treatment of Emmel is not necessary. His extraction time with ethanol was apparently of too short duration. Indeed, if this is not the case, his data lead to the conclusion that three toxic materials are present.

There seems to be agreement that the detoxified meals are digestible and may be used as sources of energy. The literature does not contain data on the quality of the protein in detoxified meals. Bryan implies, but certainly does not state, that his detoxified meals were the equal of soybean oil meal. Certainly our three meals were inferior to soybean oil meal. They were comparable to the cereals but no better. We feel that in the presence of adequate supplies of cereal grains the detoxication of tung meals or cakes solely for the production of a feedstuff would probably be an uneconomic process.

Conclusions

1. Commercial press-cakes and petroleum ether-extracted tung meals are toxic.
2. The toxicity of press-cake decreases markedly in common storage. The toxicity of oil-free meals does not decrease under such storage.
3. Many lipid solvents remove toxic material from tung products but cannot be used alone to produce a non-toxic meal.
4. Heat alone cannot detoxify tung meal or cake.
5. Moistening increases the effectiveness of the heat treatment.
6. Tung cake and meal may be detoxified by combining the moist heat treatment with solvent extraction.
7. Two toxic materials are present in tung nuts.
8. The biological value of tung protein is low insofar as the chick is concerned.
9. It seems improbable that the value of the detoxified meal as a feedstuff would justify the expense of detoxication.

Acknowledgment

We wish to thank R. S. McKinney and the staff of the Tung Oil Laboratory of the United States Department of Agriculture for obtaining the press-cakes used and for donating to us the solvent extracted meal which we used.

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The Relative Rates of Destruction of Propyl Gallate and Butylated Hydroxyanisole in Oxidizing Lard[1]

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UNTIL comparatively recently there has been little information recorded in the literature on the kinetics of the destruction of phenolic antioxidants in oils and fats. This lack of data has been attributed in part, at least, to the scarcity of satisfactory analytical procedures. However with the development of improved methods a number of studies have been reported on individual antioxidants.

Filer and co-workers (2) have studied the oxidative destruction of gallic acid in commercially refined cottonseed oil, aerated at 110°C., and reported that the rate of loss of gallic acid was approximately constant and was virtually independent of the initial concentration. They concluded that the destruction of gallic acid exhibited characteristics of a zero order reaction.

Lundberg *et al.* (7) investigated the rates of destruction of four antioxidants added to lard held at 100°C. under a stream of oxygen and simultaneously followed the change in peroxide value. They studied hydroquinone, catechol, nordihydroguaiaretic acid, and gallic acid, added separately to lard, at concentrations of 0.02, 0.10, and 0.50% by weight. These workers concluded that the deterioration of these phenolic antioxidants in oxidizing lard did not occur as a single low order reaction but was complicated by the products formed from the fat and/or the antioxidants during the oxidation. They observed a positive catalytic effect upon peroxide formation during the early stages of the oxidation of lard which accompanied the use of the higher concentrations of these antioxidants.

In 1949 Kraybill and co-workers (4) reported the development of a new antioxidant preparation designated as "AMIF-72," which consisted of 20% butylated hydroxyanisole (BHA), 6% propyl gallate, and 4% anhydrous citric acid in 70% propylene glycol. This antioxidant preparation is now widely used for the stabilization of edible fats. At that time no satisfactory methods were available for the determination of mixtures of these antioxidants. However a procedure which permits the determination of combinations of propyl gallate, butylated hydroxyanisole, nordihydroguaiaretic acid, and tocopherols in lard and shortening has now been reported by Mahon and Chapman (8). These workers have also developed a method for the estimation of the 2-tert-butyl-4-hydroxyanisole (2-BHA) and 3-tert-butyl-4-hydroxyanisole (3-BHA) isomers in lard and shortening (10). Since there were no data in the literature on the destruction of propyl gallate and butylated hydroxyanisole when combined in a fat or oil, it was considered of interest to apply the foregoing procedures to a study of the relative rates of destruction of these phenolic antioxidants in oxidizing lard.

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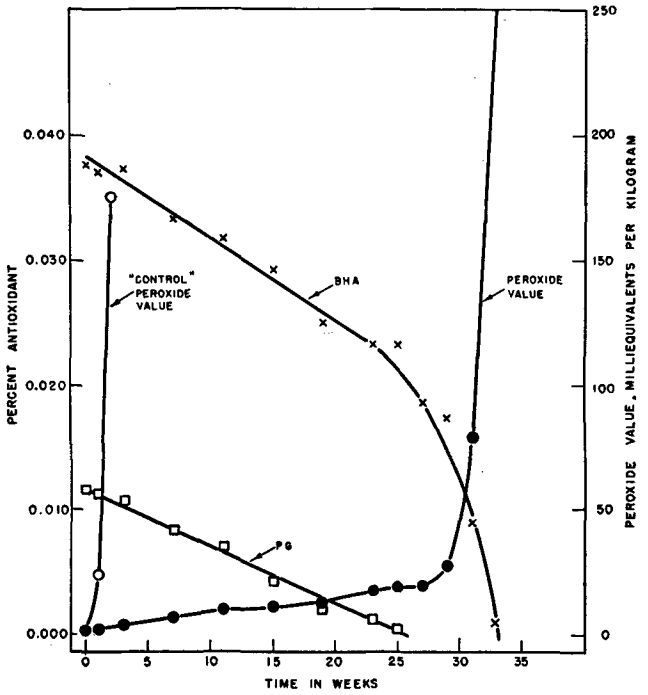

FIG. 1. Peroxide value and concentration of propyl gallate and BHA in lard sample A stored at 61°C.

Experimental

Analytical Methods

Propyl Gallate. Propyl gallate was extracted from a petrol ether solution of the lard or shortening with 1.67% aqueous ammonium acetate solution and determined in the extract employing a ferrous tartrate reagent as previously described (8). In the case of fat samples found to contain less than 0.005% of propyl gallate the following more sensitive modification was employed.

Weigh 10 grams of lard or shortening into a 500-ml. separatory funnel and dissolve the fat in 55 ml. of purified petrol ether (3 vols. of B.P. 60-100°C. and 1 vol. of B.P. 30-60°C.). Extract the fat solution with three 20-ml. aliquots of 1.67% aqueous ammonium acetate solution by continuously inverting the separatory funnel for 2.5 minutes per extraction. Finally, extract the fat solution for a few seconds with 8 ml. of water. Sufficient time must be allowed after each extraction for the complete separation of the phases before running off the aqueous layer. Dilute the combined extracts to 70 ml. with distilled water. The extract now contains 1.43% of ammonium acetate. Filter and pipette three different aliquots of the extract, not exceeding 24 ml., into 40-mm. Coleman absorption cells and dilute all aliquots to 24 ml. with 1.43% aqueous ammonium acetate solution. Add 1 ml. of freshly-prepared ferrous tartrate reagent (0.1% of $FeSO_4 \cdot 7H_2O$ and 0.5% of Rochelle salt in water) to each cell and stir the contents. After 3 minutes measure the absorbancy at 540 mμ in a Coleman Universal spectrophotometer or similar instrument equipped with wide absorption cells, relative to a blank containing 24 ml. of 1.43% aqueous ammonium acetate solution and 1 ml. of fresh ferrous tartrate reagent. Employing the Coleman spectrophotometer fitted with 40-mm. cells, the observed absorbancy divided by 0.00223 gave the concentration of propyl gallate in micrograms per aliquot used. The conversion factor should be verified by preparing a calibration curve over the range of 90 to 350 micrograms of propyl gallate per aliquot.

In the case of certain shortenings a strong tendency to emulsify was observed during the aqueous extraction of propyl gallate. To prevent emulsion formation, add 2 ml. of capryl alcohol to the fat solution before commencing the extraction. Employ 1.67% ammonium acetate in 5% ethyl alcohol to extract the propyl gallate in place of the aqueous 1.67% ammonium acetate solution. This procedure need only be employed when the normal method fails.

Total BHA. Propyl gallate must be extracted from the fat solution by using one of the foregoing methods before proceeding to the extraction of BHA. The BHA can then be extracted from the remaining fat solution with purified 72% ethyl alcohol (by volume) and determined in the extract employing the ferric chloride plus 2,2′-bipyridine reagents as previously described (8).

The 2-BHA and 3-BHA Isomers. The 72% ethyl alcohol extract obtained for the determination of "total BHA" was employed for the determination of the BHA isomers using the 2,6-dichloroquinonechloroimide-borax reagent as previously described (9). From these results the absorbancy per ml. of the BHA extract was calculated. The absorbancy per ml. of the BHA extract using ferric chloride plus 2,2′-bipyridine reagent was also determined. From these data the proportion of 2-BHA and 3-BHA isomers was estimated (10). The quantity of 2-BHA and 3-BHA was determined by multiplying the figure for "total BHA" by the proportion of these isomers found.

Peroxide Value. The peroxide value of the fat was determined by Lea's "hot-method" (6).

Experimental Conditions

Samples of fresh steam-rendered lard were obtained from two different commercial producers. Three 600-gram portions of each lard were warmed to 50°C.,

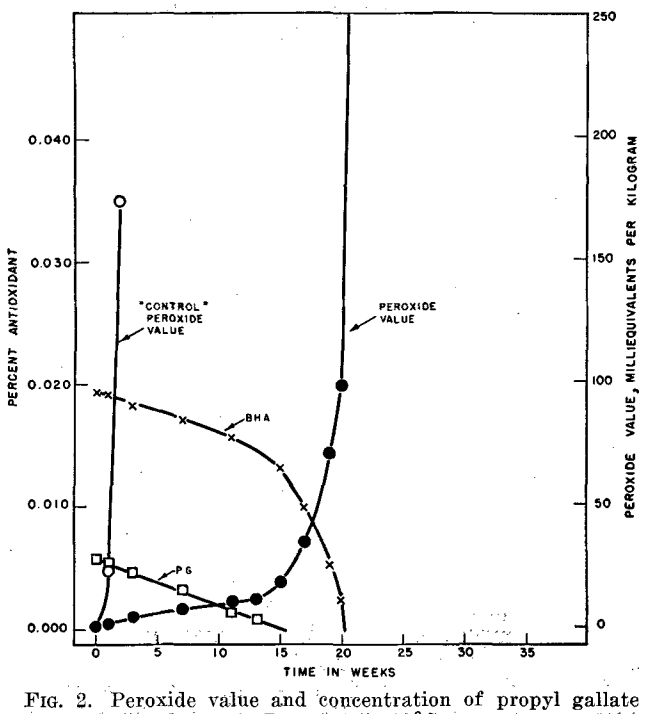

FIG. 2. Peroxide value and concentration of propyl gallate and BHA in lard sample B stored at 61°C.

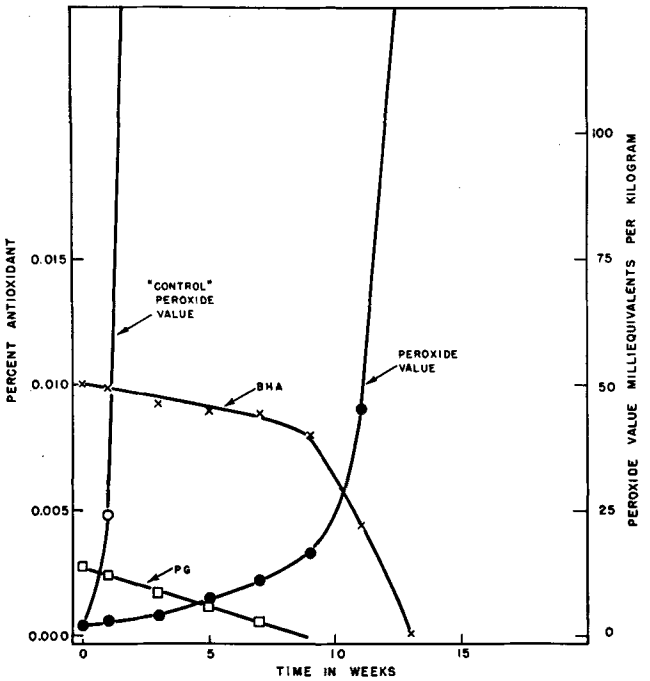

FIG. 3. Peroxide value and concentration of propyl gallate and BHA in lard sample C stored at 61°C.

and the required amount of the "AMIF-72 type" antioxidant preparation was added. The molten lard was then stirred for 10 minutes to ensure uniformity and 25-gram portions were poured into 50-ml. Pyrex beakers. The 24 beakers from each sample at each antioxidant level were divided into two groups of 12 beakers each and stored at 41° and 61°, respectively, in constant temperature ovens. At suitable intervals 12-gram portions of the lards were removed and propyl gallate, total BHA, 2-BHA, 3-BHA, and peroxide value determined. Control lard samples to which no antioxidants had been added were also stored at 41° C. and 61°C. and the peroxide value determined. In the case of the lard containing antioxidants, held at 61°C., analyses were continued until all the propyl gallate and BHA had been destroyed. Since the destruction of the antioxidants in the lard stored at 41° C. was relatively slow, this experiment was discontinued after 30 weeks.

Results and Discussion

In the initial experiments the antioxidants were added at three levels. The calculated amounts of propyl gallate, BHA, and citric acid incorporated in the lard are shown in Table I. The concentrations actually found differ slightly from these values as previous work has shown that there is frequently a small but appreciable loss immediately after the addition of these antioxidants to the lard.

The analytical data obtained for the two different lots of lard containing the "AMIF-72-type" antioxidant preparation were identical within the limits of experimental error. Therefore the results on only one lot will be presented and discussed. The results obtained for propyl gallate, BHA, and peroxide values on lard samples A, B, and C, stored at 61°C., are shown in Figures 1, 2, and 3.

Inspection of these data reveals that the rate of destruction of propyl gallate was approximately constant with respect to time for each concentration employed and therefore appeared to follow a zero order reaction. This conclusion is in agreement with those previously reported by Filer *et al.* (2) for gallic acid. Lundberg and co-workers (7) also followed the destruction of gallic acid in lard at 100°C. in an atmosphere of oxygen, employing concentrations of 0.02, 0.10, and 0.50%. Their results indicated that at concentrations of 0.02 and 0.10% the rate of destruction of gallic acid was approximately constant with respect to time, suggesting a zero order reaction. The highly irregular results obtained by these workers (7) when employing 0.50% of gallic acid was probably caused by the positive catalytic effect of this high antioxidant concentration upon the formation of fat peroxides.

TABLE I

Approximate Amounts of Antioxidant Added to Lard Samples A, B, and C

Sample	Propyl Gallate Added	BHA Added	Citric Acid Added
	%	%	%
"Control"	0.000	0.000	0.000
A	0.012	0.040	0.008
B	0.006	0.020	0.004
C	0.003	0.010	0.002

The data in Figures 1, 2, and 3 indicate that the destruction of BHA occurred in two stages. Initially there was a slow decrease in the BHA concentration which accelerated rapidly at approximately the point at which the propyl gallate could no longer be detected in the lard samples. The initial slow loss of BHA proceeded at an approximately constant rate for each concentration employed and was attributed at least in part to the volatilization of small amounts of BHA from the surface of the lard. This assumption was confirmed by placing 160 grams of lard containing 0.02% of BHA in a shallow bottle. A slow stream of nitrogen was passed over the surface of the lard, an area of approximately 150 sq. cm., and then through a series of four water-cooled traps containing absolute ethyl alcohol. This apparatus was placed in a constant temperature oven at 61°C. for 50 hours, and the BHA in the alcohol was determined. A total of 37.5 micrograms of BHA was found, corresponding to a loss of 5×10^{-3} micrograms of BHA per sq. cm. per hour. However the losses of BHA during the initial stages of the storage of samples A, B, and C amounted to 3.0×10^{-2}, 2.9×10^{-2}, and 1.1×10^{-2} micrograms per sq. cm. per hour, respectively. These values range from two to six times the amount actually recovered in the alcohol traps. It is possible that some BHA was destroyed during the process of vaporization from the lard or that during this initial period the BHA was also slowly reacting with the oxidizing fat. The second stage in the destruction of the BHA was initiated coincident with the complete disappearance of the propyl gallate and the rapid increase in the peroxide value. The rate of destruction of the BHA during this latter period was much greater than during the initial stage. This acceleration in the rate of destruction was chiefly attributed to an increase in the reactivity of BHA with the oxidizing fat.

The concentration of the isomers, 2-BHA and 3-BHA, was also determined during the storage of lard samples A, B, and C, held at 61°C. These values are shown in Figure 4. These data indicate that the de-

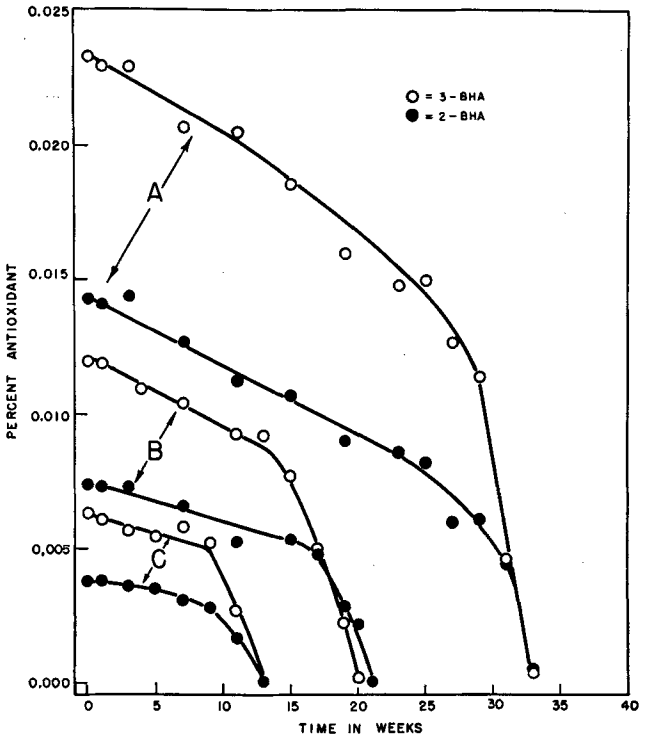

FIG. 4. The concentration of 2-BHA and 3-BHA in lard samples A, B, and C stored at 61°C.

struction of the 2-BHA and 3-BHA isomers also occurred in two stages. During the initial stage the rates of destruction of these isomers were approximately equal and remained constant with respect to time for each of the concentrations employed. As previously indicated, the evaporation of BHA is at least partially responsible for this slow initial loss. By reference to Figure 4 it can be seen that in all cases the rate of destruction of 3-BHA increased abruptly just prior to the complete disappearance of the propyl gallate (Figures 1, 2, and 3) while the rate of destruction of 2-BHA remained constant until just after the destruction of the propyl gallate and then suddenly increased. In addition, during the final stage the 3-BHA isomer was destroyed at a greater rate than the 2-BHA isomer.

The curves for the peroxide values in Figures 1, 2, and 3 exhibited similar characteristics. These data indicate that initially there was a very slow accumulation of peroxides followed by a period of rapid peroxide formation. The transition from the slow to the rapid increase was coincident with the complete destruction of the propyl gallate. It was concluded therefore that the combination of propyl gallate and BHA maintained a comparatively low peroxide value in the lard but that BHA alone was relatively ineffective in preventing further peroxide formation after the destruction of the propyl gallate.

TABLE II

Propyl Gallate and BHA Content and Peroxide Value of Lard Samples A, B, and C Stored at 41°C. for 30 Weeks

Sample	Propyl Gallate Added	Propyl Gallate at 30 Weeks	BHA Added	BHA at 30 Weeks	Peroxide Value at 30 Weeks
	%	%	%	%	*m.e./kg.*
A	0.0115	0.0080	0.0380	0.0346	7.7
B	0.0057	0.0036	0.0195	0.0172	9.8
C	0.0028	0.0011	0.0101	0.0092	20.4

The results of the experiment on lard samples A, B, and C stored at 41°C., which was terminated at the end of 30 weeks, are given in Table II.

These data indicate a trend similar to the results obtained with the same samples stored at 61°C. The loss in propyl gallate ranged from 30% in sample A to 61% in sample C. The loss in BHA however was relatively constant at an average of 10%.

In order to confirm the observation that BHA alone is relatively ineffective in maintaining a low peroxide value, it was decided to add BHA to a sample of fresh steam-rendered lard in the amounts shown in Table III. The lard samples were stored in open beakers held at 61°C. as previously described. The BHA content and the peroxide values were determined periodically with the results shown in Figure 5. These data indicate that the BHA is destroyed at a relatively rapid rate and that the peroxides accumulate with little or no induction period. Therefore it was concluded that BHA alone was relatively ineffective in preventing peroxide formation in lard stored at 61°C. as compared with a combination of propyl gallate and BHA (Figures 1, 2, and 3). This conclusion is in agreement with the statement of Kraybill and co-workers (5), who reported that BHA

TABLE III

Approximate Amount of Antioxidants Added to Lard Samples L, M, and N

Sample	BHA Added	Citric Acid Added
	%	%
"Control"	0.00	0.000
L	0.04	0.008
M	0.02	0.004
N	0.01	0.002

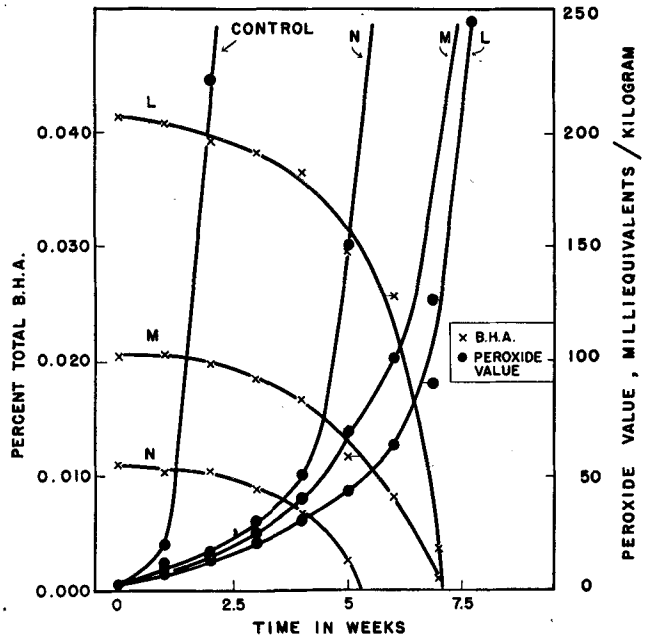

FIG. 5. Peroxide value and the concentration of BHA in lard samples L, M, and N, stored at 61°C.

when used alone was not unusually effective in increasing the stability of lard as measured by the active oxygen method. Moore and Bickford (11) in a recent study of the effectiveness of a number of antioxidants also found propyl gallate more effective than BHA in stabilizing cottonseed oil, the same oil hydrogenated to shortening consistency and lard.

The results of the determination of the 2- and 3-BHA isomers in samples L, M, and N are shown in Figure 6. These samples contain no propyl gallate, and therefore it might be assumed that these curves would resemble that portion of the curves in Figure 4 subsequent to the complete destruction of propyl gallate. Comparison of the data in Figures 4 and 6 indicates that this assumption is correct. It was noted from the data in Figure 6 that the destruction of 3-

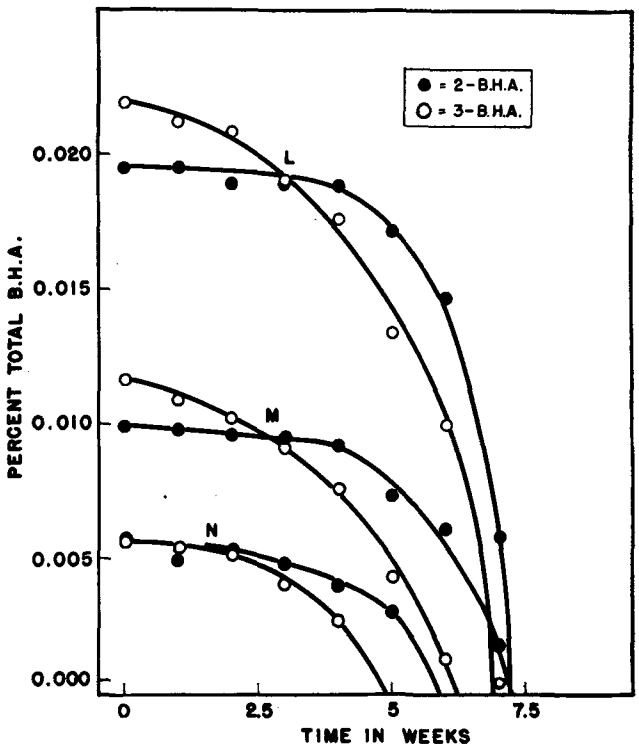

FIG. 6. Concentration of 2-BHA and 3-BHA in lard samples L, M, and N stored at 61°C.

BHA is initiated at an early stage in the storage period while 2-BHA requires an induction period of approximately 24 days in the case of lard samples L and M. It was also observed that with these samples the 3-BHA began to react at relatively low peroxide values, but a peroxide value of 25 to 35 milliequivalents per kilogram was required before the rate of destruction of 2-BHA became appreciable. No definite conclusions can be drawn from the data on sample N, owing to the very low concentration of approximately 0.005% of each of the BHA isomers. The determination of the BHA isomers is based on the color intensity obtained with the 2,6-dichloroquinonechloroimide-borax reagent relative to that obtained with the ferric chloride—2,2′-bipyridine reagent. If either of these reagents reacted with decomposition products formed during the oxidation of the fat, large errors in the determination of the iosmers would result. In order to check the validity of the determination of the 2- and 3-BHA in oxidizing fat, the following experiment was conducted. To one sample of lard 0.02% of 3-BHA was added and to a second sample an equal amount of 2-BHA. These samples were held in open beakers at 61°C., and the proportion of the isomers in each was determined until the BHA was completely destroyed. The results are given in Table IV. The ratios shown represent the optical density

TABLE IV

The Proportion of BHA Isomers as Determined in Oxidizing Lard at 61°C.

Storage Period	2-BHA		3-BHA	
	Ratio	% Found	Ratio	% Found
Weeks				
0	0.138	100.0	0.787	100.0
1	0.137	100.2	0.766	96.8
2	0.128	101.6	0.764	96.5
3	0.132	101.0	0.784	99.6
4	0.125	102.0	0.788	100.1
5	.....	.....	0.763	96.3
6	.....	.....	0.767	96.9

obtained with the 2,6-dichloroquinonechloroimide-borax reagent and divided by the optical density obtained with the ferric chloride-2,2′-bipyridine reagent. The average values of 101.2% for 2-BHA and 97.7% for 3-BHA are both within the experimental error of the method. If any substances were produced during the oxidation of the fat which interfered with the determination, the percentage of the isomers would not have remained close to 100% until completely destroyed. Therefore it was concluded that the method for the determination of the isomers was satisfactory.

Since 3-BHA began to react at a lower peroxide value than 2-BHA, it could be argued that 3-BHA would inhibit peroxide formation to a greater extent and therefore should be a more effective antioxidant.

In order to confirm this observation, amounts of 2-BHA, 3-BHA, and citric acid as shown in Table V were added to fresh steam-rendered lard.

TABLE V

Approximate Amount of Antioxidants Added to Lard Samples X, Y, Z

Sample	2-BHA Added	3-BHA Added	Citric Acid Added
	%	%	%
"Control"	0.000	0.000	0.004
X	0.020	0.000	0.004
Y	0.000	0.020	0.004
Z	0.010	0.010	0.004

The results of the experiments with lard samples X, Y, and Z, to which varying proportions of the BHA isomers plus citric acid were added and stored at 61° C., are presented in Figure 7. These data confirm the fact that 3-BHA is a more effective antioxidant than 2-BHA. Lard containing no added BHA and stored at 61°C. attained a peroxide value of 20 m.e./kg. after only 6.2 days. On the other hand, lards containing 0.02% of 2-BHA and 0.02% of 3-BHA required 9.7 and 13.8 days, respectively, to attain a peroxide value of 20. The combination of equal amounts of 3-BHA and 2-BHA was almost as effective as 3-BHA in stabilizing the lard in the initial stages of the oxidation, but at higher peroxide values this curve was approxi-

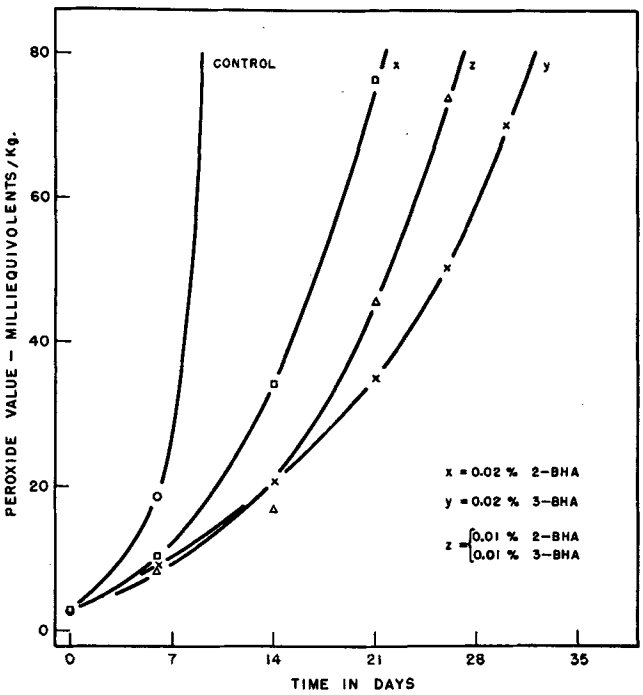

FIG. 7. Peroxide value in lard samples X, Y, and Z stored at 61°C.

mately midway between the curves for the two pure BHA isomers. These limited data indicated that the addition of 0.02% of the 2-BHA and 3-BHA isomers increased the stability of lard stored at 61°C. by factors of 1.6 and 2.2, respectively, as compared to 1.0 for lard alone. At a concentration of 0.02%, 3-BHA exhibited an inhibition ratio of approximately 1.4 relative to 2-BHA when comparison was made at a peroxide value of 20. Moore and Bickford (11) have recently stated that a more realistic evaluation of the antioxidants in lard is obtained when they are compared at a peroxide value of 100 milliequivalents per kilogram. When the data in Figure 7 are compared at a peroxide value of 80, the maximum value for which figures were available, the inhibitor ratio of 3-BHA was approximately 1.5 as compared to unity for 2-BHA.

This result is in general agreement with those reported in the literature. Dugan *et al.* (1) investigated the relative effectiveness of 2-BHA and 3-BHA alone and as mixtures of known composition on stabilization of 11 lards. They concluded that 3-BHA exhibited an inhibitor ratio of 1.5 relative to 2-BHA when 0.01% of the isomers was employed and the active oxygen method used to evaluate the relative stability of the lards. Rosenwald and Chenicek (12) have reported that 3-BHA was 2.8 times as effective as 2-BHA in inhibiting the oxidation of lard when a concentration of 0.02% was employed.

The BHA employed in these investigations contained from 54 to 62% of 3-BHA. These samples were chosen intentionally in order that sufficient 2-BHA would be present to permit accurate analysis. Commercial BHA preparations containing higher proportions of 3-BHA have been encountered (10), and these might be expected to be slightly more efficient antioxidants.

Summary

The rate of disappearance of propyl gallate and BHA has been studied in lard stored at 61°C. An antioxidant formulation containing 20% BHA, 6% propyl gallate, and 4% citric acid in 70% propylene glycol was employed. The rate of destruction of propyl gallate was approximately constant with respect to time, irrespective of the concentration, indicating a zero order reaction. The disappearance of BHA occurred in two stages, an initial slow loss which was partially attributed to evaporation of this compound from the surface of the lard, followed by a rapid decrease which was initiated coincident with the disappearance of the propyl gallate. The rapid increase of the fat peroxides in the oxidizing lard containing both BHA and propyl gallate was coincident in all cases with the complete disappearance of the propyl gallate. The results on the disappearance of the 2-BHA and 3-BHA isomers indicated that 3-BHA reacted more readily with the oxidizing fat than 2-BHA. This factor was considered to be at least partially responsible for the greater effectiveness of 3-BHA as an antioxidant. The combination of propyl gallate and BHA was much more effective than the BHA alone as judged by peroxide accumulation in lard stored at 61°C.

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