Food Sci. Biotechnol. Vol. 14, No. 6, pp. 747 ~ 751 (2005)

Food Science and Biotechnology

Optimization of Pine Flavor Microencapsulation by Spray Drying

Shin-Jo Lee, Yang-Bong Lee*, Ji-Hyang Hong[1], Jong-Hoon Chung[1], Suk-Shin Kim[2], Won-Jong Lee[3] and Jungro Yoon[3]

Department of Food Science and Technology/Institute of Seafood Science, Pukyong National University, Busan 608-739, Korea
[1]Department of Biosystems & Biomaterials Science and Engineering, Seoul National University, Seoul 151-742, Korea
[2]Department of Food Science and Nutrition, Catholic University of Korea Bucheon, Gyeonggi 420-744, Korea
[3]Department of Food Science, Kangnung National University Kangneung, Gangwon 210-702, Korea

Abstract Microencapsulation of pine flavors was investigated to determine the optimum wall material and spray drying condition. β-Cyclodextrin, maltodextrin, and a 3:1 mixture of maltodextrin and gum arabic were evaluated as wall materials. The latter mixture was determined to be the best wall material based on dispersion capacity and flavor yield. Spray drying effectiveness was evaluated using a 3^3 fraction factorial design and statistical analysis. The optimum operation condition was an inlet air temperature of 175°C, inlet airflow rate of 0.65 m^3/min and atomizing pressure of 180 kPa, which resulted in a 93% flavor yield. The best particle shape observed by SEM was a round globular shape obtained under the above spray drying condition, whereas lower temperatures and higher inlet airflow rates resulted in initial and full collapses, respectively. The round globular shapes remained stable for at least one month.

Keywords: pine flavor, microencapsulation, spray drying, optimization

Introduction

Flavor microencapsulation is commonly used to protect flavors with a coating material to enhance flavor retention during food processing and storage. Spray drying is one of the most commonly used methods for flavor microencapsulation because it is easier and less expensive than other encapsulation processes. Spray drying is performed at a high temperature to instantly dry the water phase and encapsulate flavors into the wall materials, producing stable globular granules that protect the flavors from the environment. The effect of spray drying microencapsulation on flavor quality is dependent on several factors including wall and core materials, emulsion stability, operating condition of the spray dryer and others. Emulsion stability is affected by emulsion ratio, stickiness, homogenization condition, and types of wall and core materials. The operating parameters for the spray dryer affect the flavor quality of the final product. Important settings include inlet air temperature, outlet air temperature, inlet airflow rate, feed emulsion flow rate and atomizing pressure (1). However, the effects of operating conditions during spray drying on the quality of microencapsulated flavors have not been well evaluated.

Several polymers, including carbohydrates and proteins, have been developed for use as wall materials to extend the shelf life of microencapsulated flavors. The effects of wall materials on the microencapsulation of flavors have been studied by several investigators (2-5). The most commonly used wall materials for flavor microencapsulation are maltodextrin, gum arabic and cyclodextrin (especially, β-type). Wall materials can be categorized into two classes based on their mode of action: those that encapsulate or surround the flavor agents and those in which the flavors become imbedded. The advantages of using a mixture of wall materials, rather than a single one, were investigated by Kenyon *et al.* (6).

Pine flavor and its volatile compounds were the core materials utilized in this study. Pine flavors have been isolated by several extraction methods including simultaneous distillation and solvent extraction (SDE) (7), headspace analysis of purge and trap (8), microextraction with a fiber (9), and supercritical carbon dioxide extraction (10). The relative amounts of the volatile flavor compounds in pine extracts using different extraction methods were also previously reported. Supercritical extraction was previously determined to produce higher quality pine flavor than SDE (10). Lee *et al.* (8) investigated pine flavors extracted from sprouts, twigs and needles. Among more than 20 volatile flavor compounds identified in pine extracts, the major compounds were monoterpenes such as α-pinene, β-pinene, and limonene.

Fractional factorial experimental analysis on the factors of independent variables was used to investigate their effect on optimization. Therefore, the fractional factorial design related to a surface response method was applied to the optimization of food processing. Veglio *et al.* (11) studied the application of surface responses of organic materials to manganiferous ore leaching by using whey or lactose in a sulfuric acid solution. Halliwell *et al.* (12) employed microencapsulation techniques for the immobilization of oligonucleotides on glass surfaces. The objectives of the present study were to investigate the optimum wall materials among maltodextrin, cyclodextrin, and the mixture of maltodextrin and gum arabic by evaluating flavor yield and emulsion capacity after microencapsulation of the three major volatile compounds responsible for pine aroma and to determine the optimum spray drying condition using a 3^3 fractional factorial design, multiple regression and surface response analysis of microencapsulated

---

*Corresponding author: Tel: 82-51-620-6427; Fax: 82-51-622-9248
E-mail: yblee@pknu.ac.kr
Received April 26, 2005; accepted September 15, 2005

pine flavors extracted by supercritical extraction.

Materials and Methods

Determination of optimum wall material β-Cyclodextrin (Korea Food Materials, Kimhae, Korea), 12 DE maltodextrin (Sam Yang Genex, Seoul, Korea) and a 3:1 mixture of maltodextrin and gum arabic (Sam Yang Genex, Seoul, Korea) were tested to determine the best wall material. The core materials were α-pinene, β-pinene, and limonene in a pure chemical form of major compounds of pine aroma, which were purchased from Sigma-Aldrich (St. Louis, MO, USA). The ratio of this mixture for feeding emulsion was 45-75 (w/w) and 45:5 in water for wall material and pure chemicals, respectively. The mixture was homogenized at 22,000 rpm and spray dried (EYELA spray dryer SD-10000, Japan). The optimum wall material was determined by measuring flavor yield and emulsion capacity.

Encapsulation of pine flavor Natural pine flavors used as core materials were extracted by supercritical carbon dioxide extraction (10) at 200 bar for 5 min from parts of pine (*Pinus densiflora*) sprouts collected in October of 2000 from a in forest South Korea. The ratio of emulsion mixture was 50: 45: 5 of wall material, distilled water, and pine flavor, respectively. This emulsion mixture was homogenized at 22,000 rpm for 15 min.

The spray dryer (EYELA SD-10000, Japan) was operated at a feed flow rate of 3.5 mL/min at three independent values of inlet air temperature (155°C, 165°C, 175°C), inlet airflow rate (0.55 m^3/min, 0.65 m^3/min, 0.75 m^3/min) and atomizing pressure (160 kPa, 170 kPa, 180 kPa). The 3^3 factorial design for surface response is shown in Table 1, where total essential oil, surface essential oil, flavor yield and water content were the outcomes measured by the methods shown in the next section.

Determination of flavor yield Total oil content of the microencapsulated pine flavor powders was determined by using a Clevenger hydrodistillation apparatus (Prime Biochemical, Seoul, Korea). Fifteen grams of the microencapsulated powder was added to 150 mL distilled water and heated in a 130 ethylene glycol bath for 4 hr. Surface oil determination was done by using pentane in a Soxhlet apparatus. A two-gram sample of spray-dried powder was added to a Soxhlet thimble and surface oil was extracted for 5 hr.

The flavor yield of the microencapsulated powder was calculated by subtracting the surface oil content from the total oil content. The moisture content of the microencapsulated powder was determined by the toluene distillation method (Prime Biochemical, Seoul, Korea) in a hood. A 20 g spray-dried sample was added to 250 mL toluene and heated for 5 hr.

Particle shapes of the microencapsulated powder were observed in platinum-coated samples by SEM (scanning electron microscope, LEO Electron LEO-420, England).

Statistical analysis The 3^3 fraction factorial design shown in Table 1 was used for statistical analysis of surface responses, analysis of variance (ANOVA) table and multiple regression models. As shown in Table 2, ANOVA with SAS software (SAS, Cary, NC, USA) was used to investigate the effect of three independent variables on flavor yield: inlet air temperature, inlet airflow rate and atomizing pressure. The coefficients of regression models and coefficients of determination on flavor yield of spray drying were determined using SAS and are shown in Table 3. The optimum spray drying condition for obtaining the highest flavor yield was determined by SAS programs to determine the uncoded optimum point. Minitab was used for multiple regression and surface response surface analysis.

Results and Discussion

Optimization of wall material and stable feeding ratio for emulsion stability The effectiveness of maltodextrin, gum arabic and β-cyclodextrin as wall materials was evaluated by measuring the recovery yield of the oil, including α-pinene, β-pinene, and limonene, from the microencapsulated pine extracts. Maltodextrin of 12 dextrose equivalent (DE) with 53% distilled water had an optimal yield of 90%. Kenyon *et al.* (13) studied maltodextrin as a function of DE for use as a spray drying wall material. The optimum maltodextrin used for micro-

Table 1. 3^3 fraction factorial design for spray drying of pine flavors

Inlet air temperature (°C)	Inlet airflow rate (m^3/min)	Atomizing pressure (kPa)	Total oil (%)	Surface oil (%)	Flavor yield (%)	Moisture content (%)
155	0.55	160	55	9	46	7.5
155	0.55	180	57	13	44	7.5
155	0.55	200	62	16	46	8.0
155	0.65	160	70	4	66	7.0
155	0.65	180	72	6	66	7.0
155	0.65	200	77	7	70	7.5
155	0.75	160	75	4	71	5.5
155	0.75	180	67	5	62	5.0
155	0.75	200	53	6	47	6.0
165	0.55	160	79	0	79	4.5
165	0.55	180	85	0	85	4.0
165	0.55	200	83	1	83	5.0
165	0.65	160	86	0	86	3.5
165	0.65	180	93	0	93	3.0
165	0.65	200	84	0	84	3.0
165	0.75	160	83	0	83	3.5
165	0.75	180	91	0	91	3.0
165	0.75	200	87	0	87	3.0
175	0.55	160	85	0	85	3.5
175	0.55	180	91	0	91	3.0
175	0.55	200	82	0	82	4.0
175	0.65	160	87	0	87	3.5
175	0.65	180	95	0	95	3.0
175	0.65	200	90	0	90	3.5
175	0.75	160	75	0	75	3.0
175	0.75	180	87	0	87	3.0
175	0.75	200	83	0	83	3.5

encapsulation of orange peel oil was 10 DE. β-Cyclodextrin with 50% water showed the best yield of 94% in Kenyon's study, but had poor emulsion capacity. A maltodextrin-gum arabic blend (3:1, w/w) had an optimum point of 92 % at 50% water content. This 3:1 mixture ratio was tried for encapsulation of orange flavor by Reineccius *et al.* (14). Yield was optimized by using β-cyclodextrin as the wall material, but this polymer has less dispersibility. Therefore, when considering both emulsification and flavor yield, the optimum wall material for microencapsulation of pine flavor by spray drying was the mixture of maltodextrin and gum arabic (3:1, w/w). Thevenet (15) mentioned that using mixtures of wall materials for flavor microencapsulation had intercooperative advantages for encapsulation yield and emulsion capacity. Microencapsulation of pine aroma was also evaluated by using a spray drier to determine the optimum ratio of water, wall material and flavor oil in emulsions. We also evaluated the effect of water concentration on emulsion stability as a wall material variable. Optimal water concentration was 50% for both β-cyclodextrin and the maltodextrin-gum arabic 3:1 blend and 55% for maltodextrin alone. Higher ratios of water resulted in a higher moisture content of the final product and a loss of integrity of the granule, as seen in the collapsed shape. Therefore, the optimum wall material was maltodextrin-gum arabic (3:1, w/w) mixture. The optimal stable feeding emulsion ratio was 45% wall material, 5% pine flavor oil and 50% distilled water (Fig. 1). The distilled water ratio affected stability, particle shape, and also the dry-based yield of the spray dried flavor. Reineccius *et al.* (14) studied the effect of the water ratio on emulsion stability. They determined that 50% water with 44% maltodextrin-gum arabic and 6% orange oil resulted in the most stable and best flavor yield for microencapsulated orange flavors.

Optimization of the spray drying process The optimum spray drying conditions for flavor quality in microencapsulated pine flavors were investigated by fractional factorial design analysis. The variables were total oil, surface oil, moisture content and flavor yield as affected by inlet air temperature, inlet airflow rate and atomizing pressure (Table 1). Flavor yield varied from 44-71% at an inlet air temperature of 155°C, 79-91% at 165 °C, and 82-95% at 175°C. The low flavor yield at 155°C was due to the dried powders containing a higher moisture content (5.0-8.0%) and higher surface oil content (4-16%). From the result of Table 1, the ANOVA table presenting the effect on flavor yield of three independent variables, inlet air temperature, inlet airflow rate and atomizing pressure, was obtained and is shown in Table 2. The significant factor on the treatment effects was inlet air temperature at the confidence level of 99.9%. It was concluded, therefore, that low inlet air temperature results in poor quality microencapsulation of flavors by spray drying.

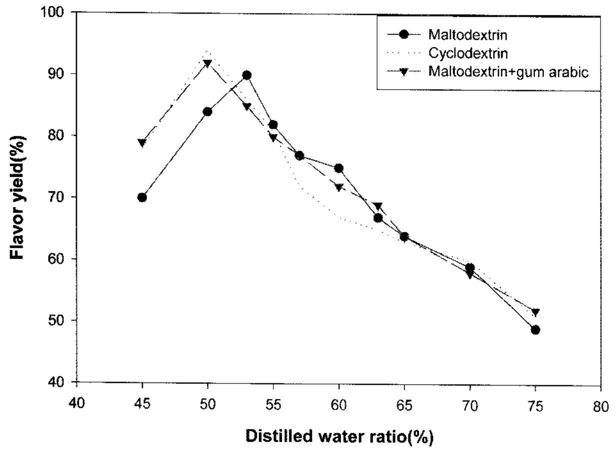


Fig. 1. Flavor yield as a distilled water ratio in feed of different wall materials in standard operation (limonene core material, 165°C, 95°C inlet and outlet air temp., 0.65 m^3/min inlet airflow rate, 180 kPa atomizing pressure, 3.5 mL/min feed flow rate)

Based on the results of Table 2, the coefficients of regression models and coefficients of determination on flavor yield of spray drying were determined using SAS and are shown in Table 3. The regression coefficient of the most fitted model of y_4 was 0.90. The following model equation was made with the variables of inlet air temperature (X_1) and inlet airflow rate (X_2):

$$y_4 = - 4758 + 49.8 X_1 + 1896 X_2 - 0.137 X_1^2 - 834 X_2^2 - 4.77 X_1X_2.$$

From the above formula, the relationships between the two variables were drawn with a Maple program to show that the maximum flavor yields at 165°C and 175°C were similar (Fig. 2). However, the flavor yield at 175°C decreased more sharply from the maximum value because of the shorter drying time at the higher temperature, which was due to reduced flavor content rather than decreased dried powder yield. Flavor yield was most affected by inlet air temperature followed by inlet airflow rate. Reineccius *et al.* (14) operated a spray dryer over an inlet air temperature range of 160°C and 200°C to investigate the effects of inlet air temperature on the microencapsulation of orange oil and determined that the optimal inlet air temperature was 175°C. Mongenot *et al.* (16) operated a spray-drier at 175°C for microencapsulation of cheese aroma.

The effect of airflow rate in spray drying on favor yield is shown in Fig. 3 from the formula of y_4. Inlet airflow rates of 0.55, 0.65 and 0.75 m^3/min produced flavor yields of 44-91%, 66-95% and 47-87%, respectively. At the

Table 2. Analysis of variance results

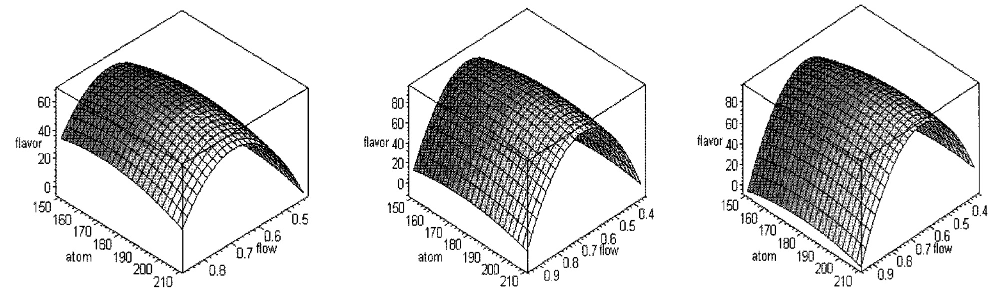
Source of variance	SS	df	MS	F	$F_{1,19,\,0.05}$	$F_{1,19,\,0.01}$
X_1[a)]	3611.0	1	3611.0	32.3**	4.3	8.2
X_2[b)]	119.0	1	119.0	1.1		
X_3[c)]	0.8	1	0.8	0.0		
X_1*X_2	272.5	1	272.5	2.4		
X_1*X_3	58.8	1	58.8	0.5		
X_2*X_3	11.4	1	11.4	0.1		
X_1*X_2*X_3	0.1	1	0.1	0		
Error	2126.3	19	111.9	1.0		
Total	6200.2	26				

[a)] X_1 : Inlet air temperature, [b)] X_2 : Inlet airflow rate, [c)] X_3 : Atomizing pressure

Table 3. Regression model for flavor retention during spray drying

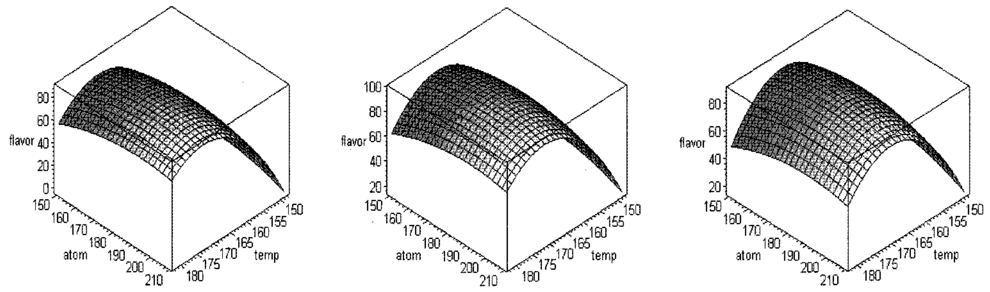
Regression model	Coefficient of determination
$y_1^{a)} = - 157 + 1.42X_1^{b)}$	0.58
$y_2 = - 3883 + 46.7X_1 - 0.137X_1^2$	0.77
$y_3 = - 4247 + 46.7X_1 + 1110X_2^{c)} - 0.137X_1^2 - 834X_2^2$	0.85
$y_4 = - 4758 + 49.8X_1 + 1896X_2 - 0.137X_1^2 - 834X_2^2 - 4.77X_1X_2$	0.90
$y_5 = - 4758 + 47.8X_1 + 1984X_2 - 2.33X_3^{d)} - 0.137X_1^2 - 834X_2^2 - 0.0107X_3^2 - 4.77X_1X_2 + 0.0111X_1X_3 - 0.488X_2X_3$	0.92

$y_{1,2,3,4,5}^{a)}$: Flavor yields for each models, $X_1^{b)}$: Inlet air temperature, $X_2^{c)}$: Inlet airflow rate, $X_3^{d)}$: Atomizing pressure



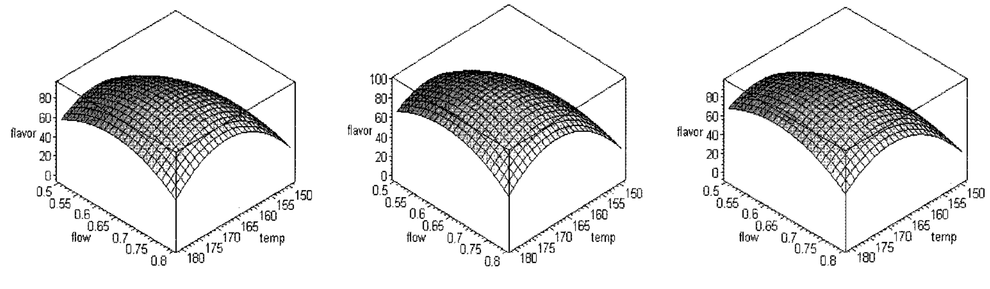
(a)155℃ (b)165℃ (c)175℃

Fig. 2. Flavor yield as a relationship between atomizing pressure and inlet airflow rate at inlet air temperatures of 155°C, 165°C and 175°C.



(a) 0.55 m^3/min (b) 0.65 m^3/min (c) 0.75 m^3/min

Fig. 3. Flavor yield as a relationship between inlet air temperature and atomizing pressure at inlet airflow rates of 0.55 m^3/min, 0.65 m^3/min and 0.75 m^3/min.



(a) 160 kPa (b) 180 kPa (c) 200 kPa

Fig. 4. Flavor yield as a relationship between inlet air temperature and inlet airflow rate at atomizing pressures of 160 kPa, 180 kPa and 200 kPa.

lowest airflow rate, the surface oil was increased and the flavor yield was decreased, because of inadequate drying. At the highest inlet airflow rate, the flavor yield was decreased, possibly due to the loss of microencapsulated flavor powders and their physical damage during the spray drying process.

The effects of spray dry atomizing pressures on the flavor yield of microencapsulated pine flavor are shown in Table 1 and Fig. 4. The flavor yield at 160, 180 and 200 kPa was 46-87%, 44-95% and 46-90%, respectively. While the lowest flavor yield showed little variation at 44-46%, the highest flavor yield was 95% at 180 kPa.

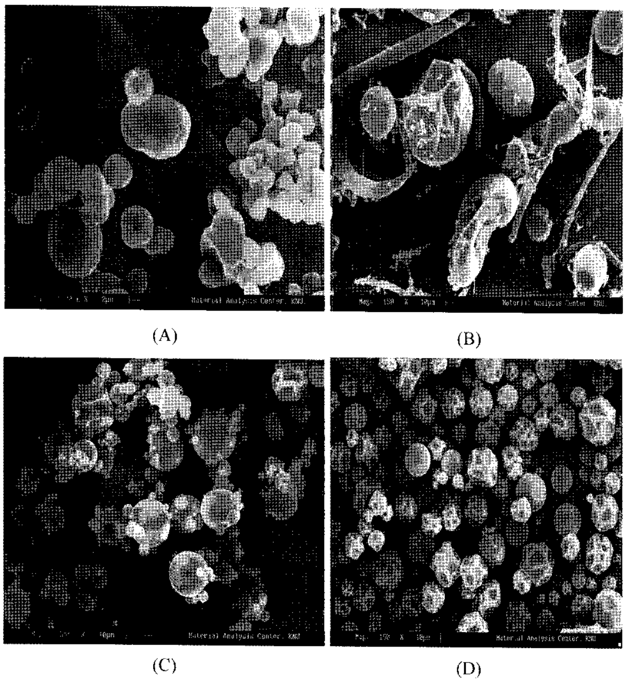


Fig. 5. SEM observations on initial (A) and full (B) collapses of the final products using several unstable sprays drying conditions, on optimum round shapes of microencapsulated pine flavors (C), and after storage for one month of the optimum shapes (D).

To observe the shapes of microencapsulated pine flavor, SEM observations were obtained from the microencapsulated pine flavors which were operated at the worst and optimum conditions of the spray dryer. The optimum microencapsulated pine flavor was stored for 1 month and the change was measured. The results are shown in Fig. 5. Under the optimum spray dryer condition, the shapes of the microencapsulated pine flavors were round, but the shapes of those under the worst condition showed initial and full collapses. After 1 month, the shapes were changed to a slightly shrunken polyhedron, showing high stability. These results on the optimization of microencapsulated pine flavors will help to determine the optimum operation conditions of a spray drier for the microencapsulation of other flavors.

Acknowledgments

This study was supported by a grant (HMP-97-F-3-0007) of the Korea Health 21 R&D Project, Ministry of Health & Welfare, Republic of Korea.

References

1. Versic RJ. Flavor encapsulation: An overview. pp.1-6. In: Flavor Encapsulation. Risch J, Reineccius GA (eds). ACS symposium series 370, Washington DC, USA (1988)
2. Bhandari BR, Dumoulin ED, Richard HMJ, Noleau I, Lebert AM. Flavor encapsulation by spray drying: application to citral and linalyl acetate. J. Food Sci. 57: 217-221 (1992)
3. Anandaraman S, Reineccius GA. Analysis of encapsulated orange peel oil. Perfum. Flav. 12: 33-39 (1987)
4. Shahidi F, Pegg RB. Encapsulation of the preformed cooked cured-meat pigment. J. Food. Sci. 56:1500-1519 (1991).
5. Trubiano PC, Lacourse NL. Emulsion-stabilizing starches use in flavor encapsulation. pp.45-54. In: Flavor Encapsulation. Risch J, Reineccius GA (eds). ACS symposium series 370, Washington D.C., USA (1988)
6. Kenyon MM. Modified starch, maltodextrin and corn syrup solid as wall materials for food encapsulation. pp. 42-50. In: Encapsulation and Controlled Release of Food Ingredients. Risch J, Reineccius GA (eds). ACS Symposium Series 590, Washington D.C., USA (1995)
7. Lee MJ, Lee YB, Yang JY, Kwon HS, Yoon JR. Isolation and identification of volatile compounds extracted from twigs of *Pinus densiflora* with Likens-Nickerson apparatus. J. Korean Soc. Food Sci. Nutr., 27, 568-573 (1998)
8. Lee MJ, Jung EJ, Lee SJ, Cho JE, Lee YB, Cho HJ, Yoon J. Comparisons of volatile compounds extracted from *Pinus densiflora* by headspace. J. Korean Soc. Food Sci. Nutr., 31: 26-31 (2002)
9. Tammela T, Nygren M, Laakso I, Hopia A, Vuorela H, Hiltunen R. Volatile compound analysis of ageing Pinus sylvestris L. (Scots pine)seeds. Flavor Frag. J. 18: 290-295 (2003).
10. Woo GY, Kim KH, Lee MJ, Lee YB, Yoon JR. A comparison of volatile compounds in pine extracts obtained by supercritical fluid extraction with those by simultaneous steam distillation and solvent extraction. Korean J. Food Sci. Technol. 31: 1268-1274 (1998)
11. Veglio F, Volpe I, Trifoni M, Toro L. Surface response methodology and preliminary process analysis in the study of manganiferous ore leaching by using whey or lactose in sulfuric acid solutions. Ind. Eng. Chem. Res. 39: 2947-2953 (2000)
12. Halliwell CM, Cass AEG. A fractional analysis of silanization conditions for the immobilization of oligonucleotides on glass surfaces. Anal. Chem. 73: 2476-2483(2001)
13. Kenyon MM, Anderson RJ. Maltodextrins and low-dextrose equivalence corn syrup solids: Production and technology for the flavor industry. pp.7-11. In: Flavor Encapsulation. Risch J, Reineccius GA (eds) ACS symposium series 370, Washington D.C., USA (1988)
14. Reineccius GA. Spray drying of food flavors. pp. 67-77. In: Flavor Encapsulation. Risch J, Reineccius GA (eds). ACS symposium series 370, Washington D.C., USA (1988)
15. Thevenet F. Acacia gums: Natural encapsulation agent for food ingredients. pp. 51-59. In: Encapsulation and Controlled Release of Food Ingredients. Risch J, Reineccius GA (eds). ACS symposium series 590, Washington D.C., USA (1995)
16. Mongenot N, Charrier S, Chalier P. Effect of ultrasound on cheese aroma encapsulation by carbohydrates. J. Agric. Food. Chem. 48: 861-867 (2000)