



## Trends in the Chemistry and Technology of Lipids

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### ABSTRACT

A review is given of the annual world production of fats and oils and their increasing availability for edible and nonedible purposes. Trends in some important industrial processes (polymerization, metathesis, oxidation, etc.) are discussed and the applicability of fatty oils as specific and characteristic base materials for the organic chemical industry is compared with developments in the field of petroleum synthetics. To improve the position of fat-intermediates and fat derivatives on the world market, future research should be particularly directed to an optimal use of their specific physical, chemical and biochemical properties, and to an increased coherence between our technological and biochemical knowhow.

### PRODUCTION AND WINNING

The primary use of fatty oils is closely related to man's need of food. Fats and oils are irreplaceable components of the daily food parcel: in the Western countries they even contribute to ca. 40% of the caloric demand (1). It is easily understood, for that reason, that modern agricultural developments have been particularly stimulated by the intensive research of food scientists and biologists.

Thus, the discovery of linoleic acid as an essential fatty acid in human food in the prevention of distinct heart and vascular diseases has promoted the production and application of vegetable oils with high linoleic content (such as soybean and sunflower oil), particularly in the United States but also in other countries. Another example of the impact of biological research is the growing evidence of the role of erucic acid in promoting distinct disorders of heart, kidney and liver. This has not only led to a voluntary reduction in use of rapeseed oil (in which erucic acid is present in amounts up to ca. 45%), but also to the successful cultivation of rapeseed varieties with much reduced erucic acid content (2-5), a fascinating result of genetic change of the composition of oils, indicating unpredictable possibilities of adjusting the production of specific fatty oils to specific needs of their consumers. Similar results have been obtained in the genetic elimination of linoleic acid from safflower oil (6). There seems to be no doubt that future developments in this area will be of crucial importance. Stein (6) points to the possibility of building short-chain fatty acids (e.g., lauric, myristic) — which only occur to a limited extent in nature (see Fig. 1) — in common oils; this certainly would largely influence the agricultural development in many countries.

Although it seems that future trends in the production of edible oils of different types thus will be stimulated to a greater extent by biological than by technological motives, it should be stressed that chemical and mechanical methods to improve the fertility of the soil and to cherish and protect the ripening crops, and appropriate facilities for storage, handling, transport, manufacture and distribution of the products, are as indispensable to obtain optimal yields of fats and oils as are the results of selective breeding and the introduction of new species. In many developing countries the percentages of the crops that get lost between harvesting and consumption amount to not less than 20-40% (7). On more than 70% of the agricultural land of the world, farm management is very primitive and the yields are not more than 20-30% of what can be produced with the aid of modern agricultural methods (8).

Figure 2 represents data of the annual world's production of fats and oils during the last 15 years (9). It is obvious that the gradual increase is mainly a result of the intensification of soybean and palm type oils. Palm oil is even expected to become the major contributor to the growth in edible oil supplies in the near future (9). The world production of soybean oil has doubled since 1968 as a consequence of the world's increasing need for proteins. Soy meal contributes as much as 65% to the world production of oil cake and meal (100 million tons in 1982). Animal fats show practically no growth, a few percent only in the last decade.

The increase of the total annual world production of oils and fats exceeds the growth of the world population in such a way that the yearly world production per capita gradually increased from 10.5 kg in 1953 to 11.4 kg in 1968 and to 13.4 kg this year. Deviations from the average, however, are very wide in different countries. Figures vary from ca. 5 kg in India (and even less in China) to ca. 25 kg in Western Europe and 35 kg in the United States. Actually, the world as a whole suffers from a chronic shortage of fats,

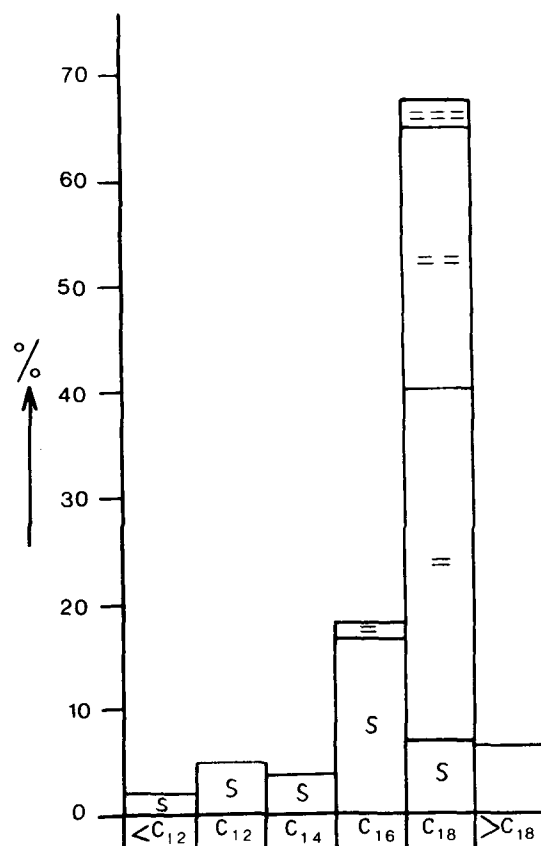


FIG. 1. The occurrence of fatty acids of different chain length and different degree of unsaturation in the 1979 world production of natural oils and fats (according to Stein (6)).

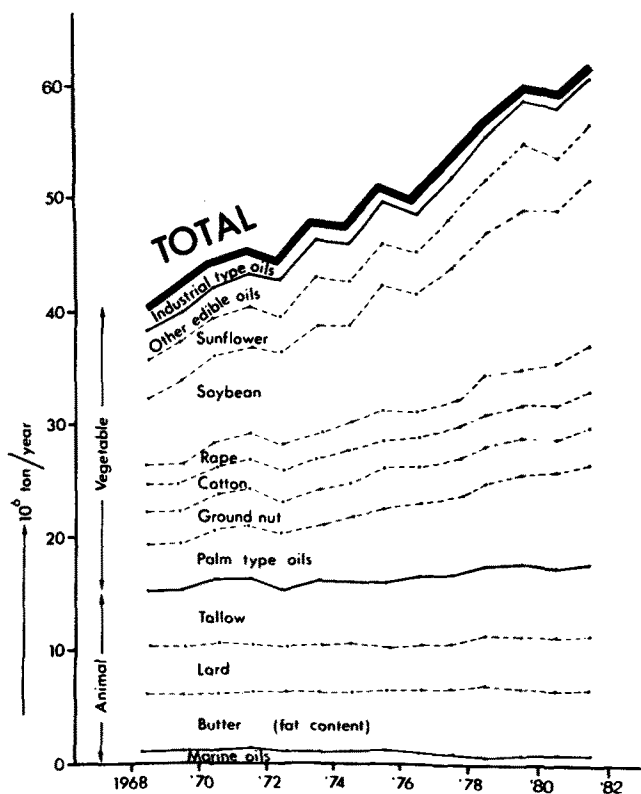


FIG. 2. Annual world production of fats and oils (9).

and the amounts, available for nonfood uses, are restricted, particularly in the Third-World countries. In the USA, not less than 35-40% of the available oils and fats are used for the manufacture of soaps, paints, varnishes, resins, lubricants, etc. In India this percentage is less than 5.

The yearly production per capita of nonedible fatty oils shows a gradual decrease. This does not hold so much for tallow, the main representative of animal origin, as for the so-called "industrial type oils": linseed, tung, oiticica, perilla and castor oil, apparently as a consequence of the still growing importance of petroleum synthetics in the paint and varnish industry. Industrial fatty oils represent ca. 15% of the total world fatty oil production, 80% being from animal origin (mainly tallow).

### FATS AS RAW MATERIALS FOR THE CHEMICAL INDUSTRY

The direct use of natural fatty oils for nonedible purposes is limited to relatively small applications of drying and semi-drying oils in the paint and varnish industry. Much larger amounts are transformed by chemical processes into basic materials for important industries: saponification, for the soap industry; hydrolysis, for the manufacture of fatty acids and glycerine; polymerization and copolymerization, in the paint and varnish industry; interesterification with glycerine, to prepare mono- and diglycerides as intermediates for the manufacture of oil modified alkyd resins; and hydrogenation, in the fat hardening industry and for the production of fat alcohols.

The main basic intermediates in the fatty oil industry are fatty acids and fatty acid esters, obtained by hydrolysis or alcoholysis of proper oils. In 1979, the production of fatty acids amounted to 1.7 million tons in the United States and 0.6 million tons in Europe, mainly: stearic, chiefly from tal-

low; oleic, from different sources; lauric, from coconut, palm kernel, babassu; linoleic and linolenic, from soybean, safflower, linseed and tall oil.

Important sources for the production of fatty acids are not only fats and oils, but also tall oil, an oleic-linoleic acid containing byproduct of sulphate "kraft" pulping in the paper industry. World tall oil production amounts to not less than 1 million tons per annum.

Table I shows the relative importance of the various fields of application (6).

TABLE I

Nonfood Applications of Fats and Fatproducts in 1979 (6)

Products	Percentage
Soaps	45
Detergents and surfactants	19.5
Surface coatings, plasticizers, rubber	18.3
Metal soaps, greases	7.9
Food additives	6.3
Cosmetics	3.0

Potential raw materials — fatty acids with unsuspected specific properties and possibilities — are present as main components of numerous less known oils and fats. Among them are cyclic acids (in chaulmoogra, hydnocarpus and gorli oils), acetylenic acids (e.g., isanic acid in boleko oil), polyunsaturated acids in a variety of fish oils, unsaturated wax esters (in jojoba oil), etc. A study group of the National Academy of Science in Washington listed numerous "under-exploited" tropical plants with promising economic value, of which further chemical, technological and agricultural research might result in valuable applications (10). Potential lipid sources are also present in algae and seaweeds (11). Use of yeasts and molds to produce microbial oils suitable for a variety of applications, has also been recognized as a realistic proposition (12,13).

On the other hand, it should be noted here that several scientists are not at all convinced of an unlimited productive capacity of our biosphere and the possibilities of larger scale application of agricultural products and byproducts for industrial purposes. According to Buringh (8), as a consequence of human activities, not only is the net primary productivity of the world land area at present not more than 70% of what it was in prehistoric times, but further, it is still rapidly decreasing, mainly due to primitive farm management methods on most of the available agricultural land and to the results of primitive, large-scale reclamation of new land, leading to deforestation and destruction of important natural resources. Although it is difficult to rate Buringh's arguments at their true value, they stress the necessity to base future strategies on realistic agrieconomic considerations and to avoid short-term would-be solutions that on a larger term might be disastrous.

### AGRICULTURAL AND PETROCHEMICAL RAW MATERIALS

#### Fatty Acids

The main sources for the manufacture of fatty acids — principal raw materials in the fat industry — are in fact surplus products: tallow from the meat and tall oil from the paper industry. Actually, even soybean oil, the most important fatty oil from a quantitative point of view, is a byproduct when we realize that the cultivation of soybeans is based

primarily on the need for proteins. Up to now this has made the position of these oils and their fatty acids (mainly  $C_{18}$ ) rather strong from an economic point of view and hampered synthetic fatty acids from petroleum feedstocks to oust agricultural acids, notwithstanding heavy efforts of the petroleum industry to enter the field.

The annual world production of synthetic fatty acids amounts to ca. 500,000 tons (14), chiefly produced in the Soviet Union by oxidation of paraffin wax. This process is particularly directed to terminal, nonbranched acids in the detergent range ( $C_{10}$  -  $C_{16}$ ), but yields also considerable amounts of lower and higher molecular products for which the market is restricted. Much more advanced and highly versatile are different versions of the Oxo-process (15): hydroformylation (addition of carbon monoxide and hydrogen) of distinct olefins and oxidation of the aldehydes formed, and hydrocarboxylation (addition of carbon monoxide and water).

Selective catalysts have been developed for these processes that promote the formation of linear above branched components (16,17). Several industries are active in developing markets for synthetic acids of a different type. Up to now, however, the production of these synthetics is of limited importance (Table II); they fail in the competition with the highly specific and easy obtainable high grade and pure products of agricultural origin. Moreover, their production is restricted to saturated acids. The unique position of the various natural unsaturates and their significance as base materials for a variety of chemical processes is practically unassailable.

#### Fat Alcohols

Much stronger than in the field of fatty acids is the position of the petrochemical industry in the fat alcohol area, par-

ticularly in the United States, where it accounts for ca. 85% of the total production (Table II).

In the fat industry, alcohols have been produced for many years by high pressure hydrogenation of fatty acids or fatty acid esters (18,19). Catalysts have been developed that are very selective, also in saving the double bonds of unsaturated starting materials (20-22). Fat alcohols have been developed to important intermediates for the manufacture of a large variety of surface active agents of different type (alkyl sulphates, polyglycoethers and polyglycoether sulphates) and have also been used as such as plasticizers and in cosmetics.

The leading position of petrochemical fat alcohols in the United States has been stimulated strongly by the development of advanced large-scale building-up processes of simple basic unsaturated building stones, of which particularly ethylene and propylene should be mentioned. In Figure 3, two important routes have been indicated for the manufacture of  $C_{10}$  -  $C_{14}$   $\alpha$ -olefins: the Ziegler-route and SHOP (Shell Higher Olefin Process) (6). According to Ziegler (23-25), the metalorganic intermediates, formed during the polymerization of ethylene in the presence of organic aluminum complexes, can be transformed directly by oxidation into aluminum alcoholates and, via subsequent hydrolysis, into *n*-alcohols. In the SHOP process (26-28), use is made of the recently developed metathesis reaction (29) to transform higher molecular oligomerization products of ethylene (after their isomerization to internal olefins) into linear iso-olefins of the desired detergent range. The metathesis process involves a *trans*-alkylidene reaction of unsaturated molecules, resulting in a completely statistical distribution of the alkylidene-moieties over the reactants. When using an excess of ethylene it is thus possible, in principle, by a proper combination of fractionation, isomerization and

TABLE II  
Production of Important Fat Derivatives in 1979 (6)

	Fatty acids		Fat alcohols		Fat amines and amides (nearly completely agricultural) (1000 tons)
	Agricultural	petrochemical (1000 tons)	Agricultural	petrochemical (1000 tons)	
USA	1020	50	54	280	96
Western Europe	630	7	140	70	64

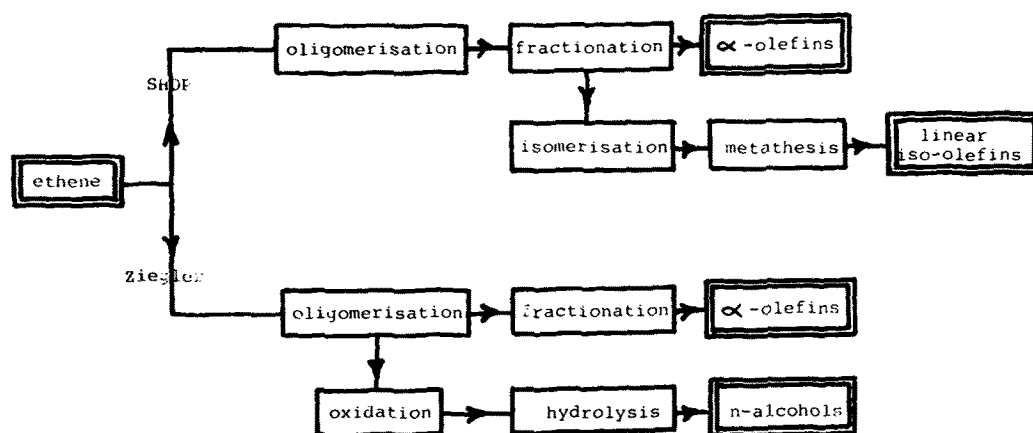


FIG. 3. Large-scale processes for the conversion of ethylene into linear olefins and alcohols (according to Stein (6)).

metathesis to transform mixtures of linear olefins completely into linear  $\alpha$ -olefins of distinct chain length.

Linear olefins in the  $C_{10}$  -  $C_{14}$  range have not only been used for the industrial production of fat alcohols (by hydration or hydroformylation) and detergents (alkene sulfonates and hydroxyalkane sulfonates (30)) but also, via epoxidation, for the synthesis of a large variety of other valuable derivatives (Fig. 4).

In the competition between oleochemical and petrochemical derivatives, the cost prices of the basic raw materials are, of course, of primary importance. Although price comparisons and price projections of mineral and fatty oils are hardly possible, it can be stated that — notwithstanding large fluctuations due to the climate-sensitive yields of the slightly tenable crops — on a larger term, the price development of fatty oils shows a more regular pattern than that of mineral crude oils. The increasing yearly production per capita of these renewable resources is in this respect a strongly stabilizing factor.

Mineral oils are highly subject to political situations and the decreasing resources have led to unprecedented rises in price — in the course of only 8 years with a factor of not less than 15. This has brought the prices of mineral oils and fatty oils closer together than ever before (Table III), a trend that, of course, stresses the importance of oilseeds as sources for the chemical industry. Nevertheless, it certainly does not motivate speculations as to a possible large-scale use of vegetable oils as substitutes for diesel oil or other fuels as has already been suggested (32,33).

The amounts of mineral oils, used in the petrochemical industry, are not more than a small fraction (ca. 5%) of the total mineral oil production. A possible decrease of this production in the future, therefore, will not directly influence their availability for the manufacture of petrochemicals, certainly not for those with a relatively high added value. It is not unlikely that the necessity to explore and apply other sources of energy in the future will even stimulate the petroleum industry, supported by their unique knowhow and their outstanding and well equipped research institutes, in finding new processes and product applications for the most optimal exploitation and use of their nonrenewable resources.

For the fat industries, it is of utmost importance to develop the utility of those physical and chemical properties of their natural raw materials that are specific and characteristic, and even irreplaceable: the strictly linear and even carbon-numbered chains with very definitely located and orientated double bonds in the unsaturates among them. The tendency of petroleum and natural gas to become shorter in supply and the much more stable position of the renewable fatty oils in this respect will sufficiently favor the significance of the latter as indispensable resources in the forthcoming decades to motivate strong efforts in research and development for their further applicability.

## PROCESSES

With all the differences, mineral and fatty oils show remarkable similarities. Important chemical transformations: polymerization, hydrogenation, hydrocarboxylation, oxidation, etc., require comparable process conditions and corresponding technical knowhow, adapted, of course, to the specific qualities of the proper base materials. It is important that fat technologists and petroleum scientists do not compete, but cooperate, and stimulate each other in finding the optimal routes that will be necessary to face future demands and developments in the field of industrial organic processes.

Throughout the years of industrial development, the fat industry has contributed considerably to chemical and tech-

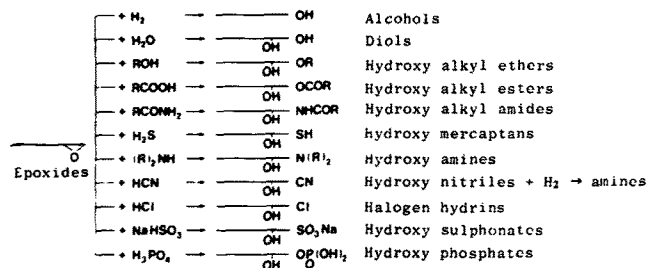


FIG. 4. Epoxides as intermediates for a variety of fat derivatives (according to Stein (6)).

TABLE 3

Current UK Prices of Some Plant Oils and Fats (31) (June 1981)

Product	Price (£/ton)
Cocoa butter	2,200
Olive oil	1,450
Groundnut oil	810
Tung oil	700
Corn oil	480
Soybean oil	335
Rapeseed oil	325
Arabian light crude oil	\$ 34/barrel (159 L)
	~ £ 135/ton

nological innovations. In the first place, of course, it has contributed in the specific field of soap and detergents, but furthermore in several other industrial areas. The polymerization of linseed oil, invented by a Dutch house-painter, is one of the oldest industrial processes, dating back to ca. 1800 (34). The first patents, concerning the copolymerization of drying oils with styrene, date from 1901 (35), large-scale applications had to wait until industrial amounts of styrene became available. The hardening of fatty oils was the first catalytic hydrogenation process adapted to commercial operation. It has stimulated further research of heterogeneous catalysis, resulting in other industrial hydrogenation processes such as the manufacture of saturated and unsaturated fat alcohols and of fat amines. The ozonolysis of oleic acid (36) to produce pelargonic and azelaic acid as valuable intermediates is the only large-scale application of ozone in the organic chemical industry.

Important chemical conversions with great potential possibilities for the fatty oil industry are hydroformylation and hydrocarboxylation, distinct polymerization reactions, and metathesis. Also, biosynthetic methods will gain in importance in the near future.

## Hydroformylation and Hydrocarboxylation

Hydroformylation and hydrocarboxylation of unsaturated esters have been realized on a laboratory scale with recently developed very specific homogeneous catalyst systems. Thus, methyl oleate has been converted nearly quantitatively by reaction with CO and  $H_2$  in the presence of rhodium triphenylphosphine at ca. 110 C and 50-150 atm. into an equimolar mixture of methyl-9 and methyl-10 formyl stearate.

From starting materials containing linoleic or linolenic esters also diformyl and triformyl stearates are formed. Formyl stearate can be easily oxidized to corresponding acids, or be transformed into acetals, hydroxy compounds,

amino compounds etc. Hydrocarboxylation has been achieved with  $\text{PdCl}_2$ -triphenylphosphine as a catalyst.

Linoleic and linolenic acids are converted in this way into tri- and tetrafunctional acids. In the petroleum industry, corresponding processes have already found application for the conversion of olefins. The fat industry can take advantage of the available know how when proper applications for the polyfunctional derivatives can be found.

### Polymerization

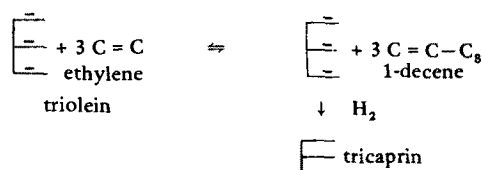
The polymerization of drying oils, containing polyunsaturated fatty acids as linoleic and linolenic, has long been one of the most valuable processes in the drying oil technology and laid the basis for the modern paint and laquer technology.

Very promising are also modern processes to dimerize mono-unsaturated fatty acids with acid catalysts of the montmorillonite type (37). The main reaction is a Diels-Alder condensation of conjugated dienoic acid molecules, formed intermediary by hydrogen transfer between two oleic acid molecules, with a third oleic acid molecule, resulting in monocyclic dimers. Dimeric acids of this type are of increasing importance for the manufacture of polyesters, polyamides and alkyl and epoxy resins with outstanding chemical, physical and mechanical properties.

### Metathesis

In 1974, Van Dam succeeded in the metathesis of unsaturated esters (38-40). The importance of this reaction is illustrated by the conversion of methyl oleate into equimolar amounts of 9-octadecene and 9-octadecene dioic methyl ester, using  $\text{WCl}_6$ - $\text{Sn}(\text{CH}_3)_4$  as a homogeneous catalyst. At 70 C, equilibrium is reached within 1-2 hr. The unsaturated dicarboxylic acid is an interesting starting material for the manufacture of unsaturated polyesters and polyamides (41), and for the synthesis of civetone, a basic material in the perfume industry (42). Corresponding reactions have been performed with a large variety of unsaturated fatty esters, including fatty oils such as olive and linseed oils.

Cometathesis of unsaturated esters, e.g., with unsaturated hydrocarbons, occurs also and has opened the route to a variety of products that often hardly can be synthesized by other methods (43,44). Thus, the synthesis of various pheromones (sex attractants) has been realized by metathesis (45,46). Very promising are cometathesis reactions with ethylene (ethylene cleavage). They offer a highly selective method for the transformation of distinct long-chain fatty acid glycerides into glycerides of the, from an industrial point of view, more important  $\text{C}_{10}$  -  $\text{C}_{14}$  acids (47). Along this line, triolein, the main component of olive oil, can be transformed by ethylene cleavage and hydrogenation into tricaprln:



1-decene is formed as a valuable byproduct.

In the same way erucic, esters can be converted into esters of myristic acid. Due to the mild process conditions, these processes are completely selective.

Recently also heterogeneous catalyst systems with a more pronounced activity have been developed (48,49). Their activity, however, is still small in comparison with catalysts developed for metathesis conversions of nonfunctionalized compounds. Industrial applications of the meta-

thesis of fatty oils will largely depend on further improvement of these catalyst systems.

### Biosynthetic Methods

Since several enzymes have become available in good quality and in relatively large quantities, more attention has been paid to their utility as selective preparative reagents, and, in distinct cases, even to commercial applications. Their highly specific and often also stereospecific activity at moderate temperatures offer many advantages above other chemical methods. In this respect the close relationship should be noticed between enzymatic and catalytic processes as to activity and specificity in dependence of the nature of the active spots, the structural properties of the substrate, the influence of coenzymes (resp. cocatalysts) and the possibility of immobilization on inert inorganic or organic carriers (50).

Examples of biosynthetic reactions are the use of lipolytic enzymes for the synthesis of distinct mono- and diacyl glycerols (51,52) and phospholipids (51-54), compounds that are very difficult to prepare by commercial means. On an industrial scale lipoxygenase present in soy flour can be used for the oxidation of linoleic acid. With an aqueous soy flour extract, the linoleic acid content of soybean soapstocks is transformed into hydroperoxides that can be easily reduced in situ by sodium bisulfite into hydroxy-conjugated octadecadienoic acids (55). These acids are valuable intermediates for the production of castor oil and tung oil substitutes.

### CONCLUDING REMARKS

In this review, only a few headlines have been indicated. Nevertheless they show the important role of the fatty oil industry in initiating and stimulating new developments. In the beginning of this century these developments — results of a breakthrough in catalytic organic synthesis — primarily served to promote the interchangeability of natural oils and fats (fat hardening), and to improve distinct chemical and physical properties. In later years, the manufacture of fat chemicals by more drastic chemical transformations extended the scope of applications across the entire industrial horizon. Future trends will definitely be influenced largely by the progress in biomolecular chemistry and biotechnology, and lead to increasing coherence between technological and biological methods.

Industrial processing of fats and oils for nonedible purposes has never reached the large-scale level of the manufacture of distinct petroleum base chemicals, which, of course, is due to their much more restricted supplies, their wide variety of raw materials and their relatively high price. On the other hand the production of oils and fats as renewable resources and irreplaceable foodstuffs warrants a relatively stable supply of their byproducts for nonfood applications. The increasing world production of oils and fats will improve the position of fat intermediates and derivatives, particularly those, in which advantage has been taken from their specific chemical and physical characteristics.

Several versatile processes, developed by the petrochemical industry, offer promising perspectives in the fatty oil area; it is of the utmost importance for petroleum and fat scientists and technologists to cooperate in facing the world's expanding need for chemicals from the available and from renewable sources.

Further intensification of the cultivation of proper oils is a challenge for many countries to provide in their shortage of essential food elements (fats and proteins) and to contribute according to ability to the world's future supply of important and indispensable fat chemicals.

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## Summary of Discussion Session H-1 on Future Trends

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Discussion session chaired by A.R. Baldwin and K.F. Gander; the panel consisted of Messrs. Boelhouwer, James, Mounts and Wiedermann.

The major interest during the discussion on long-term trends centered on palm oil and on the potential of biotechnology to affect the fats and oils industries.

Lars Wiedermann was asked for more details on why he did not think palm oil's share of world trade would increase as rapidly during the next few decades as would the market share for soybean, sunflower or rapeseed (see plenary talk). Wiedermann listed several reasons including: (a) the need for more profit-making refinery capacity if refined palm oil is to be sold on world markets, particularly as crude palm does not travel as well as refined palm oil; (b) lack of mar-

kets for palm oil byproducts compared to those of competing oilseeds; and (c) the relative narrow portion of prime liquid oil produced by crude palm oil fractionation.

Kurt Berger of the Palm Oil Research Institute of Malaysia said he expected more growth for palm oil than did Wiedermann, in part because palm is a perennial crop whereas soybeans, sunflower and rapeseed are annual crops whose production may fall dramatically in any given year if economics induces growers to plant other crops. Palm oil acreage, once planted, will continue producing for decades despite price fluctuations, Berger said. He added that the tissue culture cloning described by James (see plenary talk) and initial results in using weevils to increase fresh fruit bunch production may increase palm oil supplies at a greater rate than anticipated.

Tony James was asked about the possibility of using biotechnology to modify characteristics of oils from