

# INVESTIGATION OF ADSORPTION EQUILIBRIA OF PURE GASES (CO, CO<sub>2</sub>, CH<sub>4</sub>) AND THEIR BINARY AND TERNARY MIXTURES AT T = 293 K FOR PRESSURES $p \leq 1,1$ MPa ON INDUSTRIAL ACTIVATED CARBON (ACAL)

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## Abstract

Adsorption equilibria of pure carbon monoxide (CO), carbon dioxide (CO<sub>2</sub>) and methane (CH<sub>4</sub>) and their binary and ternary mixtures on industrial activated carbon (ACAL) have been investigated at 293 K for pressures up to 1,1 MPa. Volumetric-gravimetric experiments have been performed using a magnetic suspension balance (Rubotherm GmbH, Bochum, Germany, 3-site-type), allowing simultaneous adsorption and density measurements of the sorptive gas. For binary sorptive gas mixture measurements of this type no additional concentration measurements for the gas phase and/or for the adsorbed phase are required. However, the ternary gas mixtures adsorption equilibria require additional measurements of the sorptive gas concentrations in the equilibrium state considered by a gas chromatograph. Two thermodynamic models (the two-site Langmuir adsorption isotherm of fractal dimension (2LAI) and the Ideal Adsorbed Solution Theory (IAST)) have been used to correlate/fit the experimental adsorption equilibria data of the pure gases (CO, CO<sub>2</sub>, CH<sub>4</sub>) and their mixtures. The parameters determined for pure gas adsorption isotherms from the two model equations were used to predict coadsorption equilibria data. The relative mean deviation ( $f_m$ ) and the relative dispersion ( $\sigma$ ) of calculated and measured amounts of gas adsorbed have been determined. From these ( $f_m$ ,  $\sigma$ )-data it clearly can be seen that coadsorption equilibria can not be predicted / calculated accurately from pure adsorption isotherm data by the model equations used. The experimental equipment, procedure of measurement and the resulting data are presented and discussed.

## Introduction

In our days, most of the gas separation processes utilized for technical applications like air separation (N<sub>2</sub>, O<sub>2</sub>), cleaning of natural gas (CH<sub>4</sub>, H<sub>2</sub>S) or upgrading low energy flue gas (CH<sub>4</sub>, N<sub>2</sub>, CO<sub>2</sub>) etc., are based on adsorption phenomena of gases on highly porous solids like zeolites and activated carbons. Very often equilibrium effects are used, i. e. the selectivity of the sorbent for one component against the other in adsorption equilibrium loads at given pressure, temperature and concentrations of the sorptive gas mixture (1, 11). Hence, for proper design of gas separation processes by selective adsorption binary or even ternary coadsorption equilibrium data of the respective components should be known. There are thermodynamic models available in literature (f. e. the Ideal Adsorbed Solution Theory (IAST), Real Adsorbed Solution Theory (RAST) etc.) to predict the coadsorption equilibrium data from pure component isotherms. Due to the complexity of possible interactions of adsorbed molecules and the sorbent's surface (9), it is still not possible to calculate accurate coadsorption equilibria data ab initio or even from pure component adsorption data and (normally scarce) information available for the sorbent material. The accuracy of these predictions is

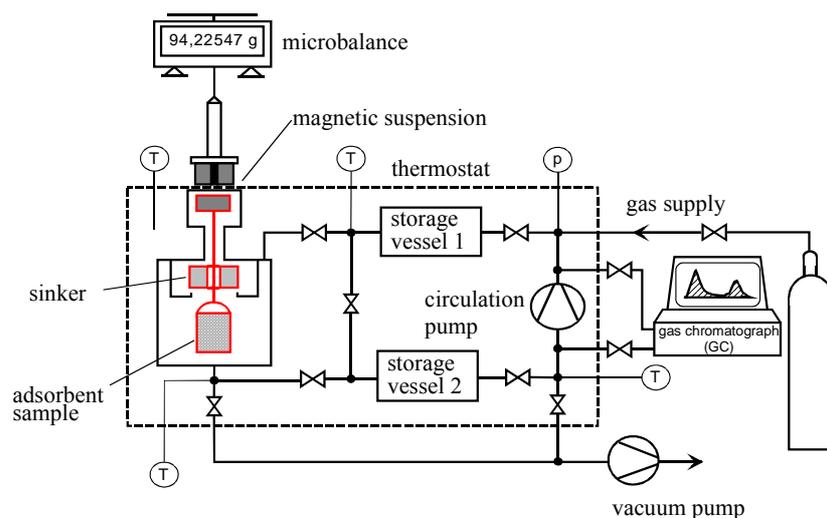
usually not sufficient to enable the economical design of adsorption separation processes. Therefore, experimental measurements of gas adsorption equilibrium data are still necessary, in particular for multicomponent systems (coadsorption equilibrium data).

As an example gas adsorption equilibria of pure gases (CO, CO<sub>2</sub>, CH<sub>4</sub>) and their 3 binary mixtures (CO/CH<sub>4</sub>, CO<sub>2</sub>/CH<sub>4</sub>, CO<sub>2</sub>/CO:  $y_1 = 0.1; 0.35; 0.65; 0.9$ ) and their ternary mixture (CO<sub>2</sub>/CH<sub>4</sub>/CO) on industrial activated carbon (ACAL) have been measured at 293 K for pressures up to 1,1 MPa by means of a magnetic suspension balance (Rubotherm GmbH, Bochum, Germany, 3-site-type). This balance allows simultaneous gravimetric measurements of the total amount of gas adsorbed and the density of the adsorptive (gas) phase. Isotherms of pure gases were measured gravimetrically. Coadsorption data of the binary mixtures were obtained by the volumetric-gravimetric method (5). For binary gas mixtures with “fairly different” molar masses ( $M_1, M_2$ ) of the gases these kind of measurements allow the direct determination of the masses of each component in the adsorbed phase without analysis of the gas phase so that no gas chromatograph and/or a mass spectrometer is needed. However, ternary gas mixtures adsorption equilibria require additional measurements of the sorptive gas concentrations in equilibrium (5). More information of the instrument and the measurements is given below. The resulting adsorption equilibria data have been correlated by means of the two model equations mentioned above. The correlation parameters corresponding to pure gas adsorption isotherms were used to calculate/predict coadsorption equilibrium data. The results of coadsorption equilibria measurements and correlations are presented and discussed to a certain extend.

## Experimental

The experimental setup for the simultaneous volumetric-gravimetric measurements of binary gas coadsorption equilibria and the density of the gas phase, designed and built in the laboratory of the Institute of Fluid- and Thermodynamics (IFT PB A 0126), University of Siegen, is sketched in Figure 1. For the determination of adsorption equilibria of pure gases (CO, CO<sub>2</sub> and CH<sub>4</sub>) the gravimetric part of the apparatus has been used. The coadsorption equilibria of binary mixed gases have been determined by the volumetric-gravimetric method (5, Chap. 4). In the case of binary mixtures with isomeric components ( $M_1 = M_2$ ) such as (N<sub>2</sub>, CO) and of ternary gas mixtures additional measurements of the sorptive gas concentration in the equilibrium state considered are necessary. For detailed information about the method refer to literature (5).

The equipment mainly consists on a microbalance (Mettler, Germany), a magnetic suspension with three working positions (Rubotherm GmbH, Bochum, Germany), an adsorption vessel (volume:  $V_{AV} \approx 0.4 \text{ dm}^3$ ), two storage vessels (*Whitey* Highland Heights, OH, USA, volume:  $V_{SV1} \approx 0.5 \text{ dm}^3$ ,  $V_{SV2} \approx 1 \text{ dm}^3$ ), a pump for gas circulation (ASF Thomas, Germany), a vacuum pump (Balzers-Pfeiffer, Germany) and a system for gas supply (GCE-DruVa, Germany). The adsorption vessel includes a basket with the adsorbent sample ( $m^S \approx 30 \text{ g}$ ) which is linked to a microbalance outside of the vessel by a magnetic suspension. The decoupling of the microbalance from the sorptive gas allows measurements in corrosive gas atmospheres like (NH<sub>3</sub>, SO<sub>2</sub>, H<sub>2</sub>S, NO<sub>x</sub> etc.). According to Archimedes' principle, the weight change of the sinker (titanium) leads directly to the density of the remaining gas in the adsorption vessel. No thermal equation of states (EOS) has to be applied for the calculation of the density of the sorptive gas and thus the implied uncertainties can be avoided. Also, the experimentally determined density is used for buoyancy corrections of the measured adsorption data (5). For detailed information about the functional principle of the magnetic suspension refer to literature (3-5).



**Figure 1.** Instrument for simultaneous volumetric-gravimetric measurements of binary gas coadsorption equilibria on porous solids and density measurements of the gas phase including a magnetic suspension balance (Rubotherm GmbH, Bochum, Germany).

After activation/regeneration of the sample (heating up to  $T = 423 - 433$  K for at least four to five hours by permanent evacuation), the gas pressure in the adsorption vessel for gravimetric measurements of the adsorption of pure gases has been stepwise increased by supplying the pure component sorptive gas. After adsorption equilibrium in the installation has been realized, i. e. the gas pressure ( $p$ ) within the system and the signal from the microbalance ( $\Delta m$ ) both remain “constant” (in practice “technical equilibrium” (5, 6)), the data ( $\Delta m$ ,  $p$  and  $T$ ) have been recorded. The procedure (increasing of the pressure in the adsorption vessel and approach to the next adsorption equilibrium state) has been repeated until the maximum pressure was achieved. It takes approximately 2 hours for  $\text{CH}_4$ , 2 - 2,5 hours for  $\text{CO}$  and 3 - 4 hours for  $\text{CO}_2$  on activated carbon (ACAL) till “technical equilibrium” is realized.

In case of binary and ternary gas mixture measurements ( $\text{CO}/\text{CH}_4$ ,  $\text{CO}_2/\text{CH}_4$ ,  $\text{CO}_2/\text{CO}$  and  $\text{CO}_2/\text{CH}_4/\text{CO}$ ) the two storage vessels were used to prepare the mixtures inside the apparatus and the circulation pump was switched on to avoid concentration differences. The purpose of the experiments was to obtain a specified concentration of the gas mixture in the adsorption chamber at a given pressure after the adsorption process was finished and adsorption equilibrium has been realized. Therefore a prediction with the two-site Langmuir adsorption isotherm of fractal dimension (2LAI) (5, 7) based on the pure gases adsorption equilibrium data was realized primarily. Adsorption equilibrium was reached, when the gas pressure ( $p$ ) within the system and the signal from the microbalance ( $\Delta m$ ) remained “constant” after stopping the gas circulator. Equilibration times were about (3 - 4) hours for the ( $\text{CO}/\text{CH}_4$ ) system, (4 - 5) hours for ( $\text{CO}_2/\text{CH}_4$ ) and ( $\text{CO}_2/\text{CO}$ ) systems and (4 - 6) hours for ( $\text{CO}_2/\text{CH}_4/\text{CO}$ ) system. After this, the instrument was evacuated and the adsorbent sample was regenerated in order to be prepared for the next measurement.

For detailed information on those measurements and the procedure of calculating the adsorption equilibrium of pure and mixed gases, based on the measured equilibria data, the reader is referred to literature (4).

## Data Correlation and Prediction of binary and ternary mixture adsorption

The experimental data of adsorption equilibria of the pure gases (CO, CO<sub>2</sub>, CH<sub>4</sub>) and their binary and ternary mixtures on industrial activated carbon (ACAL) were correlated with two adsorption isotherms:

- the two-site Langmuir adsorption isotherm of fractal dimension (2LAI) (5, 7),
- the Ideal Adsorbed Solution Theory (IAST) (8).

In this paper the form of the two site Langmuir adsorption isotherm of fractal dimension (2LAI) is cited as an example. The 2LAI reads for pure gas adsorption:

$$n(p, T) = n_{\infty}^P(T) \cdot \alpha^P \cdot \frac{(b^P(T) \cdot p)^{\alpha^P}}{1 + (b^P(T) \cdot p)^{\alpha^P}} + n_{\infty}^S(T) \cdot \alpha^S \cdot \frac{(b^S(T) \cdot p)^{\alpha^S}}{1 + (b^S(T) \cdot p)^{\alpha^S}} \quad (1)$$

The indices “P” and “S” refer to “primary” and “secondary” adsorption or pore systems, visualizing meso- and micropores (5, 7). The exponents  $\alpha^P(T)$  and  $\alpha^S(T)$  are characteristic exponents related to the diameter of molecules adsorbed and the fractal dimension of the sorbent material.

Using the parameters determined for pure gas adsorption isotherms by the two model equations the coadsorption equilibria data were calculated/predicted. Thus, the amount adsorbed of component  $i$  ( $n_i$ ) of a mixture with  $N$ -components is given by the 2LAI as:

$$n_i(p, T) = n_{\infty,i}^P(T) \cdot \alpha_i^P \cdot \frac{(b_i^P(T) \cdot p_i)^{\alpha_i^P}}{1 + \sum_{k=1}^N (b_k^P(T) \cdot p_k)^{\alpha_k^P}} + n_{\infty,i}^S(T) \cdot \alpha_i^S \cdot \frac{(b_i^S(T) \cdot p_i)^{\alpha_i^S}}{1 + \sum_{k=1}^N (b_k^S(T) \cdot p_k)^{\alpha_k^S}}, \quad i = 1, \dots, N \quad (2)$$

To check the quality of either the correlation of coadsorption data ( $n_{\text{CORR}}$ ) by the two equations used or the prediction of coadsorption data ( $n_{\text{PRE}}$ ) from pure component adsorption data by the same equations, the relative mean deviation ( $f_m$ ) between correlated or predicted ( $n_{\text{CORR}}/n_{\text{PRE}}$ ) and measured amounts adsorbed ( $n_{\text{EXP}}$ ) of the pure gases and their mixtures and the relative dispersion ( $\sigma$ ) of these data

$$f_m = \frac{1}{M} \sum_{i=1}^M \left( \frac{|n - n_{\text{EXP}}|}{n_{\text{EXP}}} \right)_i \quad \text{and} \quad \sigma^2 = \frac{1}{M} \sum_{i=1}^M \left( \frac{(n - n_{\text{EXP}})^2}{n_{\text{EXP}}^2} \right)_i \quad (3)$$

$n = (n_{\text{CORR}} \text{ OR } n_{\text{PRE}})$  have been calculated. Here  $M$  is the total number of measurements. Numerical data are given below.

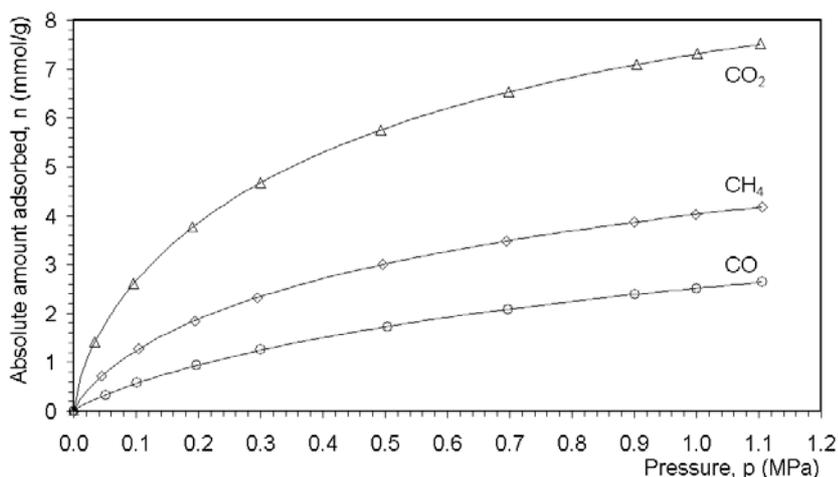
### Measured Data

Gravimetric, gravimetric-volumetric and gravimetric-volumetric-chromatographic measurements were performed to determine pure gas adsorption equilibria of (CO, CO<sub>2</sub>, CH<sub>4</sub>) and coadsorption equilibria of their binary and ternary gas mixtures on industrial activated carbon (ACAL) at  $T = 293$  K for pressures up to 1.1 MPa (2). From helium adsorption isotherm measurement at 293 K on

the activated carbon (ACAL) the so-called specific helium volume has been calculated ( $V_{\text{He}} = 0.4536 \text{ cm}^3/\text{g}$ ). The helium volume ( $V_{\text{He}}$ ) is for the approximation of the void volume of a sorbent material ( $V^s$ ,  $V^s \approx V_{\text{He}}$ ) most often used today (5). Pure sorptive gases were delivered by Messer Griesheim, Düsseldorf, with purities<sup>1)</sup> 5.5 for  $\text{CH}_4$ , 3.7 for  $\text{CO}$  and 4.5 for  $\text{CO}_2$ . Gas mixtures were produced in the experimental installation itself, Fig. 1.

The mean relative uncertainties of the resulting quantities have been determined by a standard error calculation. For gravimetric pure gas adsorption measurements the relative uncertainties of the specific excess amounts adsorbed are  $(\Delta n_{\text{ex}}/n_{\text{ex}}) < 0.1\%$ . They are smaller than the size of the graphical symbols used in the Figure 2. For volumetric-gravimetric measurements of binary coadsorption equilibria the average uncertainties are:  $(\Delta n_{\text{tot,ex}}/n_{\text{tot,ex}}) \approx 6\%$ ,  $(\Delta n_{\text{CO,ex}}/n_{\text{CO,ex}}) \approx 19\%$  and  $(\Delta n_{\text{CH}_4,\text{ex}}/n_{\text{CH}_4,\text{ex}}) \approx 10\%$  for the  $\text{CO}/\text{CH}_4$  gas mixture;  $(\Delta n_{\text{tot,ex}}/n_{\text{tot,ex}}) \approx 2\%$ ,  $(\Delta n_{\text{CO}_2,\text{ex}}/n_{\text{CO}_2,\text{ex}}) \approx 2\%$  and  $(\Delta n_{\text{CH}_4,\text{ex}}/n_{\text{CH}_4,\text{ex}}) \approx 13\%$  for the  $\text{CO}_2/\text{CH}_4$  gas mixture;  $(\Delta n_{\text{tot,ex}}/n_{\text{tot,ex}}) \approx 5\%$ ,  $(\Delta n_{\text{CO}_2,\text{ex}}/n_{\text{CO}_2,\text{ex}}) \approx 4\%$  and  $(\Delta n_{\text{CO,ex}}/n_{\text{CO,ex}}) \approx 90\%$  for the  $\text{CO}_2/\text{CO}$  gas mixture. For volumetric-gravimetric-calorimetric measurements of ternary coadsorption equilibria ( $\text{CO}_2/\text{CH}_4/\text{CO}$ ) the average uncertainties are:  $(\Delta n_{\text{tot,ex}}/n_{\text{tot,ex}}) \approx 4\%$ ,  $(\Delta n_{\text{CO}_2,\text{ex}}/n_{\text{CO}_2,\text{ex}}) \approx 2\%$ ,  $(\Delta n_{\text{CH}_4,\text{ex}}/n_{\text{CH}_4,\text{ex}}) \approx 24\%$  and  $(\Delta n_{\text{CO,ex}}/n_{\text{CO,ex}}) \approx 33\%$ .

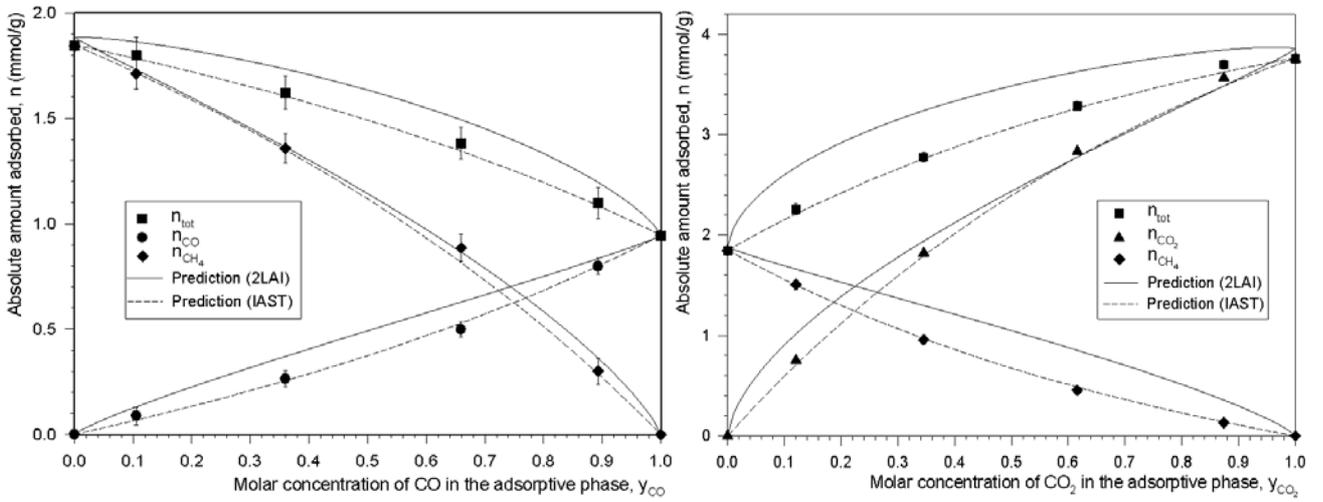
In this paper five examples of isotherms of adsorption equilibria of pure gases, binary and ternary gas mixtures are presented. Detailed information is given in the literature (2). In the next Figure 2 the absolute amount adsorbed of pure gases per unit mass of sorbent ( $n$ ) in (mmol/g) are given as function of the sorptive gas pressure ( $p$ ) in (MPa) at 293 K. The binary and ternary coadsorption data are in Figures (3 - 5) presented. All data are correlated by the two models (2LAI, IAST). The yielding correlation parameters for pure gas adsorption isotherms were used to calculate/predict coadsorption equilibrium data. The statistical uncertainties ( $f_m$ ,  $\sigma$ ) in % between correlated/predicted by 2LAI and IAST and measured adsorption data are given in the Tables below.



	2LAI	IAST
$n_{\text{CO}}$	$f_m = 0.521 \%$ $\sigma = 0.647 \%$	0.067 0.072
$n_{\text{CO}_2}$	0.124 0.209	0.203 0.228
$n_{\text{CH}_4}$	0.083 0.098	0.125 0.139

**Figure 2.** Adsorption equilibria of pure gases  $\text{CO}_2$  ( $\Delta$ ),  $\text{CH}_4$  ( $\diamond$ ) and  $\text{CO}$  ( $\circ$ ) at 293 K on activated carbon. (—) correlation with 2LAI (Eq. (1)). Experimental uncertainties are smaller than the size of the used graphical symbols. Statistical uncertainties ( $f_m$ ,  $\sigma$ ) in % between *correlated* and *measured* pure gases adsorption data by 2LAI and IAST are presented in the Table 1 (right).

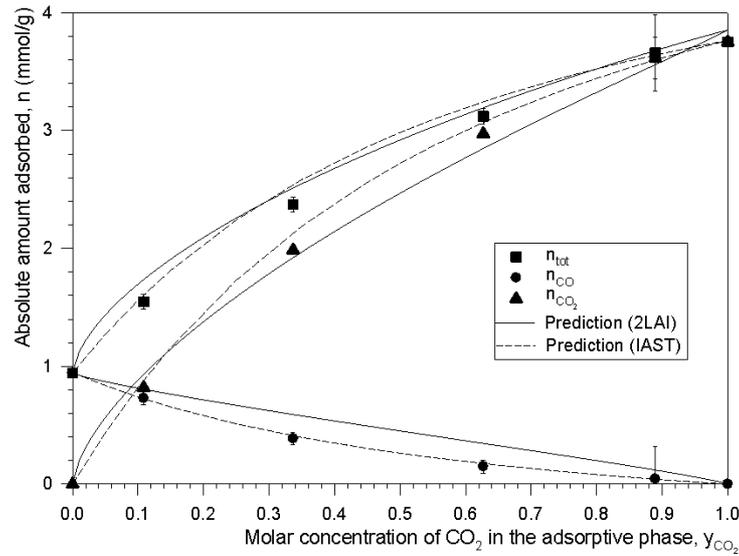
<sup>1)</sup> Example: 3.7  $\equiv$  99.97 %, 4.5  $\equiv$  99.995 %, 5.5  $\equiv$  99.9995 %.



**Figure 3.** Adsorption equilibria of binary gas mixtures CO/CH<sub>4</sub> (left) and CO<sub>2</sub>/CH<sub>4</sub> (right) (total amount adsorbed  $n_{\text{tot}}$  (■), partial amount of CH<sub>4</sub> (◆), partial amount of CO (●), partial amount of CO<sub>2</sub> (▲)) at 293 K and 0.2 MPa on ACAL. Experimental uncertainties are presented. Statistical uncertainties ( $f_m$ ,  $\sigma$ ) of coadsorption data *predicted* from pure adsorption data by 2LAI (—) (Eqs. (1, 2)) and IAS-Theory (....) are given in Table 2 (A and B).

**Table 2.** Statistical uncertainties ( $f_m$ ,  $\sigma$ ) in % of coadsorption data CO/CH<sub>4</sub> (A) and CO<sub>2</sub>/CH<sub>4</sub> (B) (total and partial loads) on ACAL at two pressures (0.2 MPa, 1.0 MPa) and 293 K *predicted* from pure adsorption data by two isotherms (2LAI, IAST).

<b>A</b>		$x_{\text{CO}}$ (adsorbed phase)	$n_{\text{CH}_4}$ (partial load)	$n_{\text{CO}}$ (partial load)	$n_{\text{tot}} =$ $n_{\text{CO}} + n_{\text{CH}_4}$
0.2 MPa	2LAI	$f_m = 24.5 \%$ $\sigma = 29.0 \%$	6.11 10.5	30.6 35.2	6.91 7.22
	IAST	8.46 11.6	3.16 4.39	7.86 11.5	1.39 1.58
1.0 MPa	2LAI	52.0 76.3	5.01 7.31	62.1 83.3	8.21 8.75
	IAST	3.55 4.07	3.33 3.64	3.82 4.51	2.67 2.86
<b>B</b>		$x_{\text{CO}_2}$ (adsorbed phase)	$n_{\text{CO}_2}$ (partial load)	$n_{\text{CH}_4}$ (partial load)	$n_{\text{tot}} =$ $n_{\text{CO}_2} + n_{\text{CH}_4}$
0.2 MPa	2LAI	$f_m = 9.94 \%$ $\sigma = 10.4 \%$	11.5 17.9	76.3 98.0	12.4 13.7
	IAST	1.49 1.97	2.78 3.25	3.35 4.34	1.31 1.50
1.0 MPa	2LAI	12.8 13.2	13.2 18.1	144 196	9.70 11.8
	IAST	2.09 2.61	6.25 7.00	21.5 26.3	6.08 6.48

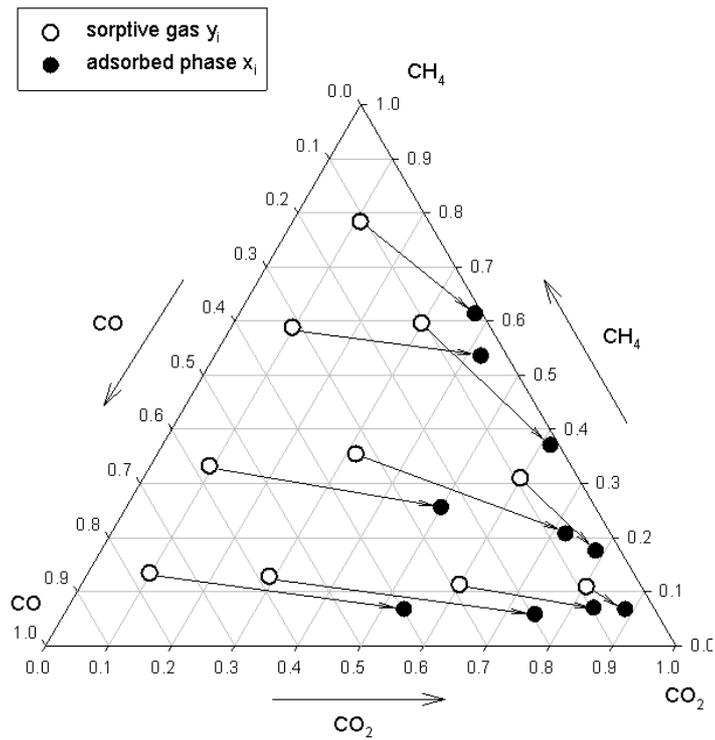


**Figure 4.** Adsorption equilibria of a binary gas mixture CO<sub>2</sub>/CO (total amount adsorbed  $n_{\text{tot}}$  (■), partial amount of CO<sub>2</sub> (▲), partial amount of CO (●)) at 293 K and 0.2 MPa on ACAL. Experimental uncertainties are presented. Statistical uncertainties ( $f_m$ ,  $\sigma$ ) of coadsorption data *predicted* from pure adsorption data by 2LAI (—) (Eqs. (1, 2)) and IAS-Theory (....) are given in Table 3.

**Table 3.** Statistical uncertainties ( $f_m$ ,  $\sigma$ ) in % of coadsorption data CO<sub>2</sub>/CO on activated carbon at two pressures (0.2 MPa, 1.0 MPa) and 293 K *predicted* from pure adsorption data by two isotherms (2LAI, IAST).

		$x_{\text{CO}_2}$ (adsorbed phase)	$n_{\text{CO}_2}$ (partial load)	$n_{\text{CO}}$ (partial load)	$n_{\text{tot}} =$ $n_{\text{CO}_2} + n_{\text{CO}}$
0.2 MPa	2LAI	$f_m = 6.21 \%$	5.67	101	3.43
		$\sigma = 6.57 \%$	7.41	130	3.80
	IAST	1.16	4.68	7.14	3.82
		1.91	5.55	9.21	4.49
1.0 MPa	2LAI	6.76	7.20	219	2.45
		7.99	7.99	292	3.20
	IAST	0.51	10.1	15.6	9.95
		0.69	11.1	19.7	10.8

As clearly can be recognized from the ( $f_m / \sigma$ )-data given in Table 2 (A and B) and Table 3, the total amounts adsorbed ( $n_{\text{tot}}$ ) can be predicted from pure adsorption isotherms with all models used by good approximation (1 - 10 %), best by the classical IAS-Theory. The amount of the component least adsorbed (CO in CO/CH<sub>4</sub>, CH<sub>4</sub> in CO<sub>2</sub>/CH<sub>4</sub>, CO in CO<sub>2</sub>/CO) cannot be predicted accurately from pure adsorption isotherms by the model equation 2LAI, but with adequate accuracy by the classical IAS-Theory.



**Figure 5.** Adsorption equilibria of a ternary gas mixture  $\text{CO}_2/\text{CH}_4/\text{CO}$  (molar concentrations of the sorptive gases  $y$  (○), molar concentrations of the adsorbed phases  $x$  (●) at 293 K and 0.2 MPa on activated carbon (ACAL).

**Table 4.** Coadsorption data of ternary gas mixture ( $\text{CO}_2/\text{CH}_4/\text{CO}$ , total and partial loads) on ACAL at  $T = 293 \text{ K}$  and  $0.2 \text{ MPa}$ .

$y_{\text{CO}_2}$ (-)	$y_{\text{CH}_4}$ (-)	$x_{\text{CO}_2}$ (-)	$x_{\text{CH}_4}$ (-)	$n_{\text{CO}_2}$ (mmol/g)	$n_{\text{CH}_4}$ (mmol/g)	$n_{\text{tot}} =$ $n_{\text{CO}_2} + n_{\text{CH}_4} + n_{\text{CO}}$
0.099	0.133	0.096	0.122	0.847	0.106	1.580
0.108	0.783	0.107	0.790	0.801	1.307	2.132
0.804	0.108	0.820	0.094	3.184	0.239	3.589
0.095	0.332	0.091	0.327	0.858	0.438	1.714
0.098	0.588	0.095	0.595	0.846	1.069	1.997
0.292	0.128	0.296	0.121	1.721	0.132	2.300
0.299	0.596	0.308	0.587	1.635	0.981	2.651
0.599	0.309	0.618	0.284	2.577	0.573	3.279
0.601	0.112	0.617	0.103	2.632	0.218	3.151
0.316	0.353	0.323	0.341	1.827	0.522	2.530

**Table 5.** Coadsorption data of ternary gas mixture (CO<sub>2</sub>/CH<sub>4</sub>/CO, total and partial loads) on ACAL at T = 293 K and 1.0 MPa.

y <sub>CO2</sub> (-)	y <sub>CH4</sub> (-)	x <sub>CO2</sub> (-)	x <sub>CH4</sub> (-)	n <sub>CO2</sub> (mmol/g)	n <sub>CH4</sub> (mmol/g)	n <sub>tot</sub> = n <sub>CO2</sub> + n <sub>CH4</sub> + n <sub>CO</sub>
0.096	0.122	0.523	0.006	1.747	0.020	3.340
0.107	0.790	0.353	0.628	1.485	2.642	4.204
0.820	0.094	0.833	0.092	5.844	0.643	7.019
0.091	0.327	0.550	0.139	1.821	0.460	3.309
0.095	0.595	0.449	0.537	1.749	2.092	3.898
0.296	0.121	0.719	0.007	3.420	0.031	4.755
0.308	0.587	0.550	0.444	3.027	2.441	5.500
0.618	0.284	0.714	0.248	4.795	1.665	6.717
0.617	0.103	0.768	0.074	4.911	0.471	6.396
0.323	0.341	0.721	0.216	3.669	1.100	5.090

**Table 6.** Statistical uncertainties (f<sub>m</sub>, σ) in % of coadsorption data (CO<sub>2</sub>/CH<sub>4</sub>/CO) on activated carbon at T = 293 K and 0.2 MPa *predicted* from pure adsorption data by two isotherms (2LAI, IAST).

	x <sub>CO2</sub> (adsorbed phase)	x <sub>CH4</sub> (adsorbed phase)	n <sub>CO2</sub> (partial load)	n <sub>CH4</sub> (partial load)	n <sub>tot</sub> = n <sub>CO2</sub> + n <sub>CH4</sub> + n <sub>CO</sub>
2LAI	f <sub>m</sub> = 156 %	23.8	4.65	80.1	16.1
	σ = 200 %	30.0	6.58	118	17.9
IAST	161	35.0	10.3	46.8	2.73
	199	39.8	12.7	71.4	3.47

**Table 7.** Statistical uncertainties (f<sub>m</sub>, σ) in % of coadsorption data (CO<sub>2</sub>/CH<sub>4</sub>/CO) on activated carbon at T = 293 K and 1.0 MPa *predicted* from pure adsorption data by two isotherms (2LAI, IAST).

	x <sub>CO2</sub> (adsorbed phase)	x <sub>CH4</sub> (adsorbed phase)	n <sub>CO2</sub> (partial load)	n <sub>CH4</sub> (partial load)	n <sub>tot</sub> = n <sub>CO2</sub> + n <sub>CH4</sub> + n <sub>CO</sub>
2LAI	f <sub>m</sub> = 14.2 %	574	9.03	705	15.4
	σ = 18.0 %	1250	11.1	1540	20.0
IAST	16.5	424	17.4	510	11.0
	18.6	924	19.2	1120	13.8

From the (f<sub>m</sub> / σ)-data given in Tables 6 and 7 can be recognized, the total amounts adsorbed (n<sub>tot</sub>) and the amount of the strongest adsorbed component (CO<sub>2</sub>, n<sub>CO2</sub>) can be predicted from pure adsorption isotherms with all models used by good approximation (3 - 20 %). The amounts of the components less adsorbed (n<sub>CH4</sub> and n<sub>CO</sub>) cannot be predicted accurately from pure adsorption

isotherms by the model equations. Hence in mixture gas adsorption there is still a need to develop more effective mixture isotherms as well as to provide more accurate and reliable experimental data of coadsorption isotherms.

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