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Significant Advances in Fat and Oil Chemistry in France During the Past Fifty Years[1]

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DURING the past 50 years or more, specifically since 1908, significant progress has been made in the chemistry of fats and oils in France in both research and industry. In 1908 the extent of chemical research was extremely modest; a couple of professors studied fats from time to time and very few industries even had a control laboratory at their disposal; those which existed were devoted solely to the commercial control of products. At that time plants simply consisted of oil mills and soap factories that operated mainly on a small scale. Today there are several important governmental research laboratories, and many industrial companies have research as well as control departments. Plants are definitely more important and operate by scientific methods. New industries have been created: cake treatment, detergent industry, lipochemistry, etc.

Progress was slow but became very significant after the second world war. To show the tremendous growth during the last 50 years it would be interesting to quote some statistics. Unfortunately they are quite difficult to compute for the first 25 years since most of the oil and soap plants were small enterprises, which generally utilized local raw products and sold their merchandise in their own area. It is helpful to consider the case of peanut oil in this connection.

Today the most important edible oil in France is imported peanut oil; in 1957 it represented 70% of the fluid edible oils. (The remainder consisted of olive oil, 8%, rape seed oil and others, 22%.) Our principal source of peanuts is Senegal, and we are practically their only customer. Therefore Figure 1, which gives the global exportations of Senegal peanuts, reckoned with hulls, is particularly significant. As can be noted, the production has increased regularly except for the periods of the two world wars when exportations decreased significantly; in 1932 the depression is also very obvious.

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[1] The original paper contained numerous photographs and charts which it is not possible to reproduce. Edited by Waldo C. Ault.

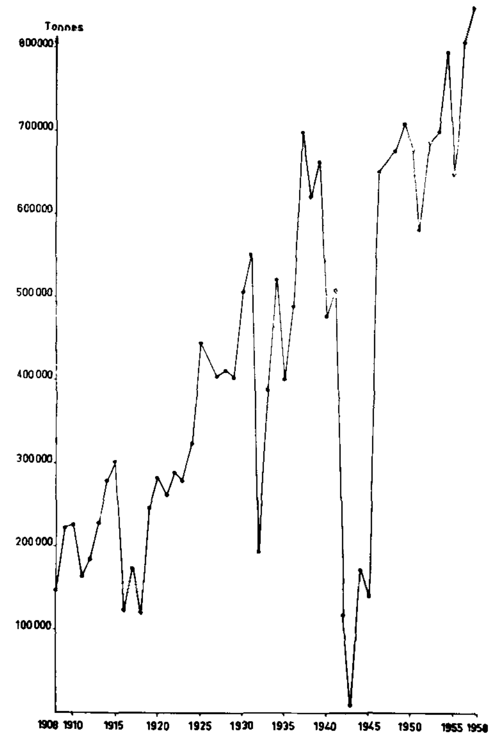

FIG. 1. Shell peanut exports from Senegal.

In southern France we have many olive-tree plantations. The average production has been approximately constant, totalling 7,000 tons per year, with maxima of 10,000 tons and minima of 5,000 tons. However since 1956, when the production suffered great losses because of the frost, the production has decreased to only 800 tons in 1957 and 2,000 tons in 1958. On the contrary, Tunisia's average production of olive oil has increased from 20,000 tons in 1910 to 26,000 in 1925, and to 60,000 in 1935. It is presently about 90,000 tons (with a maximum of 105,000 tons in 1949 and a minimum of 28,000 tons in 1955).

We import palm oil and palm kernels mainly from African territories (Cameroun, Dahomey, Belgian Congo). France has made a great effort to develop the production of palm oil in her overseas possessions. Special attention has been given to improving the extraction conditions and to building modern oil mills and thereby to obtaining a better quality of oil with low acidity. Today the Dahomey palm-oil production is roughly 20,000 tons per year, and it is progressively increasing; in Cameroun the production is only a couple of thousand tons per year.

Among the industrial products of fats, soap is the most important one. Before the second world war the production was about 350,000 tons per year. The figure decreased to 80,000 in 1944, then progressively increased to its former volume. During this period the production of surfactants increased regularly. The following quantities of detergents were produced in France in 1958:

Fatty alcohol sulphates, 30%	23,000 tons
Oxyethylamide sulphates, 20%	1,500 tons
Alkyl sulfonates, 25%	87,000 tons
Nonionics and cationics, 100%	2,500 tons

Oil Production in France

Around 50 years ago the oil industry in France was still similar to that of the previous century. Most of the oil mills were concentrated in Marseilles because the oil seeds from abroad arrived in this harbor. In addition to these relatively important oil mills, there were several local oil mills treating small quantities of seeds or fruit.

The only extraction process used was based on the hydraulic press, and only a few oil mills performed alkali refining. It was around 1908 that the first deodorizing process was carried out. The oils were first treated by steam at atmospheric pressure, then under reduced pressure; finally superheated steam was injected. The first introduction of copra on the French market in 1906 led to deodorization because those oils had a flavor that was too pronounced. The presses used with cloths were progressively replaced by cage presses. Later screw presses or continuous presses of the V. D. Anderson type were used simultaneously.

There were some early attempts to use carbon disulfide (Deiss patent of 1855) and trichlorethylene for the extraction of olive cake, but expansion of this technique awaited the use of petroleum solvents. For many years the only apparatus used for solvent extraction was of a discontinuous type. It was only about the time of the second World War that the first continuous extractors made their appearance. Today the great French oil mills operate with continuous apparatus of different types; one of the most satisfactory types is the de Smet apparatus. It should be noted that several large oil mills were bombed during the second World War and have been rebuilt with modern equipment.

As noted, it was about 1908–1910 that the refining of fats began to give satisfactory results. Progress was achieved slowly, and by the '20's refining was used regularly. Neutralizing, bleaching, and deodorizing apparatus were always of the batch type. Since 1945 the techniques of continuous refining have been worked out progressively, especially according to the Alfa-Laval and Sharples processes. For example the growth of one of the most important French societies in edible oils, Lesieur, initiated in 1908, is illustrated by their production of refined oils: 1911, 18,500 tons; 1925, 51,000 tons; 1938, 109,300 tons; 1950, 129,300 tons; and 1958, 396,000 tons.

The Margarine Industry

Besides oil mills, margarine plants also existed in France. Until 1900 the only raw material was oleo oil, obtained by the pressing of beef fat. To this oleo oil were progressively added various vegetable oils in small proportions. Cottonseed oil of American origin was used at the beginning, then coconut oil after development of the deodorizing process. Shortly thereafter margarines began to appear on the market which were exclusively vegetable.

Great progress was made in this field about 1910. Normann devised a practical method for hydrogenation of fats, based on the great discovery of the French scientists, Sabatier and Senderens. Thus was created the industry of oil hydrogenation. The first industrial hydrogenation equipmen was constructed in Marseilles. It was very well devised indeed for I saw it a couple of years ago still being used regularly with satisfactory results. The development of hydrogenation permitted the margarine plants to use many kinds of raw materials.

The technique of margarine preparation has been improved greatly during the 50 years. The wooden ripening vats of Mege-Mouries were quickly replaced by metallic drums with double walls. Once the emulsification is made, the material is cooled by a stream of ice-cold water, of which large quantities are required. Among the early processes developed was one in which a jet of emulsion and a jet of ice water were mixed; the mixture of emulsion and ice water was then decanted, after which the margarine was drained, before being plasticized. Since about 1920 the first cooling drums have been progressively improved and they are still used. Now however continuous apparatus of the Votator type is also utilized.

The annual production of margarine in France, 75%, from Astra of the Unilever group, was:

Year	1910	1930-40	1944	1945	1950	1954	1957
Tons	20,000	40,000	8,500	37,000	53,000	84,000	95,000

The Soap Industry

Fifty years ago the French soap industry was not very scientific. The only technique used to produce soap was the so-called "Marseilles technique." Soap was manufactured in open kettles from different mixtures of raw material without much use of scientific knowledge. Soap (at 72%) was spilled directly on the floor from the kettle into "mises" where it was left for two or three days to solidify. The bars of soap were then loosened, cut, and framed.

The quick cooling of soap in Buhler presses was developed later and competed with the "mises" technique. In 1940 both processes were used simultaneously; the Marseilles plants still use the "mises" process.

After 1945 new continuous processes of cooling appeared as well as moulding of the Mazzoni type. Today these processes are the most popular in France. It should be noted that the first soap flakes appeared in 1912 and that the first powder soaps date from around 1930.

The greatest contribution in France to the soap industry was the development of an entirely continuous saponification process called the Monsavon process. This development was carried out by a group of technicans and chemists of the Monsavon Company, notably Lachampt and Seemuler. After long laboratory studies a pilot plant for continuous saponification was constructed in 1945–46, and the first industrial installation was built shortly after. Today the Monsavon process is very well known, and similar plants have been built in all parts of the world. It has been estimated that the present yearly production of soap by this process is about 500,000 tons.

The Monsavon process may be outlined as follows (Figure 2). The fat and an aqueous soda lye are emulsified hot (in H); then this emulsion descends into a warm reactor in which the saponification begins; then into vat (C) in which it is held for about 30 min., the time necessary for complete saponification. The lye strength is controlled so that the soap going out of C comes into the neat phase and so that the aqueous lye into which it is mixed contains 0.2% of free soda. This mixture is then sent to the washing tower (L), which is generally constructed in six stages. The soap ascends in this tower and is washed countercurrently. There is an agitator and settling zone in each stage. Since the washing is done with a limited amount of lye and at the temperature of 85°, the decantations are very quick. The mechanical disposition of each washing stage is extremely important, and smooth working of the operation depends on it. After the last stage of washing the soap is sent to a vertical fitting box (B), in which it receives the quantity of water and lye necessary for its fitting. The fitted soap is finally sent into a settling pan to be separated from its nigre. This nigre is returned to the upper stage of the washing tower at the same time as the new lye. In general there are two settling pans; one is filled while the other empties.

The final soap is of excellent quality, with 63% of fatty acids, less than 0.2% of unsaponified matter, and at the maximum 0.1% of free alkali determined as Na_2O. The most common size of unit produces 2 tons per hour.

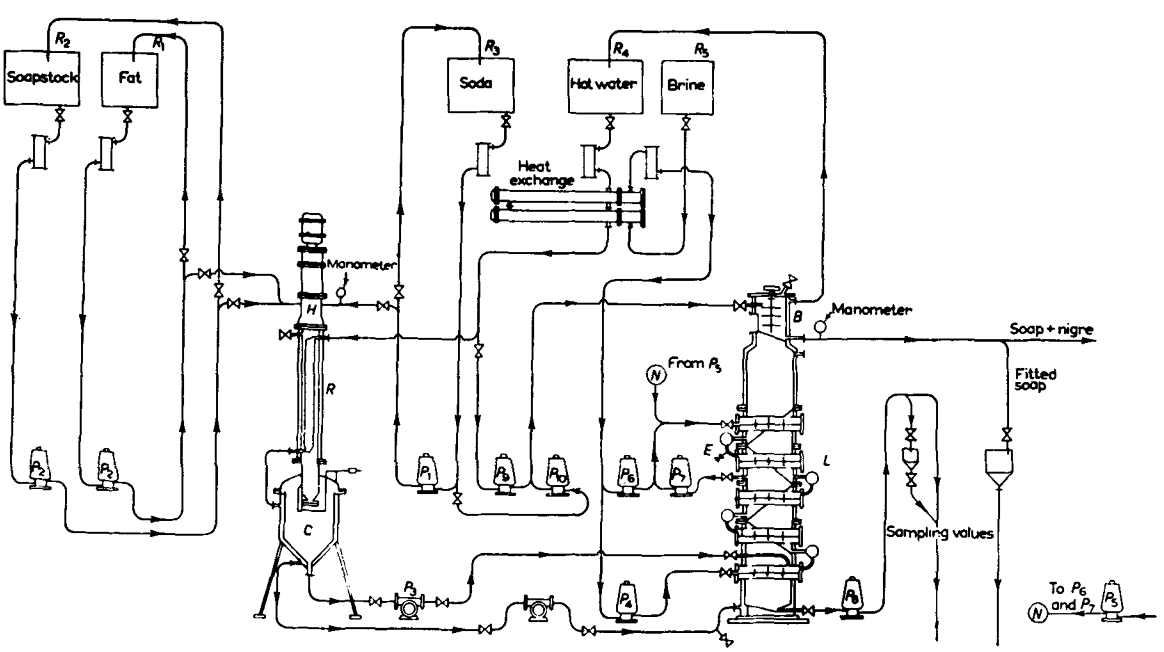

FIG. 2. Schematic diagram of the Monsavon Process.

The Monsavon process has numerous advantages: steam consumption is very low (100 kg. an hour per ton of soap), the energy consumption is low (20 Kwh), glycerol is recovered up to 95% of theory, and the operation requires only a few unskilled operators.

Detergents

Before 1939 the detergent industry was quite unimportant in France; at that time only a small number of products were manufactured for the textile industry.

The French soap industry, which consumed 220,000 tons of fats in 1938, had in 1942 only 20,000 tons at its disposal and 5,000 tons in 1944; this almost total lack of fats in the period 1940–1945 compelled the washing-products manufacturers, first to produce a soap with less and less soap (as low as 10%), then to find products where still less soap was required. Thus the production of oxyethylamide sulphates was developed, and the consumer used washing products with only 4% of fatty acids. After the war these products continued in favor for a while but little by little disappeared almost entirely.

During the same period the production of fatty alcohol sulphates increased regularly till 1956 (from 6,000 tons of products with a content of 30% active in 1952 to 35,000 tons in 1956), then it decreased a little in 1957 and 1958 (the annual production was 24,000 tons); at the same time the production of alkylaryl sulfonates climbed much higher and took the lead. There were 1,000 tons of such products with 25% active in 1952, 11,000 in 1955, and 87,000 in 1957.

It should be noted that a French company, the Sinnova, succeeded in devising an industrial process for continuous manufacture of fatty alcohols, based on the sodium reduction method of Bouveault and Blanc.

In France the development of nonionic products is growing slowly, chiefly because of the cost of ethylene oxide. It is probable however that such products will be developed in the near future.

Fat Derivatives

Among the various achievements in the field of fat derivatives in France during the last 50 years the most important one is undoubtedly "Rilsan," a polyamide produced from castor oil. Between the two world wars the Organico Society was small; its main activity was to produce Oenanthal, a base for perfumes derived from castor oil. The undecylenic acid obtained at the same time was treated as a by-product.

Beginning about 1940, the research laboratories of Organico studied the various possibilities for using undecylenic acid and, in particular, for converting it into the 11-aminoundecanoic acid. In 1949 a small unit was built to prepare this monomer in order to study its polycondensation and to study the manufacture of textile threads and fibers and of moulded objects and so forth. In 1952 this plant produced 17 tons of Rilsan per month, and production increased to 50 tons in 1955. These studies facilitated the building of large plants, such as the one in Marseilles for the production of this monomer and the one in Valence for the textile applications of Rilsan, both of which have been operating since 1956.

The uses of Rilsan are increasing both in textiles and moulded products. For example, it is used for hosiery, fine undergarments, fabrics of numerous types, brushes, cables, also for tubes, plastic panels, gears, bottles, electric insulators, and the like. Chemically Rilsan is obtained by polycondensation of the 11-aminoundecanoic acid, and its preparation from castor oil is given in Figure 4.

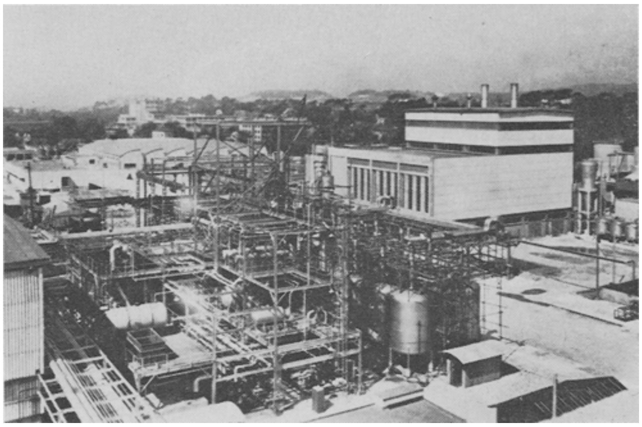

FIG. 3. Rilsan factory at Marseilles.

Basic Scientific Research in Fats

In 1908 there was no governmental scientific research institution working on fats. Some industrial firms had a small laboratory where a modest amount of research was carried out. Few members of a university faculty were interested in problems relating to fats. The Institut Colonial de Marseille, founded in 1906, took some interest in fats when Emile Baillaud became its general secretary in 1910. He was one of the promoters of the Laboratoire National des Matieres Grasses at the Science department of the University of Marseilles, which was founded at the end of the first World War. For about 15 years Pierre Desnuelle has been the director, assisted by Maurice Naudet.

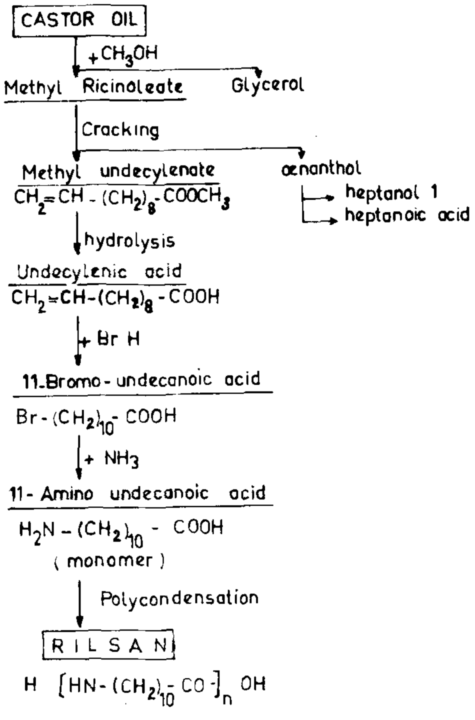

FIG. 4. Chemical steps in preparing Rilsan from castor oil.

In 1943 there were created in Paris the institutions which still have the task of studying fats. The activity of the I.R.H.O. (Institut de Recherches sur les Huiles et Oleagineux) is directed to the study of problems concerning the exploitation of oil products in Africa and the production of oils. The agronomic problems of the oil palm, coconut, peanut, and castor plant have been closely studied, not only in Paris but more particularly in test centers at Dahomey, Cameroun, etc. In the case of the oil palm tree, the result has shown significant success; the hectare output has increased tremendously. The extraction of oil has been greatly improved also by replacing part of the small oil mills by modern ones. Concurrently various laboratory studies were carried out in Paris by the I.R.H.O., such as the extraction of carotene from palm oil, the refining of shea butter, and studies on carotenes.

At about the same time members of domestic industries founded the I.T.E.R.G. (Institut Technique d'Etudes et de Recherches des Corps Gras), a professional institution financed by a self-imposed tax on production. It created a research laboratory in Paris and financed the Laboratoire National des Matieres Grasses in Marseilles. In 1950 the I.T.E.R.G. was designated the Institut des Corps Gras and became a part of the Centres Techniques Industriels (Industrial Technical Centers).

The activity of the I.T.E.R.G. is determined by its structure; its task is to be useful to domestic industries in the field of fats. In addition to an active library, the I.T.E.R.G. carries out laboratory research that is principally aimed at applied research problems.

The Laboratoire National des Matieres Grasses has studied questions related to oil refining (nature of foots, neutralizing, bleaching), to halogenated and hydroxylated by-products of fats (hydroxyacids and oxidized acids) emulsions, hydrolysis, and interesterification.

In that same year, 1943, the C.N.R.S. (Centre National de la Recherche Scientifique), the governmental institution that forms a part of the National Board of Education, founded in the group of laboratories of Bellevue (in suburban Paris) the Laboratoire des Corps Gras. Emile Andre was its director until 1945, and since then I have served. The task of this laboratory is to study the scientific aspects of problems pertaining to fats, especially those which may find application in various branches of industry. This laboratory has gradually grown in importance until today it has a staff of 25 persons. It recently increased its scope and was renamed as the Laboratoire de Lipochimie in order to stress its increased activities. Studies carried out by the Laboratoire de Lipochimie deal with the different branches of the field of fats. More than 200 articles have been published since its founding. Besides studies of secondary importance, there are today three principal fields of study: the phenomena of autoxidation and the antioxidants, the higher aliphatic peracids, and the saturated aliphatic dibasic acids.

In the field of education the I.T.E.R.G. founded in 1943 the Cycle de Preparation aux Techniques des Corps Gras; in 1952 it was reorganized into the Ecole Superieure d'Application des Corps Gras, which schedules classes and laboratory work for one year. It is open to French or foreign engineers.

The scientific and technical achievements in the field of fats are published in France chiefly in two series of reviews. The first is the Bulletin des Matieres Grasses from the Institut Colonial de Marseille. The first issue dates from 1917; it was replaced in 1946 by the review "Oleagineux," published by the I.R.H.O. The second series published by the I.T.E.R.G. successively has been constituted by the following: Corps Gras, Savons (1943–1944), Industries des Corps Gras (1945–1947), Bulletin d'Information de l'I.T.E.R.G. (1947–1954), and, finally since 1954, the Revue Francaise des Corps Gras. For 15 years engineers and chemists engaged in fat work have had a society, the Groupement Technique des Corps Gras. It participates in the Societe Internationale pour l'Etude des Corps Gras (I.S.F.).

Conclusion

From this survey of the development of industry and scientific research in the field of fats in France during the last 50 years one can conclude that very significant progress has been achieved. The position of France in this field is certainly one of stature and of honor.

Fifty Years of Fat Research in Germany

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The progress of research in fats during the past 50 years in Germany is a long period and an extensive subject. Research is not limited literally to the glycerides but includes the other lipids, such as phosphatides, sterols, lipovitamins, and lipochromes. Innumerable industrial products from fats and their substitutes, like detergents and surface-coating materials, should be included. Contemporary German literature contains thousands of papers published in this field. This very general perspective should, in no case, claim completeness, especially so in biology and nutrition physiology as well as apparatus and process technology. Where the literary references are devoid of interesting events, I shall include some personal experiences.

It has been 50 years since I was a student of chemistry at the University of Heidelberg. Eminent chemists were occupying the chairs of learning in the German universities at that time. Emil Fischer, W. Nernst, and E. Abderhalden in Berlin, A. von Baeyer (succeeded by R. Willstätter in 1915), H. Wieland and Hans Fischer in Munich, A. Hantzsch