

(43,44). Much higher solid contents are possible in these soaps as compared to the comparable coconut fatty acid soaps.

Other uses are as rust inhibitors for hydrocarbons (45), epoxy esters (46), soaps for SBR latex foam stabilization (47) and soaps for polyethylene emulsions (48).

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New Applications for Fatty Acids and Derivatives^{1,2}

M. WEISS, Gross & Co., Newark, NJ, R. ROSBERG, and N.O.V. SONNTAG, Southland Chemical Division, Dallas TX (and S. ENG, Glyco Chemicals, Inc., Painesville, OH)¹

ABSTRACT

The early development of the American fatty acid industry during 1900-1920 is usually associated with the use of stearic acid in candles for lighting purposes. Today, the use of stearic acid in candle manufacture continues; the volume consumed for this application is not small, but the use is for ornamental, decorative or festival purposes, only incidentally for emergency lighting purposes. When one considers that most candles consist of 10-20% stearic acid, which improves among other things, the appearance, burning qualities and stiffness of the candle, the volume consumed is appreciable. More than that, the growth rate in this use is impressive. In 1965, it has been estimated that the volume of double-pressed stearic acid that found its way into candles was 3.5 million pounds. Today (1979) it is conservatively estimated that from 10-12 million pounds are utilized for this purpose. Innovations, such as the development of

dripless candles, achieved by the incorporation of a somewhat larger proportion of stearic acid to the external wax coat, are now possible uses. Concrete and asphalt are examples of much-used large tonnage materials of construction which have disadvantages in use that can be at least partially or significantly improved through the incorporation of stearic acid or certain other fatty chemicals. An excellent example of the tailor-making of fatty acid derivatives to satisfy an almost desperate need is provided by the development of derivatives suited for the retardation of water evaporation from reservoirs in arid areas. In 1966 it was estimated that the evaporation from large lakes and reservoirs in 17 western states was equivalent to 14 million acre feet, an amount which could ordinarily supply 84 million people annually. The design of water insoluble long chain organic compounds with a hydrophilic group at one terminal position gives products capable of forming monomolecular films which are uniquely suited to solve this problem. Fatty alcohol ethoxylates are only one approach among several.

The early development of the American fatty acid industry during 1900-1920 is usually associated with the

¹Paper presented at Short Course by S. Eng, Glyco Chemicals, Inc., Painesville, Ohio.

²Representatives of A. Gross Candle Co. (Linden, NJ), Faroy, Inc. (Houston, Texas) and Old Harbor, Inc. (Hyanis Port, MA) supplied part of the information used in this paper.

use of stearic acid in candles for lighting purposes. Today, the use of stearic acid in candles continues; the volume consumed for this application is no longer small, but the use is for ornamental, decorative or festival (religious) purposes, only incidentally for emergency lighting purposes.

When one considers that most candles consist of 10-20% stearic acid, which improves, among other things, the appearance, burning qualities, and stiffness of the candles, the volume is appreciable. More than that, the growth rate in this use is impressive. In 1965, it was estimated that the volume of double-pressed stearic acid that found its way into candle manufacture was 3.5 million pounds (1). Today (1979) it is conservatively estimated that from 10-12 million pounds are utilized for this purpose. One of the biggest novelty items, namely the do-it-yourself candle-making kit, has contributed remarkably to this increased use of stearic acid. And ask yourself this question: the last time you ate dinner at a fashionable, prestigious restaurant, a gourmet one at that, were the premises illuminated by electric light bulbs or rather, by romantic colored candles? The softer, subdued and slightly flickering light of the candle certainly contributes much to the enjoyment of a fine dinner, whether eaten outside the home or at home on special occasions.

Beeswax and solid fats have been used in candles since antiquity, but it was only after the development of fat splitting that fatty acids supplanted solid fats in the manufacture of the modern candle. The use of solid fats in candles was always objectionable because pyrolysis of these materials produced small quantities of acrolein, a pungent-smelling and eye-watering component which gave an undesirable odor to the burning operation. Smoking from candles has always been attributed by veteran candle makers as possible from any of the usual candle ingredients; by proper design of candle diameter and wick size or by using wick impregnation techniques with salts, this objection can be eliminated. Beeswax, on the other hand, since it is composed of ca. 72% "waxlike" fatty esters, ca. 13% "wax-acids" and ca. 12.5% long chain hydrocarbons, has no triglyceride components to generate undesirable acrolein on burning; however, its high price restricts its use to only those candles prescribed specifically for certain religious ceremonies (canon law dictates the use of from 51-100% beeswax for candles for certain purposes). The substitution of fatty acids such as double-pressed stearic acid for solid fats eliminated the burning odor problem attributed to acrolein and imparted other desirable properties to the finished candle such as lower temperature lighting, improved opacity, finer luster and sheen, and better molding and mold release, better candle stiffness, an improved nongreasy feel, and, in some instances, a better dispersion of candle additives and dyes. Although most modern candles contain paraffin wax as a major component, these properties do not result from the use of paraffin alone.

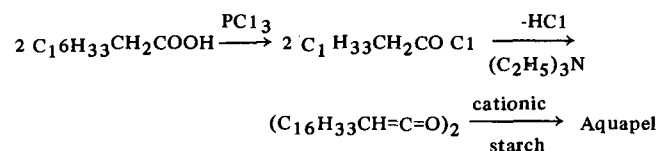
Approximately 5-30% of the total candle may be stearic acid, but the actual proportion is dependent upon the type of candle desired and its intended use. Dipping candles of the nondrip type are usually fabricated with 5-10% stearic acid for the inner 2/3 of their diameter and 20-30% stearic acid for the outer 2/3. Thus, the outside portion of the candle melts more slowly than the center, forming a cup that retains the melted wax until it is completely burned. Molded candles contain ca. 10-20% stearic acid. From this single use, the "as-is" uses of stearic acid have grown to literally hundreds of other interesting and diverse uses.

Somewhat similar, yet different in some respects, is the development of the use of stearic acid in waxy or fatty base crayon pencils. Like the use in candles, stearic acid use affords better luster, opacity, better dispersion of dyes and colors and somewhat better strength; unlike candle usage, it affords a better retention and absorption by paper and a

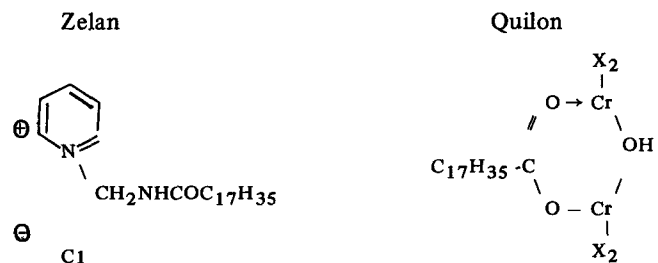
better removal from surfaces by detergents. The latter feature is indeed a desirable advantage; ask any mother of young, aspiring Rembrandts who have been left with red crayons in their possession. While the volume of stearic acid consumed in the manufacture of colored crayons is somewhat more difficult to estimate than that used for candles, perhaps 2-3 million pounds of various grades of stearic acid are now utilized for this purpose.

"Oil and water do not mix" goes the old saying, and most assuredly, fatty acids and many of their derivatives possess many water repellancy properties that are always being further developed. Stearic acid and its homologs have been used in innumerable ways to confer water repellancy to a number of other materials. With Hercules Powder Company's internal waterproof paper size line of products called "Aquapels," paper and paperboard products are afforded unique properties. Butcher paper, treated with Aquapel at little cost, increases resistance to liverblood penetration. These products, of course, are not new, having been used for 15-20 years or so, but some new innovations are beginning.

Stearic and palmitic acids were generally used for the manufacture of these products, but oleic and myristic acids were also used. The Aquapel (2) products are manufactured from fatty acids and are used in the form of reactive ketene intermediates. Fatty acids are converted to acid chlorides with phosphorus trichloride, the chlorides are dehydrochlorinated, presumably with triethylamine, and the resulting ketene dimers are emulsified and used in the paper industry to acylate cellulose under essentially neutral conditions. The Aquapel products are aqueous emulsions of appropriate ketene dimers as a dispersed phase with a cationic modified starch as an emulsifier. They are prepared in high-shear emulsification equipment such as Waring Blenders or colloid mills. The emulsions are employed as internal paper sizings and may be added to aqueous suspensions of paper pulp at any advantageous point after refining, but prior to sheet formation.



What is new is the development of improved products based upon the use of C20, 22 (behenic) and 24 fatty acids as partial replacements for the stearic acid. There are similar developments on the lasting, wash-resistant, water-repellant product called Zelan (3), developed by ICI and prepared from stearamide, formaldehyde, hydrochloric acid and pyridine and also on the Quilon (4) line duPont's durable, water and grease-repellant materials for textiles, which are water soluble, organo-inorganic chromium long chain fatty acid complexes, such as stearato chromic chloride, usually employed for the treatment of fur felt hats, finish for draperies, curtains and for treating blends of wool and synthetic fibers.



The blending of C-20-24 fatty acids that are somewhat higher priced than stearic acid is optimized with the improvement in overall water-proofing properties. The degree

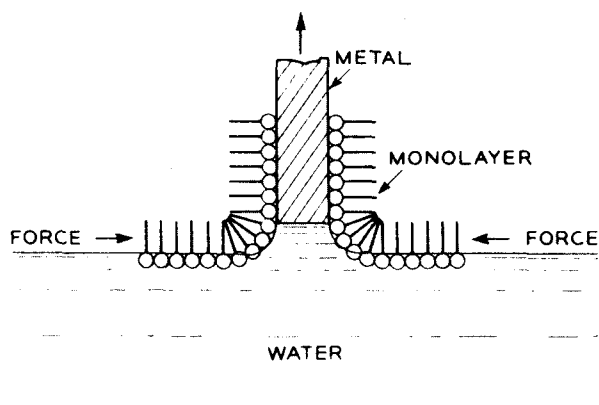


FIG. 1. Bond. Stearic acid molecules adhere to a metal plate as it's pulled up and through the monolayer on the water's surface (top diagram). The acid portion, represented by the circles, dissolves in the water just below the surface. The hydrocarbon portion (perpendicular lines) tends to stand straight up when the molecules are pushed together. The lower diagram shows the final chemical bond (metallic stearate) between the metal plate and acid portion of the molecule, and the physical bond between the hydrocarbon part of the molecule and the thermoplastic.

of water-proofing obtained from various types of long chain fatty acid derivatives varies with structural type; in some specialized applications it is even detrimental to the use intended. For example, the household fabric softener products based upon formulated suspensions of di(hydrogenated "tallyl") dimethylammonium chloride or sulfate commonly used to soften cotton or other fabrics in the household washing cycle are a case in point (5,6). Here the desired softening and antistatic effects must be balanced out against the slight water-proofing effect of the long chain quaternary salt by thinning out the application dosage to guarantee that cotton bath towels will not be rendered waterproof to the extent that they will not absorb water when they are used to dry. Downey (P & G), Sta-Puf (Staley), Nu-Soft (CP) contain di(hydrogenated "tallyl") dimethylammonium chloride or sulfate.

While the use of various fatty acids and derivatives used directly or indirectly in water-proofing paper or fabrics is substantial (estimated ca. 55 million pounds), the potential for the use of stearic acid and certain of its derivatives in concrete remains to be developed, and, in fact, could be well over 100 million pounds. Here the fatty products perform a water-proofing action for a slightly different end result. Applied to hardened concrete by a variety of techniques, they repel absorbed water and, in so doing, minimize cracking of the material when temperatures drop to below the freezing point. There are a few other fringe benefit properties that are also contributed, but, essentially, if the anticracking water-proofing job could be done reasonably well, the application could develop fast. Among the promising fatty candidates that show good promise are stearic acid itself, oleic acid chlorohydrin, 9-carboxystearic

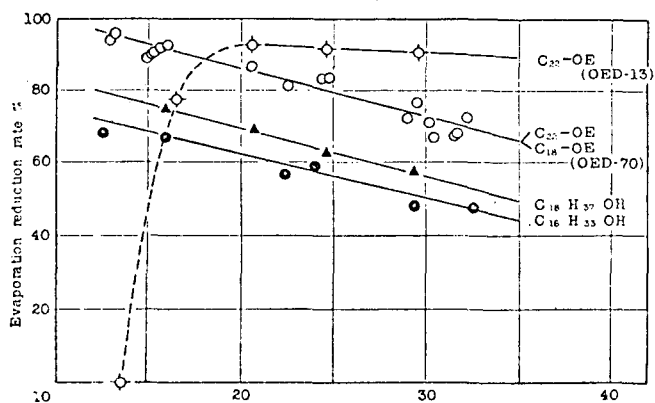


FIG. 2. Water temperature °C. Rate of reducing evaporation from water of various temperatures by the monolayer of OED-13, OED-70, octadecanol and hexadecanol.

acid, the maleic acid adduct of oleic acid (17), and, lately, trichlorosilanated tallow, promoted by the Fats and Proteins Research Foundation at Battelle Institute. The fatty silane product is a unique one; it is a product of trichlorosilane and tallow which fills the pores of concrete by reacting with free water and hydroxy-silicate groups to form a permanent water proof. It is said to be a more effective water repellent than silicones used conventionally for the water proofing of cement. TCST, as it is called, is well suited for above ground concrete applications, says Battelle (8).

Some time ago there was an inspiring report that Belle Telephone Labs had discovered that stearic acid is capable of joining aluminum to polyethylene (9); the resulting bond was more resistant to tearing than is polyethylene itself. Up to now, it hasn't been possible to form a direct metal to polyethylene bond that will withstand mechanical stress at high humidities and temperatures. An aluminum/polyethylene/aluminum bond prepared by a new technique said to be practical, has held up for months under 600 PSI stress at 100% relative humidity (at 25 to 37 C). Long chain fatty acids such as stearic acid can serve as a monomolecular layer in the metal/plastic bond. The acid end of the molecule forms a chemical bond with the metal, and the hydrocarbon chain forms a physical bond with the plastic. One end of the stearic acid molecule forms aluminum stearate with the metal plate; the other end becomes immersed in the polyethylene (Fig. 1).

Furthermore, it is also possible to bond aluminum, stainless steel and copper to polypropylene and polystyrene using octadecylamine or octadecylphosphonate as the monomolecular films. The applications in the electronics industry are obvious: a permanent bond between polyethylene insulators and copper conductors could improve mechanical properties of telephone cables. Printed circuits could be improved.

An excellent example of the tailor making of fatty acid derivatives to satisfy an almost desperate need in some countries of the world is provided by the development of retardants suited for the minimization of water evaporation from reservoirs in hot and arid areas. In 1966 it was estimated that the evaporation of water from large lakes and reservoirs in 17 western states was equivalent to 14 million acre feet, an amount of water which would ordinarily support 84 million people annually. Lake Mead losses in evaporation are about equal to the consumption of water in the entire Los Angeles metropolitan area.

Before we give the impression that we can eliminate evaporation from all the natural bodies of water in water-sparse communities in order to afford us the water our metropolitan centers need for filling swimming pools, for operating car washes, and for scores of other somewhat more essential needs, we must consider the serious potential effects of restricting natural water evaporation. A water-

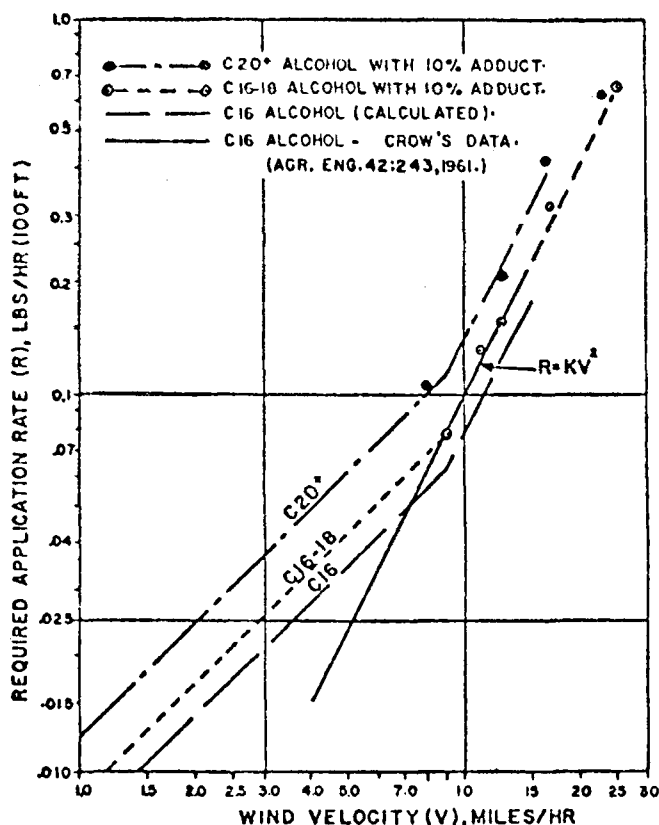


FIG. 3. Alcohol application rates required for smooth surface film. Rates determined in water-wind tunnel tests with wind meter 4 feet above water surface.

depleted atmosphere caused by the indiscriminate, well intentioned, but foolish program of water evaporation retardation from all natural sources could cause fantastic environmental problems. However, when man constructs a new reservoir in an existing hot and arid location, the minimization of evaporation from the facility is an extremely important consideration in the conservation of the water supply retained in it.

The requirements for a fatty acid derivative intended for use as a water retardation agent include insolubility of the candidate liquid in water, lighter specific gravity than water, very low volatility, ability to spread out in a thin monomolecular film on the surface with optimum effectiveness, nontoxic to fish, birds and aquatic plant life, also humans, and inexpensive operation. Some water insoluble long chain fatty compounds with a hydrophilic group at one end of the chain are uniquely suited for the solution of this problem. The fatty alcohols with carbon chains of from 16-22 atoms are potentially suited; their economic usefulness is restricted because they are not hydrophilic enough for optimum spreadability. Combined chemically with ethylene oxide at a ratio of 5 moles to 1 of alcohol, they are about optimum (10); the addition of small quantities of urea or algins makes them even more so. Figure 2 (11) illustrates performance data with monolayers of ethoxylated C_{18}/C_{22} alcohols.

If the agent is at all effective in reducing evaporation, it follows that the temperature of the water body will be higher compared to that of untreated reservoirs (evaporation is a cooling process). Actually, the advantage of somewhat higher temperatures in the rearing of water-field rice for faster growth in northeastern Japan has been noted in rice paddies treated with ethoxylated fatty alcohols as water retardants or temperature enhancers. Rice growth is substantially more rapid in water 4-5 C warmer than normal, and, hopefully, two rice crops where previously only one per season was possible is the anticipated

TABLE I

Alcohol Requirement in Phoenix Area^a

Wind, M.P.H.	% of Time prevailing	Wind demand lb./hr. 100 ft.	Reservoir requirement, lb./hr., 100 ft.
Calm	14.3	0	0
0-3	32.9	0.01	0.003
4-7	30.9	0.05	0.015
8-12	16.8	0.10	0.017
13-18	4.5	0.25	0.011
19-24	0.5	0.48	0.003
25-31	0.1	0.81	0.021

^aAv. hourly requirement, lb./hr., 100 ft. — 0.005.

TABLE II

Estimated Annual Costs

	Cost	% of Total
Alcohol, 0.06 lb./hr. 100 ft., for 8760 hr.	\$13,000	25
Labor		
Supervisor	10,000	
Maintenance man	6000	
Payroll added costs at 20%	<u>3200</u>	
Labor total	19,200	38
Other operating expenses		
Maintenance at 10% of installed cost	5000	
Taxes, etc., at 5% installed cost	2500	
Power, 6000 k.w. hr./month at 2.41¢	<u>1800</u>	
Other operating total	9300	18
Depreciation, annual, at 20%	<u>\$ 9900</u>	19
Total annual costs	\$51,400	

TABLE III

Water Costs on Circular Lakes with 3.5-Foot Savings

Reservoir area, acres	Equipment and operating cost, \$	Alcohol cost, \$	Total cost per acre ft., \$
100	\$30.30	\$7.43	\$37.73
1000	7.61	2.35	9.96
2500	4.29	1.49	5.88
10,000	1.91	0.74	2.65
35,000	0.90	0.40	1.30

advantageous result.

Using fatty acid derivatives as water evaporation retarders isn't quite as simple in practice as many have been inferred. For quiescent bodies of water, the problem is relatively simple, but when winds blow (and in few places in the world do they NOT) waves form, the surface film is either blown away, (generally at the rate of 1/30 that of the wind velocity) or washed away, and evaporation is enhanced. Application of retardant at a continuous rate is called for on the windward shore to maintain an unbroken cover. On Israel's Sea of Galilee, where winds often attain long periods of 30 mph velocities, potential losses could be quite considerable. Figure 3 (12) illustrates the correlation between wind velocity and application rate for a smooth surface film. Furthermore, the exact manner in which the retardant is applied is extremely important. Pumping all the agent that is required into a reservoir at one point isn't the answer. Precisely engineered nozzles spray the homogenized agent at specific points above the surface of the water. Statistical climatology and geology data for the area are required to determine how best this can be carried out. Tables I-III inclusive (12) illustrate alcohol requirements in

the Phoenix, AZ area, and give some 1969 projected costs, somewhat obsolete unless corrected for inflation.

While the volumes of ethoxylated alcohols used today are still relatively small, increased usage is sure to develop, despite the fact that very small quantities of retardant are employed per pound of water saved. Those areas of the world with already critical water supplies – Israel, Egypt, North Africa, India, and to a lesser extent Arizona, New Mexico and southern California in the U.S. – are certain to benefit from this use of specially tailored film-forming fatty acid derivatives. Our children are sure to be the chief beneficiaries.

There are other applications, largely unexplored, for this kind of usefulness which may be very valuable in agriculture. Consider the control of water loss through transpiration from plants and fruits by a thin monomolecular film coating; also, even potentially greater, consider the possibility of the control of evaporation from the soil surface by thin monomolecular films for those rain deficient countries that suffer from late spring or summer droughts. Both of these have potentially the greatest significance.

Fatty Alcohols

J.A. MONICK, Colgate-Palmolive Co.,
909 River Road, Piscataway, NJ 08854.

ABSTRACT

“Fatty” or higher alcohols are mostly C_{11} to C_{20} monohydric compounds. In probably no other homologous aliphatic series is the current balance between natural and synthetic products so vividly evident. Natural sources, such as plant or animal esters (waxes), can be made to yield straight chain (normal) alcohols with a terminal (primary) hydroxyl, along with varying degrees of unsaturation. In the past, so-called fatty alcohols were prepared commercially by three general processes from fatty acids or methyl esters, occasionally triglycerides. Fatty acids add hydrogen in the carboxyl group to form fatty alcohols when treated with hydrogen under high pressure and suitable metal catalysts. By a similar reaction, fatty alcohols are prepared by the hydrogenation of glycerides or methyl esters. Fatty alcohols are also prepared by the sodium reduction of esters of fatty acids in a lower molecular weight alcohol. The sodium reduction method was ordinarily too expensive; it was displaced early by the other methods; finally most unsaturated alcohols made by this route were largely replaced. Methyl ester reduction continues to provide perhaps 20% of the saturated fatty alcohols, and selective hydrogenation with the use of special catalysts such as copper or cadmium oxides was developed for the production of oleyl alcohol. Synthetic or petroleum technology for long chain alcohols include the Ziegler process, useful for straight chain, even-numbered saturated products. A second is the carbonylation and reduction of olefins affording medium or highly branched chain alcohols. Paraffin oxidation affords mixed primary alcohols. Fatty alcohols undergo the usual reactions of alcohols. They may be reacted with ethylene oxide to yield a series of polymeric polyoxyethylene alcohols or with acetylene under pressure to yield vinyl ethers or with vinyl acetate to give vinyl ethers.

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INTRODUCTION AND HISTORY

Background

The term “fatty alcohols” has become synonymous with “higher alcohols,” which is that class of monohydric aliphatic alcohols containing six or more carbon atoms, and derived from either natural or synthetic sources. The common usage of “fatty alcohols” is for those higher alcohols derived from natural fats or petroleum feed stocks, independent of the method of synthesis or procurement. The two major natural groupings of alcohols are the “coconut alcohols” and “tallow alcohols” which, of course, reflect their origin. Alcohols from petrochemical origin are usually called “synthetic higher alcohols.” Here again we have a subdivision that reflects on the chemical process for synthesis and we obtain “Oxo alcohols,” “Ziegler alcohols,” etc.

Alcohols are also classified according to their market usage. The principal market for primary alcohols between six and eleven carbon atoms is the plasticizer ester industry. The major use of higher alcohols with eleven or more carbon atoms is the detergent industry. Therefore, a logical subdivision of alcohols could be “plasticizer-range alcohols (C_6 - C_{10})” and “detergent-range (C_{11} and up) alcohols.” This division at C_{11} is arbitrary and crossover of alcohols does occur; C_{13} Oxo alcohol is used to make phthalate plasticizers, and a small amount of C_8 - C_{10} alcohols is used to make nonionic surfactants for industrial cleaning.

History and Sources

The knowledge of fermentation to produce ethyl alcohol goes back about 6,000 years. This is long before any written records, and makes ethanol one of the oldest known organic chemicals. Alcohols have two parents: “mother nature” for many centuries and “father science” for less than two. During the last 200 years, nature and science produced alcohols with a wide variety of molecular configuration and character. Simple saturated aliphatic