

REVIEWS

Silicones in Cosmetics¹

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Abstract—The structural diversity of organosilicon compounds used in decorative and care cosmetic products is studied. The unique physicochemical properties of silicones allowing their application as emollients, moisturizers, emulsifiers, film formers, viscosity regulators, and antistatic and binding agents are described. Mechanisms of action of various structure silicones, due to which organosilicon compounds are advantageously used to impart certain properties to cosmetic products, are demonstrated. A comparative analysis of the efficiency of silicones versus natural compounds serving similar functions in cosmetic products is performed. The issue of silicone safety for human health is considered.

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INTRODUCTION

Silicones are polymers constituted of repeating units composed of silicon and oxygen atoms (Si–O–Si) in the polymer backbone and organic radicals (Me, Et, Ph, etc.) in the side chain. Silicone compounds were synthesized back in the 1960s; however, their synthesis on a serious industrial scale begins in 1940 primarily due to works by E.G. Rochow [1, 2]. In our country, the main contribution to the industrial production and application of silicones was made by K.A. Andrianov [3]. At present, the annual world production of silicones, in particular, methylsiloxanes, has already exceeded 10 million tons [4]. Silicones possess exceptional physicochemical properties, such as high thermal and chemical stability, bioinertness, and low temperature dependence of physicomechanical properties. This provides their widespread application in the most diverse industries, for example, construction, medicine, the food industry, the automotive industry, and other fields.

In the past decade, silicone polymers have become increasingly important as ingredients of cosmetic products, although the history of their application dates back to the 1960s, when first patents on the use of organosilicon compounds as ingredients of lipsticks were published [5–7]. At that time, the toxicological effect of silicones on living organisms was also intensively studied. Many publications summarized by M.G. Voronkov in the book “Silicon and Life” [8]

demonstrated the biological inertness of most silicones which laid the basis for their application in the cosmetic industry. For example, in our country, siloxane fluids of various brands well compatible with vitamins, essential oils, and other kinds of cosmetic raw materials were in wide use [9, 10]. These compounds were used in lipsticks and moisturizing and protective creams. For example, protective hand care cream *Silikonovyi* was manufactured by the domestic factory Svoboda; it is still available and is in demand now [11].

At present, the global cosmetics market grows every year and, according to AdIndex Market, it is already above \$450 billion. The growing popularity of silicones in cosmetic products in the last few years is largely associated with the ongoing research of their unique functions. A deep understanding of the properties of silicones in combination with an increased ability of experts to modify compounds and synthesize silicones of various architectural forms [12–14] leads to a more effective application of these compounds in various cosmetic products. Specifically, silicones and their derivatives are used as emollients, moisturizers, surfactants (emulsifiers), antistatic and binding agents, film formers, defoamers, and viscosity regulators [15]. The physiological inertness of silicones is especially favorable in terms of cosmetic application and chemical compatibility with skin. Silicones are also employed in hair care products, shower gels, antiperspirants, deodorants, shaving products, decorative cosmetics, etc.

¹ This paper was prepared for the special issue dedicated to the 100th anniversary of birth of A.A. Zhdanov.

Table 1. Types of silicones depending on the structure of units

Unit designation	Structural features	Structural formula
M	Monosubstituted (one oxygen atom per silicon atom)	$\begin{array}{c} \text{R} \\ \\ \text{R}-\text{Si}-\text{O}\sim \\ \\ \text{R} \end{array}$
D	Disubstituted (two oxygen atoms per silicon atom)	$\begin{array}{c} \text{R} \\ \\ \sim\text{O}-\text{Si}-\text{O}\sim \\ \\ \text{R} \end{array}$
T	Trisubstituted (three oxygen atoms per silicon atom)	$\begin{array}{c} \text{R} \\ \\ \sim\text{O}-\text{Si}-\text{O}\sim \\ \\ \text{O} \\ \\ \sim \end{array}$
Q	Tetrasubstituted (four oxygen atoms per silicon atom)	$\begin{array}{c} \sim \\ \\ \text{O} \\ \\ \sim\text{O}-\text{Si}-\text{O}\sim \\ \\ \text{O} \\ \\ \sim \end{array}$

R = H, Me, Ph, Vin, Allyl, etc.

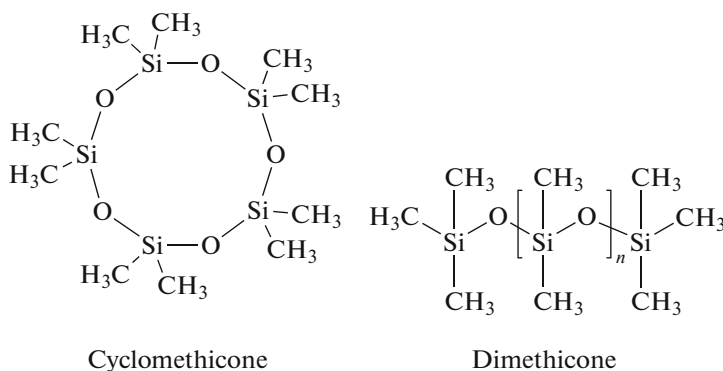
CLASSIFICATION AND STRUCTURE OF SILICONES

Term “silicone” in the cosmetic industry is frequently used with respect to diverse ingredients exhibiting different solubility and distinctive properties. They can be classified as follows: silanes; hydroxyl-containing compounds (silanols); cyclic diorganosiloxanes; linear polyorganosiloxanes; and copolymers and silsesquioxanes [16].

For convenience, silicone chemistry has its own “language” developed by Alfred Stock in 1964. This nomenclature is built using letters M, D, T, and Q and

is based on the type of groups comprising the molecule framework. Table 1 presents decoding of the mentioned designations.

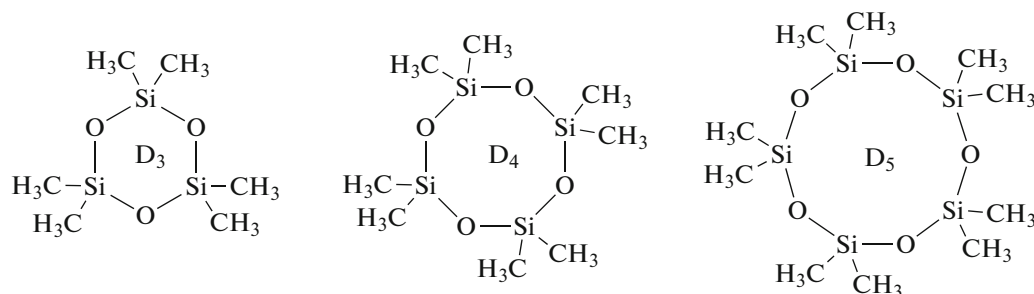
The most common ingredients of cosmetic products are methyl-substituted siloxanes known as polydimethylsiloxanes. They have structures of the two types: linear and cyclic [17, 18]. As an example, below we present the structural formulas of the two best-known silicones, cyclomethicone and dimethicone, with the cyclic and linear structure, respectively:



It is seen that these compounds are composed of type D units. Cyclic compounds are usually simply

referred to as D₃, D₄, D₅, etc., where index denotes the number of D-units in a cycle.

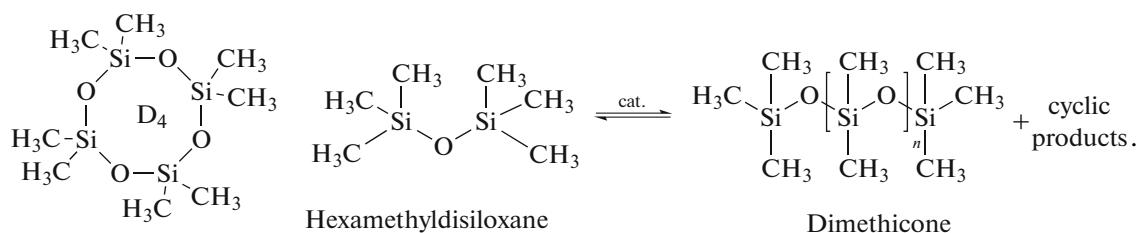
Term “cyclomethicone” refers to a number of cyclic silicone compounds:



Cyclomethicone is isolated from a mixture of hydrolysis products of dimethylchlorosilane. Octamethylcyclotetrasiloxane (D₄) is the prevailing product, while hexamethylcyclotrisiloxane (D₃) and decamethylcyclopentasiloxane (D₅) are present in smaller amounts. Cyclomethicone is available in the form of various compositions: either individual D₃, D₄, and D₅ or a more common and cheap mixture

containing 85% D₄ and 15% D₅. However, in the cosmetic industry just cyclomethicone D₅ has received the most widespread use, since it exhibits a number of favorable properties for use in cosmetics compared with other cycles [19, 20].

Dimethicone is oligodimethylsiloxane, which is synthesized by the equilibrium ring opening ionic polymerization of D₄:



Organosilicon compound hexamethyldisiloxane (HMDS) is generally used as a chain-terminating agent. By varying the D₄-to-HMDS ratio different viscosity dimethicones can be obtained; this enables their use in cosmetic products for various purposes.

ORGANOSILICON DERIVATIVES IN THE COSMETIC INDUSTRY

The composition and structure of organosilicon compounds are very diverse which entails a great variety of physicochemical properties of these compounds. Therefore, they are in widespread use in various areas of human activity. The cosmetic industry is not an exception. Along with cyclomethicone and dimethicone, polydimethylsiloxanes derivatives containing various organic substituents and terminal functional groups in the main chain at the silicon atom and nonlinear silicones are used in cosmetic products. Phenyltrimethicone is among these compounds. In chemical terms, phenyltrimethicone is oligomeric phenylsilsesquioxane (T-unit) with terminal trimeth-

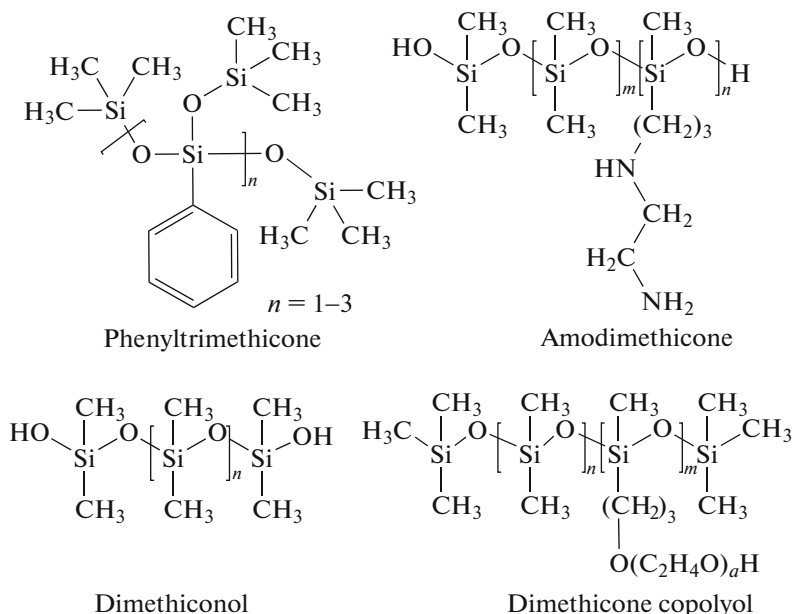
ylsilyl (M-unit) groups. It can be found in hair conditioners, sunscreens, and decorative cosmetics. Due to the presence of phenyl groups phenyltrimethicone has a high reflective index [19] which imparts hair with extra shine. Phenyl groups also ensure the effective application of phenyltrimethicone in sunscreens [21]. This component also exhibits thermal protection properties, that is, protects hair from heat damage, such as drying with a hair dryer and hair straightening and perming. Phenyltrimethicone reduces excessive foaming in skin and hair care products, and it has proved to be good in antiperspirants as an agent that prevents white spots and stains on clothes. It is an ingredient of care and decorative cosmetic products manufactured by global famous companies (L'Oréal, Schwarzkopf, etc.).

Another abundant ingredient of cosmetics, in particular, hair conditioners, is amodimethicone. This is PDMS containing units with amino groups [19]. These groups contribute to prevention of hair color degradation that may occur under exposure to UV radiation or due to frequent head washing and ensure

conditioning properties and reduction in drying time without adversely affecting hair structure and volume [22]. Products containing amodimethicone also exert the antistatic effect. Amodimethicone is an ingredient found in care products of various brands manufactured by Unilever, Astoria Kosmetik, L'Oréal, etc.

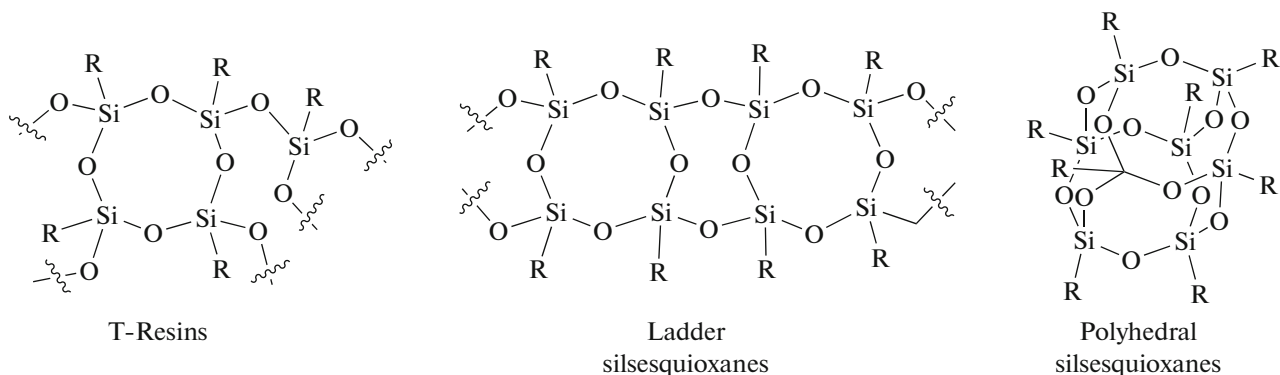
In terms of application relevant silicones are dimethiconol, which represents polydimethylsiloxane with terminal hydroxyl groups, and dimethicone copolyol—polydimethylsiloxane containing poly(eth-

ylene oxide) and/or poly(propylene oxide) fragments in the side chain [23]. These compounds are used to improve lubricating effect in richer, more nourishing skin care products, among which are night creams or after sun products [24]. Dimethiconols form water-resistant films that can extend skin care, sunscreen, or decorative products. They can be found in the products manufactured by Roche Holding, Natura Siberica, Payot, Clarins Fragrance Group, L'Oreal, etc.:



In the context of this review, it is impossible not to mention another unique class of organosilicon compounds—polyhedral oligosilsesquioxanes (POSSs/SSQs). Organosilsesquioxanes (OSSQs) by definition are compounds of the general formula $[RSiO_{3/2}]_n$. However, this formula does not reflect a great variety of structures that can be formed during

their synthesis under various reaction conditions. Silsesquioxanes may represent disordered polymer networks usually called polyorganosilsesquioxanes, also known as T-resins [25]. Sometimes ordered ladder structures are formed. Under controlled conditions synthesis enables preparation of well-defined molecular oligomeric polyhedral silsesquioxanes (**POSS**):



(R = Me, Ph, OEt, OH, NH₂ etc.)

It should be mentioned that the simplest form of POSS with the hydrogen atom at silicon has limited use in cosmetics because of low stability and solubility [26]. The attachment of organic groups to the core improves the solubility [27] and chemical stability of POSSs, rendering them more suitable for application as additives to skin care products. The introduction of either hydrophilic or hydrophobic groups can facilitate a change in the lipophilic balance. Groups R used as substituents at the silicon atom in POSSs may comprise a tertiary nitrogen atom, zwitterionic compounds, and various functional organic groups [28]. Thus, a large number of compounds with various properties can be synthesized [29] which can potentially be used in personal care products.

There is currently a lack of publications describing the application of POSS derivatives in cosmetic products. However, in recent years the number of patents on cosmetic products containing POSS derivatives has been growing rapidly. The earliest submitted patents date back to the later 1960s—early 1970s. In those years, General Electric (GE) and Dow Corning (DOW) [30] were leaders in the application of organosilicon compounds. Even then it was clear that POSSs in cosmetic products contribute to improving their quality, for example, in solid decorative cosmetics [31]. At the early 1990s first patent applications of the French company L'Oréal were published [32]. Thus, POSS derivatives have found use in various cosmetic products—decorative cosmetics, creams, lotions, sunscreens, and hair and nail care products [33].

The disordered OSSQs structures of the T-resin type are also actively used in the cosmetic industry, for example, propylsesquioxane (**PSSQ**). This compound is commercially available and is manufactured by Dow Corning in the form of dispersion in a silicone or a hydrocarbon solvent under trademarks DC670 (50% PPSSQ/D₅) or DC680 ID (72% PPSSQ/isododecane) [34]. Initially, the polypropylsilsesquioxane resin was used by L'Oréal in long-lasting foundations [35]. Owing to their loose network structure and low softening and glass transition temperatures [36–39] a T-resin is tacky and forms a cohesive adhesive film upon drying. Furthermore, PPSSQ is dispersed in a solvent; therefore, it acts as a liquid binder for pigments and fillers in cosmetic products, providing good adhesion to skin. Thus, after solvent evaporation the product forms a smooth, flexible, and strong film on the skin. T-resins are used in decorative cosmetics in products, such as lipsticks, eye shadows, and eyeliners.

Another class of organosilicon compounds that has gained recognition in the production of decorative cosmetics is MQ-copolymers, more known as MQ-resins. Name “MQ” originates from its structure composed of mono- (M) and tetra- (Q) functional siloxane fragments [40–46]. MQ-resin is brittle and cannot form films by itself; therefore, a plasticizer is needed for compositions containing MQ. MQ-resins

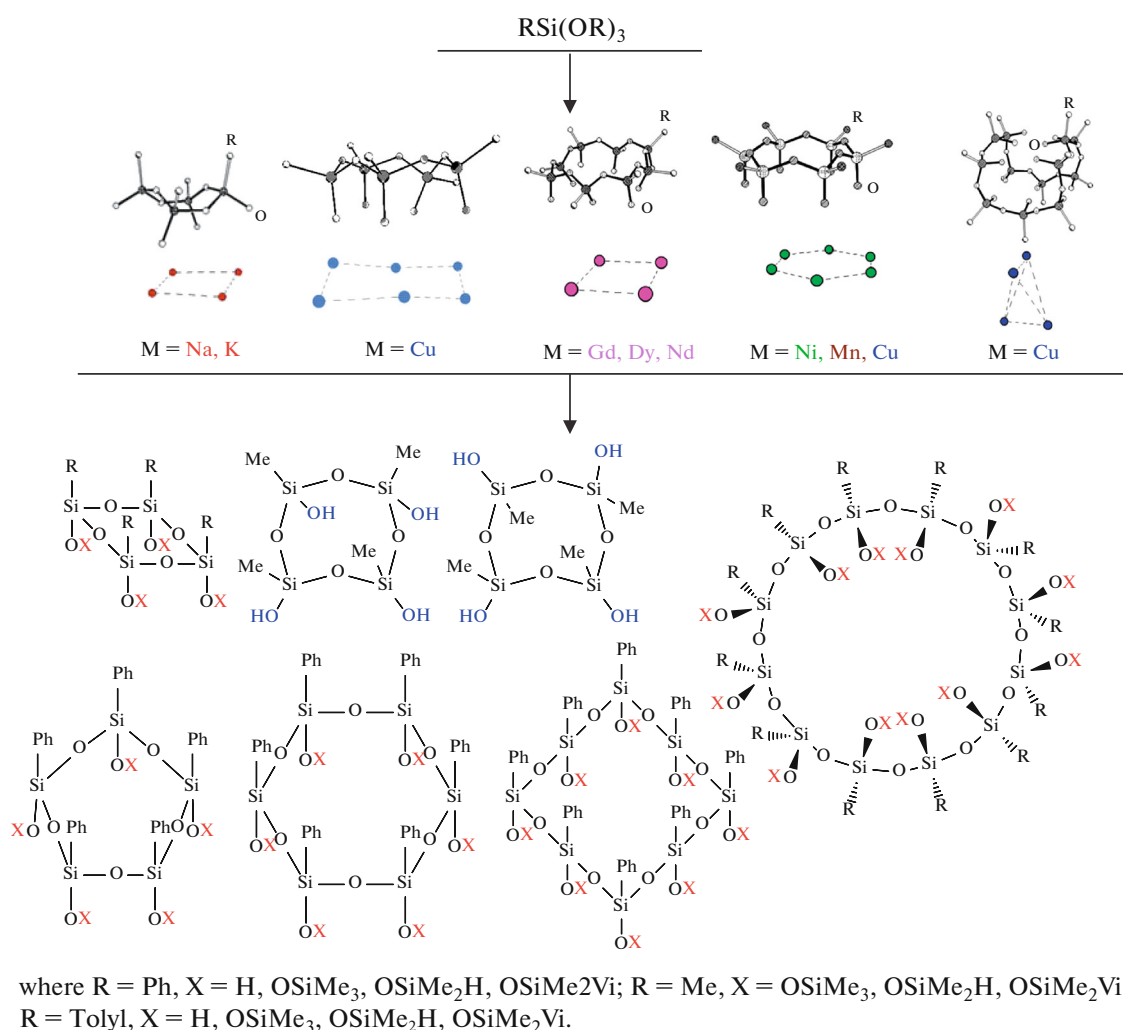
assure exceptional wear resistance and adhesion of cosmetics due to their low free surface energy similar to the free surface energy of human skin. The application of MQ-resins in cosmetics was first proposed by Shiseido [47], and over the last three decades this has been reflected in many color cosmetic products, such as long-lasting lipsticks and foundations. The interaction of MQ with solvents (PDMS or low-viscosity hydrocarbon oils), pigments, and fillers provides good adhesion to skin; that is, on the skin surface a cosmetic product forms a flexible film that exhibits high hydrophobicity and resistance to oils, sweat, and skin fat. MQ-resins ensure not only excellent stability of the cosmetic product and also its matte texture; therefore, they are ideal for use in foundations to attain lasting color and matte finish. Currently, MQ-resins are employed as ingredients of products manufactured by L'Oréal, Estée Lauder Companies Inc., etc.

Obviously, the application of silicones in cosmetics is not limited by the above-described structures. They may also contain silicones bearing a wide range of functional groups: alkyl substituents with various hydrocarbon chain length, ester groups, including triglyceride derivatives, fluoro-containing and poly(ethylene glycol)/(polypropylene glycol) fragments, carboxyl groups, charged fragments containing, for example, a quaternized nitrogen atom, and many other substituents [16, 48], and dimethicone copolymers of various structure.

NOVEL STRUCTURAL FORMS OF SILICONES

As was shown above, one of the main features of silicones is that a specified structure can be formed depending on synthesis conditions. The final structure determines properties of the target polymer. This section will cover examples of the synthesis of fundamentally new structural forms of organosiloxanes which may be promising for use in cosmetics.

Among all organosiloxanes organosilsesquioxanes are the most structurally diversified. Polyhedral, ladder, and random OSSQs were described above. These structural forms of OSSQs have been known for a long time. The polycyclic structure is inherent in their macromolecules. Another bright representative of cyclic PSSQs is stereoregular macrocycles. The approach developed by the authors of [49] allows the synthesis of these compounds with various cyclic core size and dissimilar substituents [50–54]. The general scheme for the synthesis of stereoregular organocyclosilsesquioxanes can be outlined as follows:



Structural features of the above macrocycles will permit their derivatives to exceed many times their irregular and linear counterparts in such characteristics as the possibility to control the thermal properties and viscosity of solutions and melts in a wide range. Using the example of star-shaped siloxane polymers, in which stereoregular organosilsesquioxanes function as branching centers, it was shown that in the case of these compounds the crystallization of siloxane chains can be suppressed for polymers with molecular weight $M > 20 \times 10^3$. Note that the intrinsic viscosity of star-

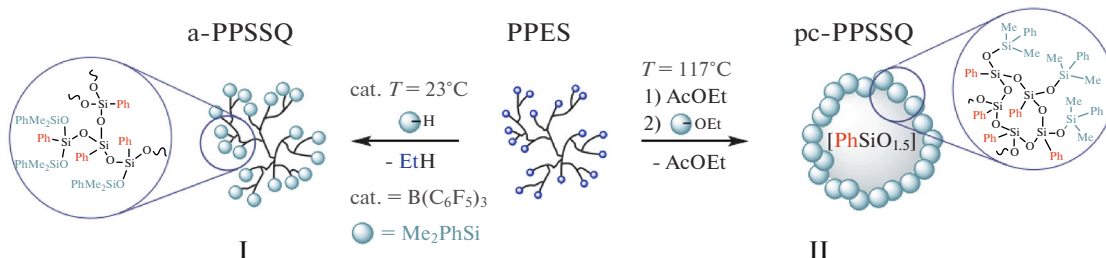
shaped siloxanes can be an order of magnitude lower than that of their linear counterparts [52–56]. Another characteristic feature of derivatives of stereoregular organocyclosilsesquioxanes is that their structure allows the introduction of hydrophilic and hydrophobic substituents into a single molecule, thereby producing unique surfactants [57].

In contrast to all above-mentioned OSSQs, hyperbranched (I) and comb-like (II) polymers have a fully acyclic structure:



As was shown in [58–66], these compounds feature unique properties among which are a low viscosity of melts and solutions, a weak dependence of hydrodynamic radius on molecular weight, and the ability to encapsulate low molecular weight compounds. Moreover, the novel structural forms of organosilsesquioxanes can easily be modified; thus,

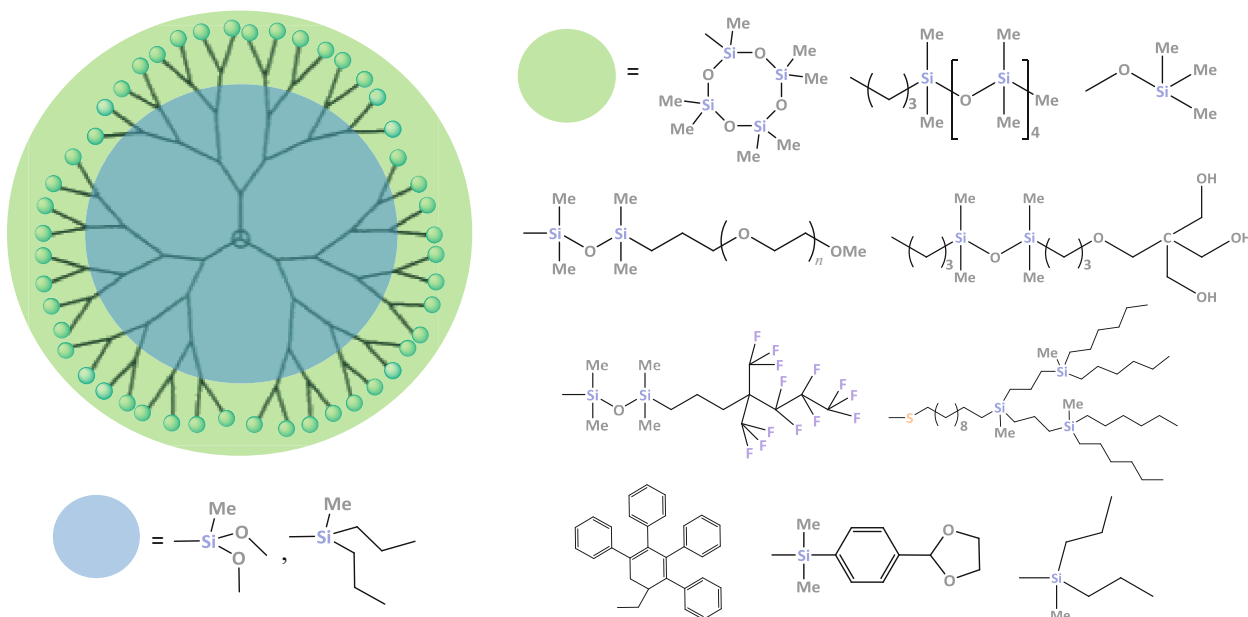
almost any desired properties can be imparted to the final polymer. Modification can proceed via both terminal functional groups with the retention of polymer acyclic structure and intramolecular cyclization. The scheme of synthesis of acyclic (I) and polycyclic (II) forms of polyphenylsilsesquioxane (PPSSQ) based on polyphenylethoxysiloxane (PPES) [62] is as follows:



The latter method provides the synthesis of particle macromolecules (nanogels) with the core-shell structure. These nanogels differ quantitatively from their acyclic analogs in properties [62, 67, 68]. Specifically, the dynamic viscosities of hyperbranched ($M = 2.15 \times 10^3$) and nanogel ($M = 2.86 \times 10^3$) PPSSQs with comparable molecular weights differ by five orders of magnitude (0.38 and 0.15×10^5 Pa s, respectively) [62]. This indicates that the representatives of this class of OSSQs are extremely promising candidates for use in cosmetics, since they demonstrate unique properties that can be adjusted to the required tasks.

Presenting particle macromolecules, we cannot but highlight such unique branched compounds as dendrimers. They have a regular structure characterized by monodispersity close to ideal and a wide range of possible functional groups which makes them the sub-

ject of ongoing studies in the past several decades. Due to the structural diversity dendrimer macromolecules can be used as photosensitive sensors, catalysts in enantioselective synthesis, molecular nanoreactors, and drug delivery systems [69]. In the context of this review, special attention should be paid to just organosilicon dendrimers. These compounds are comprised of a siloxane or carbosilane core and an organoelement outer shell. Diverse synthetic approaches to the modification of organosilicon dendrimers make it possible to prepare hybrid compounds containing a wide scope of structural fragments on the outer layer: cyclosiloxane, trimethylsilyl, linear siloxane, hydroxydine, alkyl, phenyl, ester, etc. The structures of organosilicon dendrimers with various groups on the outer shell are presented below:



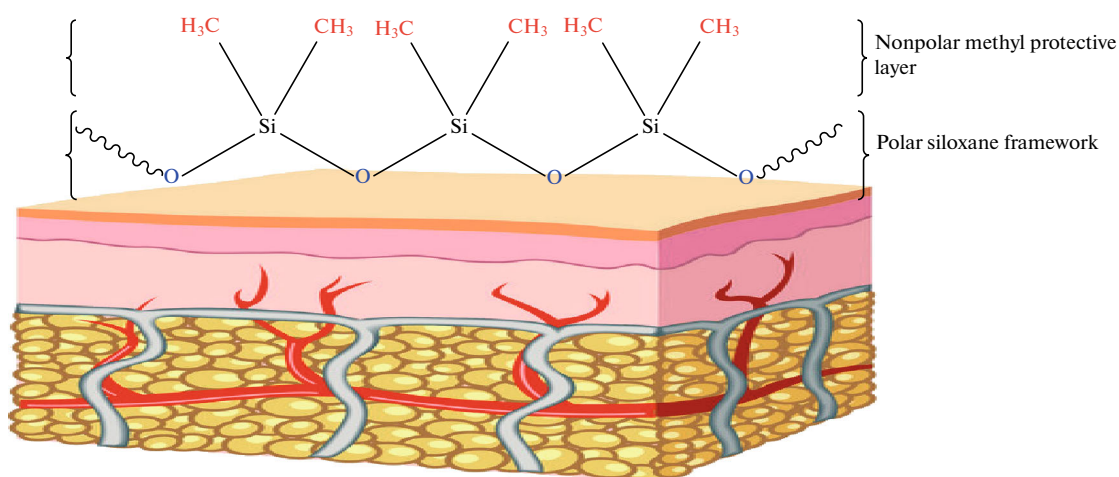
A number of works clearly showed that the properties of organosilicon dendrimers are determined by just the shell structure [70–78]. Thus, by changing the nature of the outer layer dendrimers can be provided with film-forming properties and their hydrophilicity, hydrophobicity, and rheological behavior can be controlled [79–83] which makes them very attractive for use in cosmetic products.

PROPERTIES, ACTION MECHANISMS, AND APPLICATION OF SILICONES

A popular application of silicones in cosmetics is related to their unique physicochemical properties. For example, cyclomethicone has an extremely low heat of vaporization ($\Delta H = 31$ cal/g) compared with water ($\Delta H = 539$ cal/g) and even alcohol ($\Delta H = 210$ cal/g) [16]. As a result, cyclomethicone rapidly and easily evaporates from the skin surface; therefore, it is included in deodorants and antiperspirants, hair-sprays, sunscreens, lotions, sprays, etc. A low viscosity of cyclomethicone ($\nu = 4.2$ cSt) also ensures easy and

uniform spraying of its formulations and the absence of oily spots on clothes and skin.

Another valuable property of silicones is their ability to reduce the surface tension of cosmetic products based on them. This property is especially important to promote the smearing of formulations designed for both hair and skin. F. Lardy et al. [84] and S. Budiasih et al. [85] showed that the lower the surface tension, the higher the fluidity of formulation applied to the skin. The surface tension of both linear and cyclic low molecular weight silicones is in the range of 17–22 dyn/cm², which differs appreciably from the surface tension of water (76 dyn/cm²) and mineral oils (32 dyn/cm²) [16]. The ability to reduce surface tension is a prerequisite to assuring other important surfactant properties (foaming, emulsification, and wetting). This also implies that upon contact with a polar surface, in particular, of skin or hair, a polar siloxane framework shifts to it, whereas methyl nonpolar groups are directed outwards, creating a protective layer and forming a common hydrophobic surface [20]:



This dual hydrophilic-hydrophobic behavior gives high spreadability and excellent film-forming properties to silicone oils, improving skin and hair moisturizing. A low surface tension of silicones also facilitates a good distribution of products based on them on damaged surfaces. For example, being components of hair conditioners and oils dimethicone and its derivatives fill microcracks of a damaged hair cuticle (Fig. 1), promote a healthy and tidy look of hair, make them visually thicker, impart smoothness and shine to them, and facilitate combing.

The same property determines a wide application of silicones in antiaging face creams. Silicone components fill skin folds and visually smooth out them. Thus, they successfully mask wrinkles and create the lifting effect; as a result, skin looks tightened and younger.

Silicone compounds are soluble in neither oil nor water. In the cosmetic product, they form the third phase which is simultaneously a hydrophobic and oleophobic component of the system. This key property renders cosmetic products based on silicones resistant to both oils and water; that is, they form a barrier to skin secretions and, at the same time, repel water and dirt from external sources; as a result, hair, for example, stay clean longer.

The siliphilic behavior of silicone compounds is often used in personal care products to improve emulsification which is of great importance in the manufacture of decorative cosmetics (eye shadows, lipsticks, etc.). Almost all pigments in the above products have a certain coating (oil or silicone). Coatings are applied to pigments to change their properties in terms of aesthetics and solubility. The ability to effectively

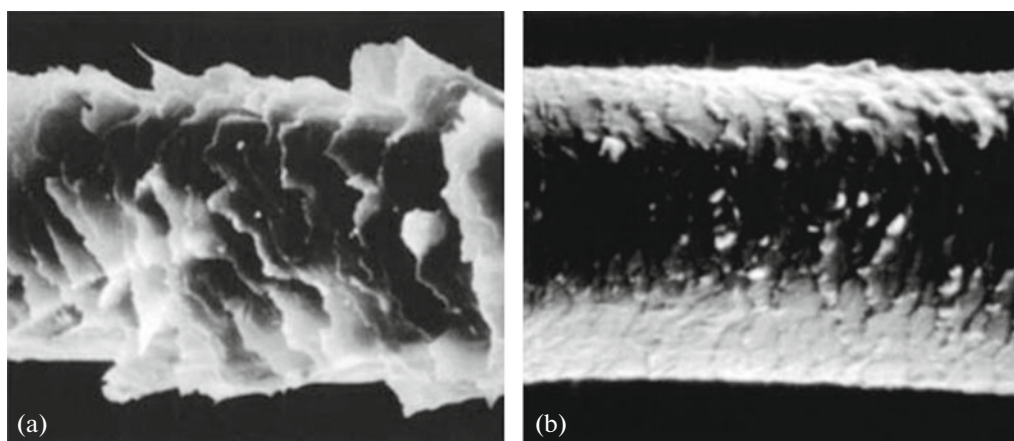


Fig. 1. Microphotographs of (a) damaged hairs and (b) hair treated with the silicone-containing product [16].

disperse a pigment in a cosmetic product is attained using a phase that most compatible with a coating. For example, a pigment with a silicone coating is well dispersed in the silicone phase. As a consequence, the pigment is distributed more uniformly, is easily applied, and adheres well to the skin.

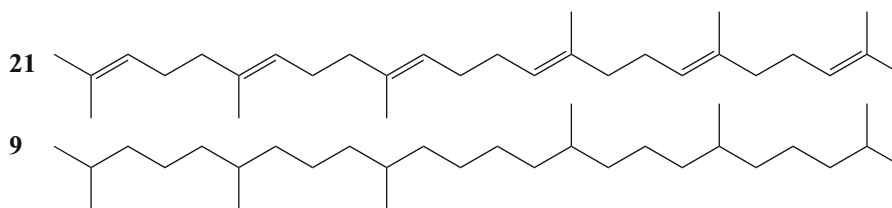
Almost all foundations contain silicones. This prevents them from drying out and makes foundations more plastic and easy to apply to skin. For example, due to low values of surface free energy the above-described T- and MQ-resins form films with a good adhesion to skin. The stability of MQ-resins can be explained by the fact that they form a tacky paste composed of MQ particles and fillers and fill irregularities on the skin surface. T-resins, on the contrary, form a liquid binding system, in which fillers and pigments can mix; this results in formation of a smoother film on the rough skin surface [34]. Thus, silicones present in the foundation smooth out irregularities and fill fine wrinkles; therefore, skin looks smoother and younger. They also make makeup last longer.

All the above features make silicones, specifically, cyclomethicone and dimethicone, effective and very popular ingredients of cosmetic products. Silicones are found in numerous care and decorative cosmetic products.

SILICONES VERSUS COMPOUNDS BASED ON NATURAL COMPONENTS

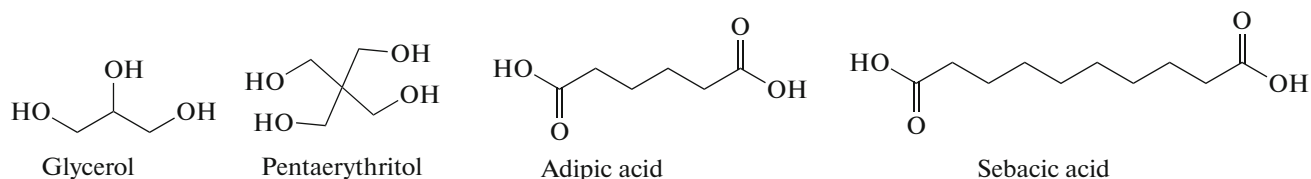
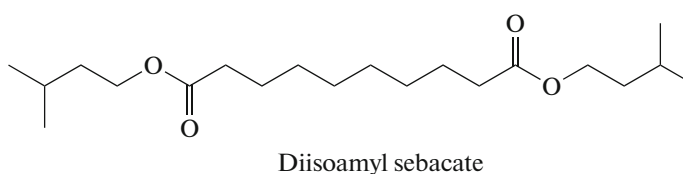
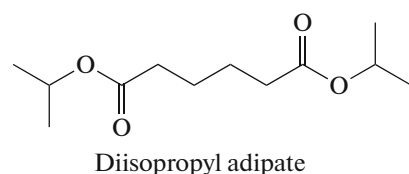
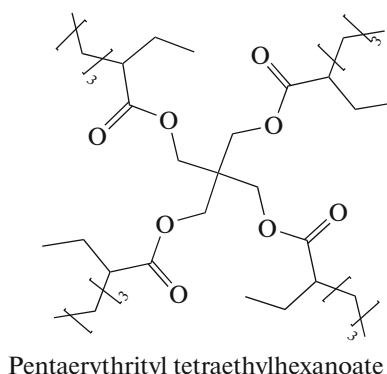
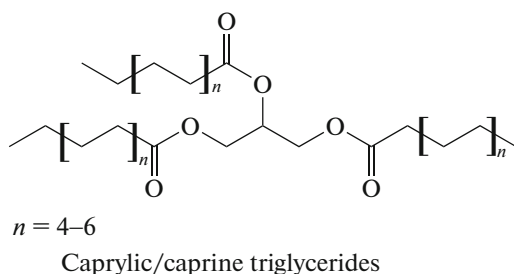
The unique properties of silicones render them almost versatile components of cosmetic products. However, whether it is possible to do without them? Let us consider the most commonly used analogs of silicones which can also serve as emollient ingredients in cosmetic products and their physicochemical properties.

Hydrocarbons are an important class of emollient ingredients known since the onset of the 21st century. For example, squalene 21 and squalane 9 are two examples of nonvolatile hydrocarbons of the triterpene series:



Squalene, which has long been extracted from short tip shark liver, is currently produced largely from olive oil [86]. Its hydrogenation product is chemically stable and inert squalane. Due to high viscosity ($\eta = 28.1$ mPa s) and nonvolatility ($T_b = 350^\circ\text{C}$) squalane manifests the occlusive behavior, facilitating strengthening of the barrier functions of the skin.

Essential oils represent the most important group of chemical compounds used as emollients [87–89]. A great diversity of initial acids and alcohols entails a wide variety of oils which explains their common use in cosmetic products. In addition, the use of polyols (glycerol or pentaerythritol) and polyacids (adipic or sebacic) further expands the list of possible candidates:

Alcohols and acidsEmollients based on esters

Depending on the length and functionality of carbon chains, the volatility of these emollients can be varied. For example, the effect of diisopropyl adipate is similar to that of cyclomethicone. Triglycerides represent a large class of ester emollients. They can be used directly as cosmetic ingredients [90] or the sources of alcohols and fatty acids. Further reactions with alcohols, polyols, or glycerol afford ethers, polyesters, and synthetic triglycerides, respectively. The combination of fatty acids and alcohols which yields esters having less than 18–20 carbon atoms is generally in demand because of their semivolatile nature. For example, coconut and palm oils are the main source of fatty acids with the average chain length (8–14 carbon atoms). Palm oil is the main source of palmitic and oleic chains. Nevertheless, its extensive use caused deforestation, especially in South America and Southeast Asia. At present, global organizations, such as World Wide Fund for Nature (WWF) or Roundtable on Sustainable Palm Oil (RSPO), confirm the need to limit its use. Other popular oils, such as soya, rapeseed, and sunflower oils, are the sources of longer chains, that is, oleic, linoleic, linolenic, and stearic acids. Esters formed from them are not volatile and in their properties are closer to linear PDMS with viscosity up to several tens of mPa s.

Fatty alcohols produced from triglycerides mostly consist of linear alkyl chains. They are usually used as auxiliary surfactants in emulsions. Fatty alcohols are also applied as rheological modifiers and texturing agents [90]. Actually, due to the presence of free hydroxyl groups viscosity increases and fatty alcohols as auxiliary surfactants commonly manifest emollient properties. Another subgroup of fatty alcohols is formed by Guerbet alcohols. These primary and β -branched alcohols synthesized by the oxidation of alcohols to aldehydes followed by the aldol reaction [91] are primarily used to change the consistency of final cosmetic products.

In general, all the above compounds do their function well in cosmetic products. However, whether their effect is comparable with the effect exerted by silicone compounds? The answer to this question lies in comparison of their physicochemical properties [20].

As is seen from Fig. 2, the emollients are situated in accordance with their boiling point and surface tension values which roughly reflect volatility and spreadability, respectively. Another property influencing spreading, namely, the dynamic viscosity, is depicted as the diameter of various dots. It is easily seen that competition with the physicochemical properties of silicone oils, especially with their very low surface tensions

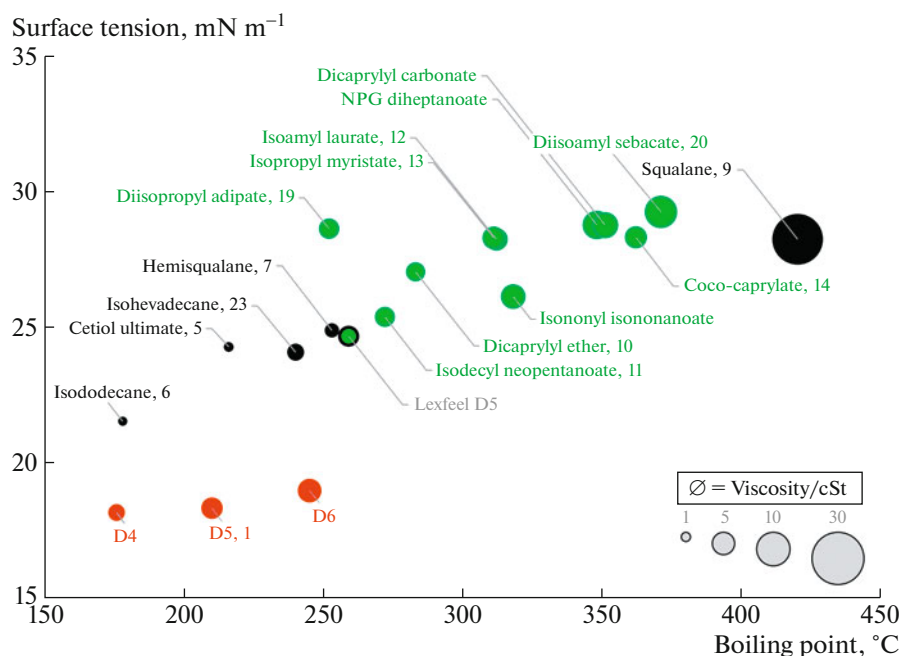


Fig. 2. Distribution of several typical commercial emollients in accordance with their chemical nature (cyclosiloxanes, alkanes, oxygen-containing emollient), boiling point, surface tension, and dynamic viscosity expressed by the size of dots [20]. Color figures are available in the electronic version.

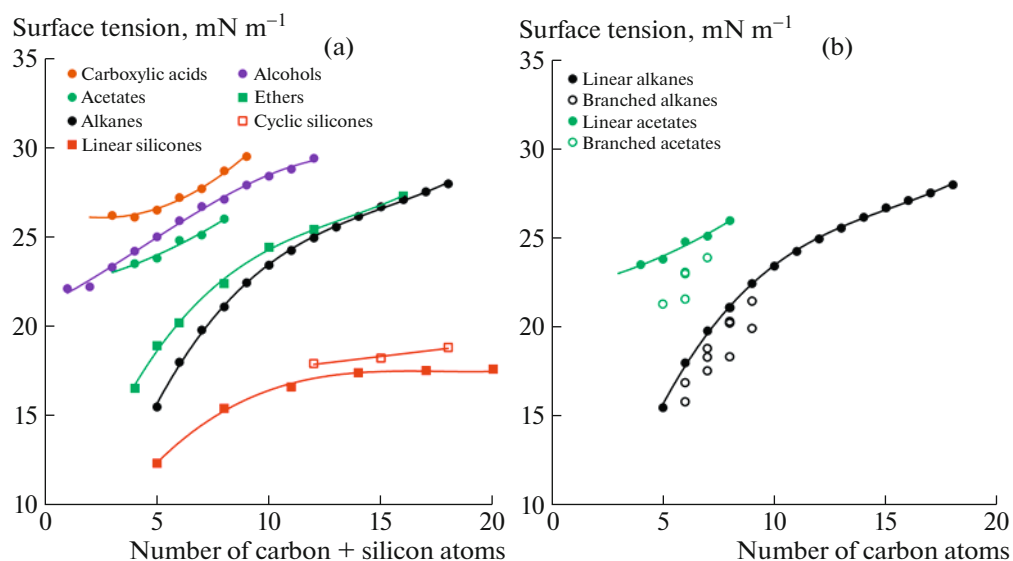


Fig. 3. Change in the surface tension of (a) homologous and (b) isomeric fluids with polar group nature, chain length, and branching degree [20].

(17–22 dyn/cm²), is hardly possible. Nonetheless, a strongly branched volatile hydrocarbon isodecane 6 (21.6 dyn/cm²) exhibits the closest physicochemical properties than all other commercially available emollients. Commercial emollients with physicochemical properties closest to the properties of volatile silicone oils are alkanes, ethers, and esters with short chains

(with 18 or less carbon atoms). It can be emphasized that the mixing of alkanes with ester (commercial product Lexfeel D5) is an effective strategy to approach the properties of cyclomethicones.

A large variety of available emollients is the direct consequence of the diversity of structural features, that is, polar group nature, chain length, and branching degree.

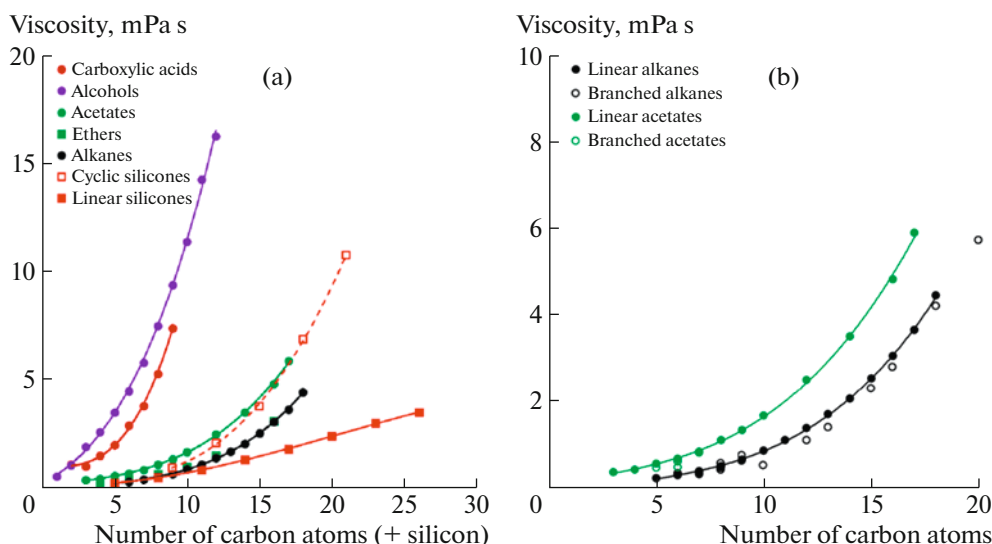


Fig. 4. Change in the viscosity of (a) homologous and (b) isomeric fluids with polar group nature, chain length, and branching degree [20].

These structural modifications have the greatest influence on the physicochemical properties of emollients. An analysis of the literature allows some structure–property relationship to be revealed. Changes in the values of surface tension and viscosity of oils with the number of carbon and silicon atoms are shown in Figs. 3 and 4 for corresponding groups of chemical compounds: alkanes, ethers, esters, acids, alcohols, and siloxanes. As expected, the values of both parameters increase with chain lengthening and enhancing polarity of molecules. A peculiar behavior of siloxanes results from the nonpolar screen created by methyl groups. Since the cyclic structure considerably decreases the degree of freedom, cyclic siloxanes have higher viscosities than linear ones [92]. It is also seen that branching of the chemical structure markedly reduces surface tension, whereas viscosity decreases slightly. Alkanes and ether compounds are characterized by extremely weak dispersion interactions which are responsible for their low values of surface tension and viscosity. In the case of polar ester groups, additional interactions arise, causing an increase in both parameters. Finally, free terminal hydroxyl groups of alcohols and carboxylic acids provide the formation of strong hydrogen bonds, increasing surface tension and viscosity.

Thus, it can be stated that an effective emollient containing no silicones should have a branched hydrocarbon chain, moderate polarity, and average chain length.

Another parameter, which is to be addressed to evaluate the efficiency of particular compounds in cosmetic products, is their volatility/nonvolatility. Recall that cyclomethicone D₅ has an extremely low heat of vaporization which makes it the effective ingredient of various sprays, antiperspirants, etc.

Figure 5 presents the volatility distribution of emollients competitive with silicones depending on their boiling points. The region including volatile compounds meets European standards on volatile organic compounds (the boiling point $\leq 250^{\circ}\text{C}$). The graph clearly shows that only two alkanes, namely, isodecane (C₁₂) and Cetiol Ultimate (a mixture of undecane and tridecane C₁₁–C₁₃), demonstrate volatility comparable with the volatility of cyclodimethicones D₄ and D₅. Despite high volatility these hydrophobic oils have no smell, since they are poorly soluble in nasal mucus. As opposed to alkanes, esters containing the same number of carbon atoms are better soluble in nasal mucus and, therefore, can interact with smell receptors. As a result, they possess a strong smell which is unacceptable for emollients. For this reason emollients carrying oxygen-containing functional groups should have from 17 to 20 carbon atoms, be branched in order to have almost no smell, and at the same time, be sufficiently volatile to easily evaporate from the skin or hair surface. However, none of modern emollients like alkanes, ethers, or esters cannot compete with cyclomethicone D₅ in properties. Actually, due to various reasons alkanes and oxygen-containing emollients are worse distributed on the skin than silicones. Alkanes have a low surface tension, facilitating spreading, but a high interfacial tension with skin unfavorable for spreading and vice versa for oxygen-containing emollients.

SAFETY OF USING SILICONES

Even though the issue of silicone toxicity was comprehensively studied in the 1960s–1970s, in recent years, disputes about the safety of silicones for human and environment have been happening more and

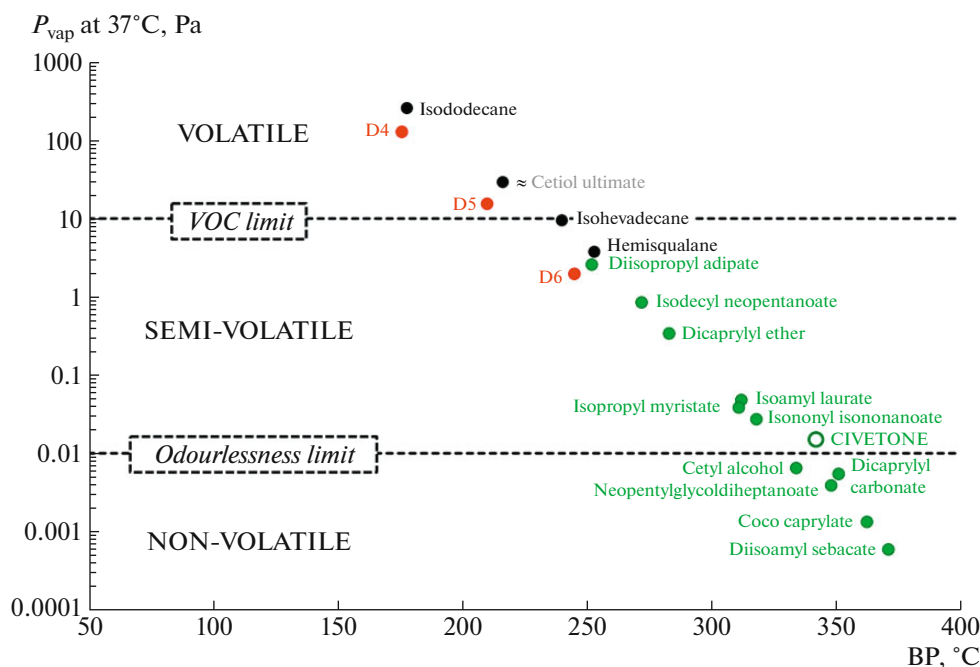


Fig. 5. Distribution of several typical emollients over volatility [20].

more frequently in the cosmetic industry and among scientists. Many myths and misconceptions about silicones have appeared. However, modern studies indicate that the use of silicones in medicine and cosmetic products is safe for organism. It is known that dimethicone and its derivatives are approved for use by the Food and Drug Administration. The European Commission also considers them safe ingredients for use in cosmetics [93]. Nevertheless, in the last decades, a real anti-propaganda of cosmetic products containing silicones has started only because of their synthetic rather than natural origin [17]. To attract more attention of consumers to their production many cosmetic companies began to focus on the pure environment concept, advertising their products as “chemical-free,” “green,” “natural,” etc. [94]. Certainly, silicones are poorly biologically degradable and their accumulation can be regarded as a long-term risk to the environment but in many cases this concept is taken to extremes, leading to conviction that silicones are fully toxic not only for the environment but also for human health [15]. Therefore, words “silicone-free” began to appear on the labels of many cosmetic products, leading the consumer to believe that the said product is safer than the products containing silicones. However, this is not the case!

There is an opinion that silicones clog pores and aggravate acne. However, according to scientific research, silicones are resistant to oxidation and non-occlusive; therefore, they are unable to seal pores and silicone oil does not affect bacterial growth [93]. On the contrary, silicones can form films on the skin sur-

face which can influence the penetration of active compounds into the skin and horny layers [95]. Therefore, some people argue that silicones can cause irritation. However, in fact, they are neutral for skin. In contrast, silicones reduce irritation [16]; therefore, they are used in soothing creams and ointments. For many years they have been applied in pharmaceutical formulations. For example, dimethicone is a component of well-known drugs against flatulence and colic [96].

It is stated that silicones accumulate on skin. Several silicones (e.g., silicone resins and silicone acrylates) create a film on the skin surface, but due to the unique structure of their molecules this film is fully permeable for water and oxygen vapors [19, 97, 98]. As a result, ideal conditions are created for skin regeneration, wound healing, and reducing scars [98, 99]. It is proved that this medium is suitable for the synthesis of collagen [100].

As evidenced by several scientific publications, silicones are suitable even for sensitive skin, since they do not irritate it, are hypoallergenic, noncomedogenic, and nonocclusive, and have no smell. Silicones can be found in auxiliary cosmetic products for eczema and acne treatment or baby skin care products, in particular in creams used to prevent and heal diaper rash [101]. Moreover, it was demonstrated that allergic reactions resulting from the use of silicones are very rare [48, 102] and are the exception rather than the rule. For example, in vivo studies of silicones also proved their safety [93]. Specifically, these authors measured transepidermal water loss (TEWL)—an

important parameter of skin integrity and degree of water evaporation from deep skin layers. The results showed that silicone-based emulsions are not occlusive and do not disturb the skin balance which allows nonirritating and breathable cosmetic formulations to be obtained. Furthermore, a survey of potential consumers was conducted who by rating skin sensations were more positive about silicone emulsions than about silicone-free ones. Silicone-based emulsions were more pleasant both at the time of application and after full distribution. These results confirm and support conscious and thoughtful application of silicones which can improve the functionality and even competitiveness of cosmetic products.

CONCLUSIONS

Summing up, it can be stated with confidence that silicones rightfully occupy a leading position among ingredients of cosmetic products. This is associated, on the one hand, with the complex of unique physicochemical properties of silicones, such as bioinertness, hypoallergenicity, volatility of cyclic and low molecular weight linear siloxanes, and ability to reduce surface tension, thus making them effective additives in decorative and care cosmetic products. On the other hand, the chemistry of silicones renders it possible to synthesize a huge number of organosilicon compounds of both various chemical composition and completely unique structural forms. This provides a way to tailor more and more novel cosmetic products from silicones and to expand their application areas. The structural diversity of organosilicon derivatives allows this branch of chemistry to develop constantly, and new methods of synthesis and modification of silicones enable the targeted synthesis of compounds with a set of necessary specified properties. Among these compounds are hyperbranched polymers and dendrimers, nanogels, various derivatives of stereoregular macrocyclic silsesquioxanes, and star-shaped and comb-like polymers. Naturally, there will be a period of time between the emergence of novel structures and their use in cosmetic formulations which is required for thorough biomedical studies. Probably, a special program is required under the auspices of the Association of cosmetics manufacturers, universities, and institutes of the Russian Academy of Sciences to implement novel cosmetic products based on silicones. It should be noted that right now the situation is very favorable in the Russian Federation. This is not only due to import substitution but primarily due to the fact that Russian scientists are leaders in the development of novel silicone compounds. This makes it possible to search for their promising applications and create fundamentally new competitive formulations of cosmetic products.

Thus, the use of silicones in cosmetic products remains an effective way of imparting desired valuable properties to cosmetic formulations, meets economic requirements in terms of production, and, most importantly, is safe for consumers.

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CONFLICT OF INTEREST

The authors of this work declare that they have no conflicts of interest.

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