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Fats and oils as oleochemical raw materials*

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Abstract: Vegetable oils and fats are important constituents of human and animal foodstuffs. Certain grades are industrially used and, together with carbohydrates and proteins, are important renewable resources compared to fossil and mineral raw materials, whose occurrence is finite. In concepts for new products, the price, performance, and product safety criteria are equally important and have a correspondingly high importance right at the start of product development. To ensure a high degree of product safety for consumers and the environment, renewable resources have often been shown to have advantages when compared with petrochemical raw materials and can therefore be regarded as being the ideal raw material basis. Results from oleochemistry show that the use of vegetable fats and oils allows the development of competitive, powerful products, which are both consumer-friendly and environment-friendly. Recently developed products, which fit this requirement profile, are the anionic surfactants cocomonoglyceride sulfate and the nonionic sugar surfactant alkyl polyglycoside. These products are used especially as mild surfactants in cosmetic formulations. In polymer applications derivatives of oils and fats, such as epoxides, polyols, and dimerizations products based on unsaturated fatty acids, are used as plastic additives or components for composites or polymers like polyamides and polyurethanes. In the lubricant sector fatty acid-based esters have proven to be powerful alternatives to conventional mineral oil products.

RAW MATERIAL SITUATION [1,2]

The sources of oils and fats are various vegetable and animal raw materials (e.g., tallow, lard) with the vegetable raw materials soybean, palm, rapeseed and sunflower oil being the most important ones regarding the amounts involved (Fig. 1). Of the approximately 101 million tonnes of fats and oils which were produced worldwide in 1998, by far the largest share was used in human foodstuffs. For oleochemistry, 14 million tonnes were available. In recent years, the amounts produced have continuously increased by approximately 3% per year. It is predicted that this trend will continue in the medium and long terms. The composition of the fatty acids contained in the oil (fatty acid spectrum) determines the further use of the oils. Special attention must be given to coconut oil and palm kernel oil (lauric oils) because of their high share of fatty acids with a short or medium chain length (mainly 12 and 14 carbon atoms: C12, C14). For example, these are particularly suitable for further processing to surfactants for washing and cleansing agents as well as cosmetics. Palm, soybean, rapeseed, and sunflower oil, as well as animal fats such as tallow, contain mainly long-chain fatty acids (e.g., C18, saturated and unsaturated) and are used as raw materials for polymer applications and lubricants (Fig. 2) [3,4]. Based on results from cycle analyses and ecological and toxicological studies for selected cases one can assume that products based on renewable resources usually are better ecologically compatible when compared with petrochemical-based substances—an important criterion in the development of a new product, just as price and performance are [5,6].

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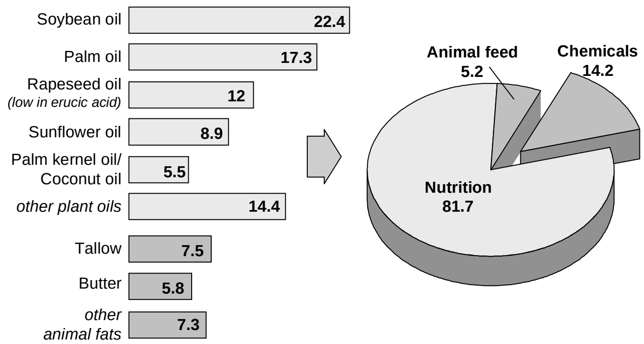

Fig. 1 World production and uses of oils and fats (1998, in million tonnes) [acc. to Oil World, Hamburg].

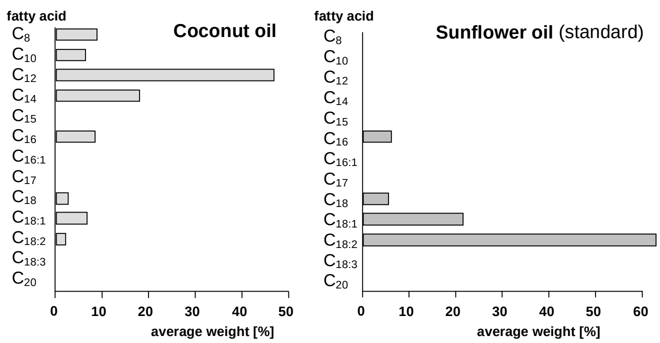

Fig. 2 Composition of coconut and sunflower oil.

EXAMPLES OF PRODUCTS

Oils and fats are triglycerides which typically consist of glycerine and saturated and unsaturated fatty acids. There are a few exeptions from this rule, such as castor oil, a glycerol triester of 12-hydroxyoleic acid (ricinoleic acid). From a chemical point of view, triglycerides offer two reactive sites, the double bond in the unsaturated fatty acid chain and the acid group of the fatty acid chain. With regard to product development based on triglycerides the majority of derivatization reactions is carried out at the carboxylic group (>90%) whereas oleochemical reactions involving the alkyl chain or double bond represent less than 10%. For most of the further uses oils and fats must be split into the so-called oleochemical base materials: fatty acid methyl esters, fatty acids, glycerol, and, as hydrogenation products of the fatty acid methyl esters, fatty alcohols [1].

Surfactants

The basic way in which surfactants act is determined by their structure. With their hydrophilic head and hydrophobic tail, surfactant molecules interpose themselves between water and water-insoluble substances. Surfactants are generally classified as being anionic, cationic, nonionic, or amphoteric surfactants depending on the type and charge of the hydrophilic groups (Fig. 3) [7a]. By far the most important field of surfactant application is the washing and cleansing sector as well as textile treatment and cosmetics; these use more than 50% of the total amount. Other uses are in the food sector, in crop protection, in mining, and in the production of paints, dyes, and paper. The basic manufacturing routes to important surfactants are laid out in Fig. 3. The surfactant with the highest volume—apart from

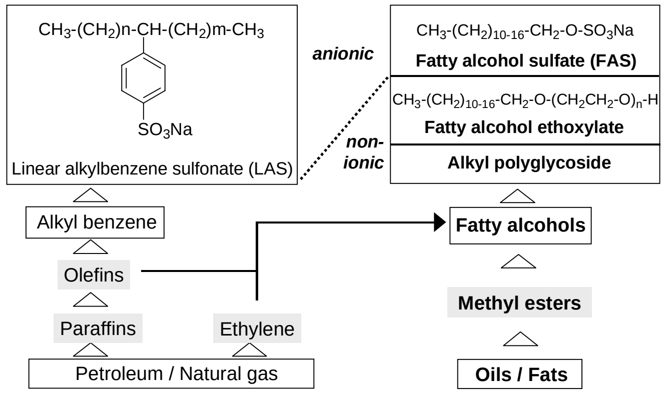

Fig. 3 Production of surfactants and examples of products.

Table 1 Surfactant production in western Europe (1000 t) [7b].

Surfactant class	1995	1996
Anionic surfactants	872	899
- alkyl benzene sulfonates	*420*	*400*
- fatty alcohol ether sulfates	*192*	*229*
- fatty alcohol sulfates	*101*	*111*
- alkane sulfonates	*77*	*77*
- others	*82*	*82*
Nonionic surfactants	1030	1068
- ethoxylates	*817*	*844*
- others (incl. APGs)	*213*	*224*
Cationic surfactants	190	170
Amphoteric surfactants	38	43
Total amount	**2130**	**2180**

soap—is still the petrochemical-based alkyl benzene sulfonate; however, in recent years a continuous trend towards surfactants based on renewable resources has become apparent. The total worldwide market amounts to approximately 9.3 million tonnes (1996, without soap). In western Europe a total of 2.18 million tonnes of surfactants was produced in 1996. The amounts involved, subdivided into the individual surfactant classes, are shown in Table 1.

Cocomonoglyceride sulfate [8]

Cocomonoglyceride sulfate (CMGS) has been known for a long time and has already been used in a few products. However, the normal manufacturing methods have various disadvantages, such as high production costs, the use of solvents or large amounts of secondary products and, as a result, a product quality which is not optimal. In a newly developed manufacturing process CMGS is obtained directly from coconut oil in a solvent-free two-stage process. In the first stage cocomonoglyceride is obtained by simple transesterification of coconut oil with glycerol in a molar ratio of 1:2 (Fig. 4). This pure vegetable raw material is converted to CMGS by reaction with sulfur trioxide gas (1–8% v/v in air or

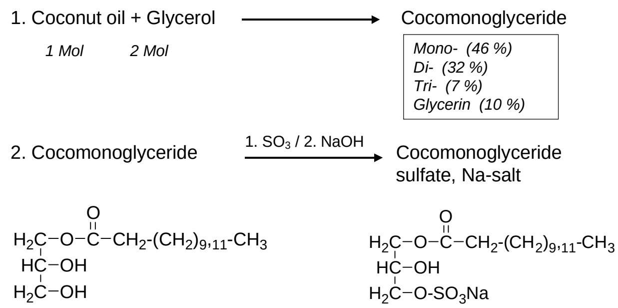

Fig. 4 Manufacture of cocomonoglyceride sulfate (CMGS).

nitrogen) in a falling-film reactor. The raw product is then neutralized with aqueous sodium hydroxide.

Because of its technical application properties CMGS is predestined for use in cosmetic products such as shower gels and foam baths or shampoos. Here it can be seen that CMGS in comparison to ether sulfate, the standard surfactant for this application, has a similarly good foaming power. Combinations of alkyl polyglycosides (APG) and CMGS, in which CMGS acts as foam intensifier, are particularly interesting. The CMGS/APG mixtures additionally show an adequate thickening ability, an important criterion for the formulation of products. An acceptable viscosity is already achieved with 10% solutions without the addition of co-surfactants by using only small amounts of sodium chloride. In dermatological tests for skin compatibility (epidermis swelling test) and mucous membrane irritation (HET-CAM) CMGS proved to be considerably less of a skin irritant than ether sulfate or other anionic surfactants such as sulfosuccinates. It is comparable to very mild surfactants, such as oleylmethyltauride and betaine (cocoamidopropylbetaine). By mixing with alkyl polyglycoside the skin compatibility of CMGS can be improved still further.

Carbohydrate surfactants – alkyl polyglycosides [9–11]

The development of surfactants based on carbohydrates and oils is the result of a product concept that is based on the exclusive use of renewable resources. In industry, saccharose, glucose, and sorbitol, which are available in large amounts and at attractive prices, are used as the preferred starting raw materials.

The selective functionalization of saccharose and sorbitol with fatty acids for the construction of a perfect amphiphilic structure cannot be realized in simple technical processes because of the polyfunctionality of the molecule. This is why the products offered on the market contain different amounts of mono-, di- and tri-esters and are therefore only suitable for particular applications, for example, as emulsifiers for foodstuffs and cosmetics or, in the case of the sorbitan esters, also in technical branches such as in emulsion polymerization.

The ideal raw material for selective derivatization is glucose. Reaction with fatty alcohol produces alkyl glucosides; N-methylglucamides are prepared by reductive amination with methylamine and subsequent acylation. Both products have proved to be highly effective surfactants in washing and cleansing agents. The alkyl glucosides have also additionally established themselves in the cosmetic products sector, as auxiliaries in crop protection formulations and as surfactants in industrial cleansing agents and today can already be said to be the most important sugar surfactants based on the yearly production amounts.

Alkyl polyglycosides have been known for a long time but only now, following several years' research work, has it been possible to develop reaction conditions that allow manufacture on a commercial scale. The structure on which these compounds are based corresponds exactly to the surfactant model described above. The hydrophobic (or lipophilic) hydrocarbon chain is formed by a fatty alcohol (dodecanol/tetradecanol) obtained from palm kernel oil or coconut oil. The hydrophilic part of the molecule is based on glucose (dextrose) obtained from starch (Figs. 5 and 6). Cognis currently has a

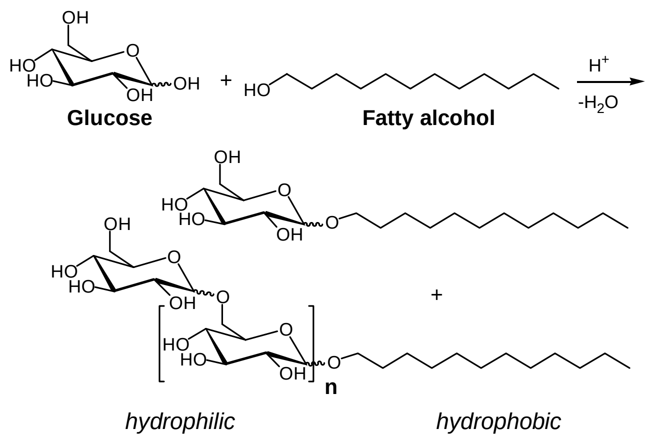

Fig. 5 Synthesis of alkyl polyglycosides.

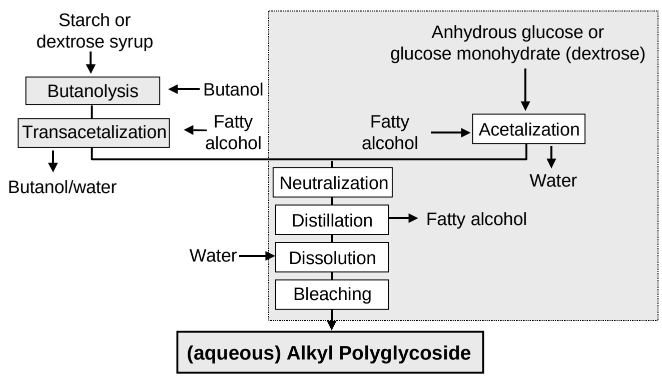

Fig. 6 Manufacturing processes for alkyl polyglycosides.

capacity of ca. 50 000 tonnes/year available for the manufacture of this class of compounds (other manufacturers are Kao, SEPPIC, Akzo Nobel, and ICI). Unique properties had previously been determined for alkyl polyglycosides, particularly in combination with other surfactants. For example, the use of alkyl polyglycosides in a light-duty detergent or shampoo formulation means that the total amount of surfactants can be reduced without sacrificing any performance. In other combinations a particularly stable and fine foam can be produced which protects sensitive textiles during the washing process. Toxicological and ecological laboratory investigations have also produced favorable results. Alkyl polyglycosides have a good compatibility with the eyes, skin, and mucous membranes and even reduce the irritant effects of surfactant combinations. On top of this they are completely biodegradable, both aerobically and anaerobically. The relatively favorable classification (for surfactants) into class I under the German water hazard classification (WGK I) results from this.

Oleochemicals for polymer applications

The use of oleochemicals in polymers has a long tradition. One can differentiate between the use as polymer materials, such as linseed oil and soybean oil as drying oils, polymer additives, such as epoxidized soybean oil as plasticizer, and building blocks for polymer, such as dicarboxylic acids for polyesters or polyamides (Table 2) [12]. Considering the total market for polymers of approximately 150 million

Table 2 Oleochemicals for polymers—selected examples [12].

	Product/use	Source
Polymer materials		
- polymerized soybean oil, castor oil	drying oils	soybean oil, castor oil
- polymerized linseed oil	linoleum	linseed oil
Polymer additives		
- epoxides	stabilizers, plasicizers	soybean oil
- soaps (Ba/Cd, Ca/Zn)	stabilizers	stearic acid
- fatty acid esters, - amides, waxes	lubricants	rapeseed oil
Building blocks for polymers		
- dicarboxylic acids	polyamides, polyesters, alkyd resins	tall oil, soybean oil, castor oil,
- ether-/ester polyols	polyurethanes	sunflower oil, linseed oil, oleic acid

tonnes in 1997 the share of oleochemically based products is relatively small—or, in other terms—the potential for these products is very high. Without doubt there is still a trend in the use of naturally derived materials for polymer applications, especially in niche markets. As an example, the demand for linseed oil for the production of linoleum has increased from 10 000 tonnes in 1975 to 50 000 tonnes in 1998 (coming from 120 000 tonnes in 1960!) [13a]. Epoxidized soybean oil (ESO) as a plastic additive has a relatively stable market of approximately 100 000 tonnes/year [13b].

Just recently, research has been started to use oleochemicals to build up matrices for natural fiber-reinforced plastics [14]. The use of natural fibers, such as flax, hemp, sisal, and yucca, is of increasing interest for various applications, among them the automotive industries, where the composites could be used in door pockets, covers, instrument panels, and sound insulation [14a]. Other applications could be in the manufacturing of furniture. In this field, Cognis is coordinating a research project, which is funded by the German Federal Ministry of Food, Agriculture, and Forestry (BML) and the National Agency for Renewable Resources (FNR), with the German Aerospace Center (DLR) as external partner. The focus is the development of a matrix-system with a high content of renewable raw materials (70–75%) and comparable or better performance compared to purely petrochemically based matrices. Oleochemically based monomers, such as epoxidized oils, maleinated oils, and amidated fats are under investigation [14b].

Oleochemically based dicarboxylic acids—azelaic, sebacic, and dimer acid (Figs. 7,8)—amount to approximately 100 000 tonnes/year as components for polymers. This is about 0.5% of the total

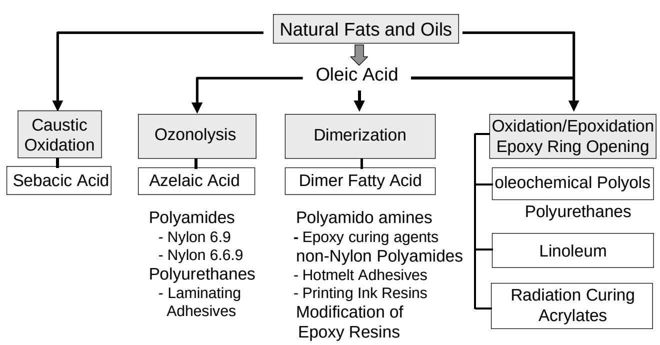

Fig. 7 Building blocks for polymers based on natural oils.

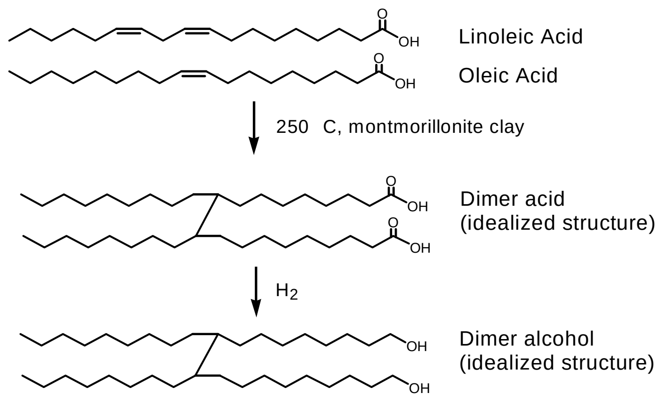

Fig. 8 Dimerization of unsaturated fatty acids.

dicarboxylic acid market for this application, where phthalic and terephthalic acids represent 87%. The chemical nature of these oleochemically derived dicarboxylic acids can alter or modify condensation polymers, and therefore will remain a special niche market area. Some of these special properties are elasticity, flexibility, high impact strength, hydrolytic stability, hydrophobicity, lower glass transition temperatures, and flexibility. The crucial reactions in the development of building blocks for polymers based on oils and fats are all carried out at the double bond of unsaturated glycerides or fatty acids: caustic oxidation, ozonolysis, dimerization, (aut)oxidation, epoxidation, and epoxy ring opening (Fig. 7). In the following sections recent developments in the field of diols and polyols for polyurethanes will be presented in more detail [15,16].

Table 3 Specifications of high-molecular-weight aliphatic diols [16].

Chemical designation	Dimer diol	Dimer diol ether	12-Hydroxy stearyl alcohol	1,10-Decane diol
outer appearance	yellow liquid/colorless liqid		white flakes	white flakes
Hydroxyl Value	180–200 / 180–210	90–112 / 50–60	345–360	625–645
Viscosity (25 °C, mPas)	3500–4300 / 1800–2800		solid	solid
Melting Point (°C)	-	-	61–65	68–73
Composition		a)		
- monomer (%)	13 / 2		-	-
- dimer (%)	68 / > 96		-	>98
- trimer (%)	19 / 2		-	-
Trademark	Sovermol 650NS / 908	Sovermol 909 / 910	Sovermol 912	Sovermol 110

a)average molecular weight = 1000 (Sovermol 909) or 2000 (Sovermol 910) by oligomerization of dimer diol.

Dimerdiols based on dimer acid [16,17]

Dimerization of vegetable oleic acid or tall oil fatty acid (TOFA) yields dimer acids, originally introduced in the 1950s by General Mills Chemicals and Emery (now Cognis Corp.), and is a complex reaction resulting in a mixture of aliphatic branched and cyclic C36-Diacids (Dimer acid) as the main product besides trimer acids and higher condensed polymer acids on one hand, and a mixture of isostearic acid and unreacted oleic and stearic acid on the other hand. Hydrogenation of dimeracid methylester or dimerization of oleyl alcohol leads to dimer alcohols (dimer diols) (Fig. 8). Oligomers based on dimer diol are industrially manufactured by acid catalyzed dehydration of dimerdiol. Oligomers in the molecular weight range of 1 000 and 2 000 are commercially available by this route. Another method used to

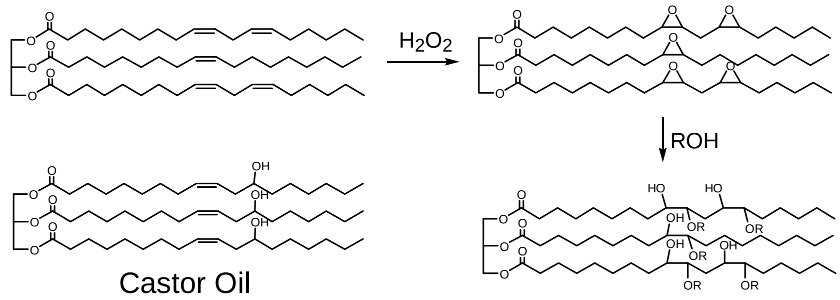

Fig. 9 Oleochemical polyols for polyurethanes (brand name: Sovermol).

produce oligomers is the transesterification of dimerdiol with dimethyl carbonate. The resulting dimerdiol polycarbonate has an average molecular weight of 2 000 (Table 3).

Due to their improved stability toward hydrolysis and oxidation, dimerdiol polyethers (and dimerdiol polycarbonates) are used as soft segments in the preparation of thermoplastic polyurethanes (TPUs). Polyurethanes prepared from these oleochemical building blocks are very hydrophobic and show the expected stability. The products were almost unaffected when stored either in 60% sulfuric acid or 20% sodium hydroxide solution (60 °C, 7 weeks). For comparison, ester-based polyurethanes as a standard were destroyed completely under these testing conditions after one week and two weeks, respectively. Soft segments based on dimerdiol ethers are used to prepare saponification-resistant TPU-sealings, which allow the contact to agressive aqueous media. A typical field of application is in nutrition technology [18].

Polyols based on epoxides [16,19]

Low-molecular-weight liquid epoxy polyol esters or ethers which can be employed as polyols for polyurethane systems are obtained by reaction of epoxidized oils with low-molecular-weight mono- or polyfunctional alcohols or acids. Depending on the reaction conditions either polyols with high OH-functionality (complete reaction) or epoxy polyol esters with remaining epoxy groups (partial conversion) are obtained (Fig. 9) [16]. Oleochemical polyols have an average molecular weight of 250 to 2500. Due to their relatively low viscosity and their compatibility with methylene di(phenylisocyanate) (MDI) they are particularly suitable for solvent-free, two-pack, full-solids polyurethane systems, to be applied as thin decorative or protective coating. They can also be applied in thick coatings, bearing even high filler loads. In industrial flooring applications, self-levelling polyurethane or epoxy/polyurethane multilayer systems offer good chemical and mechanical properties and benefits such as minimal shrinkage, high mechanical strength and durability, and favorable cost of installation. They are broadly used for wear- and crack-resistant floorings on parking decks, for concrete protection in assembly areas, as well as in large kitchens or slaughterhouses due to the ease of cleaning. They can also be used to bind porous filler materials and rubber particles to produce composites for sport tracks and playing fields [13,16].

Biodegradable fatty acid esters for lubricants [20–22]

Apart from being used as "bio-diesel", fatty acid esters, which are obtained from fatty acids (mono-, diacids) and alcohols (mono-, polyols), are gaining increasing importance as biodegradable replacements for mineral oils. In some application areas, such as chain saw oil, gearbox oils, hydraulic oils and lubricants for crude oil production, these oleochemical products are already well established. Current developments refer to the use of tailor-made fatty acid esters with specific lubricant properties. In the meantime, environment-friendly alternatives are available for almost all mineral oil-based products. In

Europe, the long-term potential is estimated to be 10–20% of the total market (500 000–1 000 000 tonnes/year. In 1997, 40 000 tonnes of biodegradable lubricants were sold in Germany alone (4.5% of the total market). An increase of this share is the aim of various measures taken by industry, government, and authorities.

PERSPECTIVES

With the examples of recent product innovations from oleochemistry the successful development of environmentally compatible and powerful products in the sense of an sustainable development has been demonstrated. Where do we go from here?

It can be assumed that in the future further possibilities for using renewable resources will continue to be sought for in an ever-increasing manner. As far as the development of new raw materials is concerned, Cognis is involved with plant breeders and government agencies within the framework of government-sponsored projects in the realization of new oils which, because of their optimized fatty acid composition, will satisfy the requirements for industrial use in a better manner. An example is sunflower oil with a high content of oleic acid (>85%) and a low stearic acid content. How the breeders will succeed, how the oil yields and, therefore, the economic efficiency of industrial use will turn out to be, remains to be seen.

The combination of various vegetable raw materials to form new products will also be a challenge for research and development in the future, as is the development of products for polymer or composite applications. Basic investigations, especially regarding the use of new long-chain fat derivatives, are needed in this respect and, in order to provide the necessary stimulus, also worthy of being sponsored.

Company Profile

Cognis (the former chemical products business of Henkel) is a leading supplier of specialty chemicals. Cognis offers customers in the whole world a complete range of products based on natural oils and fats. Cognis Oleochemicals business derives its most important raw materials from plants such as coconut and oil palms, rapeseed, soybeans, and sunflowers. The annual consumption amounts to almost 1 million tonnes. The share of renewable resources of the total amount of raw materials used by Cognis is approx. 35% (compared to 5–9% as the average number for the chemical industry). In the Care Chemicals business Cognis develops targeted solutions for the formulation and production of products that meet consumer needs in the cosmetics and toiletries and detergents and household cleaners industries. In the Organic Specialties business Cognis provides innovative solutions that boost performance and improve eco-compatibility, for example, in developing new additives and components for coatings, inks, and plastics. Ester-based lubricants are becoming increasingly widespread around the world. Cognis offers high-performance and biodegradable products for formulating hydraulic fluids, automotive and aircraft lubricants, as well as innovative drilling auxiliaries. Currently Cognis is involved in various national and European programs and projects with the goal to explore new technical uses for renewable resources. <http://www.cognis.com>

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