Separation Science and Technology

ISSN: 0149-6395 (Print) 1520-5754 (Online) Journal homepage: http://www.tandfonline.com/loi/lsst20

# Pilot Scale Study of Vegetable Oil Extraction by Surfactant-Assisted Aqueous Extraction Process

Linh D. Do & David A. Sabatini

**To cite this article:** Linh D. Do & David A. Sabatini (2011) Pilot Scale Study of Vegetable Oil Extraction by Surfactant-Assisted Aqueous Extraction Process, Separation Science and Technology, 46:6, 978-985, DOI: 10.1080/01496395.2010.541401

**To link to this article:** http://dx.doi.org/10.1080/01496395.2010.541401

Published online: 15 Apr 2011.

*Separation Science and Technology*, 46: 978–985, 2011

ISSN: 0149-6395 print/1520-5754 online
DOI: 10.1080/01496395.2010.541401

Taylor & Francis
Taylor & Francis Group

# Pilot Scale Study of Vegetable Oil Extraction by Surfactant-Assisted Aqueous Extraction Process

**Linh D. Do[1,3] and David A. Sabatini[1,2,3]**

[1]*Chemical, Biological Engineering, and Materials Science Department, University of Oklahoma, Norman, OK, USA*
[2]*Civil Engineering and Environmental Science Department, University of Oklahoma, Norman, OK, USA*
[3]*Institute for Applied Surfactant Research, University of Oklahoma, Norman, OK, USA*

**A number of aqueous extraction processes (AEP) have been studied as substitutes for hexane in oilseed extraction. In our previous batch-scale work, we have shown that the aqueous surfactant-based method could effectively extract up to 95% peanut and canola oils at 25°C. The goal of this work is to perform a semi-continuous pilot-scale study of the aqueous surfactant-based method for peanut and canola oil extraction. Two extraction strategies were evaluated including (1) a single extraction stage by aqueous surfactant solution and (2) two extraction stages, consisting of one aqueous surfactant wash and one de-ionized water wash. At optimum conditions, 90.6% and 88.1% oil extraction efficiencies of peanut and canola oil, respectively, were achieved in a single-stage extraction, while 94.5% and 92.6% were achieved in the two-stage extraction. At the highest solid/liquid centrifuge speed, the moisture level in the extracted meal was 48%. At the optimum liquid/liquid centrifuge condition, more than 90% of the oil was recovered as free oil from the extracted-oil and surfactant-wash mixture and 39–44% of the oil was recovered from the extracted oil and DI wash mixture. Total free oil recovered after the two-stage extraction was 87.1% and 85.6% for peanut and canola, respectively.**

**Keywords** interfacial tension; pilot scale study; surfactant; vegetable oil extraction

## INTRODUCTION

Vegetable oils are typically produced from oilseeds by either hexane extraction or a combination of mechanical processing and hexane extraction. However, there are growing health concerns and increased environmental regulations regarding the use of hexane in vegetable oil extraction. Exposure to hexane at 15 ppm/day for three months has been shown to cause peripheral nerve damage, and hexane is also a potential hazardous explosive material (1). In 2001, the U.S. Environmental Protection Agency (EPA) established regulations on hexane emission due to growing environmental concerns. In addition, oils produced by hexane extraction are high in free fatty acid, wax, and unsaponifiable matter, and can also exhibit a dark greenish-brown color (2).

The use of aqueous extraction processes (AEP) for vegetable oil has been studied widely (3–6). AEP for oilseed extraction eliminate the potential for explosion and emissions of the volatile organic solvent hexane. Simultaneous recovery of oil and protein by AEP is possible with lower equipment costs and energy consumption than by hexane extraction (7,8). Because of the immiscibility of water and vegetable oil, AEP have consistently been report to produce vegetable oil superior in quality (lower phosphatide levels and peroxide values) to that produced by hexane-based processes (9,10). In general, when employing AEP, the extracted oil and protein in the liquid phase distribute among three portions which are the free oil, cream (oil in water emulsions), and skim (protein and sugar-rich aqueous phase) (11). Limiting the utility of AEP is the fact that vegetable oil recovery is typically low (33–68%) (12). The vegetable oil is trapped inside the porous matrix of the meal due to high capillary forces. Low-oil extraction efficiency can be attributed to the high-interfacial tension between the water phase and the vegetable oil (8–10 mN/m for canola and peanut oil) making the oil unable to diffuse through the porous matrix of the meal (13,14). By definition, interfacial tension is the surface tension caused by intermolecular interactions at the surface separating two immiscible fluids (15)—in this case vegetable oil and the extracting aqueous solution.

Several approaches have been tested in an effort to improve the oil extraction efficiency of AEP including mechanical treatment (flaking and extruding to obtain smaller grain size) (12), enzyme-assisted treatment (EAEP) (16,17) and surfactant-enhanced extraction (14,18). Mechanical treatment by grinding has improved the oil recovery

Received 22 June 2010; accepted 16 November 2010.
Address correspondence to David A. Sabatini, School of Civil Engineering and Environmental Science, Institute for Applied Surfactant Research, University of Oklahoma, 202 West Boyd, Room 334, Norman, OK, USA, 73019-1024. E-mail: sabatini@ou.edu

from 33% to 66% when flour particles were reduced from 0.4 mm to 0.1 mm (5). Similar oil recovery (68%) was achieved when employing flaking and extruding treatment to soybean flours (12). However, mechanical treatment alone still results in insufficient oil-extraction efficiency. Consistently high oil extraction efficiency (>90%) has been reported in the literature when using enzyme-assisted (8) or surfactant-enhanced AEP (14,18). In the study of AEP extraction mechanisms of soybean oil, Campbell et al. reported that both Protease enzyme and sodium dodecyl sulfate (SDS) surfactant-enhanced AEP were able to achieve similar and higher oil-extraction efficiency than AEP alone (13). In the microscopic study (13), it was found that when employing AEP alone, the unextracted oil was trapped in an insoluble matrix of denatured proteins. The coalesced oil size was too large to diffuse through the disrupted cellular matrix (13). Further, it was found that the mechanism of oil release using Protease enzyme is by proteolytic digestion of insoluble cellular matrix (13). Alternatively, the oil-release mechanism when aqueous using a surfactant is to disrupt the oil/water interface by lowering the interfacial tension between the surfactant solution and the oil, thereby facilitating the oil droplet breakup and making it possible for the oil to diffuse through the disrupted cell (13).

The use of surfactant-enhanced AEP extraction has been investigated by our group in batch-scale studies using alkyl propoxylate-ethoxylate-sulfate surfactants (14,18). In batch studies, we have achieved up to 94% oil extraction efficiency for peanut, canola, and palm oils when the interfacial tension between the surfactant solution and the oil phase was less than 0.05 mN/m (14,18). Alkyl propoxylate-ethoxylate-sulfate surfactants are extended-surfactants, a new class of surfactant that has intermediate polar groups (i.e., propoxylate or ethoxylate) inserted between the head and tail of the surfactant molecule (19). Due to this unique structure, extended surfactants have consistently produced ultralow interfacial tension (IFT) with a wide range of vegetable oils, which is critical in oilseed extraction (14,19). We define ultralow IFT as IFT $\ll$0.1 mN/m (14). The surfactant-enhanced AEP (SAEP) for vegetable oil is particularly attractive due to the short contact time between the surfactant medium and the oil-seeds (about 30 minutes), ambient temperature extraction, and high solid-to-liquid ratios (SLR of 1 to 5 g solids/g liquid), which are desirable in industrial application (14,18). Another advantage is that the surfactant concentrations are at the critical microemulsion concentration (cμc) which are relatively low (less than 0.5 wt%) (14). Cμc is defined as the lowest surfactant concentration capable of producing ultralow IFT. At this concentration, the vegetable oil is removed mainly due to the mobilization mechanism in which the oil is liberated as a separate phase rather than solubilized into the aqueous surfactant phase (14,20). It is important to note that, when employing EAEP, the incubation time is more than one hour and the slurry temperature is in the range of 50–60°C (8,11). It is important to note that only a limited number of scale-up studies on AEP and EAEP oil extraction in the literature (11,21). Rhee et al. studied the AEP pilot plant-scale production of peanut protein concentrate, with little emphasis on oil extraction efficiency (21). The peanut protein and oil recovery processes were carried out at 60°C and pH of 4 for one-hour incubation time. Up to 88.8% oil was recovered as free oil after four consecutive washes with a SLR of 1 to 10 for the first wash and SLR of 1 to 5 for other three consecutive washes (21). It was shown that dry grinding the peanuts gave free oil while wet grinding the peanuts gave lower oil-extraction efficiency and most emulsion phases (21). Moura et al. studied the scale-up of EAEP extraction of soybeans in a two-stage counter-current process using extruded soybean flakes (11). One pilot-scale run was carried out over a two-day period (11). In the two-stage counter-current processes with a SLR of 1 to 6, at 50°C and 1 hr incubation time, up to 99% soybean oil-extraction efficiency was achieved; however, most oil was distributed among cream and skim fractions after centrifugation, requiring an additional step to obtain free oil (11). Although the oil extraction efficiency is very promising, this study did not use a continuous process (i.e., use of funnel separation to recover free oil from cream (11)).

The objectives of the current research are

1. To study the effect of processing parameters on extraction efficiency and
2. To identify potential problems related to the scale-up system of SAEP.

We thus seek to extend our prior research work in the present study, we decided to evaluate semi-continuous laboratory-based SAEP using laboratory-scale processing equipment similar to that used in industrial processes. The SAEP was scaled up from 2 grams to 150 grams of peanut and canola flours.

## MATERIALS AND METHODS

### Materials

$C_{10}H_{21}$-18PO-2EOsulfate surfactant (19.9 active%) was kindly provided by Huntsman Chemical Co. (Houston, TX) and used as received. Blanched peanut seeds were purchased from the local market. Canola seeds were kindly provided by Producers Cooperative Oil Mill, Plains Oilseed Products Cooperative (Oklahoma City, OK), and Prairie Gold Oil Seeds (Okeene, OK). Sodium chloride (99% + purity) was purchased from Sigma Aldrich and used as received.

## Methods

### *Oilseed Pretreatment*

Blanched peanut seeds were dehulled, whereas canola seeds were not since it is not economically feasible to dehull canola seeds (22). Peanut and canola seeds were ground using a food processor. The particle size used in this study was in the range of 0.21 to 0.42 mm size by using US Sieve size No. 40 and No.70, which is in the recommended range for oilseed extraction (23).

### *Oilseed Extraction by SAEP*

Figure 1 illustrates the schematic diagram of the pilot-scale process utilized in this research and Fig. 2 shows selected products at different SAEP stages using optimized extraction conditions. First, 750 grams solution of surfactant ($C_{10}H_{21}$-18PO-2EOsulfate) and sodium chloride (NaCl) mixture at concentrations specified below were placed in a two liter stainless steel-extractor vessel. Next, 150 grams of seed flours were dispensed into the solution to produce a SLR of 1 to 5 (g to g). For peanut oil extraction, $C_{10}H_{21}$-18PO-2EOsulfate was fixed at 0.15 wt% and NaCl at 6 wt%, while for canola oil extraction, the surfactant concentration was 0.35 wt% with NaCl at 5 wt%. These are optimum conditions found from our previous study (14). Dispersion of the flours in the extraction solution was performed by a four-blade 1 inch mixer attached to the Talboys light-duty mixer overhead (model 101). Oilseed flours were directly fed into the surfactant solution in the extraction vessel. The slurry was agitated at 500 rpm to ensure gentle mixing and sufficient dispersion of the flour in the solution. Preliminary studies were conducted at three agitation speeds—500, 750, and 1000 rpm. There was no statistically-significant difference in the oil-extraction efficiency when varying the agitation speeds (data not shown); therefore, an agitation speed of 500 rpm was used throughout the study. After 30 minutes of extraction, the slurry was pumped by a chemical-metering pump (Precision

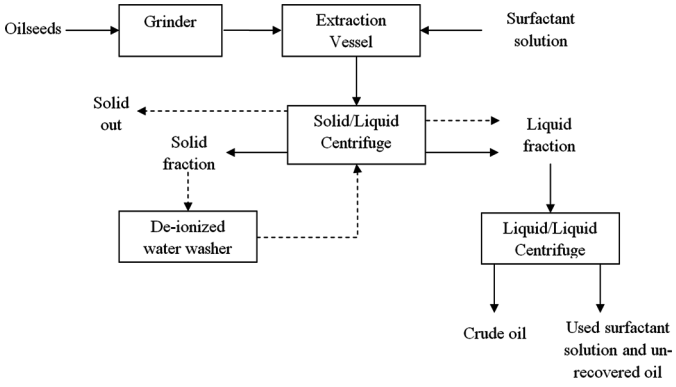


FIG. 1. Schematic diagram of laboratory–based pilot scale processing of peanut and canola oil extraction. Solid line (—): surfactant wash step; Dash line (- - -): DI washing step.

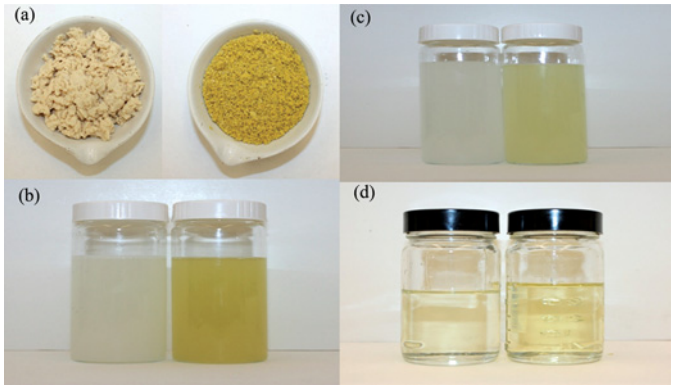


FIG. 2. Selected products at different stages of SAEP and DI washing at optimum conditions for peanut and canola from left to right, respectively. (a) peanut and canola flours (b) liquid fraction from L/L centrifuge of surfactant wash step (c) liquid fraction from L/L centrifuge of DI washing step (d) free oil crude oil recovered from L/L centrifuge.

Control Products, AMF CUNO metering pump, model 8311-11) at varying flowrates into a semi-continuous solid/liquid centrifuge (Lavin centrifuge, model L2, see reference (24) for details including picture of equipment) equipped with a 4" stainless steel bowl. The bowl was custom-perforated and used in conjunction with a 200 mesh filter cloth and a 200 mesh filter plastic to keep the filter cloth in place. They were placed inside the bowl to improve the solid/liquid (S/L) separation. The S/L centrifuge speed was varied at 1029, 2100, and 4116 × g.

The liquid portion (oil/surfactant/electrolyte/water mixture) from the S/L centrifuge was collected in a two-liter glass beaker. After collecting the liquid fraction (i.e., no more liquid was collected from the S/L outlet), the solution was then pumped by a Masterflex L/S peristaltic pump (Easyload, model 7518–00) into a continuous liquid/liquid (L/L) centrifuge (CINC model V02, see reference (25) for details/picture of the equipment) at flowrates varying from 1–5 mL/min. The L/L centrifuge was pre-filled with 150 mL of heavy-phase solution (de-ionized water) in order to obtain the best separation performance (recommended by the manufacturer). It is important to note that in the continuous large scale operation, this step will not be necessary. The L/L centrifuge speed was varied to study the efficiency in oil/surfactant solution separation. The oil from the light-phase outlet was collected in a 500 mL glass beaker and the skim fraction (most often oil in water emulsions (11)) from the heavy-phase outlet was collected in a one-liter glass beaker. The solids obtained from S/L centrifuge process were carefully scooped out. The oil residual content in the solid fraction was further analyzed. The water content in the oil or cream fraction obtained from the light-phase outlet of the L/L centrifuge was also analyzed. These test methods are discussed below. For the

de-ionized water washing step, the SAEP extracted meal (solid fraction) obtained from the S/L separation process was carefully scooped out and re-suspended in 450 mL of de-ionized water held in a two-liter stainless steel-extractor vessel. The slurry was resent to the S/L and L/L centrifuges. All data reported are average values from triplicates.

*Oil Content*

The total oil content in crude oilseeds and in residual meal obtained from S/L separation were analyzed using hexane solvent in a Soxhlet extraction apparatus following the Association of Official Analytical Chemists (AOAC) standard procedure (Method 948.22) (26). The residual meal was dried overnight in a forced oven at 104°C and re-ground for solvent extraction. In the second Soxhlet extraction step, no more oil was collected. Total oil analysis gave $46.7\% \pm 0.86\%$ peanut oil and $42.5 \pm 0.92\%$ canola oil content based on dry-weight basis, consistent with values reported in the literature (27). Oil-extraction efficiency was calculated as weight percentage of oil extracted divided by the total oil present in the seeds as determined by this method. It is important to note that in order to avoid variation in oil content and removal efficiency in different runs, the total oil content was analyzed in each run and the oil-removal efficiency was calculated based on the corresponding oil content of the same run. Oil content in the light phase obtained from the liquid/liquid centrifuge was analyzed by the temperature-modified Babcock method adapted from reference (28).

*Moisture Content*

The moisture level in the oilseeds was determined by AOAC standard procedure (Method 925.40) (26). Moisture levels in both peanut and canola seeds were in the range of 4–6 wt% which is well within the recommended range (23). The moisture content in the residual meal after S/L separation was determined by the weight difference after placing the meal in the forced oven overnight at 104°C. The water content in the extracted oil obtained from the L/L separation was determined by the Karl Fischer volumetric titration method using TitroLine KF (Schott instruments).

*Interfacial Tension Experiments*

Interfacial tension (IFT) experiments were carried out using a spinning drop tensiometer (University of Texas, model 500). To measure the IFT value between the post-wash solution and peanut and canola oils, 15 mL of the slurry obtained from S/L centrifuge were transferred into a glass tube and centrifuged at 2170 × g (IEC centrifuge, model HN). The aqueous portion obtained after the centrifuge was used for IFT measurements. IFT values were recorded at 20 minutes (14).

*Statistical Analysis*

One way ANOVA was used was for data statistical analysis and compared with p-value at 0.05.

## RESULTS AND DISCUSSION

Table 1 shows the effect of the S/L centrifuge speeds and inlet flowrate on the moisture of the meal and the total oil-extraction efficiency by SAEP. The S/L centrifuge speeds were varied at 1029, 2100, and 4116 × g (the maximum allowable speed of the equipment), and at each centrifuge speed, the inlet flowrate was evaluated at 8, 10, and 12 mL/min. It is important to note that, due to the bowl design of the S/L centrifuge, a slurry flowrate higher than 12 mL/min resulted in a significant amount of solids loss. From Table 1, it can be seen that, while the inlet flowrate had little effect on the recovery of extracted oil, the S/L centrifuge speed had a more pronounced effect. As the centrifugation speed increased, the moisture level in the meal was reduced and the total extracted oil in the liquid fraction increased. At 4116 × g (7000 rpm), the moisture level of the meals shows the lowest value at 44.8 wt% and the total oil extracted in the liquid shows the maximum value to be 90.6 wt% for peanut and 88.1 wt% for canola oil (Table 2). These values are somewhat lower than those obtained in the batch scale, which were 95 wt% and

TABLE 1
Effect of process parameters on peanut oil extraction efficiency – solid/liquid (S/L) separation

| Speed (rpm) | Speed (× g) | Inlet flowrate (mL/min) | Meal moisture content (wt%) | Oil residual[a] (wt%) | Total oil recovery[a] (wt%) |
|---|---|---|---|---|---|
| 3500 | 1029 | 8 | $78.6 \pm 0.66$ | $19.8 \pm 0.87$ | 80.1 |
| | | 10 | $80.6 \pm 1.64$ | $20.8 \pm 0.96$ | 79.2 |
| | | 12 | $78.5 \pm 0.69$ | $19.6 \pm 1.10$ | 80.4 |
| 5000 | 2100 | 8 | $64.8 \pm 0.81$ | $14.3 \pm 1.20$ | 85.6 |
| | | 10 | $63.5 \pm 2.31$ | $15.9 \pm 0.63$ | 84.1 |
| | | 12 | $65.9 \pm 1.32$ | $15.0 \pm 1.34$ | 85.6 |
| 7000 | 4116 | 8 | $44.8 \pm 2.50$ | $9.44 \pm 0.90$ | 90.6 |
| | | 10 | $46.9 \pm 3.73$ | $9.22 \pm 1.33$ | 90.1 |
| | | 12 | $48.3 \pm 1.81$ | $9.65 \pm 1.50$ | 90.2 |

[a]Amount of oil extracted via Soxhlet extraction was used as the basis. Sample calculation:

$$\begin{aligned} &\textit{Percentage of oil residual}(wt\%) \\ &= \frac{\textit{mass of oil residual}\,(g)}{\textit{mass of total oil determined by Soxhlet extraction}\,(g)} \times 100\% \\ &= \frac{6.64\,g}{70.35\,g} \times 100\% = 9.44\,wt\% \end{aligned}$$

TABLE 2
Total oil extracted at optimum conditions at 25°C[a]

| | Fraction of oil extracted from surfactant wash[b] (wt%) | Fraction of oil extracted from[b] DI wash (wt%) | Total oil extracted[b] (wt%) |
|---|---|---|---|
| Peanut | 90.6 | 3.98 | 94.5 |
| Canola | 88.1 | 4.54 | 92.7 |

[a]30 minute surfactant solution extraction, 5 minute DI wash, S/L centrifuge at 4116 × g and 8 mL/min inlet flowrate.
[b]Amount of oil extracted via Soxhlet extraction was used as the basis.

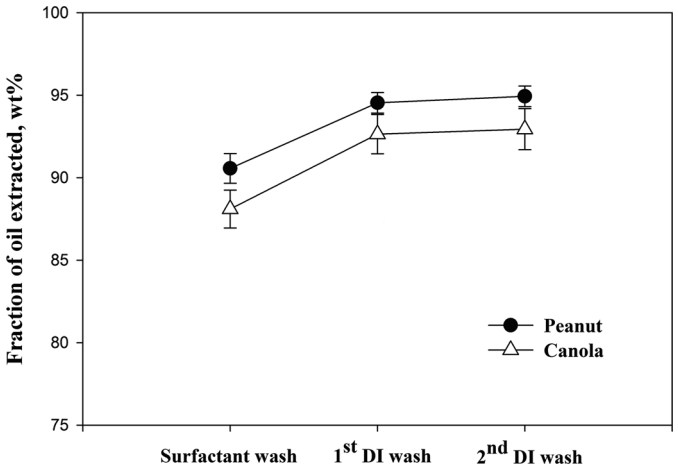


FIG. 3. Oil extraction efficiency for different consecutive extraction trials at 25°C. Extraction condition: 30 minute wash, S/L centrifuge at 4116 × g and 8 mL/min inlet flowrate. Amount of oil extracted via Soxhlet extraction was used as total oil.

93 wt% for peanut and canola oil, respectively. This difference might be due to the fact that in the batch scale we used a three-phase centrifuge, while in the pilot scale we separated this process into two different steps using the S/L separator and L/L centrifuge, suggesting that the separation of the liquid in S/L separator was not as effective in the three-phase batch centrifuge. However, it is important to note that the use of a three-phase decanter for a slurry of high solid content (up to 20 wt%) is impractical (21).

The washing step using de-ionized water was introduced to recover more oil from the SAEP extracted meal. Figure 3 shows the effect of the surfactant washing, and the first and second DI washing on the total oil extraction efficiency. It can be seen that an additional 4 to 5 wt% of total oil was recovered by the first washing step and no more appreciable amount of oil was recovered in the second washing step. The oil obtained from the washing step brought the total oil extraction efficiency to 94.5 wt% and 92.6 wt% for peanut and canola oil, respectively, approaching the results obtained in the batch scale. These results confirm that the oil was extracted effectively in the surfactant wash step but was not fully separated in the S/L separation step. The DI water in the wash step recovered the oil that was already released and stayed outside the cell structure. Table 3 shows the IFT between the peanut and canola oil with the extraction media at different washing stages by SAEP and AEP. The IFT value of the extracted oils with the first DI washing solution was about 2 mN/m for both peanut and canola oils, indicating that there was some surfactant remaining in the meal from the surfactant wash. The IFT values of the extracted oils with the second DI washing solution was 5–6 mN/m; similar to the IFT values of peanut and canola oils with DI washing only solution, which indirectly indicated that there was no appreciable amount of surfactant left in the meals. The basket centrifuge was used here because it was the only option available at the scale we needed for our system but is not the best option when operating the oilseed extraction in large-scale processes because it has limited solid holding capacity and will prevent the system from operating continuously. In a large-scale operating facility, we envision the use of the solid bowl-scrolling centrifuge or continuous pusher centrifuge which has been used widely in solid-liquid separation processes (29). However, this equipment was not available at our operational scale.

Figure 4 shows the effect of the inlet flowrate on the oil recovery and the water content in the oil phase at a constant centrifugation speed of 680 × g of the L/L centrifuge. The extracted-oil and surfactant-washed solution had 7–10 wt% of canola and peanut oil. It can be seen that the oil recovery decreased and the water content in the oil increased as the feed rate increased. The maximum moisture standard for crude peanut oil is 0.25 wt% (30) and for canola oil is 0.3 wt% (31). The highest oil recovery was achieved at the lowest inlet flowrate of 1 mL/min, corresponding to the longest residence time of 150 minutes. At this condition, the water content in the crude peanut and canola oils were 0.15 wt% and 0.22 wt%, respectively, and met the standard requirement (0.25 wt% for peanut oil

TABLE 3
IFT prewash and postwash extraction solution with refined peanut and canola oil measured at 20 minutes

| | Prewash IFT (mN/m) | Postwash IFT (mN/m) | 1st DI postwash IFT (mN/m) | 2nd DI postwash IFT (mN/m) |
|---|---|---|---|---|
| Canola[a] | 0.015 | 0.018 | 2.1 | 6.1 |
| Peanut[a] | 0.011 | 0.011 | 2.2 | 5.0 |
| Peanut[b] | 10.0 | 5.0 | NA | NA |

[a]SAEP.
[b]AEP.

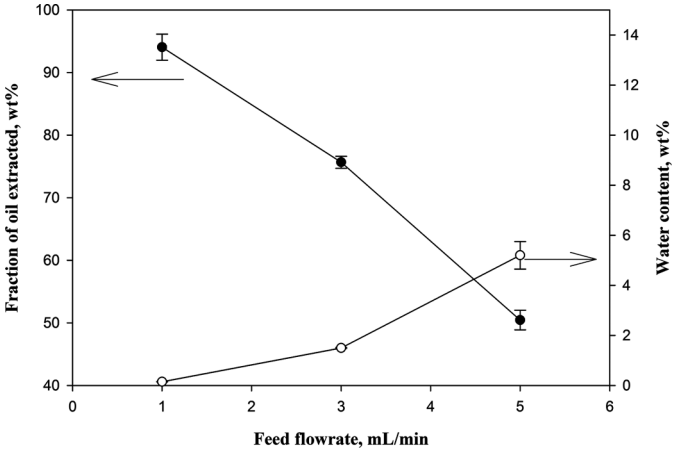


FIG. 4. Effect of feed flowrate on peanut oil recovery from liquid fraction at constant liquid/liquid centrifuge speed at 25°C. Extraction condition: 30 minute wash, S/L centrifuge at 4116 × g and 8 mL/min inlet flowrate, L/L centrifuge at 680 × g.

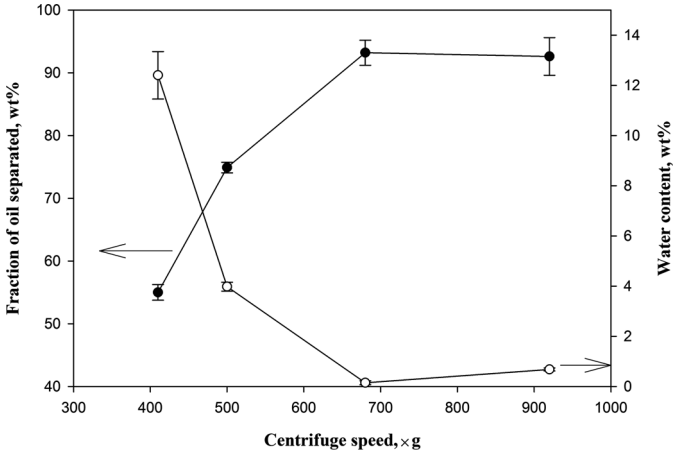


FIG. 5. Effect of centrifuge speed on peanut oil recovery from liquid fraction at a constant feed flowrate of 1 mL/min at 25°C. Extraction condition: 30 minute wash, S/L centrifuge at 4116 × g and 8 mL/min inlet flowrate.

and 0.30 wt% for canola oil). The longer residence time allowed the oil droplet to more efficiently separate from the emulsions (32). Free crude peanut and canola oils obtained from the L/L centrifuge at 1 mL/min and 680 × g are shown in Fig. 2(d). The extracted oils have excellent clarity with canola oil being more yellowish than peanut oil due to the color pigment of the oilseeds (Fig. 2a). In addition, the SAEP peanut and canola had fresh smell, whereas the hexane-extracted oil had a burnt smell. At a feed rate of 5 mL/min, there was a dramatic decrease in the free oil recovery to 51 wt% and an increase in water content of the oil phase to 5.2 wt% as the residence time decreased to 30 minutes. Although the long retention time for the demulsification process is a drawback in the aqueous-based extraction process, it might be offset by the high energy consumption and relatively long retention time to evaporate the hexane solvent and to obtain free crude oil in the hexane-extracted process. In addition, the oil obtained from aqueous-based process has been consistently reported to have superior qualities and required less refining steps than the oil from the hexane extraction process (3,8,14,18).

Figure 5 shows the effect of centrifugation speed (410, 500, 680, and 920 × g) on the oil recovery and moisture level in the oil at a constant feed rate of 1 mL/min. The effect of the centrifugation speed on the oil in water-emulsion demulsification can be understood by the following equation (32):

$$v_o = \frac{(\rho_w - \rho_o) \times r\omega^2 \times D^2}{18\mu_w} \quad (1)$$

where $v_o$ is the settling velocity of oil, $\rho_w$ is the density of water, $\rho_o$ is the density of oil, r is the radius of rotation, ω is the angular velocity of centrifugation, D is the diameter of the droplets, and $\mu_w$ is the viscosity of continuous phase, which is the aqueous surfactant solution in our case. From the equation, it is expected that the emulsion separation will be more efficient at higher centrifugation speed. The mechanism of oil in water emulsions separation by centrifugation was explained by Nour et al. (32). As the centrifuge $\omega$ rotation increases, more heat is generated, increasing the temperature of the fluid. The ratio of $\frac{\rho_w - \rho_o}{\mu_w}$ increases as the temperature increases because the water viscosity decreases much faster than the density difference (32), thereby increasing the settling velocity of the oil. When increasing temperature from 20°C to 40°C, we measured the viscosity of the aqueous phase, $\mu_w$, (surfactant/NaCl/water mixture) to be reduced by 60% whereas the change of ($\rho_w - \rho_o$) was reduced by only 4% (data now shown). In addition, during the gravimetrical separation process, oil droplets collide with each other and coalesce to form larger oil droplets, which also enhanced the oil/water separation process. From Fig. 5, it can be seen that when the centrifugation speed was increased from 410 to 680 × g, higher oil recovery and lower water content in the oil phase were obtained. However, at 920 × g, while there was no difference in the oil recovery ($p > 0.05$), the water content at 920 × g was higher than that at 680 × g. This might be due to the design of the continuous centrifuge where the fluid was introduced into a low-shear mixing chamber. When the rotation speed is too high in the continuous L/L centrifuge, it may cause the mixing to become too vigorous and worsen the separation process.

Table 4 shows the free crude-oil recovery obtained from the surfactant wash and DI wash step from the best runs. At similar L/L centrifuge condition, only 44.9% peanut oil and 38.5% canola oil was recovered as free oil phase

TABLE 4
Free crude oil recovery at optimum conditions[a]

| | Fraction of free oil recovered from SAEP[b] (wt%) | Fraction of free oil recovery from DI[b] washing (wt%) | Total free oil recovery[b] (wt%) |
|---|---|---|---|
| Peanut | 85.3 | 1.79 | 87.1 |
| Canola | 83.9 | 1.75 | 85.7 |

[a]30 minute surfactant solution extraction, 5 minute DI wash at 25°C, S/L centrifuge at 4116 × g and 8 mL/min inlet flowrate, and L/L centrifuge at 680×g and 1 mL/min inlet flowrate.

[b]Amount of oil extracted via Soxhlet extraction was used as the basis. Free crude oil has moisture level less than 0.25 wt% for peanut oil and 0.30 wt% for canola oil.

from the extracted oil-DI washing mixture versus more than 90% of free-oil recovery from the extracted oil-surfactant washing mixture. This result was expected because in the oil extracted-DI washing mixture, there was much lower oil content (1–2 wt%), therefore, the oil-in-water emulsion was much more stable and harder to separate (11). The total crude-oil recovery was at 87.1 wt% for peanut oil and 85.6 wt% for canola oil, which were lower than those obtained from the batch scale (14).

It is worth mentioning that mechanical treatment of the oilseeds for cell wall rupture is also a critical parameter in improving the oil-extraction efficiency. An approximately 50% increase in oil-extraction efficiency was achieved for soybean oil when the flour size was reduced from 0.40 mm to 0.10 mm (33). Therefore, we decided to grind the peanut to a finer size of less than 0.15 mm (mesh 120) versus the 0.21–0.42 mm (mesh 35–70) studied above to test the extraction efficiency. Table 5 shows the effect of particle size on total oil extracted and total free-oil recovery. It can be seen that while grinding improved the extraction efficiency to 93.2 wt% of oil from SAEP, the free-oil recovery dramatically decreased to 71 wt%. We attributed this result to the effect that excessively fine grinding will produce smaller oil globules, causing more stable emulsions which are harder to break (12). Recently, Lamsal et al. studied a mechanical treatment of oilseeds, whereby flaking the oilseeds first and then extruding the flakes, they could enhance the oil extraction efficiency without causing stable emulsions. While this could avoid the formation of stable emulsions, it also denatured the proteins due to the high temperature of the extruding process. This method can be employed in the case where protein recovery is not an important parameter.

TABLE 5
Effect of particle size on fraction of oil extracted and free crude oil recovery for peanut at 25°C[a]

| Mesh size | Flour size (mm) | Fraction of oil extracted from surfactant wash[b] (wt%) | Fraction of free crude oil recovery efficiency[c] (wt%) |
|---|---|---|---|
| 40−70 | 0.21–0.42 | 90.6 | 94.2 |
| Larger than 100 | <0.15 | 93.2 | 71.2 |

[a]Extraction condition: 30 minute surfactant wash, S/L centrifuge at 4116×g and 8 mL/min inlet flowrate.

[b]Amount of oil extracted via Soxhlet extraction was used as the basis.

[c]Total amount of oil in liquid fraction was used the basis; moisture level is less than 0.25 wt%.

## CONCLUSION

In conclusion, we have demonstrated that a semi-continuous pilot-scale system of aqueous surfactant-enhanced vegetable oil extraction was able to achieve a total oil-extraction efficiency similar to that obtained from batch scale (14) after aqueous surfactant and DI washing steps (25°C). However, the total crude oil recovery was at 87.1 wt% for peanut oil and 85.6 wt% for canola oil, which were lower than those obtained from batch scale. The S/L and L/L separation steps are critical parameters in oil extraction by SAEP, EAEP, and AEP. Further free-oil recovery from the skim of the L/L centrifuge outlet is very challenging. It will be worth studying the effect of SAEP on vegetable oil extraction of extruded flakes, in which the proteins were denatured, resulting in less stable emulsion problems. It is also worth studying the de-emulsification efficiency of extracted oil–surfactant solution mixture at higher temperatures, which was not within the scope of this study. Compared to other AEP processes, the SAEP process is very competitive because it achieves oil-extraction efficiency at 25°C similar to other AEP methods at 50–70°C in a reasonable time frame (30 minutes).

The scope of this study is to evaluate the pilot-scale process of vegetable oil extraction by aqueous-surfactant based process. Protein recovery from this process should be investigated in future research to evaluate the economic feasibility of this technology. Protein recovery from the aqueous-based method has been reported to have superior quality to that recovered from the hexane-based process (23) and can be used in human consumption, which has a higher market value compared to the protein produced from the hexane-extraction process which can only be used for cattle consumption. Similar to other aqueous-based process, adapting this technology was motivated by environmental issues. The vegetable oil-extraction industry has contributed the primary VOC emissions in the food industry (3). The annual hexane loss in the soybean oil-extraction process alone in the US could be as high as

210–430 million liters (3). Although the capitol cost of the aqueous-based extraction process is relatively higher than the hexane-extraction process (3), the low-surfactant concentrations (less than 0.5 wt%) and ambient operating condition might be advantageous compared to the hexane concentration at higher than 95 wt%. This should be further evaluated in future research.

## ACKNOWLEDGEMENTS

The authors would like to thank George Smith, Huntsman Petrochemical Corp. (Houston, TX) for providing us with the extended-surfactant samples; Gene Neuens from Producers Cooperative Oil Mill and Plains Oilseed Products Cooperative (OKC, OK), and Wayne Janzen from Prairie Gold Oil Seeds (Okeene, OK) for providing the canola seeds. Funding for this work has been provided through the Oklahoma Secretary of Energy, and the Oklahoma Center for Advancement of Science and Technology (OCAST). Partial support was provided by industrial sponsors of the Institute for Applied Surfactant Research at the University of Oklahoma: Akzo Nobel, Clorox, Conoco/Phillips, Church and Dwight, Ecolab, Halliburton, Huntsman, S.C. Johnson & Son, Oxiteno, Procter & Gamble, Sasol North America, Shell Chemical, and from Dr. Sabatini's Sun Oil Company endowed chair and Dr. Harwell's Conoco/Dupont endowed professorship. Brittany Pritchett and Nicole Rowlette are acknowledged for the experimental assistance.

## REFERENCES

1. N- Hexane Substance Technical Guidelines (accessed in May 2008). <http://ehs.ucsc.edu/lab_research_safety/pubs/facts/Hexane.pdf>
2. Juliano, B.O. (1985) *Rice: Chemistry and Technology,* 2nd Edn.; The American Association of Cereal Chemists, Minnesota. pp. 647–687.
3. Rosenthal, A.; Pyle, D.L.; Niranjan, K. (1996) Aqueous and enzymatic processes for edible oil extraction. *Enzyme and Microbial Technology*, 19: 402–420.
4. Hagenmaier, R.; Cater, CM.; Mattil, K.F. (1972) Critical unit operation of the aqueous processing of fresh coconuts. *J Am Oil Chem Soc.*, 49: 178–181.
5. Rosenthal, A.; Pyle, D.L.; Niranjan, K. (1998) Simultaneous aqueous extraction of oil and protein from soybean: mechanisms for process design. *Trans Inst Chem Eng Part C*, 76: 215–218.
6. Kim, S.H. (1989) Aqueous extraction of oil from palm kernel. *J Food Sci.*, 54 (2):491–492.
7. Cater, C.M.; Rhee, K.C.; Hagenmaier, R.D.; Mattil, K.F. (1974) Aqueous extraction – an alternative oilseed milling. *J Am Oil Chem Soc.*, 51: 137–141.
8. Sharma, A.; Khare, S.K.; Gupta, M.N. (2002) Enzyme-assisted aqueous extraction of peanut oil. *J Am Oil Chem Soc.*, 79 (3): 215–218.
9. Johnson, L.A.; Lusas, E.W. (1983) Comparison of alternative solvents for oils extraction. *J Am Oil Chem Soc.*, 60: 229–242.
10. Embong, M.B.; Jelen, P. (1977) Technical feasibility of aqueous extraction of rapeseed oil – a laboratory studies. *J Inst Can Sci. Technol Aliment*, 10: 239–243.
11. Moura, J.M.L.N.; Almeida, N.M.; Johnson, L.A. (2009) Scale-up of enzyme-assisted aqueous extraction processing of soybeans. *J Am Oil Chem Soc.*, 86: 809–815.
12. Lamsal, B.P.; Murphy, P.A.; Johnson, L.A. (2006) Flaking and extrusion as mechanical treatments for enzyme-assisted aqueous extraction of oil from soybeans. *J Am Oil Chem Soc.*, 83 (11): 973–979.
13. Campbell, K.A.; Glatz, C.E. (2009) Mechanisms of aqueous extraction of soybean oil. *J Agric Food Chem.*, 57: 10904–10912.
14. Do, L.D.; Sabatini, D.A. (2010) Aqueous extended-surfactant based method for vegetable oil extraction: Proof of concept. *J Am Oil Chem Soc.*, 87: 1211–1220.
15. Rosen, M.J. (1989) *Surfactants and Interfacial Phenomena*, 2nd Edn.; John Wiley & Sons, Canada. pp. 229–235.
16. Sharma, A.; Khare, S.K.; Gupta, M.N. (2002) Enzyme-assisted aqueous extraction of peanut oil. *J Am Oil Chem Soc.*, 79 (3): 215–218.
17. Moreau, R.A.; Dickey, L.C.; Johnston, D.B.; Hicks, K.B. (2009) A process for aqueous enzymatic extraction of corn oil from dry milled corn germ and enzymatic wet milled corn germ (E-Germ). *J Am Oil Chem Soc.*,86: 469–474.
18. Nasuk, A.; Sabatini, D.A.; Tongcumpou, C. (2009) Microemulsion-based palm kernel oil extraction using mixed surfactant solutions. *Industrial Crops and Products*, 30 (2): 194–198.
19. Do, L.D.; Withayyapayanon, A.; Harwell, J.H.; Sabatini, D.A. (2009) Environmentally friendly vegetable oil microemulsions using extended surfactant and linkers. *J Surf Deterg.*, 12 (2): 91–99.
20. Childs, J.D.; Acosta, E.; Scamehorn, J.F.; Sabatini, D.A. (2005) Surfactant-enhanced treatment of oil-based drill cuttings. *J Energy Resouce Technol.*, 207 (2): 153–162.
21. Rhee, K.C.; Cater, C.M.; Mattil, K.F. (1973) Aqueous process for pilot plant-scale production of peanut protein concentrate. *J Food Sci.*, 38: 126–128.
22. McCurdy, S.M. (1990) Effects of processing on the functional properties of canola/rapeseed protein. *J Am Oil Chem Soc.*, 67 (5): 281–284.
23. Carr, R.A. (1997) Oilseed Processing. In: *Technology and Solvents for Extraction Oilseeds and Nonpetroleum Oils*, Wan, P.J.; Wakelyn, P.J., eds.; AOCS Press: Champaign, pp. 101–120.
24. Lavin Centrifuge. Model L2. <http://www.lavincentrifuge.com/products/l2.html>
25. CINC Centrifuge. Model V02. <http://www.cincmfg.com/Model_V02_Centrifuges_s/29.htm>
26. Official Method of Analysis of the Association of Official Analytical Chemists, 16th edn, AOAC International, Gaithersburg, MD (1995).
27. Mattil, K.F.; Norris, F.A.; Swern, D. (1964) *Extraction of Fats and Oils. Bailey's Industrial Oil and Fat Products*, 3rd Edn.; John Wiley & Sons: London, pp. 637–717.
28. Lynch, J.M.; Barbano, D.M.; Fleming, J.R. (1997) Modification of babcok method to eliminate fat testing bias between babcock and ether extraction methods (Modification of AOAC Official Methods 989.04 and 995.18): Collaborative study. *J AOAC Int.*, 80 (4): 907–940.
29. Anlauf, H. (2007) Recent developments in centrifuge technology. *Separation and Purification Technology*, 58: 242–246.
30. Sanders, T.H. (2002) Groundut (Peanut) Oil. In: *Vegetable Oils in Food Technology: Composition, Properties and Uses*, Gunstone, F.D., ed.; CRC Press, Blackwell Publishing: USA/Canada, pp. 231–244.
31. Przybylski, R.; Mag, T. (2002) Canola/Rapeseed Oil. In: *Vegetable Oils in Food Technology: Composition, Properties and Uses*, Gunstone, F.D., ed.; CRC Press, Blackwell Publishing: USA/Canada, pp. 98–124.
32. Nour, A.H.; Mohammed, F.S.; Yunus, R.M.; Arman, A. (2009) Demulsfication of virugin coconut oil by centrifugation method: A feasibility study. *Int J Chem Technol.*, 1 (2): 59–64.
33. Rosenthal, A.; Pyle, D.L.; Niranjan, K. (2001) Simultaneous aqueous extraction of oil and protein from soybean: Mechanisms for process design. *Trans Inst Chem Eng Part C*, 76: 224–230.