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P. A. Wells[a]; N. R. Foster[a]; K. K. Liong[a]; R. P. Chaplin[a]
[a] SCHOOL OF CHEMICAL ENGINEERING AND INDUSTRIAL CHEMISTRY UNIVERSITY OF NEW SOUTH WALES, KENSINGTON, N.S.W., AUSTRALIA

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Supercritical Fluid Extraction of Triglycerides

P. A. WELLS, N. R. FOSTER, K. K. LIONG, and R. P. CHAPLIN

SCHOOL OF CHEMICAL ENGINEERING AND INDUSTRIAL CHEMISTRY
UNIVERSITY OF NEW SOUTH WALES
KENSINGTON, N.S.W., AUSTRALIA 2033

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Abstract

The demand for natural products has resulted in considerable interest in supercritical fluid (SCF) technology. The advantages of using SCFs as extraction solvents have been well documented. The ease of solute-solvent separation, low toxicity, and the ability to vary the solvent power make the use of SCFs attractive to the food industry, in particular in the area of triglyceride extraction. Recent material published has been primarily concerned with triglyceride extraction using supercritical (SC) carbon dioxide. The data published suggests limited solubilities (1-3 wt%) with the operating pressures in the vicinity of 300-500 bar, which will entail high capital and operating costs. Investigations carried out using SC propane as a solvent have indicated substantially higher loadings at much lower pressures. This is attributed to the similar chemical nature of the solute and the solvent. It is anticipated that triglycerides can be fractionated using SC propane on the basis of chain length and degree of unsaturation. In this paper the potential of using SC propane as a solvent for the extraction of triglycerides is discussed.

INTRODUCTION

The solvent properties of supercritical fluids (SCFs) were first observed over 100 years ago (*1*), but it has only been recently that SCF solvents have attracted considerable interest from both researchers and industrialists. Supercritical fluids feature properties that are intermediate between those of the liquid and gas phases. Gas-like viscosities and diffusion coef-

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ficients intermediate to those found in liquid and gas systems combine to produce a solvent with the ability to penetrate deeply and efficiently. Densities similar to those of the liquid phase engender high loading capabilities while the sensitive relationship between pressure, temperature, and SCF density allow for considerable control of solvent selectivity.

Extractions with SCFs are based on component volatilities (a distillation effect) and molecular interaction between the solute and SCF (a liquid–liquid extraction effect). The high volatility of SCFs enables economical separation of solute and solvent after the extraction has been completed with very little or no residual solvent, a feature of particular interest to the food industry.

There are a number of criteria which need to be examined when selecting the extraction solvent. To a large extent the critical temperature (T_c) and pressure (P_c) determine the temperatures and pressures at which the process will operate, and thus influence both operating and capital costs. The chemical nature of the solvent is also an important consideration as SCF extraction, in part, depends on solute–solvent interactions. The relative availability of the solvent, price, purity, and ease of use are also factors that must be considered.

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Carbon dioxide is by far the most widely investigated SCF solvent because it is inexpensive, nontoxic, nonflammable, available in abundance, and at high purity. Carbon dioxide also has a low critical temperature (31.04°C) and thus is particularly attractive for the extraction of thermally labile substances. Propane has also been extensively used as a high pressure solvent, but mainly as a near critical liquid rather than a supercritical fluid. Many excellent review papers have been written on the use of CO_2, propane, and SCFs in general (*2–4*). In this paper the major processes involving the use of propane as a solvent will be discussed as will the potential of propane to act as a solvent for the extraction of triglycerides. A comparison with extraction processes employing CO_2 will also be made.

PROPERTIES OF PROPANE AND PROPANE BINARY SYSTEMS

Propane is an odorless gas at standard temperature and pressure but can be readily liquefied by compression. The molecular weight of propane is 44, which is comparable to that of carbon dioxide. Gaseous propane is heavier than air and has a boiling point of 231 K at atmospheric pressure. Its critical pressure and temperature are 42.0 atm and 370 K, respectively.

Propane has a specific gravity ranging from 0.5 at 21.1°C (7 bar) to a critical density of 0.232. Propane is stable, noncorrosive, and inexpensive. As the critical point is approached, the surface tension and viscosity of propane become negligible and the fluid becomes highly compressible (*5*).

The major disadvantage of utilizing propane as a solvent is that it is flammable, with explosive limits in air of between 2.37 and 9.5% by volume.

As is clearly shown in Fig. 1, in the vicinity of the critical point of propane an increase in temperature at constant pressure (e.g., in going from pt 1 to pt 3) results in a large decrease in density. Similarly, a small decrease in pressure [at constant temperature (e.g., 105°C), pt 2 to pt 3], again induces a large density decrease. Both operations would have the effect of dramatically decreasing the solvent power of propane. It is this ability to control the solvent capacity in the vicinity of the critical point that makes the use of propane attractive.

A generalized pressure-temperature projection of the three dimensional P-T-X phase diagram for the propane/triglyceride system is presen-

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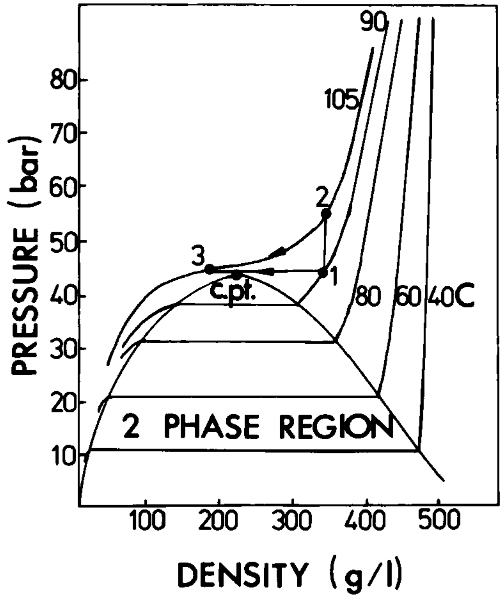

FIG. 1. Phase diagram for pure propane.

ted in Fig. 2 and is typical of binary systems that are of chemically similar nature but differ markedly in molecular size. For additional clarity, several isothermal cross sections of the P-T-X phase diagram are also included (Fig. 3). In accordance with convention, the component with the lower boiling point (propane) is labeled A and the triglyceride component B.

As can be seen from Fig. 2, there lies in the vicinity of the critical point of propane a three-phase plane bounded by the lower and upper critical end points (lcep and ucep). For temperatures below the lcep [Fig. 3(a), T_1], propane and the triglyceride component are completely miscible in the liquid phase. Between the lcep and ucep there lies a region of immiscibility in which two liquid phases, one rich in propane and the other rich in the triglyceride component, exist [Fig. 3(b), T_2]. Once the ucep has been crossed, the liquid phases once again become miscible [Fig. 3(c), T_3].

The phenomena of decreasing solubility with increasing temperature that is experienced as the lcep is crossed is contrary to that experienced

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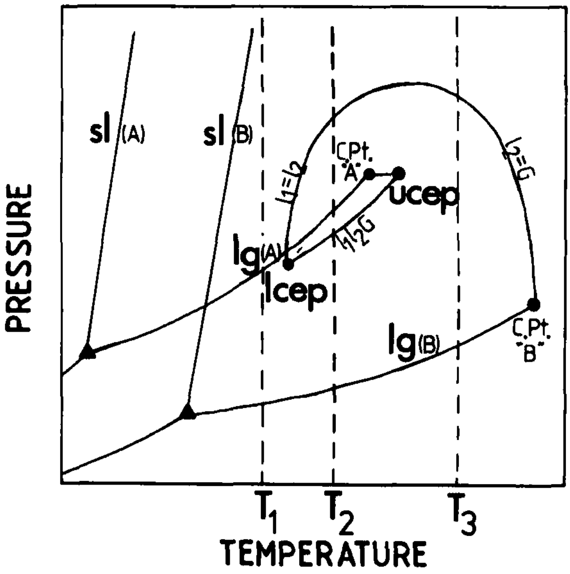

FIG. 2. P-T-X projection of a typical propane/triglyceride system. sl: solid/liquid phase boundary. lg: liquid/gas phase boundary. l_1l_2g: three-phase plane. $l_1 = l_2$: critical locus at which liquid phase l_1 and l_2 become identical. $l_2 = g$: critical locus at which liquid phase l_2 and g become identical. lcep: lower critical end point. ucep: upper critical end point. C.pt.: critical point.

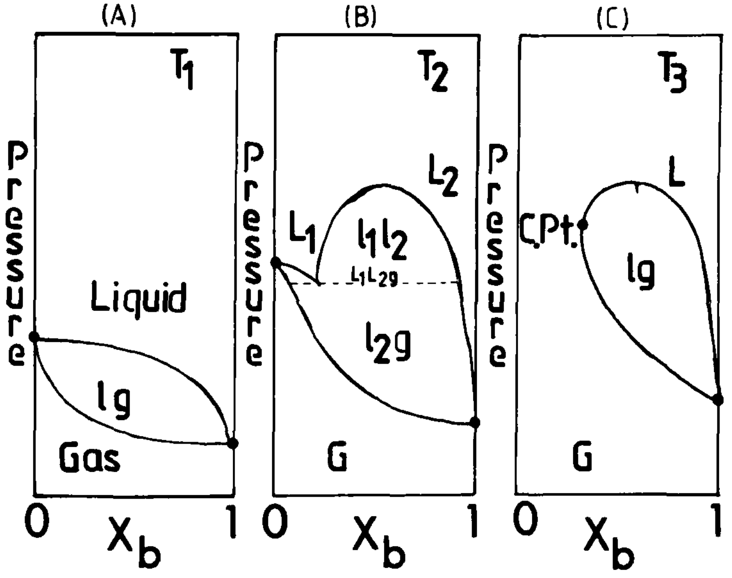

FIG. 3. P-X cross sections relating to temperatures indicated in Fig. 2.

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with normal liquid solvents. This behavior is a result of the decrease in density that propane undergoes when heated isobarically. The lcep represents the point where the propane density (and thus its solvent capacity) is such that it is no longer able to hold the solute in solution over the entire composition range.

The exact nature of the relationship between triglyceride structure and the location of the lcep will be discussed in greater depth later in this paper. At this stage it is sufficient to state that the greater the difference in chemical makeup between the solvent and solute, the lower the lcep temperature. Conversely, the greater the similarity between the solute and solvent, the smaller the region of immiscibility (the lcep moves toward the ucep), until it is possible that the region of immiscibility vanishes.

Unlike the lcep, investigations to date indicate that the location of the ucep is little affected by the nature of the solute. In the systems involving propane and a large hydrocarbon, it has been found (*6*, *7*) that the ucep is generally located close to the critical point of pure propane. Furthermore, the three-phase plane generally extends below the vapor pressure curve of propane.

PROPANE DEASPHALTING

Near-critical liquid and SCF propane extraction processes have been in use for some decades. Wilson and coworkers (*8*) examined phase equilibrium which later became the basis for a process in which heavy petroleum fractions are refined (the process is commonly called "propane deasphalting"). The process, shown schematically in Fig. 4, although not specifically a SCF extraction process, does utilize the change in solvent power of propane in the vicinity of its critical point.

A brief explanation of the process follows. Liquefied propane and the heavy crude oil fraction are both fed into the asphalt settler which operates at 40 to 60°C. At these temperatures the asphalt is only slightly soluble in the propane and precipitates. Aided by the low viscosity of the propane/oil solution, the precipitate settles quickly and is then removed for further processing. The propane/oil solution is then slightly depressurized, resulting in a decrease in solution temperature, inducing the precipitation of any waxy components. The remaining solution is then passed to the resin settler and heated to approximately 100°C where the lower solvent density (and thus solvent power) results in the successive

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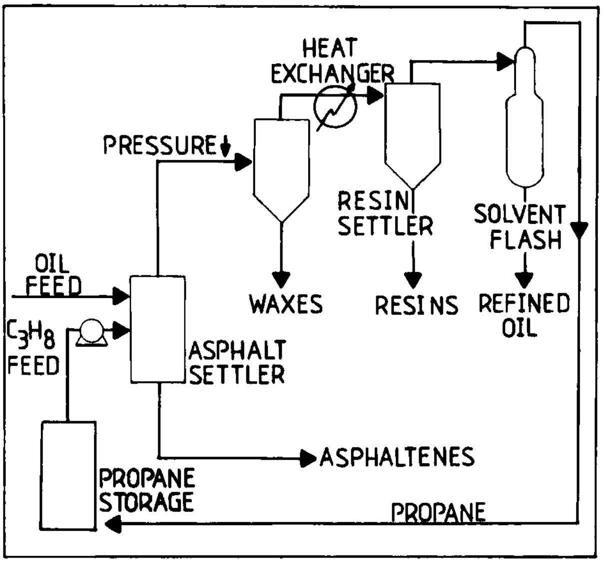

FIG. 4. Schematic of the "propane deasphalting" process.

precipitation of the resins, heavy ends, and naphthenic components. Finally, the remaining solution is stripped of propane, leaving only the lightest of oil fractions. The propane recovered is compressed and returned to the start of the process.

The propane deasphalting process is still in use today for the refining of lubricating oils. From a historical perspective, the process is important because it provided the impetus for other workers to study the unique properties of near-critical fluids and SCFs.

APPLICATION TO FATTY ACIDS AND TRIGLYCERIDES

Outside the petroleum industry there was little application of the solvent properties of propane until some pioneering work by Hixson and colleagues (*9*, *10*). Their findings showed that the solvent properties of propane could be applied to compounds of other than petroleum origin.

In their initial study, Hixson and Hixson (*9*) conducted a semiquantitative investigation into the solubilities of several phthalate esters, long chain fatty acids, and aromatic compounds in propane. A selection of results is provided in Table 1. Further investigation was then made into the solubilities of oleic and abietic acid in propane in the subcritical region. It was found that oleic acid was completely miscible up to a tem-

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TABLE 1
Solubility Results for Several Organic Compounds in Propane (*9*)

Compound	Critical solution temperature (°C)	Comments
Dimethyl phthalate	100.5	Very slightly soluble
Dipropyl phthalate	106–107	Soluble
Dioctyl phthalate	105+	Soluble
Myristic acid	104	Soluble
Stearic acid	97	Soluble
Oleic acid	90+	Soluble
Abietic acid		Limited solubility—continual precipitation above 50°C
Naphthalene		Slightly soluble at room temperature: soluble at 50°C
Phenol		Insoluble

perature of 91.1°C while abietic acid was ~10% soluble at 30°C* which decreased to 1.4% at 96°C.

In a subsequent investigation, Hixson and Bockelmann (*10*) examined the solubilities of various triglycerides, long chain fatty acids, and oils of vegetable origin in near-critical propane. An effort was made to determine the relationship between the triglyceride structure and the critical solution temperature† (CST) of the triglyceride/propane binary. It was found that each additional methylene group present in each of the triglyceride chains lowered the CST by approximately 3°C. That is, the longer the triglyceride chains, the larger the region of liquid-liquid immiscibility.

THE SOLEXOL PROCESS

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Several years after the development of the propane deasphalting process a commercial process for the countercurrent extraction of animal, vegetable, and marine oils using a liquid propane solvent was developed. The developmental work followed on from the observations of Hixson and coworkers (*9, 10*) relating to triglyceride solubility.

The Solexol process has been described in detail by Passino (*5*) and Dickinson and Myers (*11*). A schematic of the Solexol process is presented in Fig. 5. The feed oil (be it fish, vegetable, or animal) is fed into a series of extraction columns from storage tanks where temperature variations between columns as well as variations within each column can influence the solvent power of the propane solvent as was previously noted in the propane deasphalting process.

Passino (*5*) stated that two characteristics of a compound control its degree of solubility in propane:

(1) Molecular weight—As was observed by Hixson and Bockelmann (*10*), the longer the triglyceride backbone, the lower the CST; that is, the less soluble the triglyceride.

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*If no pressure figure is mentioned, the pressure corresponds to the vapor pressure of the pure propane fluid at the given temperature.

†The temperature at which liquid-liquid immiscibility first appears. The system pressure corresponds to the vapor pressure of propane at that temperature. This restriction means that the CST does not necessarily correspond to the lcep, which generally occurs at a slightly lower pressure.

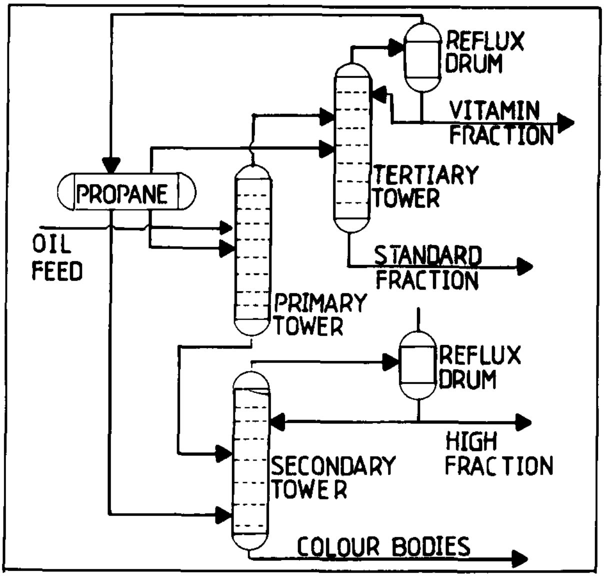

FIG. 5. Schematic of the Solexol process.

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(2) Degree of complexity—The greater the number of active sites on the triglyceride backbone, the lower the CST. Thus, the less saturated the triglyceride, the less soluble it is likely to be.

Thus by staging the temperature and pressure condition in the extraction columns, it should be possible to effect a separation on the basis of chain length and degree of saturation.

As has already been pointed out from the results of Hixson and coworkers (*9, 10*), the solubility of triglycerides in propane is generally higher at the lower temperatures (in most cases the liquids are miscible at room temperature). As the temperature of the liquid propane is increased, its density decreases. The consequent decrease in solvent power results in the precipitation of the least soluble triglycerides followed by the more soluble as the temperature is further increased. This behavior is known as retrograde behavior. By utilizing a column pressure in the vicinity of the critical point where density versus temperature relationships are at their most sensitive, it is possible to separate out oil fractions by staging the temperature above the column length. It is therefore possible to provide a reflux by employing an elevated temperature at the top of the column, inducing the triglycerides of longer chain length and lesser degree of saturation to precipitate out and wash down the column.

By further processing the initial fractions isolated in the primary tower through the secondary and tertiary towers, it is possible to separate a vitamin A concentrate, color bodies, as well as high and low iodine fractions.

CURRENT WORK

The many advantages that accompany the use of supercritical CO_2 (low cost, low toxicity, and low critical temperature) have resulted in the majority of the published work concentrating on the use of CO_2 solvent. In particular, the extraction and fractionation of oils from both plant (*12–17*) and marine sources (*18, 19*) has received considerable attention.

Results so far published indicate that the solvent capacity of carbon dioxide is quite low when extracting triglycerides. A selection of solubility data is presented in Figs. 6, 7, and 8. All data sources indicate that solubilities are in the order of 1–3% by weight, with the higher solubilities occurring at elevated pressures (300–500 bar).

The solubilities obtained in the extraction of triglycerides from natural sources are considerably higher than those typically encountered in solubility trials conducted with pure compounds (Fig. 7). Bamberger (*20*), in commenting on discrepancies between solubility values obtained in their study and an earlier investigation conducted by Chrastil (*21*), speculated that highly soluble impurities present could possibly act as entrainers, thus increasing individual component solubilities. This observation suggests that solubility trials of pure triglycerides may tend to underestimate solubilities of triglycerides obtained from natural sources.

The amount of work that has taken place utilizing propane as the supercritical/near-critical solvent is by contrast quite limited. Coorens and coworkers (*7*) determined the PTX diagram for the propane/tripalmitin binary system. As was found in the work of Hixson and Bockelmann (*10*), Coorens and coworkers (*7*) discovered within the propane/tripalmitin phase diagram a region of immiscibility. The lcep was determined to be located at a temperature of 349 K and pressure of 28.8 bar. It is thus possible to obtain complete miscibility of propane and tripalmitin in the liquid phase at quite moderate temperatures and pressures—well below that associated with the use of CO_2. Upon entering the two-phase region, the solubility of tripalmitin is still moderately high. For in-

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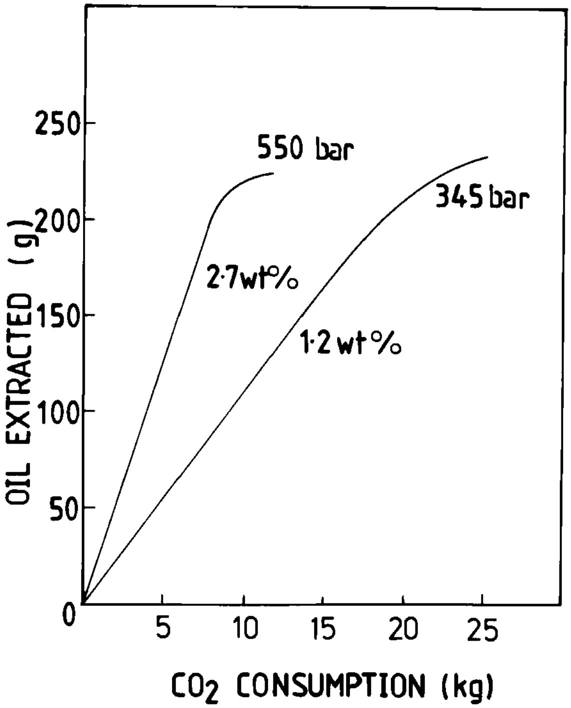

FIG. 6. Supercritical CO_2 extraction of 8% tempered and flaked dry, milled corn (*15*).

stance, at 30 bar and 350 K the propane-rich phase still contains approximately 15% by weight tripalmitin. The results of Coorens and coworkers (*7*) appear to indicate that the solvent capacity of propane with respect to triglycerides is considerably greater than that of CO_2 at considerably lower pressures.

A good solvent, however, requires both capacity and selectivity. Several articles have been published on the selectivity of CO_2 toward fatty acid esters, particularly esters of those acids found in marine oils (*18, 22–24*). In today's health conscious society, though, it is more desirable from a marketing standpoint to be able to extract and fractionate the various triglycerides directly without prior chemical processing.

Solubility data for tripalmitin (PPP), trilaurin (LLL), and trimyristin (MMM) (*20*) indicate that CO_2 should be selective in extracting the three triglycerides, with the solubilities of LLL being approximately an order of

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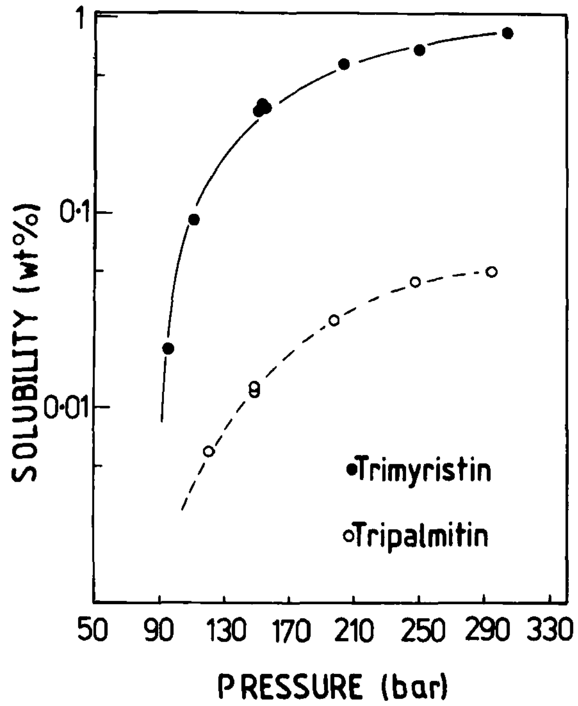

FIG. 7. Solubilities of trimyristin and tripalmitin in supercritical CO_2 at 40°C (*20*).

magnitude greater than MMM which in turn is another order of magnitude greater than the solubility of PPP. However, results presented in the same paper for ternary systems (PPP-LLL-CO_2, MMM-LLL-CO_2, etc.) and the quaternary system (PPP-LLL-MMM-CO_2) showed that the additon of other solutes brought about little change in the solubility of the more soluble component but increased the solubility of the less soluble component by up to an order of magnitude, thus considerably reducing selectivity.

The composition of triglyceride extracts taken from a mixture of various oils (50% palm oil, 25% linseed oil, 25% safflower oil%) extracted with CO_2 is presented in Table 2. Analyses were conducted on esters of triglyceride samples extracted at various temperatures and moderate pressures. Very little selectivity is apparent with respect to chain length or degree of saturation, even though the solubilities are only in the order of 1 wt%.

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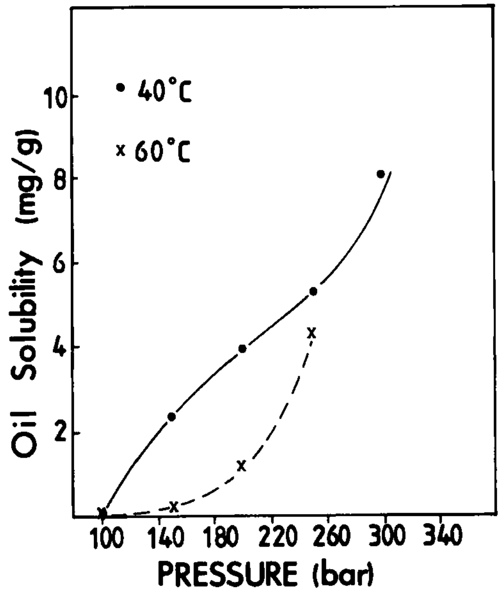

FIG. 8. Solubility of 50% palm oil/25% linseed oil/25% safflower oil mixture in supercritical CO_2 (K. Tilly, University of New South Wales).

TABLE 2
Analysis of Triglyceride Fractions Extracted from a 50% Palm Oil/25% Linseed Oil/25% Safflower Oil Mixture by Using Supercritical Carbon Dioxide

Temperature (°C)	Pressure (bar)	Analysis					
		14:0	16:0	18:0	18:1	18:2	18:3
Starting material		0.6	27.2	3.3	27.6	28.5	12.2
40	265	0.8	27.9	2.9	27.7	27.6	12.2
50	275	0.9	29.5	3.0	28.0	26.8	11.1
60	270	0.9	31.3	3.2	29.5	25.2	9.3
	285	0.8	30.1	3.1	28.2	26.2	10.7
70	290	0.8	27.2	3.3	27.6	28.5	12.2

Data relating to the selectivity of propane is not currently available for triglyceride extractions. The findings of Hixson and Bockelmann (*10*) that the location of the CST is dependent on the degree of saturation and length of the triglyceride chains indicates that selectivity on the basis of these two conditions is possible. By progressing from a region of immiscibility through the CST corresponding to triglyceride "A," a phase rich in "A" will precipitate, leaving a lighter phase rich in the other triglycerides but lacking in "A." Further heating of this phase can then take place to precipitate other triglyceride fractions.

CONCLUSIONS AND SCOPE FOR FUTURE WORK

The advantages of utilizing SCFs as extraction solvents are many and well known. The ease of solute-solvent separation, low toxicity, and the ability to vary the solvent's capacity make the use of SCFs particularly attractive to the food industry and specifically to the area of triglyceride extraction.

Recent material published has been primarily concerned with the supercritical CO_2 extraction of triglycerides and related compounds. The data published suggest that solubilities of 1-3 wt% can be expected with operating pressures in the vicinity of 300-500 bar—conditions which will entail considerable capital and compression costs.

Preliminary work on pure triglycerides suggests that CO_2 is quite selective toward triglycerides, but further work on both ternary systems and more complex mixtures indicates that the selectivity is not as pronounced when many triglycerides and impurities are present.

Investigations carried out using high pressure propane as the solvent both currently and during the development and operation of the Solexol process in the 1940s and 1950s indicate that substantially higher loadings can be achieved at considerably lower pressures (approximately 15 wt% at 30 atm, 350 K), thus substantially reducing capital and compression costs. Although data are scarce, it is also believed that triglycerides can be fractionated on the basis of chain length and degree of saturation by utilizing relationships between lcst location and triglyceride structure as postulated by Hixson and Bockelmann (*10*).

There is enormous scope for future work in this area. Among the topics that need to be investigated are:

(1) The effect of chain length and saturation on propane/triglyceride phase behavior.

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(2) The phase behavior of propane and various "impure" triglycerides (e.g., PPL, PLL, PLM, etc.) much as they are found in nature.
(3) The selectivity of propane when extracting from triglyceride mixtures.
(4) The possible entrainer effects of impurities commonly found in triglyceride sources in both CO_2 and propane/triglyceride systems.

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