Partial Defatting of Roasted Peanut Meals and Kernels by Supercritical CO_2 Using Semicontinuous and Intermittently Depressurized Processes

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Roasted peanut kernels were ground into meal and subjected to supercritical carbon dioxide (SC-CO_2) defatting using a semicontinuous process (SCP). The amount of oil recovered from meal increased with increased pressures and temperatures from 3000 to 8000 psi and from 40 to 80 °C. When SCP and an intermittently depressurized process (IDP) were applied at 8000 psi and 80 °C, the amount of oil recovered from roasted, deskinned kernels was less than that recovered from roasted peanut meals. When IDP was done at 80 °C and various CO_2 pressures, the amount of oil recovered decreased with a pressure reduction from 8000 to 4000 psi and increased with a further reduction to 2000 psi. The breaking intensity of kernels partially defatted by IDP was significantly less than that of full-fat kernels. The fatty acid composition of SCP-extracted oils from peanut meal differed insignificantly, while that of IDP-extracted oils from peanut kernels differed slightly depending on extraction conditions. Infrared and Raman spectra of the oils extracted from kernels using SCP or IDP did not vary significantly.

Keywords: *Peanut; defatting; SFE; fatty acid composition*

INTRODUCTION

Roasted peanuts have a pleasant flavor and unique texture, rendering them popular with consumers. However, since peanuts have a high oil content, some consumers are concerned about the amount of calories peanuts provide. Procedures for partially defatting roasted peanut kernels that retain indigenous textural properties have been intensively investigated (Pominski et al., 1969, 1983; Santerre et al., 1994). Nevertheless, a successful and practical defatting process has yet to be developed for peanut processors.

Supercritical fluid extraction (SFE) is recognized as a potential technique for differential extraction and has attracted the interest of food technologists. Application of SFE for food processing has been extensively investigated as a process to take the place of conventional solvent extraction processes (Dziezak, 1986). Supercritical fluid exhibits not only the liquid nature of dissolution but also the gas nature of diffusion. Supercritical CO_2 is of particular interest to food technologists, mainly because CO_2 is nontoxic, inexpensive, and unique in chemical and physical properties.

Vegetable oils can be extracted from crushed seeds with liquid or supercritical CO_2. The yield of oil depends on the pressure and temperature employed during extraction as well as the size and shape of the seeds or particle size of meals (Stahl et al., 1980). The large size of peanut meal granule makes oil extraction difficult (Snyder et al., 1984). The path of oil migration through the particle matrix hinders removal of oil and, thus, the rate and efficiency of the defatting process. In this study, a semicontinuous process (SCP) was applied for defatting of roasted peanut meals. For defatting of roasted, deskinned whole kernels, both SCP and an intermittently depressurized process (IDP) were administered for the purpose of comparing oil extraction efficiencies. The fatty acid composition of oils extracted by SCP and IDP was determined. Oils extracted from roasted, deskinned whole kernels were further subjected to infrared and Raman spectrum analyses. The partially defatted kernels were also subjected to texture analysis with a rheometer.

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MATERIALS AND METHODS

Peanuts and Equipment. Peanut kernels (Tainan 9, a Spanish cultivar) were roasted at 160 °C for 35 min, cooled, and deskinned. For peanut meal preparation, the kernels were ground with a cyclone coffee mill and sieved (1 mm). Roasted and deskinned whole kernels and meals were used as the source material for defatting. Oil prepared using a hydraulic press (150−170 kg/cm^2) served as a control for comparison of the fatty acid composition of oils produced by the experimental extraction methods. A syringe pump and an extractor (Models 100DX and SFX-10, extractor, Isco Inc., Lincoln, NE) were used for SFE. Purified CO_2 from a cylinder connected to the syringe pump was used as an extraction medium.

SCP Used for Defatting Roasted Peanut Meals and Roasted, Deskinned Whole Peanut Kernels. To determine the holding time, a series of preliminary extraction experiments were conducted. Peanut meal (1 g) was deposited in the extraction chamber (ca. 13 mL), pressurized with CO_2 at 3000 psi and 40 °C, and held for 3, 5, 10, and 15 min. The extraction medium (14 mL) was then slowly released (4.0 mL/min) and collected in a test tube. The collected oil was weighed to determine the minimal holding time required for extraction of peanut oil in the supercritical CO_2. These experiments revealed that 3 min was sufficient to dissolve peanut oil up to the saturation level in the SC-CO_2 fluid. A 3-min holding time was therefore used in subsequent experiments.

The efficiency of oil recovery from peanut meals using a semicontinuous process, various CO_2 pressures (3000, 4000, 6000, and 8000 psi at 80 °C), and various temperatures (40, 60, and 80 °C at 8000 psi) was investigated. For each extraction, 1 g of peanut meal was deposited in the extraction chamber, pressurized with CO_2 to the desired pressure, and held for 3 min, followed by the release of 14 mL of the extraction medium under the specified pressure. The oil from six replicate extractions was pooled and weighed. The pooled peanut oil was stored at −25 °C in a sealed brown vial for further analyses. The efficiency of oil recovery was determined by dividing the weight of the extracted oil by the weight of CO_2 consumed (IUPAC, 1976).

For roasted, deskinned whole kernels extracted by SCP, two roasted kernels were accurately weighed (ca. 1 g), placed in the extraction chamber, and pressurized with CO_2 to 8000 psi at 80 °C for 3 min, followed by the release of the extraction medium and collection of the oils. The extraction medium was released under constant pressure, i.e., 14 mL of the extraction medium was slowly released (ca. 4 mL/min) and collected. The extraction operation was repeated six times, and the extracted oils were weighed, pooled, and stored at −25 °C for further analyses. The amount of oil recovered was determined as described above.

IDP Used for Defatting Roasted, Deskinned Whole Peanut Kernels. Oil was extracted from roasted, deskinned whole peanut kernels at various CO_2 pressures from 8000 to 2000 psi at 80 °C using the procedure described for SCP. Two roasted, deskinned kernels were accurately weighed (ca. 1 g), placed in the extraction chamber, pressurized with CO_2 to the desired pressure, and held for 3 min before the extraction medium (ca. 13 mL) was released and collected. The extraction process was repeated by replenishing the extraction chamber with CO_2 and pressurizing to the desired pressure. The extracted oils were weighed, pooled, and stored at −25 °C for further analyses. Six replicates were done for all combinations of extraction conditions except the combination of 2000 psi and 80 °C, which was repeated 12 times. The amount of oil recovered was determined as above. The defatting percentage was determined by dividing the weight of the extracted oil by the total oil weight of meals or kernels and then multiplying by 100. The average oil content of kernels used in this study was 49%, which was determined by Soxhlet extraction method from ground peanut meals using ethyl ether as the solvent.

Peanut Oil Characterization. The fatty acid composition of peanut oils extracted from peanut meals and whole kernels was analyzed (Chiou et al., 1993). Oils extracted from roasted, deskinned whole peanut kernels by SCP (8000 psi and 80 °C) and by IDP were analyzed for infrared (IR) and Raman spectra. The IR spectra were obtained by holding oil samples between two pieces of KBr crystals (35 × 35 × 5 mm, Jasco Co., Tokyo, Japan) and then scanning between 4000 and 400 cm^{-1} with an FT-IR spectrophotometer (FT-7, Bio-Rad, Richmond, VA). Raman measurements were carried out using a Raman instrument (RFS 100, Bruker Analytische Messtechnik GmbH, Rheinstetten, Germany). Continuous wave near-IR excitation at 1.064 μm was provided by a diode laser pump (Nd:YAG laser, DPY 301, Adlas, Germany) as a light source. Each oil sample was placed in a capillary tube and focused by laser light. The scattered radiation from the sample was collected at 180° with an ellipsoidal mirror and passed through the Michelson interferometer. The modulated light was then detected by a liquid nitrogen-cooled Ga−As semiconductive detector. The data acquisition conditions for each sample were as follows: wavelength, 1.064 μm; laser power, 250 mW; spectral resolution, 4.0 cm^{-1}.

Texture Characteristics of Partially Defatted Kernels Subjected to IDP. Partially defatted peanut kernels were separated into two splits (cotyledons) and subjected to texture analysis using a rheometer (Fudoh Rheometer NRW-2010J-CW, Fudoh Ind. Co., Tokyo, Japan) adapted with an awl-shaped stainless steel probe (Fudoh probe 5) (Bai et al., 1990). Breaking intensity and hardness of the splits were determined separately. Full-fat roasted peanut kernels were used as a control.

Statistical Analysis. A minimum of two replicates was done for each experiment. Means of values are reported. Analysis of variance (ANOVA) was applied to analyze the variance of the determinations as affected by various extraction conditions. The analysis was followed by a Tukey−Kramer HSD multiple comparison test using software (JMP) for statistical visualization on the Apple Macintosh computer from SAS Institute Inc.

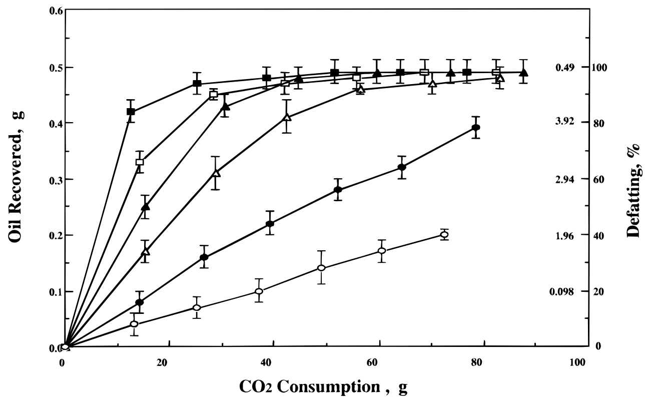

Figure 1. Peanut oils recovered and defatting percentages obtained from roasted peanut meals extracted by SC-CO_2 using a semicontinuous process: (■) 8000 psi/80 °C; (□) 8000 psi/60 °C; (▲) 8000 psi/40 °C; (△) 6000 psi/40 °C; (●) 4000 psi/40 °C; (○) 3000 psi/40 °C.

RESULTS AND DISCUSSION

The amount of oil recovered and defatting percentages obtained from roasted peanut meals extracted by SC-CO_2 at various SCP conditions are shown in Figure 1. Under the same CO_2 pressure, an increase in temperature from 40 to 80 °C resulted in increased amounts of oil extracted and defatting percentages. At a constant temperature, an increase in CO_2 pressure from 3000 to 8000 psi resulted in an increase in the amount of oil extracted. The most effective combination was 8000 psi and 80 °C. Under these conditions, approximately 80% of the oil was removed by the first extraction. The defatting percentage increased to 95% by subjecting the meal to additional extractions. In comparison to the solubility of triglycerides in SC-CO_2 (Bambeger et al., 1988; Yu et al., 1994), the oil obtained in the initial two extractions was much higher than that of the saturated dissolution point. This suggests that oil is being removed rapidly from peanut meals as a result of rapid expansion of SC-CO_2 during depressurizing. At 80 °C, peanut oil is a mobile fluid and might mix well with SC-CO_2 in the extraction chamber. Mass flow and consequent removal of peanut oil during depressurizing of SC-CO_2 resulted in enhanced efficiency of oil removal from the meal. Under other extraction conditions, the same trend was also observed.

The amount of oil recovered and defatting percentages obtained from roasted, deskinned kernels extracted by SCP at 8000 psi and 80 °C and by IDP at 80 °C are shown in Figures 2 and 3, respectively. When whole kernels were extracted by SCP at 8000 psi and 80 °C, comparatively lower amounts of oils were recovered. It has been reported that the larger the particle size used for oil extraction, the more difficult it is to extract oil by SC-CO_2 (Stahl et al., 1980; Snyder et al., 1984). Since oil was present in the interior peanut kernel matrix and the chamber pressure was kept constant during release of SC-CO_2, mass flow to remove oil from kernels is comparatively less effective than with peanut meal. In kernels extracted using IDP at the same conditions (8000 psi and 80 °C), the amount of oil recovered per unit weight of CO_2 consumption was much higher than that extracted by SCP (Figure 2). The defatting percentage reached 75% by the sixth batch (Figure 3). Compared to SCP, the chamber pressure in the IDP system rapidly declined during release of SC-CO_2 (depressurization). The pressure change resulted in a rapid expansion and flow of SC-CO_2 from the

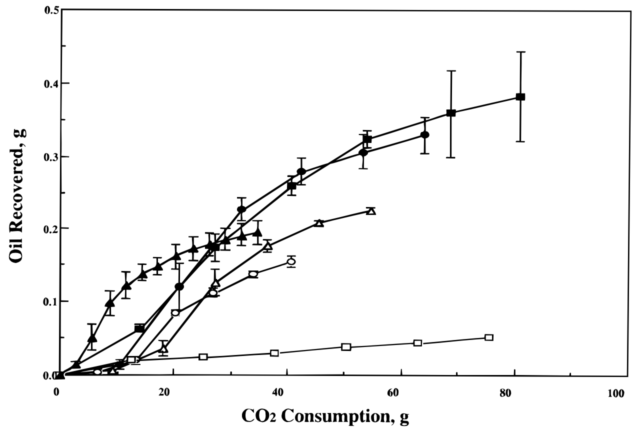

Figure 2. Peanut oils extracted from roasted, deskinned whole peanut kernels by SC-CO_2 using semicontinuous and intermittently depressurized processes: (□) SCP at 8000 psi/80 °C; (■) IDP at 8000 psi/80 °C; (●) IDP at 6000 psi/80 °C; (△) IDP at 4000 psi/80 °C; (○) IDP at 3000 psi/80 °C; (▲) IDP at 2000 psi/80 °C.

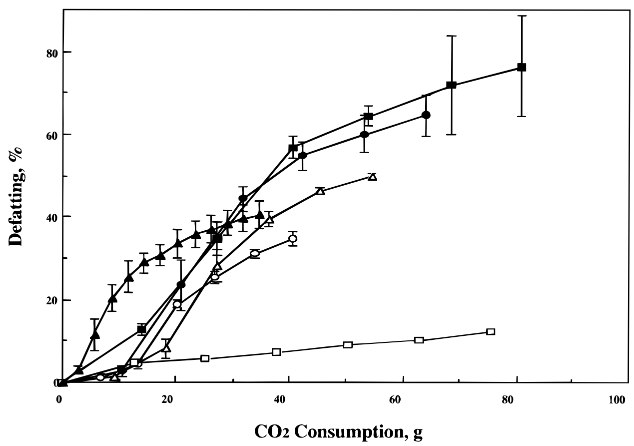

Figure 3. Defatting percentages of roasted, deskinned whole peanut kernels by SC-CO_2 using semicontinuous and intermittently depressurized processes: (□) SCP at 8000 psi/80 °C; (■) IDP at 8000 psi/80 °C; (●) IDP at 6000 psi/80 °C; (△) IDP at 4000 psi/80 °C; (○) IDP at 3000 psi/80 °C; (▲) IDP at 2000 psi/80 °C.

interior to the surface of peanut kernels. The surface oil would likely be extracted in the same manner as that in SCP. This is suggested by the fact that the amount of oil recovered in the first batch was comparatively lower than the amounts obtained in following batches (Figure 2). From a practical viewpoint, the use of 8000 psi by edible oil processors is not realistic due to limitations in the construction of scale-up equipment and to safety consideration.

When IDP was done at 80 °C and various CO_2 pressures to defat roasted, deskinned kernels (Figures 2 and 3), the amount of oil recovered and defatting percentages decreased when the CO_2 pressure was reduced from 8000 to 4000 psi and then increased with a further reduction from 4000 to 2000 psi. At each stage of the extraction process, the amount of oil recovered, expressed as weight of CO_2 consumed or as the percentage of oil in the kernels, increased with repeated extractions. At 2000 psi and 80 °C, recovered oil based on unit weight of CO_2 consumed was the highest among the extraction conditions evaluated; however, the longest extraction time was required under these conditions to achieve the desired level of oil removed from peanut kernels. From a practical viewpoint, IDP operated at

Table 1. Defatting Percentage and Texture Characteristics of Full-Fat (Control) Peanut Kernels and Kernels Partially Defatted by SCP and IDP (n = 4)

extraction condition	defatting percentage and texture characteristics[a]		
	defatting (%)	breaking intensity (10^5 kg/cm^2)	hardness (10^4 N/ cm^2)
control	0[g]	12.07[a]	1.66
8000 psi SCP	12.2[f]	—[b]	—[b]
8000 psi IDP	76.4[a]	4.46[b]	2.72
6000 psi IDP	64.6[b]	5.45[b]	2.11
4000 psi IDP	50.0[c]	4.42[b]	3.05
3000 psi IDP	34.6[e]	3.40[b]	2.88
2000 psi IDP	40.4[d]	4.10[b]	2.75
significance level of ANOVA[c]	**	*	—

[a] Values in the same column that are not followed by the same superscript letter are significantly different (p < 0.05) [b] Peanut kernels were broken after unloading from the extraction chamber. [c] "—" indicates insignificant (p > 0.05); "*" indicates significant (p < 0.05); "**" indicates very significant (p < 0.01).

a practically accepted low pressure is most promising for defatting large particulates (kernels) using a scale-up facility.

Breaking intensity and hardness of partially defatted roasted kernels were determined with a rheometer (Table 1). Breaking intensity values for partially defatted kernels were significantly lower than those of full-fat kernels. However, differences in extracting conditions did not yield differences in breaking strength. Changes in kernel hardness resulting from defatting were not significant. Defatting of kernels with SCP at 8000 psi and 80 °C resulted in broken granules. Breakage may occur at the time of depressurization after semicontinuous extraction of peanut oil and unloading of the kernels from the extraction chamber.

Peanut oils extracted from roasted peanut kernels using a hydraulic press and from meals using SCP were subjected to fatty acid composition analysis (Table 2). No significant differences were observed among samples. However, the fatty acid profiles of oils extracted from roasted, deskinned whole kernels (Table 3) by a hydraulic press (control) and by SCP or IDP indicated that the palmitic acid (18:0) and arachidic acid (20:0) contents differed significantly, depending on the extraction procedure. Differences in kernels and different defatting percentages (Table 1), in addition to different extraction conditions, may contribute to significant differences among samples. Since oil extraction was conducted in a semicontinuous process under controlled conditions up to a certain degree of defatting (Figure 3), peanut oil extracted from different areas of the kernels thus resulted in differences in fatty acid composition. Visible observations on the cross-section color of the partially defatted kernels after subjection to breaking intensity test revealed that the color of the outer areas of the kernels was lighter than that of the interior areas. Bamberger et al. (1988) reported that fatty acid composition of extracted oil varies slightly depending upon supercritical CO_2 fluid extraction conditions. However, when the oils were further subjected to IR spectrum analysis (Figure 4), differences among oils extracted using different procedures were minor. In Raman spectrum analysis (Figure 5), an almost identical spectrum pattern was also observed for all oil samples. On this aspect, more intensive investigations are needed in future studies.

Table 2. Fatty Acid Composition of Peanut Oils Extracted from Roasted Peanut Kernels Using a Hydraulic Press (Control) and from Ground Roasted Peanut Meals by SC-CO_2 Using a Semicontinuous Process (SCP) (n = 2 except Control with n = 3)

extraction condition	fatty acid (%) 16:0	18:0	18:1	18:2	20:0	20:1	22:0	24:0
control	12.90	3.54	38.57	38.57	1.57	0.83	2.86	0.99
8000 psi/80 °C	12.86	3.50	38.35	38.77	1.50	0.80	3.02	1.04
8000 psi /60 °C	12.82	3.52	38.22	38.55	1.55	0.82	3.03	1.10
8000 psi/40 °C	12.82	3.56	38.23	38.62	1.53	0.81	3.02	1.11
6000 psi/40 °C	13.03	3.56	38.28	38.66	1.49	0.78	2.90	1.00
4000 psi/40 °C	13.20	3.53	38.24	38.73	1.49	0.79	2.76	1.02
3000 psi/40 °C	13.16	3.49	38.20	38.82	1.42	0.78	2.67	0.92
significance level of ANOVA[a]	–	–	–	–	–	–	–	–

[a] "–" indicates insignificant ($p > 0.05$); "*" indicates significant ($p < 0.05$); "**" indicates very significant ($p < 0.01$).

Table 3. Fatty Acid Composition of Peanut Oils Extracted from Roasted, Deskinned Whole Peanut Kernels Using a Hydraulic Press (Control) and by SC-CO_2 at 80 °C Using Semicontinuous (SCP) and Intermittently Depressurized (IDP) Processes (n = 3)

extraction condition	fatty acid[a] (%) 16:0	18:0	18:1	18:2	20:0	20:1	22:0	24:0
control	12.90	3.54[ab]	38.57	38.57	1.57[a]	0.83	2.86	0.99
8000 psi SCP	13.91	3.45[b]	37.70	37.85	1.49[ab]	0.87	3.27	1.00
8000 psi IDP	13.18	3.52[ab]	38.51	38.73	1.48[b]	0.79	2.77	0.90
6000 psi IDP	13.29	3.58[ab]	38.08	38.91	1.47[b]	0.81	2.81	0.93
4000 psi IDP	13.23	3.49[b]	38.22	38.74	1.52[ab]	0.77	2.95	0.95
3000 psi IDP	12.91	3.32[b]	39.68	37.81	1.46[b]	0.81	2.94	0.95
2000 psi IDP	12.82	3.83[a]	38.51	38.56	1.56[ab]	0.75	2.91	0.94
significance level of ANOVA[b]	–	*	–	–	**	–	–	–

[a] Values in the same column that are not followed by the same superscript letter are significantly different ($p < 0.05$). [b] "–" indicates insignificant ($p > 0.05$); "*" indicates significant ($p < 0.05$); "**" indicates very significant ($p < 0.01$).

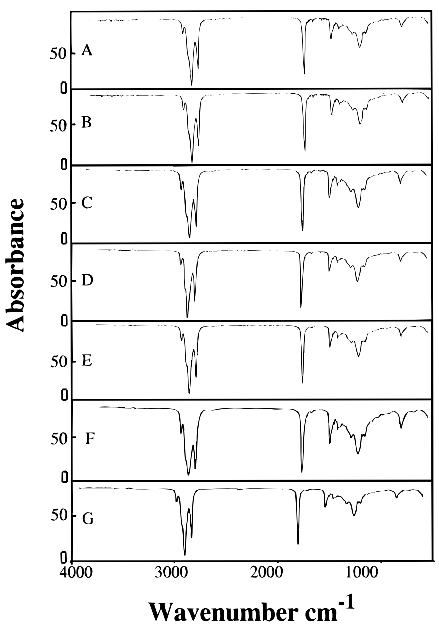

Figure 4. Infared spectra of peanut oils extracted from roasted, deskinned whole kernels by a hydraulic press (A) and by SC-CO_2 using SCP at 8000 psi/80 °C (B), IDP at 8000 psi/80 °C (C), IDP at 6000 psi/80 °C (D), IDP at 4000 psi/80 °C (E), IDP at 3000 psi/80 °C (F), and IDP at 2000 psi/80 °C (G).

In conclusion, a semicontinuous process using SC-CO_2 is suitable for defatting roasted peanut meal under high temperature and high pressure. In a recycling system, the released CO_2 after oil separation could be repeatedly pressurized and applied for defatting to save CO_2 cost. For defatting roasted, deskinned whole kernels, an intermittently depressurized process operated at a practical low CO_2 pressure is suggested. Using appropriate conditions, the appearance and texture of partially defatted kernels after treatment with IDP can be retained and perhaps improved.

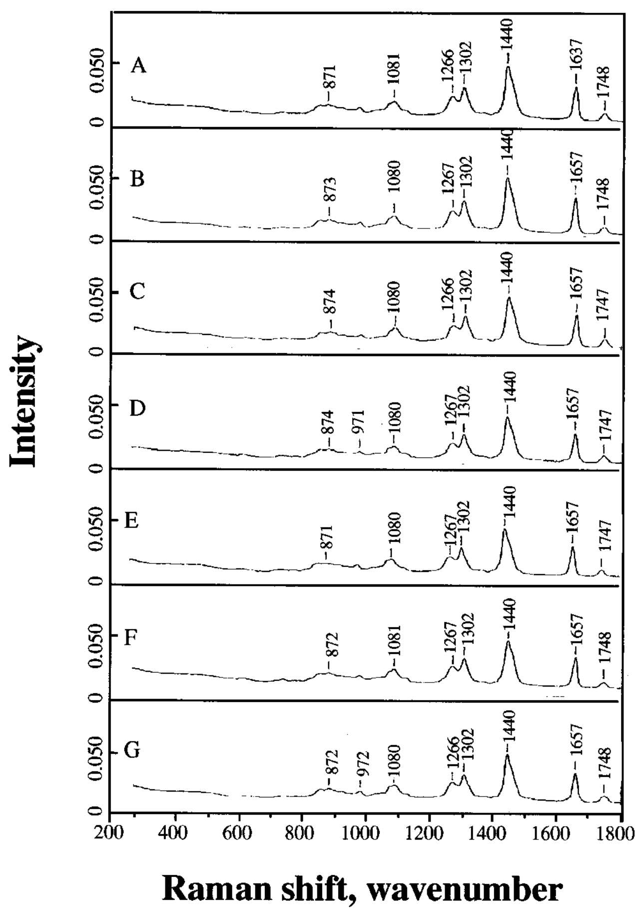

Figure 5. Raman spectra of peanut oils extracted from roasted, deskinned whole kernels by a hydraulic press (A) and by SC-CO_2 using SCP at 8000 psi/80 °C (B), IDP at 8000 psi/80 °C (C), IDP at 6000 psi/80 °C (D), IDP at 4000 psi/80 °C (E), IDP at 3000 psi/80 °C (F), and IDP at 2000 psi/80 °C (G).

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