in Figure 3a,b, and Figure 3c,d, respectively. The numerical value of the adsorption parameters (C_0 , k_0 , h_{st} , and t) are presented in Table 5. The error regression has also been calculated using the isotherm model results with the experimental data.

$$\text{error of regression} = \frac{\sqrt{\frac{1}{N} \sum_{i=1}^N (C_{\text{expt}} - C_{\text{model}})^2}}{\frac{1}{N} \sum_{i=1}^N C_{\text{expt}}} \quad (14)$$

where N is the number of data points.

The Tóth model provides a better fit than the Langmuir model to the experimental adsorption isotherms of methane for both of the adsorbents due to the presence of the heterogeneity parameter (t). However, the Langmuir model is simpler than the Tóth model and also does not deviate much from the measured uptake values for the present pressure and temperature ranges.

Similarly, the D-A isotherm model is used to fit the uptake data of methane on KT and Carbotech activated carbon as shown in Figure 3e,f, respectively. The adsorption parameters (C_0 , E , and n) are listed in Table 6. It is observed that all of the isotherm models (Langmuir, Tóth, and D-A models) are suitably accurate with experimental uncertainties of less than 5.0 % to predict the uptake of CH₄ on the activated carbons.

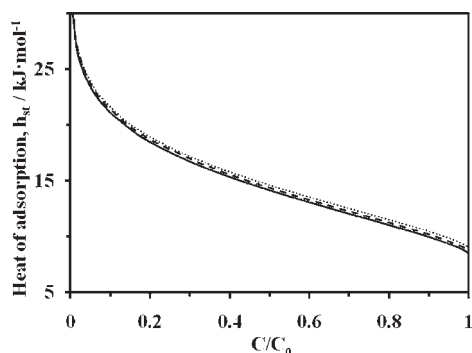


Figure 6. Isosteric heat of adsorption for CH_4 on Carbotech activated carbon using a corrected correlation (eq 13) at 300 K (solid line), 308 K (dashed line), and 318 K (dotted line).

From Figure 3, it is found that the equilibrium uptake for Carbotech activated carbon is higher compared to KT activated carbon. This may be attributed to the Carbotech having a larger surface area and total pore volume compared to KT. This is further explained in Figure 4 with the uptake value for KT, Carbotech, ACF-A20, and Maxsorb III with CH_4 at 318 K are plotted against the system pressures. Maxsorb III²¹ with the highest total pore volume has the highest uptake capacity, followed by the ACF-A20,⁹ Carbotech, and KT activated carbon.

From the Langmuir and Tóth isotherm models, the isosteric heat of adsorption, h_{st} can be obtained in the range of (11.15 to 12.54) $\text{kJ} \cdot \text{mol}^{-1}$. The isosteric heat of adsorption determined is comparable to a previous study by Loh et al.,²² that is, (11.97 to 13.39) $\text{kJ} \cdot \text{mol}^{-1}$ for methane with activated carbons Maxsorb III and ACF-A20. This is mainly due to Maxsorb III and ACF-A20 being both highly porous and having a larger total pore volume, ($2.1 \cdot 10^{-3}$ and $1.01 \cdot 10^{-3}$) $\text{m}^3 \cdot \text{kg}^{-1}$, respectively, compared to the KT and Carbotech activated carbons.

Figures 5 and 6 show the uptake-dependent heat of adsorption (h_{st}) at different isothermal conditions for both KT and Carbotech, respectively. The isosteric heat of adsorption has been extracted from eq 13. One observes that h_{st} decreases with increasing vapor uptake. It can be seen that, during the initial stage of adsorption, methane is adsorbed rapidly onto the sites of high energy. Methane molecules first penetrate into narrower pores, resulting in a stronger interaction between the adsorbate and the adsorbent. This implies a higher value of h_{st} at lower loadings. After filling up the smaller pores, methane molecules are accommodated gradually in larger pores, in which the adsorption affinity becomes weaker. A monotonic decrease in the h_{st} as a function of loading is therefore observed.

CONCLUSIONS

The adsorption isotherm of methane gas (CH_4) on activated carbon has been measured at working conditions expected of ANG in portable cylinders, namely, pressures up to 3.5 MPa and temperature ranges from (300 to 318) K using a static volumetric method. It is found that the activated carbon derived from the East of Kalimantan coal has a lower adsorption capacity than the commercially available type owing to a lower specific pore volume and surface area. The isosteric heat of adsorption is evaluated from the measured isotherm data and varies from (8.45 to 27.5) $\text{kJ} \cdot \text{mol}^{-1}$. Such basic experimental isotherms and isosteric heat of adsorption of methane with KT and Carbotech

activated carbons are useful in designing an effective ANG storage system.

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