Temperature-Dependent Diffusion Coefficient of Soluble Substances During Ethanol Extraction of Clove

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ABSTRACT: The temperature dependence of the diffusion coefficient of ethanol-soluble substances from ground cloves (particle size 250 μm) during extraction was estimated by fitting batch extraction data at several temperatures (27.8, 40, 50, and 60°C) to a previously developed mass transfer model. The model was based on spherical geometry of particles. Nonlinear regression analysis was used to develop an equation that describes the diffusivity as a function of temperature. The temperature dependence of D_A was of the Arrhenius type.
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KEY WORDS: Cloves, diffusion, eugenol, extraction, mass transfer.

Eugenol (4-allyl-2-methoxyphenol), a phenolic compound in cloves, acts as an antioxidant on oleogenous foods, as an anticarminative, antispasmodic, and as an antimicrobial agent (1–3). The diffusivity is an important transport property for predicting the mass transfer coefficients and is useful for the design of mass transfer equipment. Experimentally determined diffusivity data are used if they are available. However, up-to-date literature searches have revealed little information on experimental studies on the diffusion coefficient of ethanol-soluble eugenol-containing substances of cloves. This work investigates the temperature dependence of diffusivity of soluble substances during alcohol extraction of cloves and establishes an equation for the diffusion coefficient of alcohol-soluble substances of clove as a function of temperature.

MATERIALS AND METHODS

Materials. Fresh green cloves were obtained from Kampung Batu Laut, Banting, Selangor, Malaysia. Analytical reagent-grade, Univar, 95% ethanol was used as the extraction solvent. Eugénol with a purity of 99.9% was purchased commercially from Merck (Darmstadt, Germany). Ultraviolet (UV)-grade methanol from Lab-Scan Limited (Dublin Co., Ireland) was used as the solvent in UV spectrophotometry analysis. The quantity of cloves necessary for the whole experiment was dried and stored at room temperature. Prior to the experiment, 2 kg of dried cloves was ground with a blender (Moulinex, Bagnolex, France). The optimum particle size of 250 μm was selected by sieving with Test Sieves (Endecotts Ltd., London, England) based on a previous study (4).

Initial moisture content. The initial moisture content of the ground cloves was determined by drying at 105°C.

Mean particle size of the ground cloves. Food particles often are irregularly shaped and nonuniform in size. They can be sieved to obtain a narrow size range, and the mean size for the range is often used to determine the diffusion coefficient (5). The particle size distribution of the 250-μm clove particles was measured by the manual method of sizing particles with a microscope (Schmidt Scientific, Wild Heerbrugg, Germany) as described elsewhere (6). Microscopic measurement data of 56 particles are given in Table 1. Tabular calculations of mean size and mean surface diameter of 56 particles (7) are illustrated in Tables 2 and 3, respectively.

Analysis method. According to the Atlas for spectral data (8) eugenol possesses three peaks at 206, 230, and 281 nm in the UV spectrum of an ethanol solution. For this work, a spectrum for reference eugenol in methanol was developed by varying the eugenol concentration until a clear absorption peak on the recording scale of the UV-265 visible recording spectrophotometer (Shimadzu, Tokyo, Japan) instrument was obtained. UV scanning and peak pick-up was performed between wavelengths 200 and 300 nm, and a spectrogram is shown in Figure 1. Similarly, the same procedure was repeated on the ethanol clove extract, and a spectrogram is illustrated in Figure 2.

A curve for absorbance against concentration of standard eugenol was constructed to monitor the eugenol content in the extraction process. The calibration curve was obtained by preparing pure reference eugenol at various concentrations in methanol, spectrophotometrically taking their absorbance at the maximum absorbance wavelength of 206 nm in a UV-265 visible recording spectrophotometer (Shimadzu), and fitting a line of best fit to the data by means of the integrated fitting software in the spectrophotometer. Three replicate samples

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TABLE 1
Microscopic Measurements of 56 Ground Clove Particles

Particle size (mm)	0.02	0.03	0.04	0.05	0.08	0.09	0.1	0.11	0.15	0.2
Number of particles	1	1	2	12	7	1	29	1	1	1

TABLE 2
Relative Percentage Frequency Distribution: Tabular Calculation of Mean Size of Ground Clove Particles

Range	Interval (dx)	Average size[a] (x)	Percentage in range (dϕ)	Percentage per micrometer (dϕ/dx)	(xdϕ)
0–20	20	10	1.78	0.089	17.8
20–30	10	25	1.78	0.178	44.5
30–40	10	35	3.57	0.357	124.75
40–50	10	45	21.42	2.142	963.9
50–80	30	65	12.5	0.417	812.5
80–90	10	85	3.57	0.357	303.45
90–100	10	95	51.78	5.178	4919.1
100–110	10	10	3.57	0.357	374.85
110–150	40	25	3.57	0.089	464.1
150–200	50	175	3.57	0.020	624.75

[a] x is assumed diameter of particles, ϕ is frequency function; mean size = $\Sigma x\,d\phi/\Sigma d\phi$ = 8649.9/100 = 86.499 µm.

TABLE 3
Tabular Calculation of Mean Surface Diameter of Ground Clove Particles[a]

Particle size range	Average size (x)	Number of particles (N)	x^2	ΣNx^2
0–20	10	1	100	100
20–30	25	1	625	6250
30–40	35	2	1225	2450
40–50	45	12	2025	24300
50–80	65	7	4225	29575
80–90	85	1	7225	7225
90–100	95	29	9025	261725
100–110	105	1	11025	11025
110–150	130	1	16900	16900
150–200	175	1	30625	30625

[a] N is the number of particles, N_T is the total number of particles.
Mean surface diameter = $S_n = (\Sigma Nx^2/N_T)^{1/2} = (390175/56)^{1/2}$ = 83.47 µm.

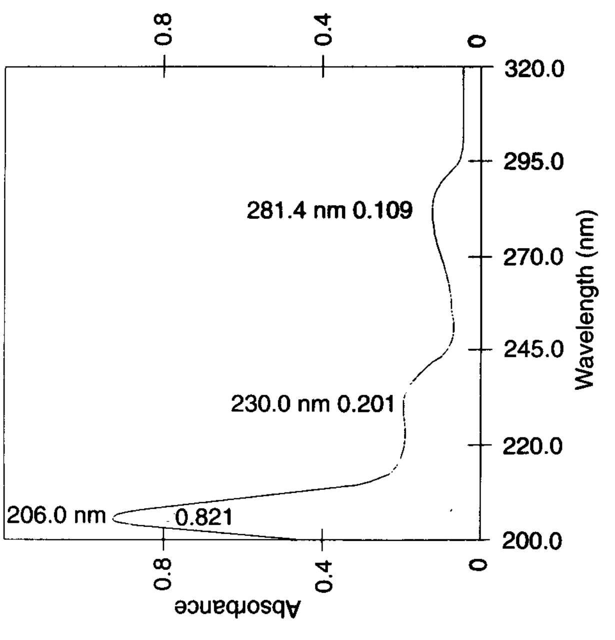

FIG. 1. Ultraviolet absorption spectrum (200–300 nm) of standard eugenol (20 mg/L) in 99.8% methanol (1-cm quartz cell), deuterium discharge lamp.

were prepared for each level of concentration, and the average results were used in curve fitting. The fit yields R^2 (0.9988), and a calibration curve obtained for eugenol is illustrated in Figure 3. Total ethanol-soluble substances were determined by drying the extract solution at 90°C. The total ethanol-soluble substances (oleoresin) were able to withstand 90°C without detectable quality deterioration, which becomes significant at temperature above 100°C (9).

Diffusion experiment. The apparatus used to carry out the batch-stirred extraction consisted of a Pyrex four-neck 500-mL flask with a hemispherical base 11 cm in diameter and 13 cm in depth. The temperature during extraction was controlled by immersing the stirred flask in a thermostatically controlled waterbath, capable of maintaining the temperature to within ±0.1°C. Ground cloves (50 g) of 250-µm particle size were agitated with 300 mL of 95% ethanol in the vessel by a marine-type propeller. The impeller speed was set at 2000 rpm, and the agitation rate was kept constant for all experiments. The

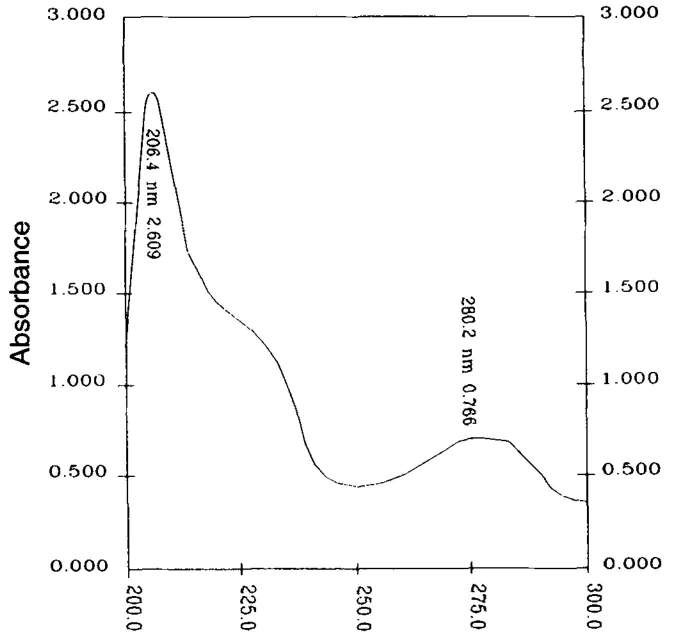

FIG. 2. Ultraviolet absorption spectrum (200–300 nm) of ethanol clove extract (800 mg/L) in 99.8% methanol; (1-cm quartz cell), deuterium

agitation method was effective in keeping the fluid well mixed and the particles in suspension. Auxiliary instruments were a temperature controller and a stirrer speed controller.

Homogeneous 2.5-mL samples of cloves and alcohol were withdrawn every 5 min by a hand-pump pipette from one neck of the vessel without stopping the stirrer, and the samples were collected in sample tubes. The samples were filtered immediately, and the eugenol content in the extract solution was measured by a UV-265 visible recording spectrophotometer (Shimadzu) at the maximum absorbance wavelength of eugenol (206 nm). The extent of extraction is characterized by the increase in concentration of eugenol in the solvent. The experiment was run continuously until equilibrium was reached, when the concentrations of several consecutive samples of bulk solution were the same. All of the extraction experiments in this study were run three times for each temperature. The average values obtained in these experiments are indicated in the study.

Mathematical model. The mathematical model for the diffusion process of solubles from the grind to the water during coffee brewing was described on the basis of the equation given by Crank (10) for a spherical particle suspended in a homogeneous medium (11):

$$\frac{M_\theta}{M_\infty}=1-\sum_{n=1}^{\infty}\frac{6\alpha(a+1)}{9+9\alpha+q_n{}^2\alpha^2}\exp(-D_Aq_n{}^2/a^2)\theta \quad [1]$$

where M_θ = mass of substance that has diffused at time t (g), M_∞ mass of substance that has diffused at infinite time (g), α = volume of solvent available per unit of grind, q_n = roots of the equation $\tan q_n = 3q_n/(3+\alpha q_n^2)$, D_A = apparent diffusivity of the substance (m²/s), α = radius of the particle (m), and θ = time (s).

Equation 1 is valid only if the flux of solubles leaving the solid is equal to the flux of solubles entering the solvent, that is, if there is no limiting layer of concentrated solution around the particles (11). Because this is also the case for ethanol extraction of eugenol from cloves particles, Equation 1 can also be used in the present study. This work is to explore the diffusivity of ethanol-soluble substances during batch extraction of cloves, taking the major phenolic compound eugenol as a representative for total solubles of the extract in lump-parameter Equation 1, in place of representing total solids as a single pseudocomponent in the diffusion of group multicomponent mixtures (12).

Setting up the modeling problem. For very large θ, only the first term in the series of Equation 1 is needed (5):

$$\frac{M_\theta}{M_\infty}=1-\frac{6\alpha(a+1)}{9+9\alpha+q_1^2\alpha^2}\exp(-D_Aq_1^2/a^2)\theta \quad [2]$$

$$\frac{M_\theta}{M_\infty}=1-A_1\exp(-B_1)\theta \quad [3]$$

where

$$A_1=\frac{6\alpha(1+\alpha)}{9+9\alpha+q_1\alpha^2}$$

$$B_1=D_Aq_1{}^2/a^2$$

and

$$\frac{M_\theta}{M_\infty}=1-A_2\exp(-B_2C_1{}^2)\theta \quad [4]$$

where

$$A_2=\frac{6\alpha(1+\alpha)}{9+9\alpha+q_1\alpha^2}$$

$$B_2=D_A/a^2$$

$$C_1=q_1$$

Regression analysis. The values of the constants A_1, B_1, A_2, B_2, and C_1 in Equations 3 and 4 at each temperature were determined by using the nonlinear regression procedure in the statgraphic statistical analysis software for the PC. The Marquardt algorithm (13) was used to fit the experimental data in Table 3 for the increase in mass of eugenol in the solvent during extraction, expressed as a ratio to the respective final concentration at equilibrium to Equations 3 and 4. The algorithm is a compromise between a straight linearization method and the method of steepest descent. The criteria used for model selection were the residual analysis and the magnitude of the correlation coefficient (r^2) being greater than 0.9.

RESULTS AND DISCUSSION

The initial moisture content of the dried clove particle sample was 5.64%, which is a realistic moisture percentage demonstrating that the extraction solvent alcohol does not pick up moisture and reduce its miscibility with eugenol.

Microscopic measurement of the 56 particles showed that 51.18% of the particles are in the 0.1 mm size range. Mean size of the 56 particles is 86.499 μm, and mean surface diameter is 83.47 μm. Diffusivities of eugenol at four temperatures are determined based on the mean size as well as on mean surface diameter.

The ultraviolet absorption spectrogram of standard eugenol in Figure 1 shows three peaks at 206, 230, and 281.4 nm and maximum absorption at 206 nm. In Figure 2, the absorption spectrogram of the ethanol extract of clove shows peaks at 206 and 280.2 nm. The broad spectrum between 225 and 250 nm indicates the presence of other minor compounds and interference in determining the eugenol content in that wavelength range.

In Figure 3, the intercept of the calibration curve of standard eugenol at 206 nm is 0.0975. The intercept is the absorbance of 99.8% methanol; the absorbance is high because the cutoff wavelength for methanol is 210 nm, and at 205 nm, the absorption of methanol is strong and equal to one. However, the intercept does not necessarily have to be zero. In this work, the calibration curve of eugenol is determined at 206 nm without avoiding cutoff of the solvent.

Foods contain many components. The extraction of a single component, e.g., sucrose or caffeine, is often desired; but for beverage extracts, extraction of all desirable flavor components is the goal. Some solutes extract more rapidly than others, e.g., lactose vs. total solubles in small-curd cottage cheese (14). However, broad groups of solutes often appear to diffuse as a single entity. This may occur because the concentration of one solute outweighs that of others, so that slower or more rapid diffusion by minor constituents is not detected. The diffusitives of trace components are probably-mutually independent, but solutes present in large concentrations can change diffusivities of other solutes (5). In coffee brewing, the apparent diffusivity of caffeine obtained by direct measurement predicts the diffusivity of total soluble coffee extract (11). In washing of cheese curd, the diffusion coefficient values of lactose are uniformly larger than the associated values for total solids, indicating that the whey proteins more significantly influence the diffusion of total solids than do the salts and low-molecular-weight components (14).

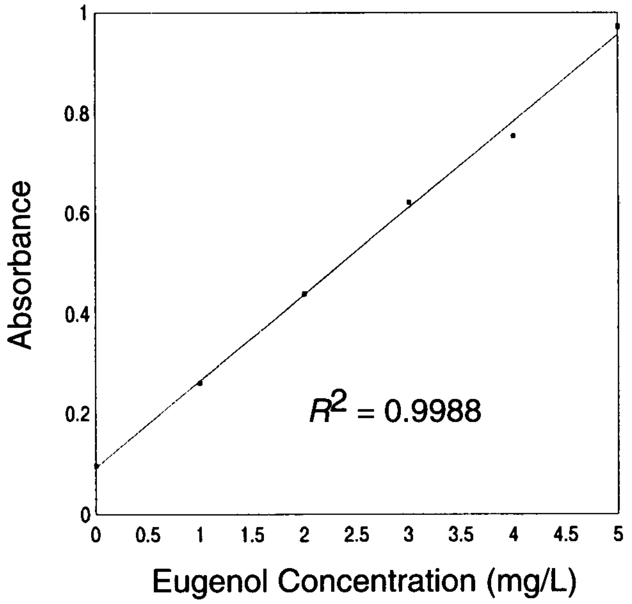

FIG. 3. Ultraviolet calibration curve for eugenol in 99.8% methanol at 206 nm.

The main volatile constituents of clove bud oil were eugenol (80.87%), caryophyllene (9.12 %), and eugenol acetate (7.33 %) (15). Previous work on analysis of the ethanol extract of clove by high-performance liquid chromatography (HPLC) (16) demonstrated that eugenol is the major phenolic compound in the ethanol extract, and that eugenol outweighs other components. Loss of volatile components can occur during removal of solvent to obtain total ethanol-soluble substances (oleoresin) of the clove extract. Phenolic antioxidants absorb strongly in the UV (17). To overcome loss of volatile components and to avoid laborious gravimetric methods, direct measurement of the increase in eugenol content in the extract with a UV spectrophotometer has been applied to characterize the extent of extraction.

In deformable solid foods, diffusion causes volume changes, such as shrinkage due to outward diffusion of water from grapefruit placed in sugar syrup (18) or in cheese undergoing brining (19). Swelling due to excess water infusion is responsible for the maintenance of cells, and osmosis can rupture the cell walls. In rigid solids, such imbalances are counterbalanced by intrinsic diffusion that generally exceeds the flow of the solute. Voilley and Cl. (11) studied the real dimensions and size changes of ground coffee as a function of time and temperature in diffusion of soluble substances during brewing of coffee. Bressan *et al.* (20) do not elucidate the various factors that affect diffusivity, such as the temperature effect on molecular diffusion and changes in curd structure and porosity with temperature, in determining the temperature dependence of the effective diffusion coefficient for total solids during washing of cheese curd. In this work, ground clove particles are taken as rigid particles, and volume changes during extraction experiments were not taken into account.

Experimental data of the ratio of increase in mass of eugenol in the solvent to final equilibrium mass at four temperatures from simple batch extraction experiments are presented in Table 4. It shows that 94.97% at 27.8°C, 96.69% at 40°C and 100% at 50 and 60°C of the equilibrium concentration of eugenol in the extract solution were extracted in 20 min. Experimental data of Table 4 were fitted onto proposed Equations 3 and 4. The model-fitting results are given in Tables 5 and 6 for the four temperatures. The magnitude of the coefficients varies with temperature. The experimental data correlated well ($r^2 > 0.98$) for all temperatures. High correla-

TABLE 4
Ratio of Mass of Eugenol in the Solvent to the Respective Final Equilibrium Values at Different Temperatures

Time (min)	M_θ/M_∞ (mass ratio)			
θ	27.8°C	40°C	50°C	60°C
5	0.9207	0.9402	0.9425	0.9528
10	0.9352	0.9549	0.9464	0.9848
15	0.9477	0.9615	0.9777	0.9938
20	0.9497	0.9669	1.00	1.00
25	0.9586	0.9685	1.00	1.00
30	0.9713	0.9947	1.00	1.00

TABLE 5
Model-Fitting Results of Ratio of Mass of Eugenol in the Solvent to the Respective Final Equilibrium Values at Different Temperatures to Equation 3

	Coefficient A_1		Coefficient β_1		
Temperature °C	Estimate	Standard error	Estimate	Standard error	r^2
27.8	0.9994	0.0469	0.4792	0.1032	0.9856
40	0.9998	0.0324	0.5445	0.0995	0.9934
50	0.9998	0.0244	0.5495	0.0768	0.9964
60	0.9999	0.0064	0.6052	0.0266	0.9997

tion coefficients and low standard errors indicate the good fit of the model. The curves from Figures 4 to 7 show that the model predictions represent the experimental data well, particularly at 40, 50, and 60°C. The apparent diffusivity determined from model-fitting results varies from 2.420×10^{-9} to 2.8932×10^{-9} m²/s with mean particle size and from 2.2581×10^{-9} to 2.6951×10^{-9} m²/s with mean surface diameter in the temperature range of 27.8–60°C. The diffusivity values calculated with particle mean surface diameters predict a closer agreement than the values with particle mean size (see Table 10 later in text).

Experimental data of the ratio of increase in mass of total ethanol-soluble substances to final equilibrium mass at optimum extraction temperature 50°C from batch extraction experiments are listed in Table 7. Only 94% of the total soluble substances are extracted in 20 min and equilibrium was reached at 90 min. Model-fitting results of the experimental data (Table 7) to the proposed Equations 3 and 4 are listed in Tables 8 and 9. The curves in Figure 8 show that the experimental data fitted well with the model equation with a correlation coefficient (r^2) of 0.9690. The apparent diffusivity of total ethanol-soluble substances at the optimum extraction temperature of 50°C, determined from the model-fitting results mentioned in Tables 8 and 9, is 2.38803×10^{-9} m²/s. The apparent diffusivity of eugenol, taking eugenol as representative for total soluble substances, is 2.8223 m²/s at the optimum temperature of 50°C. Eugenol was extracted more rapidly than the total solubles in extraction of eugenol from clove. The molecular diffusivities (Table 10) calculated from the Wilkie-Chang correlation (21) for eugenol vary from 0.6280×10^{-9} to 1.3905×10^{-9} m²/s, for caryophyllene they vary from 0.4924×10^{-9} to $0.1.0903 \times 10^{-9}$ m²/s, and for eugenol acetate they vary from 0.5590×10^{-9} to 1.2377×10^{-9} m²/s in the temperature range from 27.8 to 60°C. In Reference 22, the reported value of liquid diffusivity of phenol in ethanol at 25°C is 0.89×10^{-9} m²/s. The apparent values of diffusivities of eugenol are higher than the molecular diffusivities obtained from the correlation, but they are in the same order. At optimum extraction temperature (50°C), the sum of the molecular diffusivities of the three components eugenol, caryophyllene, and eugenol acetate is almost equal to the apparent diffusivity of eugenol and is also close to the value of the apparent diffusivity of total ethanol-soluble substances, which demonstrates that the extraction of eugenol parallelled the total solubles and that the apparent diffusivity of eugenol predicts the diffusivity of total solubles, particularly at the optimum extraction temperature.

The apparent diffusivities of eugenol at four temperatures, based on the mean size and mean surface diameter, are plotted in Figure 9. The dependence of the diffusivity on temperature is generally described by the Arrhenius equation

$$D_A = D_O e^{-E_a/RT} \qquad [5]$$

where D_A = apparent diffusivity (m²/s); D_O = constant (m²/s); E_a = activation energy (kJ/mol); R = gas constant (8.314×10^{-3} KJ/mol K); T = absolute temperature(°K).

TABLE 6
Model-Fitting Results of Ratio of Mass of Eugenol in the Solvent to the Respective Final Equilibrium Values at Different Temperatures to Equation 4

	Coefficient A_2		Coefficient B_2		Coefficient C_1		
Temperature (°C)	Estimate	Standard error	Estimate	Standard error	Estimate	Standard error	r^2
27.8	0.9994	0.0525	0.7779	87.401	0.7848	44.421	0.9856
40	0.9998	0.0362	0.8610	101.096	0.7952	47.972	0.9934
50	0.9998	0.0273	0.9249	84.373	0.7708	35.472	0.9963
60	0.9999	0.007	0.9284	32.210	0.8075	14.151	0.9994

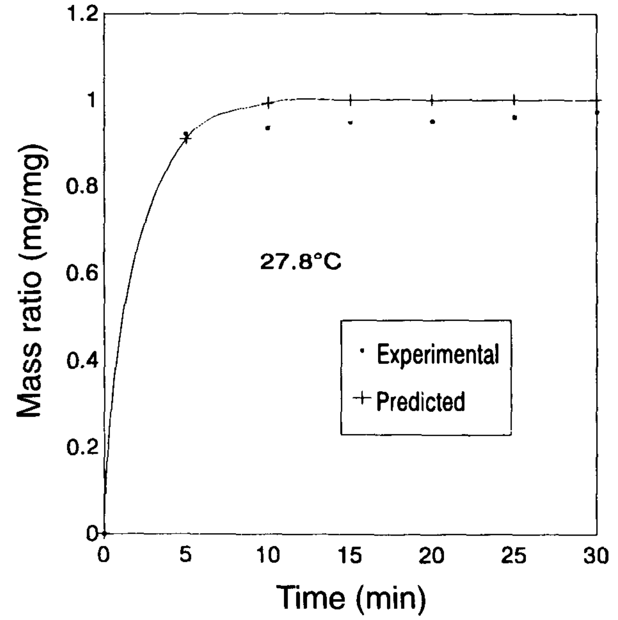

FIG. 4. Experimental and predicted results of mass of eugenol in the solvent as a ratio to final equilibrium mass during ethanol extraction of cloves at 27.8°C.

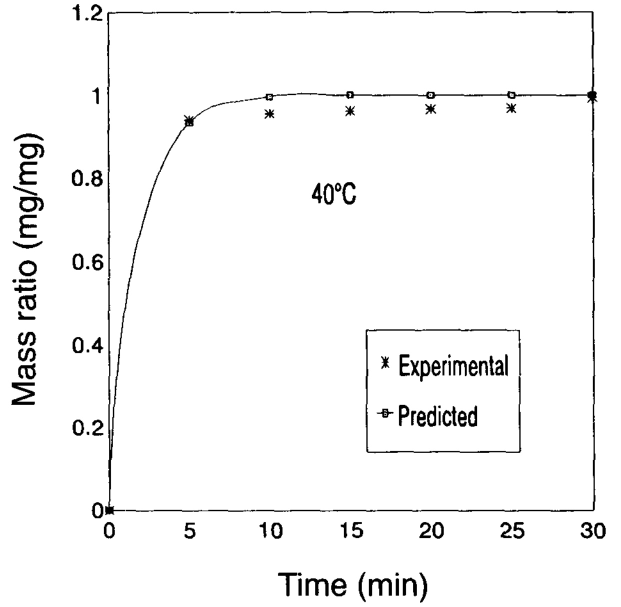

FIG. 5. Experimental and predicted results of mass of eugenol in the solvent as a ratio to final equilibrium mass during ethanol extraction of cloves at 40°C.

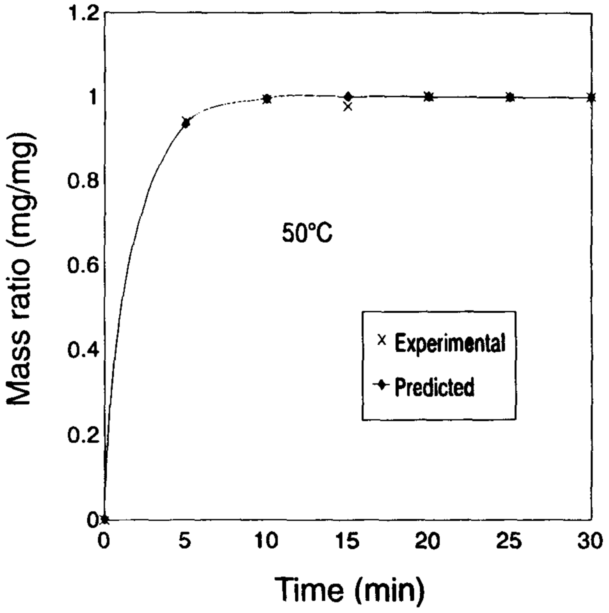

FIG. 6. Experimental and predicted results of mass of eugenol in the solvent as a ratio to final equilibrium mass during alcohol extraction of cloves at 50°C.

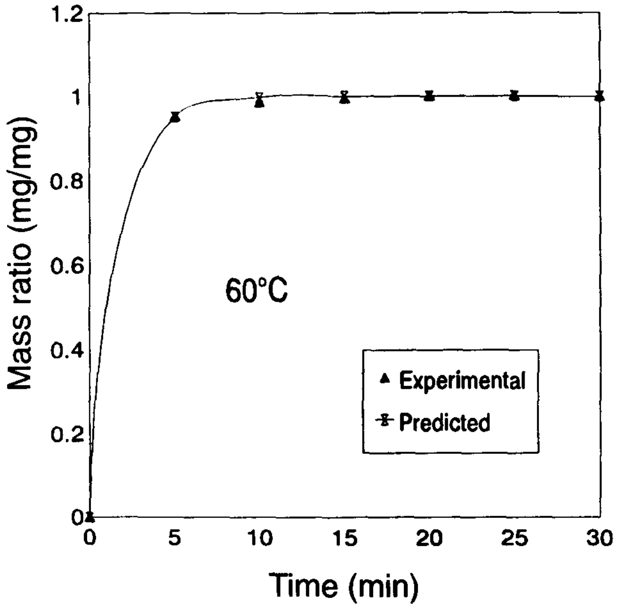

FIG. 7. Experimental and predicted results of mass of eugenol in the solvent as a ratio to final equilibrium mass during ethanol extraction of cloves at 60°C.

The logarithmic transformation of Equation 5 becomes:

$$\ln(D) = \ln(D_0) - \frac{E_a}{R}\frac{1}{T} \qquad [6]$$

The equations for the data presented in Figure 9 were, for mean size of the particle:

$$\ln(D\times 10^9) = 2.8342 - (0.5822\times 10^3)\frac{1}{T} \qquad [7]$$

for mean surface diameter:

$$\ln(D\times 10^9) = 2.7511 - (0.5782\times 10^3)\frac{1}{T} \qquad [8]$$

TABLE 7

Ratio of Mass of Ethanol-Soluble Substances of Clove to Final Equilibrium Values at Optimum Extraction Temperature 50°C

Time (min)	0	5	10	15	20	25	30	60	90	120
M_θ/M_∞ (mass ratio)	0.7623	0.8427	0.8800	0.9200	0.9400	0 9800	0.9900	1	1	1

TABLE 8

Model-Fitting Results of Ratio of Mass of Ethanol-Soluble Substances to Equilibrium Values at Optimum Extraction Temperature 50°C to Equation 3

Temperature (°C)	Coefficient A_1 Estimate	Coefficient A_1 Standard error	Coefficient B_1 Estimate	Coefficient B_1 Standard error	r^2
50	0.9855	0.05610	0.2291	0.0309	0.9690

TABLE 9

Model-Fitting Results of Ratio of Mass of Total Ethanol-Soluble Substances of Clove to Equilibrium Values at Optimum Extraction Temperature 50°C to Equation 4

Temperature (°C)	Coefficient A_2 Estimate	Coefficient A_2 Standard error	Coefficient B_2 Estimate	Coefficient B_2 Standard error	Coefficient C_1 Estimate	Coefficient C_1 Standard error	r^2
50	0.985518	0.0602	0.7659	27.605	0.5469	9.8886	0.9691

TABLE 10

Effect of Temperature on Molecular Diffusivity and Apparent Diffusivity

Temperature (°C)	Molecular diffusivity[a] ($D_M \times 10^9 m^2/s$) Eugenol	Caryophyllene	Eugenol acetate	Total	Apparent diffusivity of eugenol[b] ($D_A \times 10^9 m^2/s$) Mean size	Mean surface diameter
27.8	0.6280	0.4924	0.5590	1.6794	2.4240	2.2581
40	0.8713	0.6831	0.7756	2.330	2.6832	2.4995
50	1.0790	0.8460	0.9604	2.8854	2.8823	2.685
60	1.3905	1.0903	1.2377	3.7185	2.8932	2.6951

[a] From Wilkie-Chang correlation (Ref. 21).

[b] From model-fitting of experimental data.

The correlation coefficient (r^2) of the above was 0.9227, and the corresponding activation energies were 4.8402 and 4.8069 kJ/mol, respectively.

The nonlinear regression analysis method was used to evaluate the diffusivities of ethanol solubles from clove particles during extraction by monitoring the major phenolic compound in the extract solution at the four temperatures of 27.8, 40, 50, and 60°C. The method has made it possible to obtain reliable diffusivity more rapidly, and the results obtained herein can be used to solve the diffusion equation numerically. The temperature dependence of the diffusivity was of the Arrhenius type. The above equations are useful for engineering calculations in equipment design.

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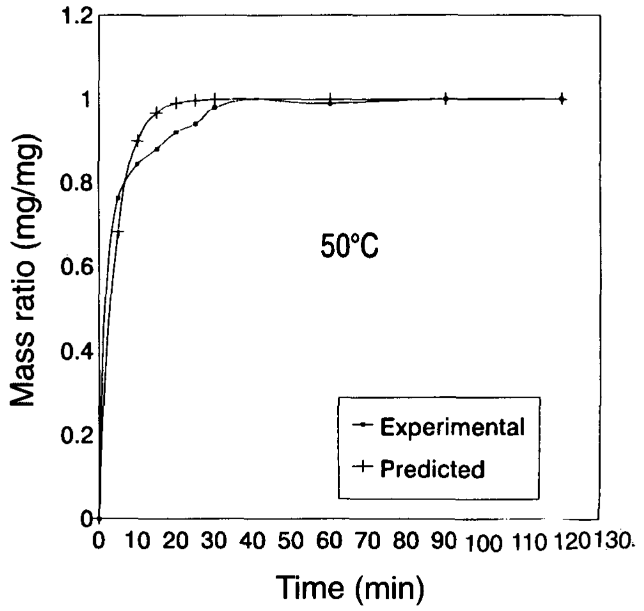

FIG. 8. Experimental and predicted results of mass of total ethanol soluble substances as a ratio to final equilibrium mass during alcohol extraction of cloves at 50°C.

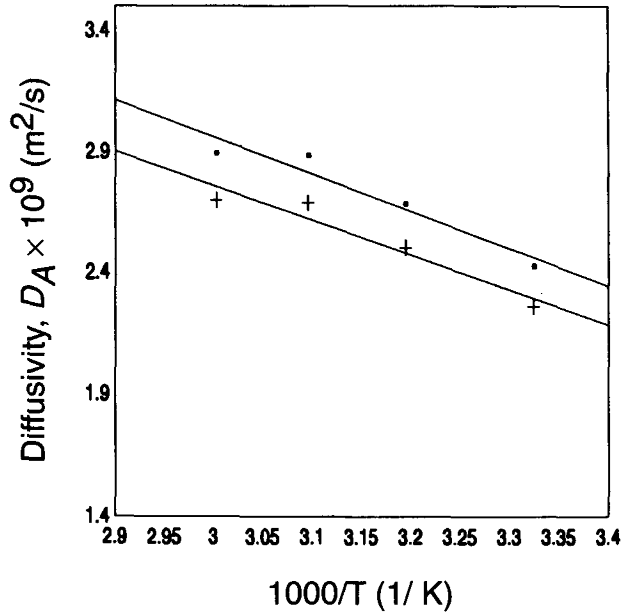

FIG. 9. Temperature-dependent diffusivity of eugenol from clove particles during alcohol extraction +, Mean surface diameter; ■, mean size.

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