Optimal Lipase-Catalyzed Esterification of Hexyl Laurate by Continuous Packed Bed Reactor

楊正剛、謝淳仁

E-mail: 9509773@mail.dyu.edu.tw

ABSTRACT

Hexyl esters, a medium-chain ester, with a fruity flavor are primarily used in personal care formulations as an important emollient for cosmetic applications. Packed bed reactors, are the most frequently used reactors for immobilized lipases. They are best used continuously on a commercial scale so as to minimize labor and overhead costs. Enzymatic synthesis with a continuous packed bed reactor can either satisfy consumers ’ need for “ natural quality ” or lower production cost on industrial applications. The ability of lipase from Rhizomucor miehei (Lipase IM77) to catalyze the direct-esterification of 1-hexanol and lauric acid in organic solvent or in the absence of organic solvent by utilizing the packed bed reactor was investigated. Response surface methodology (RSM) and 3-level-3-factor fractional factorial design were employed to evaluate the effects of synthesis parameters, such as reaction temperature, mixture flow rate and substrate molar ratio (1-hexanol to lauric acid) on molar conversion (%) and production rate (μmol/min) of hexyl laurate by direct-esterification. The results showed that hexyl laurate was successfully synthesized by the continuous packed bed reactor.

Keywords : Esterification ; Hexyl esters ; Lipase ; Packed bed reactor ; Response surface methodology

Table of Contents

REFERENCES

【中文部分】 1. 李冠汝及蕭介夫。2001。脂肪?在生物產業之應用。生物產業。12: 181 – 186。 2. 洪偉章及陳榮秀。1996。化妝品科技概論。第5 – 8頁。高立圖書有限公司。台北，台灣。 3. 洪哲穎。1998。回應曲面品質工程技術。第267 – 269 頁。義守大學。高雄，台灣。 4. 高馥君。1992。反應曲面法在食品開發上的應用。食品工業月刊。 5. 陳國誠。1990。生物固定化技術與產業應用。第121 – 125。茂昌圖書有限公司。台北，台灣。 6. 陳芳怡 。2005。以反應曲面法探討左旋丁酸薄荷酯之最優化酵素合成。第14頁。大葉大學生物產業科技研究所碩士論文。彰化，台灣。 7. 張曉莉及黃世佑。1997。生物轉換法－有機溶劑中維持酵素活性之研究。化工。44: 71 – 84. 8. 許德發、鄭智交、楊濟華、李仰川、溫慧萍、許照紅。2002。化妝品概論。華格那企業有限公司。pp.6；pp.107；pp.120。 9. 郭竹芳及劉富雄。2004。Invertase固定於Fe3O4磁性奈米載體及其在磁穩性填充床(MSFBR)之研究。第39頁。雲林科技大學化學工程學系研究所碩士論文。雲林，台灣。 10. 張麗卿。1998。現代化妝品概論。高立圖書有限公司。pp.173；pp.174；pp.159。 11. 張淑微。2002。以反應曲面法研究酵素合成己醇酯類之最優化。第64頁。大葉大學食品工程研究所碩士論文。彰化，台灣。 12. 謝至涵。2005。以反應曲面反探討月桂酸己酯之最優化酵素合成。第8頁。大葉大學生物產業科技研究所碩士論文。彰化，台灣。 【英文部分】 1. Adachi, S., Nagae, K. and Matsuno, R. 1999. Lipase-catalyzed condensation of erythritol and medium-chain fatty acids in acetonitrile with low water content. J. Mol. Catal., B Enzym. 6: 21 – 27. 2. Akoh, C. C., Sellappan, S., Fomuso, L. B. and Yankah, V. V. 2002. Enzymatic synthesis of structured

lipids. Lipid Biotechnology, p. 433 – 460. Marcel Dekker Inc, New York, USA. 3. Akoh, C. C. 1998. Structured lipids. In " food lipids: chemistry, nutrition, and biotechnology. " (ed.), p. 701 – 727. Marcel Dekker Inc, New York, USA. 4. Akoh, C. C. and Fomuso, L. B. 2002. Lipase-catalyzed acidolysis of olive oil and caprylic acid in a bench-scale packed bed bioreactor. Food Res. Intern. 35: 15 – 21. 5. Burgess, K., Henderson, I. and Ho, K. K. 1992. Biocatalytic resolution of sulfinylakanoates: a facile route to optically active sulfoxides. J. Org. Chem. 57: 1290 – 1295. 6. Bourg-Garros, S., Razafindramoboa, N. and Pavia, A. A. 1998. Optimization of lipase-catalyzed synthesis of (z)-3-hexen-1-yl acetate by direct esterification in hexane and a solvent free medium. Enzyme Microb. Technol. 22: 240 – 245. 7. Bourg-Garros, S., Razafindramboa, N. and Pavia, A. A. 1997. Synthesis of (z)-3-hexen-1-yl butyrate in hexane and solvent-free medium using Rhizomucor miehei and Candida antarctica lipases. J. Am. Oil Chem. Soc. 11: 1471 – 1474. 8. Chang, S. W., Shaw, J. F. and Shieh, C. J. 2003. Optimization of enzymatically prepared hexyl butyrate by lipozyme IM-77. Food Technol. Biotech. 41: 237 – 242. 9. Chiang, W. D., Chang, S. W. and Shieh, C. J. 2003. Studies on the optimized lipase-catalyzed biosynthesis of cis-3-hexen-1-yl acetate in n-hexane. Process Biochem. 38: 1193 – 1199. 10. Chang, S. W., Shaw, J. F., Yang, K. H., Shih, I. L., Hsieh, C. H. and Shieh, C. J. 2005. Optimal lipase-catalyzed formation of hexyl laurate. Green Chem. 7: 547 – 551. 11. Chen, J., Kimura, Y. and Adachi, S. 2005. Continuous synthesis of 6-O-linoleoyl hexose using a packed-bed reactor system with immobilized lipase. Biochem. Eng. J. 22: 145 – 149. 12. Carvalho, C. M. L., Serralheiro, M. L. M., Cabral, J. M. S. and Aires-Barros, M.R. 1997. Application of factorial design to the study of transesterification reactions using cutinase in AOT-reversed micelles. Enzyme Microb. Technol. 21: 117 – 123. 13. Costodes, T. V. C. and Lewis, A. E. 2006. Reactive crystallization of nickel hydroxy-carbonate in fluidized-bed reactor: fines production and column design. Chem. Eng. Sci. 61: 1377 – 1385. 14. Dermirel, D. and Mutlu, M. 2005. Performance of immobilized pectinex ultra SP-L on magnetic duolite-polystyrene composite particles. part II: a magnetic fluidized bed reactor study. J. Food Eng. 70: 1 – 6. 15. Dossat, V., Combes, D. and Marty, A. 1999. Continuous enzymatic transesterification of high oleic sunflower oil in a packed bed reactor: influence of the glycerol production. Enzyme Microb. Technol. 25: 194 – 200. 16. Dordick, J.S. 1989. Enzymatic catalysis in monophasic organic solvents. Enzyme Microb. Technol. 11: 194 – 221. 17. Faber, K. 1992. Biotransformation in organic chemistry. p. 2 – 4. Pringer-erlag. Germany. 18. Frings, K., Koch, M. and Hartmeier, W. 1999. Kinetic resolution of 1-phenyl ethanol with high enantioselectivity with native and immobilized lipase in organic solvents. Enzyme Microb. Technol. 25: 303 – 309. 19. Fumoso, L. B. and Akoh, C. C. 2002. Lipase-catalyzed acidolysis of olive oil and caprylic acid in a bench-scale packed bed bioreactor. Food Res. Intern. 35: 15 – 21. 20. Goddard, R., Bosley, J. and Al-Duri, B. 2000. Esterification of oleic acid and ethanol in plug flow (packed bed) reactor under supercritical conditions investigation of kinetics. J. Supercrit. Fluids. 18: 121 – 130. 21. Giorno, L., Molinari, R., Natoli, M. and Drioli, E. 1997. Hydrolysis and regioselective transesterification catalyzed by immobilized lipase in membrane reactor. J. Memb. Sci. 125: 177 – 187. 22. Hazarika, S., Goswami, P., Dutta, N. N. and Hazarika, A. K. 2002. Ethyl oleate synthesis by Porcine pancreatic lipase in organic solvents. Chem. Eng. J. 85: 61 – 68. 23. Ivancic, M., Santec, B., Novak, S. and Maric, V. 2004. Fermentative bioconversion in a horizontal rotating tubular bioreactor. Process Biochem. 39: 995 – 1000. 24. Jones, J. B. 1986. Enzymes in organic synthesis. Tetrahedron. 42: 3351 – 3403. 25. Jeong, S., Hwang, B. Y., Kem, J. and Kem, B. G. 2000. Lipase-catalyzed reaction in the packed-bed reactor with continuous extraction column to overcome a product inhibition. J. Mol. Catal., B Enzym. 10: 597 – 604. 26. Kaewthong, W., Sirisansaneeyakul, S., Prasertsan, P. and H-Kittikun, A. 2005. Continuous production of monoacylglycerols by glycerolysis of palm olein with immobilized lipase. Process Biochem. 40: 1525 – 1530. 27. Liu, Y. Y., Xu, J. J., Wu, H. Y. and Shen, D. 2004. Integration of purification with immobilization of Candida rugosa lipase for kinetic resolution of racemic ketoprofen. J. Biotechnol. 110: 209 – 217. 28. Laane, C., Boneren, S., Vos, K. and Veeger, C. 1987. Rules for optimizations of biocatalysis in organic solvents. Biotechnol. Bioeng. 30: 81 – 87. 29. Long, W. S., Kamaruddin, A. and Bhatia, S. 2005. Chiral resolution of racemic ibuprofen ester in an enzymatic membrane reactor. J. Memb. Sci. 247: 185 – 200. 30. Meijer, E. M., Boesten, W. H. J., Schoemaker, H. E. and Balken, J. A. M. V. 1985. Biocatalysts in organic synthesis. p. 135 – 156. Elsever. Amsterdam, Dutch. 31. Myers, R. H. and Montgomery, D. C. 2002. Response surface methodology: process and product optimization using designed experiments. (2nd Eds). John Wiley and Sons. New York, USA. 32. Nielsen, N. S., Yang, T., Xu, X. and Jacobsen, C. 2005. Production and oxidative stability of a human milk fat substitute produced from lard by enzyme technology in a pilot packed-bed reactor. Food Chem. 94: 53 – 60. 33. Oliveira, A. C., Rosa, M. F., Aires-Barros, M. R. and Cabral, J. M. S. 2000. Enzymatic esterification of ethanol by an immobilized Rhizomucor miehei lipase in a perforated rotating disc bioreactor. Enzyme Microb. Technol. 26: 446 – 450. 34. Piao, J., Kobayashi, T., Adachi, S., Nakanishi, K. and Matsuno, R. 2004. Continuous synthesis of lauroyl or oleoyl erythritol by a packed-bed reactor with an immobilized lipase. Process Biochem. 39: 681 – 686. 35. Ronne, T. H., Yang, T., Mu, H., Jacobsen, C. and Xu, X. 2005. Enzymatic interesterification of butter fat with rapeseed oil in a continuous packed bed reactor. J. Agric. Food Chem. 53: 5617 – 5624. 36. Shieh, C. J., Akoh, C. C. and Yee, L. N. 1996. Optimized enzymatic synthesis of geranyl butyrate with lipase AY from Candida rugosa. Biotechnol. Bioeng. 51: 371 – 374. 37. Sanchez, A., Rio del, J. L., Valero, F., Lafuente, J., Faus, I. and Sola, C. 2000. Continuous enantioselective esterification of trans-2-phenyl-cyclohexanol using a new Candida rugosa lipase in a packed bed bioreactor. J. Biotechnol. 84: 1 – 12. 38. Shieh, C. J. and Chang, S. W. 2001. Optimized synthesis of lipase-catalyzed hexyl acetate in n-hexane by response surface methodology. J. Agric. Food Chem. 49: 1203 – 1207. 39. Shaw, J. F., Chang, S. W., Liao, H. F. and Shieh, C. J. 2003. Lipase-catalyzed biosynthesis of hexyl butyrate by direct esterification: optimization by response surface methodology. J. Sci. Food Agric. 83: 1525 – 1530. 40. Sanchez, A., Valero, F., Lafuente, J. and Sola, C. 2000. Highly enantioselective esterification of racemic ibuprofen in a packed bed reactor using immobilized Rhizomucor miehei lipase. Enzyme Microb. Technol. 27: 157 – 166. 41. Thomson, D. 1982. Response surface experimentation. J. Food Process. Pres. 6: 155 – 188. 42. Villeeuve, P. and Foglia, T. A. 1997. Lipase specificities: potential application in lipid bioconversions. Inform. 8: 641 – 651. 43. Watanabe, T., Sugiura, M., Sato,

M., Yamada, N. and Nakanishi, K. 2005. Diacylglycerol production in a packed bed bioreactor. Process Biochem. 40: 637 – 643. 44. Wandrey, C. and Wichman, R. 1987. Production of L-amino acids in the membrane reactor. Biotechnology. 1: 85 – 92. 45. Xiao, Y. M., Wu, Q., Cai, Y. and Lin, X. F. 2005. Ultrasound accelerated enzymatic synthesis of sugar esters in non-aqueous solvents. Carbohydr. Res. 340: 2097 – 2103. 46. Xi, W. W. and Xu, J. H. 2005. Preparation of enantiopure (s)-ketoprofen by immobilized Candida rugosa lipase in packed bed reactor. Process Biochem. 40: 2161 – 2166. 47. Yu, Z. R., Chang, S. W., Wang, H. Y. and Shieh, C. J. 2003. Study on synthesis parameters of lipase-catalyzed hexyl acetate in supercritical CO2 by response surface methodology. J. Am. Oil Chem. Soc. 80: 139 – 144. 48. Yang, T., Rebsdore, M., Engelrud, U. and Xu, X. 2005. Enzymatic production of monoacylglycerols containing polyunsaturated fatty acids through an efficient glycerolysis system. J. Agric. Food Chem. 53: 1475 – 1481.