fatty acid composition of fats containing higher unsaturated acids.

Confirmatory evidence has been obtained for the presence of eicosenoic acid in rapeseed oil.

The nature and amount of fatty acids of yellow mustard seed oil of Indian origin do not differ in any significant manner from those of other cruciferous seed oils.

The present analysis of black mustard seed oil reveals a higher amount of linolenic acid, and the presence of a C_{20} monoethenoid acid, not heretofore reported.

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Stabilization of Vitamin A in Halibut Liver Oil With Nordihydroguaiaretic Acid (NDGA)[1]

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THE importance of adequate vitamin A in the diet is generally recognized. Certain fish liver oils, being especially rich in vitamin A, have been used for many years as a dietary supplement in American households. Halibut liver oil is one of the vitamin A oils in popular use today, but, in common with other fish liver oils, it is a relatively unsaturated oil and is susceptible to rancidification if exposed to air. It has been shown by Hilditch (1) and also by this study that the oxidation of the fresh fish liver oil to a rancid oil is accompanied by destruction of a large share of the vitamin A content. Exposure to light and warm temperatures accelerates the rate at which the freshness of the oil is lost. Most purified fish liver oils or concentrates tend to become rancid if the oil is dispensed from an open container and stored at room temperature.

To ensure the maximum freshness of the oil at the time of purchase, special precautions are taken in the commercial production, purification, and packaging of the vitamin A oil to minimize exposure to oxidizing conditions. Natural antioxidants present in the oil are important in the subsequent preservation of the freshness of the oil during the consumption period. Further stabilization of the oil may be obtained by the addition of more effective antioxidants.

Since purification and concentration of the vitamin A oil by alkali processing tends to remove natural antioxidants, the addition of antioxidant to either the purified oil or to the concentrate is highly desirable if they are to be dispensed from an open container. A good antioxidant for use in food products should be non-toxic, highly effective in low concentrations, moderately priced, and easily soluble in the product. It should produce no adverse change in the color, taste, or odor of the product. These experiments determined the effectiveness of the chemical, nordihydroguaiaretic acid (NDGA), as an antioxidant in halibut liver oil.

NDGA was developed as an antioxidant at the University of Minnesota in 1944 (2). It is produced as a pure crystalline powder from a common desert plant, one of the creosote bushes, which otherwise is quite useless.

NDGA is one of the group of polyphenolic compounds, characterized by either the ortho- or para-oxygen linkage, which are known to possess primary antioxygenic properties. According to Mattill (3), all other substances which inhibit the autoxidation of fats are properly called synergists because they merely reinforce the effect of the primary phenolic antioxidant present.

NDGA has been shown to be non-toxic in low concentrations and is soluble in hot oils at 75-125°C., ethyl alcohol, propylene glycol, and other organic solvents (2, 4). It may be incorporated easily in any fatty product by dissolving the proper amount of antioxidant in a carrier solvent, which is then mixed in with the bulk of the product. Bucher (5) noted that a 0.1-0.2% suspension of NDGA in salmon oil proved effective in retarding the development of rancidity. A report by Silver (6) showed that application of 0.2% NDGA in a coating of vegetable oil would extend the storage life of brined mackerel from 10 days for the control to 4 months for treated fish. Higgins and Black (7) found that high storage temperatures decreased the stabilizing effect of NDGA in lard.

Experimental

Crude halibut liver oil was obtained from a local extraction plant for the stability tests. The samples were blended from several lots at that plant, and the peroxide values and free fatty acid content were uniformly low. The standard Swift stability method as modified by Riemenschneider (8) was used for all tests at 97.7° C. and 80° C. The antioxidant was dissolved in ethyl alcohol, and one milliliter of the solvent containing the proper concentration for 20 grams of oil was added to the oil at the beginning of the test. The solvent evaporated easily at the temperature of the oil and was carried out of the oxidation tube in a few minutes by the current of air.

Approximately 0.25-gram samples of oil were removed at regular intervals from each tube for the determination of peroxide values and spectrophotometric determination of vitamin A. Duplicate tubes

[1] Presented at the Portland, Oregon, meeting of American Chemical Society, September 1948.

TABLE I

Effect of Various Concentrations of NDGA on Peroxide Formation in Halibut Liver Oil

Concentration of Antioxidant		Oxidation Time –Minutes at 97.7°C.							
		100′	200′	300′	400′	500′	600′	700′	800′
% NDGA	% Synergist[1]	Peroxide Value– –ME[2] of $Na_2S_2O_3$ per kg. oil							
0 (Control)	0	16	34	51	66	78	93	109	....
0.1	0	18	36	54	72	85	....	....	....
0.3	0	13	33	44	56	65	72	79	....
0.5	0	13	28	40	49	60	70	79	87
0.01	0.01 CA	24	36	47	58	69	78	84	90
0.1	0.1 CA	12	25	37	50	62	75	87	....
0.1	0.1 AA	18	33	45	54	63	72	82	91
0.3	0.15 CA	10	16	22	27	33	42	53	62
0	0.1 CA	16	34	51	66	78	....	....	....
0	0.3 AA	18	35	45	54	62	71	....	....

[1] CA = citric acid; AA = ascorbic acid.
[2] ME = milliequivalents.

were checked in most cases. Peroxide values were determined on 0.2 gram of oil by the method adapted by Riemenschneider (8). For determination of vitamin A approximately 50 milligrams of oil were weighed by use of a tared weighing wire, dissolved in a small amount of petroleum ether, and the solution was made up to volume with redistilled isopropanol. The optical density was determined on a dilution giving a reading between 0.4 to 0.8 at 328 millimicrons with a Beckman Spectrophotometer. The ratio of the $E^{1\%}_{1\,cm.}$, 328 value of the oxidized oil to that of the oil at the beginning of the test indicated the percentage of original vitamin A remaining in the sample.

An arbitrary criterion of 50% of the original vitamin A content of the oil has been used by Sanford, Harrison, and Stansby (8) in comparing the relative stability of grayfish liver oils. The protective factors in this study of the effect of antioxidant concentrations in halibut liver oil have been computed on the basis both of 20% and 50% loss of original vitamin A (Table II). The protective factor is defined as the ratio of the oxidation time for the loss in the control oil to that for the same percentage loss in the treated oil.

For the accelerated stability tests at 25° C., 0.2-gram samples of oil with proper concentration of antioxidant were measured into a number of uniform five-milliliter beakers. These were placed in a thermostatically controlled incubator at 25°C. At intervals, one or two of the beakers were removed for determination of the vitamin A.

Results

Data were obtained on the effect of various concentrations of NDGA with and without synergists on peroxide formation and on loss of vitamin A in the oil (Table I and Figure 1). Antioxidant concentrations from 0.01% to 0.5% were used; citric acid, ascorbic acid, primary calcium phosphate, and orthophosphoric acid were used as synergists. The effect of synergists alone was also determined. Limited data were obtained at 80°C. and 25°C. (Figures 6 and 7).

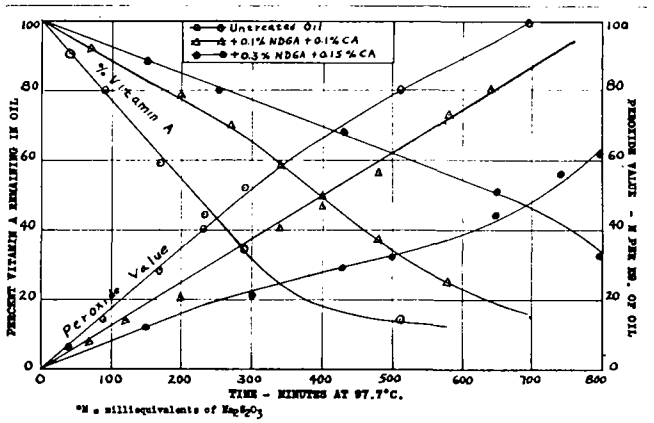

FIG. 1. Vitamin A destruction and peroxide formation in untreated and NDGA treated halibut liver oil at 97.7°C.

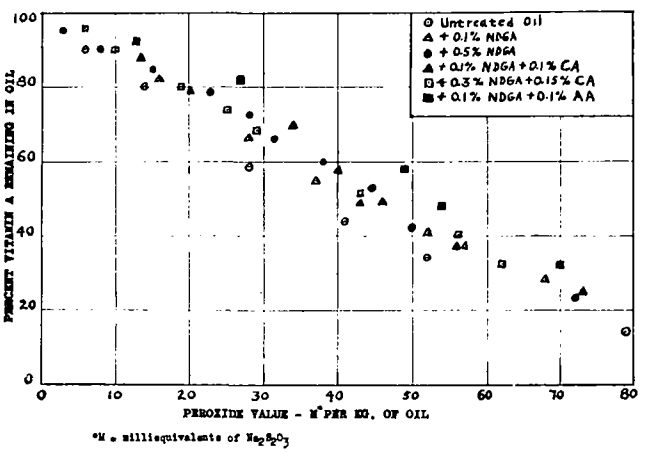

FIG. 2. Relation between vitamin A loss and peroxide formation in halibut liver oil at 97.7°C.

At the standard temperature of the Swift aeration (97.7°C.) there was no true induction period for either formation of peroxides or loss of vitamin A (Figure 1). The most important effect of the antioxidant was the decreased slope of the curve for both formation of peroxides and the loss of vitamin A. This resulted in almost a straight line relationship between peroxide values and per cent of original vitamin A in the oil (Figure 2). Stabilization of the oil with 0.3% NDGA and 0.15% citric acid effectively reduced the loss of vitamin A, but rancidity of the oil still developed although an appreciable percentage of the original vitamin A remained (Figure 1). Judged organoleptically, extreme rancidity occurred at a peroxide level of 50 to 60 in most cases.

Concentrations of NDGA of less than 0.10% had no significant stabilizing effect on the oil at 98°C. unless a synergist was present. Concentrations of NDGA up to 0.5% with no synergist present resulted in moderate stabilization of the vitamin A but not in the degree that one might expect considering the ratio of the concentrations (Figure 3). If the point at which 50% of the vitamin A was destroyed is assumed as a basis for computing the protective factor, a concentration of 0.5% NDGA resulted in a protective factor slightly less than 2 (Table II).

Use of synergists improved the efficiency of the NDGA considerably (Figure 4). A concentration of 0.1% NDGA with 0.1% citric acid was as effective as 0.5% NDGA with no synergist. Orthophosphoric acid was superior to citric acid as a synergist, and 0.1% NDGA plus 0.1% phosphoric acid was 50% more effective than 0.1% NDGA plus 0.1% citric acid in the stabilization of the vitamin A if computed for periods at which 50% loss of vitamin A had occurred. Undesirable darkening of the oil did result with the use of phosphoric acid; however, later experiments at 25°C. indicated that this was not significant at the lower temperature. Ascorbic acid was as effective

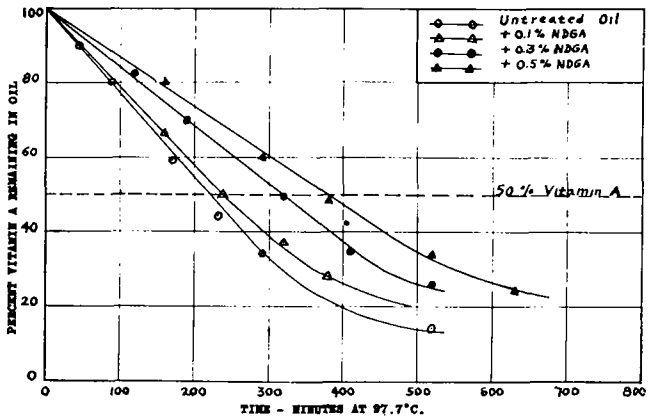

FIG. 3. Vitamin A destruction in halibut liver oil with various concentrations of NDGA.

as a synergist as citric acid. Tests with 0.3% ascorbic acid also showed that it was as effective as 0.3% NDGA in stabilization of the oil. Citric acid was slightly effective at the 0.1% level (Table 1). Presumably both citric and ascorbic acid may act as synergists with the natural antioxidant present in the crude halibut liver oil as used in these tests. In a purified oil or a concentrate where the natural antioxidant of the vitamin A oil may be reduced or removed, an antioxidant of the polyphenolic type such as NDGA would have to be added to utilize the synergistic effect of the acids. Primary calcium phosphate was added as a synergist, but in the tests at 97.7°C. there was little effect (Figure 4).

Two tests at 97.7° C. with 0.1% NDGA and 0.1% phosphoric acid indicated that an efficient antioxidant could still be used to stabilize the remaining vitamin A content of a partially oxidized oil (Figure 5). Addition of the antioxidant after 30% of vitamin A in one case and 62% in the second had been destroyed showed that the antioxidant was still effective in protecting the remaining vitamin A.

One series of tests using the Swift oxidation procedure at 80°C. indicated the possible importance of specifying the temperature at which the protective factors have been evaluated (Figure 6). At 80°C. the protective factor for 0.1% NDGA with no added synergist was equal to or slightly higher than that at 97.7°C. In the case of 0.1% NDGA with 0.1% citric acid the protective factor at the 50% vitamin A level was 3.8 at 80°C. as compared to 1.9 at 97.7°C. Another significant difference was the appearance of an induction period. If the protective factor is computed after 20% loss of vitamin A, it is 2.4 for 0.1% NDGA with 0.1% citric acid at 97.7°C. and 5.7% at 80°C. (Table II).

TABLE II

Protective Factors for Various Concentrations of NDGA in Halibut Liver Oil

Antioxidant Concentration % by Weight		Oxidation Temperature	For 20% Loss Vitamin A		For 50% Loss Vitamin A	
NDGA	Synergist[3]		P.F.[1]	P.V.[2]	P.F.[1]	P.V.[2]
		C.				
0 (Control)	0	97.7	1.0	16	1.0	40
0.10	0	97.7	1.0	16	1.1	42
0.30	0	97.7	1.6	21	1.5	45
0.50	0	97.7	1.9	21	1.7	46
0.01	0.01 CA	97.7	1.2	23	1.2	42
0.02	0.05 PA	97.7	1.4	....	1.9	....
0.10	0.10 CA	97.7	2.4	15	1.9	46
0.10	0.10 AA	97.7	2.3	30	1.9	55
0.10	0.10 PA	97.7	2.0	....	2.9	....
0.10	0.10 CP	97.7	1.3	....	1.3	....
0.30	0.15 CA	97.7	3.2	16	3.1	42
0.30	0.10 PA	97.7	2.9	....	3.9	....
0	0.10 CA	97.7	1.2	....	1.3	....
0	0.10 PA	97.7	1.7	....	2.1	....
0	0.30 AA	97.7	1.5	22	1.4	46
0 (Control)	0	80	1.0	....	1.0	....
0.10	0	80	1.6	....	1.3	....
0.10	0.10 CA	80	5.7	....	3.8	....
0 (Control)	0	25	1.0	....	1.0	....
0.10	0	25	8.8	....	7.5	....
0.10	0.10 CA	25	>28.0	....	>28.0	....
0.10	0.10 CP	25	18.0	....	14.0	....
0.30	0.15 PA	25	>60.0	....	>60.0	....

[1] P.F. = $\frac{\text{oxidation time for control oil}}{\text{oxidation time for treated oil}}$

[2] P.V. = milliequivalents of $Na_2S_2O_3$ per kg. oil.

[3] CA = citric acid; PA = orthophosphoric acid; AA = ascorbic acid; CP = primary calcium phosphate.

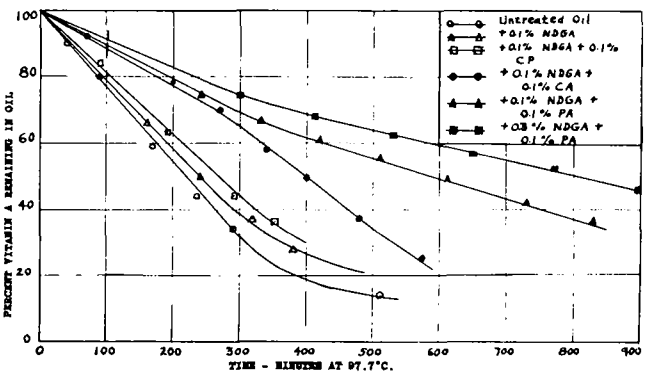

FIG. 4. Vitamin A destruction in halibut liver oil treated with NDGA and various synergists.

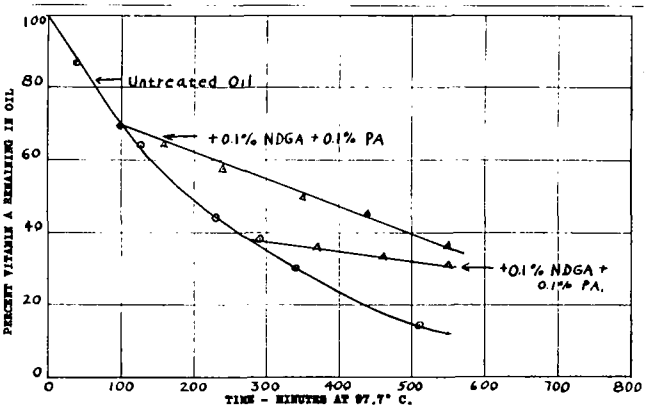

FIG. 5. Effect of addition of NDGA to partially oxidized halibut liver oil on vitamin A destruction.

The series of tests at 25°C. appeared to substantiate the results of the tests at 80°C. Protective factors were increased markedly as compared to those at 97.7°C. (Table II) and an induction period was

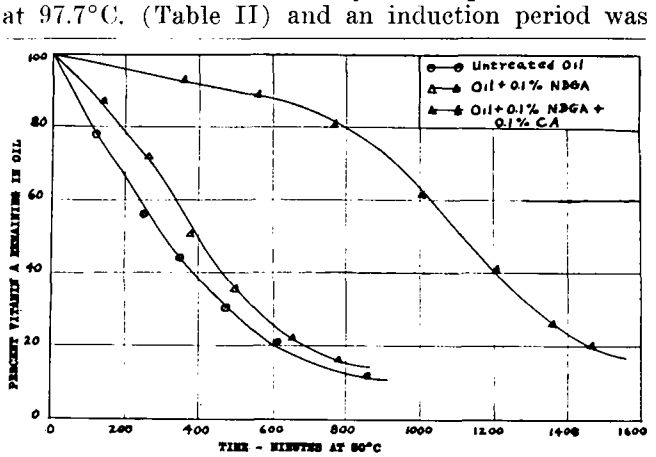

FIG. 6. Vitamin A destruction in control and NDGA treated halibut liver oil at 80°C.

present in all samples (Figure 7). NDGA was an effective antioxidant at the 0.1% level with no added synergist, and a protective factor of 7.5 was obtained at the point where 50% loss of vitamin A had occurred. Primary calcium phosphate appeared to be an effective synergist at this temperature, and with 0.1% the protective factor for NDGA was increased from 7.5 to 14. Samples with 0.1% NDGA plus 0.1% citric acid and 0.3% NDGA plus 0.1% phosphoric acid had lost less than 20% vitamin A after 700 and 1500 hours, respectively, at 25°C. compared to 25 hours for a 20% loss in the untreated oil. The tests were terminated before the end of the induction period had appeared, but a protective factor greater than 28 is indicated for 0.1% NDGA plus 0.1% citric acid under these conditions. For 0.3% NDGA plus 0.1% phosphoric acid the protection factor for stabilization of the vitamin A would be greater than 60.

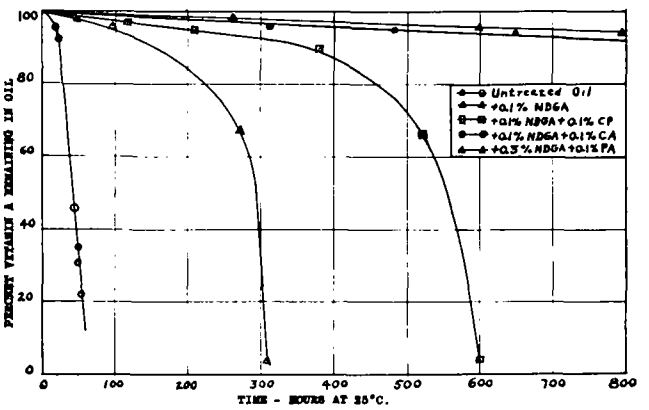

FIG. 7. Vitamin A destruction in untreated and NDGA treated halibut liver oil at 25°C.

The true vitamin A content of an oxidized or rancid oil is not necessarily measured by the optical density readings at 328 millimicrons (10). Since the absorption due to vitamin A is at a maximum at 328 millimicrons, extraneous absorption due to oxidized products in the oil is indicated by high readings at 300 and 310 millimicrons. A ratio of less than 0.73 for $E^{1\%}_{1\,cm.}\frac{300}{328}$ indicates little extraneous absorption whereas higher values indicate that absorption values are not a true measure of the vitamin A. For computation of a protection factor for vitamin A it is desirable to eliminate as much as possible the error from this source. A sample of untreated halibut liver oil was tested for absorption at 300, 310, 328, and 350 millimicrons during the course of an oxidation at 97.7°C. (Table III). With less than 25% loss of vitamin A the ratio of $E^{1\%}_{1\,cm.}\frac{300}{328}$ for the untreated oil indicated little extraneous absorption. At a greater loss than 50% vitamin A the ratios indicated considerable extraneous absorption with a probable error in the value $E^{1\%}_{1\,cm.}$ 328. For this reason the protection factors were computed on the basis of the period for 20% and 50% loss of vitamin A.

Discussion

The protection factors for both 20% and 50% loss of vitamin A are fairly close for most samples (Table 2). At lower temperatures where the induction period is present the protection factors for a 50% loss may be much less. Use of NDGA without a synergist did not give effective protection at 97.7°C. but did at considerably lower temperatures. With a synergist as citric or phosphoric acid, the antioxidant is much more effective at all temperatures; however the ratio of effectiveness appears to increase markedly with the lowering of the oxidation temperature. For 0.1% NDGA plus 0.1% citric acid the protection factor at 97.7°C. was 1.9, at 80°C. it was 3.8, and greater than 28 at 25°C. At 97.7°C. rancidity of the oil occurred concurrently with loss of vitamin A. A loss of 50% vitamin A was associated with a peroxide value from 40 to 55. Darkening of the oil and off-odors often developed at 97.7°C. with samples in which 0.3% and 0.5% NDGA were present. At lower temperatures the increased efficiency of NDGA with synergists permits concentrations of less than 0.1% NDGA for effective protection. In this case undesirable effects due to addition of antioxidant are at a minimum.

For general application the protective factors as evaluated here may not represent the average value. Other conditions of the oxidation have only been partially investigated such as the effect of temperature and effect of synergists with smaller concentrations of antioxidants. The stabilization of vitamin A in a partially oxidized oil would appear to have an application for storage of vitamin A fortified fish meals. These tests utilized only the crude vitamin A oil in which the natural antioxidant is largely present. More important applications might be extended to purified oils or vitamin A concentrates in which the natural antioxidant is not present.

TABLE III

Change in Spectrophotometric Ratios of Halibut Liver Oil During Oxidation at 97.7°C.

Oxidation time at 97.7°C.	% Loss of Vitamin A[1]	$E^{1\%}_{1\,cm.}\frac{300}{328}$	$E^{1\%}_{1\,cm.}\frac{310}{328}$	$E^{1\%}_{1\,cm.}\frac{350}{328}$
min.				
0	0	.670	.842	.625
10	3	.671	.844	.629
80	17	.717	.880	.633
120	25	.742	.889	.618
170	33	.782	.922	.599
220	42	.817	.948	.604
260	48	.867	.973	.592
290	52	.872	.990	.578
370	65	1.033	1.067	.544
470	75	1.16	1.18	.528
570	82	1.42	1.30	.490

[1] Original value $E^{1\%}_{1\,cm.} = 42.1$.

Acknowledgments

This work was partially supported by a research fellowship of the Nordigard Corporation. Acknowledgments are due the Halibut Liver Oil Producers, Seattle, Wash., for the samples of halibut liver oil, Charles Pfizer and Company, New York, for samples of citric and ascorbic acid and Nordigard Corporation, Chicago, Ill., for samples of NDGA antioxidant.

Summary

The gradual destruction of vitamin A in fish liver oils stored under oxidizing conditions occurs simultaneously with the oxidation of the oil and may be correlated with the increase of the peroxide number. Antioxidants which effectively retard the oxidation of the oil are also effective in the stabilization of the vitamin A content. The effectiveness of various concentrations of NDGA with and without added synergists was determined in halibut liver oil using a modified Swift stability procedure at 97.7°C. and at

80°C. Accelerated storage tests at 25°C. were also made. Wheeler peroxide numbers and spectrophotometric vitamin A were determined at intervals. Use of NDGA at 0.1 to 0.5% level without added synergist did not give effective protection at 97.7°C. but did at the lower temperature. NDGA was more effective at all temperatures if a synergist as citric or ortho-phosphoric acid was added; however, the relative effectiveness appeared to increase markedly with the lowering of the oxidation temperature. At lower temperatures the increased efficiency of NDGA with synergists permitted concentrations of less than 0.1% NDGA for effective stabilization. A loss of 50% vitamin A was associated with a peroxide value from 40 to 55. Undesirable effects in the oil, due to the addition of the antioxidant, were at a minimum below concentrations of 0.3%.

With 0.1% added NDGA to the oil by weight plus 0.1% citric acid as a synergist the protection factor at 25°C. was greater than 28.

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The Catalytic Isomerization of Cod Liver Oil With Sulphur Dioxide

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Introduction

AN extensive study in this laboratory of the catalytic properties of sulphur dioxide with regard to fatty oils, showed that different reactions may take place, dependent on the reaction conditions. At about 100-120°C. liquid sulphur dioxide performs a cis-trans isomerization in unsaturated fatty acids and their glycerides. When starting with low melting isomers, this reaction brings about a considerable increase in the melting point of the oil. By crystallization a still higher melting fraction may be obtained (1). If a semidrying oil is treated in the same way and the less unsaturated solid fraction thus obtained is separated, a more unsaturated liquid fraction is left, which shows very good drying properties.

A second process catalyzed by SO_2 is the "activation" of oils containing linolic or linoleic esters (2). This process is characterized by a large increase of the diene number of the oil, and this generally takes place at about 60 atm. pressure and 180-200°C.

At still higher temperatures it is found that a considerable polymerization also occurs (3), and this process has been further developed after the war (4).

The present paper will describe an application of the above mentioned research work to a new raw material, a Norwegian cod liver oil.

Experimental

The cod liver oil was freed from peroxides by heating at 180°C. for one hour while a stream of nitrogen was passed through. Its behavior towards sulphur dioxide was investigated at different temperatures by placing a sample of the oil in an autoclave of Hastalloy (58% Ni, 20% Mo, 20% Fe, 2% Mn, volume ± 120 ml.) with about the same volume of liquid SO_2, which was added afterwards.

The autoclave was then heated to the desired reaction temperature for some time. The reaction conditions for each experiment are given in Table I. After the reaction the reaction-mixture was cooled to about 90°C. and the SO_2 was blown off. The last traces of SO_2 were removed by heating the oil at 90°C. in a vacuum of about 5 mm. Hg with nitrogen ebullition. The products obtained were then analyzed by determining n_D^{20}, d_4^{20}, iodine number, viscosity, acid value, saponification value, and diene number (D.N.), which values are given in Table I. Furthermore, the ultraviolet absorption spectra of the oil and its reaction products were determined. These spectra are given in Figures 1 and 2.

Discussion of the Results

From the relatively high iodine value of the original oil it can be expected, depending on the reaction

TABLE 1

Product	Grams of oil		Grams of SO_2	p atm.	T°C.	Time, hours	n_D^{20}	d_4^{20}	r_D^{20}	I.V.*	Acid V	Sap V	η_{20}** poises	D.N.***
	Before experiment	After experiment												
Original oil	.....	.....	.....	.....	.....	.....	1.4788	0.9227	0.3072	166	1	.....	0.53	2-3
Exp. 2	nitrogen	nitrogen	.....	1	180	1	1.4789	0.9227	0.3073	165	1	201	0.53	2-3
Exp. 3	42	39	37	25	100	3	1.4780	0.9219	0.3071	165	2	.....	0.94	2-3
Exp. 3a	28	26	42	?	100	5	1.4782	0.9223	0.3070	165	4	203	0.99	.....
Exp. 4	30	27	37	25	125	3	1.4778	0.9211	0.3072	162	6	.....	0.91	2-3
Exp. 5	30	26	42	60	150	3	1.4831	0.9269	0.3082	146	15	.....	1.46	10
Exp. 6	27	23	41	80	180	3	1.4865	0.9464	0.3036	96	18	199	3.62	6

* According to Wijs. ** Determined with an Ostwald-type viscometer. *** Method of Ellis and Jones, Analyst 61, 1812 (1936).