Chapter 14

SOYBEAN OIL FLAVOR STABILITY

E. N. Frankel
Northern Regional Research Center
Agricultural Research
Science and Education Administration
U.S. Department of Agriculture
Peoria, Illinois 61604

INTRODUCTION

At one time, soybean oil was considered to be as much an industrial oil as an edible oil—and only second-rate in either application (Black and Mattil, 1951; Bradley, 1951). For example, during an edible oil shortage in World War I, oil was imported from Manchuria to the extent of 261 million pounds and 52% of this amount was used in nonfood products (U.S. Department of Agriculture, 1966). Today, soybean oil is considered primarily an edible oil and only 6% goes into nonfood uses. The American consumer views soybean oil as virtually equivalent to corn and sunflowerseed oils in a variety of attributes and actually prefers soybean to corn oil (Erickson and Falb, 1980). Soybean oil now provides the most important source of vegetable food fat. Its consumption can be estimated by 8.6 billion pounds of domestic disappearance and 1.8 billion pounds in export forecast for 1978 (U.S. Department of Agriculture, 1979).

The development of soybean oil into a quality food product has been the result of basic and applied research, especially by the Northern Regional Research Center and the industry. A number of reviews have documented the various factors involved in improving soybean oil flavor stability (Cowan, 1966; Cowan and Evans, 1962; Dutton, 1976 and 1978; Smouse, 1979). Although processed soybean oil is bland when used in margarines, shortenings, and salad oils, it may deteriorate organoleptically when repeatedly heated at cooking and frying temperatures and when exposed to light. Unless properly processed, soybean oil develops a very complex type of lipid deterioration usually but inappropriately called "flavor reversion." The present status of research
What is flavor reversion?

Flavor reversion is characteristic of soybean oil and other linolenate-containing oils and develops at low levels of oxidation; the peroxide value, if measurable, is 10 or below. In contrast, oxidative rancidity is observed at higher levels of oxidation (peroxide value of 10 or above) and is a flavor deterioration common to all unsaturated fats.

Flavor reversion is described as beany and grassy at the early stages and as fishy or painty at the more advanced stages. Because none of these flavors characterize the crude oil, "reversion" is a misnomer which still persists. This loss of flavor quality occurs at such low levels of oxidation that some workers have suggested that it is not an oxidative deterioration, because the degree of oxidation is so low that it often cannot be measured chemically though it is detected organoleptically. Other lines of evidence also suggest a nonoxidative process: (1) Antioxidants are not effective in retarding flavor reversion (Frankel et al., 1959; Mounts et al., 1978), but they are effective in retarding rancidity; and (2) Hydrogenation is not completely effective in eliminating reversion, but it surely retards rancidity. The reason for this ineffectiveness will be discussed in the last part of this chapter.

Arguments in support of an oxidative process for the flavor reversion of soybean oil include: (1) At measurable oxidation levels there is good correlation between flavor score and peroxide values (Dutton et al., 1948); (2) Antioxidants are not expected to prevent the formation of offensive flavors derived from the decomposition of unstable hydroperoxides (Frankel et al., 1959); (3) Traces of oxygen, loosely bound oxygen or metal-oxygen addition compounds (Uri, 1961) can catalyze flavor deterioration in the absence of air. Oxidatively derived polymers can also decompose to cause flavor deterioration (Chang and Kummerow, 1953; Johnson et al., 1953), and this process can occur in the absence of air; and (4) Prooxidant metal catalysts seriously accelerate the development of flavor reversion (Evans et al., 1951).

Oils susceptible to flavor reversion are unusually sensitive to the effect of metals—much more so than those fats which are prone to rancidity. Metal chelating agents are very effective in the control of flavor reversion in soybean oil (Dutton et al., 1948).

It may be concluded that flavor reversion involves some oxidation.

Causes of flavor reversion

Linolenic acid derivatives

Linolenic acid is widely accepted as the most important precursor of flavor reversion. In a classical experiment, Dutton et al. (1951) interesterified linolenic acid into cottonseed oil; the oxidized product, when examined by a taste panel, produced the same flavor response as reverted soybean oil. The decomposition of linolenate hydroperoxides is observed at relatively early stages of oxidation (Frankel, 1962). The terminal pentene radical in linolenate, \( \text{CH}_3\text{CH}_2=\text{CH=CH-CH}_2^- \), is generally thought to be associated with the flavor reversion compounds of soybean oil.

Numerous flavor compounds in oxidized soybean oil have been identified by different workers (Table 1). Many of these compounds are expected from the decomposition of linolenate hydroperoxides (Frankel, 1962). Hoffman (1961) isolated 3-cis and trans-hexenal from reverted soybean oil and described the odor of the cis isomer as green-beany. This compound is derived from the decomposition of linolenate hydroperoxides. However, in later work, Chang et al. (1966) attributed the flavor of reverted soybean oil to 2-pentyl furan, which they assumed to come from linoleate oxidation. They suggested that linolenic acid catalyzes the oxidation of linoleate. In a more recent report from the same laboratory (Ho et al., 1978), the cis and trans isomers of 2-pentenyl furan, which are expected from linolenate oxidation, were claimed to produce flavors and odors related to reverted soybean oils.

Different isomeric distributions of fatty ester hydroperoxides are found in mixtures of linoleate and linolenate oxidized to different levels (Frankel et al., 1977). Such differences might account for the unique flavor deterioration of soybean oil. For example, an unexpectedly high concentration of the 12-hydroperoxide isomer has been found in soybean oil esters oxidized at low levels, which may suggest a different pattern of hydroperoxide decomposition (Frankel and Neff, 1979). A complex mixture of volatile products is expected from decomposition of each of the multitude of hydroperoxides found in oxidized soybean oil. The number of odor and flavor compounds identified by different labora-
Class Compounds

**Aldehydes**
Alkanals: C_2^1, C_3^1, C_4, C_5, C_6, C_7, C_8, C_9
Alkenals: C_4^2, C_5^2, C_6, C_7, C_8, C_9, C_10, C_11
Alkadienals: C_8
Dienals: C_6, C_7, C_8, C_9, C_10, C_11
Dieneals: C_8, C_9, C_10
Trienals: C_9, C_{10,11}

**Ketones**
2-Alkanones: C_4, C_5, C_6, C_7, C_8
3-Alkanones: C_9
Unsaturated: 1-pentene-3-one, 4-octene-3-one

**Hydrocarbons**

**Ethyl esters**
C_1, C_2

**Dialdehydes**

**Other compounds**

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**TABLE 1**
Partial List of Volatile Compounds Identified in Oxidized Soybean Oil

<table>
<thead>
<tr>
<th>Class</th>
<th>Compounds</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aldehydes</td>
<td>Alkanals: C_2^1, C_3^1, C_4, C_5, C_6, C_7, C_8, C_9</td>
</tr>
<tr>
<td></td>
<td>Alkenals: C_4^2, C_5^2, C_6, C_7, C_8, C_9, C_10, C_11</td>
</tr>
<tr>
<td></td>
<td>Alkadienals: C_8</td>
</tr>
<tr>
<td></td>
<td>Dienals: C_6, C_7, C_8, C_9, C_10, C_11</td>
</tr>
<tr>
<td></td>
<td>Dieneals: C_8, C_9, C_10</td>
</tr>
<tr>
<td></td>
<td>Trienals: C_9, C_{10,11}</td>
</tr>
<tr>
<td>Ketones</td>
<td>2-Alkanones: C_4, C_5, C_6, C_7, C_8</td>
</tr>
<tr>
<td></td>
<td>3-Alkanones: C_9</td>
</tr>
<tr>
<td></td>
<td>Unsaturated: 1-pentene-3-one, 4-octene-3-one</td>
</tr>
<tr>
<td></td>
<td>Alkadienals: C_8</td>
</tr>
<tr>
<td></td>
<td>Alcohol: Saturated: C_2, C_3, C_4, C_5, iso C_5, C_6, C_7</td>
</tr>
<tr>
<td></td>
<td>Unsaturated: 1-pentene-3-one, 1-octene-3-one</td>
</tr>
<tr>
<td></td>
<td>Ethyl esters: C_1, C_2</td>
</tr>
<tr>
<td></td>
<td>Hydrocarbons: Saturated: C_2, C_3, C_4, C_5, C_6, C_7, C_10, C_11</td>
</tr>
<tr>
<td></td>
<td>Unsaturated: 2-pentene, 1-hexene, 2-octene, 1-decene, 1-docyne</td>
</tr>
<tr>
<td></td>
<td>Other compounds: 2-pentyl furan, lactones, benzene, benzaldehyde, acetoepheno, water</td>
</tr>
</tbody>
</table>

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In the early days, improperly refined soybean oil that contained significant amounts of phosphatides produced fishy flavors on heating. This type of deterioration would be expected from the high content of polyunsaturated fatty acids of phosphatides. However, oxidized amino compounds may also play a role in flavor reversion, and 0.7% of nitrogen-containing compounds have been isolated from reverted soybean oil (Chang et al., 1961). Compounds with dimethylamino groups were indicated. More recently a multitude of oxygenated products expected from decomposition of linoleate and linolenate hydroperoxides were identified from bitter-tasting soybean phosphatidyl cholines (Sessa et al., 1977). Therefore, any small amounts of these phosphatides left in refined soybean oil would be expected, on oxidation and decomposition, to contribute to the off-flavor.

**PHOSPHATIDES**

These minor nonglyceride components of soybean oil were implicated in the early days (Mattil, 1947), but their role has not been fully determined. Tocopherols are found in relatively high amounts in soybean oil. The stability of soybean oil was improved by removing a portion of the tocopherols (Frankel et al., 1959). Decreasing the level of unsaponifiables may also improve flavor stability because their addition lowered it (Hoffmann et al., 1962). Although sterols had little effect, squalene lowered stability.
Pigments such as chlorophyll and carotenoids play a key role on the light stability of soybean oil, and this problem is discussed below.

LIGHT EXPOSURE

Light has a much more adverse effect on soybean oil than dark storage. The effect of light on flavor stability can be divided into two mechanisms.

Direct photochemical oxidation is due to free radicals produced by ultraviolet (UV) irradiation-catalyzed decomposition of peroxides, hydroperoxides, carbonyl compounds, or oxygen complexes of unsaturated lipids. This type of oxidation proceeds by normal chain reaction (Livingston, 1961). It can be inhibited or retarded either by chain-breaking antioxidants such as butylated hydroxy anisole (BHA) or butylated hydroxytoluene (BHT), or by UV deactivators that absorb irradiation without formation of radicals. Examples of UV deactivators include carbon black, phenyl salicylate, and 2-hydroxybenzophenone (Bailey, 1962). Whether or not these UV-deactivators can be used in foods has not been established. Normally, photochemical oxidation is of little concern, because the flint glass in ordinary jar glass filters out light below 300-325 nm and window glass cuts out light below 320 nm (Gudheim, 1943).

Photosensitized oxidation is due to oxygen activated to the singlet state by visible light in the presence of photosensitizers such as chlorophylls or certain dyes (Foote, 1968). This type of oxidation proceeds about 1,500 times faster than normal free radical oxidation (Rawls and van Santen, 1970), and only traces of photosensitizers would be sufficient to initiate autoxidation of unsaturated fats. Metals can also initiate fat oxidation by reaction with oxygen and produce activated oxygen (Uri, 1961). Deterioration of vegetable oils by activated oxygen can be effectively reduced by removal of natural photosensitizers during refining and bleaching. However, refining and bleaching also remove the carotenoids that act as quenchers of activated oxygen. Although carotenoids could be restored to protect the oil against this type of deterioration, the resulting yellow coloration may be objectionable to the consumer. There is growing evidence that sufficient amounts of photosensitizers are left in commercially refined, bleached, and deodorized soybean oil to contribute to its light instability (Clements et al., 1973; Frankel and Neff, 1979). One obvious approach to control this problem is to use suitable packaging or containers that absorb visible light necessary for photosensitized oxidation or prevent such light from reaching the oil. However, there is also evidence that precursors of activated oxygen can produce their deleterious effect in the absence of light (Frankel et al., 1979).

CONTROL MEASURES

Although much progress has been made in the control of flavor reversion, there are still problems caused by light and thermal instability in processed soybean oil. The greatest improvements have been achieved through the use of metal inactivators and through better processing, selective hydrogenation, and packaging. Other approaches investigated without too much success include selective removal of linolenate triglycerides (Schwab et al., 1950), prevention of oxidation by antioxidants (Frankel et al., 1959; Mounts et al., 1978), and breeding soybeans to produce oil low in linolenic acid (see Chapter 2 by E. H. Pryde).

METAL INACTIVATORS

The effectiveness of metal inactivators in stabilizing soybean oil against oxidation has already been mentioned. Many compounds have been tested (Dutton et al., 1948; Schwab et al., 1953), and the most commonly used metal inactivators include citric and phosphoric acids. These additives are very effective for improving storage stability at normal temperatures. Their effectiveness in soybean oil is increased by a synergistic action with the naturally occurring tocopherols (Frankel et al., 1959). On the one hand, tocopherols act as free radical-chain breaking antioxidants, and on the other hand, the metal inactivators act as preventive antioxidants. They mutually reinforce each other because they suppress both initiation and propagation of free radicals. Although metal inactivators may be used in early stages of processing to protect the oil against oxidation, they are more effective after the oil is heated (Cooney et al., 1958). Apparently, metal catalysts occur naturally in the oil in the form of prooxidant complexes with oxygen (Uri, 1961) or hydroperoxides, and these complexes have to be thermally destroyed before the chelating agents become effective. Therefore, addition of metal inactivator is generally recommended on the cooling cycle of deodorization.

PROCESSING

Minimum exposure of the oil to air and oxidation is essential. Any peroxide development during processing is detrimental to flavor and oxidative stability of the finished
Conjugation of pentadiene system I gives two conjugated diene-trienes (CDT):

\[
\begin{align*}
\text{CH}_3\text{CH}-(\text{CH})_2\text{CH}=\text{CH}-\text{CH}=\text{CH}-\text{CH}=\text{CH}\text{-R} & \quad 9,12,14-\text{CDT} \\
\text{CH}_3\text{CH}-(\text{CH})_2\text{CH}=\text{CH}-\text{CH}=\text{CH}(\text{CH})_2\text{-CH}=\text{CH}\text{-R} & \quad 9,13,15-\text{CDT}
\end{align*}
\]

Depending on conditions and extent of reaction, the distribution of isomeric monoens can be broader because isomerization of the double bond can occur again the same way through most of the length of the fatty acid chain.

With linolenate we may consider two pentadiene systems.

Conjugation of pentadiene system I gives two conjugated diene-trienes (CDT):

\[
\begin{align*}
\text{CH}_3\text{CH}-(\text{CH})_2\text{-CH}=\text{CH}-\text{CH}=\text{CH}-\text{CH}=\text{CH}\text{-R} & \quad 9,12,14-\text{CDT} \\
\text{CH}_3\text{CH}-(\text{CH})_2\text{-CH}=\text{CH}-\text{CH}=\text{CH}(\text{CH})_2\text{-CH}=\text{CH}\text{-R} & \quad 9,13,15-\text{CDT}
\end{align*}
\]

Conjugation of pentadiene systems II gives:

\[
\begin{align*}
\text{CH}_3\text{CH}-(\text{CH})_2\text{-CH}=\text{CH}-\text{CH}=\text{CH}(\text{CH})_2\text{-CH}=\text{CH}\text{-R} & \quad 9,11,15-\text{CDT} \\
\text{CH}_3\text{CH}-(\text{CH})_2\text{-CH}=\text{CH}-\text{CH}=\text{CH}(\text{CH})_2\text{-CH}=\text{CH}\text{-R} & \quad 10,12,15-\text{CDT}
\end{align*}
\]

The conjugated diene system in these CDT intermediates is preferentially hydrogenated, and the following products are formed:

\[
\begin{align*}
9,12,14-\text{CDT} & \rightarrow 9,12- + 9,14- + 9,13-\text{-dienes} \\
9,13,15-\text{CDT} & \rightarrow 9,13- + 9,15- + 9,14-\text{-dienes} \\
9,11,15-\text{CDT} & \rightarrow 9,15- + 11,15- + 10,15-\text{-dienes} \\
10,12,15-\text{CDT} & \rightarrow 10,15- + 12,15- + 11,15-\text{-dienes}
\end{align*}
\]

The methylene-interrupted dienes (9,12- and 12,15-) conjugate further into a mixture of 9,11- + 10,12- + 12,14- + 13,15- dienes. These conjugated dienes are readily hydrogenated by 1,2- and 1,4-addition into cis and trans monoens, with double bonds scattered between carbon-9 and carbon-15. The dienes that are underlined are nonconjugatable because the double bonds are separated by more than one methylene group—they are also called isolinoleate. Because they have two

HYDROGENATION

This step is crucial for maintaining flavor stability and forms the basis for the shortening, margarine, and soybean salad oil industries. Partial hydrogenation and winterization of soybean oil gives a liquid cooking oil that is widely used and accepted by the U.S. consumer (Erickson and Falb, 1980). However, this oil develops off-odors and flavors that are readily detected by trained personnel on heating (Evans et al., 1972) and light oxidation (Moser et al., 1965).

A practical catalyst has long been sought for selective hydrogenation of the linolenate contributing to flavor instability of soybean oil. The mechanism of catalytic hydrogenation will now be discussed briefly to provide an insight as to how flavor reversion develops even after selective reduction of linolenate. An important pathway of hydrogenation of polyunsaturated fats involves conjugation as an intermediate step (Frankel and Dutton, 1970). The conjugated dienes formed from linoleate are very reactive with hydrogen and rapidly become reduced to monoens. The main isomeric monoens produced in hydrogenated linoleate are shown by the following overall process:

\[
\begin{align*}
\text{Linoleate} & \quad \text{cis-9,cis-12, trans-10,cis-12, cis-9,trans-11, cis-9,trans-11, cis-9,cis-12} & \quad \text{Monoens} \\
_\text{trans-10} & \quad _\text{cis-12} & \quad _\text{trans-11} & \quad _\text{cis-11} & \quad _\text{cis-9} & \quad _\text{trans-11} & \quad _\text{cis-10} & \quad _\text{trans-10}
\end{align*}
\]

The last step in soybean oil processing is the deodorization, which involves heating at 392-525°F (200-274°C) in a good vacuum. If hydroperoxides or other oxidation products are present before this step, they will polymerize or condense and the resulting oxidation polymers and polar products are, as indicated before, detrimental to flavor stability. Good processing measures therefore, include careful control of refining temperature, vacuum bleaching (King and Wharton, 1949), nitrogen blanketing (Eselgroth, 1951), and protection from light exposure. Bleaching is known to lower oxidative stability of partially processed oil (Going, 1968). Oxidation can readily occur in this critical step by exposure to large surface area to air at elevated temperatures. Furthermore, bleaching may upset the natural balance between photosensitizers that lower stability and quenchers that increase stability.

The methylene-interrupted dienes (9,12- and 12,15-) conjugate further into a mixture of 9,11- + 10,12- + 12,14- + 13,15- dienes. These conjugated dienes are readily hydrogenated by 1,2- and 1,4-addition into cis and trans monoens, with double bonds scattered between carbon-9 and carbon-15. The dienes that are underlined are nonconjugatable because the double bonds are separated by more than one methylene group—they are also called isolinoleate. Because they have two
isolated double bonds which cannot conjugate, they are unreactive and accumulate in the hydrogenated products. They are also less reactive toward oxygen and behave like two oleate functions in the same molecule.

The pentadiene systems of linoleate and linolenate permit conjugation as an important intermediate step and account for the selective hydrogenation of polyunsaturated fatty acids in soybean oil. This selectivity for nickel catalyzed hydrogenation is in the order: oleate, 1; isolinoleate, 3; linoleate, 20; linolenate, 40 (Bailey and Isher, 1946). Two questions arise: (1) Why, in going from oleate to linoleate, does the introduction of one methylene group increase the rate of hydrogenation 20 times, whereas the introduction of a second methylene group, in going from linoleate to linolenate, merely doubles the rate? The relatively low reactivity of linolenate has been attributed to the formation of unconjugatable isolinoleate (Hilditch, 1946). The other question: (2) Why is the relative hydrogenation rate of isolinoleate 3 and not 2, as one might expect from a molecule with two oleate functions? The position of the double bonds on carbon-15 apparently makes it more reactive than that on carbon-9. This fact is supported by studies with model systems showing that cis-15-monoene was 1.3 to 1.7 times more reactive than oleate (Scholfield et al., 1971).

Although soybean oil is significantly stabilized by hydrogenating linolenate into isolinoleate, the pentene function of this product can oxidize to produce off-odors and flavors. Oxidation of isolinoleate produces hydroperoxides by oxygen attack on each double bond. If 9,15-diene is used as a model, addition of oxygen to the double bond on carbon-9 would produce the same mixture of compounds as oleate with the hydroperoxide radical (−OOH) attached to carbon numbers 8, 9, 10, or 11 (Frankel, 1962). Similarly, addition of oxygen to the double bond on carbon-15 would produce a mixture of compounds with the hydroperoxide radical attached to carbon numbers 14, 15, 16, or 17 (Fig. 1). Decomposition of these four hydroperoxides, however, would produce a mixture of aldehydes similar to those formed from linolenate (Fig. 2). Many of these compounds have been identified in nonhydrogenated soybean oil (Table 1). The flavor problem is further complicated by the decomposition products arising from the internal dienoic 8-, 9-, 10-, and 11-hydroperoxides.

$$\text{Fig. 1. Hydroperoxides produced by autoxidation of isolinoleate.}$$

$$\text{Fig. 2. Decomposition of isolinoleate hydroperoxides.}$$

These unsaturated aldehydes are very potent flavor compounds and impart the so-called hardening or hydrogenated flavor to...
soybean oil (Keppler et al., 1965, 1967).

One important development is the copper-bearing hydrogenation catalysts that have a linolenate to linoleate selectivity as high as 13 (Koritala and Dutton, 1966). It is possible with these catalysts to reduce most or all of the linolenate and still produce a higher yield of liquid oil during winterization. Soybean oil hydrogenated with copper catalysts had significantly better room odor and flavor scores than nickel hydrogenated soybean oil (Cowan et al., 1970). However, even when the linolenate content was reduced to 0% with copper catalyst, the flavor stability was not as good as that of a nonlinolenate vegetable oil. This problem arises from the isolinoleic acid produced in relatively high proportion (5 to 6%) by copper catalysts (Koritala and Dutton, 1969). The isolinoleic acid contributes a sufficient amount of terminal pentene radical to cause flavor deterioration.

Hydrogenation has, therefore, provided a practical but not complete solution to the flavor reversion problem of soybean oil. By converting linolenate into isolinoleate and monoenes, the stability of the oil is increased because these products are more difficult to oxidize. However, isolinoleate and monoenes with double bonds between carbon-14 and carbon-16 produce compounds similar to those of linolenate and impart objectionable odor and flavor compounds on thermal oxidation. The ultimate solution is to find a catalyst that can selectively reduce the 15-double bond of linolenate to convert it to linoleate. Much of our research on heterogeneous and homogeneous hydrogenation catalysts (Frankel and Dutton, 1970) has been directed toward this goal.

SUMMARY

Much evidence has been reported in support of the conclusion that the flavor deterioration in soybean oil is caused mainly by linolenate autoxidation followed by decomposition and polymerization. The problem is complicated, however, because other factors contribute to off-flavors in oxidized soybean oil. For example, low concentration of linolenate hydroperoxides can catalyze the autoxidation of linoleate, the predominant unsaturated fatty acid in soybean oil. Minor constituents such as phosphatides, tocopherols, squalene, chlorophylls, and carotenoids can apparently have either beneficial or detrimental effects on flavor depending on their relative concentration. Recent research points to photochemical processes involving activated oxygen as another important pathway for generating precursors of undesirable odors and flavors in soybean oil. There is also growing evidence that oxygenated compounds can produce activated oxygen by the same mechanism as photochemical oxidation. If sufficient amounts of these oxygenated compounds develop in soybean oil, this type of oxidation can conceivably occur without catalysis by light.

In the processing of soybean oil for edible purposes, in addition to keeping the oil away from light and air, there are critical steps that should be improved including refining, bleaching, deodorization, and hydrogenation. Improved flavor stability can be expected if bleaching can be made more selective in order to increase the ratio of carotenoids over chlorophylls. To reduce the problem caused by oxidative polymers, deodorization of soybean oil should only be carried out either after ensuring that the oil is free of peroxides or after their removal. Finally, hydrogenation needs to be more selective toward the 15-double bond of linolenate. Such processing improvements will help to maintain soybean oil as the dominant edible oil of the world and will be of considerable economic importance. Further improvement in the air, heat, and light stability of edible soybean oil can only be achieved by additional research and development.

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