

Removal of Inorganic Ions in Single and Binary Mixed Electrolyte Systems by a Nanofiltration Membrane

陳柔閔、柯雅雯

E-mail: 9221183@mail.dyu.edu.tw

ABSTRACT

Abstract The research discuss the influence of concentration、pressure and crossflow to NF membrane separation mechanism and rejection effect at the condition of non-concentration polarization. Under the constant situation the rejection of Na⁺、Ca²⁺、Cl⁻、SO₄²⁻ are greater than 90 %、99 %、90 %、99 % , respectively. And the used NF-70 membrane is charge so the concentration of ions in single and binary mixed electrolyte systems effect the rejection rate and permeability more. The model of the research according Dey and others, based on the irreversibile thermodynamics approach and Donnan equilibrium theory have been used to derive an expression for Na⁺、Ca²⁺、Cl⁻、SO₄²⁻ rejection for single and binary mixed electrolyte systems. Key words : Nanofiltration ; binary mixed electrolyte ; concentration polarization

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REFERENCES

- 參考文獻? 環保署研究計畫, 「水源鹽化對加氯消毒副產物生成之影響與改善對策及研究」期末報告, 計畫主持人:蔣本基教授, 台灣大學環境工程學研究所, 計畫編號:EPA-89-U3J1-03-003, 民國89年7月。? 杜亮明, 「薄膜程序應用於淨水之探討」, 碩士論文, 國立台灣大學環境工程學研究所, 台北, 民國88年6月。? 國科會專題研究計畫, 「前處理對NF薄膜程序影響之研究」, 計畫主持人:葉宣顯教授, 成功大學環境工程學系, 計畫編號:NSC 88-2211-E-006-023, 民國88年7月。? 台灣省自來水公司專案計畫, 「澄清湖高級淨水處理模型廠試驗研究」(第一年), 計畫主持人:葉宣顯教授, 成功大學環境工程學系, 民國88年8月。? 高山鎮, 「薄膜阻塞控制之研究」, 碩士論文, 國立成功大學環境工程學研所, 民國89年6月。? 國科會專題研究計畫, 「NF薄膜程序應用於淨水處理時的阻塞行為與分離成效之研究」, 計畫主持人:柯雅雯教授, 大葉大學環境工程學系, 計畫編號:NSC, 民國90年8月。? 郭文正與曾添文, 「薄膜分離」, 高立圖書有限公司, 民國77年1月。? Ahn, K.-H., Song, K.-G., Cha, H.-Y. and Yeom, I.-T., "Removal of ions in nickel electroplating rinse water using low-pressure nanofiltration", *Desalination*, Vol.122, pp.77-84, 1999。? Alborzfar, M., Jonsson, G., and Gron, C., "Removal of natural organic matter from two types of humic ground waters by nanofiltration", *Water Research*, Vol.32, pp.2983-2994, 1998。? Allgeier, S. C. and Summers, R. S., "Evaluating NF for DBP control with the RBSMT", *Journal AWWA*, pp.87-98, 1995。? Anne, C. O., Trebouet, D., Jaouen, P. and Quemeneur, F., "Nanofiltration of seawater: fractionation of mono- and multi-valent cations", *Desalination*, Vol.140, pp.67-77, 2001。? AWWA Membrane Technology Research Committee, "Committee report: membrane processes", *Journal of American Water Works Association*, 1998。? Bohdziewicz, J., Bodzek, M. and Wasik, E., "The application of reverse osmosis and nanofiltration to the removal of nitrates from groundwater", *Desalination*, Vol.121, pp.139-147, 1999。? Bowen, W. R. and Mohammad, A. W., "A theoretical basis for specifying nanofiltration membranes — dye/salt/water streams", *Desalination*, Vol.117, pp.257-264, 1998。? Boussahel, R., Bouland, S., Moussaoui, K. M. and Montiel, A., "Removal of pesticide residues in water using the nanofiltration process", *Desalination*, Vol.132, pp.205-209, 2000。? van der Bruggen, B. and Vandecasteele, C., "Distillation vs. membrane filtration: overview of process evolutions in seawater desalination", *Desalination*, Vol.143, pp.207-218, 2002。? van der Bruggen, B., Schaep, J., Wilms, D. and Vandecasteele, C., "Influence of molecular size, polarity and charge on the retention of organic molecules by nanofiltration", *Journal of Membrane Science*, Vol.156, pp.29-41, 1999。? Bowen, W. R. and Mohammad, A. W., "A theoretical basis for specifying nanofiltration membranes-dye/salt/water streams", *Desalination*, Vol.117, pp.257-264, 1998。? Clark, S., et al., "Low-cost membrane filtration in offing", *Waterworld News*, Jan/Feb, pp.17-18, 1992。? Conlon, W. J. and McClellan, S. A., "Membrane softening: a treatment process comes of age", *Journal of American Water Works Association*, Vol.81, pp.47-51, 1989。? Deshmukh, S. S. and Amy, E. C., "Zeta potential of commercial RO membranes: influence of source water type and chemistry", *Desalination*, Vol.140, pp.87-95, 2001。? Dey, T. K., Ramachandran, V. and Misra, B. M., "Selectivity of anionic species in binary mixed electrolyte systems for nanofiltration membranes", *Desalination*, Vol.127, pp.165-175, 2000。? Donnan, F. G., "Theory of membrane equilibria and membrane potentials in the presence of non-dialysing electrolytes. A Contribution to physical-chemical physiology", *Journal of Membrane Science*, Vol.100, pp.45-55, 1995。? Escobar, I. C., Hong, S., and Randall, A. A., "Removal of assimilable organic carbon and biodegradable dissolved organic carbon by reverse osmosis and nanofiltration membranes", *Journal of Membrane Science*, Vol.175, pp.1-17, 2000。? Grib, H., Persin, M., Gavach, C., Piron, D. L., Sandeaux, J. and Mameri, N., "Amino acid retention with alumina nanofiltration membrane", *Journal of Membrane*

Science, Vol.172, pp.9-17, 2000. ? Graba, Y., Taha, S. Gondrexon, N., Cabon, J. and Dorange, G., " Mechanisms involved in cadmium salts transport through a nanofiltration membrane:characterization and distribution " , Journal of Membrane Science, Vol.168, pp.135-141, 2000. ? Kiso, Y., Nishimura, Y., Kitao, T., and Nishimura, K., " Rejection properties of non-phenylic pesticides with nanofiltration membranes " , Journal of Membrane Science, Vol.171, pp.229-237, 2000. ? Kiso, Y., Kon, T., Kitao, T., and Nishimura, K., " Rejection properties of alkyl phthalates with nanofiltration membranes " , Journal of Membrane Science, Vol.182, pp.205-214, 2001a. ? Kiso, Y., Sugiura, Y., Kitao, T., and Nishimura, K., " Effects of hydrophobicity and molecular size on rejection of aromatic pesticides with nanofiltration membranes " , Journal of Membrane Science, Vol.192, pp.1-10, 2001b. ? Kosutic, K. and Kunst, B., " Removal of organics from aqueous solutions by commercial RO and NF membranes of characterized porosities " , Desalination, Vol.142 pp.47-56, 2002. ? Koyuncu, I., " Reactive dye removal in dye/salt mixtures bynanofiltration membranes containing vinylsulphone dyes:Effects of feed concentration and cross flow velocity " , Desalination, Vol.143, pp.243-253, 2002. ? Lebrun, R. E. and Xu, Y., " Dynamic characterization of nanofiltration and reverse osmosis membranes " , Separation Science and Technology, Vol.34, pp.1629-1641, 1999. ? Marianne N., Lena, K., and Susana, L., " Fouling and retention of nanofiltration membranes " , Journal of Membrane Science, Vol.98, pp.249-262, 1995. ? Mehiguene, K. Gerba, Y., Taha, S., Gondrexon, N. and Dorange, G., " Influence of operating conditions on the retention of copper and cadmium in aqueous solutions by nanofiltration:experimental results and modeling " , Separation and Purification Technology, Vol.15, pp.181-187, 1999. ? Mehiguene, K., Taha, S., Gondrexon, N., Cabon, J. and Dorange, G., " Copper transfer modeling through a nanofiltration membrane in the case of ternary aqueous solution " , Desalination, Vol.127, pp.135-143, 2000. ? Pontalier, P.-Y., Ismail, A. and Ghou, M., " Mechanisms for the selective rejection of solutes in nanofiltration membranes " , Separation and Purification Technology, Vol.12, pp.175-181, 1997. ? Ratanatamskul, C., Urase, T. and Yamamoto, K., " Description of behavior in rejection of pollutants in ultra low pressure nanofiltration " , Wat. Sci. Tech., Vol.38, pp.453-462, 1998. ? Reiss, C. R., Taylor, J. S., and Robert, C., " Surface water treatment using nanofiltration pilot testing results and design considerations " , Desalination, Vol.125, pp.97-112, 1999. ? Shaalan, H. F., Sorour, M. H. and Tewfik, S. R., " Simulation and optimization of a membrane system for chromium recovery from tanning wastes " , Desalination, Vol.141, pp.315-324, 2001. ? Siddiqui, M., Amy, G., Ryan, J. and Odem, W., " Membranes for the contril of natural organic matter from surface wares " , Water Research, Vol.34, pp.3355-3370, 2000. ? Tang, C. and Chen, V., " Nanofiltration of textile wastewater for water reuse " , Desalination, Vol.143, pp.11-20, 2002. ? Tan, L. and Sudak, R. G., " Removing color from a groundwater source " , Journal of American Water Works Association, Vol.84, pp.79-87, 1992. ? Taylor, J., et al., " Applying membrane processes to groundwater sources for trihalomethane precursor control " , Journal of American Water Works Association, Vol.79, pp.72, 1987. ? Visser, T. J. K., Modise, S. J., Krieg, H. M. and Keizer, K., " The removal of acid sulphate pollution by nanofiltration " , Desalination, Vol.140, pp.79-86, 2001. ? Vrijenhoek, E. M. and Waypa, J. J., " Arsenic removal from drinking water by a loose nanofiltration membrane " , Desalination, Vol.130, pp.265-277, 2000. ? Xu, Y. and Lebrum, R.E., " Comparison of nanofiltration properties of two membranes using electrolyte and non-electrolyte solutes " , Desalination, Vol.122, pp.95-116, 1999. ? Xu, X. and Spencer, H. G., " Transport of electrolytes through a weak acid nanofiltration membrane:Effects of flux and crossflow velocity interpreted using a fine-porous membrane model " , Desalination, Vol.113, pp.85-93, 1997. ? Yang, J.-Z., " Transport properties in the nanofiltration of NaNO₃-water solutions with a week acid polyelectrolyte membrane " , Journal of Membrane Science, Vol.198, pp.145-148, 2002.