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APPENDICES

Appendix A Adsorbent Physical Characterization

The technical specification of the CSAC that was certified by Carbokarn Co., Ltd. is summarized in Table A1.

Table A1 Physical characteristic properties of investigated adsorbent

Physical Characterization	Adsorbent Specification
Apparent Density (g/cm^3)	> 0.48
Moisture Content (%w/w)	< 8.0
Ash Content (%w/w)	< 3.5
pH	9-11
Iodine Number (mg/g)	> 1,100
Hardness Number (%)	> 98.0

Appendix B Modeling Adsorption Isotherm

Binary gas adsorption equilibrium was measured using a volumetric-chromatographic apparatus. The experiment was taken place at room temperature, which was controlled by air-conditioning and atmospheric pressure. The gas phase composition at equilibrium was analyzed by a gas chromatograph (GC). The binary adsorption equilibrium of methane and carbon dioxide were measured on the untreated CSAC for different gas phase compositions.

The predicted co-adsorption isotherm was performed by iteration method for 0 and 1 mole fraction of carbon dioxide with the fits of the Langmuir equation for the single component data in Eq. (B.1) (Rios *et al.*, 2012). The assumptions of this prediction are; the surface containing the adsorbing sites is perfectly flat plane with no corrugations; all sites are equivalent; each site can hold only one molecule of gas; and there no interactions between adsorbates molecules on adjacent sites.

$$q_i = \frac{q_{max} \cdot b_i \cdot P_i}{1 + b_i \cdot P_i} \tag{B.1}$$

where q_i is excess amount adsorbed of the component i (mol/kg), $q_{max,i}$ is monolayer capacity of component i (mol/kg), b_i is Langmuir parameter of the component i (MPa^{-1}), and P_i is partial pressure of the component i (MPa).

To obtain the model and the accuracy of the predictions in relation to the experimental results of binary adsorption, the average relative error ($\delta_{q,i}$), as expressed in Eq. (B.2), was calculated and used as a parameter of analysis.

$$\delta_{q,i} = \frac{100}{N} \sum_{i=1}^{N} \left(\left(\frac{q_i - q_{pre,i}}{q_i} \right)^2 \right)^{1/2} \tag{B.2}$$

where $\delta_{q,i}$ is an average relative error, N is a number of isotherm points, and $q_{pre,i}$ is predicted amount adsorbed of the component i.

Table B1 Parameters of Langmuir fit of carbon dioxide and methane isotherms at room temperature and atmospheric pressure on the untreated CSAC

Isotherm	Langmuir Parameter		
	q_{max} (mol/kg)	b_i (MPa^{-1})	δ_q
CH_4	14.184	0.76	12.34
CO_2	36.656	0.89	51.77

As expected, higher q_{max} and b_i are obtained for carbon dioxide in comparison with methane, as shown in Table B1. The parameter b_i indicates how strongly an adsorbate molecule is attracted onto an adsorbent surface (Do, 1998). Experimental and predicted adsorption data of carbon dioxide and methane mixtures at room temperature and atmospheric pressure for different molar compositions is shown in Figure B1.

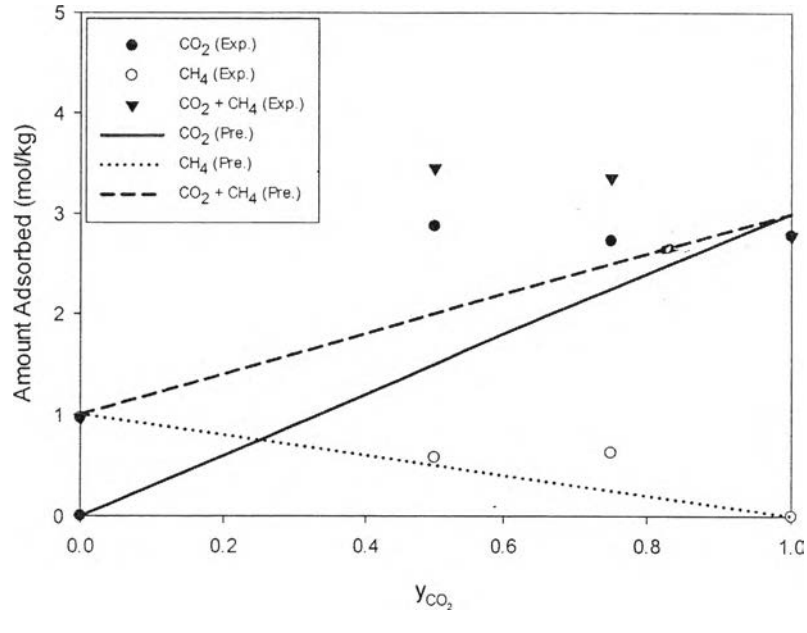

Figure B1 Binary adsorption isotherms for carbon dioxide and methane mixtures at room temperature and atmospheric pressure on the untreated CSAC.

From Figure B1, the total adsorbed amount increases along with carbon dioxide composition, while the amount of methane adsorbed decreases, indicating competition for adsorption sites and preferential adsorption of carbon dioxide over methane. This result agrees with the higher value of the parameter b for carbon dioxide in relation to methane. Moreover, carbon dioxide presents a higher polarizability, which may enhance attractive forces with the surface and a permanent quadrupole, leading to stronger interactions with the solid surface (Rios *et al.*, 2012). The isotherms from the prediction show linear relationship between the increase in the carbon dioxide composition and the increase in the total amount of adsorbed gas. Carbon dioxide is present higher adsorbed amounts than that of methane after increasing the carbon dioxide concentration more than 25%. Comparison between the predicted data with the experiment data shows that the experimental data gives the higher adsorption capacity than that of the predicted data because the predicted data was calculated from the pure component, which might not be accurate for the gas mixture. Moreover, the relative error of carbon dioxide is more than 50%, which is higher than that of methane about 4 times. Therefore, this fit might not be suitable for the prediction of carbon dioxide adsorption.

To further investigate this, a new fit of the extend Langmuir; shown in Eq. (B.3) was performed considering mixing point at 50% carbon dioxide concentration. The basic assumptions of this model consider the case when there are two distinct adsorbates present in the system and there are no interactions between adsorbate molecules on adjacent sites. The new parameters of $q_{max,i}$ and b_i are shown in Table B2.

$$q_i = \frac{q_{max} \cdot b_i \cdot P_i}{1 + \sum_{i=1}^{n} b_i \cdot P_i} \tag{B.3}$$

Table B2 Parameters of the extend Langmuir fit of carbon dioxide and methane isotherms at room temperature and atmospheric pressure on the untreated CSAC

Isotherm	Langmuir Parameter		
	q_{max} (mol/kg)	b_i (MPa^{-1})	δ_q
CH_4	12.176	1.03	10.54
CO_2	34.833	1.75	9.36

Comparison between the relative errors of methane in Table B2 shows that the error decreases from 12.34 to 10.54% similar to carbon dioxide decreases from 51.77 to 9.36%, which is acceptable in this range (Goetz *et al.*, 2006; Harlick and Tezel, 2003). For the new value of $q_{max,i}$, which was predicted by using the mixture components of methane and carbon dioxide, is lower than the previous one. Moreover, it can be observed in Tables B1 and B2 that the higher values for q_{max} are obtained for carbon dioxide because of a strong adsorption preference in the mixture.

A comparison between the experimental data and predicted data of the extend Langmuir isotherms for the binary equilibrium at atmospheric pressure is shown in Figure B2. It shows the carbon dioxide adsorption increases along with carbon dioxide composition before constant at 0.7 of carbon dioxide composition. Carbon dioxide might be saturated onto the untreated CSAC. The main reason for this behavior is the significantly higher critical temperature of carbon dioxide (304 K) in comparison with methane (190 K) (Tagliabue *et al.*, 2009). Carbon dioxide is more likely to behave as a condensable steam than as a supercritical gas, becoming less volatile and being easy saturated. Comparison between the predicted data with the experiment data shows that the experimental data and the predicted data agree very well. Therefore, this model may suit for prediction the amount of methane and carbon dioxide adsorption in the binary system than the previous model.

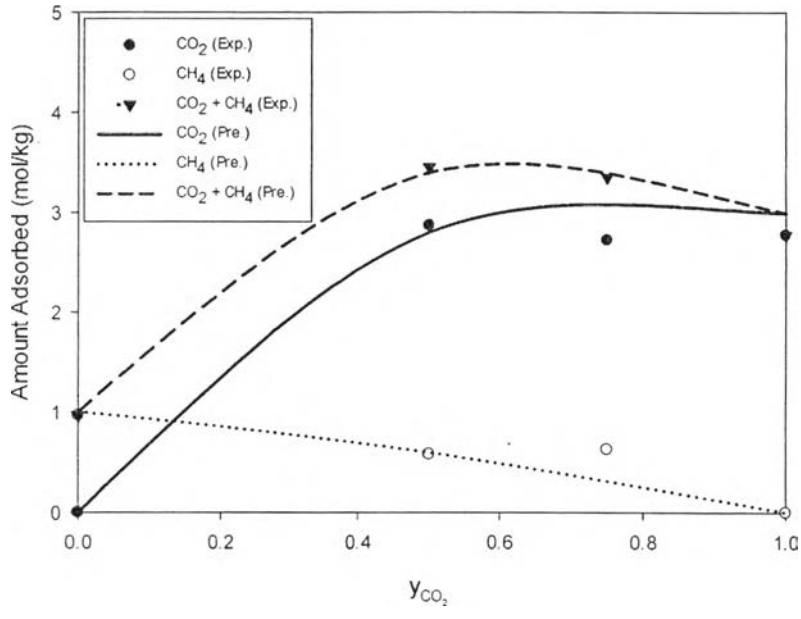

Figure B2 New binary adsorption isotherms for carbon dioxide and methane mixtures at room temperature and atmospheric pressure on the untreated CSAC.

The iteration of the Langmuir parameters for CO_2, CH_4, and CO_2-CH_4 mixture in the adsorption system is shown in Tables B3-B6, which were calculated by using Eq. (B.1) and (B.2).

Table B3 Iteration of CO_2

P_i	q_{max}	b_i	$q_{predict}$	δ_q	$*q_{max}$	$*b_i$
0.1	1	0.01	0.000999001	0.999640116	1.000999001	0.02
0.1	1.000999001	0.02	0.001998002	0.999280233	1.002997003	0.03
0.1	1.002997003	0.03	0.002999991	0.998919273	1.005996994	0.04
0.1	1.005996994	0.04	0.004007956	0.99855616	1.01000495	0.05
0.1	1.01000495	0.05	0.0050249	0.998189812	1.01502985	0.06
0.1	1.01502985	0.06	0.006053856	0.997819138	1.021083706	0.07
0.1	1.021083706	0.07	0.007097901	0.997443027	1.028181607	0.08
0.1	1.028181607	0.08	0.008160171	0.997060351	1.036341779	0.09
0.1	1.036341779	0.09	0.009243881	0.996669952	1.04558566	0.1
0.1	1.04558566	0.1	0.010352333	0.996270639	1.055937993	0.11
.	.	.	.	.	.	.
.	.	.	.	.	.	.
.	.	.	.	.	.	.
0.1	26.98220563	0.85	2.113813344	0.238512431	29.09601897	0.86
0.1	29.09601897	0.86	2.304104633	0.169961226	31.4001236	0.87
0.1	31.4001236	0.87	2.513165367	0.09464845	33.91328897	0.88
0.1	33.91328897	0.88	2.742986608	0.011856836	36.65627558	0.89
0.1	36.65627558	0.89	2.995783771	0.079211705	39.65205935	0.9
0.1	39.65205935	0.9	3.274023249	0.179445675	42.9260826	0.91
0.1	42.9260826	0.91	3.580452352	0.289834775	46.50653495	0.92
0.1	46.50653495	0.92	3.918132981	0.411482035	50.42466793	0.93

Table B4 Iteration of CH_4

P_i	q_{max}	b_i	$q_{predict}$	δ_q	$^*q_{max}$	$^*b_i$
0.1	1	0.01	0.000999001	0.998964766	1.000999001	0.02
0.1	1.000999001	0.02	0.001998002	0.997929532	1.002997003	0.03
0.1	1.002997003	0.03	0.002999991	0.996891201	1.005996994	0.04
0.1	1.005996994	0.04	0.004007956	0.995846678	1.01000495	0.05
0.1	1.01000495	0.05	0.0050249	0.994792849	1.01502985	0.06
0.1	1.01502985	0.06	0.006053856	0.993726574	1.021083706	0.07
0.1	1.021083706	0.07	0.007097901	0.992644663	1.028181607	0.08
0.1	1.028181607	0.08	0.008160171	0.991543864	1.036341779	0.09
0.1	1.036341779	0.09	0.009243881	0.990420849	1.04558566	0.1
0.1	1.04558566	0.1	0.010352333	0.989272194	1.055937993	0.11
.	.	.	.	.	.	.
.	.	.	.	.	.	.
.	.	.	.	.	.	.
0.1	12.40423319	0.74	0.854667836	0.114333849	13.25890102	0.75
0.1	13.25890102	0.75	0.925039606	0.041409735	14.18394063	0.76
0.1	14.18394063	0.76	1.001839673	0.038175826	15.1857803	0.77
0.1	15.1857803	0.77	1.085705741	0.125083669	16.27148604	0.78
0.1	16.27148604	0.78	1.177343146	0.220044711	17.44882919	0.79
0.1	17.44882919	0.79	1.277532443	0.323867816	18.72636163	0.8
0.1	18.72636163	0.8	1.387137898	0.437448599	20.11349953	0.81
0.1	20.11349953	0.81	1.507116986	0.56177926	21.62061651	0.82

Table B5 Iteration of CO_2 in equivolume mixture

P_i	q_{max}	b_i	$q_{predict}$	δ_q	$*q_{max}$	$*b_i$
0.05	1	0.01	0.00049975	0.999825871	1.00049975	0.02
0.05	1.000249875	0.02	0.000999251	0.999651829	1.001249126	0.03
0.05	1.0007495	0.03	0.001498876	0.999477744	1.002248376	0.04
0.05	1.001498938	0.04	0.001999	0.999303484	1.003497938	0.05
0.05	1.002498438	0.05	0.002499996	0.999128921	1.004998434	0.06
0.05	1.003748436	0.06	0.003002239	0.998953924	1.006750675	0.07
0.05	1.005249556	0.07	0.003506102	0.998778362	1.008755658	0.08
0.05	1.007002607	0.08	0.004011963	0.998602104	1.011014569	0.09
0.05	1.009008588	0.09	0.004520198	0.998425018	1.013528786	0.1
0.05	1.011268687	0.1	0.005031187	0.998246973	1.016299874	0.11
.	.	.	.	.	.	.
.	.	.	.	.	.	.
.	.	.	.	.	.	.
0.05	33.49267263	1.74	2.680646291	0.065976902	36.17331892	1.75
0.05	34.83299577	1.75	2.802654832	0.023465215	37.63565061	1.76
0.05	36.23432319	1.76	2.930717317	0.02115586	39.16504051	1.77
0.05	37.69968185	1.77	3.065155575	0.067998458	40.76483742	1.78
0.05	39.23225964	1.78	3.206309557	0.11718103	42.43856919	1.79
0.05	40.83541441	1.79	3.354538403	0.168828712	44.18995282	1.8
0.05	42.51268362	1.8	3.510221583	0.223073722	46.0229052	1.81
0.05	44.26779441	1.81	3.673760104	0.280055786	47.94155451	1.82

Table B6 Iteration of CH_4 in equivolume mixture

P_i	q_{max}	b_i	$q_{predict}$	δ_q	$^*q_{max}$	$^*b_i$
0.05	1	0.01	0.00049975	0.999138362	1.00049975	0.02
0.05	1.00049975	0.02	0.0009995	0.998276724	1.00149925	0.03
0.05	1.00149925	0.03	0.001499999	0.997413795	1.002999249	0.04
0.05	1.002999249	0.04	0.002001995	0.996548285	1.005001244	0.05
0.05	1.005001244	0.05	0.002506238	0.995678901	1.007507481	0.06
0.05	1.007507481	0.06	0.003013482	0.994804341	1.010520963	0.07
0.05	1.010520963	0.07	0.003524488	0.993923297	1.014045451	0.08
0.05	1.014045451	0.08	0.004040022	0.993034445	1.018085473	0.09
0.05	1.018085473	0.09	0.004560861	0.992136447	1.022646333	0.1
0.05	1.022646333	0.1	0.005087793	0.991227944	1.027734126	0.11
.	.	.	.	.	.	.
.	.	.	.	.	.	.
.	.	.	.	.	.	.
0.05	11.07962067	1.01	0.532623364	0.081683855	11.61224403	1.02
0.05	11.61224403	1.02	0.563486628	0.028471332	12.17573066	1.03
0.05	12.17573066	1.03	0.596338687	0.028170149	12.77206935	1.04
0.05	12.77206935	1.04	0.631319017	0.088481064	13.40338837	1.05
0.05	13.40338837	1.05	0.668577567	0.152719943	14.07196593	1.06
0.05	14.07196593	1.06	0.708275588	0.221164807	14.78024152	1.07
0.05	14.78024152	1.07	0.750586541	0.294114727	15.53082806	1.08
0.05	15.53082806	1.08	0.795697073	0.371891506	16.32652513	1.09

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