

Comparison of carbon dioxide adsorption performance by adsorbents under low partial pressures

周豐志、余世宗、申永順

E-mail: 321426@mail.dyu.edu.tw

ABSTRACT

In this study an equipment for carbon dioxide adsorption was designed to evaluate the adsorption performance of four adsorbents: activated carbon, zeolite, silica xerogel and amine-grafted silica xerogel for possible application on the removal of carbon dioxide from indoor air. Silica xerogel and amine-grafted silica xerogel were synthesized in the lab. To simulate indoor conditions adsorption experiments were conducted with the initial carbon dioxide concentration between 1000 and 4000 ppm and temperature between 20 and 43 °C. Of the four adsorbents, amine-grafted silica xerogel has the highest specific amount of carbon dioxide adsorbed, and zeolite has almost not any amount on it. Higher removal percentage of carbon dioxide were obtained at 35 °C using amine-grafted silica xerogel than any other adsorbent within the test concentration range. The highest removal percentage of carbon dioxide were 34% with the initial carbon dioxide concentration of 1192 ppm. Fitting tests with Freundlich and Langmuir equations were demonstrated to established the relationship between the specific amount of carbon dioxide adsorbed and the equilibrium carbon dioxide concentration. It was shown that Langmuir equation is suitable for the adsorption process under low partial pressure. Based on the dynamic change of carbon dioxide concentration during the adsorption process in this equipment, a pseudo-second-order kinetic model was also demonstrated to be the suitable model.

Keywords : indoor carbon dioxide、Freundlich、Langmuir

Table of Contents

目錄 封面內頁 簽名頁 授權書.....	iii 中文摘要.....
iv 英文摘要.....	v 誌謝.....
vi 目錄.....	vii 圖目錄.....
x 表目錄.....	xiv
第一章 緒論.....	1 1.1 研究動機.....
1 1.2 研究目的與內容.....	2 第二章 文獻回顧.....4
2.1 二氧化碳物理與化學特性.....	6 2.2 目前國外管制室內二氧化碳濃度的指標.....7
室內二氧化碳累積原因之敘述.....	2.3 影響 9 2.4 二氧化碳處理技術.....10
.....11 2.6 吸附理論.....	2.5 吸附材料評估.....
.....16 2.6.2 物理吸附.....	14 2.6.1 化學吸附.....
.....17 2.7 等溫吸附模式.....	16 2.6.3 固體的活性衰退.....
整.....19 第三章 研究方法與步驟.....	18 2.8 吸附性材料吸附二氧化碳之文獻彙 24 3.1 吸附性材料選擇與實驗流程.....
24 3.2 實驗材料與設備.....	26 3.3 二氧化碳吸附試驗與裝置.....28
吸附模式計算.....	3.3.1.1 Pseudo – first – order kinetic model.....31
.....30 3.3.1.2 Pseudo – second – order kinetic model.....31	3.3.1.3 Intraparticle diffusion model.....32
.....31 3.3.1.4 Amine-grafted silica xerogel合成方法.....33	3.3.2 等溫吸附模式迴歸計算.....36
.....38 4.1 不同溫度與起始二氧化碳濃度沸石吸附過程二氧化碳濃度變化動態.....38	4.1 不同溫度與起始二氧化碳濃度沸石吸附過程二氧化碳濃度變化動態.....38
.....38 4.2 不同溫度與起始二氧化碳濃度活性碳吸附過程二氧化碳濃度變化動態.....38	4.2 不同溫度與起始二氧化碳濃度活性碳吸附過程二氧化碳濃度變化動態.....38
.....40 4.3 活性碳吸附二氧化碳平衡吸附量與平衡濃度之關係.....43	4.3 活性碳吸附二氧化碳平衡吸附量與平衡濃度之關係.....43
.....43 4.4 不同溫度與起始二氧化碳濃度silica xerogel吸附過程二氧化碳濃度變化動態.....44	4.4 不同溫度與起始二氧化碳濃度silica xerogel吸附過程二氧化碳濃度變化動態.....44
.....44 4.5 silica xerogel吸附二氧化碳平衡吸附量與平衡濃度之關係.....49	4.5 silica xerogel吸附二氧化碳平衡吸附量與平衡濃度之關係.....49
.....50 4.6 不同溫度與起始二氧化碳濃度amine-grafted silica xerogel二氧化碳濃度變化動態.....49	4.6 不同溫度與起始二氧化碳濃度amine-grafted silica xerogel二氧化碳濃度變化動態.....49
.....55 4.7 amine-grafted silica xerogel吸附二氧化碳平衡吸附量與平衡濃度之關係.....50	4.7 amine-grafted silica xerogel吸附二氧化碳平衡吸附量與平衡濃度之關係.....50
.....56 4.8 吸附動力比較.....55	4.8.1 Pseudo-first-order kinetic model.....56
.....58 4.8.2 Pseudo-second-order kinetic model.....56	4.8.2 Pseudo-second-order kinetic model.....58
.....58 4.8.3 Intraparticle diffusion model.....58	4.8.3 Intraparticle diffusion model.....58
.....65 4.9.1 Freundlich吸附模式.....61	4.9.1 Freundlich吸附模式.....65
.....65 4.9.2 Langmuir吸附模式.....61	4.9.2 Langmuir吸附模式.....65
.....68 4.10 amine-grafted silica xerogel吸附劑特性分析.....80	4.10 amine-grafted silica xerogel吸附劑特性分析.....76
.....80 5.1 結論與建議.....80	5.1 結論與建議.....81

.....81 參考文獻.....83 圖目錄 圖2.1大眾聚集的公共場所及辦公大樓室內二氣化碳濃度彙整圖.....
.....25 圖3.2實驗反應槽示意圖.....6 圖3.1研究流程.....
.....29 圖4.1以沸石吸附不同起始濃度的二氣化碳的吸附變化，溫度 26 0C.....39 圖4.2以沸石吸附二氣化碳其溫度與濕度，溫度26 0C相對濕度47.1(%).....
.....39 圖4.3以活性碳吸附不同起始濃度的二氣化碳的吸附變化，溫度2641 圖4.4以活性碳吸附不同起始濃度的二氣化碳的吸附變化，溫度36
.....41 圖4.5以活性碳吸附不同起始濃度的二氣化碳的吸附變化，溫度4541 圖4.6以活性碳吸附二氣化碳(1000 ppm)其溫度與濕度變化，溫度25 相對濕度62.5(%).....
.....42 圖4.7不同溫度活性碳吸附二氣化碳之平衡吸附量.....44 圖4.8以silica xerogel吸附不同起始濃度的二氣化碳的吸附變化，溫度4545 圖4.9以silica xerogel吸附二氣化碳其溫度與濕度變化，溫度2646 圖4.10以silica xerogel吸附二氣化碳其溫度與濕度變化，溫度3046 圖4.11以silica xerogel吸附二氣化碳其溫度與濕度變化，溫度3547 圖4.12 silica xerogel吸附二氣化碳(1000ppm)其溫度與濕度變化圖起始溫度20
.....47 圖4.13不同溫度silica xerogel吸附二氣化碳之平衡吸附量.....47 圖4.14 相對濕度39.4(%).....
.....49 圖4.14 amine-grafted silica xerogel吸附不同起始濃度的二氣化碳的吸附變化，溫度2051 圖4.15 amine-grafted silica xerogel吸附不同起始濃度的二氣化碳的吸附變化，溫度2
.....51 圖4.16 amine-grafted silica xerogel吸附不同起始濃度的二氣化碳的吸附變化，溫度3051 圖4.17 amine-grafted silica xerogel吸附不同起始濃度的二氣化碳的吸附變化，溫度35
.....52 圖4.18amine-grafted silica xerogel吸附二氣化碳(1000 ppm)其溫度與濕度變化圖起始溫度20 相對濕度39.4(%).....53 圖4.19不同溫度amine-grafted silica xerogel吸附二氣化碳之平衡吸附量.....
.....55 圖4.20活性碳在不同初始濃度下之Pseudo-first-order模式.....57 圖4.21 silica xerogel在不同初始濃度下之Pseudo-first-order模式.....
.....57 圖4.22 aminosilane-modified silica SBA-15在不同初始濃度下之Pseudo-first-order模式.....58 圖4.23活性碳在不同初始濃度下之Pseudo- second-order模式.....
.....59 圖4.24 silica xerogel在不同初始濃度下之Pseudo- second-order模式.....60 圖4.25 amine-grafted silica xerogel在不同初始濃度下之Intrapariticle diffusion model模式.....
.....60 圖4.26活性碳在不同初始濃度下之Intrapariticle diffusion model模式.....61 圖4.27silica xerogel在不同初始濃度下之Intrapariticle diffusion model模式.....
.....62 圖4.28 amine-grafted silica xerogel在不同初始濃度下之Intrapariticle diffusion model模式.....62 圖4.29在不同溫度下活性碳吸附二氣化碳之Freundlich模式.....
.....67 圖4.30在不同溫度下silica xerogel吸附二氣化碳之Freundlich模式.....67 圖4.31在不同溫度下amine-grafted silica xerogel吸附二氣化碳之Freundlich 模式.....
.....68 圖4.32在不同溫度下活性碳吸附二氣化碳之Langmuir模式.....68 圖4.33在不同溫度下silica xerogel吸附二氣化碳之Langmuir模式.....
.....70 圖4.34在不同溫度下amine-grafted silica xerogel吸附二氣化碳之Langmuir模式.....70 圖4.35活性碳吸附二氣化碳之平衡吸附模式與實驗數據比較.....
.....71 圖4.36 silica xerogel吸附二氣化碳之平衡吸附模式與實驗數據比較.....72 圖4.37amine-grafted silica xerogel吸附二氣化碳之平衡吸附模式與實驗數據比較.....
.....72 圖4.38 silica xerogel的EDX分析圖.....73 圖4.39 amine-grafted silica xerogel的EDX分析圖.....
.....78 圖4.40 amine-grafted silica xerogel的SEM分析圖.....78 圖4.41 silica xerogel的SEM分析圖.....
.....79 表目錄 表2.1 大眾聚集的公共場所及辦公大樓室內二氣化碳濃度彙整表.....79 表目錄 表2.1 大眾聚集的公共場所及辦公大樓室內二氣化碳濃度彙整表.....
.....5 表2.2 二氣化碳物理與化學特性整理表.....7 表2.3各國室內二氣化碳管制標準彙整表.....
.....8 表2.4香港行政區二氣化碳室內管制指標彙整表.....8 表2.4香港行政區二氣化碳室內管制指標彙整表.....
.....9 表2.5吸附性材料吸附二氣化碳處理研究整理表.....9 表2.5吸附性材料吸附二氣化碳處理研究整理表.....
.....20 表3.1 Testo-535儀器偵測極限.....20 表3.1 Testo-535儀器偵測極限.....
.....27 表3.2 HT-305儀器偵測特色儀器偵測極限.....27 表3.2 HT-305儀器偵測特色儀器偵測極限.....
.....28 表4.1活性碳吸附二氣化碳平衡吸附量與平衡濃度之結果.....28 表4.1活性碳吸附二氣化碳平衡吸附量與平衡濃度之結果.....
.....43 表4.2 silica xerogel吸附二氣化碳平衡吸附量與平衡濃度之結果.....48 表4.3 amine-grafted silica xerogel吸附二氣化碳平衡吸附量與平衡濃度之結果.....
.....54 表4.4在不同溫度下活性碳三種動力模式之比較.....54 表4.4在不同溫度下活性碳三種動力模式之比較.....
.....63 表4.5在不同溫度下silica xerogel三種動力模式之比較.....63 表4.5在不同溫度下silica xerogel三種動力模式之比較.....
.....63 表4.6在不同溫度下amine-grafted silica xerogel三種動力模式之比較.....64 表4.7不同溫度下活性碳吸附二氣化碳Langmuir參數比較.....
.....74 表4.8不同溫度下活性碳吸附二氣化碳Freundlich參數比較.....74 表4.9不同溫度下silica xerogel吸附二氣化碳Langmuir參數比較.....
.....74 表4.10不同溫度下silica xerogel吸附二氣化碳Freundlich參數比較.....74 表4.11不同溫度下amine-grafted silica xerogel吸附二氣化碳Langmuir參數比較.....
.....75 表4.12不同溫度下amine-grafted silica xerogel吸附二氣化碳Freundlich參數比較.....75 表4.12不同溫度下amine-grafted silica xerogel吸附二氣化碳Freundlich參數比較.....
.....76 表4.13 silica xerogel分析結果.....76 表4.13 silica xerogel分析結果.....
.....77 表4.14 amine-grafted silica xerogel分析結果.....77 表4.14 amine-grafted silica xerogel分析結果.....

REFERENCES

- 1.Bandar Fadhel , Milton Hearn , Alan Chaffee , “ CO₂ adsorption by PAMAM dendrimers: Significant effect of impregnation into SBA-15, ” , Microporous and Mesoporous Materials 123 (2009) 140 – 149。 2.D. Darmana, R.L.B. Henket, N.G. Deen*, J.A.M. Kuipers “ Detailed modelling of hydrodynamics, mass transfer and chemical reactionsin a bubble column using a discrete bubble model: Chemisorption of CO₂ intoNaOH solution, numerical and experimental study, ” , Chemical Engineering Science 62 (2007) 2556-2575。 3.Gregory P. Knowles, Seamus W. Delaney, and Alan L. Chaffee, “ Diethylenetriamine[propyl(silyl)]-Functionalized (DT) Mesoporous Silicas as CO₂ Adsorbents, ” Ind. Eng. Chem. Res. 2006, 45, 2626-2633 4.Hyun Tae Jang, YoonKook Park, Yong Sig Ko, Ji Yun Lee , Bhagiyalakshmi Margandan, “ Highly siliceous MCM-48 from rice husk ash for CO₂ adsorption, ” , International Journal of Greenhouse Gas Control 3 (2009) 545 – 549。 5.Helen Y. Huang and Ralph T. Yang, “ Amine-Grafted MCM-48 and Silica Xerogel as Superior Sorbents for Acidic Gas Removal from Natural Gas ” , Ind. Eng. Chem. Res. 2003, 42, 2427-2433。 6.Hyun-Kon Song, Kil Won Cho, Kun-Hong Lee “ Adsorption of carbon dioxide on the chemically modified silica adsorbents ” , Journal of Non-Crystalline Solids 242 (1998) 69-80。 7.Hall, K. R., Eagleton, L. C., Acrivos, A., and Vermeulen, T., Pore – and solid – diffusion kinetics in fixed – bed adsorption under constant – pattern conditions, 1996, Industrial and Engineering Chemistry Fundamentals, 5, 212 – 223。 8.Joey K. Parkerson, “ Atmospheric Carbon Dioxide Capture Technology ” , THE UNIVERSITY OF MISSISSIPPI SCHOOL OF ENGINEERING。 9.L.-C. Lora Huang and Huan-Cheng Chang* “ Adsorption and Immobilization of Cytochrome c onNanodiamonds ” , Langmuir 2004, 20, 5879-5884。 10.Moo-Hyun Kim , Ji-Hyeon Hwang, “ Performance prediction of a hybrid ventilation system in an apartment house ” , Energy and Buildings 41 (2009) 579 – 586。 11.Meyer, B., and K. Hermanns “ Reducing Indoor Air Formaldehyde concentration, ” J. APCA, vol.35,pp816-821,1985。 12.M. Santamouris, A. Synnefa, M. Asssimakopoulos, I. Livada, K. Pavlou,M. Papaglastra, N. Gaitani, D. Kolokotsa, V. Assimakopoulos “ Experimental investigation of the air flow and indoor carbon dioxide concentration in classrooms with intermittent natural ventilation ” , Energy and Buildings 40 (2008) 1833 – 1843。 13.Norihiro Hiyoshi 1, Katsunori Yogo , Tatsuaki Yashima, “ Adsorption characteristics of carbon dioxide onorganically functionalized SBA-15 ” , Microporous and Mesoporous Materials 84 (2005) 357 – 365。 14.Norihiro Hiyoshi, Katsunori Yogo, and Tatsuaki Yashima “ Adsorption of Carbon Dioxide on Amine Modified SBA-15 in the Presence of Water Vapor ” , Chemistry Letters Vol.33, No.5 (2004)。 15.Rahul Banerjee, Anh Phan,1 Bo Wang,Carolyn Knobler, Hiroyasu Furukawa, Michael O ’ Keeffe, Omar M. Yaghi “ High-Throughput Synthesis of Zeolitic Imidazolate Frameworks and Application to CO₂ Capture, ” , SCIENCE VOL 319 15 FEBRUARY 2008。 16.Ryosuke O. Suzuki, “ Calciothermic reduction of TiO₂ and in situ electrolysisof CaO in the molten CaCl₂ ” , Journal of Physics and Chemistry of Solids 66 (2005) 461 – 46。 17.Robert S. Franchi, Peter J. E. Harlick, and Abdelhamid Sayari, “ Applications of Pore-Expanded Mesoporous Silica. 2. Developmentof a High-Capacity, Water-Tolerant Adsorbent for CO₂ ” , Ind. Eng. Chem. Res. 2005, 44, 8007-8013。 18.Soo Chool Lee, Ho Jin Chae, Soo Jae Lee, Bo Yun Choi, ChangKeun Yi, Joong Beom Lee, Chong Kul Ryu, and Jae Chang Kim “ Development of Regenerable MgO-Based Sorbent Promoted with K₂CO₃ for CO₂ Capture at Low Temperatures, ” , Environ. Sci. Technol. 2008, 42, 2736 – 2741。 19.Sangil Kim, Junichi Ida, Vadim V. Gulians,* and Jerry Y. S. Lin, “ Tailoring Pore Properties of MCM-48 Silica for Selective Adsorption of CO₂ ” , J. Phys. Chem. B 2005, 109, 6287-6293。 20.V. Zelenak , D. Halamova, L. Gabrova, E. Bloch, P. Llewellyn, “ Amine-modified SBA-12 mesoporous silica for carbon dioxide capture:Effect of amine basicity on sorption properties, ” , Microporous and Mesoporous Materials 116 (2008) 358 – 364。 21.Wang Zuo-tang, Fu Zhen-kun, Zhang Bang-an, Wang Guo-xiong,Rudolph Victor, Huo Li-wen, “ Adsorption and desorption on coals for CO₂ sequestration, ” Mining Science and Technology 19 (2009) 0008 – 0013。 22.YAO Yadong, GUO Xiangli, KANG Yunqing, LI Xieji, CHEN Aizheng,YANG Weizhong, YIN Guangfu, “ Degradation of Residual Formaldehyde in Fabric by Photo-catalysis, ” Journal of Wuhan University of Technology-Mater. Vol.23 No.2 pp147-151,2008. 23.Yingjie Li,Changsui Zhao,Chengrui Qu,Lunbo Duan,Qingzhao Li1,Cai Liang, “ CO₂ Capture Using CaO Modified with Ethanol/Water Solution during CyclicCalcination/Carbonation, ” , Chem. Eng. Technol. 2008, 31, No. 2, 237 – 244。 24.Yuzuru Sakamoto, Kensuke Nagata, Katsunori Yogo, Koichi Yamada, “ Preparation and CO₂ separation properties of amine-modified mesoporous silica membranes ” , Microporous and Mesoporous Materials 101 (2007) 303 – 311。 25.張修齊,胡忠信,葉啟輝 “ 咖啡渣吸附銅離子之研究 ” ,大葉大學環境工程學系研究所碩士論文 , 2010. 26.戴川發,曾昭衡,李彥頤,王鳳瑾 “ 校園室內空氣品質自主管理推行之研究 ” ,中華民國環境工程學會 2009 空氣污染控制技術研討會。 27.謝祝欽,陳怡茜,郭柏成, “ 利用沸石觸媒轉化二二氧化碳之研究 ” ,中華民國環境工程學會 2009 空氣污染控制技術研討會。 28.邱瑞宇, 邱春惠, 賴東璟, 林佳昫, 許湘翎 “ 以VOC、CO、CO₂ 及總菌落數探討大班教室室內空氣品質 ” 中華民國環境工程學會 2009 空氣污染控制技術研討會。 29.財團法人中華民國消費者文教基金會,2000-2008。 30.陳治宇,鐘竺均, “ 校園環境中二二氧化碳濃度之調查與研究 ” ,醒吾學報第,31期,129-146,2008. 31.黃詣迪, 曾亮, “ 從裝修型態探討室內空氣品質之研究 以逢甲大學與台中商務旅館為例 ” 逢甲大學建築學系研究所碩士論文 , 2007. 32.林厚順、賴榮平, “ 公寓大廈地下停車場空氣品質之研究 ” 成功大學建築學系研究所碩士論文,2006. 33. “ 室內空氣汙染物健康風險評估及管制成本效益分析 ” 行政院環境保護署,2004。 34.江欣宸, 郭乃文, “ 台灣旅館客房室內空氣品質之評估-研究生 ” 台北護理學院旅遊健康學系研究所論文,2004. 35.蔡東翰,張仁瑞, “ 改進活性碳之再生研究 ” 中正大學化學工程研究所碩士論文,2003. 36.劉明翰,袁中新, “ 粉狀活性碳吸附氯相氯化汞之研究操作參數之探討及恆溫吸附模式之建立 ” 中山大學環境工程研究所碩士論文,2001. 37.李明澤, “ 非均勻系觸媒反應的理論與應用 ” ,復文書局, 1992出版。 38.陳海曙(1990), “ 空氣品質不佳之案例研究 ” ,中華民國建築學會第三屆建築學術研究發表會論文集 , 263-266. 39.杜逸虹, “ 物化原理 ” ,三民書局, 1982出版。 40.林俊一, “ 反應工程學 ” ,文京書局, 1982出版。