Sesame Oil. I. Properties of a Solvent-Extracted Sesame Oil[1]

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SESAME oil is obtained from the seed of the plant *Sesamun indicum* Loew, which is a member of the Pedaliaceae family. Sesame has been cultivated since ancient times in tropical and sub-tropical regions. During recent years the annual world production of sesame seed has been approximately 1.5 million tons, of which approximately one-half is grown in China and one-third in India. During World War II there was a marked stimulation of sesame production in the Western Hemisphere amounting to more than 100,000 tons annually in 1945 of which Mexico produced about 88,000 tons. In Central and South America production began in the late 1930's, especially in Nicaragua, Guatemala, Costa Rica, El Salvador in Central America, and Colombia and Venezuela in South America.

Sesame seed contains approximately 50% of an oil which is valuable for edible purposes and, after extraction, yields a residual meal which is a high-protein feed. Despite the valuable properties of its seed and the adaptability of the plant to a variety of climatic and soil conditions, the production of sesame in many countries, among them the United States, has been uneconomic, owing to the fact that all known varieties of sesame have had the tendency to shatter on ripening. Consequently the crop has not been adaptable to mechanical harvesting.

Recently however as the result of an intensified breeding program begun in 1940 in Venezuela by Langham (1), an indehiscent or non-shattering type of sesame has been under development which is adaptable to mechanical harvesting. The completion of this development will have a far-reaching effect on the economics of the production of sesame as an oilseed crop. At the present time work aimed at perfecting non-shattering varieties is in progress at the South Carolina, North Carolina, and Nebraska Agricultural Experiment stations, The United States Department of Agriculture Experiment Station, Beltsville, Maryland, and at the Texas State Research Foundation, Renner, Texas. These already fruitful investigations have a reasonable expectation of attaining their objective during the next several years.

Existing literature on sesame oil deals chiefly with the glyceride composition of the oil with little pertinent data with respect to its extraction, processing, and its industrially important characteristics. Part of these data and information are supplied in this and succeeding reports on sesame seed oil. The present report deals with the extraction, processing, and characteristics, including the stability, of oil extracted from a sample of white sesame seed.

Methods of Analyses

Free fatty acids, smoke and flash points, Reichert-Meissl and Polenske values, titer, saponification and iodine values were determined by methods of the American Oil Chemists' Society (2). Unsaponifiable matter (3) and hydroxyl numbers (4) were determined by the methods described in the references cited. Saturated acids were determined by a modification of the method of Pelikan and von Mikusch (5), using sintered glass filter sticks for filtration. Thiocyanogen values were determined by the modified method and with the reagent described by Lambou and Dollear (6) and were employed with the iodine values in equations recommended by the American Oil Chemists' Society (2) for calculating the glyceride composition of oils.

Spectrophotometric analyses of the oils and hydrogenated oils were made by a modification of the method of Mitchell *et al.* (7), which is under collaborative test by the Spectroscopy Committee of the American Oil Chemists' Society. In this method samples of 0.1 gram were isomerized for 25 minutes at 180°C. with a 1.3 *N* potassium hydroxide-ethylene glycol reagent and were protected from oxidation by a stream of purified nitrogen. The results of the spectrophotometric analyses were calculated by the method of Swain *et al.* (8). Saturated acids, as well as iso-oleic acids, were determined by the Twitchell lead salt-alcohol method (2). Micropenetrations were measured by the method of Feuge and Bailey (9).

Experimental

Crude Oil. Ninety pounds of white Nicaraguan-grown sesame seed were flaked to 0.004 inch thickness using 12-inch diameter flaking rolls. The flaked seed were extracted with hexane in a vertical batch extractor, the operation and design of which has been previously described (10). A total of 202 gallons of hexane at 20°C. were pumped through the flaked seed at the rate of 20 gallons per hour. The seed contained: moisture 4.9%, total lipids 49.5%, nitrogen 4.2%, protein (N × 6.25) 26.4% and the extracted meal moisture 8.8%, total lipids 3.7%, nitrogen 7.0%, protein (N × 6.25) 43.8%,

The miscella from the extractor was concentrated to an oil content of 73.3% by evaporating the solvent at atmospheric pressure, after which it was filtered. A portion of the miscella was stripped with steam (11) at *ca.* 140°F. and 14-mm. pressure. The stripped oil was centrifuged to break the emulsion which formed during the stripping operation. The stripped, centrifuged oil contained 0.26% volatile matter and moisture. It was found that bubbling nitrogen through the miscella at 90° C. under reduced pressure removed solvent from the oil in an equally satisfactory manner. The physical and chemical characteristics of the crude oil are shown in Table I.

Refined Oil. The crude oil was refined with caustic lye using several variations of the refining method described in the Official and Tentative Methods of the American Oil Chemists' Society for refining degummed soybean oil (2). Additional refinings were made to determine the effect of variations in the

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TABLE I

Physical and Chemical Characteristics of Crude Sesame Oil

Characteristic	Value
Free fatty acids (as oleic), %	1.0
Color, Lovibond, 5¼″ cell	35Y, 4.3R
Unsaponifiable matter, %	2.3
Smoke point, °F	330
Flash point, °F	607
Specific gravity, 25°/25°	0.918
Refractive index, N_D^{50}	1.4629
Hydroxyl number	5.3
Reichert-Meissl value	0.51
Polenske value	0.47
Titer, °C	22.0
Saponification value	185.8
Iodine value	112.0
Thiocyanogen value	76.3

amount and concentrations of lye, the effect of refining degummed oil, and the effect of adding sodium silicate and glycerin was investigated. A degummed oil was prepared by vigorously stirring the crude oil at approximately 70°C. while spraying it with hot water (7.5% of weight of oil). Centrifugation was used to break an emulsion which formed during the degumming operation. This treatment reduced the phosphorous content from 0.003 to 0.001%.

TABLE II

Refining Data on Sesame Oil [a]

No.	Lye		Time of stirring in cold, min.	Refining loss per cent	Consistency of foots	Refined oil, Lovibond color 35Y/xR
	°Bé	Excess,[b] per cent				
R-1	12	0.1	90	2.8	Firm	1.4
R-2	12	0.1	45	2.7	Soft	1.5
R-3[c]	12	0.1	90	2.2	Very firm	1.2
R-4[c]	12	0.1	45	2.3	Very firm	1.3
R-5[d]	12	0.1	90	2.4	Very firm	1.3
R-6	12	0.2	90	2.7	Very firm	1.5
R-7	12	0.2	45	3.2	Very firm	1.5
R-8[c]	12	0.2	90	3.0	Very firm	1.2
R-9	14	0.1	90	2.8	Very soft	1.6
R-10[e]	14	0.1	90	3.8	Very soft	1.3
R-11[d]	14	0.1	90	2.9	Very soft	1.5
R-12	14	0.1	45	2.9	Very soft	1.5
R-13	14	0.25	90	3.9	Very soft	1.6
R-14	14	0.3	90	4.4	....	1.6
R-15	14	0.5	90	3.9	....	1.6
R-16	14	0.6	90	3.3	....	1.6
R-17	16	0.2	90	4.1	Very soft	1.6
R-18	16	0.4	90	5.0	Very firm	1.6
R-19	20	0.1	90	6.3	Very firm	1.6
R-20[e]	20	0.1	90	3.5	Very firm	1.4
R-21	20	0.3	90	5.7	Firm	1.6

[a] Refined by Official American Oil Chemists' Society Method Ca 9C-46 (2) except as otherwise specified.
[b] Excess solid sodium hydroxide based on weight of oil used.
[c] Degummed oil.
[d] 1.0% glycerin mixed with lye.
[e] 1.5% sodium silicate mixed with oil 5 minutes before lye was added.

In refining tests which are summarized in Table II uniformly low refining losses were obtained with various amounts and strengths of alkali. The refining losses were not appreciably affected by prior degumming the crude oil, or by the addition of glycerin or sodium silicate during refining. The addition of sodium silicate produced better settling of the foots and subsequent decantation of the neutralized oil.

Bleached Oil. Refining reduced the color of the crude oil from 35 yellow and 4.3 red to 35 yellow and 1.4 red Lovibond units. The refined oil was bleached according to the method of the American Oil Chemists' Society (2), except that in some tests the effect of variations in the quantity and nature of bleaching earths was investigated as indicated in the data in Table III. From the data included in Table III it is evident that the sesame oil was readily bleached by as little as 2% neutral clay with a reduction in color to 3.0 yellow and 0.6 red Lovibond units.

Hydrogenated Oil. Some data have been published relative to the hydrogenation of sesame oil. Joglekar and Jatkar (12) investigated the hydrogenation of sesame oil at temperatures ranging from 100° to 180°C., using a semi-dried nickel catalyst, but their results do not show any selectivity of hydrogen addition. Fiero (13) hydrogenated sesame oil and investigated its antioxidant properties. Hilditch *et al.* (14) hydrogenated sesame oil in a stepwise manner in connection with an investigation of the glyceride structure of this oil. Two conditions of hydrogenation were used one of which was apparently selective.

Table III

Effect of Bleaching on the Color of Sesame Oil

Oil	Lovibond color	
	Yellow	Red
Crude	35	4.3
Refined	35	1.5
Bleached [a]		
(1) Official Fuller's earth, 6%	3	0.8
(2) Neutral clay, 4%	3	0.5
(3) Neutral clay, 2%	3	0.6
(4) Neutral clay, 4% + active carbon, 1%	3	0.5
(5) Neutral clay, 2% + active carbon, 1%	3	0.6
Degummed, refined	35	1.2
Degummed, refined and bleached with 2% neutral clay	3	0.5

[a] Bleached by Official American Oil Chemists' Society Method Cc 8a-47 (2).

To supplement the above-mentioned information on the hydrogenation of sesame oil, an investigation of the effect of selective conditions of hydrogenation on the composition, stability, and plasticity of hydrogenated oil was included in the present investigation.

Hydrogenation was accomplished in a vertical, cylindrical steel hydrogenator provided with a motor-driven stirrer of the paddle type. The hydrogenator was charged with 16 pounds of refined and bleached sesame oil and a catalyst containing 0.1% nickel based on the weight of the oil. The catalyst was of the precipitated type supported on filter cel and dry-reduced at 900°F. The oil was hydrogenated at 375°F. and 15 p.s.i., which conditions correspond to those previously found to be selective (15). Samples were withdrawn at intervals from the hydrogenator as the oil successively decreased in iodine value.

Iodine and thiocyanogen values, spectrophotometric analyses, the Twitchell lead salt-alcohol, and the Bertram oxidation methods were applied in determining the composition of the original and hydro-

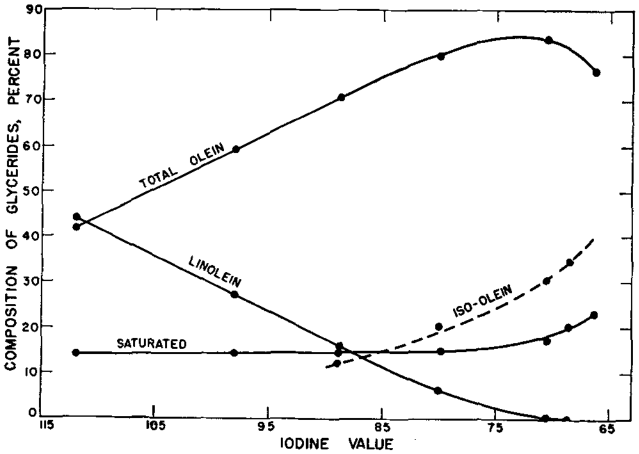

FIG. 1. Effect of hydrogenation on the composition of sesame oil.

TABLE IV

Composition, Refractive Index, Micropenetration, and Stability of *Unhydrogenated* and Hydrogenated Sesame Oil

Sample	Iodine value	SCN value	*Percentage composition of glycerides*											Refractive index, 50°C.	Micro-penetration, mm./10 at 25°C.	Keeping time, A.O.M., hours
			Linolein		Olein					*Saturated*						
								ISO	Normal							
			A_1	B_2	A_1	B_2	D_4	D_4	D_4	A_1	B_2	C_3	D_4			
SO-0	112.1	76.3	44.0	46.0	41.7	33.4	......	......	......	14.3	20.0	14.1	4....	1.4629	......	19
SO-I	98.2	76.0	27.1	27.7	59.5	55.7	......	......	......	13.4	16.6	14.5	......	1.4615	......	43
SO-II	88.8	75.4	16.2	13.7	70.6	74.5	71.5	12.3	59.2	13.2	11.8	14.7	14.4	1.4607	......	96
SO-III	79.9	75.4	6.7	6.6	79.4	79.0	77.2	20.6	56.5	13.9	14.4	15.6	16.3	1.4590	......	159
SO-IV	71.5	71.2	0.0	0.1	83.1	82.9	80.2	30.5	49.5	16.9	17.0	17.3	19.4	1.4578	145	682
SO-V	68.7	68.4	0.0	0.0	79.9	79.9	79.0	34.7	44.4	20.1	20.1	20.4	21.0	1.4579	68	895
SO-VI	66.3	66.6	......	......	77.1	......	......	......	......	22.9	......	......	......	1.4576	44	......
SO-VII	64.0	64.3	......	......	74.4	......	......	......	......	25.6	......	......	......	1.4572	28	1094
SO-VIII	61.7	61.5	......	......	71.7	......	......	......	......	28.3	......	......	......	1.4571	21	1305
SO-IX	58.8	58.0	......	......	68.4	......	......	......	......	31.6	......	......	......	1.4569	19	......
SO-X	45.5	42.0	......	......	52.9	......	......	......	......	47.1	......	......	......	1.4553	......	1926

A_1: Iodine-thiocyanogen method; saturated acids calculated by difference. B_2: Spectrophotometric method. C_3: Modified Bertram oxidation method. D_4: Twitchell lead salt-alcohol method.

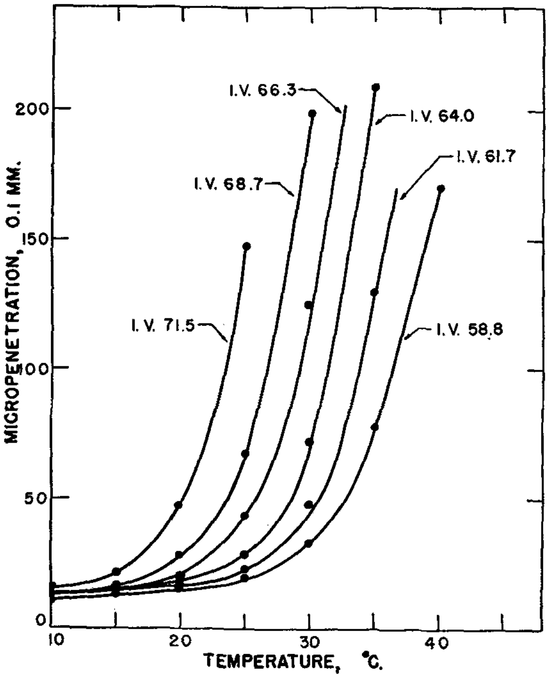

FIG. 2. Micropenetration values of sesame oil hydrogenated to various iodine values.

genated oils. The plasticity of the hydrogenated oils was determined with a micropenetrometer. Stabilities of the oils were determined by the active oxygen method. These data are given in Table IV and in Figures 1 and 2.

Calculation of the composition from iodine-thiocyanogen values yielded results for the saturated acids (as glycerides) which agree closely with the corresponding values determined by the modified Bertram oxidation method. The spectrophotometric method gave values for the content of linoleic and saturated glycerides somewhat at variance with the corresponding values determined by the iodine-thiocyanogen method for samples SO-0, SO-I, and SO-II. The content of glycerides of saturated acids determined by the spectrophotometric method were also at variance with the results determined by the modified Bertram oxidation method for the same samples. The Twitchell lead-salt alcohol method gave values for the content of saturated glycerides which were generally slightly higher than those obtained by the modified Bertram oxidation method.

Measurements of the plasticity of the various samples by the micropenetration method indicate that samples SO-IV and SO-V, which had iodine values of 71.5 and 68.7, had micropenetration at 25°C. of 145 and 68 (mm./10). respectively, hence their consistencies were in the plastic range of ordinary shortenings.

The original and especially the hydrogenated sesame oils exhibited marked stabilities. For example, samples SO-IV and SO-V, which had consistencies approximately equivalent to that of shortening, had stabilities of 682 and 895 hours, respectively, or approximately four times the stability of acceptable commercial all-hydrogenated type shortenings (200 hours).

The stabilities of unhydrogenated and hydrogenated sesame oils have been mentioned in the literature. Johnson and Frey (16) measured the induction periods of such oils with Barcroft-Warburg apparatus and Fiero (13) reported that hydrogenated sesame oil exhibited antioxygenic activity when added to other fats in large amounts. Some years previous to these observations Grettie (17) stated that the addition of from 5 to 10% of hydrogenated sesame oil to other fats exerted a marked antioxygenic effect. These observations were confirmed and extended by observations of the effect of blending hydrogenated sesame oil with unhydrogenated and hydrogenated cottonseed oils as shown in Table V. These data indicate that, at least in the case of sesame-cottonseed oil mixtures, a high proportion of sesame oil is necessary to obtain a marked antioxygenic effect.

The unusual stability of hydrogenated sesame oil suggests that the oil contains one or more antioxidants of greater activity than the antioxidants present in most of the other vegetable oils of commerce. On the other hand, the failure of the addition of hydrogenated sesame oil to produce any appreciable

TABLE V

Stability of Blends of Hydrogenated Sesame Oil with Unhydrogenated and Hydrogenated Cottonseed Oils

Sample No.	Composition of oils			
	Hydrogenated sesame,[a] per cent	Cottonseed, per cent	Hydrogenated cottonseed,[b] per cent	Stability, A.O.M., hours
1	0	100	0	9
2	10	90	0	10
3	15	85	0	11
4	25	75	0	14
5	40	60	0	19
6	0	0	100	180
7	10	0	90	250
8	25	0	75	335
9	50	0	50	493
10	100	0	0	1926

[a] Wijs iodine value, 45.5. [b] Wijs iodine value, 65.9.

stabilizing effect on unhydrogenated or hydrogenated cottonseed oils is similar to the previously observed ineffectiveness of adding tocopherols (18) or concentrates of vegetable oil antioxidants (19) to vegetable oils. An investigation of the nature and action of the antioxidants present in sesame oil forms the subject of another communication in this series.

Summary

Data and information have been presented with respect to the extraction, processing characteristics, and the chemical and physical characteristics of oil obtained from white sesame seed.

Extraction of sesame seed with hexane yielded a crude oil low in free fatty acids and in color. The oil was refined with caustic soda under a variety of conditions with low losses and bleached with comparatively small quantities of several bleaching earths each of which produced a light-colored oil.

Data have been presented on progressive changes which occurred in the composition, stability, plasticity, and refractive index of the fat during selective hydrogenation of the refined and bleached oils.

Sesame oil hydrogenated to shortening consistency exhibited extremely high stability when tested by the accelerated active oxygen method. This stability confirms previous suggestions that sesame oil contains one or more antioxidants of greater activity than those present in most of the other vegetable oils of commerce.

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Lespedeza Seed Oil

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Introduction

LESPEDEZA has become one of the major crops of the southern states. Rather large quantities of the seed are harvested annually. The 1948 crop in North Carolina alone is estimated at 36 million pounds (1). Possibly because of its recent growth to such production no data are available in the literature on the isolation or characterization of the oil from this seed. Not only the potential availability, but also the fact that as a member of the *Leguminoseae lespedeza* seed oil might resemble soy bean oil, creates an interest in this oil. The present investigation was therefore undertaken, and we wish to report at this time preliminary data on the isolation and characterization of lespedeza seed oil.

Experimental

Seed Used: The seed used in this study was 1948 crop of the Korean variety obtained from open stock in the Farmers Exchange, Carrboro, N. C. The seed are small, black ovals about $\frac{1}{16}$ inch in length and are covered with a light tan hull.

Extraction: The seed were prepared for extraction by grinding in a ball mill. No attempt was made to separate the hulls prior to extraction. The seed sample, 100 g., were placed in a 1-qt. porcelain ball mill containing 50 steel balls, $\frac{3}{8}$ inch in diameter, and rotated at 100 R.P.M. for four hours. The seed were crushed to a fine powder by this treatment.

Various solvents were used in extraction of the powdered seed to determine which was most effective. The sample to be extracted, 10.0 g., was extracted for three hours in a Soxhlet extractor. The sample was then removed, crushed in a mortar and pestle, and returned for an additional hour of extraction. The solvent and extract were evaporated in a tared evaporating dish on a steam bath to give the weight of extracted oil. Data with the four solvents used are given in Table I. The crude oil is a yellow-

TABLE I

Extraction of Lespedeza Seed With Various Solvents

Solvent	Per cent oil extracted
Diethyl ether	11.7
Petroleum ether, 60-90°	11.7
Hexane	11.5
Tetrachloroethylene	11.1

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