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26.1 Introduction

Emolliency can be translated as “softening ability,” and emollients are substances that make something softer to the touch. In cosmetic and personal care applications, an emollient is usually an oily substance, which when applied to the skin, makes it softer and more lubricated, enhancing the sensory properties of the skin. Emolliency is also an important parameter in skin moisturization as many emollients also contribute to regulating the moisture balance in the skin. In clinical practice, the term emollient is also used for formulated products used for treating dry skin conditions.

Emollients are normally volume-wise the biggest ingredient group in a skin care formulation, after water. The emollient phase may comprise 5–30% of the formulation in oil-in-water emulsions. Anhydrous systems and water-in-oil emulsions can contain even higher emollient concentrations. Understanding the different emollient types and their chemical and physical behavior is consequently important for the formulator, especially when optimizing the emollient composition to meet requirements on functionality and cost.

This chapter reviews several emollient technologies used in skin care products and describes the similarities and differences between the different substances that are commonly used for emolliency. The focus is on chemical and physical parameters differentiating the different emollient types, with the purpose of giving the formulator better understanding of this ingredient group.

26.2 Emollient Functionality Is Complex

Emollients are, along with emulsifiers and actives, the most important functional ingredient groups in skin care formulations. The emollient will influence the performance of the formulation in several ways: consistency of the formulation, skin feel, moisturization and lubricity on skin, delivery of actives, and the marketability of the product. It is therefore important to be able to optimize the properties of the emollients phase and tailor-make it for the intended use.

26.2.1 Emollients Determine the Structure of the Formulation

In the formulation, the emollient phase composition will determine the consistency of the cream or lotion. The viscosity and polarity, together with the presence of solid emollients, can be used to make the formulation more fluid or more solid, depending on the intended use.

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The emollient polarity will determine the solubility of active components, especially lipophilic ones. Correct choice of emollients can potentiate the activity of an active substance by controlling its bioavailability, by considering the properties of the active ingredient, stratum corneum and the formulation [1].

The interaction with emulsifiers and the choice of emulsifiers are also determined by the polarity and viscosity of the emollient. The solubility of the emulsifier and its ability to form liquid crystals and other surfactant aggregates are strongly dependent on the polarity of the oil phase in the system. This phenomenon is reflected in the use of the HLB system for characterizing emulsifiers as well as the phase inversion temperature (PIT) concept [2, 3].

26.2.2 Skin Feel Is Influenced by the Choice of Emollient

When applied to the skin, the polarity and viscosity of the emollient phase will determine “the skin feel.” This elusive term is a concentrate of everything which can be classified as the sensory properties of the formulation: lubricity, spreadability, absorption into the skin, and duration on the skin surface. These factors are closely linked to the molecular structure of the emollients [4–6].

The functionality of the emollient when absorbed into the skin is also important to consider. In contrast to common belief, the emollient is seldom an inert substance that does not influence the physiology of the skin. The emollient may interact with the skin lipids and also be metabolized in the skin if the chemical structure is suitable [7, 8].

26.3 Principles for Emollient Classification

There are many reasons to classify emollients: for understanding the behavior of emollients in formulations, for simplifying formulations, for making alternative formulations, and for solving

problems due to emollient behavior. The classification of emollients can be based on the chemistry of the substances used, but sometimes, especially when dealing with complex ingredients or mixtures of ingredients, a physicochemically based classification is more appropriate.

26.3.1 Emollient Classification Based on Molecular Structure

From a chemical point of view, most commercially available emollients come from three different types of chemistries: hydrocarbon based, ester based, or silicone based. Emollients utilizing moieties containing nitrogen and phosphorus are not common, and combinations with halogens are normally too toxic to be of interest. Table 26.1 illustrates some typical emollient structures and structural variations in each group.

Hydrocarbons, in this case without functional groups other than unsaturation and ring formation, are the common base for many classes of emollients. In this group, we find naturally occurring hydrocarbons such as squalene and squalane derived from biomaterials, simple hydrocarbons from mineral oil deposits (petroleum based), and synthetic hydrocarbons, normally derived from petroleum sources. Saturated hydrocarbons are usually chemically inert, being resistant to oxidation and hydrolysis.

Fatty alcohols such as oleyl alcohol or isostearyl alcohol are also frequently used as emollients or emollient modifiers in skin care formulations. These substances combine a long hydrocarbon chain with a primary hydroxyl group. In some cases, especially in short chain alcohols used in esters, the alcohol group can also be a secondary one.

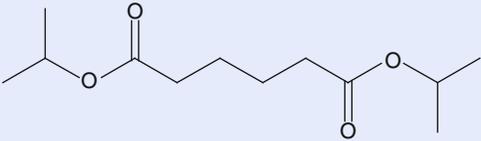
Ester-based emollients combine a carboxylic acid with an alcohol. Depending on the chemical structure of the acid and the alcohol, we can talk about simple esters, polyhydric alcohol (polyol) esters, and complex esters. A specific group of polyhydric alcohol esters are the naturally occurring vegetable and animal oils and fats, which are based on fatty acid esters of glycerol. The common

Table 26.1 Grouping of commonly used skin care emollients and typical structural variations

Emollient type	Examples	Variations	Molecular structure
Mineral oil hydrocarbons	Paraffin oil	Chain length and branching	
Naturally occurring hydrocarbons	Squalane	Unsaturation	
Synthetic hydrocarbons	Isohexadecane	Monomer type, chain length, and branching	
Fatty alcohols	Cetearyl alcohol	Chain length and unsaturation	<chem>HOCCCCCCCCCCCCCCCCCCCC</chem>
Guerbet and oxo alcohols	2-octyldodecanol	Chain length and branching	
Natural fatty acids	Oleic acid	Chain length and unsaturation	
Synthetic fatty acids	Isostearic acid	Chain length and branching	
Simple esters	Isopropyl myristate	Acid and alcohol structure	
Natural triglycerides	Olive oil	Fatty acid and triglyceride composition	
Synthetic triglycerides	Caprylic/capric triglyceride	Fatty acid type and ratio	

(continued)

Table 26.1 (continued)

Emollient type	Examples	Variations	Molecular structure
Complex esters	Diisopropyl adipate	Acid and alcohol structure	

trait of all esters is their ability to react with water, releasing acid and alcohol, making esters sensitive to formulation conditions. Unsaturated esters are also sensitive towards oxidation reactions.

The fourth group of emollients, based on silicon chemistry, is usually referred to as silicone oils. More appropriately named siloxanes, they are combinations of silicon, oxygen, and hydrocarbons, offering a different set of physicochemical behavior compared to the hydrocarbons and esters. This group of emollients, although commercially and technically important, will not be covered in this chapter.

26.3.2 Classification Based on Physical Properties

Many different approaches to classifying emollients based on their physicochemical properties have been proposed [9]. The methods used should be easy to apply but also give enough information to distinguish between closely related substances. Methods proposed for the classification include rheology, interfacial and surface tension measurements, thermal properties, dielectric constants, and partition coefficients. The main directions for all the different methods can be divided into polarity and rheology.

26.3.2.1 Factors Affecting the Polarity of Emollients

The polarity of a substance can be described in many ways and is manifested in a variety of directions, depending on the chemistry involved [10]. The first prerequisite for polarity is the presence of polarizable functional groups in the molecular structure. In emollients such functional groups include double bonds, aromatic structures, ester groups, and carboxylic acids and alcohols.

The polarizability of double bonds increase in the following order: isolated double bonds < conjugated double bonds < aromatic double bonds as the presence of mobile electrons increases. Isolated ester bonds are less polarizable than conjugated ones of the same reason.

Polarizable groups can also present permanent dipoles if the functional groups are separated by an appropriate distance.

Permanent polarity is also caused by hydrogen bonding. Here, free carboxylic acid, alcohol, and amino groups are important functionalities to consider.

A classification of emollients based on polarity looks into the presence and concentration of these polarizable structural elements, permanent dipoles, and the presence or absence of hydrogen bonding.

26.3.2.2 Rheology as a Tool to Describe Emollients

The viscosity of a substance is to a large degree dependent on the size of the constituent molecules, larger molecules have larger surface areas, increasing the possibility of van der Waals interactions. Structural factors such as branching and cyclization will affect the molecular size and shape leading to effects on viscosity and self diffusion coefficients [11]. Increasing the molecular interactions by introducing polarizability and polarization will also strongly influence the rheological properties.

In complex emollient mixtures comprising solid crystals, such as in petrolatum, the rheology is more complex, and viscoelasticity is commonly encountered. For viscoelastic systems, viscosity as commonly measured is an insufficient tool to describe the rheological properties, and more sophisticated rheometers must be used to get a correct evaluation of such systems. The elastic

modulus is often giving good correlation to formulation stability and to sensory properties [12].

Classification of emollients based on rheology is a simple tool and gives a rough guideline for the estimation of the applicability of each emollient.

26.4 Individual Emollient Groups

The commercially available emollients can be described in order of increased complexity, starting with simple hydrocarbons and adding functional groups to influence the physicochemical properties.

26.4.1 Hydrocarbon-Based Emollients

Although emollients based on vegetable and animal oils and fats were probably the first ones to be used in skin care, petroleum-based materials rapidly became the standard when the cosmetic industry developed in the early 1900s. Petroleum-based materials were abundant, had good technological properties, and gave an excellent moisturization and emolliency. Petrolatum and mineral oil are still the most commonly used emollients globally although alternative technologies are being developed, basically due to the nonrenewability of the petroleum resources.

26.4.1.1 Naturally Occurring Hydrocarbons

Squalane and Squalene

Squalene and squalane are naturally occurring hydrocarbons with a specific metabolic function, being precursors for the formation of steroidal components in animals and plants [13]. Pure squalene is extracted from shark liver oil and from olive oil processing residuals. The shark oil squalene is not a sustainable alternative due to overfishing of the shark, and most squalene on the market is now from vegetable sources. The main raw material is olive oil deodorizer distillate which can contain sufficiently high levels of squalene to be commercially viable [14].

Squalene is highly unsaturated and oxidizes rapidly. Oxidized squalene is associated with comedogenicity. Squalene with a high peroxide

value was tested and shown to be comedogenic. After purification to remove the oxidized squalene, the comedogenicity disappeared, linking oxidized lipids to inflammatory reactions associated with comedogenesis [15–18].

The squalene is hydrogenated to produce squalane, a very stable, low-melting branched hydrocarbon with low viscosity. It is used to decrease the oiliness of the formulation, increasing the slip and for a generally pleasant and light skin feel.

Petrolatum and Mineral Oil

Petrolatum is a fraction of mineral oil with semi-solid consistency. Physicochemically, it is an oleogel, an oil-based, lipophilic, gel stabilized by network forming crystals of high-melting hydrocarbons. The composition of petrolatum is usually complex, comprising hundreds of individual hydrocarbons [19, 20]. It is derived from paraffinic mineral oils which is purified and hydrogenated to remove unsaturation and improve stability and color. Liquid mineral oils or paraffin oils are chemically similar to petrolatum but do not contain the high-melting waxes that give petrolatum its consistency.

26.4.1.2 Synthetic Hydrocarbons

Synthetic hydrocarbons are obtained from simple building blocks such as ethene, propene, isobutene, and decene by oligomerization reactions. Ethene produces polyethylene which is a linear hydrocarbon resulting rapidly in high melting points with increased molecular weight while propene, isobutene, and decene can be used to make low-melting branched hydrocarbons. These saturated, branched, emollients are hydrophobic and stable against oxidation. Synthetic hydrocarbons can also be obtained by catalytic cracking of heavier mineral oils and fractional distillation to obtain fractions with desired melting points and viscosities.

26.4.2 Alcohol-Based Emollients

A few long chain alcohols are used as emollients on their own, but both long-chain and short-chain alcohols are important building blocks for

ester-based emollients. They also introduce an important additional functionality to the hydrocarbons: the hydroxyl group. Free hydroxyl groups increase the polarity and have the capacity of forming hydrogen bonds with water, allowing alcohols to participate in liquid crystals and interact with skin lipids.

26.4.2.1 Long Chain Fatty Alcohols

Saturated fatty alcohols such as cetyl (palmitoyl), stearyl, or cetostearyl alcohols are not primarily used as emollients in skin care creams and lotions, their primary function being emulsion stabilizer and consistency factor. They do, however, contribute to the moisturization and emolliency by interacting with the emulsifier system when formulated at 1–2% in the formulation. These saturated alcohols are obtained from fully saturated palm oil, soybean oil, or animal fats after a catalytic reduction of the corresponding acids or methyl esters.

Oleyl alcohol is a constituent of some plant and animal waxes. A traditional source is whale oil which was obtained from the blubber of the sperm whale. Small amounts of oleyl alcohol are present in jojoba oil (*Simmondsia chinensis*) where the seed oil contains esters of unsaturated long chain alcohols with long chain fatty acids (wax esters) [21]. Most industrial oleyl alcohol is produced by catalytic reduction of oleic acid or its methyl esters. Oleyl alcohol is available in different grades characterized by the iodine value, reflecting the degree of unsaturation. Stearyl and palmitoyl alcohol are present as by-products, depending on the raw material origin (palm oil or animal fats).

Isostearyl alcohol is a branched fatty alcohol which is formed by a high-temperature isomerization reaction of unsaturated fatty acids, producing a mixture of randomly branched isomers. After hydrogenation to remove remaining double bonds and to convert the acid to alcohol, isostearyl alcohols with a low melting point and high stability are obtained.

Guerbet alcohols are a special class of branched alcohols formed from primary alcohols via an oxidation and aldol condensation mechanism [22]. The resulting alcohols are primary

alcohols with a long hydrocarbon branching in the beta position. Typical Guerbet alcohols are 2-ethylhexanol (from *n*-butanol), 2-octyldodecanol (from *n*-decanol), and 2-hexyldecanol (from *n*-octanol). The branched Guerbet alcohols have lower melting points than their corresponding linear isomers with the same carbon number. Being saturated, Guerbet alcohols are normally resistant against oxidation.

Synthetic alcohols from petroleum sources include the oxo alcohols which are derived from unsaturated hydrocarbons by the addition of carbon monoxide and hydrogen in a hydroformulation process. The resulting aldehydes can further react by the aldol condensation forming branched alcohols after reduction of the aldehyde. Oxo alcohols are available in chain lengths from C8 (2-ethylhexanol) to C15.

26.4.3 Ester-Based Emollients

Esters are a popular and versatile group of emollients, due to the availability of a large number of ingredients with large differences in properties. This versatility can be used by the formulator to bring various functions to the skin care product, influencing stability, aesthetics, skin feel, and delivery of actives.

26.4.3.1 Simple Esters

Simple esters can be defined as esters of monohydric alcohols with acids with only one acid group. When used as emollients, the molecular weights, expressed as carbon numbers, range from C16 to C36, and the melting points from about -30°C up to 40°C . If the melting points exceed 40°C , the ingredients are often too waxy and hard to be used alone. However, high melting esters can be used at low concentration to modify the consistency and skin feel of the formulations.

Raw Materials for Simple Esters

Two types of raw materials are needed for ester production: an alcohol and a fatty acid. As there are numerous possibilities of combining an acid with an alcohol, a large variety in properties and behavior can be seen. Many groups of emollients

are the result of the development of oleochemistry which combines a natural raw material with a synthetic one.

Carboxylic Acids for Ester Production

All types of carboxylic acids are useful reactants for the formation of esters. Emollient esters, however, are usually based on acids that are longer than 8 carbons due to the higher aggressiveness of shorter acids and due to the normally offensive smell associated with shorter fatty acids. These long chain carboxylic acids are normally called fatty acids, especially if they originate in natural raw materials.

Several types of structural variations are relevant when describing the fatty acids used in emollient esters: chain length, even/odd carbon chains, unsaturation, branching, position on the chain of various functional groups, and so on. All these parameters contribute to the performance of the resulting ester as a cosmetic emollient.

Natural Fatty Acids

Natural fatty acids are obtained from vegetable or animal oils and fats by splitting (hydrolysis) of the fat into glycerol and acids [23]. The most important sources for fatty acids are coconut/palm kernel oil for lauric and myristic acids, palm oil for palmitic and oleic acids, and soybean/rapeseed oil for oleic, linoleic, and linolenic acids. Palm oil, soybean oil, and rapeseed oil are also important sources of stearic acid after hydrogenation. Animal fats such as lard and tallow are sources of palmitic, oleic, and stearic acids but are also often used after hydrogenation producing palmitic and stearic acids.

Vegetable-derived fatty acids are usually even numbered with chain lengths ranging from C8 to C24, with predominance for C12–C18. Fatty acids derived from animal fats can also contain fatty acids with an uneven number of carbons, typically C15 and C17.

The longer chain length fatty acids, starting from C16, can contain one or more double bonds. The number of double bonds and their location are important characteristics for unsaturated fatty acids. In life sciences, the normally used nomenclature is the *n*-*x* or omega-*x* system which

indicates the position of the first double bond, when calculated from the terminal end of the fatty acid chain. Oleic acid is thus an omega-9 fatty acid, linoleic acid an omega-6 fatty acid, and alpha-linolenic acid is an omega-3 fatty acid. Gamma-linolenic acid, in contrast, is an omega-6 fatty acid with three double bonds, changing its functionality and metabolism in the skin.

Unsaturated fatty acids from plants are normally in the all-*cis* configuration. Partially hydrogenated fatty acids and animal fatty acids can also contain trans-configured double bonds.

Natural-based fatty acids also comprise hydroxy acids which are found in, for example, castor oil and in lanolin (wool fat). Lanolin is also a source for branched fatty acids as well as dicarboxylic acids.

Synthetic Fatty Acids

Isostearic acid is produced as a by-product from oleic acid dimerization by a high-temperature reaction using montmorillonite clay or zeolites as catalyst [24]. Other long chain branched fatty acids are obtained by oxidation of the corresponding Guerbet alcohol or oxo alcohol.

Alcohol Sources for Esters

Alcohols and acids are related to each other in many ways. During chemical synthesis, an acid may be formed which is later reduced to an alcohol via an aldehyde. In a similar fashion, the first step in a chemical synthesis may result in an alcohol which later is oxidized to an acid. This means that for many alcohols and acids, there is a common hydrocarbon background, and the chemistry of these substances has many properties in common.

Low Molecular Weight Alcohols

Low molecular weight alcohols used in the manufacture of cosmetic emollients include isopropanol and ethanol as well as methanol and 2-ethylhexanol. Isopropanol and 2-ethylhexanol are obtained from petrochemistry while ethanol and methanol are also available through fermentation.

Low molecular weight polyhydric alcohols are also often used, for example, 1,2-propanediol

(propylene glycol). Glycerol (1,2,3-propantriol) is available both from natural sources (vegetable and animal oils and fats) and from petroleum chemistry.

Long Chain Alcohols

All types of long chain fatty alcohols described above are also useful as reactants in ester formation. Branched alcohols are often used due to their stability and low melting points. The nomenclature does not always reveal if the alcohol is branched or not: octanol can be either n-octanol obtained from caprylic acid or 2-ethylhexanol obtained from petrochemical sources.

Production Methods

Classic Methods: Direct Esterification and Transesterification

Any carboxylic acid and alcohol may be used to produce an ester by direct esterification. Normally, catalysts such as strong acids or tin derivatives are used, together with a sufficiently high temperature to remove the water produced in the reaction. A more efficient route to esters is to do transesterification reactions starting from a vegetable or animal triglyceride-based oil [25]. In the first step, the oil can be reacted with methanol to produce methyl esters which can be further transesterified with other alcohols, releasing the methanol. The methanol is subsequently removed by distillation and recycled.

Esters of high molecular weight alcohols can also be produced by direct transesterification from the triglyceride oil, but reaction conditions and yields will be dependent on the structure of the alcohol used. Secondary alcohols will be less reactive in the transesterification process, and better yields are obtained via the methyl ester route.

New Developments in Esterification:

Enzyme Catalysis

Biocatalysis, such as enzymatic catalysis using lipases supported by a carrier are increasingly used for the production of emollient esters. The reaction can be run at lower temperatures, saving energy but also leading to decreased formation of byproducts [26, 27].

26.4.3.2 Emollients Based on Triglycerides

Triglycerides are naturally occurring, renewable sources of emollients that can be used directly in skin care formulations. Plants and animals use triglycerides to store energy for future uses. The plants need energy in the seeds to produce a seedling that can start to photosynthesize. Animals store energy in fat depots to be used when the availability of food is scarce due to seasonal variations. Triglycerides are also a good source of raw materials for further processing to oleochemical derivatives.

Natural Oils and Fats

The fat depots in vegetable material and in animal material can be used to obtain triglyceride oils and fats that can be further developed into cosmetic ingredients. Many emollients and emulsifiers used in skin care formulations are derived from the easily accessible vegetable and animal oils and fats.

Due to purity and sustainability considerations, the vegetable fats have grown in importance over the last years and constitute an important raw material base for the cosmetic industry.

Raw Materials Based on Natural Oils and Fats

There are many possible raw materials available for making emollients based on natural oils and fats; however, most of them are produced in small quantities and are not always commercially feasible. Table 26.2 lists the vegetable oils and fats that are most common as raw materials for making cosmetic ingredients.

Animal fats are principally available from three sources: tallow from cattle, lard from pigs, and fish oil from marine fish species. Other sources do exist, but these are the main ones from an industrial point of view. Of these sources, the fish oil triglycerides are less common as very few fisheries remain to produce a cheap raw material. Lard and tallow are also less attractive today as raw materials due to issues related to sustainability, social, and cultural preferences.

The majority of industrially produced vegetable oils and fats come from four sources: *palm oil*, *soybean oil*, *rapeseed oil*, and *sunflower seed oil*.

Table 26.2 Approximative fatty acid composition ranges for commonly used vegetable oil raw materials

	C8	C10	C12	C14	C16	C18:0	C18:1	C18:2	C18:3	C22:1
Soybean oil					9-13	3-5	17-30	48-58	5-11	
Rapeseed oil (LEAR)					4-5	1-2	60-64	18-21	7-10	
Rapeseed oil (HEAR)					2-7	1-3	12-22	10-16	4-12	40-50
Rapeseed oil (HORO)					3-4	1-2	73-78	11-16	2-4	
Sunflower seed oil					4-9	1-7	14-40	48-74		
High-oleic SFO					2-4	3-4	80-84	7-10		
Shea butter					4-5	40-43	45-46	6-7		
Liquid shea butter					4-6	25-28	56-58	8-9		
Olive oil					7-20	0-4	56-85	4-20	0-1	
Sweet almond oil					4-9	0-3	62-86	17-30		
Palm oil				0-1	43-45	4-5	38-42	9-10		
Coconut oil	5-11	4-9	40-50	15-20	7-12	1-5	4-10	1-3		

Source: AathusKarlshamn Sweden AB, Karlshamn, Sweden

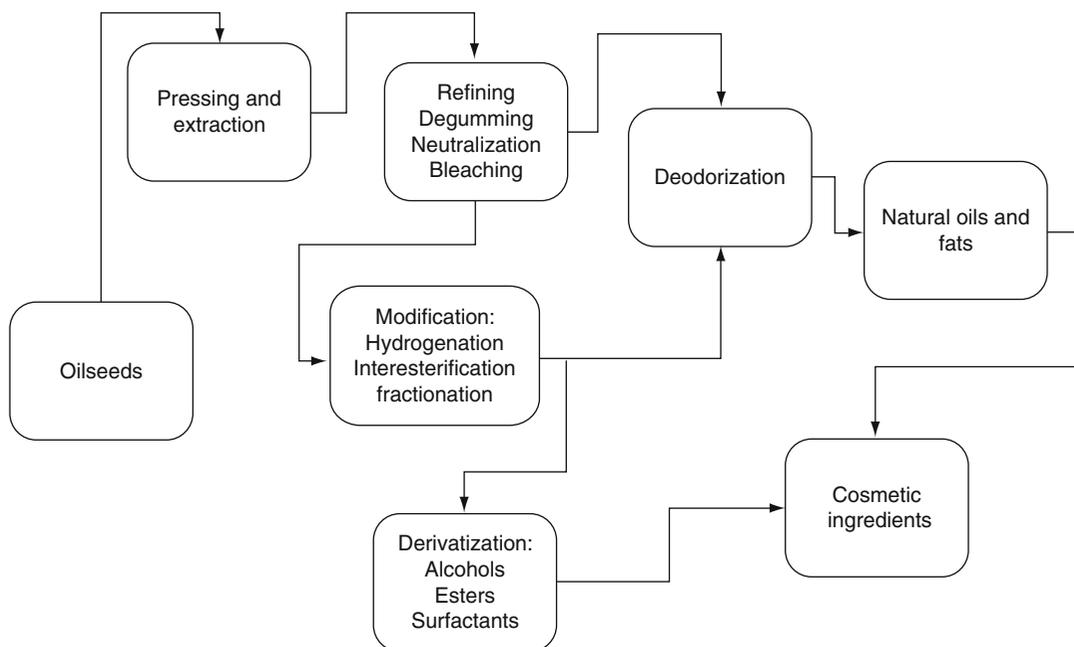


Fig. 26.1 Oilseeds of different kinds are converted to emollients, emulsifiers, and surfactants for skin care use via refining, modification, and derivatization steps

The three latter are liquid oils with high degree of unsaturation as they have high levels of linoleic (C18:2) and linolenic acids (C18:3). The unsaturation of soybean, rapeseed, and sunflower oil restricts their use in skin care as they are difficult to stabilize against oxidation. However, they are good starting points for modification as they are easily accessible and cost-effective.

Palm oil is a source for oleic (C18:1) and palmitic (C16:0) acids. It has a semisolid consistency at room temperature and is taken from the fruit pulp of the oil palm (*Elaeis guineensis*). The widespread cultivation of palm oil in Malaysia and Indonesia has led to concerns about habitat destruction and deforestation, and mechanisms for sustainable production of palm oil have been implemented. The Round Table for Sustainable Palm Oil (RSPO) is a multistakeholder organization for promoting and certifying the production of palm oil which do not contribute to social and environmental degradation in the producing counties [28].

There are two major sources of short and medium chain fatty acids (C8, C10, C12, and C14): *palm kernel oil* and *coconut oil*. These two

oils are the main raw material sources for producing surfactants and simple esters used in cosmetics and personal care applications.

Extraction, Purification, and Modification of Vegetable Oils and Fats

Natural oils and fats which are used as cosmetic emollients need to be purified to remove undesired naturally occurring seed constituents and environmental pollutants [29]. Apart from undesired minor components, vegetable oils usually also contain functional lipids such as tocopherols (vitamin E) and phytosterols. A balanced production efficiently removes the contaminants but preserves the functional minor lipids in the oil. Figure 26.1 gives a simplified outline of the processing involved in converting oilseeds to cosmetic ingredients.

The first step in all oil processing is to remove the oil from the seeds, kernels, fruits, and nuts. This is usually done by decorticating and drying the seed raw material before crushing it by different types of mills. The resulting cake is pressed to extract the oil. For most seed materials, the resulting pressed seed cake is further extracted

with a solvent, usually hexane. The crude oil is sometimes used as such, but normally the oil is further processed via refining, bleaching, and deodorization.

The refining of a vegetable oil can be regarded as a washing process using water with different pH values. The first washing, the degumming, is done using acidic water and removes phospholipids, gums, and sugars and decreases the level of metals and proteins present in the oil. The second washing is done with a weak alkali to remove free fatty acids. Again, water-soluble substances and metals are decreased. After drying, the oil is now ready for bleaching.

The bleaching step in vegetable oil refining comprises adding one or more absorbents to the oil and filtering off the absorbent after sufficient contact time. The absorbent is usually activated montmorillonite clay, also known as bleaching earth. The oil can also be treated with activated carbon to remove oil soluble contaminants. After the bleaching, the oil is free from phospholipids, free fatty acids, metal traces, protein residuals, and other degradation products and contaminants.

The final step in oil refining is the deodorization. This step, which is a steam distillation in vacuum, removes volatile components that cause odor and flavor. It also eliminates the last traces of free fatty acids. If done at too high temperature and extreme vacuum, this process also removes tocopherols and phytosterols, substances that are beneficial in skin care.

In case the properties of the oil are not suitable for the intended application, the vegetable oil can be modified using chemical and physical methods [30]. The most common method to modify the properties of unsaturated oils is to use hydrogenation. Hydrogen is added to the double bonds removing the unsaturation step by step. The melting point increases, but also the oxidative stability. Fully saturated vegetable oils are often used for further processing into esters and emulsifiers. Chain lengths from C14–C18 are preferred in this context.

Interesterification is another chemical modification method which is used to modify the melting and crystallization properties of oils and fats.

This process rearranges the fatty acids on the glycerol backbone and with an altered triglyceride composition, the melting behavior and sensory properties are modified. Oxidative stability is not affected as the fatty acid composition is not changed.

The physicochemical properties as well as chemical behavior can also be modified using fractionation techniques [31]. Methods which can be used for low molecular weight substances, such as fatty acids, alcohols, and esters, are not suitable for triglyceride oils. The high boiling point of triglycerides is due to their large size and prevents separation based on distillation. Instead, high-melting solids can be frozen out of the oil phase by lowering the temperature. Dry fraction is the term used when the cooling of the oil is done without solvent and is frequently used for palm oil and its derivatives. Dry fractionation uses high-pressure filters to separate the solid and liquid fractions from each other. If a better separation of the solid and liquid fractions is needed, a solvent must be used for the fractionation process. Hexane or acetone is the industrially used solvent for this type of process.

After physical or chemical modification processes, the solvents, catalysts, and by-products are removed by bleaching and deodorization to produce an emollient with optimized properties and high purity.

Some Common Natural Oil-Based Emollients and Their Properties [23]

Soybean oil is the oil from the soya bean, *Glycine soja*, of the Leguminosae family. It is highly unsaturated and has a short shelf life in cosmetic applications. Soybean oil is a good source for linoleic and linolenic acids; tocopherols, especially gamma-tocopherol; and phytosterols. Soybean oil is frequently derived from GMO soybeans, and special care must be taken in sourcing if non-GMO soybean oil is required. Soybean oil can be used in skin care to provide essential fatty acids (linoleic and linolenic acids), but due to the low oxidative stability, the level in the formulation is usually lower than 1–2%.

Rapeseed oil is found at high concentration in the seeds of the oil rape, *Brassica campestris*. The dominant fatty acids are oleic acid and linoleic acid, but also, linolenic acid is abundant. Several varieties of oil rape have been developed over the years. Currently, at least three different types of rapeseed oil are available: low-erucic acid rapeseed (LEAR, canola oil), high-erucic acid rapeseed oil (HEAR), and high-oleic rapeseed oil (HORO). HEAR oil has a high level of the long-chain monounsaturated erucic acid (C22:1) while high-oleic rapeseed oil has a significantly lowered content of linolenic and linoleic acids for improved oxidation stability. Rapeseed oil is also rich in tocopherols and phytosterols. Hydrogenated and fractionated rapeseed oil having an elevated level of tocopherols and phytosterols can be used to increase recovery of surfactant damaged skin [32]. The high oleic and the hydrogenated rapeseed oil can be used at 1–5% in the formulation without adverse effects on smell and stability.

Sunflower seed oil is obtained from the seeds of the sunflower plant (*Helianthus annuus*). The traditional sunflower oil has a high content of linoleic acid and is prone to oxidation. Newer varieties with low-linoleic and high-oleic acid content are also available (high-oleic sunflower oil). Sunflower oil is rich in tocopherols but requires normally removal of the seed coat waxes by winterization to obtain clear oil. Sunflower seed oil of the traditional, high-linoleic acid type can be used to provide essential fatty acids to the formulation. Low oxidative stability limits the use to 1–2% in the formulation. High-oleic sunflower seed oil can be used as a basic emollient at higher concentrations.

Shea butter is a unique vegetable fat with a high functionality in skin care [33]. The main fatty acids are stearic (C18:0) and oleic (C18:1) acids. The triglyceride composition yields a solid fraction primarily comprising stearic-oleic-stearic triglycerides and a liquid fraction dominated by stearic-oleic-oleic triglycerides. This combination gives shea butter a semisolid consistency and high emolliency and moisturization when added to a skin care formulation.

The uniqueness of shea butter also lies in the presence of high concentrations of triterpene cinnamates and acetates which are sometimes useful as bioactive additives in skin care [34]. A variety of shea butter, meeting the criteria on stability and low melting point, is liquid shea butter, obtained by fractionation of the semi-solid material. It has a high level of oleic acid but contributes also with a significant level of triterpene esters.

Two oils with a high content of monounsaturated fatty acids are *olive oil* and *sweet almond oil*. These oils are both popular in skin care formulations. Olive oil can be used as nonrefined but contributes with a typical greenish color and olive flavor to the formulation. These two oils are reasonably stable against oxidation due to the lack of sensitive linolenic acid and a moderate content of linoleic acid.

Numerous other vegetable oils are used in cosmetic formulations, mainly due to their usefulness as marketing ingredients. The composition normally falls into one of the four groups described above. In this group, we find vegetable oils and fats such as cocoa butter, murumuru butter, avocado oil, and babassu oil.

Synthetic Triglycerides and Analogues

The most common synthetic triglyceride used as emollient in skin care formulations is “caprylic/capric triglyceride,” also known as medium chain triglyceride (MCT). This synthetic triglyceride has intermediate viscosity and polarity and a high resistance against oxidation as it is fully saturated.

Caprylic/capric triglycerides are produced by esterification of glycerol by a mixture of caprylic and capric acids. Coconut oil or palm kernel oil is hydrolyzed to fatty acids and glycerol; the fatty acids are separated by distillation into fractions (C8–C10, C12–C14, and C16–C18). The C8–C10 fraction is added to glycerol, and the esterification reaction is carried out by removing reaction water. The process can be run without added catalyst or using tin-based acidic catalysts. After reaction, the oil can be deodorized to improve flavor and odor.

Emollients which are analogues to the caprylic/capric triglycerides are, for example, diesters of propylene glycol and triesters of glycerol and synthetic ethylhexanoic acid (“octanoic acid”).

26.4.3.3 Complex Esters

Esters with poorly defined compositions and which are often made from constituents with several reaction centers can be summarized by the name complex esters. This group also comprises lanolin, which is a natural product with complex composition.

Polyacid and Polyhydric Esters

In the last years, a new group of emollient esters have been introduced. These complex esters are based on different combinations of polybasic acids and polyhydric alcohols and are usually more viscous than traditional esters. They have also higher molecular weights, and by the selection of constituents, the polarity can be modified.

Esters based on polyhydric alcohols such as trimethylolpropane (three hydroxyl groups) and pentaerythritol (four hydroxyl groups) are also available. These complex esters are normally produced using branched fatty acids or unsaturated acids as the long chain linear esters tend to become too high melting.

Esters based on polycarboxylic acids with simple alcohols are also available. In this case, the viscosities are low, and the polarity is high due to the presence of several ester groups. Examples of this category include esters of adipic acid and citric acid with low molecularweight alcohols.

Lanolin and Lanolin Derivatives

Lanolin is another naturally occurring oleogel, characterized by having a high content of cholesterol and lanosterol esters. It is also rich in esters between dicarboxylic acids, long-chain diols, and hydroxy acids. Lanolin can be fractionated to yield different qualities of emollients, and it can be derivatized to increase and tailor-make the functionality for different applications [35–38].

26.5 Important Properties to Consider When Selecting Emollients

When formulating skin care products, the emollient is often combined with water, actives, and emulsifiers. In the formulation, several aspects must be optimized: stability, aesthetics, and delivery of actives. By a careful selection of emollients and by systematic variation of the emollient composition, formulations meeting the desired characteristics may be developed.

26.5.1 Compositional Aspects of Emollient Selection

The composition of commercially available emollients is usually difficult to investigate and analyze. Two emollients with the same INCI name, for example, “isopropyl palmitate” may differ in properties because the “palmitate” part may be derived from different starting materials. The palmitic acid used for this ester is seldom pure, and various amounts of stearic acid may be present as well as shorter fatty acids such as myristic and lauric. If the palmitic acid is derived from animal fats, it may also contain fatty acids with 15 and 17 carbons as well as branched fatty acids.

Many other types of emollients are equally difficult to characterize due to the complex composition of structurally similar and isomeric forms. This can be the case with petrolatum and mineral oils which can have different compositions depending on their origin. For this reason, it is important to remember that there cannot be a direct comparison from a technological point of view of two emollients, even if they are nominally the same and fulfill the same specification.

26.5.1.1 Contaminants: Environmental and Processing Dependent

All raw materials can be more or less contaminated with environmental pollutants. Such pollutants come from the soil, from air, from the processing, and from transports of raw materials

and intermediates. Contaminants can also leach into the emollient from packaging materials and from process equipment. Since contaminants of different types are ubiquitous and often represent higher toxicity than the emollients themselves, monitoring the presence of contaminants is important and constitute a part of the new Cosmetic Regulation of the European Union [39].

Contaminants which are of specific concern for emollients are heavy metals, pesticide residuals for plant derived materials, and polyaromatic hydrocarbons. Allergens, both those associated with skin sensitization as well as food allergens, should be absent. Even if emollients normally do not sustain microbial growth due to low water activity in the ingredient itself, packaging materials, and insufficiently heated materials can cause transfer of microbial contamination.

26.5.1.2 By-Products, Processing Aids, and Other Residues from Manufacturing

Normally, emollients do not contain high levels of toxic or irritating by-products from the reactions used to manufacture the ingredient, unless the material is highly unsaturated or has been subjected to very high temperatures for a prolonged time. It is important to check the level of residual processing aids and catalysts. Processing aids include solvents, filter aids, and acids/alkalis used to neutralize the material after esterification or other reactions. Catalyst residuals may be strongly acidic or alkaline or contain heavy metals such as nickel or tin.

26.5.2 Chemical Properties to Consider when Selecting Emollients

Chemical reactions usually need a reaction center and one or more reactants and frequently the presence of a catalyst. In emollients, the reaction centers can either be located in the hydrocarbon chain or it can be one or more of the functional groups present in the structure. Only two reactants will be considered in this review: oxygen and water. The two reaction types are consequently oxidation and hydrolysis, two reactions

that degrade the emollients and can have large impact on the quality and shelf life of the formulation.

26.5.2.1 Oxidation: Rancidity and Skin Damage

In emollient technology, oxidation can be regarded as a simple reaction between oxygen and the emollient. The oxidation results in breakdown products which can be smelly or irritating, depending on their size and volatility. Oxidation can also cause changes in the color and appearance of the formulation. In the worst case, oxidation can cause changes in metabolic pathways in the skin, resulting, for example, in inflammatory reactions, mutations, and apoptosis [40, 41]. Understanding emollient oxidation is therefore important for the formulator and product designer.

Oxidation requires an activated reaction center to proceed rapidly. Saturated hydrocarbons contain few activated carbons that can serve as reaction centers and are usually resistant against oxidation. The introduction of one double bond creates two active reaction centers adjacent to the double bond, and oxidation rates are more than doubled. When more double bonds are added, more active reaction centers are introduced and the oxidative stability decreases rapidly. Carboxylic acids and hydroxyl groups also contribute to active reaction centers. In case an oxygen molecule reacts with an activated carbon moiety, a hydroperoxide is formed. This hydroperoxide is a primary oxidation product and is measured by the “peroxide value” which is normally stated as a quality parameter for emollients.

If more oxygen is available, the oxidation process can proceed in two directions: formation of additional hydroperoxides or further oxidation of the hydroperoxide to form aldehydes, ketones, and hydrocarbons. This secondary oxidation breaks the hydrocarbon chains and produces fragments of different sizes and polarities. Fragments with low molecular weights are volatile and enter the headspace above the emollient. These volatile aldehydes and hydrocarbons have low odor thresholds and can be detected by the human nose in concentrations that are in the ppb-ppm range. If the oxidized fragment is larger, it

does not volatilize and stays in the emollient. These components are usually not smelly, but they can contribute to skin irritation.

Oxidation is catalyzed by metal ions, by certain enzymes and by the presence of oxidation products (autocatalysis) [42]. Iron and copper are the most active oxidation catalysts seen in emollient systems, and already, ppm levels of these metals can cause problems. Oxidation is also catalyzed by UV radiation and by visible light (photooxidation). Unstable emollients, such as highly unsaturated oils, as well as sensitive actives need to be formulated with care and the product packaged in opaque packaging to prevent photooxidation.

Oxidation can be prevented by the use of antioxidants. Most antioxidants are free radical scavengers and will eliminate the oxygen before it reacts with the activated reaction centers, alternatively deactivating the hydroperoxide and preventing the secondary oxidation. A good principle to prevent oxidation in formulations is to combine a water-soluble antioxidant with an oil-soluble one. Further protection is obtained if a surface active antioxidant is added to act at the interface between oil and water. The choice of antioxidant is often regulated by the legal restrictions, and care must be taken to use only permitted antioxidants.

Avoiding oxidation is also important from a product safety aspect. Free radicals and reactive oxygen species can start and maintain inflammatory reactions in the skin. They can also contribute to the oxidation of proteins and structural lipids in the skin, reducing the ability of the skin to maintain its elasticity and moisture barrier properties.

26.5.2.2 Hydrolysis of Esters Gives Texture Changes in the Formulation

A second chemical reaction of importance for ester-based emollients is hydrolysis. Obviously, hydrocarbons and alcohols do not react with water, but all esters are more or less sensitive towards hydrolysis. The hydrolysis is the reversed version of the esterification reaction: water reacts with the ester bond and liberates the acid and the

alcohol. As this is an equilibrium reaction, esterification occurs if the water is continuously removed while the hydrolysis takes place when there is an excess of water. Hydrolysis of esters is catalyzed by strong alkalis, strong acids, and by enzymes such as lipases and esterases. Most esters are stable in the pH range 5–8, especially if they are encapsulated inside emulsion droplets. Hydrolysis of esters in skin care formulation can cause the emulsion structure to change, leading to texture changes and instability. The pH value of the formulation drops, and the liberated acid or alcohol can increase the irritancy of the formulation. High molecular weight esters with low polarity have better hydrolysis resistance as the ester bond concentration is lower and because water solubility in the ester is restricted. Formulating with barrier forming emulsifiers and polymers can also protect the oil phase against hydrolysis.

26.5.3 Selecting Emollients Based on Physicochemical Behavior

Controlling the polarity and rheology of the emollient blend in a formulation is necessary in order to obtain the desired skin feel and stability of the product. Many practical emollient mixtures contain high- and intermediate-melting components which give a semisolid consistency to the ingredient. Understanding crystallization and melting behavior for texture and rheology control is important, especially when formulating anhydrous ointments and high emollient content W/O emulsions.

26.5.3.1 Melting Points Versus Melting Ranges

For many semisolid emollients and for mixtures of emollients, the melting behavior is an important selection criterion. As most emollients and emollient mixtures have complex compositions, the melting point is no longer a good characteristic for comparison. In this case, the melting range or the melting profile is a more appropriate parameter. The melting profiles can easily be measured using differential scanning calorimetry

(DSC), also in formulations. The DSC results, when standardized, give a direct comparison of the melting behavior of an emollient mixture and can also give insight into interactions between the emollients and emulsifiers. Pure emollients and emollient blends can be characterized using low-resolution NMR techniques. These measurements are strongly influenced by water so the measurements are restricted to dry emollients and to anhydrous systems.

Analysis of the emollient melting behavior can be used to optimize the formulation properties. Important temperature ranges to consider are storage temperatures (normal, too low, too high), body temperature, and temperatures during product stability testing. The formulation should be possible to dispense from its container (at storage temperature), meaning that the solids content must not be too high. On the other hand, in order to achieve high temperature stability and stability at varying storage conditions, increasing the solids content can be the solution as the solids will help the emulsifier and stabilizer system to maintain consistency. Finally, the solids can also be used to modify skin feel and moisturization. The higher the solids content, the heavier the skin feel is, but the perceived moisturization is improved.

26.5.3.2 Optimizing the Polarity by Mixing Emollients

As the polarity of an emollient or an emollient mixture is difficult to measure directly, the selection of emollients to optimize properties is at best a semiempirical procedure. In general, the polarity is additive, meaning that mixtures of emollients have properties proportional to the mixing proportions. However, some properties associated with polarity are strongly influenced by the purity of the system. For example, interfacial and surface tension of an emollient is sometimes used to evaluate polarity. These two parameters are strongly influenced by the presence of small amounts of surface active contaminants, and the measured values can be strongly misleading if the emollients are not properly purified before the measurements are taken.

In order to manipulate the skin feel and the solubility of actives, changing the polarity of the emollient system is a useful tool. A hydrocarbon-based emollient is normally nonpolar, and adding a more polar emollient such as an ester or a fatty alcohol can strongly influence the behavior.

26.5.3.3 Controlling Viscosity and Lubricity for Improved Skin Feel

Viscosities of emollient blends in the liquid state are normally additive, and the viscosity of the blend usually reflects the composition of the mixture and its individual constituents. Viscosity and perceived lubricity are often connected, but quite often there is an optimal viscosity associated with best skin feel and other sensory properties. If the emollient viscosity is too low, the skin feel is thin and watery while too high viscosities are perceived greasy and unpleasant. For sensitive skin, a high viscosity can also be disadvantageous as the drag will be perceived as irritating. High emollient viscosity is often preferred to reduce emulsifier and stabilizer content. High-viscosity oils can reduce emulsion instability, especially as higher viscosity is also often associated with higher density.

26.5.3.4 Spreading and Spreadability

Spreading and spreadability of the formulation on skin are important parameters for consumer appeal and acceptance. A formulation should be easy to apply, spread rapidly on the skin, and give a rapid disappearance from the skin surface. The spreading ability of emollients and emollient mixtures directly on skin is difficult to measure accurately, and various techniques have been applied to estimate the spreading properties [10, 43]. Spreadability and viscosity are dependent on each other as a rapid spreading is often observed for low-viscosity emollients. The rapid spreading is also associated with lubricity or slip, again favoring emollients of low viscosity. Equilibrium spreadability, on the other hand, is a different parameter and also important for the perceived emolliency of the formulation. High equilibrium spreading will yield a thin film on the skin surface and may lead to a disappearance of the perceived emolliency and moisturization over time.

26.5.4 Environmental Properties

Although the environmental properties of emollients are usually not considered to be a primary concern for skin care products, recent changes in legislation, especially in Europe, has brought the focus to this issue. When considering the use of skin care product, it is obvious that part of the emollients and other ingredients are absorbed into the skin, while other parts are rubbed off into clothes or washed off during showering and cleansing procedures. Other aspects concern the manufacturing, storage, and transport of emollients in large scale and the results locally and regionally of accidental discharge of chemicals into the environment. All this points to the importance of environmentally safe ingredients, and emollients, being used in relatively high concentrations in the formulations, are of special concern.

Biodegradability has become an important property for emollients and other cosmetic ingredients in the past few years. In a sustainability perspective, with life cycle analysis as a defining principle, not only the production and use of chemical substances but also the disposal after use and accidental discharge into the environment have become important to consider. Most straight-chain hydrocarbon-based emollients are readily degraded by sewage bacteria and can be considered readily biodegradable. This is also the case for straight chain fatty acids and alcohols. Esters are usually first hydrolyzed to the constituents which are then degraded. High-melting and high molecular weight substances are generally degraded more slowly due to low solubility and bioavailability. Although ultimately biodegradable, they can sometimes cause visible problems and disturb flora and fauna locally.

Another aspect of biological degradation is the possibility for substances to accumulate in the environment. Lipophilic substances, such as emollients, have a higher potential for bioaccumulation. However, since chemical structures that are known to cause toxic metabolites and show a high tendency to accumulate in the environment are today not used as emollients, most

cosmetics are also safe in this aspect. Again, high molecular weight lipophilic substances with high melting points are more of a problem, especially since the data available is scarce.

Conclusions

Emollients are necessary ingredients for skin care preparations, and the selection of ingredients is a difficult task when optimizing properties and performance. This chapter has reviewed hydrocarbons, alcohols, and esters used for emolliency and pointed to important chemical and physical properties that must be considered when selecting emollients.

Each group of emollient technologies has its benefits and disadvantages, and it is obvious that the perfect emollient does still not exist. A skilled formulator can utilize the benefits of each emollient while minimizing the adverse effects. It is also evident that many of the properties of a finished formulation are the result of ingredient interactions, and the winning formulation is often achieved by skillful manipulation of the interactions.

Take Home Messages

When reading this chapter you should learn about the following points:

- Emollient classification based on chemistry of physical properties
- The importance of optimizing physical properties for the application
- The chemical reactions that must be considered for emollients
- How emollients are produced
- Different raw materials for emollients
- Considerations when selecting emollients

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