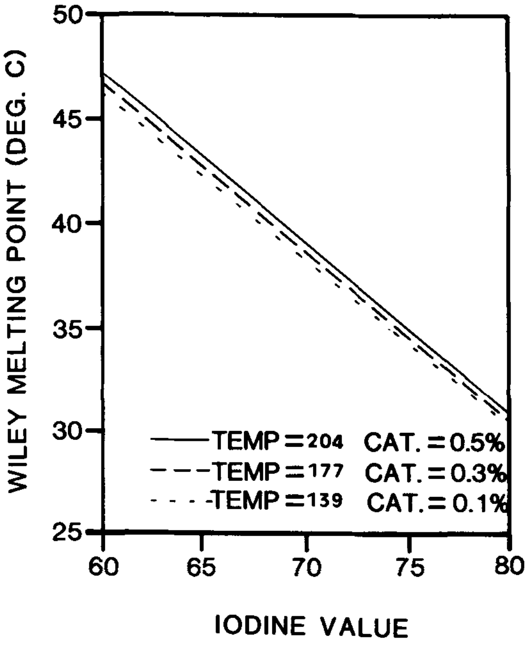

FIG. 5. Wiley melting point (°C) as a function of iodine value, based on model predictions.

quantitatively the reaction rate, *trans*-isomer content and melting point is valuable for improving process and product development. Models for other oils or products could be developed easily, using the techniques presented here.

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High Pressure Extraction of Oil Seed[1]

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ABSTRACT

For calculation of phase equilibria of the system seed oil/CO_2, an equation of state published in the literature has been fitted to experimental data of that system. The results thus obtained are of only limited use in designing a supercritical extraction process. The experimental investigation of the mass transfer kinetics is much more significant. Mechanical processing of the oil seed's cell wall structure has been shown to be of great importance. The best specific yields were achieved with material that had been mechanically pre-deoiled and thereby broken open. Yields are increased considerably by use of the gas mixture CO_2/propane or other special gas mixtures or by the addition of refrigerants. However, the extraction times achieved in batch operation, together with the mass product nature of oil seed, make a continuous supercritical extraction essential if operation is to become economic relative to the conventional hexane extraction. To this end, the energetics of the process have been calculated, and practical possibilities for continuous operation are discussed.

INTRODUCTION

During the past two decades, the development of high pressure extraction has resulted in intensive research in the processing of natural products (1). The solvent CO_2 has become important for reasons of safety and economics. The extraction of solids is especially of interest for materials which are present in small amounts and which yield high quality products after extraction. The reason lies in the relatively high investment and energy costs of a high pressure plant, where at present only batch processing is possible, with the associated loading and emptying times. Because of the problems that have been encountered in extraction of the mass-product oil seed with conventional solvents the question has been raised whether high pressure extraction, using solvents which do not pose safety and health hazards, might replace the conventional, potentially explosive, hexane extraction.

COMPARISON OF HEXANE EXTRACTION AND HIGH PRESSURE EXTRACTION

In conventional oilseed processing, a distinction is made between oil-rich seeds (for example rape, corn germ, sunflower) and less oil-rich seeds (for example soybean, cottonseed) (Fig. 1). The latter generally are extracted directly, after mechanical and thermal treatment to break open the cells. The most significant oil crop is without doubt the soybean, for which the worldwide seed oil production was 15 million tons in 1982 (2). Seeds with a higher oil content, on the other hand, are mechanically pre-deoiled in a cage screw press prior to extraction. In the case of rapeseed, which is increasingly important in the European market, the resulting press cake has a residual oil content of 18 to 20%. This material is extracted afterwards; the oil thus obtained must be separated from solvent by distillation,

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and the remaining solid material, the meal, is freed from adhering solvent in a desolventizing stage. The two processes, distillation and desolventizing, could be omitted if a supercritical gas were used as the solvent. The disadvantage is that the phase equilibria so far measured for seed oil/supercritical fluid show rather low oil concentrations in the supercritical fluid phase at the pressures and temperatures, about 300 bar and 20 to 80 C, typical for high pressure extraction. In CO_2, for example, the values are less than 1% by mass. In comparison, the fraction by mass of oil in hexane in the conventional miscella extraction (3) is 30 to 35%, so that extraction times are considerably shorter. The most significant industrial advantage of hexane extraction lies in the continuous processing of 1000-2000 T daily.

AIMS

In order to help determine if supercritical fluid extraction can, in the future, be used for the production of edible oils, we have carried out the following theoretical and experimental work:

- Calculation of phase equilibria in seed oil/compressed fluid systems, taking into account available experimental data.
- Improvement of the mass transfer kinetics during the extraction process.
- Energetic investigation of various process variants in comparison with conventional hexane extraction.
- Development of a continuous high-pressure extraction of oil seeds.

CALCULATION OF PHASE EQUILIBRIA

Hederer (4) has developed a modification of the Redlich-Kwong equation especially for calculation of phase equilibria in highly compressed fluids:

$$p = \frac{R\ T}{V - b} - \frac{a}{V\,(V + b)}. \qquad [1]$$

This equation of state gives the pressure p as a function of temperature T and of molar volume V, using the universal gas constant R, the attraction parameter a and the covolume b. In multi-component systems, a is dependent on temperature and composition of the system, while b depends on composition alone. a and b are calculated from the coefficients a_{ii}, a_{jj} and b_i of the pure substances according to the usual mixing rules:

$$a = \sum_{i=1}^{N} \sum_{j=1}^{N} x_1\ x_j\, a_{ij}\,(T) \qquad [2]$$

$$b = \sum_{i=1}^{N} x_i\, b_i \qquad [3]$$

and the combination rule

$$a_{ij}\,(T) = [a_{ii}\,(T)\ a_{jj}\,(T)]^{0.5}\,(1 - \theta_{ij}) \qquad [4]$$

together with the mole fractions x_i, x_j. N is the number of components in the system, and θ_{ij} is the binary interaction parameter. The temperature-dependence of the coefficients a_{ii}, a_{jj} is given by

$$a_{ii} = a_i^*\ T^{\alpha_i};\ a_{jj} = a_j^*\ T^{\alpha_j}. \qquad [5]$$

In order to use the modified Redlich-Kwong equation [1] to [5] on multi-component systems, the values of a_i^*, a_j^*, α_i, α_j, b_i and b_j must be known for each component, together with the binary interactions parameters θ_{ij} for each possible binary system within the complete system.

The equation of state is employed here for the calculation of phase equilibria of seed oil/CO_2 system. A seed oil is a complex mixture of various substances. In the calculation, this multiple component mixture is treated as a single substance, so that the system seed oil/CO_2 is regarded as a quasi-binary system.

The coefficients a_i^*, α_i and b_i for the solvent CO_2 were taken from Brunner (5). The coefficients a_j^*, α_j and b_j for seed oil were at first calculated from the empirical expressions given by Hederer (4), and the binary interaction parameter θ_{ij} was fitted to experimental phase equilibrium data (6-9). The experimental data were reproduced very inaccurately due to the poorly characterized properties of seed oil. For this reason, the coefficients of seed oil, together with the binary interaction parameter, were fitted to Quirin's (6) phase equilibrium data on the dew line (i.e., solubility data).

The results of this optimization, i.e. the phase equilibrium (solubility) data which are calculated with the modi-

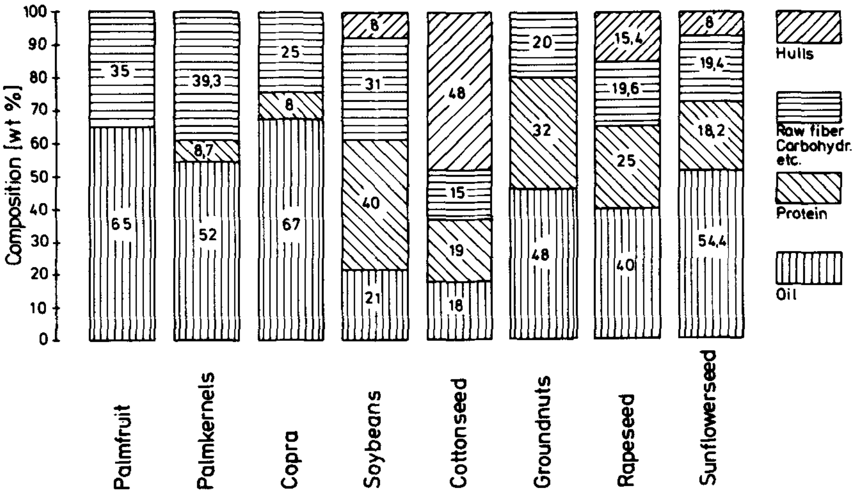

FIG. 1. Composition of prepared seeds and fruits (dehydrated basis).

fied Redlich-Kwong equation with the thus determined coefficients and interaction parameters, are illustrated for the system soybean oil/CO_2 in Figure 2 together with the experimental data from Quirin (6). Only part of the data shown in Figure 2 is represented by the equation of state within their experimental error. The equation does not reproduce the measured solubility maxima. The values for low extraction temperatures ($T \leqslant 300$ K) and pressures ($p < 300$ bar) are relatively imprecise. A computer calculated working diagram is shown in Figure 3. In addition to the above-mentioned inaccuracy in reproducing the data, it must be mentioned that the molecular mass of seed oils needed for the calculation fluctuates according to the proportion of mono-, di- and triglycerides, and according to the composition of the constituent fatty acids. Figure 4 shows the influence of molecular mass variation.

In summary, even after optimization of an equation of state that has been introduced specifically for supercritical fluid extraction, the solubility of seed oil in CO_2 cannot be calculated with sufficient accuracy to describe seed oil processing. The calculated data can serve as rough estimates of the solubility. It therefore still remains necessary to measure phase equilibria for the case of specific interest and to carry out experiments on the kinetics of mass transfer.

INVESTIGATION OF MASS TRANSFER KINETICS

Methods of improving mass transfer kinetics were examined in order to achieve shorter and therefore more economic extraction times. Experience in the solid-liquid extraction of oil seed with hexane shows that every possible oil cell must be broken open before the extraction, to facilitate mass transfer over the shortest possible diffusion distance. Figure 5 shows the structure of such an oil cell. The cell walls are embedded in a matrix of fibrous material. Figure 6 shows the cross-section through a single rapeseed cell. The oil is present in small droplets [0] of about 1 μm in diameter, which are distributed throughout the entire cell. The protein bodies [P] have diameters up to 10 μm and contain embedded phosphate salts, the globoids [G] (10). It is clear that the cell wall must be disrupted if extraction is to be successful.

In a test series of high pressure extractions of rapeseed,

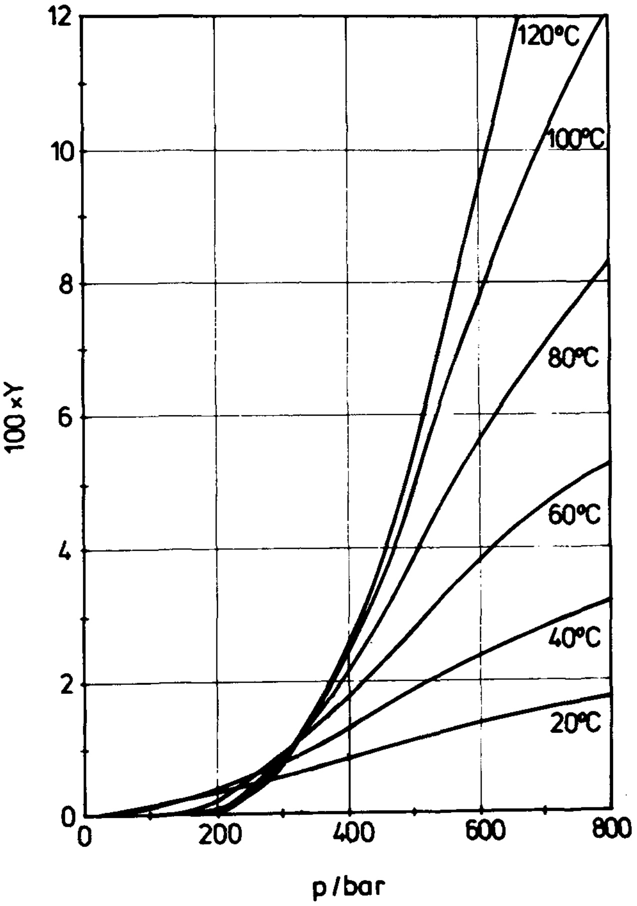

FIG. 3. Solubility Y of soybean oil in CO_2 as a function of pressure p at different temperatures. Calculated data.

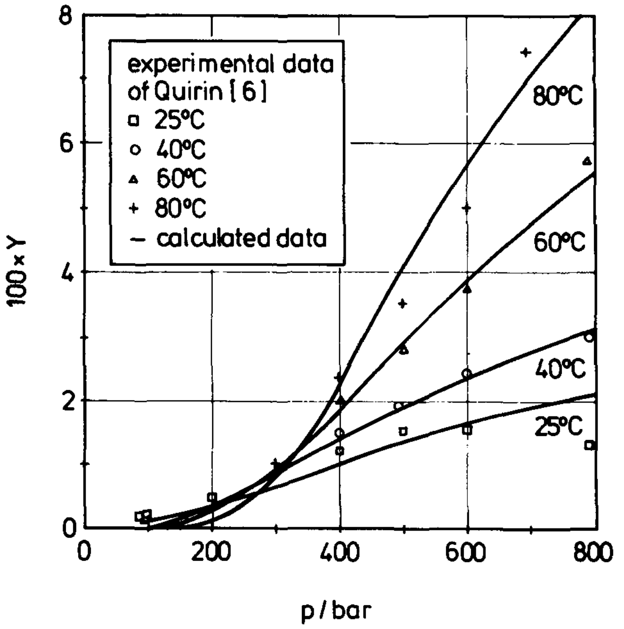

FIG. 2. Solubility Y of soybean oil in CO_2 as a function of pressure p at different temperatures. Comparison between calculated and experimental data.

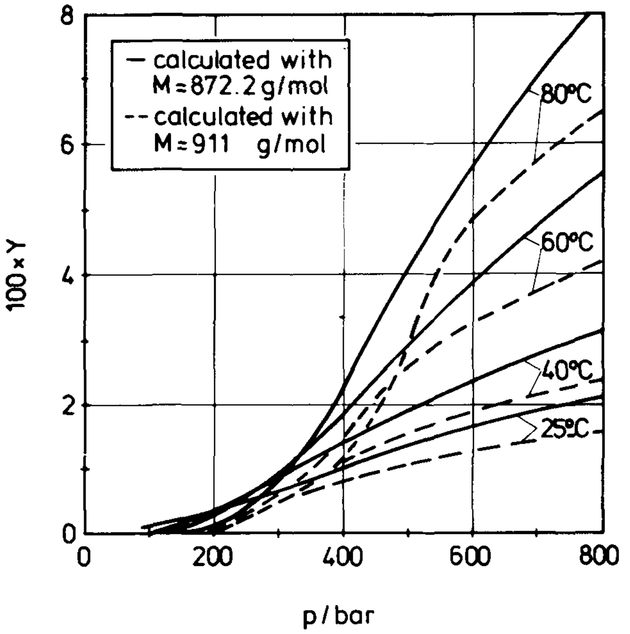

FIG. 4. Solubility Y of soybean oil in CO_2 as a function of pressure p at different temperatures. Influence of the molecular mass on calculated data.

the effect of different preparative sequences on the extraction behavior has been examined:

- Extraction of non-decorticated, flaked rapeseed.
- Extraction of decorticated (11), unflaked rapeseed.
- Extraction of decorticated, flaked rapeseed.
- Extraction of decorticated rapeseed that was previously subjected to "cell cracking" by means of sudden pressure reduction (12).
- Extraction from flaked rapeseed press cake.

Rapeseed is flaked on a flaking mill with a gap of 0.1 to 0.2 mm. The flaked rapeseed has a bulk density of 312 kg/m^3, an oil content on dry basis of 42% by mass and a water content of 9% by mass. The flaked rapeseed press cake has a bulk density of 561 kg/m^3, an oil content on dry basis of 22% and a water content of 11 mass-%.

The resulting extraction curves are shown in Figure 7.

The mass of extract is plotted against the mass of solvent CO_2. The limiting solubility is derived from the initial linear portion of the curve. Comparisons were made under extraction conditions of 750 bar and 40 C. The dashed line is the ideal extraction curve for phase equilibrium through-

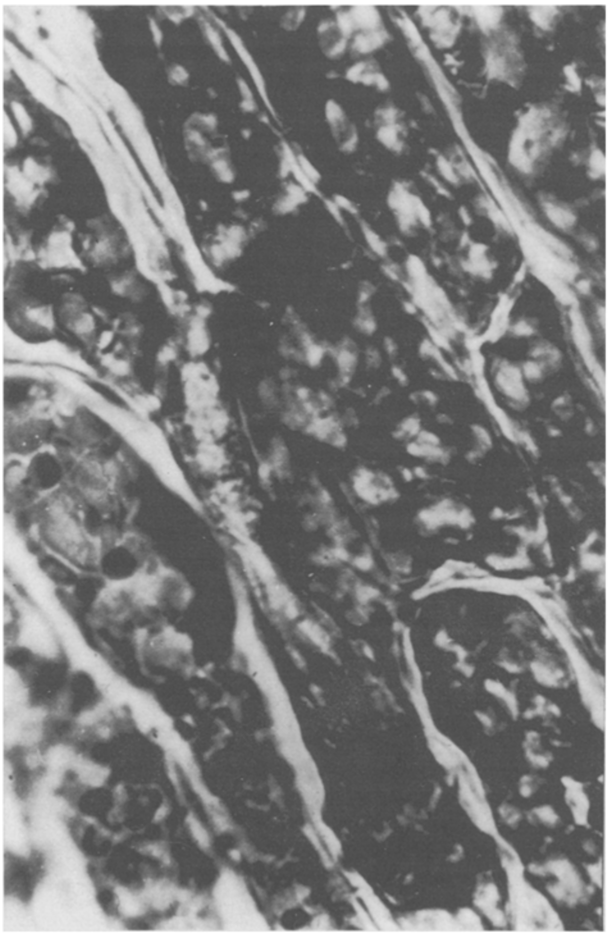

FIG. 5. Seed structure.

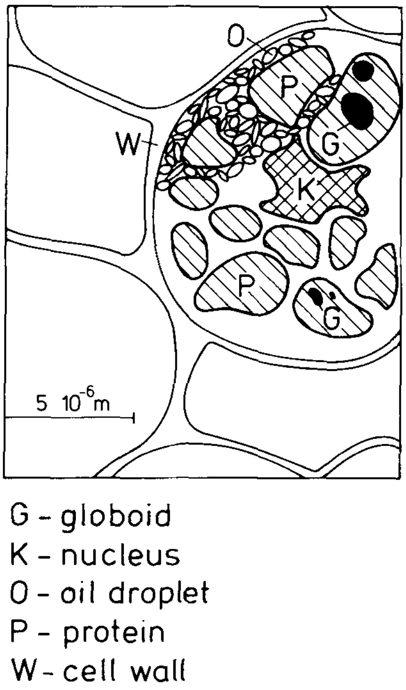

FIG. 6. Microsection of rapeseed.

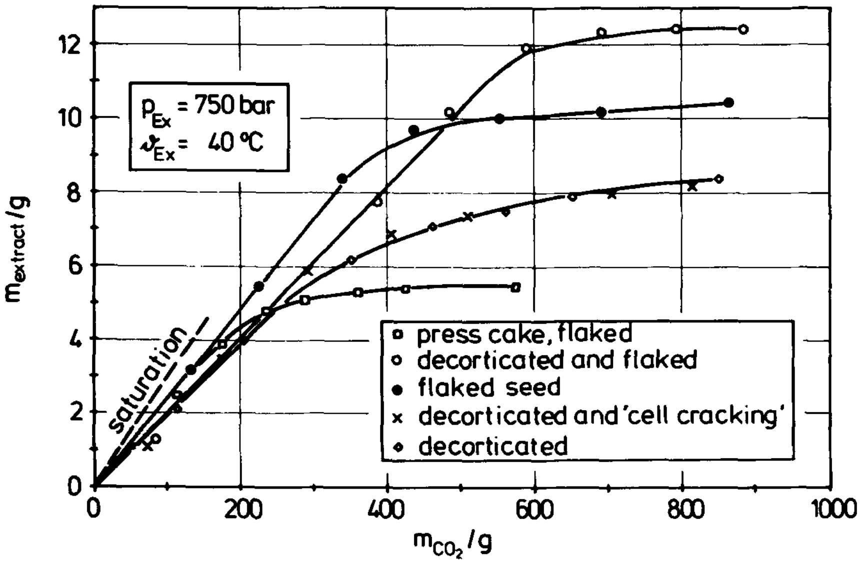

FIG. 7. High pressure extraction of differently prepared rapeseed.

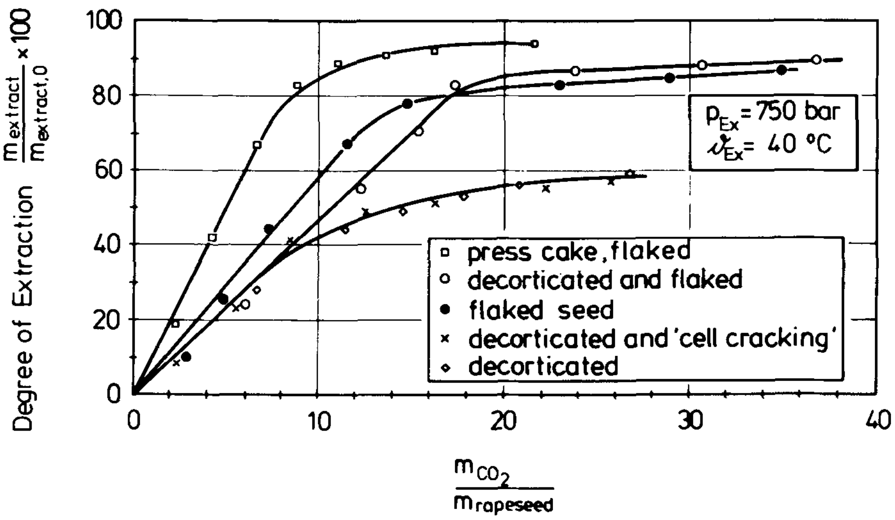

FIG. 8. High pressure extraction of differently prepared rapeseed.

out the extraction. In all cases, a constant loading between 2.3 and 2.5% is reached initially, compared to the calculated equilibrium value of 3%. On the other hand, the duration of this constant loading varies, taking into account the proportionality of CO_2 amount (abscissa) to extraction time. The earliest decrease in mass transfer occurs, as expected, when unflaked material is processed. Decortication alone does not break open the cells. When the autoclave was emptied at the end of the experiment, cell bursting could be heard clearly during outgassing of the CO_2. Unexpectedly, a similar extraction curve results for the material subjected to "cell cracking" by rapid pressure reduction from 100 bar to ambient pressure. In addition, the extraction autoclave was depressurized at three separate times during the experiment, to disrupt any gas channels formed. However, this brings no improvement in mass transfer relative to the untreated material. This is explained in terms of the cell structure (Figs. 5 and 6) if "cell cracking" disrupts only some cell walls, or disrupts them insufficiently. This process, in contrast to mechanical flaking, does not reduce the diffusion paths, and no additional diffusion paths are opened. These mass transfer kinetics show that the diffusion resistance of the natural product matrix is at least of the same magnitude as the transport resistance of the cell wall. Mass transfer is correspondingly better after mechanical treatment of the seed. The duration of saturation loading is considerably increased when flaked rapeseed is extracted, and the overall extraction time is correspondingly shorter. Decortication before flaking further increases the duration of constant loading. This must, however, be attributed to the higher initial oil concentration of the decorticated material.

The effort of extraction is seen clearly in the dependence of the degree of extraction $E = m_{extract}/m_{extract,\,0}$ on the specific solvent requirement $m_{CO_2}/m_{rapeseed}$ (Fig. 8). Here, too, it is seen that cell cracking brings no improvement. The advantage of decortication is cancelled out by the higher initial oil content of the starting material. Mechanically predeoiled rapeseed press cake is most readily extracted, this being the result of the intense shearing and compression forces produced in the mechanical press. If the degree of preparation AG is defined as

$$AG = 1 - z/x$$

where x is the oil content of the native seed and z is the oil content which depends on the length of the screw press, the value for a mechanical pre-press is over 90%. Figure 9 shows the observed degree of preparation of press material during operation for rapeseed. This result, of advantage for the mass transfer kinetics of high pressure extraction, is not achieved by flaking in a rolling mill with or without decortication, by decortication alone, or by cell cracking.

To determine whether mechanical pre-deoiling remains desirable, or whether direct extraction with supercritical gases is superior, extraction experiments with supercritical CO_2 at 300 bar and 40 C were performed on mechanically pre-deoiled rapeseed press cake. The CO_2 requirement in

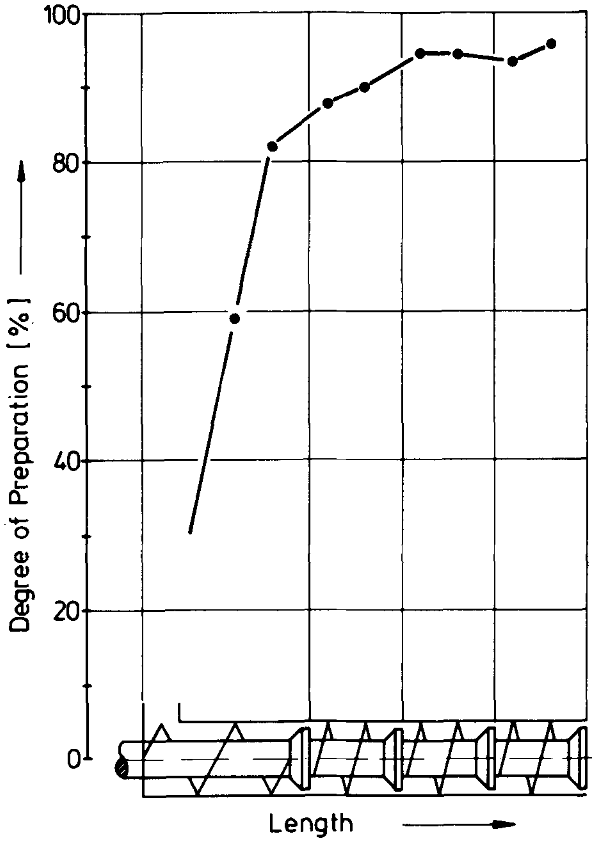

FIG. 9. Degree of preparation of press material during operation (rapeseed).

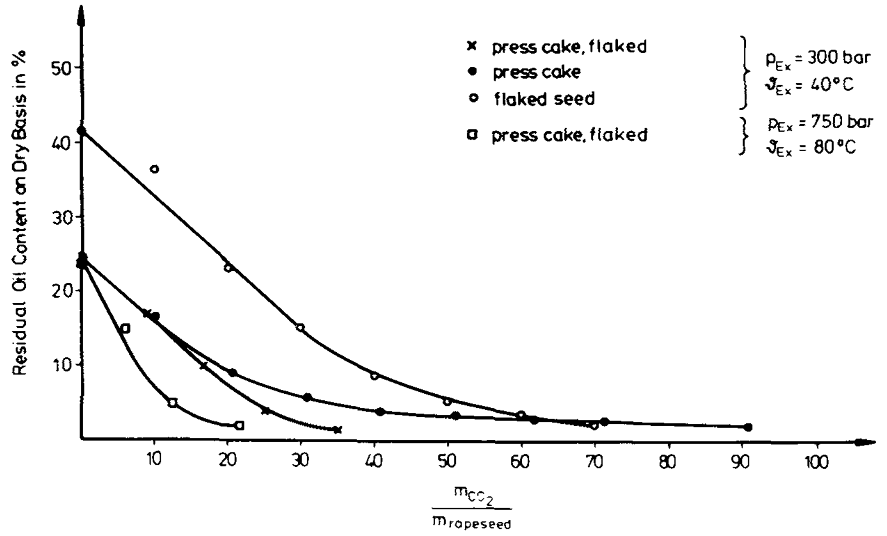

FIG. 10. Specific solvent requirement in the high pressure extraction of rapeseed.

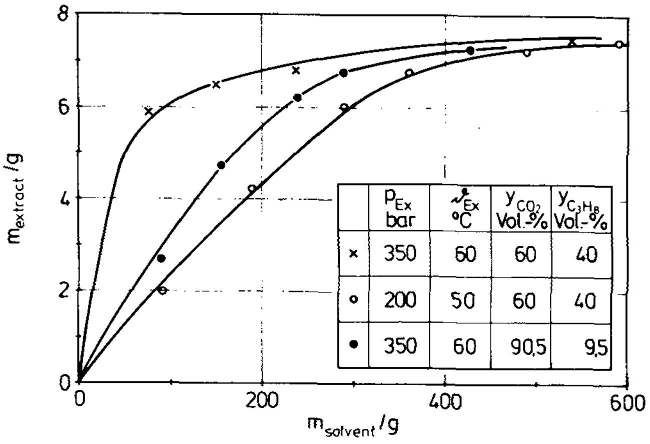

FIG. 11. High pressure extraction of rapeseed press cake with CO_2/propane gas mixture.

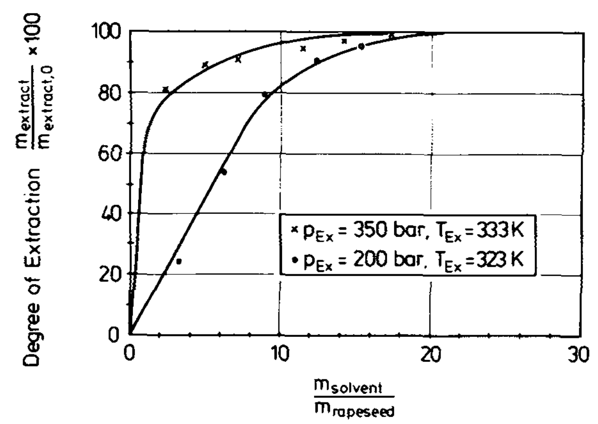

FIG. 12. High pressure extraction of rapeseed press cake with a gas mixture of CO_2:propane (60:40, v/v).

terms of the mass of starting material is shown in Figure 10 for de-oiling down to a residue of 2% by mass. Pressing extraction of 1 t rapeseed yields 750 kg of press cake, so that the direct extraction of 1 t of rapeseed requires a throughput of 70 times this quantity of CO_2, a value which is reduced to 26 times when press cake is extracted. In both cases, the same ratio of CO_2 mass flow to mass of starting material was used (10 kg CO_2/h/kg starting material), so that a shorter specific extraction time and smaller extraction autoclaves are needed to process rapeseed press cake. This clearly shows that mechanical pre-deoiling remains of advantage in the supercritical extraction of oilseed.

To improve the kinetics of mass transfer further, rapeseed press cake was extracted with a gas mixture containing 60% CO_2 and 40% propane by volume. The results are shown in Figure 11. At the start of extraction, saturation loads are achieved which are considerably in excess of the loadings for the seed oil/CO_2 system, which also are shown. It is apparent that the solvent requirement for complete extraction at 350 bar is practically the same as at 200 bar, although the solubility at 350 bar is considerably greater. This is seen more clearly in Figure 12, where the results are presented in terms of specific mass. The solvent requirement is about 20 kg gas mixture/kg rapeseed press cake, or 15 kg gas mixture/kg rapeseed if mechanical prepressing is taken into account.

The use of propane, however, is less safe than that of CO_2 alone. The inert CO_2 reduces the danger of explosion, but the lower flame limit of the mixture must be taken into account, because leaks or de-gassing procedures can bring the mixture into contact with air. For the system air/propane/CO_2, the maximum permissible propane concentration in the initial CO_2/propane mixture is derived from Figure 13 (13). The diagram shows the confluence of lower and upper flame limits in the presence of an inert gas—here CO_2—at ambient conditions. If a tangent to the lower flame limit is drawn through the point 100% air, a mixture by volume of 12.1% propane and 87.9% CO_2 is found on this line for the air-free system. The admixture of air, caused for example by a leak into the surroundings, can never create an inflammable mixture if this initial concentration is used.

The result of an extraction with 9.5% propane and 90.5% CO_2 (at 350 bar and 60 C) is, therefore, included in Figure 11. The initial constant load of 3% is a significant improvement on the use of pure CO_2 (about 1%). The addition of this safe proportion of propane results in an overall extraction comparable to that achieved with CO_2 alone at an extraction pressure of 750 bar.

Finally, solvent mixtures of CO_2 with Frigen have been investigated. These also greatly increase the yield of extracted seed oil (14). Disadvantages are that distillation of the oilseed-solvent mixture and the removal of Frigen from the extracted meal become necessary, and a residue-free product is no longer guaranteed.

The following conclusions may be drawn from these experiments to improve the extraction kinetics of the supercritical processing of oilseeds:

1. The mechanical treatment is of great importance for the yield of oil.
2. Extraction of material pre-deoiled in a cage screw press is preferable to direct extraction of oilseeds.
3. The use of propane/CO_2 mixtures or other gas mixtures with a high loading of seed oil to increase the yield and to shorten the extraction time brings possible disadvantages in safety, production and product quality. Nevertheless, even small, safe quantities of such additives significantly increase the yield.
4. To reduce the extraction time for oilseeds, only the initial linear extraction curve at saturation loading should be used. Either a higher residual oil concentration in the extraction residue must be accepted, or a plant must be designed in which oilseeds are transported continuously into, through and out of a pressure vessel.

ENERGY COMPARISON WITH THE CONVENTIONAL HEXANE EXTRACTION

Now, the energy requirement per unit mass of starting material will be considered. A distinction is made between mechanical and thermal energy, which is further subdivided into the requirements for heat addition (heating) and heat removal (cooling). Various stationary-state high pressure extraction cycles are compared, making use of the law of conservation of energy and the T, s-diagram of CO_2 or new tables of its thermodynamic properties (15).

The non-stationary compression and decompression conditions arising in the batch processing of solids are neglected.

The energy considerations are based upon the observations on rapeseed press cake with CO_2 at 300 bar and at 750 bar (Fig. 14). The operating schemes listed in Table I (sub- or supercritical separation, compression by liquid-gas pump and by gas compressor) are investigated and compared with the results for the conventional hexane extraction.

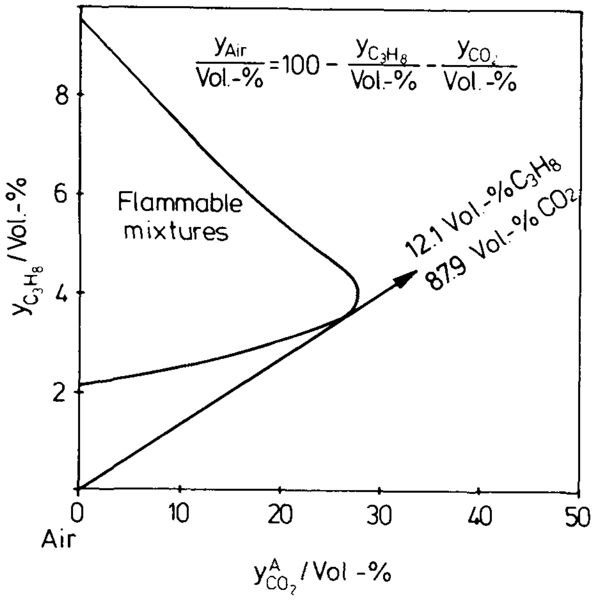

FIG. 13. Limits of flammability of propane/CO_2/air system at atmospheric conditions.

It must be noted that the energy expenditure for mechanical pre-pressing of rapeseed is included under the heading for mechanical energy requirement. This quantity was determined to be 35 kWh/t seed in another experimental series for cold pre-pressing. The values calculated lead to the following conclusions:

- High pressure extraction of oilseed requires considerably more mechanical energy than does conventional hexane extraction.
- The thermal energy requirement favors operation with supercritical separation, because the required thermal energy is introduced directly into the system via the temperature rise during compression.
- The energy requirement for cooling is higher than for hexane extraction, because the high pressure cycles considered make no use of the heat recovery which is used during hexane extraction. Operation with supercritical separation is, nevertheless, comparably good.

Taking the ideal case, the CO_2 is saturated with oil throughout the extraction period; the operating procedures B to I have theoretical energy requirements shown by the dashed lines in Figure 14. In particular, operation at 750 bar becomes energetically comparable to the conventional hexane extraction. It also seems possible that the energy requirement can be decreased using suitable gas mixtures with a higher solubility for the seed oil. Energy considerations show very clearly that high pressure extraction of oilseeds will become economical only when the mass transfer rate, which declines with extraction time, can be

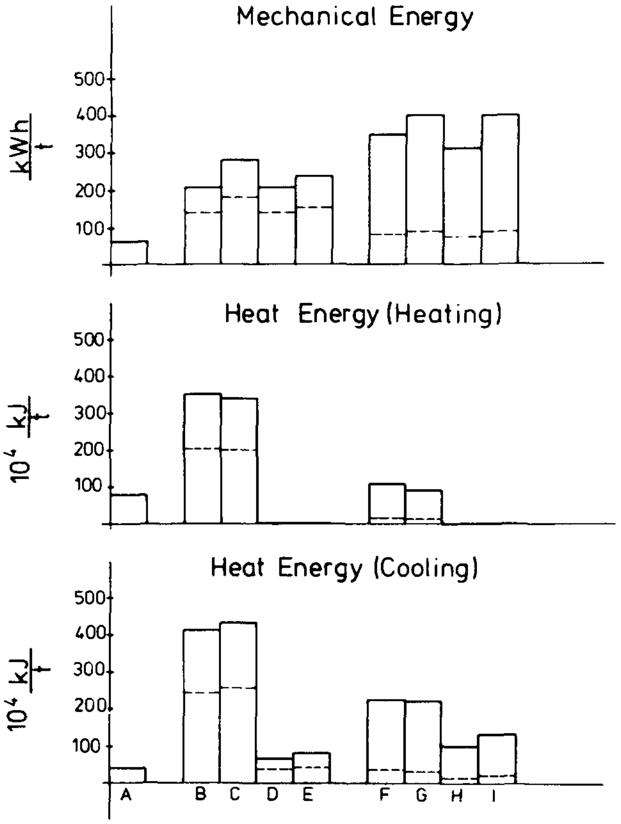

FIG. 14. Energy consumption in various high pressure extraction cycles.

TABLE I

Energy Consumption in Various High Pressure Extraction Cycles

		Mechanical energy kWh/t	Heating $10^4 \cdot$kJ/t	Cooling $10^4 \cdot$kJ/t
A	Hexane extraction	75	75	42
B	$p_{Ex} = 300$ bar; $p_{se} < p_c$; $t_{Ex} = 40$ C ; $t_{se} < t_c$; pump operation	213	352	415
C	as B ; compressor operation	287	341	431
D	$p_{Ex} = 300$ bar; $p_{se} > p_c$; $t_{Ex} = 60$ C ; $t_{se} > t_c$; pump operation	213	–	64
E	as D ; compressor operation	228	–	82
F	$p_{Ex} = 750$ bar; $p_{se} < p_c$; $t_{Ex} = 80$ C ; $t_{se} < t_c$; pump operation	348	113	226
G	as F ; compressor operation	400	92	223
H	$p_{Ex} = 750$ bar; $p_{se} > p_c$; $t_{Ex} = 80$ C ; $t_{se} > t_c$; pump operation	313	–	100
I	as H ; compressor operation	400	–	131

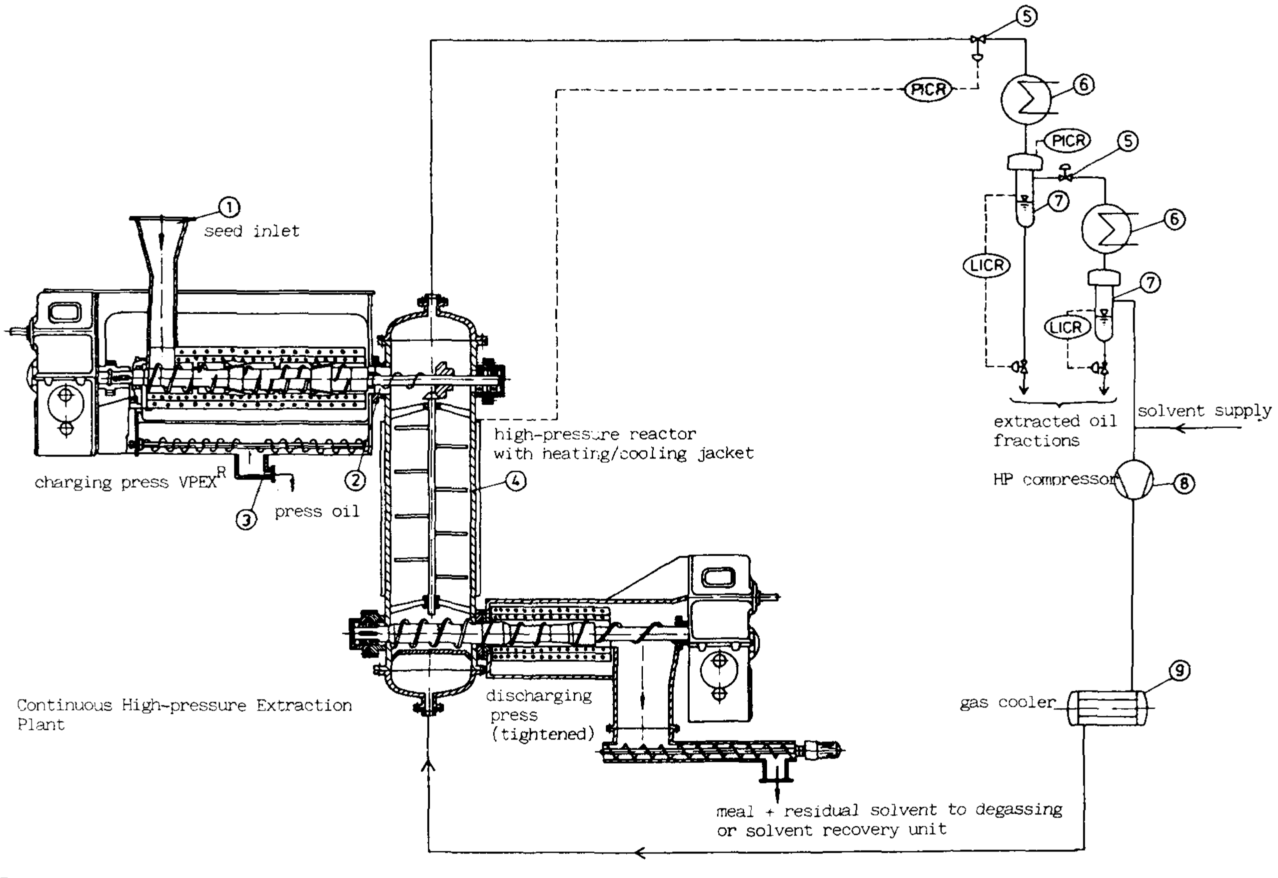

FIG. 15. Continuous high pressure extraction plant.

brought to a constant value which is as close as possible to the saturation solubility. This will be possible only when continuous operation of the supercritical extraction of oilseeds is achieved.

CONTINUOUS HIGH PRESSURE EXTRACTION OF OILSEED

The daily turnover of an oil mill lies between 1000 and 2000 t. Experimental and theoretical investigations of high pressure oilseed extraction clearly show the necessity of continuous operation without loading, de-gassing or emptying periods (16). A quasi continuous process with a series connection of high pressure vessels requires too much expenditure for device and controlling.

The following engineering solutions are available.

- Entry and exit of the solid material through high pressure airlocks, the solid being transported countercurrent to the solvent by means of a transport screw. In

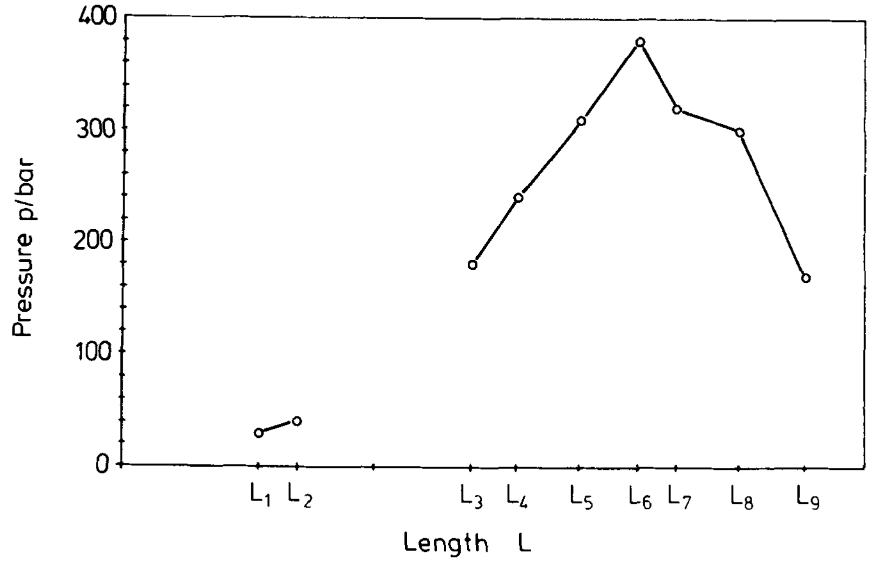

FIG. 16. Pressure distribution in the cage screw press.

principle, this involves quick opening seals that are opened and closed periodically, permitting only quasi-continuous operation. The pressure in the airlocks has to be increased from atmospheric to operating pressure after each entry of material.

- Entry and exit of the seed in mobile pistons, again allowing intermittent operation only, which in addition requires a high degree of process control. Also, a suitable liquid must be found which will convert the seed into a compressible slurry.
- Entry and exit of solid material through a screw press. The advantage of fully continuous extraction is thereby achieved (Fig. 15). The entering seed is mechanically predeoiled by the build-up of pressure in the press. It is of great advantage for the oil quality that this step occurs under exclusion of air directly before the extraction step, so that no additional enzymic reactions can result. The geometry of the screw press can be such that the pressure required for extraction is reached on entry to the extraction autoclave. The solid material can be transported—with the help of a stirrer—countercurrent to the solvent. Exit is possible through a screw press with an enclosed jacket, allowing a steady reduction of pressure. To develop this possibility further, the pressure profiles of various screw geometries are being measured. An example is shown in Figure 16. A steady build-up of pressure is seen, reaching a clear maximum, which is followed by a slower decline in pressure. During the continuous entry and exit of solid material, the maximum is prolonged by the residence time in the extraction autoclave, and the declining portion of the curve represents the separate exit screw.

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