Optimization of Enzymatic Synthesis of Octyl Hydroxyphenylpropionate by Response Surface Methodology

凌逸凡、謝淳仁

E-mail: 9318514@mail.dyu.edu.tw

ABSTRACT

Phenolic acids are widely known as efficient antioxidants in biological systems. Generally, the solubility of natural antioxidants might be a restriction to the practical applications in hydrophobic media. Therefore, the esterification of phenolic acid with alcohol can be a tool to alter physical properties like solubility and activity of the lipophilic antioxidants in oil based formulae and emulsions. Phenols can be converted into esters by esterification with acid chlorides or acid anhydrides; however, these routes do not meet the requirements necessary for food applications. Recently the esterification of organic acids via enzymatic routes has been successfully reported in some studies. The ability for immobilized lipase Candida antarctica (Novozyme?435) to catalyze the direct esterification of p-hydroxyphenylpropionic acid and octanol was investigated in this study. Response surface methodology (RSM) and 5-level-4-factor central composite rotatable design (CCRD) were employed to evaluate the effects of synthesis parameters, such as reaction time (24?72 h), temperature (25?65 ?C), enzyme amount (10?50%, w/w), and pH memory (pH 5?9) on percentage molar conversion of phenolic acid ester. Reaction time, temperature and enzyme amount were the most important variables. Based on ridge max analysis, the optimum synthesis conditions with 96% molar conversion were: reaction time 58 h, temperature 53 ?C, enzyme amount 38%, and pH memory 7.

Keywords : phenolic ; direct esterification ; lipase ; optimization ; response surface methodology

Table of Contents

REFERENCES

參考文獻 1. 朱燕華。2000。植物類機能性成分介紹。食品工業月刊。32: 48—52。 2. 江文德。1999。脂肪胺在油脂加工上的應用。食品工業月刊。31: 10—19。 3. 李昌憲、洪哲穎及熊光濱。1992。利用反應曲面法進行以 Streptococcus faecalis 生產酪胺酸脫羧胺之培養基最適化研究。中國農業化學會誌。30: 264—272。 4. 李根永和李孟修。1998。 Corynebacterium glutamicum 在高濃度鹽份培養基脯胺酸發酵之研究。中國農業化學會誌。36: 57—64。 5. 林志城和孫璐西。1995。食品添加物。食品化學/張為憲等編著。華香園出版社。台北市。 6. 拱玉郎。1997。天然抗氧化劑發展近況。食品工業月刊。29: 29—35。 7. 郁凱衡。1999。酚類抗氧化劑(上)－合成類。食品資訊。157: 34—38。 8. 高馥君。1992。反應曲面法在食品開發上的應用。食品工業月刊。24: 32—41。 9. 張基郁和張為憲。1995。酵素。食品化學/張為憲等編著。華香園出版社。台北市。 10. 張淑微。2002。以反應曲面法研究酵素合成己醇酯類之最優化。大葉大學食品工程研究所碩士論文。 11. 張曉莉和黃世佑。1997。生物轉換法－有機溶劑中維持酵素活之研究。化工。44: 71—84。 12. 陳如茵。2002。抗氧化成分對動脈粥樣硬化影響的機制。食品工業月刊。34: 34—42。 13. 陳昭雄和孫璐西。1995。脂質。食品化學/張為憲等編著。華香園出版社。台北市。 14. 楊海明。1986。脂解?在油脂交酯化上的應用。食品工業月刊。18: 34—43。 15. 劉冠汝。1996。應用脂肪?在有機溶劑與超臨界流體中合成高價產品。國立海洋大學水產食品科學研究所博士論文。 16. 賴永沛。1999。小籽立大功－葡萄籽抽出物。食品資訊。162: 20—23。 17. Arouma, O. I. 1994. Nutrition and health aspects of free radicals and antioxidants. Food Chem. Toxic. 32: 671—683. 18. Ballesteros, A., Bornscheuer, U., Capewell, A., Combes, D., Condoret, J. S., Koening, K., and Kolisis, F. N. 1995. Enzymes in non-conventional phases. Biocatal. Biotransfor. 13: 1—42. 19. Bowman, L. and Geiger, E. 1984. Optimization of fermentation conditions of alcohol production. Biotechnol. Bioeng. 26: 1492—1497. 20. Box, G. E. P. and Wilson, K. B. 1951. On the experimental attainment optimum conditions. J. Roy. Statist. Soc. B13: 1—45. 21. Box, G. E. P., Hunter, W., and Hunter, J. S. 1978. Statistics for experimenters. John Wiley and Sons, New York. 22. Branen, A. L. 1975. Toxicology and biochemistry of BHA and BHT. J. Am. Oil Chem. Soc. 52: 59—65. 23. Buisman, G. J. H., Helteren, C. v., Kramer, G. F. H., Veldsink, J. W., Derksen, J. T. P., and Cuperus, F. P. 1998. Enzymatic esterifications of functionalized phenols for the synthesis of lipophilic antioxidants. Biotechnol. Lett. 20: 131—136. 24. Chalas, J., Claise, C., Edeas, M., Messaoudi, C., Vergnes, L., Abella, A., and Lindenbaum, A. 2001. Effect of ethyl esterification of phenolic acids on low-density lipoprotein oxidation. Biomed. Pharmacother. 55: 54—60. 25. Chen, Q.Y., Shi, H., and Ho, C. T. 1992. Effects of rosemary extracts and major constituents on lipid oxidation and soybean lipoxygenase activity. J. Am. Oil Chem. Soc. 69: 999—1002. 26. Chen, W. K., Tsai, C. F., Liao, P. H., Kuo, S. C, and Lee, Y. J. 1999. Synthesis of caffeic acid esters as antioxidants by esterification via acyl chlorides. Chin. Pharm. J. 51: 271—278. 27. Cheynier, V., Feinberg, M., Chararas, C., and Ducauze, C. 1983. Application of response surface methodology to evaluation of bioconversion. Appl. Environ. Microbiol. 45: 634—639. 28. Chimi, H., Cillard, J., Cillard, P., and Rahmani, M. 1991. Peroxyl and hydroxyl radical scavenging activity of some natural phenolic antioxidants. J. Am. Oil Chem. Soc. 68: 307—312. 29. Pirozzi, D. and Greco Jr., G. 2004. Activity and stability of lipases in the synthesis of butyl lactate. Enzyme Microb. Tech. 34: 94—100. 30. Dziezak, J. D. 1986. Antioxidants: the ultimate answer to oxidation. Food Technologist 40: 94—102. 31. Frankel, E. N., Kanner, J., German, J. B., Parks, E., and Kinsella, J. E. 1993. Inhibition of oxidation of human low-density lipoprotein by phenolic substances in red wine. Lancet 341: 454—457. 32. Graf, E. 1992. Antioxidant potential of ferulic acid. Free Radic. Biol. Med. 13: 435—448. 33. Gutman, A. L., Shkolnik, E., and Shapira, M. 1992. A convenient method for enzymatic benzyl-alkyl transesterification under mild neutral conditions. Tetrahedron 48: 8775—8780. 34. Guyot, B., Bosquette, B., Pina, M., and Graille, J. 1997. Esterification of phenolic acids from green coffee with an immobilized lipase from Candida antarctica in solvent-free medium. Biotechnol. Lett. 19: 529—532. 35. Himmelblau, D. M. 1970. Process analysis by statistical methods. John Wiley and Sons, New York. 36. Ito, N., Fukushima, S., and Tsuda, H. 1985. Carcinogenicity and modification of the carcinogenic response by BHA, BHT, and other antioxidants. Crit. Rev. Toxicol. 15: 109—150. 37. Jaeger, K. E., Ransac, S., Dijkstra, B. W., Colson, C., and Misset, O. 1994. Bacterial lipases. FEMS Microbiol. Rev. 15: 29—63. 38. Jogiekar, A. M. and May, A. T. 1987. Product excellence through design of experiments. Cereal Food World 32: 857—868. 39. John, V. T. and Abraham, G. 1991. Lipase catalysis and its applications. In Biocatalysis for Industry. By Dodrick, J. S. Plenum Press, New York. 40. Kajiyama, T. and Ohkatsu, Y. 2001. Effect of para-substituents of phenolic antioxidants. Polym. Degrad. Stabil. 71: 445—452. 41. Karra-Chaabouni, M., Pulvin, S., Touraud, D., and Thomas, D. 1998. Parameters affecting the synthesis of granyl butyrate by esterase 30,000 from Mucor miehei. J. Am. Oil Chem. Soc. 75: 1201—1206. 42. Kittss, D. 1997. An evaluation of the multiple effects of the antioxidant vitamins. Trends Food Sci. Tech. 8: 198—203. 43. Kochnar, S. P. and Rossel, J. B. 1990. Detection, estimation and evaluation of antioxidants in food systems. In Food antioxidant. By Huson, B. J. F. Elsevier Science Publishers, New York. 44. Kris-Etherton, P. M. and Nicolosi, R. J. 1995 Trans fatty acid and coronary heart disease risk. ILSI press, Washington DC. 45. Lee, Y. J., Liao, P. H., Chen, W. K., and Yang, C. C. 2000. Preferential cytotoxicity of caffeic acid phenethyl ester analogues on oral cancer cells. Cancer Lett. 153: 51—56. 46. Liu, X. Y., Guo, F. L., Wu, L. M., and Liu, Z. L. 1996. Remarkable enhancement of antioxidant activity of Vitamin C in an artificial bilayer by making it lipo-soluble. Chem. Phys. Lipids 62: 345—350. 47. Marangoni, A. G. and Rousseau, D. 1998. The influence of chemical interesterification on physicochemical properties of complex fat systems 1. Melting and crystallization. J. Am. Oil Chem. Soc. 75: 1265—1271. 48. Marinova, E. M. and Yanishlieva, N. V. 1994. Effect of lipid unsaturation on the antioxidative activity of some phenolic acid. J. Am. Oil Chem. Soc. 71: 427—434. 49. Massaeli, H., Sobrattee, S., and Pierce, G. N. 1999. The importance of lipid solubility in antioxidants and free radical generating systems for determining lipoprotein peroxidation. Free Radic. Biol. Med. 26: 1524—1530. 50. Maugard, T., Boulonne, M., Rejasse, B., and Legoy, M. D. 2001. Enzymatic synthesis of water-soluble derivatives of salicylic acid in organic media. Biotechnol. Lett. 23: 989—993. 51. Montgomery, D. C. 1984. Design and analysis of experiments. John Wiley and Sons, New York. 52. Moresi, M., Colicchio, A., and Sansovini, F. 1980. Optimization of whey fermentation in a jar fermenter. Eur. J. Appl. Microbiol. Biotechnol. 9: 173—183. 53. Moure, A., Cruz, J. M., Franco, D., Dominguez, J. M., Sineiro, J., Dominguez, H., and Nunez, M. J. 2001. Natural

antioxidants from residual sources. Food Chem. 72: 145—171. 54. Nardini, M., D''Alessandro, N., Tomassi, G., Gentili, V., Di Felice, M., and Scaccini, C. 1995. Inhibition of human low-density lipoprotein oxidation by caffeic acid and other hydroxycinnamic acid derivatives. Free Radic. Biol. Med. 19: 541—552. 55. Natella, F., Nardini, M., Felice, M. D., and Scaccini, C. 1999. Benzoic and cinnamic acid derivatives as antioxidants: structure-activity relation. J. Agr. Food Chem. 47: 1453-1459. 56. Novozymes Co. 2001. Product Sheet. https://www.novozymes.com 57. Ohta, T., Nakano, T., Egashira, Y., and Sanada, H. 1997. Antioxidant activity of ferulic acid beta-glucuronide in the LDL oxidation system. Biosci. Biothchnol. Biochem. 61: 1942—1943. 58. Pokorny, J. 1987. Major factors affecting the antioxidant of lipids. In Autoxidation of unsaturated lipids. By Chan, H. Academic Press, London. 59. Pratte, D. E. and Hudson, B. J. F. 1990. Natural antioxidants not exploited commercially. In Food Antioxidants. By Hudson, B. J. F. Elsevier Science Publishers, New York. 60. Retsky, K. L., Freeman, M. W., and Frei, B. 1993. Ascorbic acid oxidation product(s) protect human low density lipoprotein against atherogenic modification: anti-rather than prooxidant activity of vitamin C in the presence of transition metal ions. J. Biol. Chem. 268: 1304—1309. 61. Rice-Evans, C. A., Miller, N. J., and Paganga, G. 1997. Antioxidant properties of phenolic compounds. Trends Plant Sci. 2: 152—159. 62. Schuler, P. 1990. Natural antioxidants exploited commercially. In Food Antioxidants. By Hudson, B. J. F. Elsevier Science Publishers, New York. 63. Seriburi, V and Akoh, C. C. 1998. Enzymatic transesterification of triolein and stearic acid and solid fat content of their products. J. Am. Oil Chem. Soc. 75: 511—516. 64. Shahidi, F., Amarowicz, R., Yuehua, H., and Mahinda, W. 1997. Antioxidant activity of phenolic extracts of evening primrose (Oenothera biennis): a preliminary study. J. Food Lipids 4: 75—86. 65. Sherwin, E. R. 1990. Antioxidants. In Food antioxidants. By Branen, A. L., Davidson, P. M., and Salminen, S. Marcel Dekker Inc., New York. 66. Stamatis, H., Sereti, V., and Kolisis, F. N. 1999. Studies on the enzymatic synthesis of lipophilic derivatives of natural antioxidants. J. Am. Oil Chem. Soc. 76: 1505—1510. 67. Stamatis, H., Sereti, V., and Kolisis, F. N. 2001. Enzymatic synthesis of hydrophilic and hydrophobic derivatives of natural phenolic acids in organic media. J. Mol. Catal. B-Enzym. 11: 323—328. 68. Thomson, D. 1982. Response surface experimentation. J. Food Process. Pres. 6: 155—188. 69. Villeneuve, P. and Foglia, T. A. 1997. Lipase specificities: potential application in lipid bioconversions. INFORM 8: 640—651. 70. Wada, S. and Fang, X. 1992. The synergistic antioxidant effect of rosemary extract and ?-tocopherol in sardine oil model system and frozen-crushed fish meat. J. Food Process Preserv. 16: 263-274. 71. Wanasundara, U. N. and Shahidi, F. 1998. Antioxidant and pro-oxidant activity of green tea extracts in marine oils. Food Chem. 63: 335—342. 72. Yagi, K. and Ohishi, N. 1979. Action of ferulic acid and its derivatives as antioxidants. J. Nutr. Sci. Vitaminol. 25: 127—130. 73. Zertuche, L. and Zall, R. R. 1985. Optimizing alcohol production from whey using computer technology. Biotechnol. Bioeng. 27: 547—554.