# **Evaluation of the Diffusion Coefficient of Rapeseed Oil During Solvent Extraction with Hexane**

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**ABSTRACT:** The diffusion coefficient of rapeseed oil was determined from data obtained during time-varied solvent extraction experiments. The experiments were carried out in a Gülbaran extractor-diffuser with hexane as the solvent. A relationship was found between the slope of the diffusion line and the shape and size of the rapeseed particles. This relation can be used to calculate the diffusion coefficient. A diffusion coefficient of  $3.4 \times 10^{-8}$  cm<sup>2</sup>/s was determined from the experimental data.

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**KEY WORDS:** Diffusion, diffusion coefficient, rapeseed oil extraction, solvent extraction.

Although the principle of solvent extraction is simple, in reality it entails a complex mechanism, especially for oil-bearing materials, because of the cellular structure of vegetable bodies. Therefore, it is not possible to explain all phenomena that take place during solvent extraction with a single theory, such as surface washing, elution, osmosis, or diffusion (1). Molecular diffusion is an important mechanism in the extraction of oil from seeds after the removal of surface oil by washing, followed by diffusion of the oil from capillary channels. To explain the diffusion rate during solvent extraction of oilbearing seeds, empirical equations based on experimental data are usually employed, instead of the theoretical diffusion rate equation. However, it is possible and useful to obtain an estimate, based on Fick's second law of unsteady-state molecular diffusion (2). Studies are reported here in which the diffusion coefficient of rapeseed oil has been determined during extraction with hexane.

#### **EXPERIMENTAL PROCEDURES**

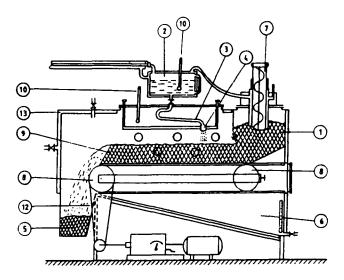
Experiments were conducted at different diffusion times, in a Gülbaran extractor-diffuser (3,4). The solid phase was ground rapeseed. The particles were spherical in shape  $(\alpha = 0.608, B = 9.87)$ , with an average particle diameter of 2R = 0.58 mm (x = R = 0.29 mm where R is radius of spherical particles (cm);  $\alpha$ , B are constants of theoretical diffusion equation; X is diffusion distance (cm). Hexane is used as the solvent.

The Gülbaran extraction system used in the study consists of a percolation type extractor, a rolling mill, solvent and water heating vessels, a screw-type conveyor, and an oil re-

covery distillation column. In the Gülbaran extractor, which is principally a percolation-type extractor, solvent is contacted with the bed of oil-bearing material once, and the miscella is not recyled. In this manner, every particle of the crushed material is brought into contact with the pure solvent during the entire residence time. This always provides a high driving force, i.e., a high difference in between the oil-bearing material and the solvent. Consequently, the extraction rate and the driving force are held at high values. The extractor is 2.1 m in length, 0.60 m in width, and 0.82 m in height. A cross-sectional diagram of the extractor is given in Figure 1.

The diffusion temperature is kept at about 55°C by circulating hot water in the extractor-diffuser jacket. The solvent is heated in a separate vessel and then drained to the particle bed at about 53°C. In the experiments, two different solvent drain rates (SDR) of 0.024 and 0.033 kg hexane per kg of raw material per min were used (5).

Crushing and grinding of cleaned rapeseed were conducted with a pair of rolling cylinders (35 cm in length, 15 cm in diameter). The opening between the cylinders was adjusted by a screw system. The speed of the cylinders was ap-



**FIG. 1.** Vertical cross-section of the Gülbaran extractor–diffuser. 1, Raw material feeding; 2, heated solvent tank; 3, solvent feeding pipe; 4, solvent spray feeder; 5, meal discharge; 6, miscella collection; 7, screw feeder for raw material; 8, cylinder; 9, bed; 10, thermometer; 11, motor; 12, transmission; and 13, hot water jacket.

TABLE 1 Sieve Analysis of Crushed Rapeseed

Sieve opening (mm)	Amount of crushed seed, m		
	Range, %	Average, %	
1.0-0.8	10–18	14	
0.8-0.63	2024	22	
0.63-0.50	4046	43	
0.50-0	1824	21	

proximatelly 300 rpm. The cylinders are grooved to crush the seeds. The differential sieve analysis of crushed rapeseed is given in Table 1. The average particle diameter was computed by the following formula.

$$2R = \sum_{i=1}^{n} m_i * (2R)_{i,av}$$
 [1]

where  $m_i$  is the amount of crushed seeds between sieve number i and i+1 (g/100 g), and  $(2R)_{i,av}$  is the average sieve opening of the sieves in series. The initial average oil content of the rapeseeds before diffusion was determined by laboratory soxhlet extraction as 43.82% by weight. The particles also contained 6.43% moisture. Thus, calculated oil concentration of particles at the beginning —t = 0—  $(C_0)$  was 88.10% on a dry basis.

After predetermined diffusion times, residual oil in the spent particles (C) was determined by Soxhlet extraction (6).

$$C = \frac{Y}{100 - Y} \text{(kg oil/kg inert material)}$$
 [2]

where Y is the percentage of oil content in dried meal, obtained directly from the Soxhlet. Experimental results and the calculated values of  $C/C_0$  are listed in Table 2 at the two different solvent drain rates (SDR).

# **RESULTS AND DISCUSSION**

The unsteady-state diffusion equation is:

$$-(\mathrm{d}C/\mathrm{d}t) = K(C - C_s)$$
 [3]

TABLE 2
Residual Oil Content (C) of Dried Rapeseed Meal at the End of Predetermined Diffusion Times, and the Calculated Values of  $C/C_0$ 

Diffusion time	Oil content of dried	Calculated value of	
t (min)	meal, Y	C from Equation 9	$C/C_0$
Solvent drain rat	e (SDR) = 0.024 kg solve	ent/(kg raw material • n	nin)
75	11.50	12.99	14.74
90	7.97	8.66	9.83
105	4.35	5.59	6.35
120	3.88	4.04	4.59
SDR = 0.033  kg	solvent/(kg raw material	• min)	
70	9.97	11.07	12.57
90	5.34	5.64	6.40
105	3.78	3.93	4.46
120	2.97	3.06	3.47

where K is the extraction constant, and its numerical value depends on the particle shape and thickness, permeability of the cell, and temperature.  $C_0$  is the homogeneous solute concentration in the particle initially, C is the concentration at the end of a certain extraction time, and  $C_{\rm s}$  is oil concentration of the hold-up solution (function of diffusion time). The integral form of Equation 3 can be written as:

$$\frac{C - C_s}{C_0 - C_s} = A \cdot \exp(-K \cdot t)$$
 [4]

where A is an integration constant, and its numerical value depends on the particle geometry, which is assumed to be unchanged during the extraction period.

Rearrangement of Equation 4 yields:

$$\frac{C_0 - C}{C_0 - C_s} = 1 - A \cdot \exp(-Kt)$$
 [5]

Because  $C_s$  will be much lower than  $C_0$  ( $C_0 >> C_s$ ), Equation 5 can be written as:

$$\frac{C}{C_0} = A \cdot \exp(-Kt)$$
 [6]

On the other hand, the molecular diffusion equation in a binary mixture, assuming one-dimensional transport, is as follows:

$$\frac{\partial c}{\partial t} = D_{AB} \cdot \frac{\partial^2 c}{\partial x^2}$$
 [7]

where  $D_{AB}$  is diffusion coefficient (cm<sup>2</sup>/s). Equation 7 is known as Fick's second law for diffusion. This transient diffusion equation can be solved only for known rigid particle geometries, such as flakes, cylinders, or spheres (7):

$$\frac{C - C_s}{C_0 - C_s} = \alpha \cdot \exp(-B \cdot D_{AB} / x^2) t$$
 [8]

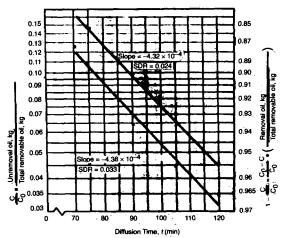
In this equation,  $C - C_s$  represents the amount of solute still unremoved, and  $C_0 - C_s$  is the amount of the total solute removable.

It is clearly seen that, using experimental values of C and  $C_0$ , when  $\log (C/C_0)$  is plotted against t, a straight line (called the experimental diffusion line) will be obtained from Equation 6. The slope of this line must be equal to the slope of the theoretical diffusion equation. Hence, it is possible to obtain a relation between K and the diffusion coefficient  $D_{AB}$  as follows:

$$D_{AB} = (K \cdot x^2)/B$$
 [9]

The above equation shows that, if experimental C values are known, the diffusion coefficient  $D_{AB}$  and the liquid-phase concentration  $C_s$  can be calculated from experimental data.

Evaluation of diffusion coefficient,  $D_{AB}$ . From data given in Table 1, two lines may be obtained for two separate solvent drain rates (Fig. 2). The slopes of both lines are calculated to be about  $-4.35 \cdot 10^{-4}$ , and the diffusion coefficient is calcu-



**FIG. 2.** Values of  $(C/C_0)$  as a function of diffusion time. SDR, solvent drain rate.

lated to be  $3.4 \cdot 10^{-8}$  cm<sup>2</sup>/s. These values are similar to those reported in the literature (8–10).

Determination of oil concentration of liquid phase as a function of diffusion time. After the value of the diffusion coefficient has been established, oil concentration of the liquid solution into which particles are immersed at any diffusion time can be determined by using Equations 8 and 6. Rearrangement of these two equations gives:

$$C_s = \frac{(1-a) \cdot C_0 \cdot C}{C_0 - a \cdot C}$$
 [10]

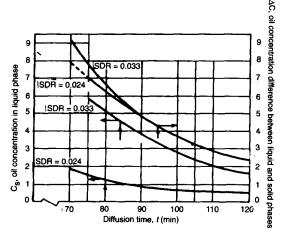
where a is a constant  $(a = \alpha/A)$  a = 0.608/1.04 = 0.585 for 0.024 solvent drain rate, and a = 0.608/0.720 = 0.844 for 0.033 SDR. (The values of 1.04 and 0.72 are obtained from diffusion line equations for corresponding SDR values.)

The proper driving force for diffusion  $\Delta C$ , the oil concentration difference between the particles (solid phase) and hold-up liquid (liquid phase) may be evaluated directly. Calculated values are shown in Table 3 and plotted in Figure 3, and it is clear that both C and  $C_{\rm s}$  strongly depend on SDR, because both C and  $C_{\rm s}$  increase when SDR is decreased.

The results obtained in this study may be summarized as follows: The diffusion coefficient of rapeseed oil in hexane

TABLE 3
Oil Concentration of Liquid Phase  $C_s$ , and the Value of Driving Force  $\Delta C$ , as a Function of Diffusion Time

Diffusion time (min)	С	$C_{\rm s}$ from Equation 9	ΔC
Solvent drain rate = 0.024	a = 0.585		
75	12.99	5.90	7.09
90	8.66	3.81	4.85
105	5.59	2.41	3.18
120	4.04	1.72	2.32
Solvent drain rate = 0.033	a = 0.844		
70	11.07	1.93	9.14
90	5.64	0.93	4.71
105	3.93	0.64	3.29
120	3.06	0.49	2.57



**FIG. 3.** Values of oil concentration in liquid phase and driving forces as a function of diffusion time. Abbreviation as in Figure 2.

has been determined to be  $3.40 \cdot 10^{-8}$  cm<sup>2</sup>/s from the experimental conditions employed in this study. The oil concentration difference between the solid and liquid phase, which is the driving force for diffusion, depends on the solvent drain rate at the beginning of the process but becomes independent toward the end. Thus, during extraction of oil from seeds, high solvent drain rates are advantageous at the beginning, and a lower rate prevails at the end of the process.

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