

## Comparison between ethanol and hexane for oil extraction from *Quercus suber* L. fruits

By Suzana Ferreira-Dias<sup>\*(1)</sup>, Dina G. Valente<sup>(1)</sup> and José M.F. Abreu<sup>(2)</sup>

<sup>(1)</sup> Depart. Agro-Indústrias e Agronomia Tropical, Centro de Microbiologia e Indústrias Agrícolas.  
e-mail: suzanafdias@mail.telepac.pt

<sup>(2)</sup> Depart. Produção Agrícola e Animal, Instituto Superior de Agronomia,  
Tapada da Ajuda, 1349-017 Lisbon; Portugal.

### RESUMEN

#### Comparación entre el etanol y el hexano para la extracción del aceite de los frutos de *Quercus suber* L..

El objetivo de este estudio fue (i) evaluar la posibilidad de reemplazar el *n*-hexano por el etanol para la extracción del aceite de los frutos de *Quercus suber* y (ii) optimizar, a escala de laboratorio, la preparación de la muestra y el tiempo de extracción para ambos solventes (*n*-hexano vs. etanol anhidro). Para ambos solventes, se ha evaluado el efecto del proceso de acondicionamiento en el rendimiento de la extracción. Para ello, se ha empleado un diseño factorial completo como función de cuatro variables: descascarillado (con vs sin cascarilla), y tratamiento térmico del material triturado (40°C vs. 75°C), a diferentes tiempos (5 min vs. 120 min) y presiones (10 kPa vs. 100 kPa). Los rendimientos más elevados de extracción de aceite se obtuvieron con *n*-hexano cuando los frutos descascarillados se acondicionaron a presión atmosférica. Cuando el material descascarillado fue tratado a 75°C por 90 min, se obtuvieron mejores rendimientos con *n*-hexano. El etanol demostró no ser adecuado para la extracción del aceite de los frutos de *Quercus*, debido a la extracción conjunta de otros materiales a la vez del aceite.

**PALABRAS-CLAVE:** Aceite-Bellotas-Etanol-Extracción-Hexano-*Quercus*.

### SUMMARY

#### Comparison between ethanol and hexane for oil extraction from *Quercus suber* L. fruits.

The aim of this study was to (i) evaluate the feasibility of replacing *n*-hexane with ethanol for the extraction of oil from *Quercus suber* fruits and (ii) optimize, at lab scale, sample preparation and extraction time for both solvents used (*n*-hexane vs. anhydrous ethanol). For both solvents, the effect of the conditioning process on extraction yield was evaluated. Therefore, a full factorial design was used as a function of four variables: dehulling (with vs. without husks), and thermal treatment of the crushed material (40°C vs. 75°C), at different times (5 min vs. 120 min) and pressures (10 kPa vs. 100 kPa). Higher oil yields were obtained with *n*-hexane when dehulled fruits were conditioned under atmospheric pressure. Better yields were obtained with *n*-hexane, when dehulled material was treated at 75°C for 90 min. Ethanol was not adequate for oil extraction from *Quercus* fruits, since other materials rather than oil were also extracted.

**KEY-WORDS:** Acorns-Ethanol-Extraction-Hexane-Oil-*Quercus*.

### 1. INTRODUCTION

*Quercus* genus plants are characteristic of the Mediterranean Flora. For instance, Portugal has more than one million hectares with *Quercus* trees which represents an annual production of approximately 300,000 tons of acorns. This production is close to the annual production of wheat, higher than the production of other grains (barley, oats, rye and rice) and accounts for about half of the production of corn in this country. The extraction of the oil of *Quercus* fruits in a profitable way could reduce the importation of edible oils. The oil content of the fruits from *Quercus rotundifolia* Lam. may go up to about 14% (Ferrão and Ferrão, 1988). Until the early seventies, the extraction of the oil from *Q. rotundifolia* acorns was carried out in some oil extraction plants in Portugal (Ferrão and Ferrão, 1988). In spite of the low oil content of the fruits from *Quercus suber* (4.4%-9.1%), this oil has a fatty acid composition similar to olive oil (Ferrão and Ferrão, 1988) and, therefore, its extraction would increase revenues to the *Quercus suber* industry. The exploitation of this tree mainly lies on the use of its cork. Moreover, the extracted meal could be used as animal feedstuff (Ferrão and Ferrão, 1988).

Currently, hexane, a solvent obtained from petrochemical sources, is the solvent used for oil extraction. This solvent can be emitted during extraction and recovery and has been identified as an air pollutant since it can react with other pollutants to produce ozone and photochemical oxidants (Wan *et al.*, 1995a; Hanmoungjai *et al.*, 2000). Safety, environmental and health concerns have increased the interest in alternative solvents to hexane to reduce the emissions of volatile organic compounds to the atmosphere as well as potential traces of hexane in edible oils after refining.

As an alternative to organic solvent extraction, the extraction of oil from oil-containing materials with aqueous solutions (acidic or alkaline at 45°C to 85°C) has been investigated (Rhee *et al.*, 1972; Hagenmaier *et al.*, 1973; Hagenmaier, 1974; Lusas *et al.*, 1982; Kim, 1989; Southwell and Harris, 1992;

Rosenthal *et al.*, 1996; Rosenthal *et al.*, 1998; Hanmoungjai *et al.*, 2000). However, due to the low selectivity of aqueous solutions for lipids, simultaneous extraction of proteins, carbohydrates and other compounds also occur. Also, aqueous extraction is limited by (i) the lower efficiency of oil extraction, (ii) the need of an additional de-emulsification step to recover the oil and (iii) the production of an aqueous effluent which requires further treatment (Hanmoungjai *et al.*, 2000).

The most feasible alternative to hexane extraction seems to be the replacement of this solvent by other organic solvents recognized as environmentally safer. Several studies have been carried out, both at laboratory and pilot scales, aimed to replace hexane with other hydrocarbons (Wan *et al.*, 1995a; Wan *et al.*, 1995b; Conkerton, *et al.*, 1995) or alcohols (Abraham *et al.*, 1988; Rittner, 1992; Hron *et al.*, 1994; Sineiro *et al.*, 1998) as solvents for oil extraction. Among hydrocarbon solvents, heptane and isohexane were recommended as potential substitutes for hexane to extract oil from cottonseed (Wan *et al.*, 1995a; Wan *et al.*, 1995b; Conkerton *et al.*, 1995). With respect to the use of alcohols, isopropanol and ethanol are the most promising solvents for the oil extraction from cottonseed (Abraham *et al.*, 1988; Hron *et al.*, 1994), sunflower seed (Sineiro *et al.*, 1998) and soybean (Baker and Sullivan, 1983; Rittner, 1992). Ethanol is a worthy candidate to investigate as an alternative solvent since its cost is low and it may be produced from a large variety of biological materials using simple technology. In addition, although flammable (flash point= 8.9°C; ignition temperature= 425°C), this alcohol is recognized as non-toxic and has less handling risks than hexane (flash point = -23°C; ignition temperature= 225°C) (Rittner, 1992). It can also be obtained by fermentation and therefore labelled as "natural". The use of this alcohol as an extraction solvent also avoids eventual toxicity problems of meals for animal feedstuff.

The aim of this study was to (i) evaluate the feasibility of replacing *n*-hexane with ethanol for the extraction of the oil from *Quercus suber* fruits and (ii) to optimize the extraction conditions, namely preparation of samples (dehulling and conditioning), and extraction time for both solvents used (*n*-hexane v.s. ethanol), at laboratory scale.

For seeds with high oil content, a conditioning treatment ("cooking") is currently carried out prior to mechanical pressing/expelling oil extraction. In general, a thermal treatment (conditioning technique) of seeds or other fatty materials with low oil content (lower than 20-25%), prior to solvent extraction, may also increase oil yield and quality (Hoffman, 1989). For instance, the best extraction conditions for cracked soybeans are achieved after heating at 55-75°C for about 20-30 min to reach 11%

moisture. For cottonseed, the best results are obtained with a treatment at 65°C for 10-12 min (Hoffman, 1989).

In this study, the effect of conditioning on the yield of extracted oil was investigated, both for *n*-hexane and ethanol. Therefore, a full factorial design was used as a function of the following variables: dehulling, thermal treatment of the crushed material at different times and pressures.

## 2. MATERIALS AND METHODS

### 2.1. Materials

Ripened fruits from a *Quercus suber* L. tree were obtained in Lisbon, Portugal. Anhydrous ethanol p.a. and *n*-hexane p.a. were used.

### 2.2. Methods

#### Size reduction

The fruits (with or without husks) were crushed in a coffee mill (knife cutter type) for about 20 sec.

#### Moisture content of the fruits

Samples (5g) of both whole and dehulled crushed fruits were dried at 103°C to attain a constant mass. Moisture content was expressed on a dry basis. Analyses were run in triplicate.

#### Optimization of fruit conditioning

The effect of dehulling the fruits, conditioning temperature (40°C vs.75°C), time (5 min vs.120 min) and pressure (10 kPa vs.100 kPa) on the yield in oil was investigated, both when *n*-hexane or ethanol were used as extraction solvents. A full factorial design 2<sup>4</sup> (2 levels and 4 factors) was followed and Table I contains the decoded experimental design (2<sup>4</sup> = 16 experiments).

With this experimental design, several variables (factors) are tested simultaneously with a minimum number of trials. In addition, variables and interactions with significant effect on the extraction yield will be identified (Gacula and Singh, 1984; Haaland, 1989; Montgomery, 1991).

For these experiments, the extracts were obtained in a Soxhlet apparatus for 5 h at the boiling points of the solvents tested (78°C and 67-69°C, for ethanol and *n*-hexane, respectively). A ratio of crushed fruits to solvent of 1:6 (m/v) was used. Subsequently, the solvent was evaporated under reduced pressure and the amount of extracted material evaluated by weighting. The yields of extract (%) were expressed on a dry basis, i.e., mass of extract per mass of dry matter.

After full factorial experiments, additional experiments were carried out in an attempt to determine the near-optimum extraction time with *n*-hexane. In these experiments, dehulled *Quercus* acorns were used and conditioning occurred at 75°C under normal pressure.

#### Water activity assay

The water activity ( $a_w$ ) of the crushed fruits after conditioning was measured at 25°C in a Rotronic Hygroskop DT with a lithium chloride humidity sensor (DMS-100H).

#### Optimization of extraction time

After defining the best conditions for sample preparation for oil extraction, the extraction time was optimized. The oil was obtained from the fruits by solvent (*n*-hexane *p.a.*) extraction in a Soxhlet apparatus. To optimize the extraction time, the

solution was recovered in a flask, at different extraction times, the solvent evaporated under reduced pressure and the extracted oil quantified. Experiments were carried out in duplicate and the results were expressed on a dry basis.

#### Statistical analysis

The analysis of the results of the full factorial experiments was performed by using the software "Statistica™", version 5, from Statsoft, USA.

The linear effects of each of the 4 factors under study (dehulling, temperature, pressure and conditioning time), as well as their linear interactions, on the yield in oil were calculated, when both ethanol and *n*-hexane were used. Their significance was evaluated by analysis of variance. The results of the full factorial experiments were used to establish first-order models. First-order coefficients were generated by regression analysis. The goodness of fit of the models was evaluated by the determination

Table I  
Decoded Full Factorial Design, 2<sup>4</sup>: the effect of different conditioning conditions (i) on the water activity ( $a_w$ ) values of crushed fruits, after conditioning and before extraction, and (ii) on the amount of extracted material (dry basis), when *n*-hexane or ethanol were used. (Factors: with (1) or without husks (0), temperature, T, time, t, and pressure, P)

Experiment	Experimental Design (Decoded Matrix)				Experimental Results		
	Husks	T (°C)	t (min.)	P (kPa)	$a_w$	Hexane extraction (%)	Ethanol extraction (%)
1	0	40	5	10	0.948	3.5	25.9
2	1	40	5	10	0.914	3.4	22.9
3	0	40	5	100	0.936	3.0	36.0
4	1	40	5	100	0.946	3.6	19.3
5	0	75	5	10	0.933	5.3	21.2
6	1	75	5	10	0.949	4.2	20.7
7	0	75	5	100	0.935	3.7	22.5
8	1	75	5	100	0.940	3.9	21.4
9	0	40	120	10	0.277	3.3	19.9
10	1	40	120	10	0.302	3.8	20.9
11	0	40	120	100	0.935	3.7	21.9
12	1	40	120	100	0.921	4.6	18.9
13	0	75	120	10	0.250	4.6	21.2
14	1	75	120	10	0.377	4.3	18.1
15	0	75	120	100	0.237	5.1	23.6
16	1	75	120	100	0.155	4.9	26.5

coefficients (squared correlation coefficient,  $R^2$ ) and adjusted  $R^2$  ( $R^2_{adj}$ ) (Weisberg, 1985; Haaland, 1989) as well as by the plot of predicted values by the model vs. observed experimental values (Doran, 1995). High values of both  $R^2$  and  $R^2_{adj}$  suggest a good fit of the model to the experimental data points (Weisberg, 1985).

### 3. RESULTS AND DISCUSSION

#### 3.1. Moisture content of the fruits

The average moisture content (dry basis) of whole and dehulled *Quercus suber* fruits were 57.3% ( $\sigma=0.04$ ) and 59.4% ( $\sigma=0.03$ ), respectively. These results were used to express the amount of extracts obtained with ethanol or *n*-hexane on a dry basis.

Optimization of the conditioning operations of the fruits: Both the decoded matrix and the obtained results (i.e.,  $a_w$  values of crushed fruits, after conditioning and before extraction, and the amount of extracted material by *n*-hexane or ethanol) are shown in Table I. These data were used to calculate the significant linear effects of each variable and their

interactions on the experimental responses to select the best operation conditions (Table II).

With respect to  $a_w$  values of the crushed fruits, after conditioning and before extraction, only conditioning time had a significant and negative effect on them. As expected, the water activity decreased when longer drying times were used. The other factors tested had no significant effect on  $a_w$  values.

When hexane was used for oil extraction, the temperature and conditioning time, as well as the interaction (Pressure) x (Time), had a significant and a positive effect on the yield of oil (Table II). The interaction (Husks) x (Temperature) showed to have a negative significant effect on the extraction yield. Therefore, the fruits should be dehulled, crushed and conditioned under atmospheric pressure, at higher temperatures and times, prior to hexane extraction.

Both for water activity values and oil extracted by *n*-hexane, high values of  $R^2$  and  $R^2_{adj}$  were obtained for the models fitted to the experimental data points. However, the goodness of fit and therefore the selection of the most suitable model should not be done on the basis of  $R^2$  and  $R^2_{adj}$  values alone. The  $R^2$  and  $R^2_{adj}$  indicators must be complemented by

Table II  
The effects of different conditioning factors and respective significance levels ( $\alpha$ ) on (i) the water activity ( $a_w$ ) of crushed fruits, after conditioning and before extraction, and (ii) on extract yield when either hexane or ethanol were used (\* at  $\alpha<0.05$ ; \*\* at  $\alpha<0.01$ ; (a) not significant effects). Determination coefficients ( $R^2$ ) and adjusted  $R^2$  ( $R^2_{adj}$ ) of the fitted models to experimental data

Factor	$a_w$	Hexane extraction	Ethanol extraction
(1) Husks	0.0066(a)	0.0625(a)	-4.7400(a)
(2) Pressure	0.1319(a)	0.0125(a)	0.6100(a)
(3) Temperature	-0.1754(a)	0.8875**	-3.1150(a)
(4) Time	-0.5058**	0.4625*	-0.5600(a)
(1) x (2)	-0.0269(a)	0.3125(a)	-3.3400(a)
(1) x (3)	0.0099(a)	-0.4125*	0.6850(a)
(1) x (4)	0.0074(a)	0.1625(a)	4.1900(a)
(2) x (3)	-0.1924(a)	-0.2125(a)	-1.0150(a)
(2) x (4)	0.1286(a)	0.5625**	2.0900(a)
(3) x (4)	-0.1786(a)	-0.0125(a)	5.0650(a)
$R^2$	0.9141	0.9496	0.7455
$R^2_{adj}$	0.7423	0.8487	0.2365

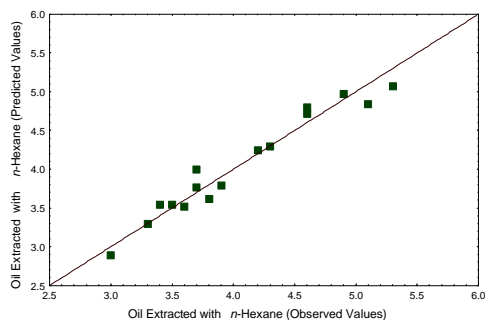


Figure 1  
Relationship between the experimental values of the extracts obtained with *n*-hexane and the corresponding values estimated by a first-order model (line).

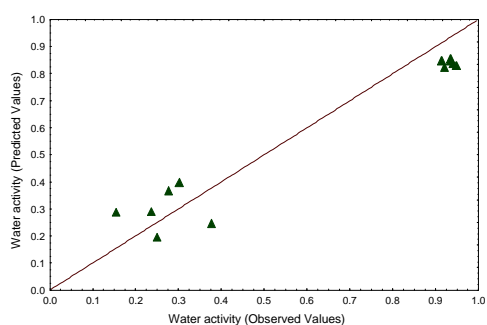


Figure 2  
Relationship between the experimental values of water activity of the samples, after conditioning and before extraction, and the corresponding values estimated by a first-order model (line).

Table III  
Additional experiments for the establishment of a near-optimum conditioning time of dehulled *Quercus* fruits at 100 kPa and 75°C (experiments were run in duplicate; oil extracted with *n*-hexane)

Conditioning Time (min)	Average Oil Yield (% w/w)
60	4.4
90	5.1
120	5.1

other tools, such as the graphic plots of theoretical values predicted by the model vs. observed experimental values, to decide on the goodness of fit

of the models (Doran, 1995). In fact, for hexane extraction, a linear relationship between the predicted values and the experimental values was observed (Fig. 1), confirming the adequacy of the first order model. However, for the water activity values of the samples before extraction, experimental points are scattered around the theoretical line "observed values vs predicted values" (Fig. 2) indicating a certain lack of fit of the model.

For the extraction of oil with *n*-hexane from dehulled *Quercus* fruits, the following first order polynomial equation can be used to estimate the yield in oil:

$$\text{Oil Yield} = 4.06 + 0.89 T + 0.46 t + 0.56 (P \times t)$$

where the oil is expressed in mass percentage on a dry basis, *T* is the temperature (°C); *t* is time (min) and *P*, pressure (kPa) of conditioning of the acorns prior to solvent extraction.

In addition, complementary experiments were carried out in an attempt to determine the near-optimum conditioning time, for the oil extraction with *n*-hexane, when the highest temperature tested (75°C) was used (Table III). The best yield in oil was obtained after a treatment at 75°C for 90 min.

When ethanol was used, the amounts of extracts obtained were from 4 to 12 times higher than the extracts achieved with *n*-hexane (reference solvent) (Table I). However, in spite of the observed variation, no significant effects of the conditioning factors tested were observed on ethanol extraction (Table II). This may be ascribed to a lower selectivity of ethanol for the oil with a consequent extraction of other compounds such as phosphatides, polyphenols, pigments and soluble sugars (Hron *et al.*, 1982; Hron *et al.*, 1994; Pomeranz and Meloan, 1994; Sineiro *et al.*, 1996). In fact, in acorns from *Q. rotundifolia*, contents of total phenolic compounds up to 6.8%, in acorn pulp, and 9.3%, in acorn hulls, were quantified

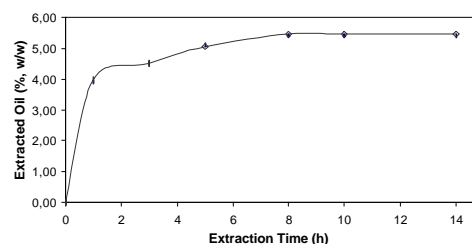


Figure 3  
Selection of extraction time- Extraction of the oil from dehulled *Quercus suber* fruits with *n*-hexane, after conditioning at 75 °C and 100kPa for 90 min (experiments were run in duplicate).

(Bruno-Soares *et al.*, 2000). Ethanol showed not to be an adequate solvent for the direct extraction of the oil from *Quercus* acorns.

### 3.2. Optimization of the oil extraction time

Due to the low selectivity of ethanol for oil extraction from *Quercus* fruits, the optimization of the extraction time was only carried out for *n*-hexane extraction. When this solvent was used in a Soxhlet equipment, after about a 8-hour extraction time, no significant increase in extracted oil was observed (Fig. 3).

Alternative organic solvents to hexane for oil extraction from *Quercus* fruits have to be investigated.

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### REFERENCES

- Abraham, G., Hron, R.J., Koltun, S.P. (1988). Modeling the solvent extraction of oilseeds. *J. Am. Oil Chem. Soc.*, **65**, 129-135.
- Baker, E.C., Sullivan, D.A. (1983). Development of a pilot-plant process for the extraction of soy flakes with aqueous isopropyl alcohol. *J. Am. Oil Chem. Soc.*, **60**, 1271-1277.
- Bruno-Soares, A.M., Farinha, S., Abreu, J.M. (2000). Effects of the ripening on chemical composition and gas production of *Quercus rotundifolia* acorn, in *Animal Production for a Consuming World*, AAASP/ASAP International Congress, Australia (in press).
- Conkerton, E.J., Wan, P.J., Richard, O.A. (1995). Hexane and heptane as extraction solvents for cottonseed: a laboratory-scale study. *J. Am. Oil Chem. Soc.*, **72**, 963-965.
- Doran, P.M. (1995) *Bioprocess Engineering Principles*, pp 27-48. Academic Press, London.
- Ferrão, J.E.M., Ferrão, A.M.B.C. (1988). A bolota e a gande, potencialidades em óleo e farinha. *Revista de Ciências Agrárias*, **2**, 15-29.
- Gacula, M.C. Jr., Singh, J. (1984) *Response Surface Designs and Analysis in Statistical Methods in Food and Consumer Research. Food Science and Technology. A Series of Monographs*, pp 214-273. Academic Press Inc., New York.
- Haaland, Perry D. (1989) *Experimental Design in Biotechnology, Statistics: Textbooks and Monographs*, D.B. Owen, (Ed.). Marcel Dekker, Inc., New York and Basel, pp. 258.
- Hagenmaier, R., Cater, C.M., Mattil, K.F. (1973). Aqueous processing of fresh coconuts for recovery of oil and coconut skim milk. *J. Food Sci.*, **38**, 516-518.
- Hagenmaier, R.D. (1974). Aqueous processing of full-fat sunflower seeds: yields of oil and protein. *J. Am. Oil Chem. Soc.*, **51**, 470-471.
- Hanmoungjai, P., Pyle, L., Niranjan, K. (2000). Extraction of rice bran oil using aqueous media. *J. Chem. Technol. Biotechnol.*, **75**, 348-352.
- Hoffman, G. (1989). *The Chemistry and Technology of Edible Oils and Fats and their High Fat Products*, pp 63-64, Academic Press, New York.
- Hron, R.J. Sr., Koltun, S.P., Graci, A.V. Jr. (1982). Biorenewable solvents for vegetable oil extraction. *J. Am. Oil Chem. Soc.*, **59**, 674A-684A.
- Hron, R.J. Sr., Kuk, M.S., Abraham, G., Wan, P.J. (1994). Ethanol extraction of oil, gossypol and aflatoxin from cottonseed. *J. Am. Oil Chem. Soc.*, **71**, 417-421.
- Kim, S.H. (1989). Aqueous extraction of oil from palm kernel. *J. Food Sci.*, **54**, 491-492.
- Lusas, E.W., Lawhon, J.T., Rhee, K.C. (1982). Producing edible oil and protein from oilseeds by aqueous processing. *Oil Mill Gazetteer*, **4**, 28-34.
- Montgomery, D.C. (1991) *Design and Analysis of Experiments*, pp 197-256, John Wiley and Sons, New York.
- Pomeranz, Y., Meloan, C.E. (1994) *Food Analysis. Theory and Practice*, 3<sup>rd</sup>. Ed. pp. 625-677, Chapman & Hall, New York.
- Rhee, K.C., Cater, C.M., Mattil, K.F. (1972). Simultaneous recovery of protein and oil from raw peanuts in an aqueous system. *J. Food Sci.*, **37**, 90-93.
- Rittner, H. (1992). Extraction of vegetable oils with ethyl alcohol. *Oléagineux*, **47**, 29-42.
- Rosenthal, A., Pyle, D.L., Niranjan, K. (1996). Aqueous and enzymatic processes for edible oil extraction. *Enzyme Microb. Technol.*, **19**, 402-420.
- Rosenthal, A., Pyle, D.L., Niranjan, K. (1998). Simultaneous aqueous extraction of oil and protein from soybean: mechanism for process design. *Trans IChemE*, **76C**, 224-230.
- Sineiro, J., Domínguez, H., Núñez, M.J., Lema, J.M. (1996). Ethanol extraction of polyphenols in an immersion extractor. Effect of pulsing flow. *J. Am. Oil Chem. Soc.*, **73**, 1121-1125.
- Sineiro, J., Domínguez, H., Núñez, M.J., Lema, J.M. (1998). Ethanolic extraction of sunflower oil in a pulsing extractor. *J. Am. Oil Chem. Soc.*, **75**, 753-754.
- Southwell, K.H., Harris, R.V. (1992). Extraction of oil from oilseeds using hot water floating method. *Trop. Sci.*, **33**, 251-262.
- Wan, P.J., Pakarinen, D.R., Hron, R.J. Sr., Richard, O.L., Conkerton, E.J. (1995a). Alternative hydrocarbon solvents for cottonseed extraction. *J. Am. Oil Chem. Soc.*, **72**, 653-659.
- Wan, P.J., Hron, R.J. Sr., Dowd, M.K., Kuk, M.S., Conkerton, E.J. (1995b). Alternative hydrocarbon solvents for cottonseed extraction: plant trials. *J. Am. Oil Chem. Soc.*, **72**, 661-664.
- Weisberg, S. (1985) *Applied Linear Regression*, pp. 217-218, John Wiley and Sons, New York.

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