A Twin-Screw Extruder for Oil Extraction: II. Alcohol Extraction of Oleic Sunflower Seeds[1]

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ABSTRACT: Our work is about the extraction of sunflower seed oil in a twin-screw extruder with or without the injection of 2-ethylhexanol and acidified 2-ethylhexanol. 2-Ethylhexanol is mixed with phosphoric acid. The oil recovery is increased to 90% by the co-injection of acidified alcohol. Mixing phosphoric acid with the alcohol enhances the lability of the oily spherosomes. Its addition increases the destruction of the membranes enveloping the lipid-containing organelles to release the oil more easily. Phosphoric acid exhibits an extracting and a degumming role. The best oil quality was obtained at a low extraction temperature (80°C), when 88% of the oil was removed. After alcoholic distillation, the oil exhibited a total acid value (mineral acidity plus organic acidity) of 4 mg KOH/g of oil and an organic phosphorus content below 30 ppm.

Paper no. J9106 in *JAOCS 76*, 1081–1086 (1999).

KEY WORDS: Acidic alcohol extraction, acid value, 2-ethylhexanol, high-oleic sunflower seeds, oil extraction yields, oil phosphorus content, oleic oil, phosphoric acid, twin-screw extruder.

The direct expression of oil from oleic sunflower seeds in a twin-screw extruder is an efficient alternative to conventional mechanical pressing in a single-screw press (1,2). Two twin-screw extruders of type BC21 and BC45 (Clextral, Firminy, France) were suitably configured for the pressing operation. The screw profile was progressively improved and the influence of pressing temperature and moisture content of the raw material was examined to obtain optimal oil yields. Yields of 80% have already been attained, and a good quality oil with a total phosphorus content of about 100 ppm has been produced. However, the resulting cake contains 15% residual oil (1,2).

In conventional trituration, this residual oil can be recovered by solvent extraction, and the oil yield can thereby be increased to 98%. The quality of this crude oil is poor and refining is required to make it suitable for subsequent use. The most frequently used solvent is hexane. It has, however, been reported to be neurotoxic so replacement of it is desirable (3). The alcoholic extraction of oleaginous seeds has been tried and tested (4–6), and isopropanol (6) and ethanol (7) have already been used for seed extraction. Isopropanol is the first branched alcohol tested and is in principle preferable as its heat of vaporization is of the order of 159 cal/g whereas that of ethanol is of 204 cal/g. These values are expressed in cal/g because this parameter is often used in the trituration industry where values to define the heat necessary to vaporize quantities of solvent are needed.

Jacks *et al.* (8,9) have shown that high extraction yields can be obtained by using hexane mixed with acetic acid for the extraction of cottonseed oil. The membranes of the cells containing the lipid droplets became labile owing to the acid, so that the oil was more easily liberated. The yield of the extraction of classic sunflower seed was also increased by approximately 5% by the addition of acetic acid (9) or sulfuric acid (10). We chose to use phosphoric acid solubilized in 2-ethylhexanol. 2-Ethylhexanol is a branched, 6 carbon-chain. Its heat of vaporization, about 93 cal/g, is close to that of hexane, about 75-80 cal/g. Its linear chain is similar to that of hexane and its branching to that of isopropanol. It seems to be an ideal candidate for oil extraction.

We had the idea of combining seed pressing and solvent extraction with 2-ethylhexanol and acidified 2-ethylhexanol in a single step within the twin-screw extruder, which would thus serve as a press, extractor, and oil separator (11). Our principal objective was to increase the extraction yield of sunflower seed oil using a single-step process. Another content of aims was to obtain a high-quality oleic sunflower oil, with a lower content of free fatty acids and phospholipids than that of oil obtained simply by pressing of sunflower seeds in a single-screw or twin-screw press.

This article is divided into two sections. The preliminary tests (experiments 1 and 2) served to determine the optimal ratio of injected 2-ethylhexanol/seeds. An experimental design technique was then set up which led to a study of the effect of the extraction temperature and of the injection of phosphoric acid on the extraction yield of oleic sunflower seed oil. Both factors influenced the quality of the collected oils and especially their organic phosphorus contents.

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[1]This work was presented as an oral communication at the 2nd American Oil Chemists' Society Europe Symposium, October 1–4, 1998, at Cagliari, Italy.

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EXPERIMENTAL PROCEDURES

Twin-screw extruder design. Oil expression was carried out using a BC21 co-rotating twin-screw extruder made by Clextral. The machine was operated by the Terminal Operator Intouch version 1.00 software (Clextral) so that the parameters controlling the twin-screw extruder function (input flow rate, torque) and the temperatures of the different barrel modules could be measured. The barrel consisted of seven modules 10 cm in length, and the input of whole seeds into the first module was controlled by a weight feeder of LWF-D55 type (Ktron; Niederlanz, Switzerland). A filtration module was inserted at position 6, the filters consisting of four hemispherical dishes with perforations 1 mm in diameter, with an open outlet on the seventh barrel module (no frontal plate). Modules 2, 3, 4, 5, and 7 were heat-regulated. The extruder is described in more detail in paper I (1).

The screw profiles consisted of trapezoidal double-thread screws to ensure efficient transport, monolobed paddle screws (designated DM) which exerted considerable pressure on the sides of the barrel, and bilobed paddle screws (BB) which had a pronounced shearing effect (1). The presence of a reverse screw, or screw elements carrying the contents in the opposite direction, immediately beneath the filtration module, guaranteed the formation of a so-called "dynamic plug". The screw profile tested is the same as Profile 4 in Figure 1 of Reference 1. The nature of the screw is indicated together with the pitch or mounting angle of the bilobed paddle screws (BB) and monolobed paddle screws (DM). This screw profile had previously been shown to correspond to the optimal oil expression yield from seeds alone (1). 2-Ethylhexanol, acidified or not with phosphoric acid, was injected into module 2.

Raw materials and chemicals. The sunflower seeds (variety Olbaril, 1996 harvest, La Toulousaine de Céréales, St Orens, France) had a high oleic acid content, ranging from 88 to 90% [NFT 60-223 and NFT 60-23 (12)], and a protein content of approximately 18% [NFV 18-100 (12)]. Dry matter content was 96.3% [NFV 03-903 (12)], and oil content was 50.9% [NFV 03-905 (12)].

2-Ethylhexanol was 99% pure (Prolabo, Gradignan, France). Mixing studies had confirmed that oleic sunflower oil was totally miscible with this alcohol. Phosphoric acid was 99% pure and in the form of white crystals (Prolabo). It was dissolved in the 2-ethylhexanol at ambient temperature before being injected into the extruder.

Operating conditions. The first tests, involving the injection of nonacidified alcohol, were carried out at 90°C with variable seed input flow rates, alcohol input flow rates, and screw rotation speeds.

In the second series of tests (injection of acidified alcohol) the seed input flow rate was fixed at 8 kg/h and the screw rotation speed at 125 rpm. The mass ratio of alcohol/seeds varied from 0.65 to 1.95 and the mass ratio of phosphoric acid/seeds from 0.10 to 0.42 (dissolution of variable quantities of phosphoric acid in the alcohol). The selected extraction temperature range was 80 to 90°C.

In the study to determine the effect of the extraction temperature and the amount of injected phosphoric acid (experiment 3) a mean alcohol/seeds ratio of 1.3 was selected (twice the alcohol stoichiometry in relation to the triglycerides). The seed input flow rate was fixed at 8 kg/h and the screw rotation speed at 125 rpm. A higher seed input flow rate with coinjection of alcohol and an alcohol/seed ratio of 1.3 led to excessive filling of the machine and forced the mixture back into the feeder module. The temperature varied from 80 to 100°C, the latter corresponding to the limit above which an excessive amount of alcohol evaporated. The mass ratio of H_3PO_4/seeds varied from 0.12 to 0.36.

Samples of filtrate (mixture of alcohol and oil containing the foot) and of cake were taken after the machine had been running for 5–10 min to ensure flow rate and temperature stability. They were handled in hermetic bottles made of glass and stored at +5°C.

Samples analysis. The filtrate, a mixture of liquid and some solid particles (foot), was centrifuged to separate the foot from the liquid. The triglycerides were quantified by thin-layer chromatography coupled with flame-ionization detection (13) and the extraction yield of triglycerides calculated from the relationship

$$\text{extraction yield (\% mass)} = 100 \cdot (Q_F \cdot T_F)/[(Q_F \cdot T_F) + (Q_S \cdot T_S)] \quad [1]$$

where Q_F is output flow rate of the filtrate (oil + alcohol) in kg/h; T_F is triglyceride content of the filtrate (determined after removal of the foot) as a percentage; Q_S is output flow rate of the cake in kg/h, and T_S , is triglyceride content on the cake as a percentage. The matter was extruded out of the machine as pellets or powder depending on the operating conditions. The residual triglyceride content of this cake was also measured.

In the third series of tests, the oil purity was measured by determining the total acidity of the centrifuged filtrate, expressed in mg KOH/g of triglycerides [NFT 60-204 (12)]. This indicated the free fatty acid and phosphoric acid (organic and mineral acidity) contents of the oil. For some tests, the phosphorus content (P), expressed in mg of phosphorus per kilogram of oil, of the oils obtained after distillation of the alcohol was also determined [NFT 60-227 (12)]. This indicated the content of inorganic phosphorus compounds (phosphate ions derived from phosphoric acid) and of organic phosphorus (from phospholipids). By determining the total phosphate content (PO_4^{3-}) by ionic chromatography, expressed in mg of phosphates per kilogram of oil, the amount of organic phosphorus present (phospholipids) could be calculated from the difference between the organic phosphorus and the inorganic phosphorus.

RESULTS AND DISCUSSION

Experiment 1: Study of the expression of sunflower oil assisted by 2-ethylhexanol. Yields of almost 80% were attained (Table 1). The addition of 2-ethylhexanol in certain cases facilitated the extraction by solubilizing the triglycerides, thus

TABLE 1
Oil Extraction Yield for the Extraction of Whole Sunflower Seeds by 2-Ethylhexanol in a BC21 Twin-Screw Extruder

Test	Operating conditions[a]	Mass ratio 2-ethylhexanol/ seeds	Extraction yield (without alcohol injections) (%)	Extraction yield (with alcohol injection) (%)
1	Q_I = 12 kg/h Ss = 125 rpm Q_A = 5.46 kg/h	0.45	72.7	79.1
2	Q_I = 12 kg/h Ss = 150 rpm Q_A = 8.09 kg/h	0.67	72.1	59.6
3	Q_I = 14 kg/h Ss = 125 rpm Q_A = 6.07 kg/h	0.43	73.1	78.6
4	Q_I = 14 kg/h Ss = 150 rpm Q_A = 9.11 kg/h	0.65	71.8	62.4

[a]Ss, screw rotation speed; Q_I, seed input flow rate; Q_A, alcohol input flow rate.

increasing the extraction yield by approximately 6% (tests 1 and 3, Table 1). The pressing efficacy could, however, be affected by a change in consistency of the mixture made of triturated seeds and alcohol (tests 2 and 4), the liquid/solid separation being less satisfactory in certain cases than when the seeds were expressed in the absence of alcohol. The extraction yield could then be reduced by approximately 10%. This phenomenon was all the more obvious when the amount of alcohol involved in relation to the seed was high (test 2). In fact, enough solid matter is needed to guarantee the formation of the dynamic plug that is necessary to press the matter in the filtration module.

Experiment 2: Study of the expression of sunflower oil assisted by 2-ethylhexanol acidified with phosphoric acid. The operating conditions are presented in Table 2 and associated yields are shown in Figure 1. Yields exceeding, 75% were obtained in all cases and could attain 88.5%. The first tests, performed with a mass ratio of 2-ethylhexanol/seed of 0.65, clearly showed that the injection of 2-ethylhexanol acidified with phosphoric acid improved the triglyceride extraction yield by 3.0 to 7.0% (tests 5 to 7, Fig. 1). Increasing the amount of acid injected was reflected by a better solid/liquid separation. The oil gain increased by 3.0 to 5.5% (tests 5 and 6, Fig. 1). The same occurred with a increase temperature increase, the oil gain increased by 3.0 to 7.7% for a temperature increase from 85 to 90°C (tests 5 and 7, Fig. 1).

At a mass ratio of 2-ethylhexanol/seeds of 1.95 the oil extraction yield was more sensitive to an increase in temperature (tests 10 and 11, Fig. 2) than to an increase in the level of phosphoric acid in the alcohol (tests 9 and 11). Increasing the amount of 2-ethylhexanol injected, with a mass ratio phosphoric acid/seeds content of 0.24 and a temperature of 90°C (tests 8 and 9) improved the triglyceride extraction yield by 3.5%. Increasing the 2-ethylhexanol/seed ratio was therefore apparently favorable for the oil extraction. But it is not necessarily of interest in that the oil would then have to be separated from the excess alcohol. It would be better to act on the phosphoric acid/seed ratio and the extraction temperature.

Oil extraction was improved by addition of the acid solution. This may be explained by the lability of the oil vacuoles to acid. During a study of the extraction of cotton and soybean seeds with a hexane/acetic acid mixture, carried out by

TABLE 2
Experimental Domain for Study of the Influence of Extraction Temperature and Quantity of Phosphoric Acid Injected on Extraction Yields of Whole Sunflower Seeds by Acidified 2-Ethylhexanol in a BC21 Twin-Screw Extruder

Test	Temperature (°C)	Mass ratio 2-ethyl-hexanol/ seeds	2-Ethyl-hexanol flow rate (kg/h)	Mass Ratio phosphoric acid/seeds	Phosphoric acid flow rate (kg/h)
5	85	0.65	5.2	0.10	0.80
6	85	0.65	5.2	0.16	1.28
7	90	0.65	5.2	0.10	0.80
8	90	1.30	10.4	0.24	1.92
9	90	1.95	15.6	0.24	1.92
10	80	1.95	15.6	0.42	3.36
11	90	1.95	15.6	0.42	3.36

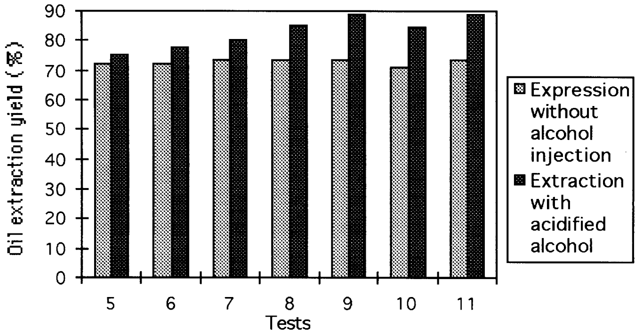

FIG. 1. Influence of extraction temperature and quantity of phosphoric acid injected on extraction yields of whole sunflower seeds by acidified 2-ethylhexanol in a BC21 twin-screw extruder. Test 5, T = 85°C; 2-ethylhexanol/seeds = 0.65; H_3PO_4/seeds = 0.10. Test 6, T = 85°C; 2-ethylhexanol/seeds = 0.65; H_3PO_4/seeds = 0.16. Test 7, T = 90°C; 2-ethylhexanol/seeds = 0.65; H_3PO_4/seeds = 0.10. Test 8, T = 90°C; 2-ethylhexanol/seeds = 1.30; H_3PO_4/seeds = 0.24. Test 9, T = 90°C; 2-ethylhexanol/seeds = 1.95; H_3PO_4/seeds = 0.24. Test 10, T = 80°C; 2-ethylhexanol/seeds = 1.95; H_3PO_4/seeds = 0.42. Test 11, T = 90°C; 2-ethylhexanol/seeds = 1.95; H_3PO_4/seeds = 0.42.

Jacks *et al.* (8,9), examination of the extracted cotton seeds with an electron microscope revealed that the addition of acid solution resulted in disintegration of the membranes enveloping the lipid-containing organelles so that the oil could more easily be released. Phosphoric acid possesses the required acid character to obtain spherosome lability.

Experiment 3: Study of the influence of the extraction temperature and the injection of phosphoric acid on the extraction of oleic sunflower oil assisted by 2-ethylhexanol. The Doehlert's method for the design of the experiments was used (14). The operating conditions and the results corresponding to the Doehlert experimental matrix for the factors "extraction temperature" and "mass ratio of H_3PO_4/seeds" are reported in Table 3. The variation of the extraction yield and the obtained oil quality were considered in relation to the organic phosphorus content of the oil.

A yield of 90.5% was attained at a temperature of 100°C and with a mass ratio of phosphoric acid/seeds of 0.24. Analysis of the isoresponse curves produced with the Nemrod software (15) for statistical analysis of polynomial models (Fig. 2), relating the response to the two factors, revealed that increasing the amount of added phosphoric acid had an adverse effect on the triglyceride extraction yield when the ratio of H_3PO_4/seeds exceeded 0.18. Thus, although a very low amount of phosphoric acid increased the lability of the oil-containing spherosomes and facilitated oil extraction by 2-ethylhexanol, excess mineral acid could result in transformation of the triglycerides. Furthermore, the minimal triglyceride extraction yield is dependent on temperature, this being even more pronounced when the ratio of H_3PO_4/seeds is low.

Yields of 90% are therefore attained at a very low temperature (80°C) and with a moderate H_3PO_4/seeds ratio (0.24). Increasing the temperature to 90°C brought the yield down slightly to 85%. At a high temperature, in contrast, the favorable effect of phosphoric acid on oil release seemed to be more important than the phenomenon of triglyceride degradation. Extraction yields exceeding 90% were obtained at 100°C, especially when the H_3PO_4/seed ratio was very low (<0.18).

As for the phosphorus content of oils obtained by extraction of oleic sunflower seeds with injection of 2-ethylhexanol and phosphoric acid (Table 3), a very high quality oil with an organic phosphorus content of 29 ppm was obtained with a pressing temperature of 80°C and a mass ratio of H_3PO_4/seeds of 0.24. Simple pressing of the seeds under identical conditions of temperature and seed input flow rate, but without the co-injection of alcohol or phosphoric acid, releases an oil with a total phosphorus content of 235 ppm. The injection of phosphoric acid is therefore highly favorable to the production of pure oil. Phosphoric acid shows an extracting role and a degumming role.

The same performances in terms of extraction yield and oil quality were obtained when the temperature was in-

TABLE 3
Experimental Domain for Study of the Effects of Extraction Temperature and Mass Ratio H_3PO_4/Seeds on the Extraction of Oleic Sunflower Seeds with 2-Ethylhexanol[a]

Temperature		Mass ratio H_3PO_4/seeds		Results			
X1	U1 (°C)	X2	U2 (kg/kg)	Triglycerides extraction yield (%)	Total acidity (mg KOH/g oil)	Organic phosphorus content (mg/kg oil)	Oil content of the cake (%)
1	100	0	0.24	90.5	8	n.d.	6
–1	80	0	0.24	87.5	8.5	29 (235[b])	8
0	90	0.866	0.36	79	7.5	n.d.	9.5
–0.2	88	–0.866	0.12	86	6	262 (235[b])	10
0	90	–0.866	0.12	84.5	6	n.d.	11.5
–0.7	83	0.866	0.36	82.5	14	n.d.	11
0	90	0	0.24	88.5	10.5	98 (220[b])	8.0

[a]X1,X2, coding values for the temperature and mass ratio H_3PO_4/seeds in relation to Nemrod software (15); U1, U2: experimental values; n.d., not determined.
[b]Values obtained for total phosphorus content of related oils obtained by pressing of oleic sunflower seeds in the BC21 twin-screw extruder without alcohol injection (Ref. 1); temperature, screw speeds and seed flow rate similar.

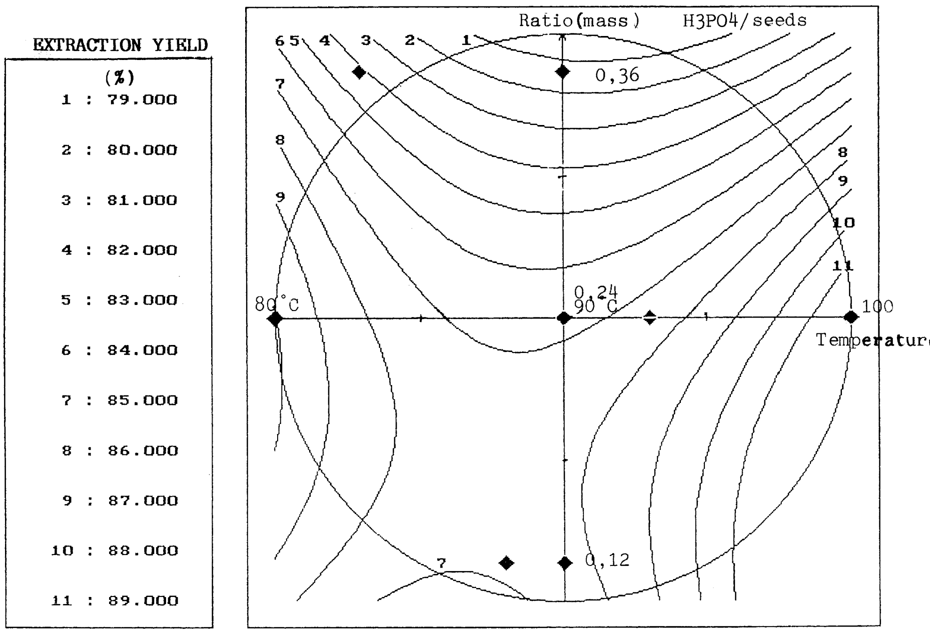

FIG. 2. Isoresponse curves for the oil extraction yield as a function of the extraction temperature and the mass ratio phosphoric acid/seeds (from Nemrod Software).

creased. An organic phosphorus content of 98 ppm was observed for an extraction temperature of 90°C and a mass ratio of H_3PO_4/seeds of 0.24. When the quantity of phosphoric acid was very low (ratio of H_3PO_4/seeds of 0.12), the oil purity remained equivalent to that of oil obtained by direct pressing of oleic sunflower seed without any injection of alcohol or phosphoric acid. It should be noted that phosphoric acid is traditionally used as a degumming agent of oils during refining. We can therefore consider that we are integrating a seed extraction operation with a simultaneous degumming operation within the twin-screw machine and thereby obtaining a high-quality oil. Most of the phospholipids seem to be complexed with proteins of the cake (16). They are hydrophilic constituents that are not soluble in the hydrophobic oil/2-ethylhexanol mixture.

The combination of pressing in a twin-screw extruder, having a suitably adapted configuration, together with the injection of 2-ethylhexanol acidified with phosphoric acid led to a oil extraction yield of over 90%. This oil is of very high quality, with an organic phosphorus content below 30 ppm (17).

However, the proposed twin-screw procedure is characterized by elimination of the seed-flattening and cooking steps, essential to the pretreatment of seeds prior to pressing in the conventional single-screw press with subsequent hexane extraction (5).

Using an alcohol as extracting solvent leads to a medium that can be directly used in lipid chemistry. The residual phosphoric acid is a good catalyst for the transesterification of the oil triglycerides into 2-ethylhexyl esters (2). Oleic oils, obtained by direct expression and acidified 2-ethylhexanol extraction of oleic sunflower seeds in a twin-screw extruder, are useful for oleochemical purposes and reactions (17) as oleic sunflower seeds are an important plant source of oleine and high purity oleic acid (>85%).

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[Received December 21, 1998; accepted April 14, 1999]