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U형 초단파 추출시스템에서 치자분말 추출상내

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Flow Characteristics in Packed Bed of Cape Jasmine Powders in U-column Microwave Assisted Extraction System

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Abstract

A Microwave Assisted Extraction (MAE) system was fabricated with a U-column of Pyrex glass tubing placed in cylindrical cavity having central focusing and dipolar power mode of 2450 MHz microwave and was applied to the extraction of yellow pigment from Cape Jasmine (*Gardenia Jasminoides Ellis*) with the result of 50% yield increase. The pulsating flow characteristics of MAE were investigated by monitoring pressure changes during MAE process with and without loading Cape Jasmine powder. Water vapor pressure in the heating column of U-column built up to 4.11 kPa in 5 sec to obtain the first elution in unpacked U-column followed by consecutive pulsating changes between 1.57~1.96 kPa. The first break through pressure of the packed bed was 4.11 kPa and gradually decreased to 1.96 kPa. The effluent volume of the extract through the packed column increased from 5 mL to 11 mL during leaching period of 4 min. The micro-structural changes in Cape Jasmine powder were examined by scanning electron microscopy and the increased porous structure was observed. The displacement theory was employed to elucidate flow mechanism in the packed bed of U-column MAE system.

Key word: microwave, extraction, cavity design, Cape Jasmine, leaching column, electron micrograph

Introduction

Since Maheshwari *et al.* (1988) noted micro-structural changes caused by microwave during oil extraction from rape-seed, the studies on food extraction using microwave have been reported. Lopez *et al.* (1996) introduced the MAE process as an alternative method to the conventional extraction process and researches including pectin extraction of fruit by Manabe *et al.* (1996) and essential oil extraction of peppermint leaves by Spiro *et al.* (1995) showed that microwave treatment enhanced their extraction rates. Yeum (1996) observed increased yield of soluble solids from coffee, green tea, red pepper and barley tea in batchwise microwave extraction, and Bureau *et al.* (1996) reported a similar result of flavor extraction from grape juice.

Recently, Jun and Chun (1998) designed a U-column MAE system which could utilize efficiently the power mode of microwave energy in the cylindrical microwave applicator, and noted that the U-column MAE system revealed two characteristic properties with 50% more yield than conventional method. Firstly, the extract showed a pulsating elution pattern. Secondly, the heating rate was self-regulated during the leaching process. The pulsating flow pattern and the pulse time interval were affected by the size of column and the heating rate of food inside the heating column under the focused electrical field mode in the cavity. This indicates that the flow properties of the packed bed in U-column MAE system can be different from the conventional one and that the extraction equipment should be matched with the energy distribution in the cavity.

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For further application of U-column MAE system to biological materials, the characteristics of flow and bed structure under microwave leaching should be investigated. Our objective was to elucidate the flow mechanism in the packed bed of U-column MAE system.

Materials and Methods

Theory

Mass transfer in a packed bed leaching can be correlated to the contacting time of solvent with solute particles in the bed. In MAE system the leaching process is completed within short period of time ranging in 10~20 seconds and as such contacting time is an order of second. Diffusion mechanism, therefore, is not applicable to this MAE system but the displacement model may be used. The displacement extraction model in the packed bed developed by Danckwerts (1953) was applied to barley tea extraction by Park (1984).

Danckwerts model is described as the following Equation 1.

$$\frac{C}{C_0} = 1 - \frac{1}{\pi R^2 \bar{\mu}} \int^{R\sqrt{1-1/2\bar{\mu}t}} 2\bar{\mu}\left(1 - \frac{r^2}{R^2}\right) 2\pi r \, dr \quad (1)$$

where C/C_0 is concentration ratio at time t. After integration, it can be expressed by simplified equation as follows;

$$\frac{C}{C_0} = \frac{l^2}{4\bar{\mu}^2 t^2} \quad (2)$$

C/C_0 is inversely proportional to two variables, the mean velocity and time. At a given length of packed bed, velocity is affected by the porosity of the bed, which may also be associated to the structure of solid tissue of Cape Jasmine under MAE environment. The hydraulic diameter can be described as a function of the porosity of the bed as in Equation 3.

Any change in hydraulic diameter by microwave may affect the dynamic properties of fluid around the solid particles such as thickness of laminar layer across which the solutes leach out.

$$d = \frac{4\varepsilon}{S} \quad (3)$$

Along with fluid properties, the steam generation and the characteristic pulsating flow in U-column MAE system must be explained.

The structure of waveguide and cavity is an important design factor for MAE system and the guide wavelength of the waveguide (λ_{g1}) can be designed based on Equation 4 for TE_{10} mode with 2.45 GHz source (Roussy and Pearce, 1995).

$$\lambda_{g1} = \frac{\lambda}{\sqrt{1 - \left(\frac{\lambda}{\lambda_{c1}}\right)^2}} = \frac{\lambda}{\sqrt{1 - \left(\frac{\lambda}{2a}\right)^2}} \quad (4)$$

The waveguide length (L_w) can be determined to be the integral multiples of $\lambda_{g1}/2$.

To build a cylindrical cavity having an axial focusing electric field, TM_{nm} mode is adapted and the cavity length (L_c) is calculated with Equation 5.

$$L_c = n\frac{\lambda_{g2}}{2} = \frac{n}{2}\frac{\lambda}{\sqrt{1 - \left(\frac{\lambda}{\lambda_{c2}}\right)^2}} = \frac{\lambda}{\sqrt{1 - \left(\frac{\lambda 2\pi r}{\rho_{nm}}\right)^2}} \quad (5)$$

The details of the designing the cavity are found elsewhere reported by the authors (1998).

Equipment

Microwave applicator: The magnetron of 2.45 GHz frequency producing 700 watt output was used and the power was transmitted through WR 340 type rectangular waveguide (Roussy and Pearce, 1995). The dimensions of the waveguide fabricated were 90 mm in width and 45 mm in length, and the waveguide length (L_w) was 166 mm by Equation 4.

The cylindrical cavity of TM_{01} mode with ρ_{01} of 2.405 was built to have 50 mm radius and 224 mm height (L_c), which was calculated according to Equation 5, as depicted in Fig. 1. A stub tuner was provided at the waveguide in order to obtain the maximum power transfer into the cavity.

Extraction unit: The U-column MAE system reported by Jun and Chun (1998) was used with modification at top of the inlet section of the heating column by installing a manometer (P) to measure the pressure. The U-column was made of Pyrex glass and consisted of two separable columns, the

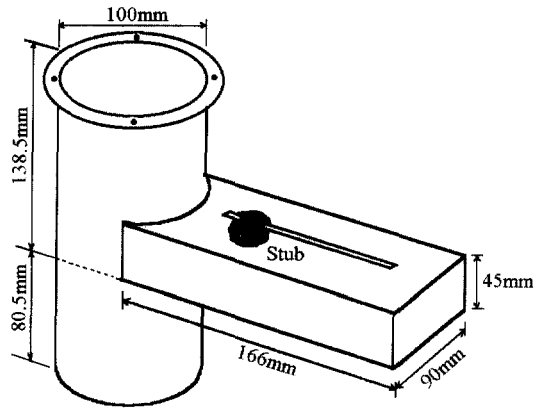

Fig. 1. Structure of the cylindrical cavity for MAE system.

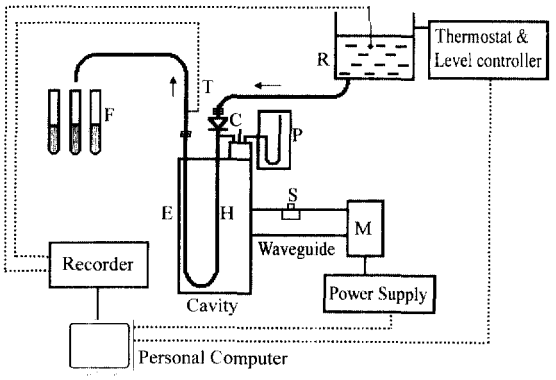

Fig. 2. Experimental set up for U-column MAE system. E: extraction column, H: heating column, M: magnetron, S: stub, R: reservoir, C: check valve, F: fraction collector, T: thermocouple, P: manometer

solvent heating column (H, 200 mm in length, 15 mm in diameter) and the packed-bed extraction column (E, 100 mm in length, 12 mm in diameter) as shown in Fig. 2. The heating and the extraction columns of the U-column were positioned along the central axis and 10 mm apart from the wall of the cylindrical cavity, respectively. The set-up of U-column MAE system is shown schematically in Fig. 2.

Methods

MAE procedures and data analysis: After dissembling the extraction column (E) of the U-column from the cavity, 0.7 grams of dry powder (-8+10 mesh, Tyler) of Cape Jasmine were packed in the extraction column and wetted by soaking into distilled water at 20°C for 2 min to expel air in the packed bed. The packed extraction column was connected to the heating column (H) to form U-column. The solvent, water at 20°C, was supplied from the reservoir (R) to flow through the check valve (C) up to the level of the reservoir.

The MAE procedure was begun by energizing the magnetron (M) and the extracts from the packed bed were collected with a fraction collector. The pigment content of the effluent was measured with a spectrophotometer (Ultraspec III, Sweden) at 440 nm. Non-energized MAE system served as the conventional extraction system for the control run, and the temperature and flow rate of the solvent were adjusted to have nearly identical conditions to MAE procedure.

Network Analyzer (Hewlett Packard, Model 8753C, USA) was used to monitor and measure the power properties in the cavity. The simulation of electric field intensity in the cavity was carried out with I-DEAS Master Series3 (Structural Dynamics Research Co., USA) and the resulting power intensity was expressed as the color image with the relative gray unit of arbitrary scale ranging 0~100.

Temperature and pressure measurements: After quick disassembling of the MAE unit, the surface temperature of U-column was measured with Hybrid recorder (Yokogawa, HR-2300, Japan, with T-type thermocouple), assuming the proportionality of the inner temperature of material loaded and the surface temperature.

Steam pressure at inlet end of the heating column was measured by U-tube manometer filled with water and a graduate cylinder (100 mL) trap was used between two U-tubing sets.

Scanning electron microscopy (SEM): For SEM, particles of Cape Jasmine (dimension: 7 mm × 7 mm) were taken from the packed U-column MAE system and the conventional extraction unit, respectively, under identical thermal treatment and effluent flow rate. After leaching for 5 minutes, the leached particles from both systems were observed with a low-temperature scanning electron microscope. The inoculated specimens of extracted Cape Jasmine were mounted to standard stubs using double-stick tapes and plunged into a liquid nitrogen

chamber for cryo-fixation. The specimens were monitored for sublimation process at 5 kV, sputtered with gold and observed with a Scanning Electron Microscope (JSM-5410V, JEOL Ltd., Japan) interfaced with a cryo-transfer system (CT1500 Cryotrans, Oxford Instruments Ltd., UK) at 15 kV.

Results and Discussion

Electrical power distribution in cylindrical cavity for MAE system

The simulation result of TM_{01} mode exhibited a dipolar power distribution as shown in Fig. 3. Maximum power field intensity (100 scale) occurred at the upper part of the zone, and gradually decreased to 25 scale and thereafter, it increased again to 50 scale at the center of the lower section forming another minor focusing mode. The radial power field intensity in the cylinder decreased as the radius increased approaching to the wall.

Power absorption at U-column under material loaded

The actual power absorption in the U-column placed in the cavity with 2% agar loaded was differentiated from the power mode of the empty cavity (Fig. 3). The highest thermal energy was found in mid section of the heating column as confirmed by the picture of melted agar as noticed in Fig. 4(a). The discrepancy between the power distributions in the empty cavity and thermal energy absorbed in the packed agar column was probably arisen by the broken modes, which were reformed by microwave reflection at the U-column unit installed. When water was filled in the U-column, hot water was ejected abruptly through the open tube of U-column and the hottest zone occurred at the similar zone of the agar column. Fig. 4(b) shows the location of heating and how the heated water flows out.

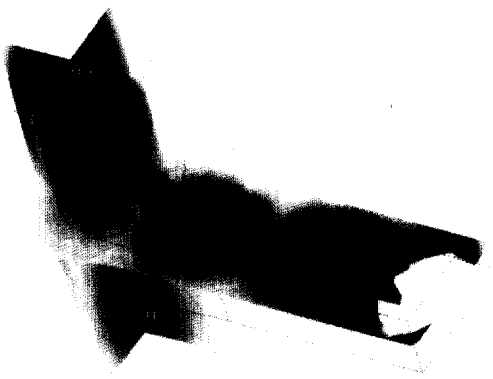

Fig. 3. Three dimensional power mode of microwave applicator.

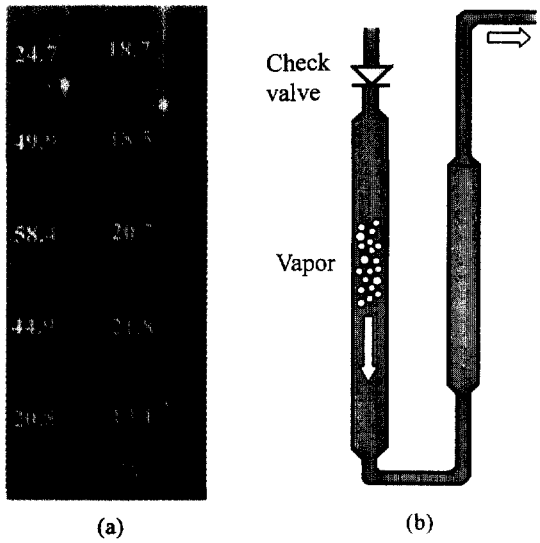

Fig. 4. Changes in flow rate and temperature of the effluent during U-column MAE operation. (a) Agar block melt; numeric figures are surface temperatures (°C) measured after 12 sec heating, (b) Model of vapor generation and solvent flow.

Flow characteristics of U-column MAE system

In our study on U-column MAE system (Jun and Chun, 1998), the pulsating elution and the self-regulatory heating pattern were observed during the leaching process. Fig. 5 showed a similar pattern between effluent volume and temperature measured in outlet tubing. The average volume fraction of the pulsating effluents was 30.7 mL with time intervals of 20 sec and the temperature fluctuations of the effluent fraction were self regulated within the range of 70~95°C despite of high steam pressure at the heating column.

The focused energy brought the solvent in mid section of the heating column to boil instantly and vapor bubbles were grown very rapidly at this section until the pressure was high enough to eject the solvent toward the extraction column. The va-

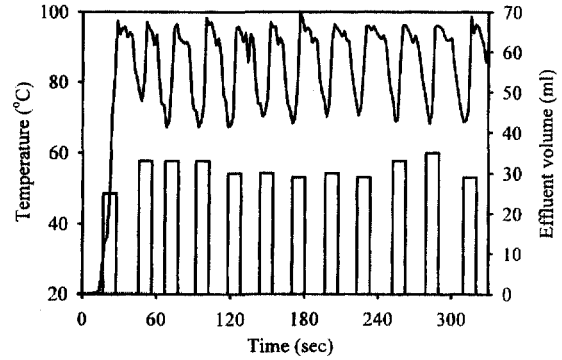

Fig. 5. Changes in flow rate and temperature of the effluent during U-column MAE operation with Cape Jasmine. —: Temperature, □: Effluent volume

porization and the release of its pressure were repeated until the end of the leaching process and it made the pulsating flow.

Pressure changes in U-column filled with water during MAE operation

To understand the mechanism of pulsating elution, the time course changes of pressure at inlet end of the heating column were plotted together with effluent volumes at outlet of the extraction column as shown in Fig. 6. Comparison of the pulsating effluent curve with pressure reveals a good agreement in their patterns suggesting that vaporization of solvent controls the flow pattern.

The initial pressure at the inlet of the column was 200 mmH_2O because the solvent was fed by gravity flow, and the effluent was to flow out when the pressure increased over this initial pressure, or break through pressure. Onset of MAE, vapor pressure in heating column reached 600 mmH_2O, which made the abrupt effluent flow out until the pressure was lowered down to the initial pressure of 200 mmH_2O. Fig. 6 shows the pattern of the pulsating pressure during MAE with unloaded state and the pressure remained at steady value near 160 mmH_2O at the effluent amount of 8 mL. The peaks of pressure pulse and the volume of the effluent decreased similarly until the pressure was dropped to 160 mmH_2O and thereafter the pulses disappeared. Despite of the disappearance of the peaks, the pulsating effluents continued. This disappearance of the pressure pulses could be explained by the effect of trap installed between two U-tube columns as shown in Fig. 2. The trap was gradually filled with the unheated solvent in the tubing outside the cavity and the solvent increased 36 mL with the rate of 0.4 mL/sec after 4 effluent fractions. At this moment the pressurized vapor was ejected from the heating column into the trapped water and condensed momentarily without any influence to the manometric readings of water leg of U-tube manometer.

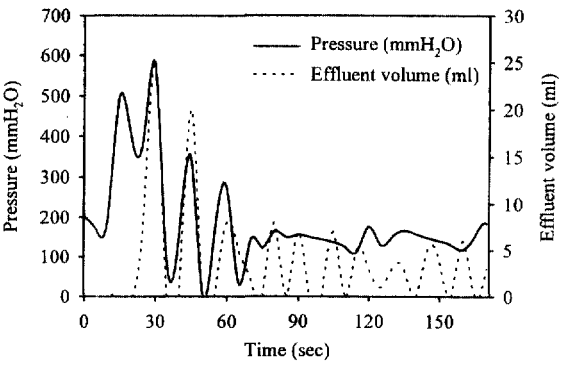

Fig. 6. Changes of pressure and effluent volume in U-column filled with water during MAE operation.

Pressure changes in packed U-column during MAE operation

After U-column packed with Cape Jasmine, pressure pulses were appeared with the pressure difference in average 200 mmH_2O during MAE as shown in Fig. 7(a) and the highest pressure was recorded with 400 mmH_2O, which was almost two times of the average pressure difference. The first peak pressures, 600 mmH_2O, were gradually decreased to 200 mmH_2O at the end of the leaching. On the contrary, the amounts of elution fractions showed an increasing trend as noticed in Fig. 7(b).

These phenomena suggested that the packed column restricted the flow rate of solvent through the bed at remarkable extent and extended the residence time of the solvent at heating column. The increase of residence time allowed for the solvent stream to absorb more energy and as such it increased vaporization rate, which substantially resulted the increase of effluent.

These data were very informative in that some structural changes might occur in the fluid path of the packed bed during MAE operation. The increase

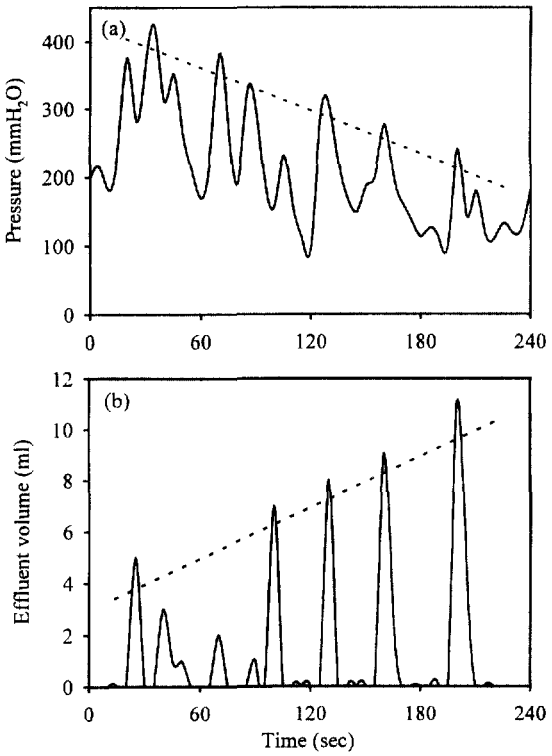

Fig. 7. Changes of pressure and effluent volume in U-column packed with Cape Jasmine during MAE operation. Dotted line represents trend. (a) Pressure, (b) Effluent volume

of elution volume could be resulted by an increase of hydraulic flow diameter in the packed bed as described in Equation 3.

Effect of MAE on the microstructure of Cape Jasmine

In order to investigate the effect of microwave irradiation on the microstructure of the tissue of Cape Jasmine, scanning electron microscopy was conducted. SEM images obtained after MAE operation shows remarkable structural changes compared to the specimen of the conventional extraction. The cutin layer and surface of tissue were severely damaged as convinced in Fig. 8. Even though we did not identify the damaged substances it might be wax layer which surrounds the cellular component (Juniper, 1995). Loss of the surface layer was so significant that intracellular structure was exposed and flat surface of the tissue was altered to the hollow structure and that the porosity

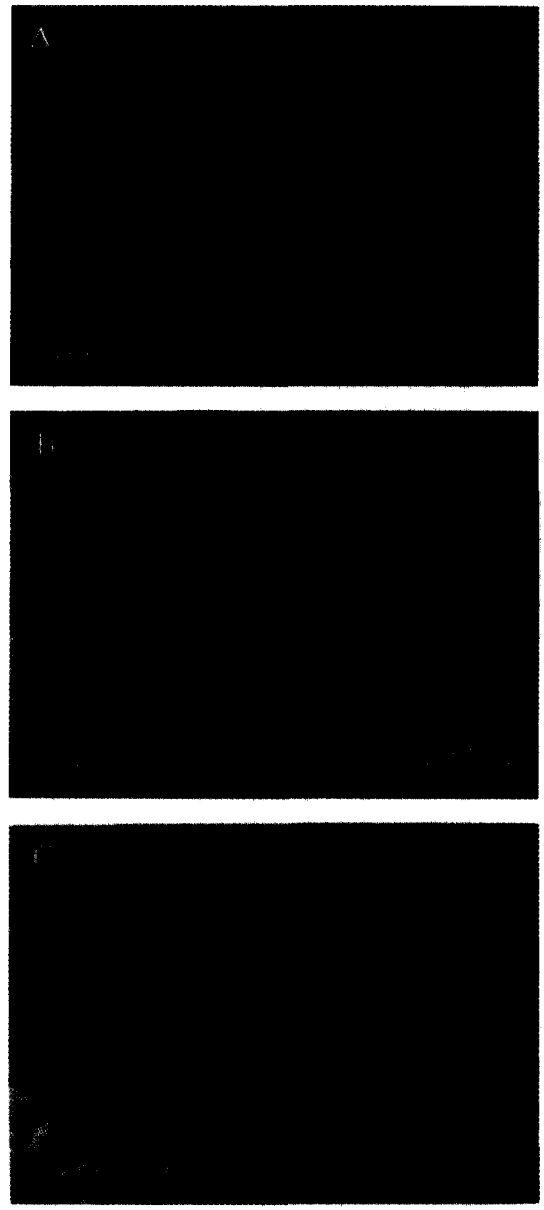

Fig. 8. Scanning electron micrographs of Cape Jasmine. A: Unleached (Control), B: Leached by conventional method, C: Leached by MAE method

fraction was increased several times.

The structural modification can affect the flow rate through the packed bed by enlargement of hydraulic diameter and can increase the contacting area with the solvent through more complicate capillary structure. These results strongly support that micro-structural change can be the main cause of the yield increase in MAE system.

Flow mechanism in packed bed of U-column at MAE processing

In spite of the peculiar nature of pulsating flow under focused power mode of microwave, we tried to employ Dankwerts model to elucidate this leaching process. Plotting of the concentration ratio (C/C_0) against the reciprocal of square time showed that MAE process data for Cape Jasmine fitted the displacement equation described as Equation 2 (Fig. 9). In the second run which was carried out one hour after the first MAE run, it also fitted well (Fig. 10).

The slopes of the fitted lines were clearly differentiated between the U-column MAE system and the conventional one as shown in Figs. 9 and 10. The increase of slope means the decrease of solvent velocity in the packed bed of MAE system as shown in Equation 2. Even though the plot was obtained under the pulsating flow conditions unlike those of Danckwerts model, the linearity of the plot suggested that it apparently followed the displacement mechanism.

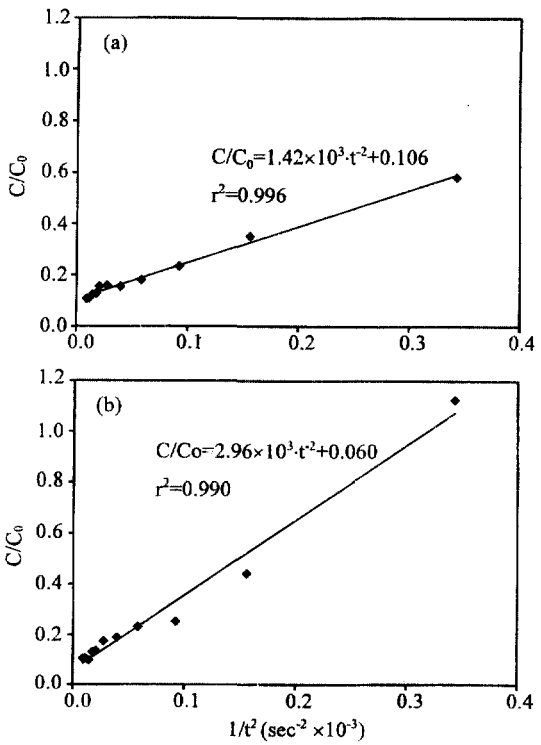

Fig. 9. Plots of the pigment concentration ratio against the reciprocal of square time in the first MAE run. (a) Conventional system (b) U-column MAE system

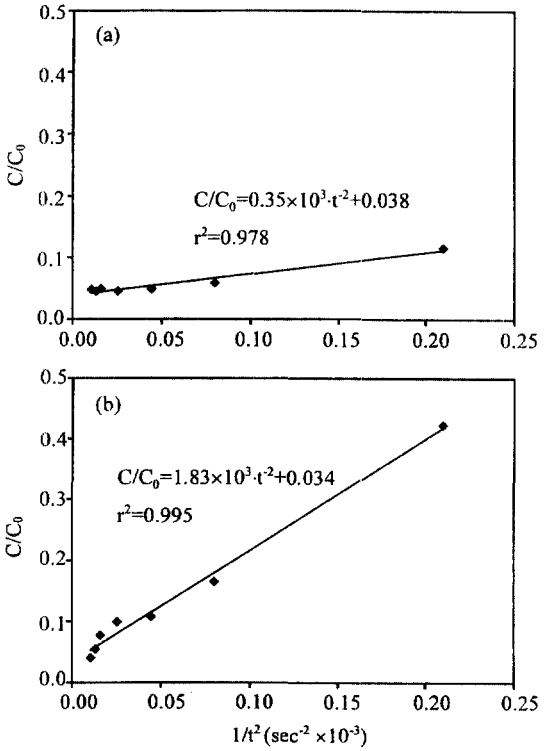

Fig. 10. Plots of the pigment concentration ratio against the reciprocal of square time in the second MAE run. (a) Conventional system (b) U-column MAE system

As noticed in Table 1, at the same volume flow rate mean linear velocity was reduced to 30% at the first run and 50% at the second run. It indicates that pore area is increased because we can confirm the hollow structure in the image of SEM (Fig. 8).

Table 1. Mean velocity and pore area in packed column at MAE and conventional extraction systems

	First run		Second run	
	MAE	Conventional	MAE	Conventional
Slope[1] ($\times 10^3$)	2.96	1.42	1.83	0.35
Mean velocity[2] (cm/sec)	0.09	0.13	0.12	0.27
Volume flow rate[3] (mL/sec)	0.97	0.97	0.71	0.71
Pore area[4] (cm^2)	10.52	7.28	6.12	2.68

[1]Coefficient of linear equation in Figs. 9 and 10.
[2]Calculated by Equation 3.
[3]Adjusted manually.
[4]Volume flow rate divided by mean velocity.

Considering the fluid characteristics of MAE system, the packed bed in column is altered to more porous structure and the bed is impacted by pulsating flow, which may affect the mass transfer rate of the pigment. For practical application the relationship between bed size and pressure development must be studied.

Nomenclature

a, b : width and height of waveguide, m
C : concentration of extract, %w/v
C_0 : initial concentration of extract, %w/v
d : hydraulic diameter, m
l : distance, m
L_w : length of waveguide, m
L_c : length of cylindrical cavity, m
n : integer
R : outer radius of packed bed, m
r : radius, m
S : cross section area, m^2
t : time, sec
ε : porosity
λ : wavelength in free space, m
λ_{c1} : cutoff wavelength in waveguide, m
λ_{c2} : cutoff wavelength in cylindrical cavity, m
λ_{g1} : guide wavelength in waveguide, m
λ_{g2} : guide wavelength in cylindrical cavity, m
μ : mean velocity, m/sec
ρ_{nm} : m th root of Bessel function J_n (ρ)

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References

Bureau, S., Razungles, A. and Bayonove, C., 1996, Glycosylate flavor precursor extraction by microwave from grape juice and grape, *Journal of Food Science,* **61**(3): 557-561

Danckwerts, P.V., 1953, Continuous flow systems: Distribution of Residence Times. *Chemical Engineering Science,* England, **2**(1): 1-13

Jun, S.J. and J.K. Chun, 1998, Design of Microwave Applicator for Packed Column Extraction and Its Application to Food Extraction, *Food and Bioproducts Processing,* submitted

Juniper, B.E. and C.E. Jeffree 1983, The anatomy, ultrastructure and biosynthesis of the plant surface, In: *Plant surfaces,* Edward Arnold, London, UK, pp.10-21

Lopez Avila, V., Young, R. and Teplitsky, N., 1996, Microwave-assisted extraction as an alternative to Soxhlet, sonication, and supercritical fluid extraction, *J. AOAC. Int'l,* **79**(1): 142-156

Maheshwari, P.N., D.W. Stanley, F.R. Van De Voort and J.I. Gray, 1981, Effect of microwave treatment on the microstructure of dehulled rapeseed, *Cereal Chem.,* **58**: 381-384.

Manabe, M., Naohara, J., Sato, T. and Okada, J., 1988, The extraction of pectin by microwave heating, *Journal of the Japanese Society for Food Science and Technology,* **35**(7): 497-501

Park, S.G., 1984, Extraction Mechanism of Barley Tea in Cylindrical Packed Column, Ph.D. thesis, Seoul National University

Ruossy, G. and J.A. Pearce, 1995, *Foundations and Industrial Applications of Microwaves and Radio Frequency Fields,* Wiley, USA

Spiro, M. and Chen-Sau Soon, 1995, Kinetics of isothermal and microwave extraction of essential oil constituents of peppermint leaves into several solvent systems, *Flavour and Fragrance Journal,* **10**(4): 259-272

Yeum, K.S., 1996, Effect of microwave heating in the extraction yield of soluble solid in food, M.S. thesis, Seoul National University