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Carbon dioxide-methane mixture adsorption on activated carbon

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Abstract In this work, we report new experimental data of pure and binary adsorption equilibria of carbon dioxide and methane on the activated carbon RB2 at 273 and 298 K. The pressure range studied were 0–3.5 MPa for pure gases and 0–0.1 MPa for mixtures. The combination of the generalized Dubinin model to describe the pure CO_2 and CH_4 isotherms with the IAST (Ideal Adsorbed Solution Theory) for the mixtures provide a method for the calculation of the binary adsorption equilibria. This formulation predicts with acceptable accuracy the binary adsorption data and can easily be integrated in general dynamic simulation of PSA (pressure swing adsorption process) adsorption columns. It involves only three parameters, independent of the temperature, and directly determined with only one adsorption isotherm of CO_2.

Keywords Adsorption · Mixture · Activated carbon · Volumetric method · Experimental data · Modelling

1 Introduction

Gas adsorption is of particular interest especially because it is involved in numerous processes linked to environmental protection. Gases such as carbon monoxide, carbon dioxide, and hydrogen sulphide are removed by pressure swing adsorption (PSA). PSA is also involved in organic solvent vapour recovery or separation and purification of hydrogen from steam-methane reforming. In many gas adsorption processes activated carbons operate as adsorbents and the basic physical phenomenon is the adsorption of a gas mixture.

Numerous experimental data are available in literature for pure gas isotherms. At the opposite, development and validation of multi-components theoretical models still suffer of a relative lack of data (Siperstein and Myers, 2001). The first objective of this paper is to provide a new set of data for the binary mixture CO_2/CH_4. It is currently involved in hydrogen production and is, to date, one of most studied mixture. Adsorption data are available in the open literature (Buss, 1995; Dreisbach et al., 1999; Van Der Vaart et al., 2000) allowing comparisons with previous experimentations.

Representative chemical engineering modelling of fixed bed adsorbers needs to take into consideration coupling phenomena between multicomponent adsorption equilibria and the heat and mass transfers inside the adsorbers that generate pressures and temperatures gradients (Yang, 1986; Warmuzinski and Tanczyk, 1997). Within this framework, validation over a wide range of temperature and loading of robust, simple and self-consistent multicomponent adsorption equilibria models is the basic step. The semi-empirical Dubinin approach for pure gas isotherms modelling and more particularly the Dubinin-Astakhov (DA) (Dubinin, 1975) equation has demonstrated its ability to predict

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the adsorption capacities of activated carbons. It takes advantage to involve parameters independent of the temperature and linked to the microtexture of the active carbon. In the case of carbon dioxide storage (Goetz and Guillot, 2001) and methane storage by adsorption (Biloé et al., 2001; Goetz and Biloé, 2005), the DA equation has been already integrated with success in macroscopic models coupling adsorption and heat and mass transfer in a storage vessel. For binary or multicomponent gas mixtures, the Ideal Adsorbed Solution Theory (IAST) (Myers and Prausnitz, 1965) is a reference model. The combination of the IAST with a formulation of the pure gases isotherms with the DA equation (Lavanchy et al., 1996) provides a simple and general mathematical formulation to estimate equilibrium conditions with a minimum of parameters and data input. The second objective of the present paper is to test this formulation in the case of the CO2/CH4 mixture at pressure range corresponding to current PSA processes.

2 Experimental study

2.1 Experimental laboratory apparatus and measurement method

Various experimental methods can be considered for co-adsorption isotherms achievement. The major ones are gravimetric method (Buss, 1995; Van der Vaart et al., 2000), gravimetric-volumetric method (Dreisbach et al., 1999; Keller et al., 1999) and volumetric method (Lewis and al., 1950; Kaul, 1987; Costa et al., 1991; Russel and Le Van, 1997). The last one corresponds to a full measurement of equilibria in the sense that the amount of each adsorbed species is directly determined by experimental measurements (Talu, 1998). Kaul (1987) presented the modern version of volumetric apparatus for measuring gas-solid equilibrium data. Like most of the volumetric devices encountered in the devoted literature, the apparatus developed at the laboratory and sketched in Fig. 1 takes inspiration of this system.

The room containing the equipment is maintained under air-conditioning to keep an almost constant ambient temperature equal to 298 K with a maximum variation of 2 K. The apparatus is composed of a close loop containing the adsorption cell with an approximate capacity of 10^{-3} kg of activated carbon. A pump (Sercom GK-M24/02) circulates the gas mixture in the overall circuit to avoid concentration gradient and to reduce the time needed to reach the equilibrium conditions. During measurements, the circulating loop was put inside a thermostated bath (Lauda RE230) which provides a temperature control with accuracy better than 0.1 K. The pressure of the gaseous phase in the circuit

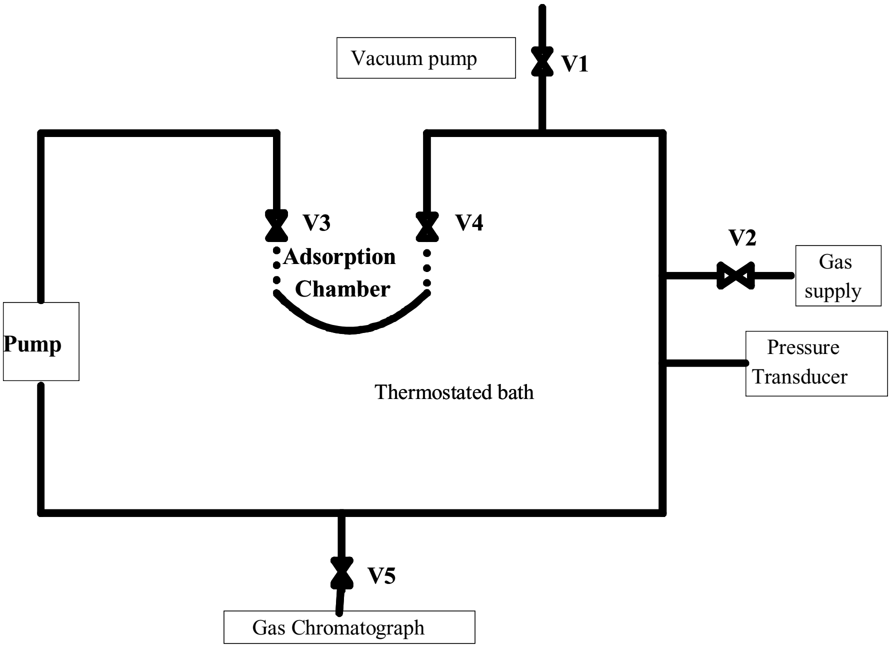


Fig. 1 Schematic diagram for volumetric-chromatographic measurements of multicomponent gas adsorption equilibria (V: Valve)

Table 1 Concentration (molar percentage) of the different initial gas compositions

	Mixture 1	Mixture 2	Mixture 3
CH_4	25.0	49.5	74.9
CO_2	75.0	50.5	25.1

was measured with a pressure sensor (Mensor 2100) with a full scale equal to 4 MPa and a precision of 0.4×10^{-3} MPa. In the case of gas mixture, composition was analysed with a gas chromatograph (GC 3800 Varian) with accuracy, expressed in mole fraction, better than 2%. The gas circuit is evacuated with a vacuum system composed of a primary and a turbomolecular (Edwads EXT70H) pumps which allows pressure down to 10^{-3} Pa. Finally, the circuit is connected to a gas supply system.

The adsorbent used for this investigation was the commercial activated carbon Norit RB2. The pure gases, carbon dioxide and methane, as well as the gas mixtures (Table 1) were purchased from Air Liquide at a purity grade higher than 99.99%.

Before every experiment, the activated carbon was dried and degassed at 523 K and at a pressure lower than 10^{-3} Pa during 12 hours. Then, the gaseous circuit was dipped into the thermostated bath. The mass balance applied to the gaseous phase according to the following equation directly provides the quantity of adsorbed gas:

$$(N_{ads})_i = (N_I)_i - (N_G)_i \tag{1}$$

where i represents (in the case of a gas mixture) the considered species, N_I is the number of mole of gas introduced and N_G is the number of remaining mole of gas when the equilibrium conditions are reached. The numbers of mole of gases are calculated using a PVT measurement with compressibility factors calculated by the Virial equation of state terminated after the second virial coefficient (Reid et al., 1987).

2.2 Experimental results

The gas adsorption isotherms for pure methane and pure carbon dioxide on the activated carbon Norit RB2 at the two temperatures of 273 K and 298 K and pressure up to 3.5 MPa are presented in Fig. 2.

For the binary gas-mixture, the pressures range from 0.1 MPa to 1 MPa (Tables 1 and 2). Adsorbed quantities as a function of the gaseous composition at an "almost" constant pressure of 0.5 MPa are presented in Fig. 3. The experimental values (at 298 K and 273 K) present a coherent and continuous evolution of the adsorbed amount between the two particularly points corresponding to adsorption of pure methane ($y_{CO_2} = 0$) and pure dioxide of carbon ($y_{CO_2} = 1$). Results available in the literature for activated carbons with similar microtextures show a similar evolution of the adsorbed phase composition (Fig. 4).

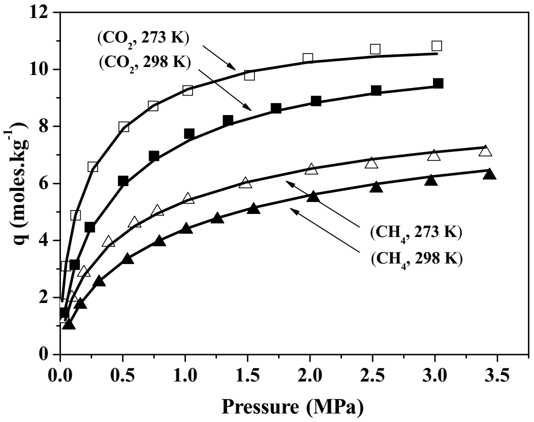


Fig. 2 Adsorption isotherms for pure methane (△: 273 K, ▲ 298 K) and pure carbon dioxide (□: 273 K, ■: 298 K). Solid lines correspond to calculated values using the Dubinin Astakhov equation

3 Prediction of the equilibrium adsorption of the binary CO_2/CH_4 mixtures

3.1 Modelling pure gas isotherms with the theory of the micropores volume filling

The adaptation of the Polanyi potential theories leads to the Dubinin-Astakhov (DA) equation (Dubinin, 1975):

$$W = W_o \cdot \exp\left[-\left(\frac{\varepsilon}{\beta \cdot E_o}\right)^n\right] \tag{2}$$

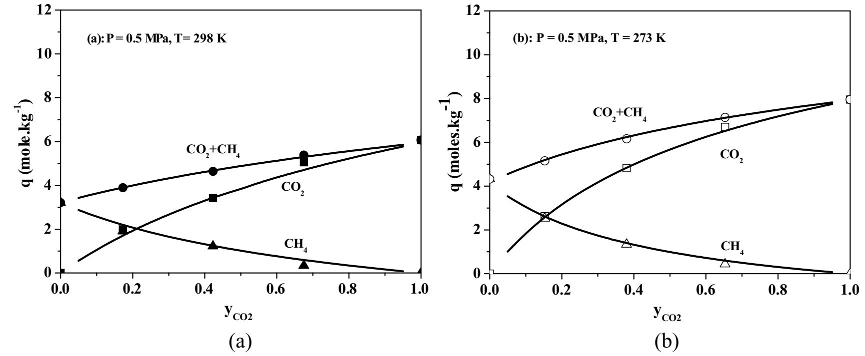
where ε, the Polanyi potential, is:

$$\varepsilon = R \cdot T \cdot \ln\left(\frac{P_{sat}(T)}{P}\right) \tag{3}$$

W is the volume of adsorbed gas at the relative pressure of (P_{sat}/P) and W_o is the total micropore volume. The exponent n lies in the range of 1–3; its value depends mainly on the pore size distribution; E_o, the

Table 2 Adsorbed phase compositions of methane – carbon dioxide at 298 K experimentally measured and calculated with MPD model

P MPa	Y_{CO_2}	q_{CO_2} exp mole · kg^{-1}	q_{CH_4} exp mole · kg^{-1}	q_{TOT} exp mole · kg^{-1}	q_{CO_2} cal (MPD) mole · kg^{-1}	q_{CH_4} cal (MPD) mole · kg^{-1}	q_{TOT} cal (MPD) mole · kg^{-1}	Relative error for q_{TOT} (%)
0.1016	0.62	2.19	0.29	2.49	2.03	0.35	2.38	4.4
0.0995	0.35	1.41	0.61	2.02	1.27	0.69	1.96	2.9
0.1109	0.16	0.68	0.95	1.63	0.69	1.105	1.74	6.7
0.5015	0.68	5.05	0.33	5.38	4.65	0.60	5.25	2.4
0.4963	0.42	3.42	1.22	4.64	3.37	1.26	4.63	0.2
0.4997	0.17	1.99	1.90	3.89	1.67	2.22	3.89	0
1.0087	0.70	6.54	0.31	6.85	6.12	0.67	6.79	0.8
1.0094	0.45	4.73	1.29	6.03	4.59	1.52	6.11	1.3
0.9973	0.20	2.69	2.47	5.17	2.50	2.75	5.25	1.5



(a)

(b)

Fig. 3 Co-adsorption isotherms on a commercial activated carbon for the binary mixture methane/carbon dioxide at 0.5 MPa (a): 298 K (solid symbols), b): 273 K (open symbols). (circles: total amount adsorbed; triangles; amount of adsorbed CH_4; squares: amount of adsorbed CO_2). Solid lines correspond to calculated values with MPD model

characteristic energy is connected to the mean pore size. The affinity coefficient β depends of the adsorbate. In the case of benzene, the reference vapour, its value is equal to one.

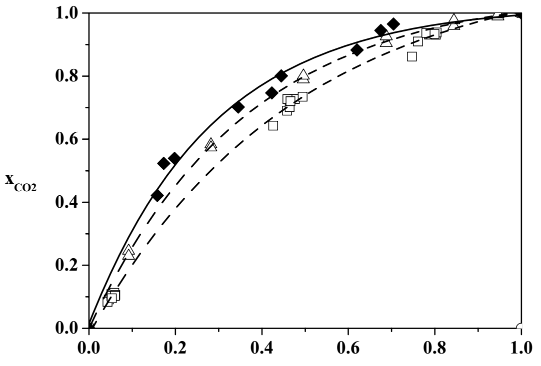


Fig. 4 Comparison between our laboratory experimentals results (♦) and those available in the literature for the binary mixture CO_2/CH_4 at several MPa : △ (Buss ,1995); ☐ (Dreisbach et al., 1999)

The adsorbent capacity of the activated carbon (expressed in moles of adsorbed fluid per kilogram of activated carbon) is given by:

$$q(T, P) = \frac{\rho_{ads}(T)}{M} \cdot W \qquad (4)$$

with ρ_{ads} the density of the adsorbed phase estimated from the relationship proposed by Ozawa et al. (1976):

$$\rho_{ads}(T) = \frac{\rho_{boil}}{\exp(2.5 \cdot 10^{-3} \cdot (T - T_{boil}))} \qquad (5)$$

with the properties at the boiling point replaced by the properties at the triple point in the case of CO_2.

Finally, beyond the critical point, the saturated vapour pressure (P_{sat}) is determined by the extrapolation

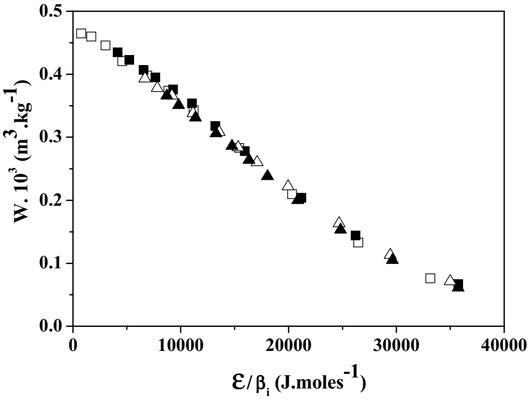


Fig. 5 Characteristic curves for pure methane (△: 273 K, ▲ 298 K) and pure carbon dioxide (□: 273 K, ■: 298 K) with $\beta_{CO_2} = 0.34$ and $\beta_{CH_4} = 0.35$

proposed by Dubinin (1960):

$$P_{sat}(T) = P_{crit} \cdot \left(\frac{T}{T_{crit}}\right)^2 \tag{6}$$

At high pressure, gases can no longer be considered as ideal gases, eqs. 2 and 3 are expressed in terms of fugacity. The fundamental requirement of the Dubinin's theory is the temperature and the adsorbate invariance of the parameters W_o, E_o, n. This is illustrated in Fig. 5 by the single experimental characteristic curve ($W = f(\varepsilon/\beta)$) for CO_2 and CH_4 at the two temperatures.

The best fit of the CO_2 adsorption isotherm at 273 K is provided with $W_o = 0.45\ 10^{-3}$ m^3.kg, $E_o =$ 23800 J.mole^{-1} and $n = 1.66$. This isotherm is particularly interesting in our case because it corresponds to the largest range of Polanyi potential covered by the experimental measurements and leads to an almost saturated adsorbent (Fig. 5). Those parameters used directly as model inputs, leads to a prediction of the other experimental isotherms with accuracy higher than 5% (Fig. 2). In other terms, after the needed identification step of the three parameters related to the microtexture of the considered activated carbon, the DA equation is an efficient tool to extrapolate the adsorption capacity at different temperatures but also different adsorbates.

3.2 Binary equilibria

Lavanchy et al. (1996) derived an analytical solution for the integration of DA equation to calculate the spreading pressures (π) given by the Gibbs isotherm. That leads to the simple expression for the integral corresponding to the ψ_i functions:

$$\begin{aligned}\psi_i &= \frac{A \cdot \pi}{R \cdot T} = \int_0^{f^\bullet_{\pi,i}} \frac{q^\bullet_i}{f^\bullet_i} df \\ &= W_o \cdot \frac{\rho_{ads,i}(T)}{M} \cdot \frac{\beta_i \cdot E_o}{n \cdot R \cdot T} \cdot \\ &\quad \Gamma\left[\frac{1}{n}, \left(\frac{R \cdot T}{\beta_i \cdot E_o} \cdot \ln\left(\frac{f^\bullet_{sat,i}(T)}{f^\bullet_{\pi,i}}\right)\right)\right]\end{aligned} \tag{7}$$

with Γ the incomplete gamma function.

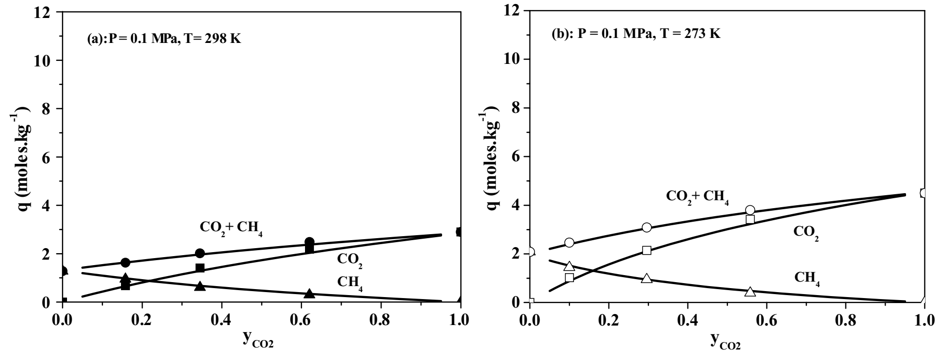
The fundamental requirement for the co-adsorption equilibria of binary or multi-component gas mixtures estimated with the IAST (Myers and Prausnitz, 1965) is the equality of the ψ functions of each species and the equality of the chemical potentials of the gaseous and the adsorbed phases (Eq. 8). The Gibbs free energy change during the mixing provides the total number of adsorbed moles (Eq. 9).

$$f^\bullet_{\pi,i} \cdot x_i = \phi_i \cdot P \cdot y_i \tag{8}$$

$$\frac{1}{q_t} = \sum_i \frac{x_i}{q^\bullet_{\pi,i}} \tag{9}$$

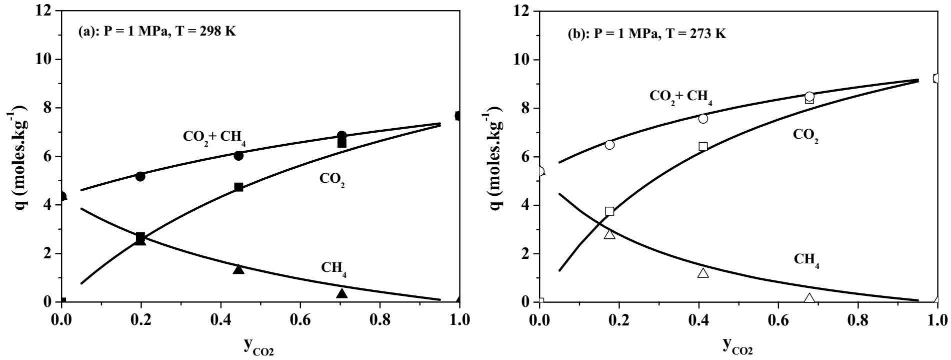
The combination of the IAST with a formulation of the pure gas isotherms with the help of DA equation was named Myers-Prausnitz-Dubinin (MPD) model (Lavanchy et al., 1996). There are different fundamental bases between the Dubinin's approach and the IAST. The DA equation is particularly adapted to heterogeneous surfaces while IAST pertains to homogeneous surfaces. Nevertheless, there is no mathematical inconsistency to use the equations involve in the Dubinin's approach for single gas isotherm and in the IAST for the mixture. As stated by Valenzuela and Myers (1989) who tested the combination of the IAST with the Toth equation, the IAS predictions are independent of the particular equation adopted for the pure gas isotherms. The requirement is a high fit quality between the single gas experimental data and those calculated with the selected model. In this way, the DA equation can be considered only as the function that fits the pure gas data, and nothing more.

In the objective to test a model adapted to a chemical engineering approach, the DA equation presents the advantage to involve parameters that are independent of the temperature.



(a) (b)

Fig. 6 Co-adsorption isotherms CO_2-CH_4 at 0.1 MPa (a): 298 K (solid symbols), b): 273 K (open symbols). (circles: total amount adsorbed; triangles: amount of adsorbed CH_4; squares: amount of adsorbed CO_2). Estimated values with MPD theory (solid lines)



(a) (b)

Fig. 7 Co-adsorption isotherms CO_2-CH_4 at 1 MPa (a): 298 K (solid symbols), b): 273 K (open symbols). (circles: total amount adsorbed; triangles amount of adsorbed CH_4; squares: amount of adsorbed CO_2). Estimated values with MPD theory (solid lines)

The MPD model was previously tested in the case of different binary equilibria of vapour at low pressure (Garrot et al., 2002). It was also used with success for the calculation of breakthrough curves for pressure swing process in the case the ternary system 2-chloropropane-chlorobenzene-carbon tetrachlorure (Lavanchy and Stoeckli, 1997). Thus, it is of particular interest to test its ability in the case of co-adsorption of binary mixtures at several bars.

Figures 3, 6 and 7 demonstrate the good agreement between the calculated equilibria conditions and the experimental values. Relative errors obtained for the total number of moles adsorbed are less than 5% (Tables 2 and 3). It is important to underline that for the whole pressure range (0.1–1 MPa) and for the two temperatures (273 and 298 K) the co-adsorption equilibria are calculated with only three parameters (W_o,E_o, n) provided by the isotherm of CO_2 at 273 K.

Figure 8 summarizes the results corresponding to the evolution of the composition of the adsorbed phase as a function of the composition of the gaseous phase. It confirms the global coherence of the model that allows a correct estimation of the adsorbed amount of each component whatever are the conditions of temperature, pressure and gas composition. As previously suggested (Costa et al., 1989; Van der Vaart et al., 2000), the introduction of activity coefficients related to the adsorbed phase could improve the small discrepancy between calculated and experimental values. Nevertheless, to date, there are not reliable correlations available to predict the activity coefficients.

Van der Vaart et al. (2000) measured the adsorption equilibria of CO_2/CH_4 mixtures of different compositions on the microporous activated carbons Norit RB1. This adsorbent presents a very similar microtexture to that of the RB2. Like for the RB2, identification

Table 3 Adsorbed phase compositions of methane – carbon dioxide at 273 K experimentally measured (exp) and calculated with MPD model

P MPa	Y_{CO_2}	q_{CO_2} exp $mole \cdot kg^{-1}$	q_{CH_4} exp $mole \cdot kg^{-1}$	q_{TOT} exp $mole \cdot kg^{-1}$	q_{CO_2} cal (MPD) $mole \cdot kg^{-1}$	q_{CH_4} cal (MPD) $mole \cdot kg^{-1}$	q_{TOT} cal (MPD) $mole \cdot kg^{-1}$	Relative error for q_{TOT} (%)
0.1119	0.56	3.41	0.39	3.80	3.36	0.50	3.86	1.6
0.1065	0.29	2.14	0.94	3.08	2.14	0.98	3.12	1.3
0.1029	0.10	1.03	1.44	2.47	0.88	1.55	2.43	1.6
0.5194	0.65	6.70	0.43	7.13	6.52	0.61	7.13	0
0.5156	0.38	4.82	1.13	6.15	4.83	1.43	6.26	1.8
0.5125	0.15	2.62	2.53	5.15	2.57	2.65	5.22	1.4
1.0111	0.68	8.36	0.12	8.48	7.89	0.63	8.52	0.4
1.0173	0.41	6.42	1.15	7.57	6.17	1.54	7.71	1.9
1.0508	0.18	3.75	2.74	6.49	3.62	3.06	6.68	2.9

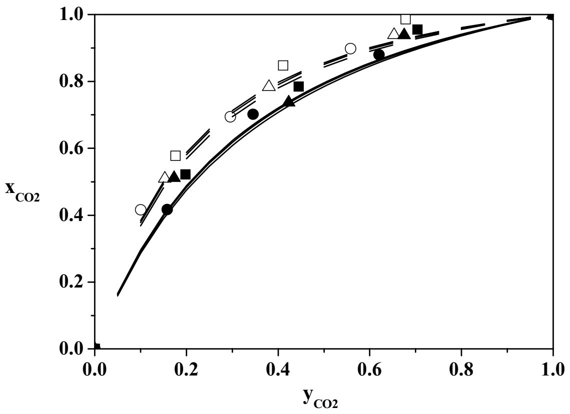


Fig. 8 Adsorbed phase composition at 298 K (solid symbols) and 273 K (open symbols), and at different pressure (circles: 0.1 MPa, triangles: 0.5 MPa, squares: 1 MPa). Modelisation with MPD theory (solid lines: 298 K, dashed lines: 273 K)

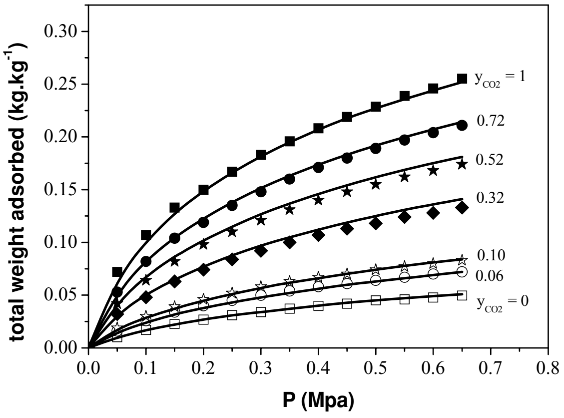


Fig. 9 Adsorbed weight of CO_2-CH_4 mixture in RB1 at 302.9 K; comparison between predicted values with MPD model (solid lines) and experimental data of Van der Vaart et al. (2000): $Y_{CO_2} = 0$ (□); 0.06 (○); 0.1 (◇); 0.32 (♦); 0.52 (⋆); 0.72 (●); 1 (■)

of the parameter ($W_o = 0.43\ 10^{-3}$ m^3.kg, $E_o = 23340$ J.mole^{-1}, $n = 1.68$) was made with the adsorption isotherm of pure gases (302.9 K in this case). The comparison between the experimental isotherms and the ones calculated by the MPD model (Fig. 9) demonstrates for this other activated carbon the ability of the MPD model to predict with an acceptable accuracy the equilibria.

4 Conclusion

Experimental equilibrium measurements were performed for adsorption of carbon dioxide, methane and their mixtures at two different temperatures (273 and 298 K) and for pressure up to 1 MPa (3.5 MPa for pure gases). Single-gas isotherms were analysed in terms of Dubinin-Astakhov equation with the three parameters connected to the microtexture of the activated carbon. For the studied system, we have outlined the efficiency of the MPD model to predict the experimental binary CO_2/CH_4 mixture adsorption equilibria on an activated carbon. When the adsorbed phase can be considered as ideal, the main advantage of the MPD model, a combination of IAST with DA, is to provide a predictive method that needs very few experimental results. Basically, (W_o, E_o, n) can be obtained with the pure CO_2 isotherm at 273 K. To check the validity of the suggested model for adsorption of CO_2/CH_4 mixture, these results have to be extended at pressure upper than 1 MPa and in the case of others activated carbons with different microtexture properties. In the same way, future work will be devoted to the study of strongly non-ideal adsorbed phase to test the combination of the real adsorbed solution theory with modelisation of the isotherms with the Dubinin's theory. Finally, and as claimed at the beginning of the paper, the tested model is intended as a practical engineering approach rather than a fundamental description of multicomponent adsorption equilibrium. Engineering requires rapid methods with a set of parameters as few as possible for obtaining good approximation and prediction of co-adsorption equilibria over a wide range of pressures, compositions and temperatures; in that role the MPD model could be extremely successful.

Notations

E_o	caracteristic energy ($\mathrm{J \cdot mole^{-1}}$)
f	fugacity (Pa)
M	molar mass ($\mathrm{kg \cdot mole^{-1}}$)
N	number of moles (mole)
n	coefficient of DA equation (–)
P	pressure (Pa)
q	mole adsorbed ($\mathrm{mole \cdot kg^{-1}}$)
R	gas perfect constant ($\mathrm{J \cdot K^{-1} \cdot mole^{-1}}$)
T	temperature (K)
W	adsorbed volume ($\mathrm{m^3 \cdot kg^{-1}}$)
W_o	total microporous volume ($\mathrm{m^3 \cdot kg^{-1}}$)
x	molar fraction in the adsorbed phase
y	molar fraction in the gaseous phase

Greek Letters

β	affinity coefficient (–)
ϕ	fugacity coefficient (–)
π	spreading pressure ($\mathrm{J \cdot m^{-2}}$)
ρ	density ($\mathrm{kg \cdot m^{-3}}$)

Subscript/Superscript

ads	adsorbed
boil	liquid at the boiling point
crit	critical point
i	component i
π, i	component i at spreading pressure π
sat	saturated
trip	triple point
$\bullet$	pure component (in the case of binary mixture)

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