At 2,750 angstroms the three esters showed marked differences in their behaviors. Quite highly oxidized ethyl oleate showed no appreciable increase in absorption in alcohol but developed a maximum at 2,750 in alkali. The linoleate after oxidation showed no appreciable increases in either alcohol or alkali unless oxidized under more drastic conditions calculated to destroy the more stable peroxides; in the latter case an increased absorption in alcohol was obtained which was not appreciably enhanced in alkali. With the linolenate, oxidation readily produced increases in the absorption in alcohol and a further marked increase occurred when alkali was added. Thus, under the conditions used, oleates and linolenates, to a much greater extent than linoleates, appeared to develop carbonyl derivatives from their peroxides which by the addition of alkali could be enolized to conjugated triene systems.

Supplementary studies indicated that in the cases of the oxidized oleates the increased absorptions at 2,750 could not be attributed to alpha dihydroxystearic or oxidostearic acids, which may, however, have been present. The presence of alpha ketolstearic and alpha diketostearic acids, except in trace amounts, was precluded.

In conclusion, no attempt has been made in the present report to thoroughly analyze the limited spectroscopic data that have been presented. The purpose has been rather to emphasize that there is promise of much knowledge to be gained concerning the course and mechanism of the autoxidation of fats, at least in the early stages, through spectroscopic studies.

REFERENCES

1. Filer, L. J., Mattil, K. F., and Longenecker, H. E., Paper presented at Meeting of American Oil Chemists' Society, New Orleans, May, 1944.
2. Holman, R. T., Lundberg, W. O., Lauer, W. M., and Burr, G. O., J. Am. Chem. Soc. *67*, 1285 (1945).
3. Holman, R. T., Lundberg, W. O., and Burr, G. O., J. Am. Chem. Soc. *67*, 1386, 1390 (1945).
4. Mitchell, J. H., Jr., and Kraybill, H. R., J. Am. Chem. Soc. *64*, 988 (1942).
5. Atherton, D., and Hilditch, T. P., J. Chem. Soc. *1944*, 105.
6. Deatherage, F. E., and Mattill, H. A., J. Ind. Eng. Chem. *31*, 1425 (1939).
7. Farmer, E. H., Bloomfield, G. H., Sudralingam, A., and Sutton, D. A., Trans. Faraday Soc. *98*, 348 (1942).

Modification of Vegetable Oils.

V. Relative Reactivities Toward Hydrogenation of the Mono-, Di-, and Triethenoid Acids in Certain Oils

A. E. BAILEY and G. S. FISHER
Southern Regional Research Laboratory[1]
New Orleans, Louisiana

THE process of catalytic hydrogenation has long been the most useful available tool for modification of the chemical and physical properties of vegetable oils.

That the diethenoid (linoleic) acids in certain oils are hydrogenated much more readily than the monoethenoid (oleic) acids was perhaps first pointed out by Moore, Richter, and Van Arsdel (9) and has since become a matter of common observation.

The work of Richardson, Knuth, and Milligan (10) and of Hilditch and coworkers (5, 6) on the hydrogenation of marine oils has suggested that, as the number of double bonds in fatty acid chains is increased beyond two, there is not a further comparable increase in the readiness with which they add hydrogen. However, in the past it has not been possible to make any quantitative observations on the ease of hydrogenation of different unsaturated acids in natural fats because of the inadequacy of analytical methods for mixtures of such acids.

Recently a more satisfactory means of estimating certain polyethenoid acids has been provided by the spectral method of analysis of Mitchell, Kraybill, and Zscheile (8). One of the authors and other workers in this laboratory (4) have used the spectral method and other methods to determine the fatty acid composition of a number of series of hydrogenated cottonseed, peanut, soybean, and linseed oils. Selected portions of these data have provided a basis for calculation of the relative readiness with which hydrogen adds to oleic, linoleic, and linolenic acids, and certain isomers of these acids, upon catalytic hydrogenation of the oils.

Hydrogenation and Analysis of the Oils

The methods followed in preparing and analyzing the various oil samples are described in detail elsewhere (4). Of the oils with which the present discussion is concerned, the hydrogenated cottonseed and soybean oils were prepared under "selective" conditions of hydrogenation whereas one of the series of linseed oils was hydrogenated selectively (at 375° F. and 15 p.s.i. hydrogen pressure) and another was hydrogenated relatively nonselectively (at 250° F. and 60 p.s.i.). Hydrogenation was carried out in each case with a dry-reduced, electrolytically precipitated nickel catalyst (1), in the apparatus described previously (3). Each series represented samples withdrawn successively from a single batch of oil.

Analyses of the oils, in terms of their component fatty acids, are given in Table 1. The compositions of the cottonseed oils were calculated from iodine and thiocyanogen values. Briefly, in the case of the soybean and linseed oils, the method of analysis consisted of the following: *(a)* determination of the saturated acids by the Bertram oxidation method, *(b)* determination of linoleic and linolenic acids by the spectral method, and *(c)* estimation of total oleic and isolinoleic acids according to the preceding deter-

---

[1] One of the laboratories of the Bureau of Agricultural and Industrial Chemistry, Agricultural Research Administration, U. S. Department of Agriculture.

minations and the iodine value of the oil. Determinations of iso-oleic acid were made by the official method of the American Oil Chemists' Society.

It is to be noted that, according to the above method of calculation and the terminology adopted by Lemon (7), "isolinoleic acid" refers to all diethenoid acids which have their double bonds in such positions that they will not form a conjugated system upon alkali isomerization. In the case of the hydrogenated soybean and linseed oils there appears to be no reason to doubt that the isolinoleic acid largely consists, as postulated by Lemon, of a 9:10, 15:16 acid produced by hydrogenation of linolenic acid at the middle double bond.

It is recognized that the above distribution of fatty acids disregards the presence of minor amounts of conjugated fatty acids in the oils and the possible presence of triethenoid isomers which could fail to yield conjugated double bonds upon isomerization. However, it is believed that the analyses are reasonably accurate with respect to contents of the major fatty acids and that they are sufficient for the purposes of the present discussion.

The Concept of Affinity Constants

In the case of a number of different fatty acids existing together in an oil and competing for hydrogen it appears reasonable to assume that, of an infinitesimal amount of hydrogen absorbed, the fraction going to each acid will depend upon the concentration of the acid and a constant expressive of the affinity of the acid for hydrogen under the particular conditions of hydrogenation then obtaining.

In dealing with the relative reactivities of two fatty acids, *A* and *B*, let H_A be the fraction of hydrogen absorbed by *A* in any interval of hydrogenation, and let H_B be the fraction absorbed by *B*. Let A_m be the average concentration of *A* over the interval, and let B_m be the average concentration of *B*. Let the affinities of *A* and *B* for hydrogen be respectively represented by the constants k_A and k_B. Then, from the assumptions outlined above,

$$H_A = k_A A_m/(k_A A_m + k_B B_m)$$

$$\text{and, } H_B = k_B B_m/(k_A A_m + k_B B_m)$$

$$\text{or, } k_A/k_B = H_A B_m/H_B A_m$$

The ratio k_A/k_B is a measure of the relative readiness with which *A* and *B* undergo hydrogenation; if, for example, it is equal to 2.0, we may say that the reactivity of *A* is twice that of *B*, or that *A* hydrogenates twice as readily as *B*.

While the concept of affinity constants is useful, and indeed appears to be essential to any quantitative treatment of the present subject, it has certain limitations which should be recognized in applying it to practice. A given set of affinity constants is applicable only to a single set of hydrogenation conditions. These conditions comprise not only such obvious variables as, for example, the temperature of the reaction, but probably also such difficultly definable factors as the ratio of oil to hydrogen adsorbed on the catalyst (2), and the configuration of the glycerides in the oil. All conditions may not remain constant throughout the course of hydrogenation hence the possibility should be recognized of different affinity constants obtaining for different phases of the hydrogenation reaction.

The affinity constants are to be distinguished from velocity or rate constants, for they do not embody the dimension of time. However, they derive from a similar concept, and in the theoretical case where the hydrogenation of each acid assumes the form of a monomolecular reaction, and hydrogenation condi-

TABLE 1

Composition of the Unhydrogenated and Hydrogenated Oils

Sample		Iodine value	Percentage composition of fatty acids					
			Saturated	Oleic (total)*	Linoleic	Isolinoleic	Linolenic	Iso-oleic
Unhydrogenated cottonseed oil	CO-0	101.7	26.6	29.1	44.3	......	......	......
Selectively hydrogenated cottonseed oil	CO-1	76.6	27.3	56.6	16.1	......	......	......
	CO-2	71.9	27.5	61.6	10.9	......	......	......
	CO-3	67.0	29.0	64.2	6.8	......	......	......
	CO-4	62.6	31.2	64.8	3.9	......	......	......
	CO-5	57.8	33.9	65.0	1.1	......	......	......
	CO-6	52.8	38.6	61.4	0.0	......	......	......
	CO-7	46.7	45.7	54.3	0.0	......	......	......
Unhydrogenated soybean oil	SO-0	129.6	14.3	28.2	51.0	0.0	6.49	......
Selectively hydrogenated soybean oil	SO-1	116.2	14.4	40.7	41.0	0.0	3.85	......
	SO-2	108.5	14.5	47.9	33.4	1.7	2.55	......
	SO-3	98.0	14.6	58.5	23.4	2.3	1.23	......
	SO-4	90.4	14.8	66.1	14.8	3.8	0.50	......
	SO-5	81.1	15.7	74.5	5.7	3.9	0.2	......
	SO-6	71.2	20.5	76.3	0.9	2.2	0.1	......
	SO-7	60.2	30.9	68.2	0.00	0.9	0.00	......
	SO-8	51.2	40.5	59.5	0.00	0.0	0.00	......
Unhydrogenated linseed oil	LO-0	188.9	8.9	17.0	17.1	5.0	52.0	0.7
Selectively hydrogenated linseed oil	LO-2-1	162.9	9.6	28.4	15.6	11.1	35.3	2.6
	LO-2-2	143.6	9.8	37.5	13.2	16.8	22.7	5.9
	LO-2-3	124.4	10.0	46.1	8.4	25.6	9.9	12.1
	LO-2-4	104.5	11.1	57.7	4.8	25.5	0.9	18.5
	LO-2-5	85.7	17.7	65.4	0.02	16.7	0.2	27.3
	LO-2-6	66.0	31.0	61.5	0.00	7.6	0.1	29.0
	LO-2-7	47.1	48.1	49.1	0.00	2.8	0.00	......
	LO-2-8	29.3	66.4	33.1	0.00	0.5	0.00	10.7
Nonselectively hydrogenated linseed oil	LO-1-1	163.4	9.4	27.7	17.6	10.6	34.7	2.6
	LO-1-2	141.8	10.3	36.5	15.6	17.0	20.6	5.6
	LO-1-3	119.7	13.2	44.0	11.7	22.3	8.8	7.4
	LO-1-4	96.3	18.8	52.7	5.5	21.1	1.9	11.6
	LO-1-5	75.8	28.6	55.1	1.3	14.9	0.1	11.2
	LO-1-6	54.6	43.6	49.4	0.1	6.9	0.00	13.3
	LO-1-7	31.3	64.7	34.3	0.00	1.0	0.00	9.9
	LO-1-8	22.2	74.2	25.8	0.00	0.0	0.00	6.8

* Includes both normal and "iso"-oleic acids.

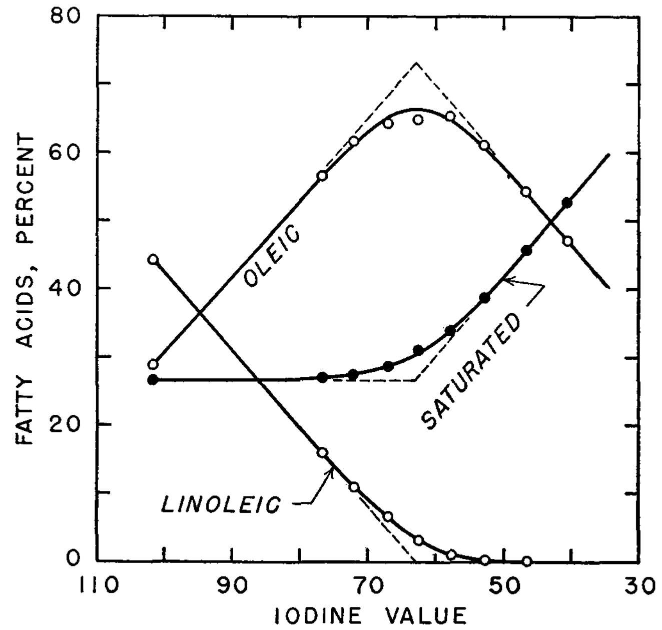

FIG. 1. Composition of selectively hydrogenated cottonseed oil.

tions do not vary, the ratio k_A/k_B will be numerically equal to the ratio of the corresponding velocity constants.

Relative Reactivities of Oleic and Linoleic Acids

In the hydrogenation of cottonseed oil or other oil whose unsaturated acids consist substantially of oleic and linoleic acids, the total oleic acid content rises to a maximum and thereafter declines (Fig. 1). At the maximum, the hydrogenation of oleic acid to stearic acid is proceeding at the same rate as the hydrogenation of linoleic acid to oleic acid, hence, from the preceding equations,

$$k_{linoleic}/k_{oleic} = [\text{conc. oleic}]/[\text{conc. linoleic}]$$

In the present experiment wherein cottonseed oil was hydrogenated selectively, the oleic acid content reached a maximum of about 66% when the linoleic acid content was about 3.5%, hence $k_{linoleic}/k_{oleic} = 19$. In previous experiments (3) in which hydrogenation conditions were varied to produce differing degrees of selectivity, calculated values of $k_{linoleic}/k_{oleic}$ vary from about 5 to 23.

Data on the hydrogenated soybean and linseed oils do not lend themselves so readily to calculation of this ratio. However, it is obvious that in these oils, also, it is relatively high.

Relative Reactivities of Oleic and Iso-oleic Acids

The term "iso-oleic acid" refers to the monoethenoid acids which appear with the saturated acid fraction in the Twitchell lead soap separation. It is probable that they comprise more than one isomer, and more or less indeterminate proportions of the entire amount of each isomer in the oil. For this reason, it is hardly possible to arrive at any precise conclusions regarding the readiness with which this class of acids hydrogenates. However, in both selective and nonselective hydrogenation, iso-oleic acid reaches a maximum at very nearly the same iodine value as total oleic acids (Fig. 2). It appears, therefore, that there is no great difference in the ease of hydrogenation of the normal and isomeric monoethenoid acids and that hydrogenation of the two is affected similarly by a change from selective to nonselective hydrogenation conditions.

Reactivity of Isolinoleic Acid in Comparison with Other Unsaturated Acids

From the persistence of isolinoleic acid in the soybean oil after linoleic acid has disappeared (Fig. 3), it is evident that the former acid is hydrogenated with relative difficulty; in the hydrogenated linseed oils, this effect may be more closely evaluated. The relative reactivities of oleic and isolinoleic acids may be estimated from the change in composition of the oils toward the end of each series after these two acids have become virtually the only unsaturated acids remaining in the oil. For the present purpose of approximation it appears sufficient to take the average concentration of each acid in the interval between two successive samples as a simple arithmetical average. If this is done, and the increase in saturated acids is taken as a measure of the oleic acid hydrogenated, and the decrease in isolinoleic acid is taken as a measure of the amount of this acid hydrogenated, the data of Table 2 emerge.

TABLE 2

Calculation of Relative Reactivities (Expressed as Ratio of Affinity Constants) of Oleic and Isolinoleic Acids in the Hydrogenation of Linseed Oil

Selective Hydrogenation

	Interval (in terms of iodine value)		
	85.7 to 66.0	66.0 to 47.1	47.1 to 29.3
H_{oleic}	13.3	17.1	18.3
$H_{isolinoleic}$	9.1	4.8	2.3
$[\text{oleic}]_m$	63.5	55.3	41.1
$[\text{isolinoleic}]_m$	12.2	5.2	1.7
$k_{isolinoleic}/k_{oleic}$	3.6	3.0	3.0

Nonselective Hydrogenation

	Interval (in terms of iodine value)
	54.6 to 31.3
H_{oleic}	21.1
$H_{isolinoleic}$	5.9
$[\text{oleic}]_m$	41.9
$[\text{isolinoleic}]_m$	4.0
$k_{isolinoleic}/k_{oleic}$	2.9

On the basis of these data it appears that isolinoleic acid hydrogenates only about three times as readily as oleic acid and that the relative affinities of the two acids for hydrogen are but little dependent upon the conditions of hydrogenation.

During the course of hydrogenation, isolinoleic acid, like total oleic acids, rises to a maximum. At the maximum, the respective hydrogenations of isolinoleic acid to oleic acid and linolenic acid to isolinoleic acid are proceeding at equal rates hence calculations may be made of the ratio $k_{linolenic}/k_{isolinoleic}$, as outlined previously for the ratio $k_{linoleic}/k_{oleic}$. If calculations are made upon the basis of the total amount of linolenic acid in the oil, the ratios are 29.0/3.5, or 8.3 for the selective hydrogenation and 22.5/6.5, or 3.5 for the nonselective hydrogenation. Actually, however, linolenic acid goes about equally to normal linoleic acid and isolinoleic acid upon hydrogenation (see the succeeding section) so that in this case the values of linolenic acid concentration which should properly be used in the calculation are half the total values. Recalculation on this basis gives for $k_{linolenic}/k_{isolinoleic}$, about 16 in the case of selective hydro-

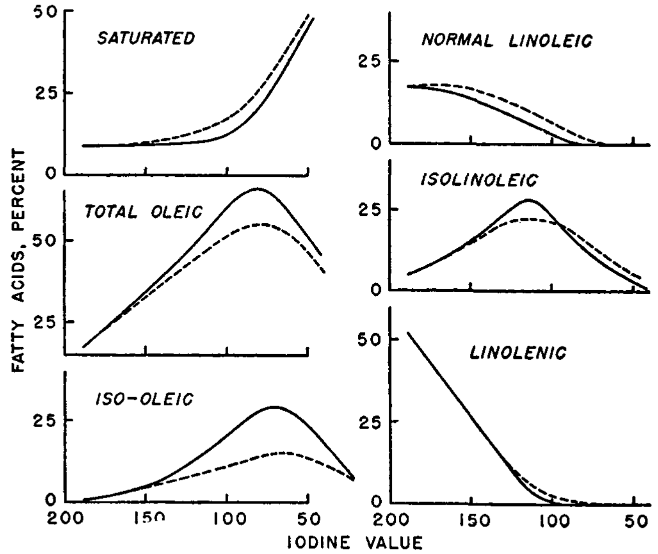

FIG. 2. Composition of hydrogenated linseed oil. Selective hydrogenation (solid lines) *vs.* nonselective hydrogenation (dotted lines).

genation and about 7 in the case of nonselective hydrogenation.

The relatively low reactivity of isolinoleic acid toward hydrogenation is consistent with its apparent resistance toward oxidation, and its high thiocyanogen absorption (4), all of which are to be expected from the widely separated position of its double bonds.

Relative Productions of Normal and Isolinoleic Acids

A rough estimate of the proportions of linolenic acid hydrogenating respectively to linoleic and isolinoleic acids may be made from the change in composition of the linseed oils during the initial period of hydrogenation. During the first part of the reaction the concentration of isolinoleic acid is low, and since this acid hydrogenates with relative difficulty, the amount appearing during any interval approximates the amount actually formed. The amount of isolinoleic plus the amount of linoleic acid formed is, of course, measured by the decrease in linolenic acid content.

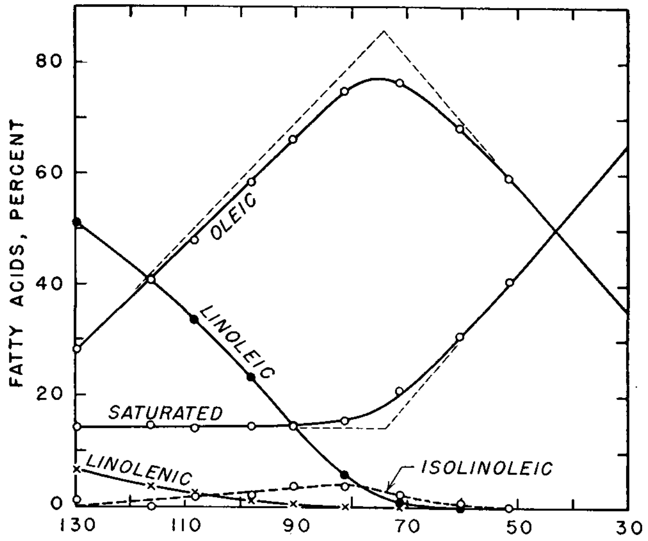

FIG. 3. Composition of selectively hydrogenated soybean oil.

Calculations from the first two intervals of hydrogenation in each of the two experiments are outlined in Table 3. When allowance is made for the circumstance that the recorded percentages of linolenic acid converted to isolinoleic acid are actually minimum values, it is evident that the proportions of the two isomers produced are roughly equal, and that the proportions are not to any large degree changed by altering the hydrogenation conditions.

TABLE 3

Calculation of Relative Proportions of Linolenic Acid Hydrogenating to Normal Linoleic and Isolinoleic Acids in Linseed Oil

Selective Hydrogenation

	Interval (in terms of iodine value)	
	188.9 to 162.9	162.9 to 143.6
Total linolenic acid hydrogenated, %	16.7	12.6
Amount to isolinoleic	6.1	5.7
Amount to linoleic (by difference)	10.6	6.9
Per cent to isolinoleic	37	45
Per cent to linoleic	63	55

Nonselective Hydrogenation

	Interval (in terms of iodine value)	
	188.9 to 163.4	163.4 to 141.8
Total linolenic acid hydrogenated, %	17.3	14.1
Amount to isolinoleic	5.6	6.4
Amount to linoleic (by difference)	11.7	7.7
Per cent to isolinoleic	32	45
Per cent to linoleic	68	55

Relative Reactivities of Linoleic and Linolenic Acids

The relative reactivities of linoleic and linolenic acids in soybean oil may readily be estimated from data from the first part of the hydrogenation, where the amount of hydrogen added by the linolenic acid is measured by the decrease in linolenic acid content, and the amount added by linoleic acid is approximated by the increase in oleic acid content. Calculations made on the basis previously outlined are shown in Table 4.

TABLE 4

Calculation of Relative Reactivities (Expressed as Ratio of Affinity Constants) of Linoleic and Linolenic Acids in the Hydrogenation of Soybean Oil

Selective Hydrogenation

	Interval (in terms of iodine value)		
	129.6 to 116.2	116.2 to 108.5	108.5 to 98.0
$H_{linolenic}$	2.64	1.30	1.32
$H_{linoleic}$	12.5	7.2	10.6
$[linolenic]_m$	5.17	3.20	1.89
$[linoleic]_m$	46.0	37.2	28.4
$k_{linolenic}/k_{linoleic}$	1.9	2.1	1.9

Under the selective conditions obtaining in this case, the linolenic acid in soybean oil hydrogenates with approximately twice the readiness of the linoleic acid.

In linseed oil, where the ratio of linolenic to linoleic acid is much higher, a wholly different ratio of reactivities is observed, as indicated in Table 5. In the calculations outlined in this table, the increase in total oleic acids plus the increase in saturated acids has been taken as a measure of the hydrogen added to linoleic acid, whereas the hydrogen going to linolenic acid is of course measured by the decrease in linolenic acid content. Some degree of uncertainty in the calculations is occasioned by the circumstance that the isolinoleic acid content in the interval is relatively

TABLE 5

Calculation of Apparent Relative Reactivities (Expressed as Ratio of Affinity Constants) of Linoleic and Linolenic Acids in the Hydrogenation of Linseed Oil

	Selective hydrogenation I.V. 188.9 to 162.9	Nonselective hydrogenation I.V. 188.9 to 163.4
$H_{linolenic}$	16.7	17.3
$H_{linoleic}$	12.1	11.2
$[linolenic]_m$	43.7	43.4
$[linoleic]_m$	16.4	17.4
$k_{linolenic}/k_{linoleic}$	0.5	0.6

high and that an indeterminate small proportion of the oleic acid produced must actually have been derived from the hydrogenation of this acid. However, even when a maximum allowance is made for this effect, it is still apparent that the calculated reactivity of linolenic acid, in comparison with the reactivity of linoleic acid, is much lower in linseed oil than in soybean oil.

It seems unlikely that, through the existence of a peculiarity in glyceride structure or from other causes, linolenic acid could actually react with hydrogen less readily than linoleic acid. A possible explanation of the effect observed is that in a certain proportion of molecular collisions a single contact of fatty acid with hydrogen and active nickel may serve not only to convert linolenic acid to linoleic acid but also to further convert linoleic acid to oleic acid without intermediate desorption of linoleic acid. In other words, the series of reactions may be in part,

$$\begin{aligned}&\text{Ni} + \text{linolenic acid} + 2\text{H}_2 \rightarrow\\&\quad[\text{Ni} — \text{linolenic acid} — \text{hydrogen}]\ \text{complex} \rightarrow\\&\qquad\text{Ni} + \text{oleic acid}\end{aligned}$$

rather than wholly,

$$\begin{aligned}&\text{Ni} + \text{linolenic acid} + \text{H}_2 \rightarrow\\&\quad[\text{Ni} — \text{linolenic acid} — \text{hydrogen}]\ \text{complex} \rightarrow\\&\qquad\text{Ni} + \text{linoleic acid}\end{aligned}$$

and,

$$\begin{aligned}&\text{Ni} + \text{linoleic acid} + \text{H}_2 \rightarrow\\&\quad[\text{Ni} — \text{linoleic acid} — \text{hydrogen}]\ \text{complex} \rightarrow\\&\qquad\text{Ni} + \text{oleic acid}\end{aligned}$$

In the case of soybean oil, which has a relatively low linolenic acid content, the operation of a mechanism such as the one suggested would cause but a comparatively slight error in calculation of the ratio, $k_{linolenic}/k_{linoleic}$. However, in linseed oil, where the concentration of linolenic acid is high in relation to linoleic acid, it would have the effect of making the calculated ratio much too low.

It is obvious that changing the hydrogenation conditions so as greatly to change the relative reactivities of oleic acid and linoleic acid has little or no effect on the relative reactivities of linoleic and linolenic acids. With respect to the so-called factor of "selectivity" the unsaturated fatty acids may be divided into two groups comprising oleic and isolinoleic acids in one group and linoleic and linolenic acids in the other. The readiness with which one group hydrogenates relative to the other may be greatly changed by altering the conditions of hydrogenation, but such alteration has but little effect with respect to the hydrogenation of the two different fatty acids within the same group.

Summary

From the analysis of several series of hydrogenated cottonseed, soybean, and linseed oils, made by the spectral and other methods, estimates have been made of the relative readiness of hydrogenation of oleic, iso-oleic, linoleic, and linolenic acids, and an isomer of linoleic acid which presumably has its double bonds in the 9:10 and 15:16 positions.

The conclusions reached are as follows:

(a) Under selective conditions of hydrogenation, the relative reactivities of the unsaturated fatty acids toward hydrogen may be approximately represented by the following whole numbers: oleic acid, 1; iso-oleic acid, 1; isolinoleic acid, 3; linoleic acid, 20; linolenic acid, 40.

(b) Use of so-called "nonselective" conditions of hydrogenation reduces the reactivity of linoleic and linolenic acids as a group in comparison to that of oleic and isolinoleic acids as a group but does not appreciably alter the relative reactivities of the two acids within the same group. In nonselective hydrogenation the ratio of reactivity of linoleic acid to that of oleic acid may be as low as 5, as compared with 20 or above under selective conditions.

(c) The hydrogenation of linoleic acid to stearic acid appears to proceed substantially in two stages, with intermediate desorption of oleic acid from the catalyst, but there is evidence that the hydrogenation of linolenic acid may in part proceed directly to oleic acid, without intermediate desorption of linoleic acid.

(d) In both selective and nonselective hydrogenation, the hydrogenation of linolenic acid to linoleic acid and to isolinoleic acid appears to proceed at approximately equal rates.

Acknowledgment

The authors wish to acknowledge the assistance of R. T. O'Connor, Dorothy C. Heinzelman, F. G. Dollear, G. E. Mann, and B. A. Smith, who were responsible for a large part of the analytical data upon which the present analysis of the hydrogenation reaction is based.

REFERENCES

1. Bailey, A. E., *Industrial Oil and Fat Products*, Interscience Publishers, New York, 1945, pp. 591-93.
2. Bailey, A. E., *ibid.*, p. 578.
3. Bailey, A. E., Feuge, R. O., and Smith, B. A., Oil & Soap *19*, 169-76 (1942).
4. Fisher, G. S. and coworkers, in manuscript.
5. Harper, D. A. and Hilditch, T. P., J. Soc. Chem. Ind. *56*, 322-329T (1937).
6. Hilditch, T. P. and Terleski, J. T., J. Soc. Chem. Ind. *56*, 315-22T (1937).
7. Lemon, H. W., Can. J. Research *22F*, 191-98 (1944).
8. Mitchell, J. H., Kraybill, H. R., and Zscheile, F. P., Ind. Eng. Chem. Anal. Ed. *15*, 1-3 (1943).
9. Moore, H. K., Richter, G. A., and Van Arsdel, W. B., J. Ind. Eng. Chem. *9*, 451-62 (1917).
10. Richardson, A. S., Knuth, C. A., and Milligan, C. H., Ind. Eng. Chem. *17*, 80-83 (1925).