as catalyst (Bespyatov, Trudy Khar'kov Politekh Inst. im V. I. Lenina, Ser. Khim. Tekhnol. 13, No. 4, 105-110 (1957). The negative catalysts present in rancid oils brought about a rapid decrease in the amount of Twitchell reagent during hydrolysis of the oils (Kanno, Kogyo Kagaku Zasshi, 64, 311-315 (1961).

VEGETABLE AND ANIMAL FATS AND OILS

Reviews were published on modern technology of fats and fatty products and high pressure method of oil extraction (Kaufmann and Grothues, Fette, Seifen, Anstrichmittel, 62, 1085-1090 (1960); Products from sunflower seed (Mizuno and Guerrero, Rev. Arg. de Grasas y Aceites, 3, 39-44 (1961); the relationship between hull content of sunflower seed kernel and oil losses during processing (Matsuk, Masloboino-Thiro-vaya Prom. 27, No. 1, 7-10 (1961); and castor oil processing and industrial utilization of by-products (Geleji, et al., Maguar Kem. Lapja, 15, 298-303 (1960). The industrial uses of castor oil were discussed (Iovehev, Drived (Setti) C. N. 6, 57, 50 (1057).

Priroda (Solia), 6, No. 6, 57-59 (1957). Recent inventions in the field of oils and fats were surveyed (*oils and oil-seeds J.*, (India) 9, No. 11, 10-12 (1957). Various aspects of palm oil industry were described (Raymond, Tropical Sci., 3, 69-89 (1961)

8, 69-89 (1901). Refining and bleaching in air decreased the stability of olive oil (Nosti and de la Borbolla, Grasas y Aceites 11, 139-150 (1960). Changes in the quality of cottonseed oil during storage were discussed (Sterlin and Burnasheva, Uzbek. Khim. Zhur. No. 3, 54-57 (1960).

Optimum conditions for processing cottonseed and soybean oils were suggested (de Castro and Ramos, Grasas y Aceites, 11, 97-101 (1960). The effect of the extent of milling of cottonseed meat on the oil content of the cake was investigated

(Glushenkova and Markman, Masloboino-Zhirovaya Prom. 27, Oil from fish, fish waste, and whale meat was recovered by

disintegrating and homogenizing the material for 5-6 hr followed by centrifugation (Ehlert and Mikkelson, Norw. 96,177). Inadequate filtration after rendering and contamination with iron and copper decreased the stability of lard (Vargas, et al., Grasas y Aceites, 11, 243-247 (1960).

BY-PRODUCTS

A semi-countercurrent process for extraction of woolwax used trichloroethylene as the solvent at a temperature of $45 \pm 2C$ (Kamada and Inone, Yukagaku, 5, 239-240 (1956). Waxy components were separated from sterols by extracting a mixture obtained from tall oil or crude "cane oil" with ethylene dichloride (Miller, et al., U.S. 3,004,992). Lecithin products were recovered from soapstocks by treatment with mineral acids at temperature and pressure insufficient to split or char the phosphatides (Thurman, U. S. 2,970,910). A patent was issued for the recovery of fatty material from

wash water obtained during refining of vegetable oil (Allen and Wack, U. S. 2,993,006). Lysine values for sesame protein from solvent extracted meal were much higher than those obtained for screw pressed meals (Contor et al. 1400CS 28, 148, 150, (1961)). meals (Carter, et al., JAOCS, 38, 148-150 (1961). A new process using acetone as solvent permitted the processing of cottonseed to produce a meat containing neither free nor bound gossypol (Vaccarino, JAOCS, 38, 143-147 (1961). Defatted soybean flakes were successfully debittered by countercurrent washing with aqueous ethyl or isopropyl alcohol. The entrained solvent was removed by flash distillation without excessive denaturation of protein (Mustakas, et al., JAOCS, 38, 473-478 (1961).

Mustard Seed Processing: Bland Protein Meal, Bland Oil, and Allyl Isothiocyanate as a By-product'

G. C. MUSTAKAS, L. D. KIRK, and E. L. GRIFFIN, JR., Northern Regional Research Laboratory,² Peoria, Illinois

Abstract

Mustard seed like rapeseed is characterized by the presence of glucosides, which are readily hydrolyzed under certain conditions by enzymes in the seeds to produce pungent "mustard oils." This property is utilized in the preparation of condiments from some varieties of mustard, but when the seed is processed to yield a palatable oil and feed meal, this pungent factor must be removed. This was accomplished in bench-scale studies at the Northern Laboratory by converting the mustard glucoside enzymatically and removing the converted product, allyl isothiocyanate, during a subsequent cooking step before filtration-extraction of the meal.

Introduction

THE AGRONOMIC POTENTIAL of oriental mustard seed 1 as an erucic acid-containing oilseed has been demonstrated in several parts of the world, e.g. India, Pakistan, Canada, Western Europe, Southern Italy, the Soviet Union and the United States. It is high in oil (42%) and in meal protein (46%). In this country, the seed is grown primarily for condiment purposes. Workers at this laboratory engaged in the screening of new crops for commercial development

in the United States have reported favorably on the utilization aspects and potential of oriental mustard seed (9). Nutritional values for mustard meals were recently reported as quite promising by K. J. Goering et al. (6). Opportunities for mustard seed as an oilmeal crop could develop in areas that produce a wheat surplus and particularly in areas that are rather far removed from soybean production and are in need of high-protein animal feeds. Possible uses of erucic acid oils are in industrial lubricants and as sources of the lower molecular weight cleavage products, brassylic and pelargonic acid.

Because oriental mustard seed is grown principally for a condiment, little information has been published dealing with the economic processing of the seed into oil and meal products. Also, little information is available on methods for removal of the glucosides that impart pungent factors to the oil and meal fractions. The presence of the essential oil presently limits the proportion that can be used in cattle rations. K. J. Goering has patented a process (5) for obtaining a bland feed material from mustard seed. By his method, mustard seed is first processed to separate the lipid from the glucoside-containingmeal fraction. The meal is then treated in a series of additional steps to remove the essential oil, allyl isothiocyanate. The present paper discloses a simplified process in which the glucosides are removed before the lipids are separated. This procedure eliminates many operating steps. The new process devi-

¹ Presented at the Annual Spring Meeting of the American Oil Chem-ists' Society, May 1-3, 1961, St. Louis, Mo. ² A laboratory of the Northern Utilization Research and Development Division, Agricultural Research Service, USDA.

ates little from commercial processing of soybeans and cottonseed and little or no difficulty should be anticipated in its commercial application.

Principles of Enzyme Hydrolysis

Successful processing of mustard seed involves removal of the glucoside in addition to the normal conversion of the seed into nonvolatile lipids and defatted oil meal. The pungent factor, allyl isothiocyanate, occurs naturally in mustard seed as the water-soluble thioglucoside, sinigrin. The isothiocyanate in the presence of moisture and heat is released from the glucoside by the enzyme "myrosinase" which occurs naturally in the seed. In the hydrolysis reaction developed for sinigrin by Ettlinger et al. (3), the sugar moiety of the thioglucoside is glucose and the aglycone fraction is allyl isothiocyanate:

$$CH_{2} = CH - CH_{2} - C$$

$$enzyme + H_{2}O$$

$$C_{6}H_{12}O_{6} + KHSO_{4} + CH_{2} = CHCH_{2}N = C = S$$

Conditions of the enzyme reaction are favorable for integration into the filtration-extraction process, a relatively new extraction method developed by the Southern Utilization Research and Development Division, which is now used commercially to recover oil from soybeans, cottonseed, rice bran, sunflower seed, and more recently from rapeseed and flaxseed.

Material, Methods, and Equipment

Oriental mustard seed, Brassica juncea, was furnished by Montana State College. It had been harvested in Montana during the 1959 crop year. Weight per thousand seeds averaged 3.4g. Moisture content varied between 4 and 6%. Analyses of seed showed 42% oil, 26.5% protein, and approximately 15% hull content. Glucoside content, expressed as the converted product, allyl isothiocyanate, was 0.7-0.8%mfb. Commercial grade n-hexane was the solvent for the filtration-extraction studies.

Allyl isothiocyanate was determined in the various process streams by Wetter's procedure (8), a modified argentimetric method. Purity of the essential oil was determined by a modification of this procedure in which an aliquot of the oil in ethanol was added directly to the ammoniacal silver nitrate solution. The wet-screen analysis followed was that described by the Southern Utilization Research and Development Division (4). Crude fat was determined by extraction with pentane-hexane in a Bütt extraction apparatus for 6 hr and drying overnight in a vacuum oven at 80C.

Comminution of the mustard seed was carried out in the following equipment: 6 in. diameter rolls with 12 corrugations per inch; 6 in. diameter rolls with 16 corrugations per inch; and 12 in. diameter smooth rolls.

Converting and cooking steps were conducted in a cylindrical vessel (Figure 1) which held up to 550g of material. Upper and lower electric heating mantles were used to control heat input. Mixing was provided by a multiple rod agitator revolving at 9 rev/min. Intermeshing rods were fixed to the top of the cooker for an efficient baffle system. A spray

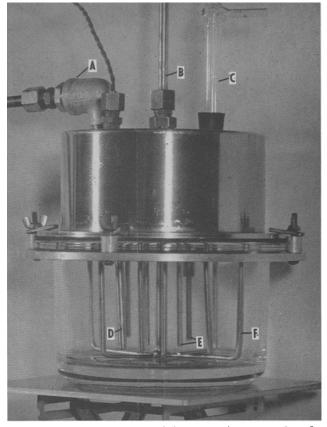


FIG. 1. Converter-cooker vessel for processing mustard seed. Key: (A) Vapor outlet to condenser. (B) Agitator shaft. (C) Glass spray unit. (D) Thermocouple well. (E) Stationary rod baffles (6) ($\frac{3}{16}$ -in. diameter). (F) Multiple rod agitator ($\frac{3}{16}$ -in. rod diameter).

nozzle for introducing moisture, a vapor opening, and a thermocouple were installed in the top head of the vessel. For distillation of volatile oil, the top vapor opening was connected to a water-cooled condenser and receiver apparatus.

Bench-scale apparatus, similar to that described by A. V. Graci et al. (7) was used for evaluating the characteristics of mustard seed for filtration-extraction. In our experiments, the filtration unit was modified to use a $1\frac{3}{4}$ in. diameter funnel.

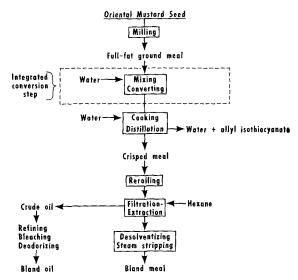


FIG. 2. Flowsheet of simplified mustard seed process integrated with filtration-extraction.

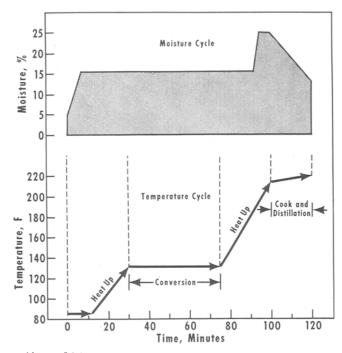


FIG. 3. Moisture and temperature variation of the mustard meal during conversion and cook steps.

Procedure

A flowsheet of the integrated process is shown in Figure 2.

Mustard seed was cracked in 2 passes, through rolls with 12 corrugations per in. set at a clearance of 0.008 in. for the first pass and through rolls with 16 corrugations per in. set at a clearance of 0.003 in. for the second pass. The product was then rerolled in three passes through smooth rolls to obtain a finely ground meal. This full-fat ground meal was then charged to the cooker and moistened by a fine spray at room temperature to 15--16%. A slow-moving agitator distributed the moisture evenly throughout the meal. Moisture and temperature profile curves of the meal during moistening, converting, and cooking are shown in Figure 3. After a mixing period of approximately 5 min, the wetted meal was heated to a temperature of 131F, and conversion was carried out with agitation for 15-45 min at constant temperature. The converted meal was then heated to 185F where additional moisture was introduced by a fine spray to raise the moisture content to 25%. Heating was continued to 212F where cooking and distilling of moisture and volatile oil were carried out simultaneously. During the 20 min cook cycle, the temperature increased from 212 to 220F, and the moisture was reduced from 25% to approximately 13%. Allyl isothiocyanate and moisture distilled out of the meal, condensed, and separated into two liquid phases. The hot meal was removed from the cooker, passed through a 5-mesh sieve to break up small balls, allowed to air cool and crisp, and then rerolled through smooth rolls to comminute the material for subsequent extraction.

Filtration-extraction of the rerolled material was carried out according to Graci's procedure (7) to determine extraction efficiency, as well as to evaluate other critical processing variables. Conditions suitable for extraction were: slurry for 60 min, solvent ratio 1.3-1, 3 washes, cake thickness of 2 in., and temperature extraction of about 140F. Slurry mis-

cella contained 10% oil in hexane, and washes contained 5, 1, and 0% oil in hexane.

Spent meal from the extraction was air-desolventized and then stripped with open steam on a 60-mesh screen Büchner funnel for 30 min to remove the last traces of volatile oil in the meal.

A new technique of double-soak filtration-extraction as described by E. L. D'Aquin et al. (2) was also evaluated for mustard seed. This process is very similar to the procedure originally described by A. V. Graci et al. (7), except that it requires an additional soaking step followed by refiltration. Miscella in the first slurry contained 10% oil in hexane; after filtration the meal was washed with a 5%-oil miscella. For the second slurry step, a 1.5%-oil miscella was used, and after filtration the meal was washed successively with 0.5%-oil miscella and pure hexane.

Process Variables

Meal Preparation. Size distribution of the whole mustard seed was as follows:

Screen size		Weight percent retained		
No.	Opening, in.	Individual	Cumulative	
10	0.0787	0	0	
12	0.0650	9	9	
14	0.0555	25	34	
16	0.0460	61	95	
20	0.0331	5	100	

Studies were carried out to determine the feasibility of separating the hulls. A hull fraction could be separated by tempering, cracking in three stages, and screening with aspiration. This fraction however was high in lipid and protein, 15.5% and 22.4%, respectively. The hull-separated fraction represented about 15-20% of the seed weight, so that dehulling would probably not be economically attractive. Dehulling increased protein content of the defatted meal by about 5%. No dehulling was carried out on the experiments reported in this paper.

Comminution studies were conducted to determine optimum preparation of the solids for solvent extraction. Because a finely ground feed resulted in low residual lipids, early experimental work was directed at fine grinding. Grinding to obtain over 50% of -300 mesh material was facilitated by predrying of the seed to 1-3% moisture. Our succeeding work however showed that rerolling of the crisped meals before extraction compensated for the initial requirement of a fine grind to give low residual lipids in the extracted meal. Rerolling also eliminated the need to predry the seed and was adopted as an improvement in the process. Without rerolling the air-dried marc, the residual lipid was roughly proportional to the fineness of initial grind (Table I). Particlesize distribution of mustard meal between processing steps is shown graphically in Figure 4.

TABLE I Effect of Size Reduction and Rerolling on Residual Lipids in Extracted Mustard Meal

Grind specification of starting material Fraction through 300 mesh, ^b %	Rerolling after crisping ^a	Residual lipid in extracted meal, %	
12.0	No	6.56	
50.3	No	4.45	
60.4 °	No	2.50	
19.2	Yes	0.94	
66.3	Yes	0.78	

^a Rerolled in 2 passes through smooth rolls.
^b Wet-sieve analysis with hexane as the immersion liquid.
^c Seed predried before comminution.

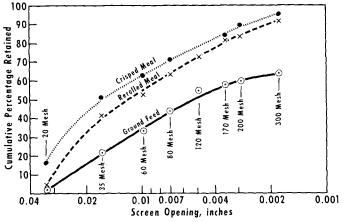
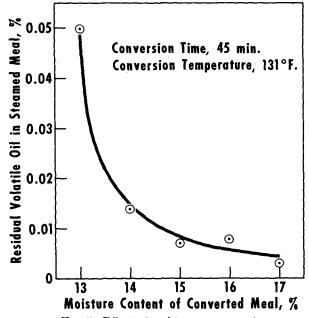


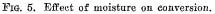
Fig. 4. Particle-size distribution of mustard meal after milling, cooking, and rerolling.

Thioglucoside Conversion. Optimum conversion of the thioglucoside depends on moisture content, temperature, and retention time. The enzyme was furnished by the myrosinase naturally occurring in the original seed. The curve in Figure 5 shows the effect of moisture level during conversion on volatile oil removed in the detoxification process. Conversion conditions for these studies were 131F, and a 45 min retention period. At 15–16% moisture, the solids remained granular and were easy to handle.

Moisture levels above 16% became undesirable when a finely ground meal was prepared from the seed. During the conversion period the high moisture meals experienced the "Skippin" effect (1) whereby oil phase separation or oil release occurred. Solid material became pasty and apparently sufficient paste grinding of the meal occurred so that the mass filtration rates in the subsequent extraction step were lowered to inoperable values.

The effect of temperature is shown in Figure 6, where conversion was carried out at 15.5% moisture for 45 min. Good conversion was obtained over the temperature range of 115–150F. The data in Figure 7 indicate that conversion time can be reduced to 15 min when at the 15.5% moisture level at 131F. The





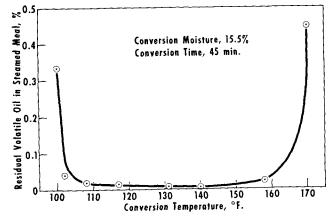


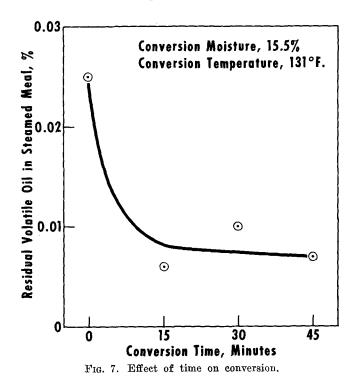
FIG. 6. Effect of temperature on conversion.

longer interval of 45 min was used in most of the experimental work, however, in order to insure good conversion.

Cooking. During the cooking operation, protein was coagulated and coalescing of the oil phase occurred. Under the conditions of heat and moisture, oil permeability through the cell walls was greatly improved, so that oil diffused readily into the solvent phase during the soaping step before filtration-extraction. In addition, suitable conditions were provided for distillation of the volatile oil simultaneously with water removal.

Secondary moisture addition on the heat-up cycle at 185F prior to the cook step was used to improve volatile oil separation. This step provided additional moisture for the distillation and permitted the volatile oil to be separated at lower meal temperatures. Without the additional moisture, meal would need to be heated to 240–260F to achieve the same degree of volatile oil removal.

The hot material after air cooling formed a crisp, porous solid, which was relatively granular and incompressible and which possessed porosity characteristics favorable for rapid filtration. When the crisped



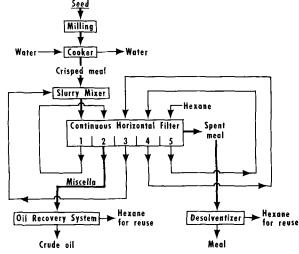


FIG. 8. The filtration-extraction process developed by the Southern Utilization Research and Development Division.

solids were rerolled for filtration-extraction, a lowmoisture level contributed to excessive comminution with production of fines and low mass-filtration rates. Moisture content was reduced by roughly 12% during cooking. Thus, if additional moisture were not added, the crisped meal would be unduly dried. However with the secondary moisture addition, the hot cooked meal contained about 13% moisture (air cooling will reduce the moisture an additional 3-4%) so that rerolling could be controlled to give fewer fines and adequate filtration rates in the subsequent extraction step.

Volatile oil and moisture were condensed and collected as two liquid phases; the essential oil was the lower phase. Because the specific gravities of allyl isothiocyanate and water are close, the phase separation was not always clean, and it was further hampered by surface tension effects. Two techniques were used in our laboratory studies for recovery of the volatile oil from the two-phase system. The simplest procedure consisted of saturating the water phase with calcium chloride after which separation of the two liquid phases in a separatory funnel was direct and complete. In the alternative procedure, haxane was used to dissolve the volatile oil. The hexanevolatile oil phase is then decanted and the hexane evaporated off. The calcium chloride method was used in these studies because of simpler operation and better yields of volatile oil.

Filtration-Extraction. Extraction variables were studied on the basis of determining the optimum conditions with respect to extraction efficiency and mass filtration rates. Extrapolation of the benchscale data indicates that large-scale filter operations in a commercial unit will be feasible. A flowsheet of the filtration-extraction commercial process is shown in Figure 8. On the basis of previous investigations conducted with other oilseeds by the

TABLE II Optimum Conditions for Filtration-Extraction of Mustard Seed

	· · · · · · · · · · · · · · · · · · ·
Rerolling	Crisped meal rerolled through
	smooth rolls
Particle size after rerolling	
	mass velocities below 2,000 lb/hr/ft ²
Solvent ratio	
Slurry time	60 min
Temperature of hexane	140F
Vacuum	4 inch Hg
Cake thickness	2 inch
Mass velocity during filtration	2.000-4.000 lb/hr/ft ²

Southern Utilization Research and Development Division, mass filtration rates of 2,000-4,000 lb/hr ft² were deemed suitable for operation of a commercial horizontal rotary filter.

As shown in Table I, and discussed previously under meal preparation, rerolling was successful in obtaining a good extraction efficiency even when a relatively coarse grind was used for the preliminary steps of conversion and cooking.

Solvent ratios in the range of 1.1–2.0 were tested. Equally good extraction efficiencies were obtained throughout this range. Although the 1.1 ratio gave efficient extraction, the quantity of free liquid available for slurrying at this ratio was quite small.

The effect of slurry time upon lipid removal and filtration rate was investigated briefly. The results shown below indicate that lower residual lipid content in the extracted meal and essentially no change in mass filtration rate occurred with a 60-min slurrying period:

Slurry	Residual	Mass filtra-
time,	lipid,	tion rate,
min	% mfb	lb/hr/ft ²
30 60	$\begin{array}{c} 2.31 \\ 1.62 \end{array}$	2140 2170

Cake thickness was studied between 1.0 and 2.5 in.; although poor consistency was obtained in the data, it is roughly estimated that a cake thickness of 2 in. will give good filtration characteristics with high extraction efficiencies.

Optimum conditions, found in these laboratory tests, for the filtration-extraction of mustard seed are given in Table II. The double-soak extraction technique yielded meals with slightly lower lipid content than did the conventional single soak. Approximately 1% residual fat in the extracted meal was achieved with single-soak extraction; whereas values of approximately 0.5% were obtained by a double soak.

Integrated Runs. Table III presents data that demonstrate the effect of process variables. The effect of comminution upon extraction efficiency is shown in Runs 1A, 2A, and 3. These represent both coarse and fine initial grinds. To facilitate fine grinding in Run 3, the seed was oven-dried before grinding. Extraction efficiency was increased significantly by rerolling the crisp meal prior to extraction as demonstrated in Runs 1B, 2A, and 4A.

Poor thioglucoside conversion was obtained in Run 6 due to insufficient moisture (13%). Conversion time was reduced to 15 min for Run 5, and an excellent conversion was obtained at high lipid-extraction efficiency.

The effect of extraction variables upon residual fat content is shown in Runs 2, 4, and 7. Slightly better extraction is demonstrated by a double-soak extraction over the single-soak procedure. Solvent ratios of 1.3 and 1.5 (Runs 7A and 7B) gave the same extraction efficiency.

Evaluation of Products

Mustard Oil Meal. Bland meals after desolventization and steaming contained less than 0.01% residual allyl isothiocyanate, approximately 1% lipid, and approximately 46% protein. Although tolerance limits for allyl isothiocyanate in defatted mustard meals have not been established, the extent of removal of this factor in the present studies is believed adequate. Preliminary analyses for lysine indicate that this

TABLE III						
Bench-Scale	Filtration-Extraction Oil, Meal, and			or Production	of	

Run No.	1	2	3	4	5	6	7
Seed preparation							
Predrying	No	No	Yes	No	No	No	No
Fraction through 300 mesh, %	12.0ª	50.9	60.4	62.5	53.1	61.6	19.2
Conversion-cook conditions							
Moisture content, %)			1	
Feed	4.3	4.3	3.6	4.3	4.3	4.8	6.0
Conversion	15.5	15.5	15.5	15.5	15.5	13.0	15.5
Maximum in cook	25.0	25.0	15.5	25.0	25.0	25.0	25.0
Cooker discharge	13.2	13.1	2.0	13.4	13.4	13.6	12.4
After rerolling	9.0	10.0	•••••	9.5	9.0	9.9	8.7
Temperature, F			1	į ·	i I		1
Moisture addition	75	75	75	75	75	75	75
Conversion	131	131	122	131	131	131	131
Second moisture added	185	185		185	185	185	185
Distribution range	216	215	226	213	215	213	215
	221	220	260	219	221	221	220
Time, minutes						1	
Moisture mixing	5	5	30	5	5	5	5
Conversion	45	45	45	45	15	45	45
Total for conversion cook	126	122	100	135	90	125	120
	A B		1				
Reroll	No Yes	No	No	Yes	Yes	Yes	Yes
Clearance	0.003			0.003	0.003	0.003	0.003
	contact			contact	contact	contact	contact
Extraction conditions				·			
	A B	A B		A B			A B
Cake thickness, inch	2 2	1.75 1.75	1.5	2 2	2	1.75	2 2
Slurry time, min	60 60	60 60	6 0	60 60	60	6 0 .	60 60
Slurry temp., F	140 - 140	140 140	140	140 140	140	140	140 140
Vacuum, inch Hg	4 4	4 4	4	4 4	4	4	4 4
Hexane/meats ratio	1.3 1.3	1.3 1.3	2.0	1.3 1.3	1.3	1.3	1.3 1.5
Soak method (single/double)	sing sing	sing dbl	sing	sing dbl	sing	sing	sing sing
	A B	AB	1	A B	([A B
Results							
Spent meal crude fat, % mfb	6.56 1.58	4.93 3.60	2.50	0.98 0.45	1.20	1.20	0.94 0
		3600		1720			
Mass velocity lb/hr/ft ²	4610 2040	5450 4100	4820	1880 2950	2720	2420	2330 3060
Extraction efficiency, % ^b	90.3 97.7	92.8 94.8	96.4	98.6 99.3	98.3	98.3	98.6 98.6
Extraction efficiency, % ^b Allyl isothiocyanate, % mfb ^c							
Urisp meal	0.018 0.018	0.081 0.081	0.019	0.017 0.017	0.026	0.101	0.052 0.052
Spent meal Steamed meal	0.014 0.013	0.030 0.029	0.021	0.016 0.023	0.020	0.132	0.022 0.019
Steamed meal	0.014 0.010	10.005	0.002	0.010 0.007	0.003	0.050	0.004 0.002

^a To prepare this material, seed was tempered to 8% moisture content and flaked in 3 passes through smooth rolls.

Extraction officiency =
$$\frac{100}{100} \left[\frac{A-R}{A-R} \right] \times 100 = \frac{100}{100} \left[\frac{42-0.99}{42-0.99} \right] \times 100 = 98.6\%$$

^b Extraction efficiency = $\frac{1}{A} \left[\frac{100 - R}{100 - R} \right] \times 100 = \frac{100 - 100}{42} \left[\frac{100 - 0.99}{100 - 0.99} \right] \times 100 = 98.6\%$

where A = % crude fat of whole mustard (mfb) and R = % residual fat after extraction (mfb).

^c Full-fat meal = 0.734% allyl isothiocyanate, mfb.

meal is subject to some loss of nutritional quality due to heat damage, but this loss occurs with many other oilseed cake meals. Certain modifications of this process that produce a meal of higher nutritional quality are presently under investigation and will be reported in a subsequent paper.

Mustard Oil. Crude nonvolatile oils recovered in the process contained 0.01 to 0.04% allyl isothiocyanate. Volatile oil can be completely removed by the usual refining, bleaching, and deodorizing. Organoleptic evaluation of the deodorized mustard oil compared with a commercial soybean oil for control gave the following results:

Oil	Flavor score a		
011	Initial	4-day storage b	
Mustard Soybean	7.6 7.5	5.3 6.2	

^a Range = 1-10 units with 10 rated best. ^b 60C in presence of air.

This test shows that mustard seed oil is comparable to soybean oil when freshly prepared but develops a lower flavor score after 4 days' aging at 60C. This response might be expected since mustard seed oil is slightly higher in linolenic acid. Additional physical and chemical properties are given below for a refined, bleached, and deodorized mustard oil: density 0.9467 g/cc (27.2C); refractive index 1.4709 (25C); melting point -13.0 color, Gardner scale (2); color, photometric method 2.44; I.V. 118.0; saponification value 186.4; peroxide value 0.59; acid value 0.07; unsaponifiable 1.04%.

Volatile Oil. The crude volatile oil was a clear, light yellow material which analyzed 88-93% allyl isothiocyanate. The liquid measured 1.0108 g/cc (20C) in density and 1.525 (25C) in refractive index. For many applications the volatile oil product should be usable without further purification.

Acknowledgments

We acknowledge the assistance of Lynn T. Black, and J. E. McGhee for analytical determinations; the advice of C. H. VanEtten and M. E. Daxenbichler on analytical methods; C. R. Evans, H. A. Moser, and the laboratory taste panel for organoleptic evaluations; R. L. Reichert for construction of the benchscale cooker; E. A. Gastrock of the Southern Utilization Research and Development Division, New Orleans, La., for helpful advice on filtration-extraction; and Dr. K. J. Goering and Montana State College for furnishing the oriental mustard seed.

REFERENCES

- 1. Bailey, A. E., "Cottonseed and Cottonseed Products," Interscience Publishers, Inc., New York, 1948, p. 625. 2. D'Aquin, E. L., A. V. Graci, Jr., H. L. E. Vix, and E. A. Gas-trock, JAOCS, 38, 258 (1961).
- 8. Ettlinger, M. G., and A. J. Lundeen, J. Am. Chem. Soc., 78, 4172 (1956).
- Gardner, H. K., E. L. D'Aquin, J. S. Parker, and E. A. Gastrock, Ind. Eng. Chem., 44, 2263 (1952).
 Goering, K. J. (Oilseed Products, Inc.)., U. S. 2,987,399 (1961).
- 6. Goering, K. J., O. O. Thomas, D. R. Beardsley, and W. A. Curran, Jr., J. Nutrition, 72, 210 (1960).
- 7. Graci, A. V., Jr., J. J. Spadaro, M. L. Paredes, E. L. D'Aquin, and H. L. E. Vix, JAOCS, 32, 109 (1955).
 - 8. Wetter, L.R., Can. J. Biochem. and Physiol., 33, 980 (1955). 9. Wolff, I. A., and Q. Jones, Chemurgic Dig., 19, 4, July 1960.