# Carbon Dioxide Extraction of Canola Seed: Oil Solubility and Effect of Seed Treatment

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**The extraction of oil from fixed beds of canola seed (*Brassica napus*) was studied using carbon dioxide at temperatures and pressures ranging from 25 to 90°C and 10 to 36 MPa, respectively. The oil solubility in $CO_2$ was found to be strongly dependent on $CO_2$ pressure and weakly dependent on the system temperature. The highest observed oil solubility was 11 mg/g $CO_2$ and occurred at 36 MPa and 55°C. The manner in which different methods of seed pretreatment (flaking, cooking, pressure rupturing, chopping and crushing) affected the extraction process also was studied. The total amount of oil recovered from the seeds by $CO_2$ extraction was found to be strongly dependent on the pretreatment. No measurable quantity of oil chould be recovered from whole, intact seeds. The amount of oil extractable from flaked and cooked seeds was comparable to that recoverable by conventional hexane extraction.**

The unusual solvent properties of supercritical fluids were first reported during the last century (1). However, extraction technology based on such fluids has developed slowly. This is due, in part, to the difficulty in understanding the behavior of supercritical fluids. Moreover, the price of petroleum-based solvents used in conventional solvent extraction has, until recently, been low. This has discouraged research into alternative technologies. During the last decade, however, this situation changed, and the petroleum solvent costs have, in some cases, increased twentyfold. Furthermore, as shown during the oil crisis of 1973, the supply of these solvents is not always secure.

During the same period, there has been a growing concern regarding health issues. Many food additives have become suspect, and petroleum solvent residues in food products are far less acceptable (2).

Supercritical fluid extraction (SFE) can use safe, low-toxicity gases such as $CO_2$ for the extraction of food products (3,4). Interest in SFE is growing, and some promising results have been obtained. For example, SFE has been used to extract oil from soybeans (5), corn germ (6) and rapeseed (7,8). Because the gases used for extraction are very volatile, virtually no solvent residue remains in the collected extract. Desolventizing, a costly and sometimes lengthy procedure when using conventional solvents, is greatly simplified (9).

Although several gases can be used for extraction, carbon dioxide is most commonly employed. The preference for $CO_2$ is due in large part to its physical and chemical properties. It is nonflammable and nontoxic, its critical temperature and pressure are low (31°C, 7.38 MPa) and its solvent capacity for numerous compounds is appreciable at pressures of 10 to 40 MPa (10,11). These conditions fall well within the capabilities of current technology. Other advantages are the low cost and easy availability of $CO_2$.

In Canada, the largest oilseed crop is canola (composed of *Brassica napus* and *Brassica campestris*). In 1981 over 2.5 million tons of the seed were produced (12). The majority of the seed is processed to obtain canola oil by pressing and/or hot hexane extraction.

This project was undertaken to assess the feasibility and merits of supercritical $CO_2$ extraction as an alternative to hexane extraction in the canola oilseed industry. The project examined SFE of canola seed from both physical and chemical perspectives. The present paper reports some of the physical data obtained from this study.

## MATERIALS AND METHODS

*Extraction equipment.* The basic instrument used in this work was a Model 1081B, Hewlett Packard High Performance Liquid Chromatograph (HPLC), which had been modified to permit operation with liquid carbon dioxide. The general operation of the extraction system is shown in Figure 1. Liquid $CO_2$ from a storage cylinder (a) passes through the shutoff valve (b), through a Nupro 7 μm sintered metal filter (c) and then into the pump head (d) which operates at -5°C. The cooled liquid $CO_2$ then flows through a pressure-flow monitoring device (e) and into the HPLC oven. In the oven, the $CO_2$ temperature can be adjusted to the desired value by passing the fluid through seven m of stainless steel

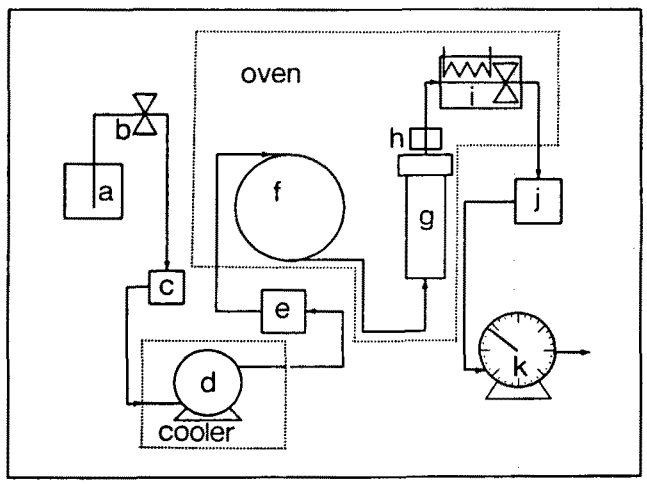


**FIG. 1. Schematic diagram of the experimental supercritical fluid extraction system. a, solvent ($CO_2$) reservoir; b, shut-off valve; c, sintered steel filter; d, diaphragm pump; e, flow and pressure transducer; f, temperature equilibration coil; g, extraction vessel; h, sintered steel filter; i, temperature controlled restricter valve; j, sample collection vessel; k, volumetric flow meter.**

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TABLE 1

**Errors Associated With the Various System Parameters as Determined Experimentally**

| Parameter | Average error |
|---|---|
| Oven temperature | <±2% |
| Extractor pressure | ±2% |
| $CO_2$ flowrate | <±3% |
| Total $CO_2$ volume | <±0.5% |

tubing (f) (0.010″ i.d., 0.062″ o.d. The $CO_2$ subsequently enters the extraction vessel (g) and then passes through a two-μm frit (h) and flow restrictor (i) where its pressure is reduced to about 0.1 MPa. It then flows through 0.6 m of narrow bore (0.25 mm i.d.), fused silica tubing into the sampling section (j) where the liquid solute is collected. The $CO_2$ is subsequently passed through a test meter (k) and finally vented to the atmosphere. A two-channel chart recorder is also incorporated into the system to provide a continuous record of $CO_2$ flowrate and pressure.

Two cylindrical extraction vessels of 12.7 mm i.d. and 82 mm or 114 mm in length were used in this study. The volumes of the vessels were 10.4 ml and 14.4 ml, respectively.

The extraction equipment was calibrated for solvent flowrate, oven temperature and system pressure, and the accuracies of the parameters are listed in Table 1.

*Carbon dioxide.* The carbon dioxide which was used in the experiments (USP Siphon grade), was obtained from pressurized steel cylinders. Each cylinder held 30 kg of $CO_2$ which was withdrawn in liquid form through an educter tube within the cylinder. Approximately 200-300 hr of system run time were obtained with each cylinder.

*Seed samples.* Two principal varieties of rapeseed are grown currently in Canada: the *Brassica campestris* and *Brassica napus.* Genetic strains, or cultivars, within these varieties are numerous due to continuous breeding programs designed to improve both the agronomics of the plants and their adaptability to various soils and climatic conditions. Unlike the seed originally introduced into Canada in 1943, most of the rapeseed presently grown in Canada contains only small amounts of erucic acid and glucosinolates. These modern versions of rapeseed are called canola.

One cultivar of *Brassica napus* was obtained from a commercial processor (CSP Foods, Nipiwan, Saskatchewan) and was used in this work.

## SEED PRE-TREATMENT METHODS

The canola seed used during the course of this work was physically ruptured prior to extraction using five different pre-treatments.

*Crushing.* The seed was crushed using a 20-cm mortar and pestle. Approximately 10 g of whole seed were placed into the mortar. The seed was crushed by hand over a five-min period. The crushed seed material was free flowing and contained particles ranging from ca. 0.1 mm to 0.5 mm in diameter.

*Chopping.* Finely chopped seed material was produced by placing 50 g of whole seed into a two-l Osterizer blender, model "Cyclo-Trol-Ten" for five min on the "blend" setting. The seed material produced in this manner was more homogeneous than the crushed material. Individual particles ranged in size from about 0.05 mm to 0.1 mm.

*Flaking and cooking.* The flaked seed material was procured from a commercial seed processor (CSP). This material was produced by passing whole seed through a series of roller mills. During the flaking process, the seed was crushed and flattened (rupturing most of the seeds' cell walls), thereby rendering the material more susceptible to solvent extraction. Typical thicknesses of the seed flakes ranged from 0.2 to 0.5 mm. After flaking, the seed was subjected to a short heating process (90°C, 0.5 hr). This served to inactivate certain undesirable enzymes and to enhance the extractability of the material (13). Seed treated by this method is referred to as having been "cooked."

*Pressure rupturing.* Pressure rupturing is a technique which involves placing a biological material in a high pressure gas for an appropriate length of time (typically one hr). During this time the pressurized gas penetrates the material. The pressure is then quickly released and, as the contained gas expands, some cells are ruptured.

The procedure for preparing pressure-ruptured seed was as follows. Samples of whole seed (typically 12 g) were placed in a 40-ml extraction vessel. The vessel was then placed in the HPLC oven for about 0.5 hr and allowed to equilibrate at 55°C. The pump was then turned on and the system pressurized to 36 MPa. Because the restricter valve was closed, there was no flow of

TABLE 2

**Distribution of Seed Particle Sizes for Different Methods of Seed Treatment**

| Size range (mm) | Percent of total mass | | | | | |
|---|---|---|---|---|---|---|
| | Whole seed | Crushed | Chopped | Flaked | Cooked | Exploded |
| <0.149 | - | 0.8 | 8.6 | 0.2 | 0.2 | - |
| 0.149 - 0.589 | - | 41.3 | 70.8 | 19.2 | 12.6 | - |
| 0.590 - 0.850 | - | 30.2 | 15.5 | 24.8 | 24.3 | - |
| 0.851 - 1.00 | 1.1 | 12.1 | 1.5 | 11.5 | 22.8 | 0.4 |
| 1.00 - 1.40 | 25.8 | 9.7 | 1.1 | 19.0 | 33.3 | 19.5 |
| >1.40 | 73.1 | 5.9 | 2.4 | 25.2 | 6.7 | 80.1 |

$CO_2$ through the vessel. At the end of one hr, the restricter valve was opened fully, thereby allowing the $CO_2$ to escape rapidly. The ruptured seed material was then removed from the vessel.

The pressure-ruptured seeds exhibited a wide variation in particle sizes ranging from one mm (whole seed) to 0.05 mm in diameter; the majority of the fragments were greater than 0.5 mm.

*Seed particle size.* Table 2 presents the range of seed particle sizes resulting from the various treatment procedures. The analysis was performed by passing the seed material through sieves of various mesh sizes and weighing the fraction associated with each tray.

*Extraction procedure.* At the beginning of each experiment, a weighed amount of seed material, typically four or seven g depending on the extractor size, was loaded into the vessel. Fine-spun glass wool was placed at both ends of the vessel to prevent small particles from entering the tubing. The extractor was then placed in the oven for 0.5 hr to allow for temperature equilibration, and the pump was turned on. When the system had reached the desired pressure, the sampling was started.

The latter consisted of fitting pre-weighed, 1.8-ml glass vials to the sampling tube for varying lengths of time. From the weight change of each vial, the oil collected during the sampling period could be determined. The volume of gaseous $CO_2$ used during the sampling period, as well as the $CO_2$ flowrate, were determined using a wet test meter. The corresponding mass of $CO_2$ was calculated from the known $CO_2$ molar volume at the temperature and pressure of the wet test meter.

*Solubility determination.* The average oil concentration in the $CO_2$ at the extractor outlet was determined from the mass of oil collected and the mass of $CO_2$ which had passed through the bed during the sampling interval. A typical extraction curve is shown in Figure 2. The concentration of the oil in the $CO_2$ collected during the initial linear portion of the extraction curve corresponds to its solubility, provided that the $CO_2$ becomes saturated on its passage through the vessel.

Two methods were used to ensure $CO_2$ saturation. In the first method, two separate extractions were performed at the same pressure, temperature and carbon dioxide flowrate. The only difference between the two extractions was that, in the second extraction, the height of the seed bed was increased. Provided the slopes of the linear part of the extraction curves were identical, the $CO_2$ was saturated with oil and the slopes could be used to calculate the oil solubility.

In the second method, the extraction was stopped during the "linear phase" and the bed of seed removed from the extractor in sections. Each section was analyzed for its seed-oil content. If the oil content of the seeds nearest to the extractor outlet remained unchanged, it indicated that the carbon dioxide had become saturated prior to reaching the extractor outlet. This, in turn, indicates that it is possible to calculate the oil solubility in $CO_2$ from the slope of the linear portion of the extraction curve.

## RESULTS AND DISCUSSION

*Oil solubility.* The solubility of canola oil in $CO_2$ at different temperatures and pressures was determined from the corresponding extraction curves using the procedures outlined above. The error bars shown on the figures represent the maximum and minimum slopes of the lines which could be drawn through the initial linear portion of the extraction curves.

In Figure 3 the solubility of canola oil in $CO_2$ is plotted as a function of pressure at different temperatures. The figure indicates that, as the pressure of the $CO_2$ increases, the oil solubility also increases for all temperatures studied.

In each case, the maximum solubility was found at 36 MPa, but it is possible that even higher solubilities may occur at more elevated pressures. Due to equip-

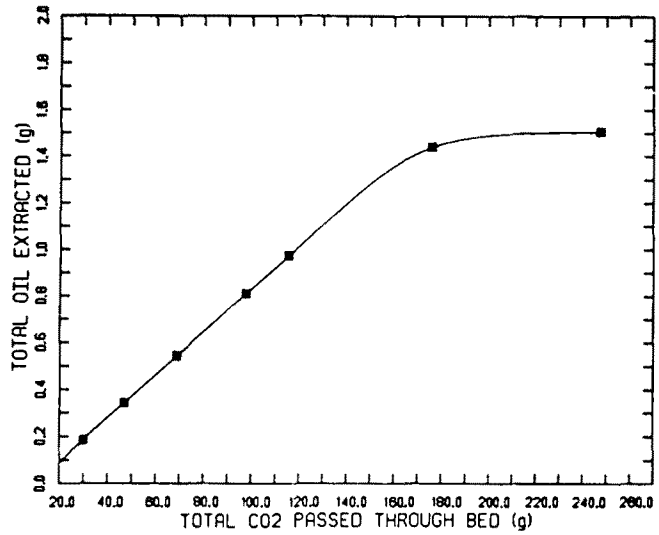


FIG. 2. Extraction curve for a 4-g sample of commercially cooked canola seed. Conditions: 36 MPa, 55°C, $CO_2$ flow rate 0.7 g/min.

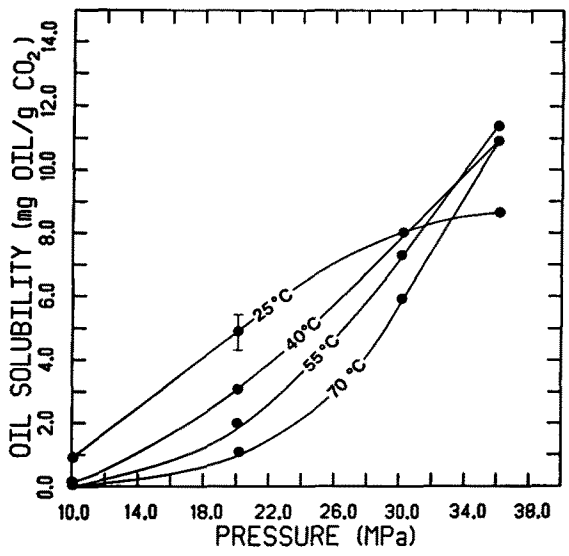


FIG. 3. Solubility of canola oil in $CO_2$ as a function of pressure at four temperatures. Conditions: vessel #2, $CO_2$ flow rate 0.7 g/min, 7 g flaked seed.

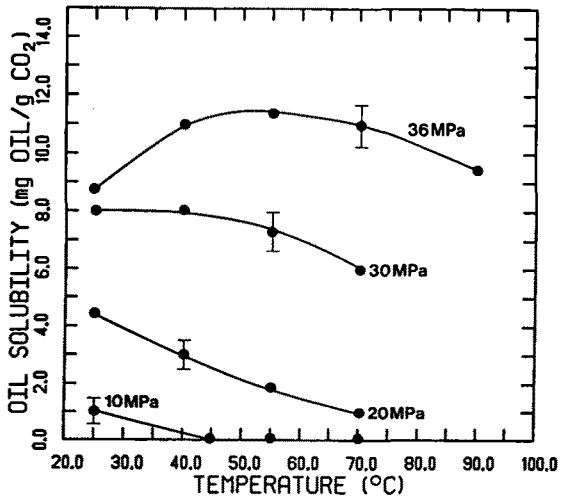


FIG. 4. Solubility of canola oil in $CO_2$ as a function of temperature at four pressures. Conditions: vessel #2, $CO_2$ flow rate 0.7 g/min, 7 g flaked seed.

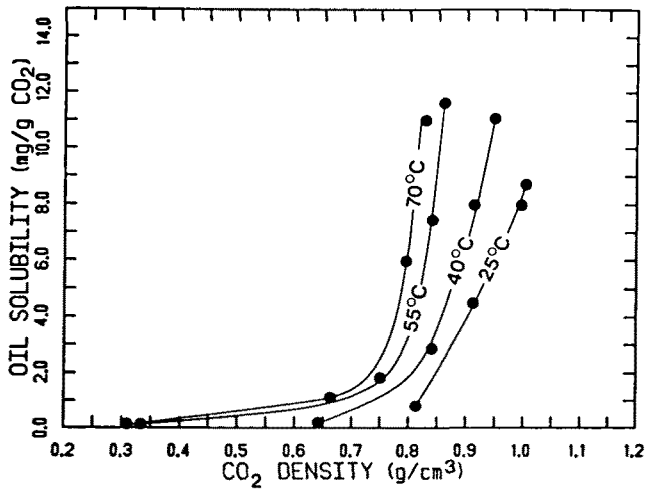


FIG. 5. Solubility of canola oil in $CO_2$ as a function of $CO_2$ density at four temperatures. Conditions: vessel #2, $CO_2$ flow rate 0.7 g/min, 7 g flaked seed.

ment limitations such pressures could not be explored in the present study. The temperature cross-over is not unusual and has been reported previously by others (14,15).

In Figure 4 the solubility of the oil is plotted as a function of temperature, at different pressures. At low pressures, the oil solubility was found to decrease with temperature, whereas at higher pressures the solubility exhibits a maximum. This temperature effect is not ususual and has been reported for naphthalene dissolved in supercritical ethylene (16) and in $CO_2$ (11).

In Figure 5 the solubility of canola oil is plotted as a function of $CO_2$ density, at different temperatures, while in Figure 6 the solubility is plotted as a function of temperature at different $CO_2$ densities. These plots show that a simple monotonic relationship exists between oil solubility and $CO_2$ density. The complex relationship between oil solubility and pressure (at different temperatures) can be explained by referring to Figure

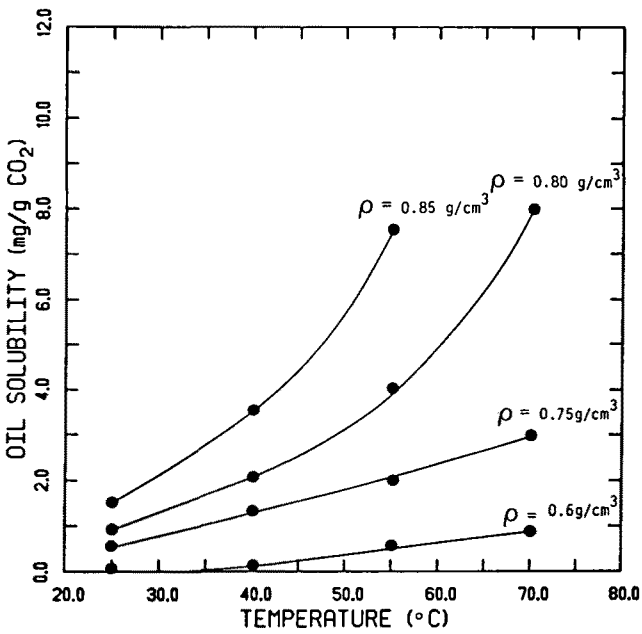


FIG. 6. Solubility of canola oil in $CO_2$ as a function of temperature at different $CO_2$ densities. Conditions: vessel #2, $CO_2$ flow rate 0.7 g/min, 7 g flaked seed.

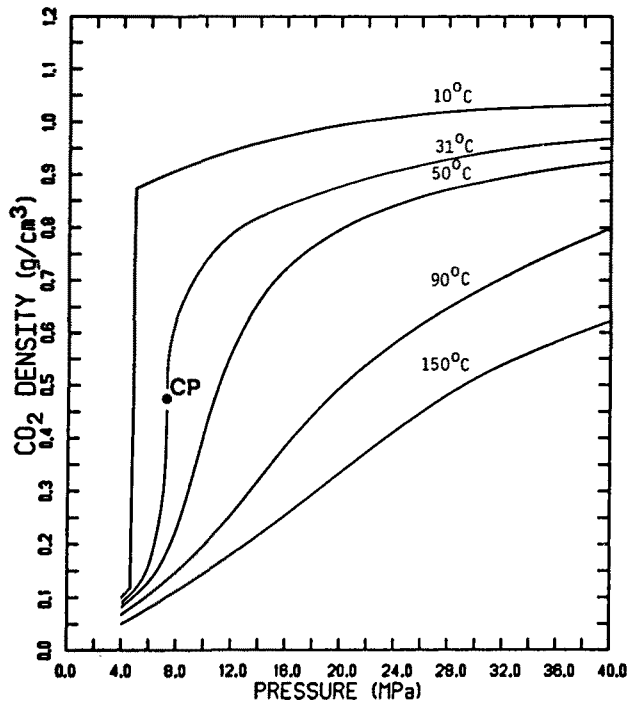


FIG. 7. Density of carbon dioxide as a function of pressure at different temperatures. The critical point (CP) of the $CO_2$ is indicated on the diagram (17).

7. As indicated in this figure, a rise in temperature, at constant pressure, leads to a decrease in $CO_2$ density. On the other hand, a rise in temperature also leads to an exponential increase in the vapor pressure of the oil (18,19). Near the critical point of the $CO_2$ the density changes rapidly with temperature. A small temperature change in this region may lead to a large change in

$CO_2$ density and a commensurate change in oil solubility. At higher pressures, however, the same temperature change has a smaller effect on the fluid density. In this case, the increase in the vapor pressure of the oil may more than offset the decreased solvent capacity of the fluid due to its decreased density. The net effect is an overall increase in solubility.

These results follow a common trend that: (i) the solvent power of a supercritical fluid increases with density at a given temperature, and (ii) the solvent power of a supercritical fluid increases with temperature at constant density (20).

## PRACTICAL IMPLICATIONS OF SOLUBILITY DATA

Figure 3 indicates that excellent separation of canola oil and $CO_2$ solvent can be achieved by a simple pressure reduction. The figure also shows that the pressure need not be reduced to atmospheric levels because the oil's solubility in $CO_2$ at 10 MPa is almost insignificant. This fact is important because it indicates that, in an extraction system where the $CO_2$ is recycled, the costs of repressurizing the $CO_2$ could be reduced. It has been suggested that, in some cases, oil separation based on pressure reduction is too expensive and that separation based on a temperature change may be more desirable (19). However, it seems unlikely that canola oil can be separated economically from $CO_2$ solely by changing the temperature. As indicated above, the most effective separations of this kind involve the use of a supercritical solvent near its critical point. For $CO_2$ this would require pressures and temperatures in the region of 7 to 10 MPa and 30 to 40°C, respectively. However, under these conditions the solvation capacity of $CO_2$ for canola oil is too low to make such an extraction worthwhile.

A temperature-based separation at higher $CO_2$ pressure would also appear to be impractical. For example, if the extractor operates at 55°C and 36 MPa and the separator operates at 25°C and 36 MPa, only a small amount of the oil would precipitate on passing through the separator (Fig. 3). The recirculated $CO_2$ would thus enter the extraction chamber with a high oil content, thereby lowering the driving force in the extraction and consequently the rate of extraction.

A separation based solely on temperature might, however, be possible provided a suitable entrainer can be found for the $CO_2$. Peter and Brunner (18) have demonstrated such a separation using $CO_2$ charged with 10 wt% ethanol. At 50°C and 17.5 MPa, the solubility of palm oil in this mixture was reported as approximately 8 wt%. At the same pressure but at 90°C, the solubility was only 2 wt%. This indicates that an effective separation of palm oil from the solvent could be achieved with a temperature change of only 40°C. Because palm oil and canola oil are similar, it would be reasonable to assume that similar separations could be performed with canola oil.

An alternate approach to using an entrainer might be to perform the separation at a lower temperature and pressure.

*Effect of seed treatment.* Initially, attempts were made to extract whole, unbroken canola seeds with carbon dioxide at 36 MPa and 55°C. Virtually no oil was extracted over a five-hr period using a $CO_2$ flow-

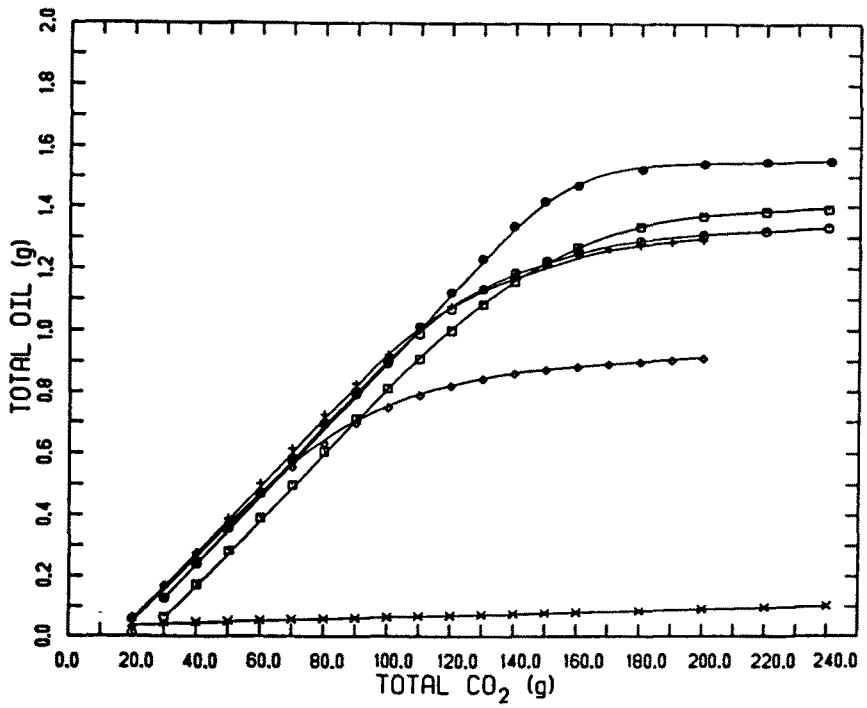


**FIG. 8. Extraction curves for 4-g samples of canola seed subjected to different pretreatments. (O, flaked; +, finely chopped; ◆, crushed; X, exploded; □, cooked; •, glass beads). For comparison purposes, the extraction curve for Canola oil on glass beads is also shown. Conditions: vessel #1, pressure 36 MPa, temperature 55°C, $CO_2$ flow rate 0.8 g/min.**

rate of 0.8 g/min. This result agrees with the findings of Othmer and Agarwal (21), who extracted whole and laterally sectioned soybeans with hexane for 166 hr. It was determined that less than 0.08% of the oil originally in the whole beans and less than 0.19% of oil in the half beans was extracted. This indicates that hexane is unable to penetrate and remove oil from unbroken cells. Commercial canola seed extraction, for similar reasons, is therefore preceded by cooking and flaking (22,23).

In Figure 8 typical extraction curves are shown for seeds having undergone various pretreatments. All extractions were carried out at 36 MPa, 55°C and a $CO_2$ flowrate of 0.7 g/min. In each case, a four-g sample was used. For comparative purposes the extraction curve of pure canola oil supported on a bed of 0.3-mm glass beads is also shown. The amount of oil on the beads (1.6 g) was equivalent to the oil content of four g of oilseed. It is evident from this figure that seed pretreatment greatly affects the total quantity of oil removed from the seeds. This finding is also in accordance with the observations of Snyder et al. (24).

In Figure 9 the extraction data are plotted in a different form. The curves were generated by fitting a second order polynominal function to the data points in Figure 7 and differentiating the function at selected positions. The values on the vertical axis correspond to the oil concentrations in the $CO_2$ at the outlet of the extractor. The horizontal axis represents the percentage of the theoretical maximum amount of oil in the seed samples (40 wt%) as determined by exhaustive extraction with hot hexane.

As indicated in Figure 8, the least effective pretreatment procedure was the "exploding" technique. After extracting this material for five hr, less than 10% of its oil was removed. The figure also illustrates that the $CO_2$ did not reach saturation. This seed pretreatment technique differs from all of the others in this respect and indicates that the cellular disruption was only slight. Hence, either the $CO_2$, which originally was used to explode the seeds, did not penetrate to a significant depth and disrupted only the cells on the surface of the seeds or, alternatively, the cell walls were sufficiently robust to withstand the large pressure differential generated during decompression.

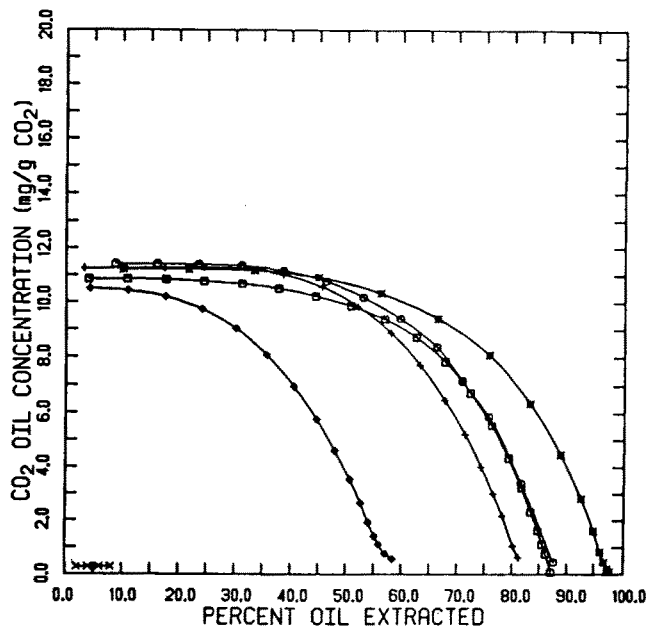


**FIG. 9. Transformed extraction curves for several canola seed pre-extraction treatments. (O, flaked; +, finely chopped; ◆, crushed; ×, exploded; □, cooked; •, glass beads). The Y axis of the graph represents the concentration of oil in the $CO_2$ at the extractor outlet.**

The crushing procedure, although considerably more effective than the pressure-rupturing treatment, evidently left much of the seed intact as well. As indicated in Figure 9, concentrations at the extractor outlet began to decline rapidly after ca. 10% of the oil had been removed. By the time 60% of the seed oil had been removed, the oil concentration in the outlet $CO_2$ had decreased to less than 5% of its saturation value.

For the finely chopped seed, the outlet oil concentration began to decrease after about 35% of the oil had been extracted. The oil concentration in the $CO_2$ fell to 5% of its saturation value only after 80% of the seed oil had been removed. The flaked and cooked seeds had extraction characteristics similar to those of the finely chopped seed in the initial stage of the extraction. However, the amount of oil extracted from these samples exceeded 85% of total before the oil concentration of the extractor outlet fell to 5% of saturation.

As can be seen from Figure 8, the canola oil was most effectively removed from the glass beads. Over 95% of the oil in the bead matrix was removed before the outlet oil concentration in the $CO_2$ fell to 5% of its saturation value. This result is not unexpected, because the beads are nonporous and all of the oil lies on the bead surface. Furthermore, the bead beds are composed of many open channels through which $CO_2$ can pass freely. By contrast, the oil in beds of crushed or flaked seed may be trapped in regions between particles impervious to the $CO_2$ or within intact cells. Much of the oil contained within these regions is not exposed to the moving stream of $CO_2$ and is only transferred out of these channels by molecular diffusion, which is a very slow process.

## ACKNOWLEDGMENT

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