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Characteristics and Composition of Watermelon Seed Oil and Solvent Extraction Parameters Effects

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Abstract Watermelon seed oil characteristics were evaluated to determine whether this oil could be exploited as an edible oil. Hexane extraction of watermelon seeds produced yields of 50% (w/w) oil. The refractive index, saponification and iodine value were 1.4712 (at 25 °C), 200 mg KOH/g and 156 g I/100 g, respectively. The acid and peroxide values were 2.4 mg KOH/g and 3.24 mequiv/kg, respectively. The induction time of the oil was also 5.14 h at 110 °C, which was measured for the first time. Total unsaturation contents of the oil was 81.6%, with linoleic acid (18:2) being the dominant fatty acid (68.3%). Considering that the watermelon seed oil was highly unsaturated, the relatively high induction time might indicate the presence of natural antioxidants. In addition, the influence of extraction parameters on extraction of oil from watermelon seed with hexane as a solvent was studied at several temperatures (40, 50, and 60 °C), times (1, 2, and 3 h) and solvent/kernel ratios (1:1, 2:1, and 3:1). The oil yield was primarily affected by the solvent/kernel ratio and then time and temperature, respectively. The protein content of the oil-free residue was 47%.

Keywords Watermelon seed · Solvent extraction · Oil characteristics · Fatty acids · Taguchi

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Introduction

The world's supply of vegetable oils is currently in excess of 100 million metric tonnes (MMT). The demand is increasing at a rapid pace due to increasing demand for non-food uses of vegetable oil, for example in biodiesel, oleochemicals, lubricants, pharmaceuticals, and cosmetics. However, only about 12 of the ∼500,000 known plant species are currently commercially exploited to produce vegetable oils in order to meet the world's increasing demand [1].

Watermelon (*Citrullus lanatus*) is one of the major under-utilized fruits grown in the warmer part of the world. The juice or pulp from watermelon is used for human consumption while rind and seeds are major solid wastes [2]. The rind is utilized for products such as pickles and preserves as well as for extraction of pectin [3], whereas seeds are a potential source of protein [4–6] and lipids [7]. Melon seeds are used for oil production at the subsistence level in Nigeria, in several other African countries, and in the Middle East. In Nigeria, such seeds are used for oil extraction at the village level but not on an industrial scale for oil or protein production. The dry seed of watermelon has been reported to contain on average about 32 g of protein and 51.4 g of fat per 100 g sample [6]. There are a few reports in the literature about the fatty acid compositions of seed oils from some varieties of watermelons and some acknowledgement of the good quality of these seed oils [7, 8].

The objective of this study was to determine the physicochemical properties and the fatty acid composition of watermelon seed oil and for the first time, to measure the induction time of watermelon seed oil. The present work also compares the effects of temperature, mixing time and solvent/kernel ratio on the solvent extraction of watermelon seed oil, which was done using the Taguchi method to reduce the time and cost for experimental investigations and to improve the performance characteristics.

Experimental Procedures

Materials

Ten batches of certified watermelon (*Citrullus lanatus*) were purchased from a local market (Chaf Village, Langrod, Iran) during the summer season of 2007. The process of selecting the watermelon seed was done randomly.

Methods

Watermelons were cut by a sharp knife and the seeds were hand-collected and washed with tap water, then sun-dried at 30 °C for a week. The seeds were then shelled by cracking them with a small iron rod and manually peeled to remove the kernels. The kernels were ground using a coffee grinder (Black & Decker, Towson, Maryland, USA) and the fine flour of the kernels placed in a refrigerator (5 °C) until analysis.

Physicochemical Properties

The crude fat by the Soxhlet method (7.056) and the total nitrogen (micro-Kjeldahl; 2.057) were determined according to the methods of the AOAC [9]. The protein was calculated as N × 5.3 [9]. The refractive index (RI, at 25 °C), the acid value (AV, Cd 3a–63), the peroxide value (PV, Cd 8–53), the saponification value (SV, Cd 3–25), the iodine value (IV, Cd 1–25) and the unsaponifiable matter (UM, Ca 6a–40) of the oil samples were determined according to the AOCS [10].

The oxidative stability of watermelon seed oil was evaluated by an accelerated automated test using the 743 Rancimat (Metrohm, Herisau, Switzerland) at 110 °C. Oil (3 g) was placed into the Rancimat apparatus and heated under an airflow rate of 4 L/h. When the temperature reached 110 °C (35 min), the vessels head outlets were connected to the conductivity cells, the air flow rate was increased to 20 L/h, and the measurement was started. The time taken until there was a sharp increase in conductivity is termed the induction time (IT), and it is expressed in hours [11].

All analyses were carried out three times and expressed as means ± SD.

Fatty acid Composition

The methyl ester (969.33) of crude oil was prepared according to the AOAC [9], using boron trifluoride. A PR2100 gas chromatograph (Perichrom, France) was used to separate the methyl esters using helium as the carrier gas. A CP SIL 8CB column (60 m × 0.25 mm × 0.39 mm i.d., Varian Inc., USA) at temperature 210 °C was used. Reference compounds were standard FAME mixtures from Supelco and Sigma (Sigma Chemical Co., St Louis, MO, USA).

Solvent Extraction Parameters

For the purpose of analyzing the effects of temperature, time, and solvent ratio on solvent extraction of oil from watermelon seeds, 4 g of the flour samples was extracted with hexane as a solvent under different combination of parameters (temperature at 40, 50, and 60 °C, extraction time for 1, 2, and 3 h and solvent/kernel ratio 1:1, 2:1, and 3:1 w/w). The solution was continuously stirred with a magnetic stirrer for selected periods in a water bath at the desired temperature. After cooling, the solution was centrifuged in a cooling centrifuge at 10,000 rpm for 20 min at 4 °C. Then, the solvent was removed with a rotary evaporator and the remaining oil was weighed.

The Taguchi method was employed to reduce the number of experiments and improve the performance characteristics. The Taguchi method uses special orthogonal arrays to study all the design parameters using a minimum number of experiments. Orthogonal array means that parameters can be evaluated independently of one another; the effect of one parameter does not interfere with the estimation of the influence of another parameter [12]. In the present study, three levels are defined for each of the parameters and a L9 orthogonal array scheme was adapted, which required nine experiments instead of 27 individual experiments to complete the process [13]. After conducting the experiments, the results were converted into signal-to-noise (S/N) ratio data. In the Taguchi method, the 'S/N' data is subjected to analysis of variance (ANOVA) [12].

Results and Discussion

Physicochemical Properties

The physicochemical properties of watermelon seed oil are compared with average properties of soybean and sunflower seed oils in Table 1. The yields of watermelon seed oil are appreciably higher than many other oily vegetables such as palm (40%), soybeans (18–22%), rapeseed (41%), and sunflower (40%) [14]. The protein content of watermelon seed is 47% (w/w), which is higher than sunflower seed (40%) and lower than soybean (51%) base on oil-free residue [15].

Table 1 Physicochemical properties of watermelon seed oil

Property	Watermelon seed oil	Sunflower seed oil	Soybean oil
Refractive index (25 °C)	1.4712 ± 0.001	1.4735	1.4728 [14, 15]
Saponification value (mg KOH/g oil)	200 ± 0.1	194 [14, 15]	191
Iodine value (g I/100 g oil)	156 ± 0.2	137	131
Acid value (mg KOH/g oil)	2.4 ± 0.1	4	1.4
Peroxide value (mequiv O_2/kg oil)	3.24 ± 0.1	10 [14, 15]	2.4 [14, 15]
Unsaponifiable matter (% g/g oil)	3 ± 0.2	1.02	1.6 [14, 15]
Induction time (h) at 110 °C	5.41 ± 0.1	3.7 [18]	8.25 [17]
Oil yield (g/100 g dry seed)	50 ± 0.2	40 [14, 15]	21 [14, 15]
Protein yield (g/100 g oil-free seed)	47 ± 0.1	40	51 [15]

Values represent the averages of three analyses ± SD

Table 2 Fatty acid compositions (%, w/w) of watermelon seed oil

Fatty acid (%)	Watermelon seed oil	Sunflower seed oil [14, 15]	Soybean oil [14, 15]
Palmitic (C16:0)	11.36 ± 0.1	6.8	11
Stearic (C18:0)	7.04 ± 0.03	4.7	4
Oleic (C18:1)	13.25 ± 0.1	18.6	23.4
Linoleic (C18:2)	68.3 ± 0.03	68	53.2
Others	0.05	2	7.8
Total saturated fatty acids	18.4	11.5	15
Total unsaturated fatty acid	81.55	88.6	84.4

Values represent the averages of three analyses ± SD

The RI for watermelon seed oil and those of soybean and sunflower were similar. This similarity in RI suggests that these three sets of oils have similar degrees of unsaturation.

The SV of watermelon seed oil (200 mg KOH/g oil) clearly suggests that watermelon seed oil consist mainly of medium-chain fatty acids (i.e. C16 and C18). The IV for watermelon seed oil, 156 g I/100 g oil (Hanus), is higher than the range of IV for soybean and sunflower oils, 110–143 g I/100 g Oil, reported in [14, 15]. However, the differences in the IV were small and this again corroborates the inference drawn from the RI value regarding the similarity of unsaturation in these oils.

The AV and PV for watermelon seed oil of 2.4 mg KOH/g oil and 3.24 mequiv O_2/kg oil, respectively, fall well within the Codex standards for vegetable oils. These measurements indicate the amount of primary oxidation products and free fatty acid released by hydrolysis (generally deterioration) in watermelon seed oil was lower than sunflower seed oil and a little higher than soybean oil. It must be mentioned that refined oils have an AV and a PV of less than one.

The UM of watermelon seed oil was more than soybean and sunflower seed oils. The UM of oil include tocopherols, sterols, triterpenic alcohols, hydrocarbons, aliphatic alcohols, and waxes [14]. Generally, these results were in good agreement with those reported by El-Adawy [7] for watermelon seed oil except IV (115 g I/100 g Oil) and UM (0.93% g/g oil) values were less than the ones in our results.

Fatty Acid Composition

The fatty acid compositions of the watermelon seed oil are given in Table 2, which shows the principal fatty acid components in the watermelon seed oil to be linoleic (68.3%), oleic (13.3%), palmitic (11.4%), and stearic (7%) acids. The results are comparable to the findings of El-Adawy [7]. However, the samples have less oleic (18%) and higher linoleic (59.6%) reported by El-Adawy [7]. From Table 2, the close resemblance between the watermelon seed oil and the oils from soybean and sunflower is very apparent. Palmitic acid content in the watermelon seed oil is comparable with those in soybean and sunflower oils. Stearic acid composition in the watermelon seed oil is higher than it is in the commercial oils. Thus, the level of total saturation (18.4%) in the watermelon seed oil is higher than that in the commercial oils (4–17%, Table 2). The unsaturation profiles of the watermelon seed oil are similar to those of soybean and sunflower oils, where both sets of seed oils have linoleic acid as the most abundant fatty acid, followed by oleic acid. However, the oleic acid content in watermelon seed oil is lower than the two other oils. In general, higher polyunsaturation in soybean oil is due to α-linolenic acid, 18:3n-3 (5.5–10%), which is not present in watermelon seed oil.

Table 3 The results of orthogonal test L9

Tests	Solvent/kernel ratio	Temperature (°C)	Time (h)	Oil extraction (w/w)
1	1:1	40	1	9 ± 0.1
2	1:1	50	2	11.3 ± 0.1
3	1:1	60	3	17.5 ± 0.2
4	2:1	40	2	22.5 ± 0.1
5	2:1	50	3	34.8 ± 0.1
6	2:1	60	1	26.4 ± 0.2
7	3:1	40	3	33.3 ± 0.1
8	3:1	50	1	25.3 ± 0.1
9	3:1	60	2	31.5 ± 0.1

Values represent the average of three analyses ± SD

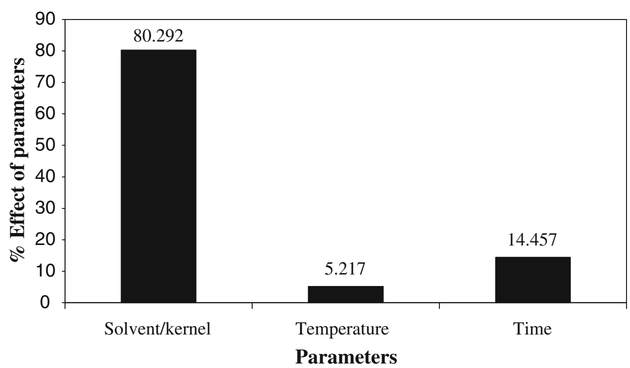

Fig. 1 Significant parameters influences on watermelon seed oil extraction

From a technological point of view, the Rancimat technique is usually applied to oils in order to measure their oxidative stability. In other words, the induction time indicates the length of time that oil may be expected to resist oxidation [16]. The induction time (Table 1) for watermelon seed oil was 5.41 h at 110 °C, which is lower than those of soybean oil, 8.25 h [17], 16 h at 100 °C [15] and 4 h at 120 °C [18], and higher than sunflower seed oil, 3.7 h [18] and 6 h at 100 °C [15]. Considering the degree of unsaturation of soybean oil, the high oxidative stability of soybean oil was probably due to the content of antioxidants (total tocopherols: 60–340 mg/100 g [14]). However, watermelon seed oil has higher fatty acid saturated levels, the 5.41-h induction time could also be due to the present of natural antioxidants in this oil. No data on the oxidative stability of watermelon seed oils were available in literature for comparative purposes.

Effects of Solvent Extraction Parameters

The extraction results performed under orthogonal design conditions are shown in Table 3. The results show that by increasing the three parameters, time, temperature and solvent/kernel ratio, the oil extraction yield was improved. The significant parameters on oil extraction as a percentage of influences indicate that the solvent/kernel ratio had a greatest influence on oil extraction (Fig. 1).

Conclusion

Data obtained from watermelon seed oil characteristics compare well with those of soybean and sunflower oils. Watermelon seeds could be utilized successfully as a source of edible oils for human consumption. Watermelon seed oil might be an acceptable substitute for highly unsaturated oils. Considering that the watermelon seed oil was a highly unsaturated oil, the induction time of 5.41 h at 110 °C might indicate the presence of natural antioxidants in this oil. The Taguchi model developed for oil yield demonstrated that the solvent/kernel ratio had the greatest effect on oil extraction. These results will be helpful in designing the best conditions to maximize the oil extraction. Finally, the utilization of watermelon seed for oil production could provide extra income and at the same time help to minimize waste disposal problems.

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