

## REFERENCES

- Alcañiz-Monge, J., De La Casa-Lillo, M.A., Cazorla-Amoros, D., and Linares-Solano, A. (1997) Methane storage in activated carbon fibres. Carbons, 35(2), 291-297.
- Arami-Niya, A., Daud, W.M.A.W., and Mjalli, F.S. (2011) Comparative study of the textural characteristics of oil palm shell activated carbon produced by chemical and physical activation for methane adsorption. Chemical Engineering Research and Design, 89(6), 657-664.
- Bagheri, N., and Abedi, J. (2011) Adsorption of methane on corn cobs based activated carbon. Chemical Engineering Research and Design, 89(10), 2038-2043.
- Bastos-Neto, M., Canabrava, D.V., Torres, A.E.B., Rodriguez-Castello'n, E., Jimenez-Lo'pez, A., Azevedo, D.C.S., and Cavalcante Jr, C.L. (2007) Effects of textural and surface characteristics of microporous activated carbons on the methane adsorption capacity at high pressures. Applied Surface Science, 253, 5721-5725.
- Brady, T.A., Rostam-Abadi, M., and Rood, M.J. (1996) Applications for activated carbons from waste tires: natural gas storage and air pollution control. Gas Separation and Purification, 10(2), 97-102.
- Bremmell, K.E., Jameson, G.J., and Jameson, S. (1999) Adsorption of ionic surfactants in particulate systems: flotation, stability, and interaction forces. Colloids and Surfaces, 146, 75.
- Chen, J.P., and Wu, S. (2004) Acid/Base-treated activated carbons: characterization of functional groups and metal adsorptive properties. Langmuir, 20(6), 2233.
- Daud, W.M.A.W., and Ali, W.S.W. (2004) Comparison on pore development of activated carbon produced from palm shell and coconut shell. Bioresource Technology, 93, 63-69.
- Dipendu, S., and Shuguang, D. (2010) Hydrogen adsorption on metal-organic framework MOF-177. Tsinghua Science and Technology, 16, 363-376.

- Düren, T., Sarkisov, L., Yaghi, O.M., Snurr, R.Q. (2004) Design of new materials for methane storage. Langmuir, 20, 2683-2689.
- Esteves, I.A.A.C., Lopes, M.S.S., Nunes, P.M.C., and Mota, J.P.B. (2008) Adsorption of natural gas and biogas components on activated carbon. Separation and Purification Technology, 62(2), 281-296.
- Farzad, S., Taghikhani, V., Ghotbi, C., Aminshahidi, B., and Lay, E.N. (2007) Experimental and theoretical study of the effect of moisture on methane adsorption and desorption by activated carbon at 273.5 K. Journal of Natural Gas Chemistry. 16(1), 22-30.
- Geankoplis, C. (1993) Transport Process and Unit Operations. 3<sup>rd</sup> ed., New Jersey, Prentice Hall.
- Ginn, M.E., Kinney, F.B., Harris, J.C., and Am, J. (1961) Experimental studies on adsorption of surfactants onto cellulosic surface and its relevance to detergency. Journal of the American Oil Chemists Society, 38, 138.
- Hasan S., Hasim M.A. and Gupta B. S. (2000). Adsorption of NiSO<sub>4</sub> from Malatsian Rubber wood ash. Bioresource Technol., 72(1), 153-158.
- Inomata, K., Kanazawa, K., Urabe, Y., Hosono, H. and Araki, T. (2002) Natural gas storage in activated carbon pellets without a binder. Carbons, 40, 87-93.
- Jeong, B.M., Ahn, E.S., Yun, J.H., Lee, C.H., and Choi, D.K. (2007) Ternary adsorption equilibrium of H<sub>2</sub>/CH<sub>4</sub>/C<sub>2</sub>H<sub>4</sub> onto activated carbon. Separation and Purification Technology, 55(3), 335-342.
- Juergen, G., Irena, S., Dirk, W., Michael, T., David, F., Tina, D., Jasper M., B., Rajamani, K., and Stefan, K. (2010) Methane storage mechanism in the metal-organic framework Cu<sub>3</sub>(btc)<sub>2</sub>: An in situ neutron diffraction study. Microporous and Mesoporous Materials, 136, 50-58.
- Kavalov, B. (2004) Techno-economic analysis of natural gas application as an energy source for road transport in the EU. [N/A]. 15 Jun 2013. <[http://www.edis.sk/ekes/eur21\\_013en.pdf](http://www.edis.sk/ekes/eur21_013en.pdf)>.
- Li, L., Liu, S., and Liu, J. (2011) Surface modification of coconut shell based activated carbon for the improvement of hydrophobic VOC removal. Journal of Hazardous Materials, 192, 683-690.

- Lozano-Castelló, D.; Alcañiz-Monge, J.; De La Casa-Lillo, M.A. Cazorla-Amorós, D. and Linares-Solano, A. (2002a) Advances in the study of methane storage in porous carbonaceous materials. Fuels, 81, 1777-1803.
- Lozano-Castello, D., Cazorla-Amoros, D., Linares-Solano, A., and Quinn, D.F. (2002b) Activated carbon monoliths for methane storage: Influence of binder. Carbons, 40(15), 2871-2825.
- Ma, S., Sun, D., Simmons, J.M., Collier, C.D., Yuan, D., Zhou, H.C. (2008) Metal-organic framework from an anthracene derivative containing nanoscopic cages exhibiting high methane uptake. Journal of the American Chemical Society, 130, 1012-1016.
- Najibi, H., Chapoy, A., and Tohidi B. (2008) Methane/natural gas storage and -delivered capacity for activated carbons in dry and wet condition. Fuels, 87(1), 7-13.
- Nameni M., Alavi M. R. and Arami M. (2008) Adsorption of hexavalent chromium from aqueous solutions by wheat bran. International Journal of Environmental Science and Technology, 5(2), 161-168.
- Namvar-Asl, M., Soltanieh, M., Rashidi, A., and Irandoukht, A. (2008) Modeling and preparation of activated carbon for methane storage I. Modeling of activated carbon characteristics with neural networks and response surface method. Energy Conversion and Management, 49, 2471-2477.
- Park, S., Jang, Y., Shim, J., and Ryu, S. (2003) Studies on pore structures and surface functional groups of pitch based activated carbon fibers. Journal of Colloid Interface Science, 260, 259-264.
- Paulo, C., Eduarrdo, L., De Gilmar, A., and Joao, B. (1999) Adsorption of sodium dodecylsulfate on a hydrotalcite-like compound. Effect of temperature, pH and ionic strength. Colloids and Surfaces, 154, 399.
- Patel, S. and Desai, H. (2011) Potential of *Moringa oleifera* seeds leaves and barks for removal of Hexavalent Chromium from aqueous solution with reference to adsorption isotherm. Journal of Environmental Research And Devolpment, 3(2), 41-57.

- Pfeifer, P. (2011) Advanced natural gas fuel tank project. [N/A]. [10 April 2013] [.www1.eere.energy.gov/cleancities/pdfs/ngvtfl1\\_pfeifer.pdf](http://www1.eere.energy.gov/cleancities/pdfs/ngvtfl1_pfeifer.pdf).
- Prauchner, M.J., and Rodríguez-Reinoso, F. (2008) Preparation of granular activated carbons for adsorption of natural gas. Microporous and Mesoporous Materials, 109(1-3), 581-584.
- Prasetyo, I., and Do, D.D. (1998) Adsorption rate of methane and carbon dioxide on activated carbon by the semi-batch constant molar flow rate method. Chemical Engineering Science, 53(19), 3459-3467.
- Pupier, O., Goetz, V., and Fiscal, R. (2005) Effect of cycling operations on an adsorbed natural gas storage. Chemical Engineering and Processing, 44(1), 71-79.
- Qiao, S., and Hu, X. (2000) Effect of micropore size distribution induced heterogeneity on binary adsorption kinetics of hydrocarbons in activated carbon. Chemical Engineering Science, 55, 1533-1544.
- Qing, H., Yingbo, X., Chenghui, W., Shung, Z., and Ran, W. (2011) Silane modification and characterization of activated carbon. Adsorption, 23-29
- Rahman, K.A., Loh, W.S., Chakraborty, A., Saha, B.B., and Choon, K. (2010) Adsorption thermodynamics of natural gas storage onto pitch-based activated carbons. Paper presented at Proceedings of the 2<sup>nd</sup> Annual Gas Processing Symposium, 187-195.
- Rashidi, A.M., Lotfi, R., Nouralishahi, A., Khodaghali, M.A., Zare, M., and Eslamipour, F. (2011) Nanoporous carbons as promising novel methane adsorbents for natural gas technology. Journal of Natural Gas Chemistry, 20(6), 664-668.
- Rodríguez-Reinoso, F., Nakagawa, Y., Silvestre-Albero, J., Juárez-Galan, J.M., and Molina-Sabio, M. (2008) Correlation of methane uptake with microporosity and surface area of chemically activated carbons. Microporous and Mesoporous Materials, 115(3), 603-608.
- Saha, D., and Deng, S. (2010) Hydrogen adsorption on metal-organic framework MOF-177. Tsinghua Science and Technology, 1(16), 363-376.
- Salehi, E., Taghikhani, V., Ghotbi, C., Nemati Lay, E., and Shojaei, A. (2007) Theoretical and experimental study on the adsorption and desorption of

- methane by granular activated carbon at 25 °C. Journal of Natural Gas Chemistry, 16(4), 415-422.
- Shao, X., Wang, W., and Zhang, X. (2007) Experimental measurements and computer simulation of methane adsorption on activated carbon fibers. Carbons, 45, 188–195.
- Shen, W.Z., Li, Z.J., and Liu, Y.H. (2008) Surface chemical functional groups modification of porous carbon. Recent Patents on Chemical Engineering, 1, 27-40.
- Shengqian, M., Daofeng, S., Jason M., S., Christopher D., C., Daqiang, Y., and Hong-Cai Z. (2007) Metal-organic framework from an anthracene derivative containing nanoscopic cages exhibiting high methane uptake. Journal of the American Chemical Society, 9, 130(3), 1016.
- Solar, C., Blanco, A., Vallone, A., and Sapag, A (2010) Adsorption of methane in porous materials as the basis for the storage of natural gas. Natural Gas, 978-953-307-112-1.
- Somasundaran, P., Fuerstenau, D.W., and Phy, J. (1966) Adsorption of surfactants on minerals for wettability control in improved oil recovery processes. Journal of Physical Chemistry, 70, 90.
- Soo-Jin, P., and Min-Kang, S. (2011) Solid-Gas Interaction. Interface Science and Composites, 70-104.
- Sun, J., Brady, T.A., Rood, M.J., Lehmann, C.M., Rostam-Abadi, M., and Lizzio, A.A. (1997) Adsorbed natural gas storage with activated carbons made from Illinois coals and scrap tires. Energy & Fuels, 11, 316-322.
- Tagliabue, M., Farrusseng, D., Valencia, S., Aguado, S., Ravon, U., Rizzo, C., Corma, A., and Mirodatos, C. (2009) Natural gas treating by selective adsorption: Material science and chemical engineering interplay. Chemical Engineering Journal, 155(3), 553-566.
- Vasiliev, L.L., Kanonchik, L.E., Mishkinis, D.A., and Rabetsky, M.I. (2000) Adsorbed natural gas storage and transportation vessels. International Journal of Thermal Sciences, 39(9-11), 1047-1055.

- Xue , C., Zhou , Z., Yang , Q., and Zhong , C. (2009) Enhanced methane adsorption in catenated metal-organic frameworks: A molecular simulation study. Chinese Journal of Chemical Engineering, 17(4), 580-584.
- Yang, H., Gong, M., and Chen Y. (2011) Preparation of activated carbons and their adsorption properties for greenhouse gases: CH<sub>4</sub> and CO<sub>2</sub>. Journal of Natural Gas Chemistry, 20(5), 460-464.
- Zeng, -X. and Osseo-Asare, K. (2001) Partition of pyrite and hematite in aqueous biphasic system: effects of pH and surfactants. Colloids and Surfaces A, 17, 247-254.
- Zhou, H. Metal-Organic Frameworks for Adsorbed Natural Gas Fuel Systems. Department of Chemistry Texas A&M University. [N/A]. [27 April 2013]. <[http://growingtexas.tamu.edu/Resources/documents/Zhou\\_2012.pdf](http://growingtexas.tamu.edu/Resources/documents/Zhou_2012.pdf)>.
- Zhou, W. (2011) Methane storage in porous metal-organic frameworks: Current records and future perspectives. Materials Science and Engineering, 10, 200-204.
- Zor, S. (2004) Investigation of the adsorption of anionic surfactants at different pH values by means of active carbon and the kinetics of adsorption. Journal of the Serbian Chemical Society, 69(1), 25-32.
- Coconut Shell Charcoal Granules manufacturers. [N/A]. [15 March 2013]. <[www.chemechel.com/products.htm](http://www.chemechel.com/products.htm)>.
- Low Pressure Adsorbed Natural Gas for Vehicles. [N/A]. [24 March 2013]. <[www.glnobledenton.com/assets/downloads/ANG\\_for\\_Vehicle\\_truck\\_DS.pdf](http://www.glnobledenton.com/assets/downloads/ANG_for_Vehicle_truck_DS.pdf)>.
- Adsorbed Natural Gas Technology. Enertek Company. [N/A]. [9 April 2013]. <[www.energtek.com/t/1008-ang-technology](http://www.energtek.com/t/1008-ang-technology)>.
- Green car reports. [N/A]. [9 April 2013]. <[www.greencar.com/articles/clean-burning-natural-gas-vehicles.php](http://www.greencar.com/articles/clean-burning-natural-gas-vehicles.php)>.
- Natural Gas. [N/A]. [16 April 2013]. <[www.naturalgas.org](http://www.naturalgas.org)>.
- Adsorption. [N/A]. [25 April 2013]. <<http://en.wikipedia.org/wiki/Adsorption>>.
- Renewable energy. [N/A]. [25 April 2013]. <[www.eon.com/en/business-areas/mobility/gas-mobility.html](http://www.eon.com/en/business-areas/mobility/gas-mobility.html)>.
- Silica gel. [N/A]. [27 April 2013]. <[http://en.wikipedia.org/wiki/Silica\\_gel](http://en.wikipedia.org/wiki/Silica_gel)>.

Silica gel. Grace Enriching Lives, Everywhere company. [N/A]. [27 April 2013].  
<[www.grace.com/EngineeredMaterials/MaterialSciences/SilicaGel](http://www.grace.com/EngineeredMaterials/MaterialSciences/SilicaGel)>.

Zeolite Molecular Sieve. Xinyuan Technology Co., Ltd. [27 April 2013].  
<[www.molecularsieve.org/Zeolite\\_Molecular\\_Sieve](http://www.molecularsieve.org/Zeolite_Molecular_Sieve)>.

Polymer adsorption. [27 April 2013]. <[http://en.wikipedia.org/wiki/Polymer\\_adsorption](http://en.wikipedia.org/wiki/Polymer_adsorption)>.

MOF technology. [2012]. [27 April 2013]. <[www.moftechnologies.com/gas-storage.html](http://www.moftechnologies.com/gas-storage.html)>.

## APPENDICES

### Appendix A Adsorbent Physical Characterization

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

**Table A1** Physical characteristic properties of investigated adsorbents

Physical Characterization	Adsorbent Specification	
	Coconut Shell Activated Carbon (CSAC)	Palm Shell Activated Carbon (PSAC)
Apparent Density (g/cm <sup>3</sup> )	> 0.48	> 0.48
Moisture Content (%w/w)	< 8.0	< 8.0
Ash Content (%w/w)	< 3.5	< 5.0
pH	9-11	9-11
Iodine Number (mg/g)	> 1,100	> 1,100
Hardness Number (%)	> 98.0	> 98.0

The zeta potential of all samples as a function of pH ( $\text{pH}_{\text{pzc}}$ ) is summarized in Table A2.



**Table A2** Zeta potential measurements of activated carbon

<b>Adsorbents</b>	<b>pH<sub>pzc</sub></b>	<b>Adsorbents</b>	<b>pH<sub>pzc</sub></b>
CSAC – Untreated	4.5	CSAC – Treated by H <sub>2</sub> SO <sub>4</sub>	3.3
CSAC – Treated by KOH	7.6	CSAC – Treated by HNO <sub>3</sub>	2.5
CSAC – Treated by NaOH	7.5	CSAC – Treated by H <sub>3</sub> PO <sub>4</sub>	2.7
CSAC – Treated by NH <sub>4</sub> OH	7.3	PSAC – Untreated	5.2

Nitrogen adsorption-desorption isotherms of the adsorbents were conducted using an Autosorb-1MP (Quantachrome Instrument) in Table A3.

**Table A3** BET surface area, micropore volume, and average pore diameter of investigated adsorbents

Adsorbent	Physical Characterization			
	BET	Micropore	Total pore	Average pore
	Surface area (m <sup>2</sup> /g)	volume (cm <sup>3</sup> /g)	volume (cm <sup>3</sup> /g)	diameter (Å)
CSAC – Untreated	1,062	0.57	0.59	22.45
CSAC – Treated by KOH	900	0.48	0.50	22.29
CSAC – Treated by NaOH	1,019	0.55	0.57	22.20
CSAC – Treated by NH <sub>4</sub> OH	1,022	0.55	0.57	22.15
CSAC – Treated by H <sub>2</sub> SO <sub>4</sub>	996	0.53	0.55	22.26
CSAC – Treated by HNO <sub>3</sub>	988	0.53	0.55	22.30
CSAC – Treated by H <sub>3</sub> PO <sub>4</sub>	922	0.49	0.51	22.28
PSAC – Untreated	1,058	0.56	0.59	22.28

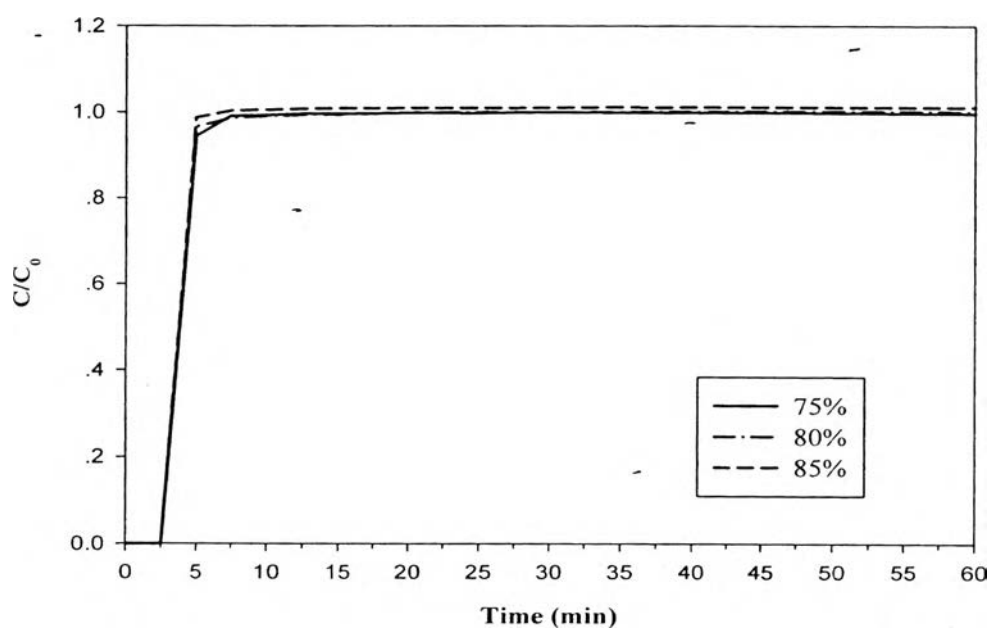
Fourier transform infrared spectroscopy (FTIR) was used to qualitatively evaluate the chemical structure of carbon materials in Table A4.

**Table A4** IR assignments o functional groups on carbon surfaces

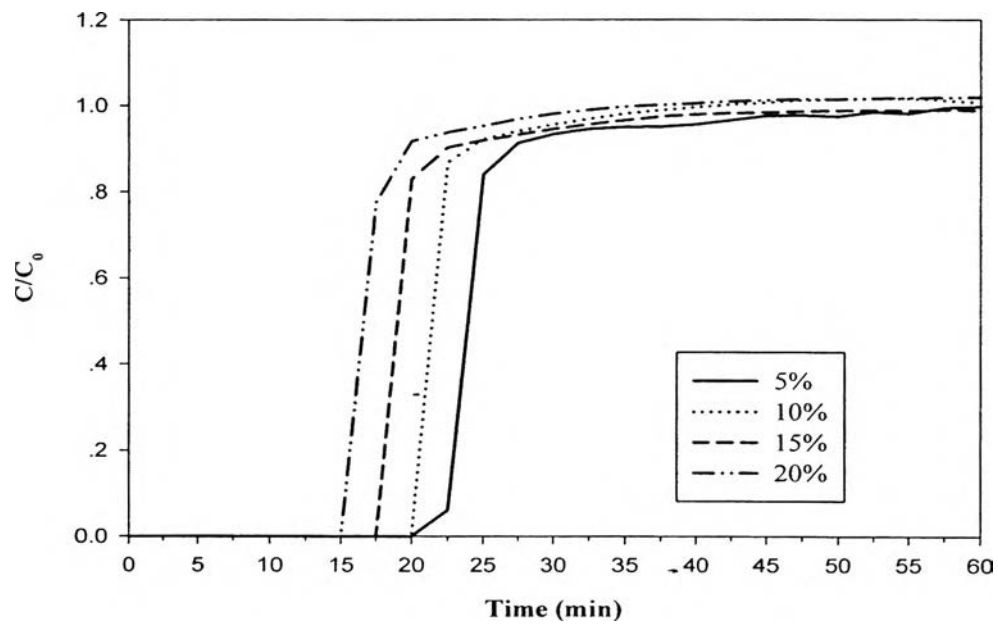
Adsorption peaks (cm-1)							
Surface group	In reference	This work					
		CSAC	CSAC/NaOH	CSAC/NH <sub>4</sub> OH	CSAC/H <sub>2</sub> SO <sub>4</sub>	CSAC/HNO <sub>3</sub>	CSAC/H <sub>3</sub> PO <sub>4</sub>
-OH	3435	3430	3400	3408	3409	3394	3436
CH <sub>2</sub>	2920	2925	2922	2925	2918	2921	2923
C=C	1629	1633	1628	1630	1629	1615	1631
C=O	1558	-	1558	1572	-	1566	-
COOH	1382-1392	-	1385	-	-	1390	1384
COC	1157	-	-	-	-	1134	-
COH	1118	1115	1120	1133	1118	-	1130

## Appendix B Adsorption and Desorption Curves in Different Scale

The breakthrough curves of methane and carbon dioxide were plotted in terms of concentration ratio versus time, as shown in Figures 4.3 and 4.4 are shown in Figure B1 and B2.

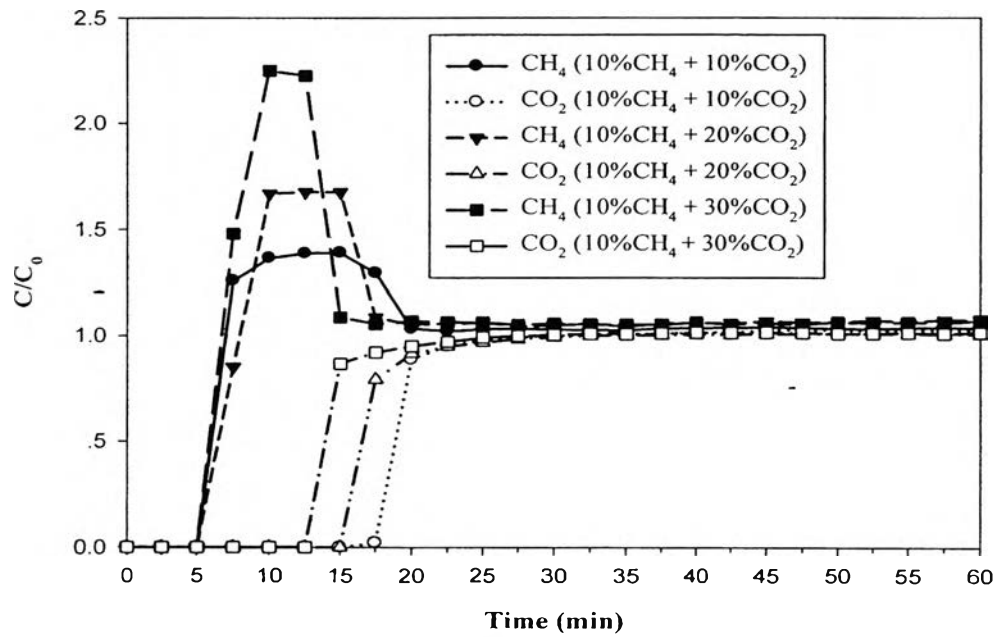


**Figure B1** Breakthrough curves of methane from the adsorption on the CSAC with the initial concentration of methane at 75, 80, and 85 vol% at room temperature.



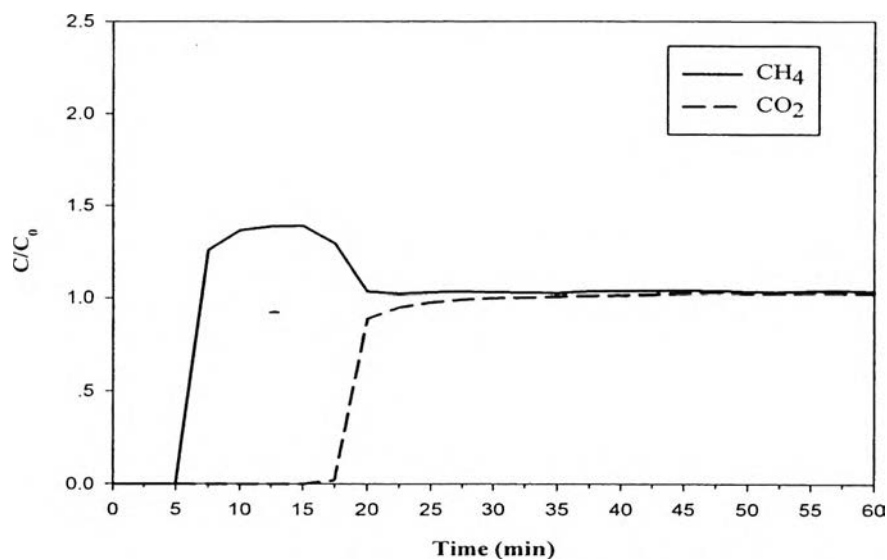
**Figure B2** Breakthrough curves of carbon dioxide from the adsorption on the CSAC with the initial concentration of carbon dioxide at 5, 10, 15, and 20 vol% at room temperature.

Some of the adsorption and desorption curves from the breakthrough experiments are rewritten in a different scale. The breakthrough curves in Figure 4.8 are shown here in Figure B3.

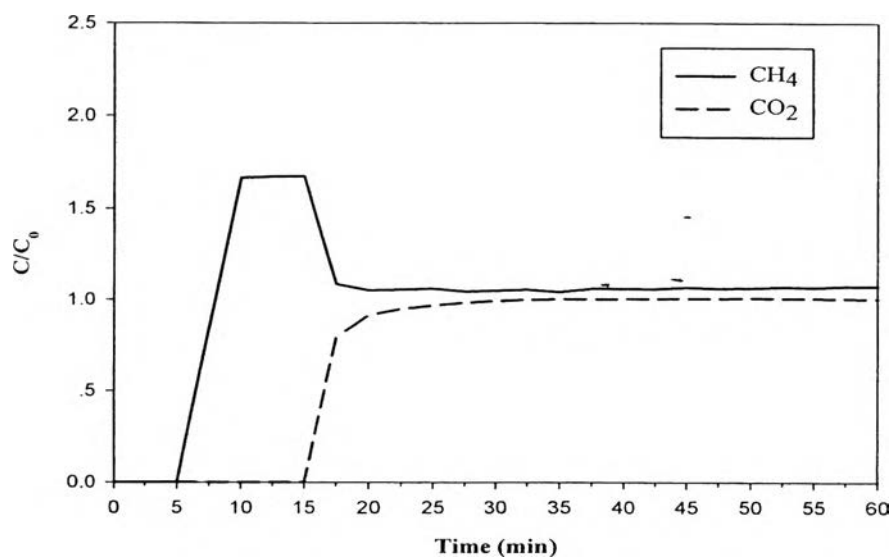


**Figure B3** Breakthrough curves of methane and carbon dioxide from the competitive adsorption on the CSAC with the initial concentration of methane at 10 vol% and carbon dioxide at 10, 20, and 30 vol% at room temperature.

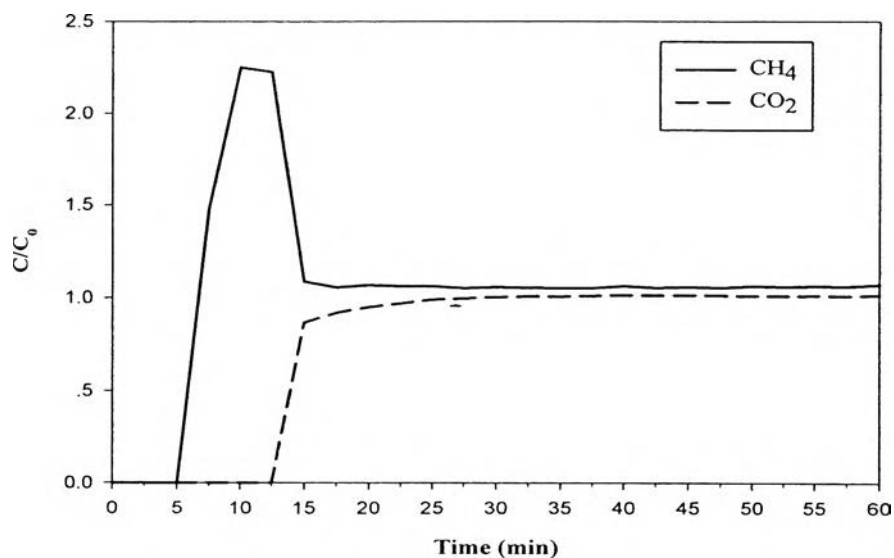
Competitive adsorption at 10 vol% and carbon dioxide composition was varied from 10 to 30 vol% profiles in Figures 4.5 to 4.7 are shown in Figures B4 to B6.



**Figure B4** Breakthrough curves of methane and carbon dioxide from the competitive adsorption on the CSAC with the initial concentration of methane at 10 vol% and carbon dioxide at 10 vol% at room temperature.



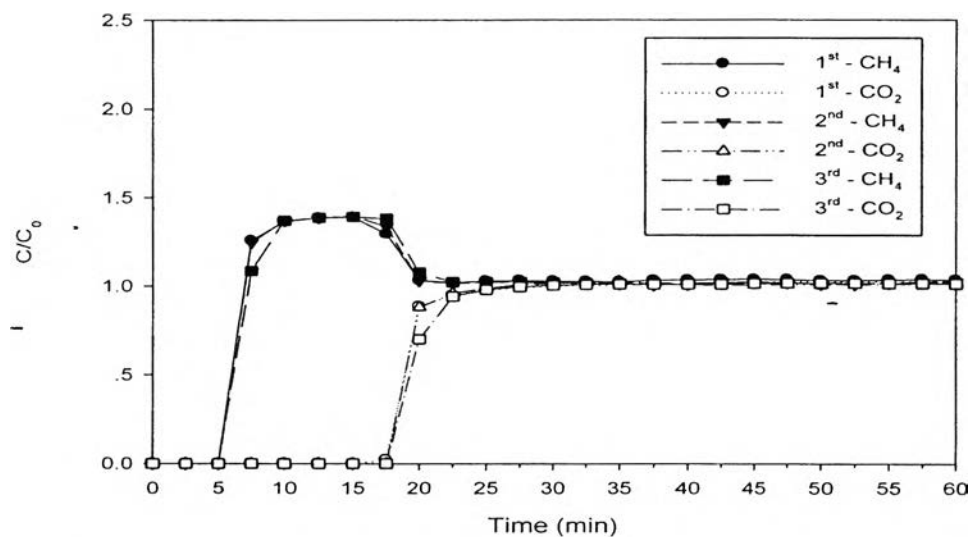
**Figure B5** Breakthrough curves of methane and carbon dioxide from the competitive adsorption on the CSAC with the initial concentration of methane at 10 vol% and carbon dioxide at 20 vol% at room temperature.



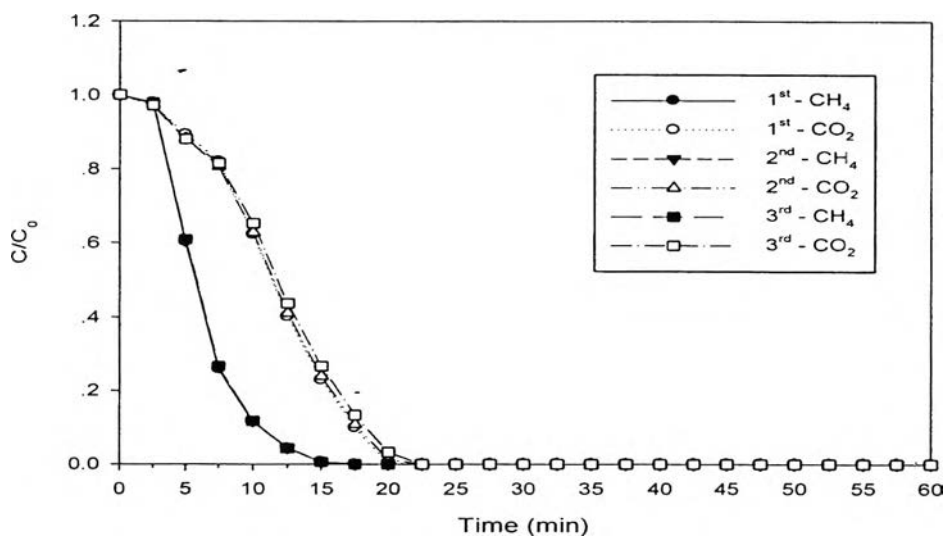
**Figure B6** Breakthrough curves of methane and carbon dioxide from the competitive adsorption on the CSAC with the initial concentration of methane at 10 vol% and carbon dioxide at 30 vol% at room temperature.

The breakthrough curves and the desorption cycles in Figures 4.9 to 4.22 are shown in Figures B7 to B21.

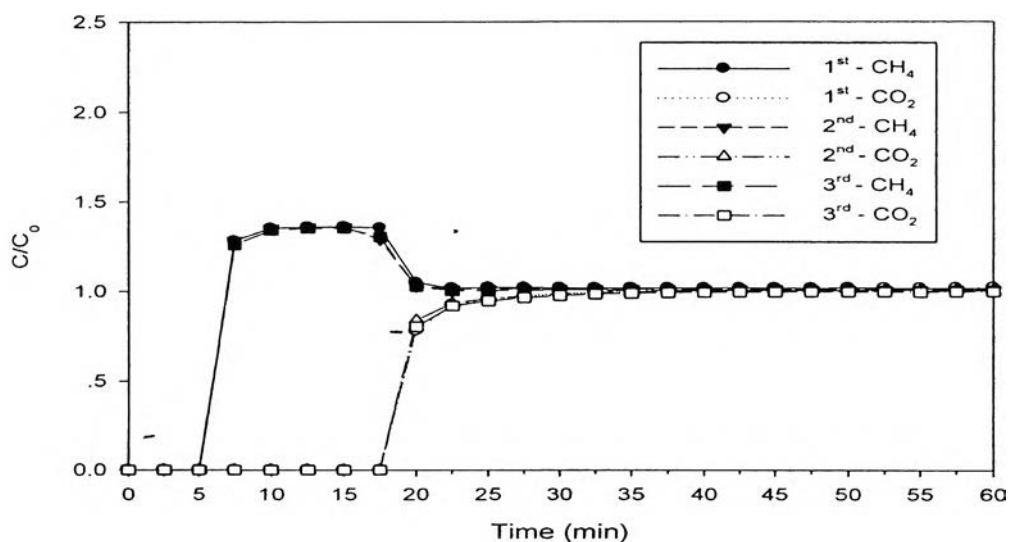




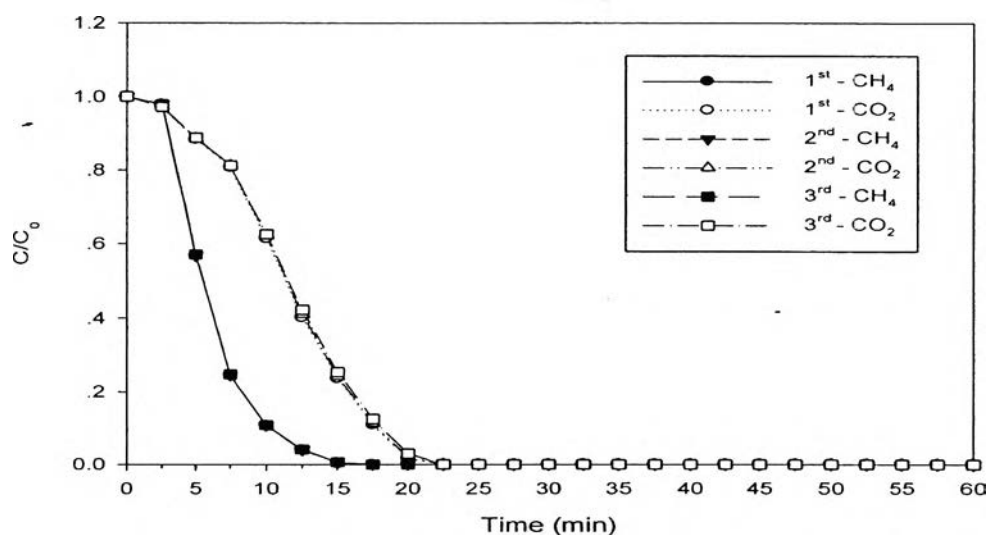
**Figure B7** Breakthrough curves of methane and carbon dioxide from the 3-cycle adsorption process on untreated CSAC with the initial concentration of methane at 10 vol% and carbon dioxide at 10 vol% at room temperature.



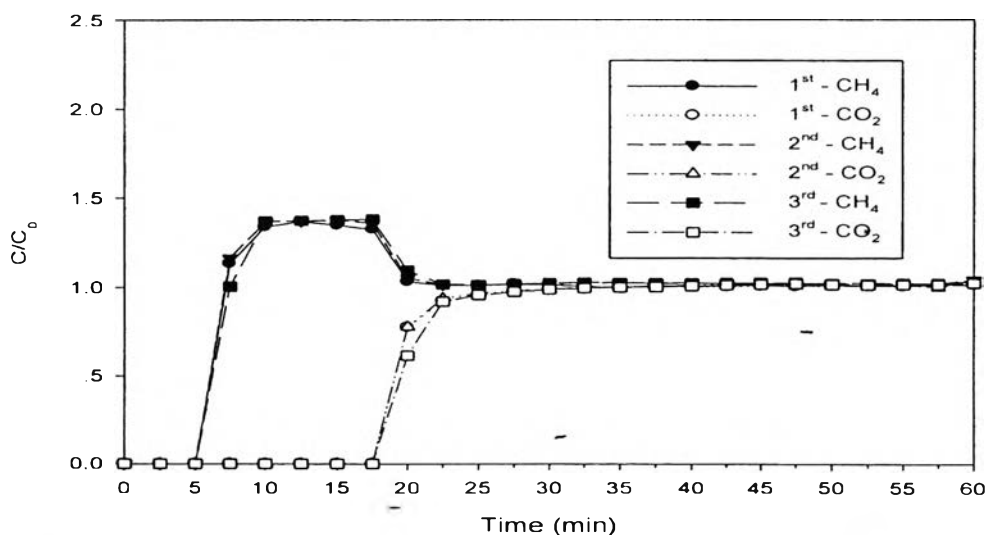
**Figure B8** Three desorption cycles of methane and carbon dioxide from the CSAC with the initial concentration of methane at 10 vol% and carbon dioxide at 10 vol% at room temperature.



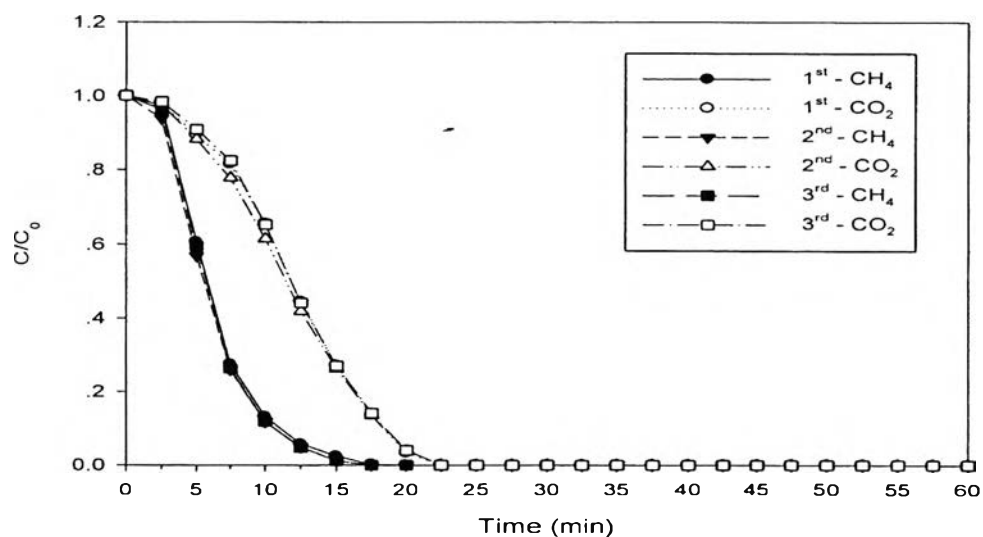
**Figure B9** Breakthrough curves of methane and carbon dioxide from the 3-cycle adsorption process on the CSAC treated by sodium hydroxide with the initial concentration of methane at 10 vol% and carbon dioxide at 10 vol% at room temperature.



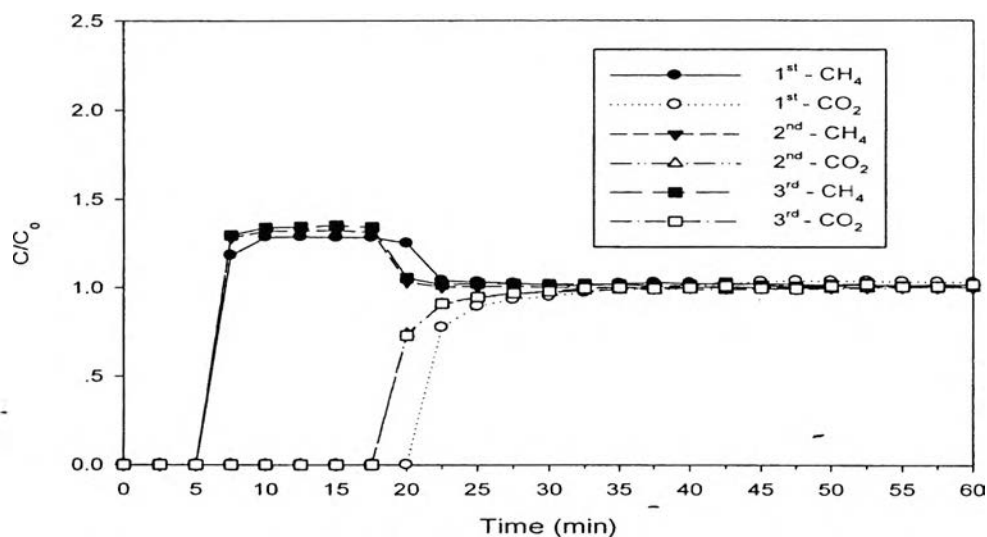
**Figure B10** Three desorption cycles of methane and carbon dioxide from the CSAC treated by sodium hydroxide with the initial concentration of methane at 10 vol% and carbon dioxide at 10 vol% at room temperature.



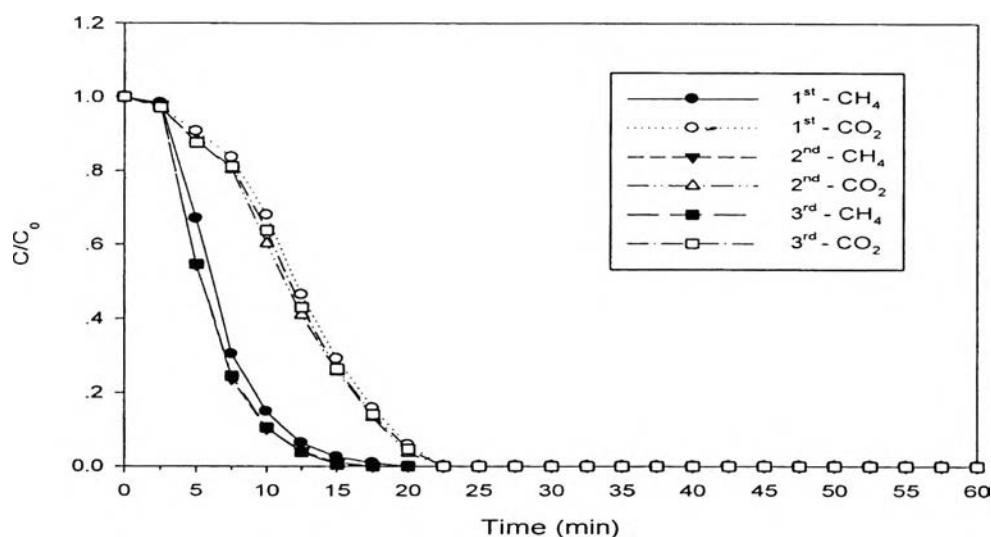
**Figure B11** Breakthrough curves of methane and carbon dioxide from the 3-cycle adsorption process on the CSAC treated by ammonium hydroxide with the initial concentration of methane at 10 vol% and carbon dioxide at 10 vol% at room temperature.



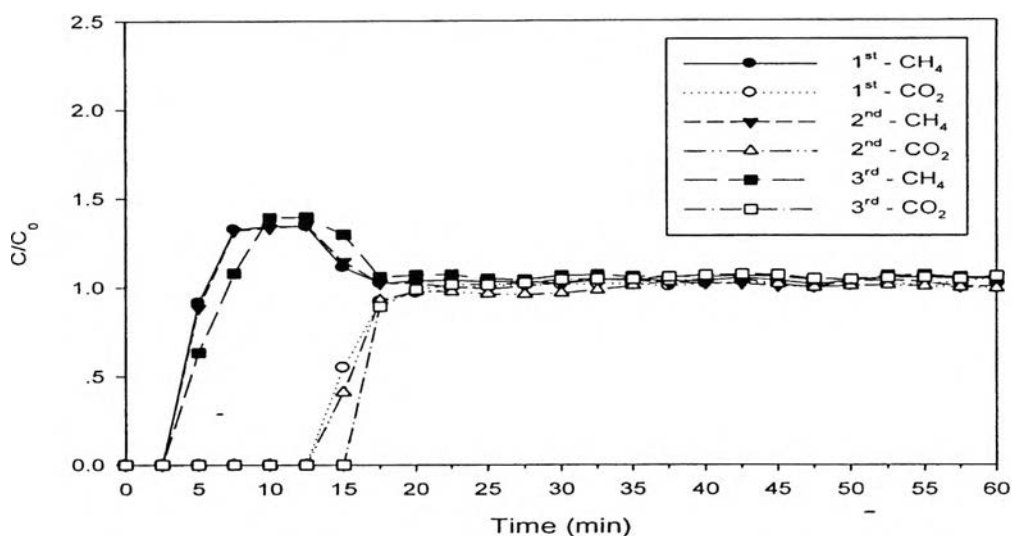
**Figure B12** Three desorption cycles of methane and carbon dioxide from the CSAC treated by ammonium hydroxide with the initial concentration of methane at 10 vol% and carbon dioxide at 10 vol% at room temperature.



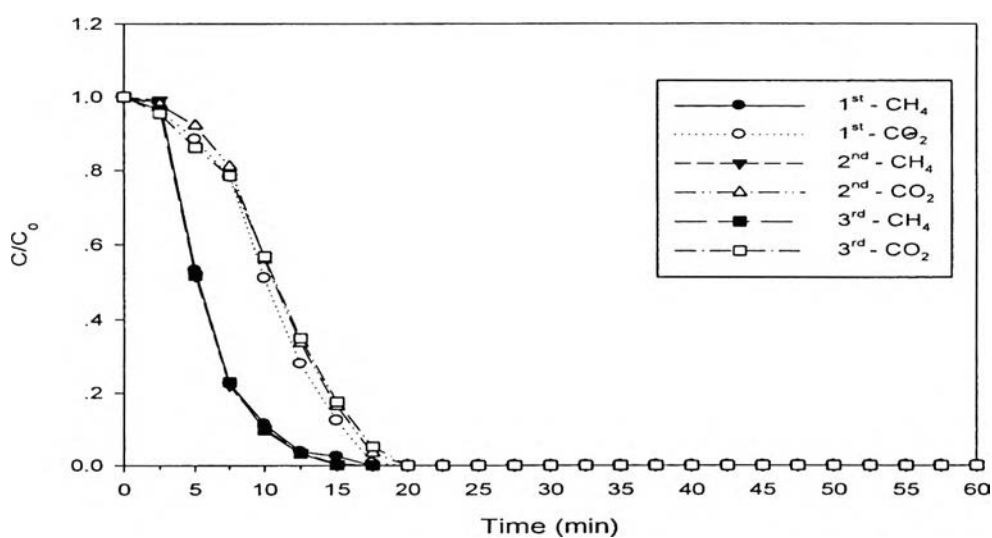
**Figure B13** Breakthrough curves of methane and carbon dioxide from the 3-cycle adsorption process on the CSAC treated by potassium hydroxide with the initial concentration of methane at 10 vol% and carbon dioxide at 10 vol% at room temperature.



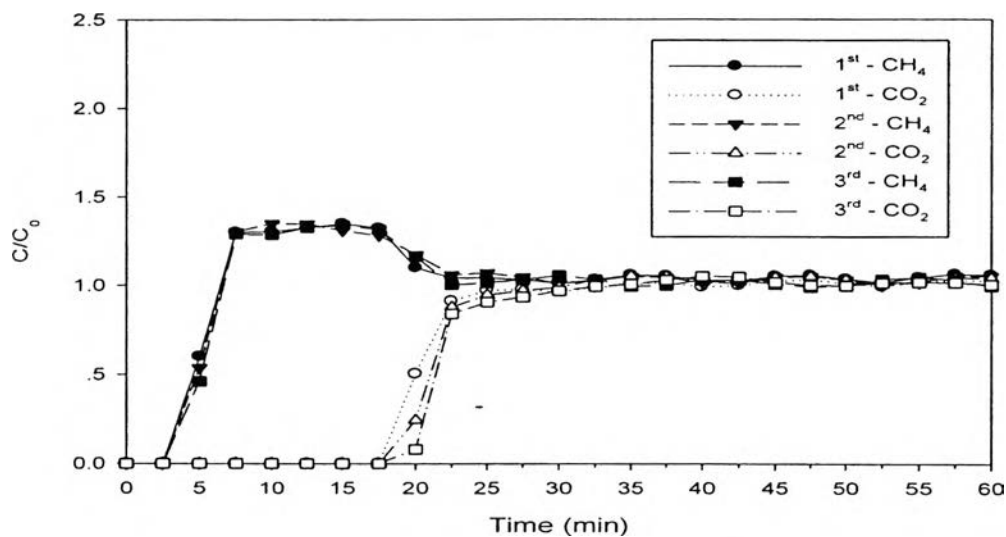
**Figure B14** Three desorption cycles of methane and carbon dioxide from the CSAC treated by potassium hydroxide with the initial concentration of methane at 10 vol% and carbon dioxide at 10 vol% at room temperature.



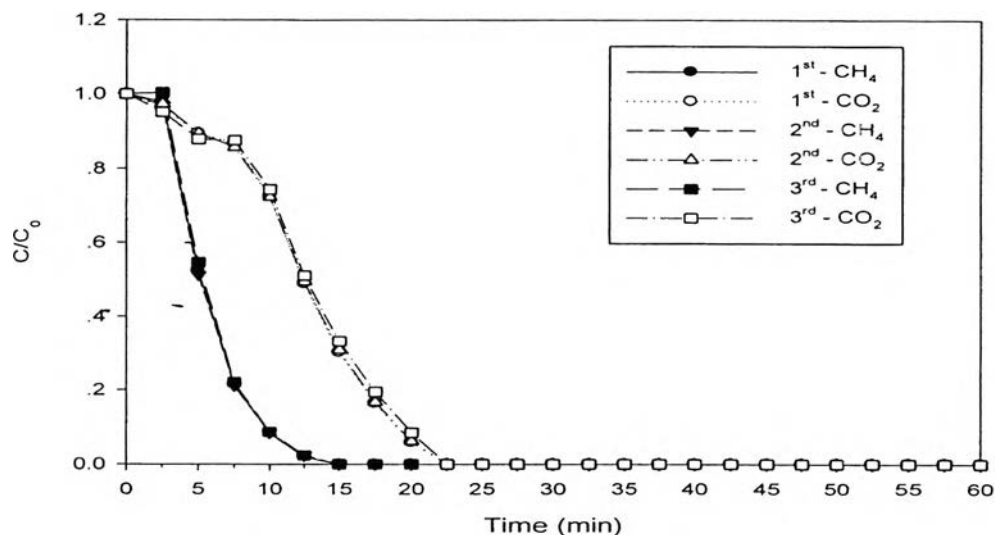
**Figure B15** Breakthrough curves of methane and carbon dioxide from the 3-cycle adsorption process on the CSAC treated by sulfuric acid with the initial concentration of methane at 10 vol% and carbon dioxide at 10 vol% at room temperature.



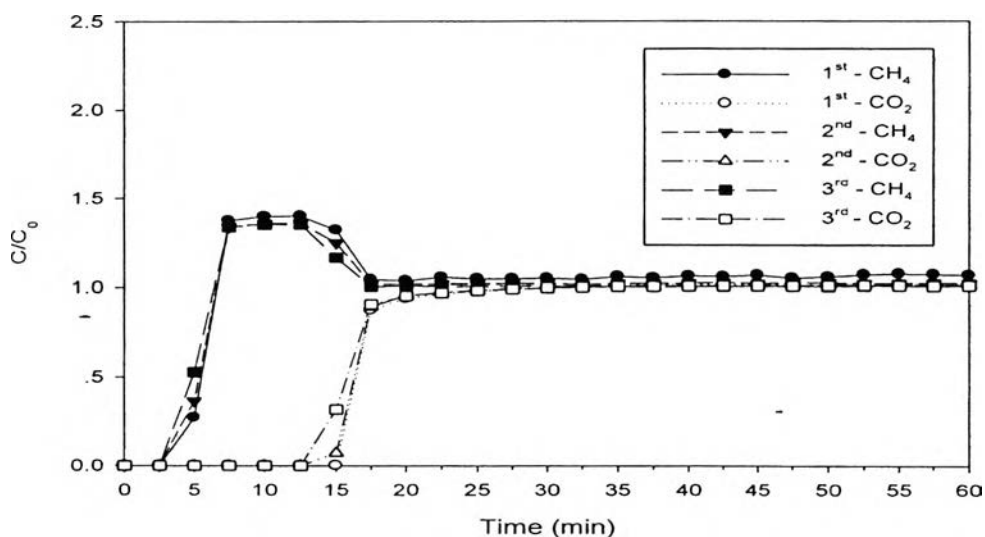
**Figure B16** Three desorption cycles of methane and carbon dioxide from the CSAC treated by sulfuric acid with the initial concentration of methane at 10 vol% and carbon dioxide at 10 vol% at room temperature.



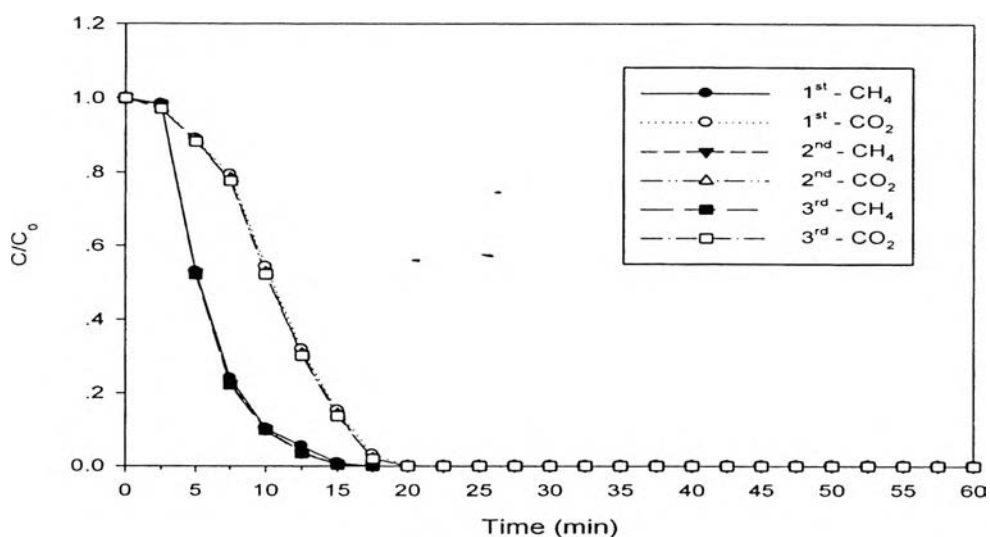
**Figure B17** Breakthrough curves of methane and carbon dioxide from the 3-cycle adsorption process on the CSAC treated by nitric acid with the initial concentration of methane at 10 vol% and carbon dioxide at 10 vol% at room temperature.



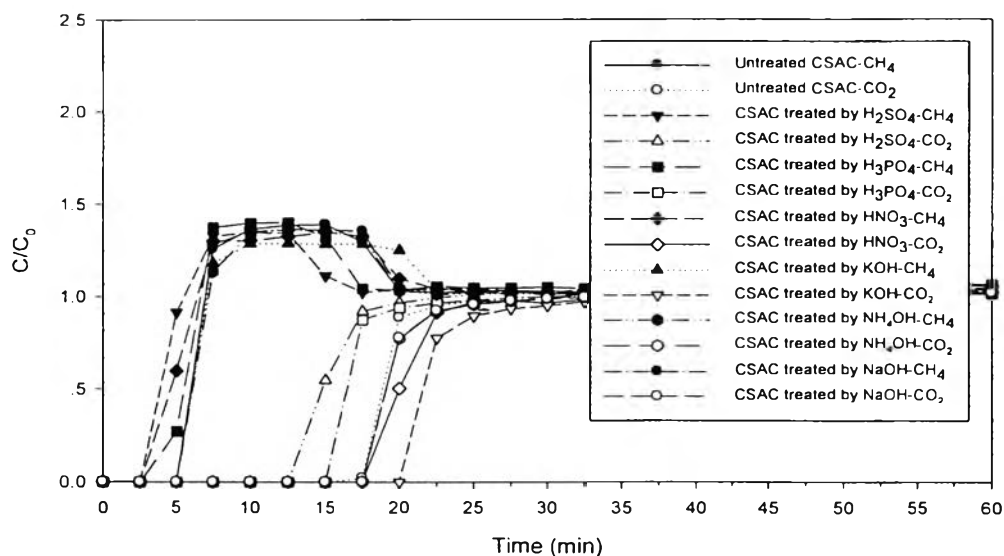
**Figure B18** Three desorption cycles of methane and carbon dioxide from the CSAC treated by nitric acid with the initial concentration of methane at 10 vol% and carbon dioxide at 10 vol% at room temperature.



**Figure B19** Breakthrough curves of methane and carbon dioxide from the 3-cycle adsorption process on the CSAC treated by phosphoric acid with the initial concentration of methane at 10 vol% and carbon dioxide at 10 vol% at room temperature.



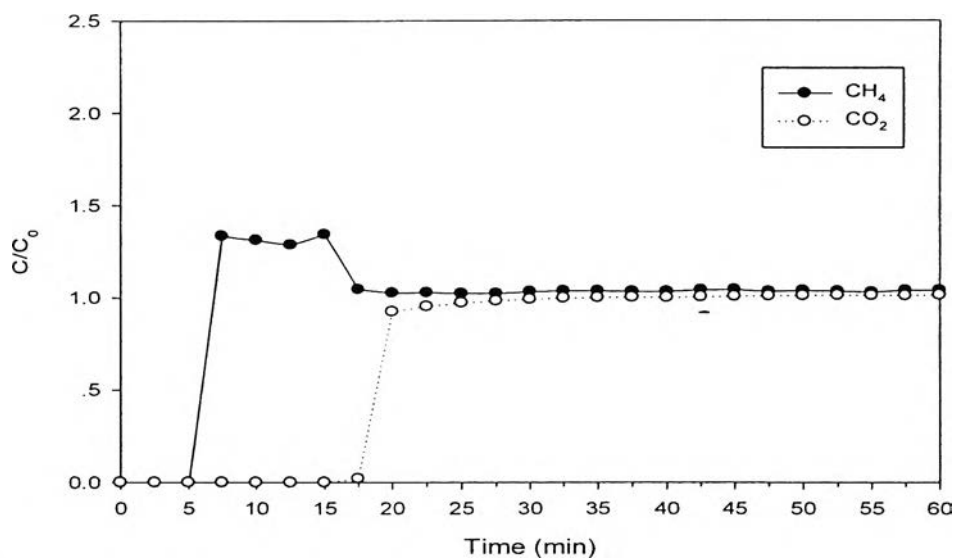
**Figure B20** Three desorption cycles of methane and carbon dioxide from the CSAC treated by phosphoric acid with the initial concentration of methane at 10 vol% and carbon dioxide at 10 vol% at room temperature.



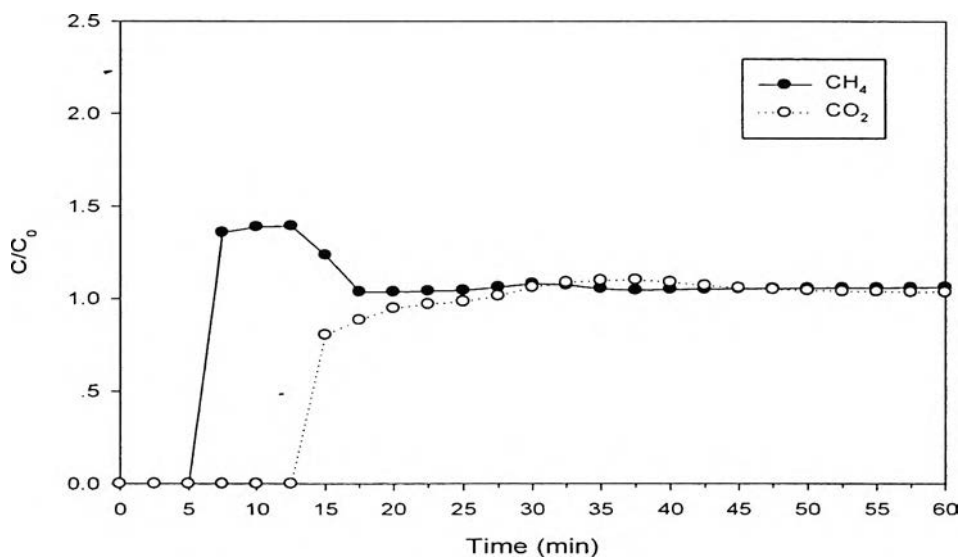
**Figure B21** Breakthrough curves of methane and carbon dioxide from the competitive adsorption on the untreated CSAC, CSAC treated by sulfuric acid, CSAC treated by phosphoric acid, CSAC treated by nitric acid, CSAC treated by potassium hydroxide, CSAC treated by ammonium hydroxide and CSAC treated by sodium hydroxide with the initial concentration of methane at 10 vol% and carbon dioxide at 10 vol% at room temperature.

The breakthrough curves of CSAC treated MES on varies concentration in Figures 4.31 to 4.34 are shown in Figures B22 to B25.

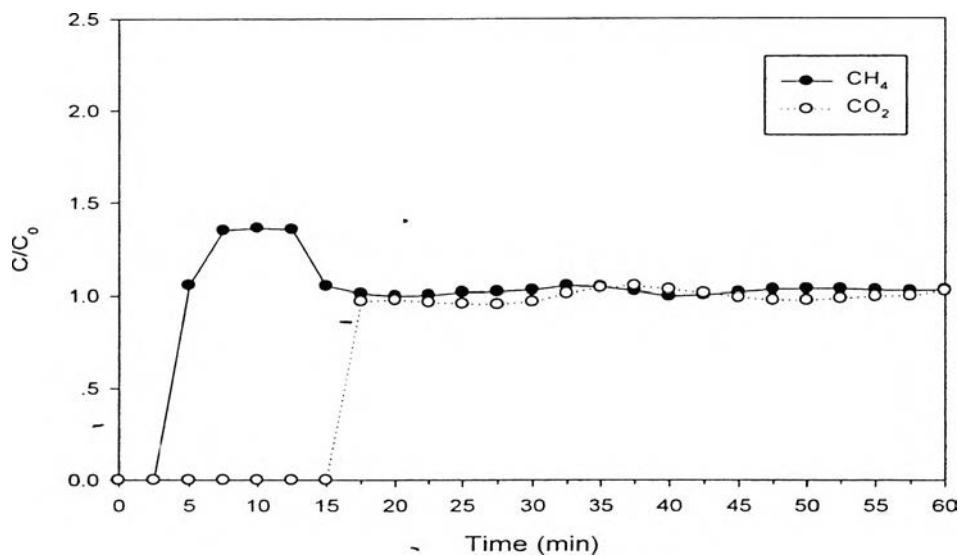




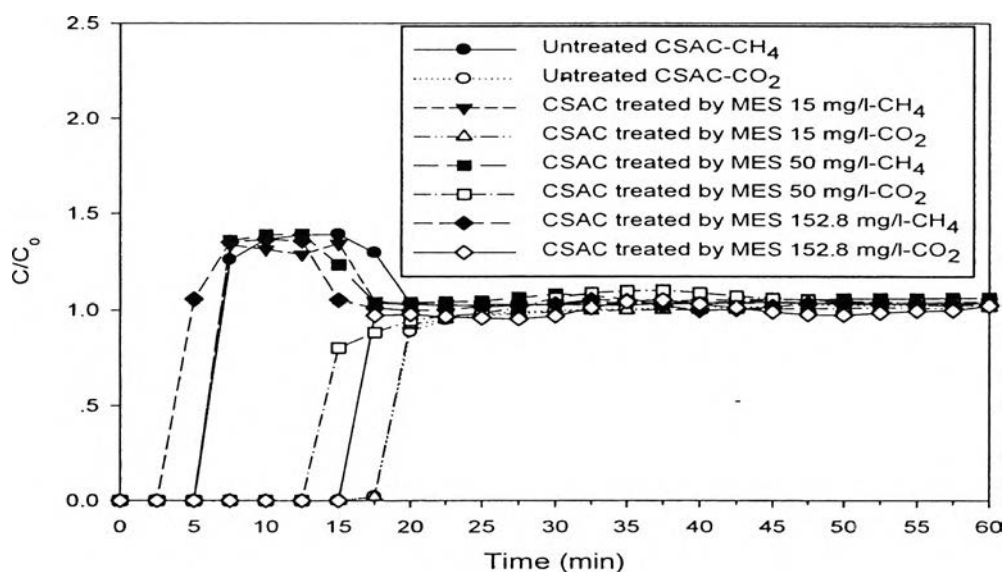
**Figure B22** Breakthrough curves of methane and carbon dioxide from the competitive adsorption on the CSAC treated by MES at 15 mg/l with the initial concentration of methane at 10 vol% and carbon dioxide at 10 vol% at room temperature.



**Figure B23** Breakthrough curves of methane and carbon dioxide from the competitive adsorption on the CSAC treated by MES at 50 mg/l with the initial concentration of methane at 10 vol% and carbon dioxide at 10 vol% at room temperature.



**Figure B24** Breakthrough curves of methane and carbon dioxide from the competitive adsorption on the CSAC treated by MES at 152.8 mg/l with the initial concentration of methane at 10 vol% and carbon dioxide at 10 vol% at room temperature.



**Figure B25** Breakthrough curves of methane and carbon dioxide from the competitive adsorption on the untreated CSAC, CSAC treated by MES at 15mg/l, 50 mg/l, and 152.8 mg/l with the initial concentration of methane at 10 vol% and carbon dioxide at 10 vol% at room temperature.

**Table B1** Summary of breakthrough time adsorption capacity and selectivity of investigate CSAC

Adsorbent	Breakthrough time (min)		Total adsorption (mmol/g)	CH <sub>4</sub> adsorption (mmol/g)	CO <sub>2</sub> adsorption (mmol/g)	Adsorption selectivity
	CH <sub>4</sub>	CO <sub>2</sub>				
CSAC	3.46	17.00	3.41	0.58	2.84	0.20
CSAC/H <sub>2</sub> SO <sub>4</sub>	1.76	14.64	2.74	0.29	2.44	0.12
CSAC/H <sub>3</sub> PO <sub>4</sub>	2.26	16.53	3.13	0.38	2.76	0.14
CSAC/HNO <sub>3</sub>	2.22	20.19	3.74	0.37	3.37	0.11
CSAC/KOH	4.91	21.80	4.46	0.82	3.64	0.23
CSAC/NH <sub>4</sub> OH	5.24	19.51	4.13	0.87	3.25	0.27
CSAC/NaOH	4.60	19.41	4.01	0.77	3.24	0.24
CSAC/MES at 15 mg/l	3.92	18.97	3.82	0.65	3.16	0.21
CSAC/MES at 50 mg/l	3.18	12.80	2.67	0.53	2.13	0.25
CSAC/MES at 152.8 mg/l	2.21	17.00	3.20	0.37	2.84	0.13

