

Hexane Elimination from Soybean Oil by Continuous Packed Tower Processing with Supercritical CO₂

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ABSTRACT: Hexane elimination is the most energy-consuming step in the industrial extraction of soybean oil. It utilizes three sets of equipment: two evaporation stages in series followed by a stripper at a pressure of about 0.07 bar. The final hexane residue in the oil is about 1000 ppm. We propose an alternative to the present process for hexane elimination, based on the extraction of the soybean oil/hexane mixture with supercritical CO₂ in a continuous countercurrent packed tower. In this work, we tested a soybean oil/hexane mixture feed containing 10% by weight of hexane. Various pressures and temperatures of the column were tested to reduce hexane residue in the oil. The extraction process was demonstrated to be very effective for hexane separation. Indeed, at the bottom of the column we recovered soybean oil containing quantities of hexane as low as 20 ppm when we operated at 120 bar, 40°C. The effect of process parameters is also discussed.

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KEY WORDS: Desolventizing, hexane, oil purification, supercritical fluids.

Soybean oil is industrially produced by hexane extraction. The largest operative cost in this process is solvent elimination (1). The equipment used to carry out this operation consists of two rising film evaporators followed by a packed stripper operated under reduced pressure (0.067 bar, 100°C). As a rule 500–1000 ppm of hexane residue remains in the oil (2). Further reduction is very difficult owing to the requirement for higher temperatures that also reduce the oil stability. However, the need for further reduction of hexane residues in food is under examination since its toxicity has been demonstrated. O'Quinn *et al.* (3) found that hexane may be responsible of some degeneration of the digestive system of animals. Furthermore, hepatic absorption of hexane can lead to toxicity problems in the long term.

Supercritical CO₂ extraction of seed oil has been widely studied as an alternative to the traditional hexane extraction process. Soybean (4–8), wheat germ (9), corn germ (10), cottonseed (4,8,11), rice bran (12), rapeseed (6), peanut (8), sunflower (13), and almond (14) are some of the vegetable oils

extracted using supercritical CO₂ in laboratory plants. However, the industrial application of supercritical extraction of oils is not straightforward. Indeed, this process requires higher investment and operating costs than the traditional process. In particular, the operating costs will be comparable to those of the traditional process only if specific process schemes are adopted (15). Reverchon and Sesti Osséo (15) analyzed three different process schemes in the supercritical extraction of soybean oil and pointed out that only in the case of a near isobaric process with heat transfer integration are the operating costs in a similar range as the conventional extraction plants.

Supercritical CO₂ has also been used by some authors to study the post-processing of vegetable oils. A literature survey regarding de-acidification of oils and extraction of other impurities was given by Peters (16). However, until now, no attempt at hexane/oil mixture processing has been proposed.

In this work, we propose a new process for hexane elimination from soybean oil using a continuous countercurrent packed tower operating with supercritical CO₂. The tower is filled by packings characterized by high specific surface and wettability. It is fed by the oil/hexane mixture from the top and by supercritical CO₂ from the bottom. At the selected operating conditions hexane is soluble in supercritical CO₂ and thus is removed from the oil. This process could potentially substitute the units used for hexane elimination with a single-step operation which may have lower costs. The analysis of the process performance and of the effect of process parameters on the hexane residue is presented.

EXPERIMENTAL PROCEDURES

Experimental apparatus. A schematic representation of the experimental unit is presented in Figure 1. It mainly consisted of a 1920-mm long column (C) with an internal diameter of 17.5 mm. The column was packed with stainless steel packings 5 mm nominal size with 1600 m²/m³ specific surface and 0.9 voidage. The column was formed by five AISI 316 cylindrical sections (Autoclave Engineers, Erie, PA) each 305 mm long, connected by six four-port elements. Temperature along the column was controlled by five PID (proportional-integral-derivative) controllers (model 965; Watlow, Richmond, IL). The column was thermally insulated by ceramic cloths. CO₂ was fed to the column by a high-pressure diaphragm pump

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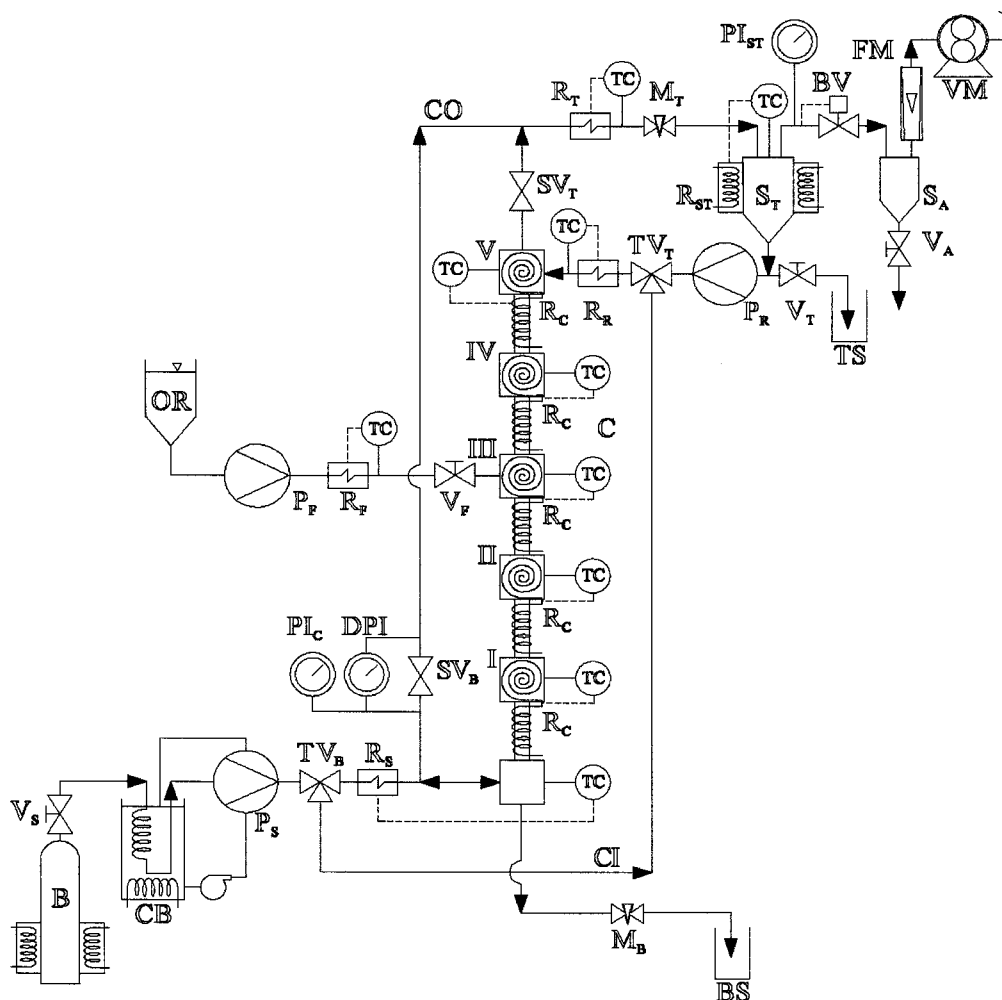


FIG. 1. Experimental apparatus. B, CO₂ cylinder; BS, bottom sample; BV, back pressure valve; C, column; CB, cooling bath; CI, cleaning line-in; CO, cleaning line-out; DPI, differential pressure indicator; FM, rotameter; M_B, regulating valve of the bottom product; M_T, regulating valve of the top product; OR, essential oil reservoir; P_F, feed pump; P_R, extract reflux pump; P_S, solvent pump; P_C, column pressure indicator; P_{IST}, top separator pressure indicator; R_C, column heater; R_F, feed heater; R_R, extract reflux heater; R_S, solvent heater; R_{ST}, top separator heater; R_T, top product heater; S_A, atmospheric separator; S_T, top separator; SV_B, sphere valve of cleaning line-out (open only while cleaning); SV_T, sphere valve of the column outlet (closed only while cleaning); TC, temperature controller; TS, top sample; TV_B, three-way valve at column bottom (deviates CO₂ flux toward CI while cleaning); TV_T, three-way valve at column top (takes CO₂ flux from CI and sends it to the column top while cleaning); V_A, atmospheric separator valve; V_F, feed valve; V_S, solvent valve; V_T, top product valve; VM, volumetric flow meter.

(P_C) (model Milroyal B; Milton Roy, Pont Saint-Pierre, France). The hexane/oil mixture was withdrawn from a reservoir and fed to the column by a high-pressure piston pump (P_F) (model Minipump; Milton Roy). The four-port connections allowed feeding of the oil at different column levels. At the top of the column a separator collected CO₂ and hexane. In all the tests reported in this work the experimental conditions were such that the oil solubility in the CO₂ phase was negligible. According to the data in the literature, in fact, the solubility of triglycerides is on the order of a magnitude of 0.1 weight fraction or lower (17,18). The oil feed was placed at the column top. The pressure in the separator was regulated by a back-pressure valve (model 26-1725-24; Tescom, Elm River, MN). Ahead of the vent, flow rate and total quantity of

CO₂ used were measured by a rotameter (model 605; Matheson, Parsippany, NJ) and dry test meter (model B10; Sim Brunt, Milano, Italy), respectively. Further details on this apparatus appear elsewhere (19).

Materials. A commercial soybean oil (Sagra; Salov s.p.a., Viareggio, Italy) was used to perform the experiments. We added hexane (purity 99.9% from Fluka, Neu-Ulm, Germany) to the oil to simulate hexane/oil mixtures obtained during the solvent elimination process. Pure CO₂ (99.9%) was purchased from SON (Naples, Italy).

Procedures. Two measures of the hexane content can be performed: free hexane and total hexane. Free hexane is the quantity of hexane desorbed from the sample when heated. Total hexane is obtained by adding a fixed quantity of water

to the sample, then heating it to desorb the hexane. We used the ISO Method number 9832 (20) procedure since it is a widely accepted international standard. It is based on the desorption of volatile compounds by heating at 80°C in a closed vessel after the addition of an internal standard (cyclohexane). The determination of the hexane content in the headspace was performed by gas chromatography using a capillary column. An HP 5890 gas chromatograph was used, which was coupled to a flame-ionization detector. A 25-m-long, 20- μ m film thickness Poraplot Q (Hewlett-Packard, Palo Alto, CA, USA) capillary column was used for the separation. Before starting the sample analysis, the whole procedure was repeatedly tested and calibrated. A detailed description of this procedure is reported elsewhere (21).

RESULTS AND DISCUSSION

We tested a soybean oil/hexane mixture containing 10% hexane by weight. This mixture is representative of the composition at the exit of the second evaporation stage in the conventional industrial process. It represents a good starting point for supercritical packed tower processing since the final stripper is the most expensive part of the solvent elimination process owing to its operation under vacuum (0.067 bar and 100°C).

Thermodynamic feasibility of hexane elimination by supercritical CO₂ is based on the solubilities of hexane and of soybean oil components in the supercritical solvent. Figure 2 is a diagram representing hexane-supercritical CO₂ solubilities at different temperatures. The experimental data were adapted from Wagner and Wichterle (22). More experimental data on equilibrium concentrations for CO₂-*n*-hexane mix-

tures are given by Li *et al.* (23) and Ohgaki and Katayama (24). Figure 2 shows that for operations, for example, at 40°C, hexane is completely soluble in supercritical CO₂ for pressures greater than about 80 bar. Therefore, operating at pressures equal or larger than 80 bar ensures the complete solubility of hexane in supercritical CO₂. In Table 1 a typical soybean oil composition is reported (after Ref. 2). It appears that soybean oil is essentially made of triglycerides. At 40°C these compounds have appreciable solubilities in supercritical CO₂ at pressures above 150–200 bar (25). Reverchon (17) and Maxwell (18) reported a selection of experimental data on triglycerides from the literature that confirm this general trend. Therefore, at 40°C, hexane is completely miscible with supercritical CO₂ for pressures higher than 80 bar, whereas the oil is substantially insoluble up to 150 bar. Although the behavior of the ternary system CO₂-hexane-oil has not been studied, we assumed that experimental data derived from the two binary systems CO₂-hexane and CO₂-oil are applicable; i.e., hexane and oil solubilities in CO₂ are not affected by the simultaneous presence of each other.

The process feasibility using supercritical CO₂ depends on a compromise between the maximization of the hexane solubility and the minimization of oil components solubility in supercritical CO₂. Therefore, we selected a range of temperatures between 40 and 60°C and two pressure levels of 90 and 120 bar for our experiments. The experiments were performed in the packed column designed using the conventional criteria for this kind of equipment. The fluid dynamic behavior of such columns is generally related to the gas flooding rate, G_{ff} , that is, a limit value of the gaseous stream (in our case supercritical CO₂). This stream flows upward in the column and exchanges momentum with the liquid phase flowing downward. When the momentum exchange balances the weight of the liquid, the downward flow stops: we say that the flooding condition has been reached. Of course, this is an undesirable process condition since it does not allow the proper column operation. The most convenient operating conditions are attained at gas velocities just below flooding (the so-called loading condition). The flooding gas flow rate depends strongly on the difference in density between the two phases; therefore, in our system it changes when different op-

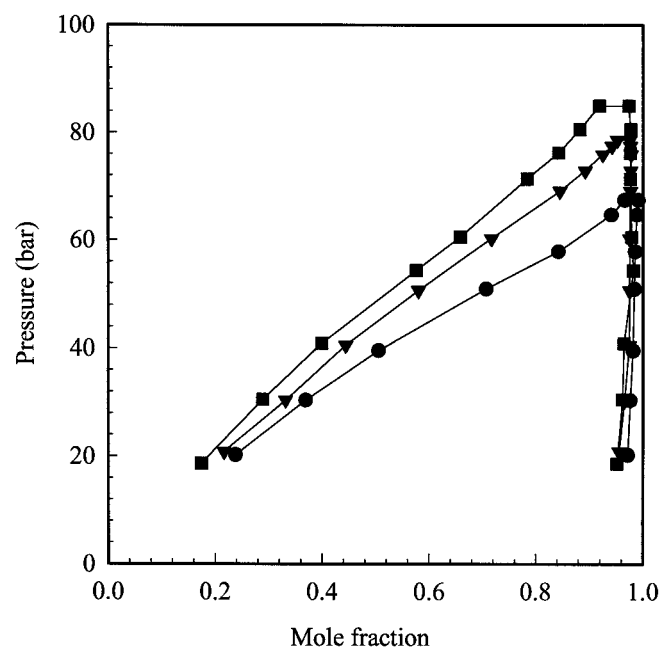


FIG. 2. Hexane solubility in supercritical CO₂ at various temperatures [adapted from (22)]. Temperatures: ●, 30°C; ▼, 40°C; ■, 50°C.

TABLE 1
A Typical Soybean Oil Composition

	Crude oil	Refined oil
Triglycerides (%)	95–97	>99
Phosphatides (%)	1.5–2.5	0.003–0.045
Unsaponifiable matter (%)	1.6	0.3
Plant sterols (%)	0.33	0.13
Tocopherols (%)	0.15–0.21	0.11–0.18
Hydrocarbons (%)	0.014	0.01
Free fatty acids (%)	0.3–0.7	<0.05
Trace metals		
Iron (ppm)	1–3	0.1–0.3
Copper (ppm)	0.03–0.05	0.02–0.06

^aAdapted from Reference 2.

erating conditions are chosen. Table 2 provides a list of the operating conditions tested. Besides temperature and pressure, CO₂ densities are reported in this table. The last row of Table 2 refers to the ratio (G/G_{fl}) between the CO₂ flow rate used during the experiment and the one calculated at the flooding conditions at the same pressure, temperature, and liquid flow rate. The liquid flow rate was fixed at $0.15 \times 10^{-3} \text{ m}^3 \text{ h}^{-1}$ which, considering an oil density of approximately 920 kg m^{-3} , corresponds to a mass flow rate of the liquid of about 0.138 kg h^{-1} . Solvent flow rate was fixed at 0.7 kg h^{-1} , which corresponds to a solvent-to-liquid feed ratio of about 5 and which allows small solute concentrations in the solvent phase inside the column and large driving forces.

Such low values of G/G_{fl} indicate that all our operating conditions are sufficiently far from flooding to operate safely. They also imply that the operating conditions tested are far from the best loading conditions for the operation. Research on loading flow rates deserves further experimental efforts and studies which are beyond the purpose of this work.

When the process is started in the column, it is necessary to wait to allow the system to reach steady-state conditions. This start-up time must be determined first for a correct evaluation of the process performance. Steady-state conditions have been reached when the composition of products at the top and at the bottom of the column no longer varies with time. In this process, we are particularly interested in the composition of the bottom product; i.e., the purity of the soybean oil; therefore, we measured the hexane content in the bottom product at increasing processing times for different process conditions. These results are reported in Figure 3 in terms of hexane content (ppm) in the bottom product vs. time. For all the process conditions tested, we registered no significant variations of the bottom content of hexane for processing times greater than 120 min. We assumed therefore that at that time steady-state conditions had been reached.

Although the density of the supercritical fluid is not an independent variable, results have been frequently expressed in terms of CO₂ density; indeed, CO₂ density results from a combination of temperature and pressure but is the most important parameter related to solubility. In Figure 4 we report the content of hexane at the bottom of the column at steady-state conditions as a function of the supercritical CO₂ density. In spite of simultaneous changes in pressure and temperature, in this case also the density alone correlates fairly well with separation efficiency. In particular, by increasing the CO₂

TABLE 2
Experimental Conditions

Test run	1	2	3	4
Pressure (bar)	90	120	120	120
Temperature (°C)	40	40	50	60
CO ₂ density (kg m ⁻³)	484	716	581	437
G/G_{fl}	3.83	6.54	4.35	3.50

^a G , gas rate (kmol s⁻¹m⁻²); G_{fl} , gas rate at flooding conditions (kmol s⁻¹m⁻²); thus G/G_{fl} represents the CO₂ flow rate used during the experiment and the calculated flooding rate at the same temperature and pressure.

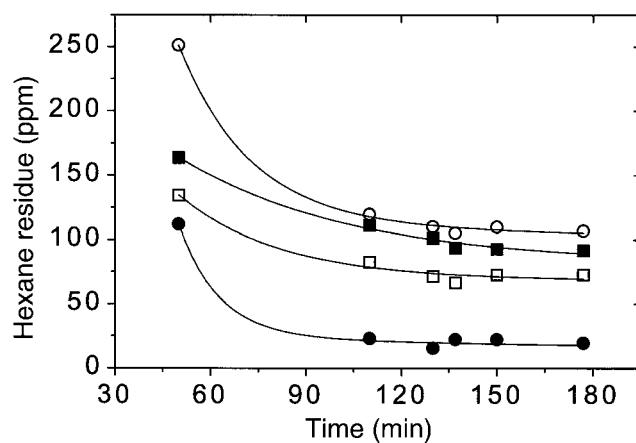


FIG. 3. Hexane residue in the bottom product vs. time. ○, 120 bar 60°C (716 kg m^{-3}); ■, 90 bar 40°C (581 kg m^{-3}); □, 120 bar 50°C (494 kg m^{-3}); ●, 120 bar 40°C (437 kg m^{-3}).

density the separation performance is improved, reaching the best performance with 20 ppm of hexane in the refined phase operating at a CO₂ density of 716 kg m^{-3} . This density corresponds to operating conditions of 120 bar and 40°C in the column. A possible explanation is that, at the operating conditions we used, the solubility of hexane in supercritical CO₂ is the controlling parameter of the process rather than the mass transfer rate. Within certain temperature ranges, in fact, solute solubility can be related with only minor deviations to the supercritical solvent density alone.

In Figure 5, the experimental results obtained by operating at 120 bar are shown as a function of temperature. If thermodynamics is controlling the column operation, we expect the temperature to play an important role. This is confirmed by the strong variation with temperature of the hexane content of the refined phase shown by Figure 5.

We carried out some calculations regarding the mass transfer inside the column using engineering literature equations

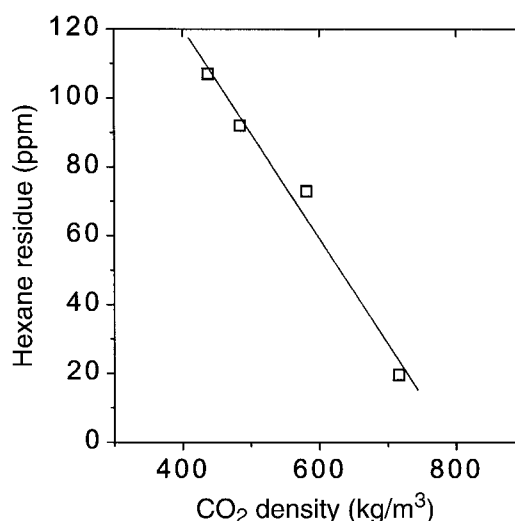


FIG. 4. Hexane content in the refined oil plotted against CO₂ density.

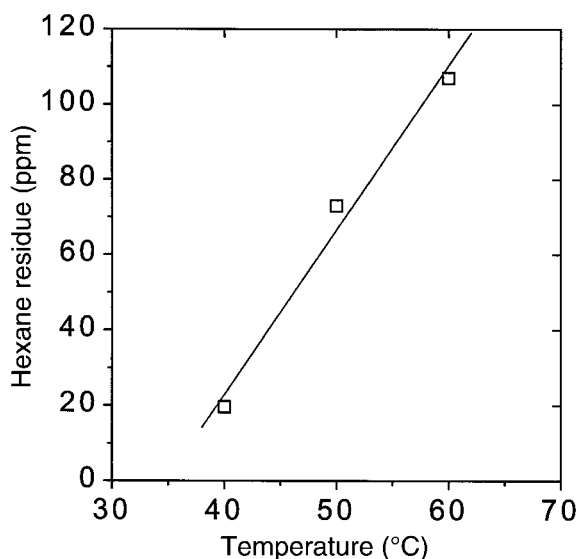


FIG. 5. Hexane content in the refined oil plotted against the process temperature at 120 bar.

developed for extraction columns using two fluid phases (26). As usual, the height of the column H_c (m) was set as

$$H_c = \text{HTU} \times \text{NTU} \quad [1]$$

In this equation, HTU is the height of the overall mass transfer unit in meters referred to the gaseous phase; it includes the features of mass transfer between the phases (the larger the mass transfer resistance is, the larger is HTU), and it is hypothesized to be constant along the column. In the same equation NTU is the number of mass transfer units. It is a function of the inlet and outlet concentrations and of the equilibrium partition coefficient between phases. Therefore, it is a measure of how difficult the separation is in terms of thermodynamic and separation objectives. HTU depends on the mass transfer coefficient in the gaseous phase k_G ($\text{kmol s}^{-1}\text{m}^{-2}$) and in the liquid phase k_L ($\text{kmol s}^{-1}\text{m}^{-2}$). Therefore, it is a measure of how fast the separation occurs. In the simplifying assumption of dilute systems we have

$$\text{HTU} = \frac{G}{a} \left(\frac{1}{k_G} + \frac{m}{k_L} \right) \quad [2]$$

where G is the molar flux in the gaseous phase ($\text{kmol s}^{-1}\text{m}^{-2}$), a is the specific interfacial area (m^{-1}), and m is the equilibrium partition coefficient (dimensionless). The calculation of the height of the mass transfer coefficients was performed using the method suggested by Onda *et al.* (27). From these calculations it appeared that, under the experimental conditions tested, the mass transfer resistance is almost completely concentrated in the gaseous phase, i.e., $1/k_G \gg 1/k_L$. Therefore, according to Equation 2 the partition coefficient is not important in the definition of HTU. Table 3 reports the values of HTU calculated in this way together with some of the system properties relevant to the calculation of k_G . Pure carbon dioxide viscosity was evaluated at atmospheric pressure and system temperature according to Chung *et al.* (28).

TABLE 3
Evaluation of Some Fluid and Process Characteristics^a

Test run	1	2	3	4
CO ₂ viscosity ($\text{Pa}\cdot\text{s} \times 10^5$)	3.34	5.25	4.07	3.22
Diffusivity of hexane in CO ₂ ($\text{m}^2 \text{s}^{-1} \times 10^8$)	3.71	1.61	1.99	2.37
Partition coefficient m	0.018	0.027	0.023	0.021
HTU (m)	0.33	0.51	0.47	0.45
NTU	5.9	3.7	4.1	4.2

^aHTU, height of overall mass transfer unit, in meters, referred to the gaseous phase; NTU, number of mass transfer units.

The correction due to the effect of the pressure followed Jossi *et al.* (29). Diffusion coefficients of infinitely dilute hexane in CO₂ were evaluated according to Takahashi (30). Table 3 also reports the number of mass transfer units (NTU) which was evaluated from the solute concentration in the fluid streams entering and exiting the system. The equation used was proposed by Colburn, as reported in Perry and Green (31), under the hypotheses of linear equilibrium curve, i.e., a constant equilibrium partition coefficient, and a linear operating line:

$$\text{NTU} = \frac{1}{1 - mG/L} \ln \left[\left(1 - \frac{mG}{L} \right) \left(\frac{y_b - mx_t}{y_t - mx_t} \right) + \frac{mG}{L} \right] \quad [3]$$

where y_b is the solute mole fraction in the gaseous phase (CO₂) at the column bottom (in our case it was assumed $y_b = 0$); y_t and x_t are the solute mole fractions at the column top in the gaseous phase and in the liquid phase, respectively; and L is the liquid rate ($\text{kmol s}^{-1}\text{m}^{-2}$).

According to this equation, NTU depends on the value of the dimensionless partition coefficient m . This value was chosen in order to have $\text{HTU} \cdot \text{NTU} = H_c$ (1.92 m). The values of the partition coefficient m obtained in this way are also given in Table 3. These are qualitatively consistent with what was expected from the variation of the operating conditions, supporting the soundness of the procedure followed. The values of HTU vary with the operating conditions, but these variations are of minor importance with respect to the thermodynamic constraints in the determination of the column effectiveness.

The effectiveness of the process is likely to be improved by using higher-efficiency packing and a different ratio between oil and supercritical fluid flow rate.

Some tests that extend this process to the extraction of hexane from other seed oils have also been performed (32).

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