

Biodegradation of Silicones (Organosiloxanes)

Part 9. Miscellaneous Biopolymers and Biodegradation of Polymers

Prof. Dr. Habil. Jerzy Lukasiak , Dr. Agnieszka Dorosz , Dr. Magdalena Prokopowicz ,
Doc. Eng. Pawel Rosciszewski , Dr. Bogdan Falkiewicz

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Abstract

Introduction

Historical Outline

Chemical Structures

Occurrence and Functions

Properties, Degradation and Biodegradation

 Degradation and Biodegradation *In vitro*

 Environmental Fate and Degradation of Silicones

In vitro Biodegradation of Silicones

 Biodegradation *In vivo*

 Silicone Implants and Their Components

 Other Silicones

Silicone Industry

 Production of Silicones

 Main Commercial Silicone Producers

Patents

Outlook and Perspectives

1 Introduction

The overwhelming majority of synthetic polymers are characteristic of strong resistance to action of biological factors, and upon their application in practice, they become an extremely inconvenient burden to the environment. This results not so much from their specific “durability”, but rather from the fact that they originated relatively recently in comparison with the period of formation of evolutionary processes in nature. There are no natural, specific mechanisms of decomposition for synthetic polymers in the environment, unless they happen to fit into the degradation routes that exist in the natural world.

There are differences between chemical and biological decomposition (Lenz, 1993; Huang and Huang, 1994). Biodegradation is a term for all transformations to which a polymer (a chemical compound) is subjected under the influence of biotic factors, e.g., biotransformation by microorganisms or metabolisms in higher organisms. Biodegradation takes place mainly under the influence of enzymes. Active microorganisms in a biodegradation process are mainly bacteria, fungi, algae, and yeasts (Lenz, 1993), which are capable of secreting active agents to the natural environment, e.g., enzymes, organic acids. In the first stage of the interaction, microorganisms are subjected to adhesion on the polymer's surface, and subsequently, the entire process of reaction of bacteria cell metabolism products with substrate is started.

Research into the unification of the criteria of assessment of the biodegradation processes has been in progress; however, so far there are no international unequivocal methods or procedures of testing biological degradation of polymers. The applied methods must be test-material specific (Stevens and Annelin, 1997).

2 Historical Outline

The degradation and biodegradation of polydimethylsiloxane (PDMS) and other siloxanes in natural and anthropogenic environments have been poorly examined (Lukasiak and Falkiewicz, 1996, 1998a). Initially, they were treated as non-degradable in the environment, and for many years, they were considered to be inert with respect to living organisms. Then, at the turn of the 1960s and 1970s, the possibility of, respectively, their chemical (environmental) degradation and biodegradation was proven. Until recently, no more than 100 papers focusing on the biodegradation of various siloxanes and their products could be found. Nevertheless, this lack of studies did not affect the general assessment that polysiloxanes are a group of polymers that is hard to biodegrade.

3 Chemical Structures

In a broader sense, all organosilicon compounds are called silicones (Smith, 1991). However, for many years now the use of the name silicones has been limited to oligo- and polymeric organosiloxanes that contain alternating silicon and oxygen atoms in the chain (Si-O-Si), which are the subject matter of this report.

From the point of view of environmental fate and transport characteristics, three major classes of organosiloxane compounds may be singled out (Allen et al., 1997):

- volatile methylsiloxanes (VMS) (Hobson et al., 1997);

- PDMS and its derivatives, excluding polyethermethylsiloxanes (Fendinger et al., 1997a);

- polyethermethylsiloxanes (PEMS) (Powell and Carpenter, 1997).

There has been very little research concerning biodegradation of organosiloxanes; until recently, only some data on degradation, and its products in the case of a few structures containing siloxanes, have been published (Table 1). They are presented in this review.

Table 1. Some organosiloxanes studied in the context of their biodegradation, covered by the chapter

Chemical name	Formula	Abbreviation
Polydimethylsiloxane	$(\text{CH}_3)_3\text{SiO}[(\text{CH}_3)_2\text{SiO}]_n\text{Si}(\text{CH}_3)_3$	PDMS
Cross-linked polydimethylsiloxane	-	Cross-linked PDMS
Copolymer of dimethylsiloxane and vinylmethylsiloxane	$(-[(\text{CH}_3)_2\text{SiO}]_x[(\text{CH}_3)(\text{CH}_2=\text{CH})\text{SiO}]_y)_n$	-
(Dialkyl or alkylaryl)cyclotetrasiloxanes	$(\text{CH}_3\text{RSiO})_4$ R=CH ₃ or (CH ₂) ₃ CH ₃ , CH(C ₂ H ₅)CH ₃ , C ₆ H ₅ , (CH ₂) ₃ Cl	-
Linear poly(dialkyl or alkylaryl)siloxanes	$\text{XO}(\text{CH}_3\text{RSiO})_n\text{X}$	
n>15; X=H or SiCH ₃ , R=CH ₃ or (CH ₂) ₃ CH ₃ , CH(C ₂ H ₅)CH ₃ , C ₆ H ₅ , (CH ₂) ₃ Cl, (CH ₂) ₂ CF ₃ , C ₆ H ₂ Cl ₃	-	
Branched or cross-linked poly(dialkyl or alkylaryl)siloxanes	$\text{HO}(\text{R}_2\text{SiO})_a(\text{RSiO}_{1.5})_b$ a+b>10 and a : b=0.5–0.7 R=CH ₃ or (CH ₂) ₃ CH ₃ , CH(C ₂ H ₅)CH ₃	-
Polydimethylcyclsiloxane	$[(\text{CH}_3)_2\text{SiO}]_n$	cPDMS

4 Occurrence and Functions

Polyorganosiloxanes, both in their liquid form and in solid plastics, are broadly utilized in different household and industrial applications (Tomanek, 1991; ECETOC, 1994; Chandra et al., 1997). Because silicones are generally very stable polymers, they are used in various adverse environments, such as those with corrosive chemicals, high humidity, high temperature, electric fields, etc. (Tomanek, 1991). They are applied in such areas as protective encapsulating

materials for semiconductors in computers, bases for coolants in transformers, foam-control agents in laundry detergents, lubricants for bearings, coatings to protect facades and historical monuments, rubber-like sealants for windows and bathrooms, basis of gaskets used for engine parts, adhesives for fixtures and fittings, and protectors of electric circuits (Tomanek, 1991). They are also used in several kinds of domestic products, such as detergents, shampoos, conditioners, deodorants, creams, gels, textiles, water repellents, and other products that after use are directly evaporated into the air (only the low-molecular-weight ones) or released into the environment through sewage and wastewater treatment plants (Chandra et al., 1997).

Silicones can be found in all environments: soil, air, and water

(Pellenbarg, 1979a, 1979b, 1982, 1988; Watanabe et al., 1984a, 1988; Pellenbarg and Tevault, 1986; Pellenbarg and Carhart, 1990, 1991; Lukasiak et al., 1993a, 1993d, 1998; Pellenbarg et al., 1997; Fendinger et al., 1997b; Grümping et al., 1998; Grümping and Hirner, 1999; Wang et al., 2001).

Because of the high-scale production and wide range of applications of silicones, human exposure to silicones and their health effects are under strict control in the European Union, the United States, and Japan (Wischer and Stevens, 1997; Hatcher and Slater, 1997; Miyakawa, 1997).

Octamethylcyclotetrasiloxane (D₄, OMCTS) and PDMS, as well as the majority of other organosilicon products, do not meet the requirements to be regarded as substances hazardous to the environment, according to the criteria of the European Community (ECETOC, 1994; Stevens, 1998a). Non-toxicity of selected siloxanes toward many species of organisms, including man, has been verified in numerous tests (Hobbs et al., 1975; Siddiqui and Hobbs, 1982, 1983, 1984; Bruggemenn et al., 1984; Aubert et al., 1985; Guillemaut-Drai et al., 1988; Frye, 1988; Isquith et al., 1988a; Kolesar et al., 1989; Craig and Caunter, 1990; Siddiqui and York, 1993; ECETOC, 1994, and references cited therein; Kent et al., 1994; Siddiqui et al., 1994a, 1994b; Sousa et al., 1995; Hobson and Silberhorn, 1995; Tolle et al., 1995; Fackler et al., 1995; Spivack et al., 1997, and references cited therein; Stevens, 1998a and references cited therein; Burns-Naas et al., 1998a, 1998b; Utell et al., 1998; Looney et al., 1998; Klykken et al., 1999; Padros et al., 1999), but some of these substances were clastogenic *in vitro* (Isquith et al., 1988b). Nonetheless, it is worth noting that early tests of the behavior of PDMS with respect to higher organisms indicated a possibility of bioconcentration of PDMS by animals (Watanabe et al., 1984b; Opperhuizen et al., 1987). On the other hand, these results are disputable and are not proven in other studies (Hobbs et al., 1975; Bruggemenn et al., 1984; Guillemaut-Drai et al., 1988; Annelin and Frye, 1989; ECETOC, 1994).

5 Properties, Degradation and Biodegradation

The knowledge of the physicochemical properties of an individual siloxane is crucial for understanding and prediction of its possible fate in the environment or even under *in vivo* conditions. The properties vary significantly, and therefore the fates of various silicone materials may vary from almost complete inertness to easy degradability in the environment. In order to predict their environmental fate, all silicone materials manufactured in significant amounts were divided into the three classes mentioned above (or, more precisely, into the following nine classes: organochlorosilanes; organoalkoxysilanes; hexamethyldisilazanes; volatile methylsiloxanes; polydimethylsiloxanes; modified polydimethylsiloxanes, excluding polyethermethylsiloxanes; polyethermethylsiloxanes; silicone resins; and silicone elastomers [Allen et al., 1997]) based on their physicochemical properties rather than their chemical structures. Some of them are normally used under both *in vitro* and *in vivo* conditions, some only *in vitro*. Unfortunately, only a few were tested for their susceptibility to being biodegraded.

5.1 Degradation and Biodegradation *In vitro*

A majority of studies dealing with the processes of biodegradation of silicone materials were performed under *in vitro* conditions. Their results are strongly connected with the environmental fate, analysis, and environmental degradation of silicones; hence, the review should be commenced with these issues.

5.1.1 Environmental Fate and Degradation of Silicones

PDMSs are characterized by their very low water solubility (usually <1 part per million [ppm]). In the case of high-molecular-weight PDMSs, the water solubility is approximately 1 ppt (parts per trillion) or even lower (Mazzoni et al., 1997; Varaprath et al., 1996; Stevens, 1998a). Low solubility is also characteristic of low-molecular-weight siloxanes, e.g., D₄ (approximately 0.05 mg/L [Varaprath et al., 1996] or 0.07 mg/L [Hobson and Silberhorn, 1995]). Hence, they are simply hard to test with respect to their biodegradation, as even recommended tests applicable for sparingly soluble substances require a concentration of the examined substance in a water solution of approximately 10 to 100 mg/L, which is impossible to achieve for PDMS (Stevens and Annelin, 1997; Stevens, 1998b). Only upon the introduction of isotopic marking of PDMS by means of ¹⁴C (Varaprath, 1999; Stevens, 1998b) and upon the establishment of new methods of extraction (Lehmann, 1993; Varaprath et al., 1998; Varaprath and Cao, 2000) and analysis of organosiloxanes in biological matrices (Cavic-Vlasak et al., 1996; Carpenter and Gerhards, 1997; Varaprath and Lehmann, 1997; Lugowski et al., 1998; Lukasiak and Falkiewicz, 1998b; Grümping, et al., 1998; Grümping and Hirner, 1999; Kennan et al., 1999), including analyses of changes in the siloxanes' structures by means of the NMR method (Pfleiderer et al., 1993a, 1993b, 1993c, 1995, 1996, 1999; Pfleiderer and Garrido, 1995; Garrido, 1997; Garrido et al., 1993, 1994, 1996, 1998, 1999; Garrido and Young, 1999; Jamrógievicz et al., 1997), was the observation of the degradation process of siloxanes in the environment, with their *in vitro* and *in vivo* biodegradation, possible.

Because of their volatility and very high value of Henry's law constant, volatile low-molecular-weight siloxanes, e.g., D₄, evaporate to a large extent by penetrating mainly to the atmospheric air, where they are detectable and where they undergo degradation (Mueller et al., 1995; Hamelink et al., 1996; Mazzoni et al., 1997; Wang et al., 2001). Part of them may be sorbed by soil because of their high absorption coefficient (e.g., log K_{OC}=4.5 for D₄ [ECETOC, 1994]; for more data, see Mazzoni et al., 1997). All the other siloxane products eventually undergo decomposition as solid wastes or liquid wastes (Griessbach and Lehmann, 1999; Chandra et al., 1997).

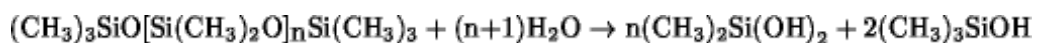
Insoluble in water, high-molecular-mass liquid siloxanes have a high adsorption coefficient (approximating log K_{OC}=3.69 for PDMS 10,000 cSt, containing siloxanols [Watanabe et al., 1985; ECETOC, 1994], or log K_{OC}=5.8 for PDMS 350 cSt [Lehmann et al., 1995]; for more data, see Mazzoni et al. 1997). Therefore, they strongly bind with other organic molecules and they become part of sewage sludge during wastewater treatment in sewage treatment plants (Watts et al., 1995; Fendinger et al., 1997b). Their concentrations there approximate 300 to 5000 mg/kg dry matter (Fendinger et al., 1997a). Research by Watts and coworkers ascertained that almost the entire amount of PDMS (> 99.9%) added to activated sewage is combined with microbial mass (Watts et al., 1995). Further fate of siloxanes depends on their behavior in sewage and on the manner of the treatment of that sewage. The three most frequently applied methods of sewage utilization (Stevens, 1998a, 1998b; Fendinger et al., 1997b) are incineration, which causes complete mineralization of siloxanes to CO₂ and SiO₂, landfill, and soil amendment.

As a consequence of the increasing significance (particularly in the countries of the European Community) of slurry's application in soil amendment, soil becomes a basic environmental compartment in which degradation takes place for the majority of liquid siloxanes wastes (Griessbach and Lehmann, 1999). In the study by Fendinger et al. (1997b), the concentration of PDMS in soils enriched with slurry equaled from 0.41 to 10.4 mg/kg of soil in different regions of the U.S.

Hydrolysis and re-arrangement of the silicon–oxygen bond in PDMS, catalyzed by contact with dry soil, were observed as early as in 1979 (Buch and Ingebrigtsen, 1979). The formation of cyclic (primarily D₄ and decamethylcyclopentasiloxane [D₅]) and linear low-molecular-weight siloxanes was observed mainly in pure kaolinite clays, and the formation of silanols in montmorillonite clays was observed under the conditions of oven-dried soils and very high loadings (1%) of PDMS. In further research, at lower concentrations of PDMS in soil, silanols were observed as the product of PDMS degradation (mainly very easily soluble dimethylsilanediol [DMSD]), with only a minority of other products (various siloxanes having a molecular weight of approximately 160, with the initial at some 7000) (Lehmann et al., 1994a, 1995; Fendinger et al., 1997b). At the same time, no products of siloxanes' demethylation were revealed (Mahone et al., 1983). (Nonetheless, the possibility of chemical methylation of mercury(II) salts by various polydimethylsiloxanes in aqueous solution has been

assumed; the best capacity for methyl mercury [highly toxic compound] production was shown in the case of hexamethyldisiloxane [Watanabe et al., 1986; Nagase et al., 1988]). Main catalysts of degradation (re-arrangements or hydrolysis) are clays' minerals, whereas humus and sand components are much less active (Buch and Ingebrigtsen, 1979; Xu et al., 1998; Xu, 1998). Catalytic properties of soil appear to be associated with acid catalysis, typical of clays (Stevens, 1998b). Out of the examined mineral components of clays, the most active ones with respect to catalysis are kaolinite, gibbsite, beidellite, and nontronite, whereas goethite and allophane are less active (Xu et al., 1998). This degradation seems to be quite effective; in the case of soil, to which subsequent portions of slurry are added, usually no PDMS accumulation is revealed (Siebert, 1988; Fendinger et al., 1997b). However, in the examination by McAvoy et al. (1996), PDMS was still found in soil after 20 years of its enrichment with slurry. Degradation to water-soluble silanols takes place in very different types of soil, coming from various geographic regions (Lehmann et al., 1994a, 1994b, 1995, 1996, 1998a, 2000; Singh et al., 2000); degradation is also apparent when dry EPA standard soil matrix is used (Carpenter et al., 1995). Detailed investigations have revealed that the rate of degradation is several times greater in the case of a dry soil versus a wet one (e.g., approximately 3% degradation in 6 months in wet soil compared with 50% degradation in several days in the same dry soil) (Lehmann et al., 1994a, 1994b, 1998a; Xu, 1998; Griessbach and Lehmann, 1999; Singh et al., 2000); consequently, the rate of PDMS degradation *in situ* is also affected by climatic conditions, clay type, exchangeable cation type and concentration, and siloxane concentration (Singh et al., 2000; Lehmann et al., 1995, 2000; Xu, 1998). In tests done by Lehmann et al. (1995), the half-life period of PDMS persistence in the dry soil ranged from 4 to 28 days, depending on the initial content of siloxane in the mixture. Notwithstanding, degradation also occurs in wet soil and even in freshwater sediments, where after approximately 1 year of incubation, 5% to 10% of [¹⁴C]PDMS undergoes hydrolysis to DMSD, while approximately 0.25% is oxidized to CO₂ (Carpenter, 1996).

The reaction of PDMS hydrolysis may be summarized as follows (Fendinger et al., 1997a; Carpenter et al., 1995; Lehmann et al., 1994a, 1994b; West, 1997):



Hydrolysis of PDMS is expected to be the main source of low-molecular-weight methylsilanols in the environment (Spivack et al., 1997).

5.1.2 *In vitro* Biodegradation of Silicones

PDMS is much less susceptible to microbiological degradation than its soluble derivatives. Modified siloxanes could even have some antimicrobial properties (Lukasiak et al., 1999a). Because of its high-molecular nature (> 700 D for viscosities of 10 cSt or above), PDMS is not an object that would pass through cellular membranes. Additionally, while forming a hydrophobic

surface of methyl groups along the chain, it effectively protects the oxygen–silicon bond, one of the bonds most resistant to enzymatic hydrolysis, against hydrolytic attack (Stevens, 1998b). The classical intracellular biodegradation of PDMS is also rather unlikely (Stevens, 1998b). However, some microorganisms and enzymes gather on some part of the hydrophilic surface.

Biodegradation of organosilicone polymers probably consists of the shortening of the polymer chain and formation of silanol groups at places of the chain scission (possibly also at places of detachment of alkyl groups). Therefore, upon a partial hydrolysis of PDMS to smaller molecules, biodegradation may proceed, which has been demonstrated many times.

Original research into biodegradation of silicone oils (based mainly on PDMS and polyphenylmethylsiloxane) by soil micromycetes and aquatic diatoms was unsuccessful (Glazer 1954; Sharp and Eggins, 1978; Buch et al., 1984). The tests reported by Frye (1983, 1988) or by Lehmann et al. (1996) did not show PDMS degradation in sewage. In addition, some experiments indicated that [^{14}C]PDMS is not degraded to the marked $^{14}\text{CO}_2$ by anaerobic sewage bacteria (Hobbs et al., 1975). In the case of 15% emulsion [^{14}C]PDMS of viscosity 300 cSt exposed to the action of sludge during the period of observation (70 days), no presence of $^{14}\text{CO}_2$ was found in the products of decomposition (Calandra et al., 1976). In the same conditions, a comparative substance ([^{14}C]octadecane) underwent decomposition to $^{14}\text{CO}_2$ and other volatile mineral products. Liquid silicone polymers (polydimethylsiloxanes) also do not appear to be degraded during the composting process (Smith et al., 1998). Some other similar studies on biodegradation of silicones were also unsuccessful (ECETOC, 1994, and references cited therein).

Microorganisms capable of biodegrading low- and/or high-molecular-weight siloxanes are certain bacteria (Heinen, 1978; Wasserbauer and Zadak, 1990; Lukasiak et al., 1993b, 1993c, 1995a, 1995b, 1997; Rosciszewski et al., 1994, 1995a, 1995b, 1997, 1998; Dorosz, 1997; Dorosz et al., 1997; Lukasiak and Dorosz, 1997) and fungi (Baud-Grasset and Palla, 1997). Among bacteria, this capability is demonstrated by species from the genera *Pseudomonas*, *Proteus*, *Escherichia*, *Bacillus*, and *Klebsiella* and among fungi by species belonging to the Corticiaceae family. Fungi from the species *Phanerochaete chrysosporium* belonging to the family are known to be capable of catalyzing numerous reactions of oxidation by means of extra-cellular secretion of peroxidases, whereas the reactions occur with the use of a hydroxyl radical derived from hydrogen peroxide (Stevens, 1998b).

Among the first organosiloxanes examined with respect to microbiological biodegradation, hydrolysis of ethoxysilanes, which was catalyzed by soil bacteria and growth of these bacteria with the use of ethanol produced in the process, was observed in 1978 (Heinen, 1978).

Research conducted by Wasserbauer and Zadak (1990) demonstrated that bacteria from the genus *Pseudomonas* (*P. putida* and *P. fluorescens*) are capable of growing on oligomeric and polymeric dimethylsiloxanes, with the growth being the function of the increasing viscosity of

siloxanes. PDMS microbiological biodegradation took place in aerobic conditions. Nevertheless, these experiments must be interpreted very carefully, as the bacteria growth within 12 days, as indicated therein, is at variance with observations that we may make on a daily basis, where we do not notice fast degradation of polydimethylsiloxanes, which are applied in numerous areas (see also Stevens, 1998b).

Our initial research on siloxanes' biodegradation was done on a group of pathogenic bacterial strains isolated directly from clinical materials (Lukasiak et al., 1993b, 1993c, 1995a, 1997; Rosciszewski et al., 1994, 1995a, 1997; Dorosz, 1997; Dorosz et al., 1997; Lukasiak and Dorosz, 1997). These include *Staphylococcus aureus*, *Staphylococcus epidermidis*, *Staphylococcus saprophyticus*, *Streptococcus fecalis*, *Streptococcus viridans*, other group B or A streptococci, *Bacillus subtilis*, *Escherichia coli*, *Klebsiella pneumoniae*, *Shigella sonnei*, *Salmonella enteritidis*, *Yersinia* sp., *Proteus* sp., and *Pseudomonas aeruginosa*. Incubation in a liquid culture with the addition of glucose and approximately 3% concentration of one of the siloxanes tested (see Tables 2 and 4) was conducted by a maximum of 30 days at 37°C. After extraction of organosilicon products, the biodegradation degree test was done by using the ^{13}C -NMR method. The tests revealed that many of the tested bacteria demonstrated the capability of degrading the examined siloxanes in the applied conditions (see Tables 2-5 and Figure 1).

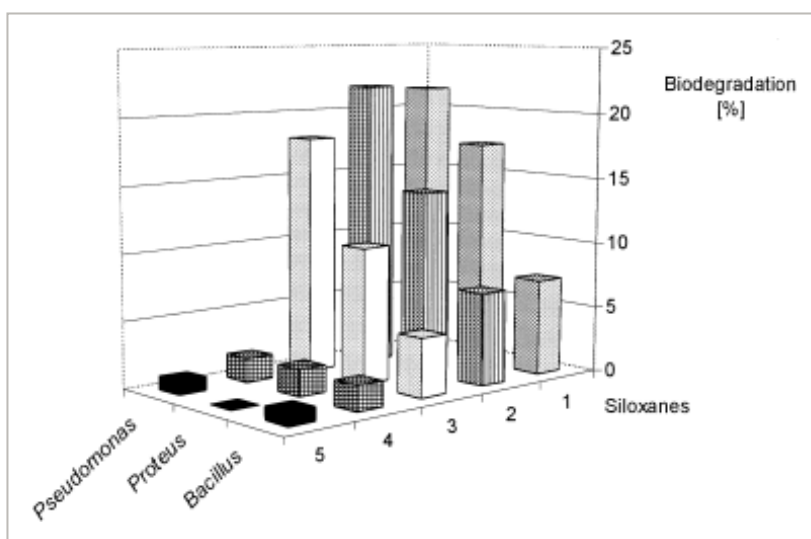


Figure 1

[Open in figure viewer](#) | [PowerPoint](#)

Biodegradation of cyclosiloxanes after 21 days of incubation with bacteria. 1: octamethyltetracyclosiloxane; 2: cyclotetra(methyl-*n*-butyl)siloxane; 3: cyclotetra(methyl isobutyl)siloxane; 4: cyclotetra(methylphenyl)siloxane; 5: cyclotetra[methyl-(3-chloropropyl)]siloxane.

Table 2. Three types of organosiloxanes tested by our group ^a

Type	I	II	III
Name	Poly(dialkyl or alkylaryl)cyclosiloxanes	Linear poly(dialkyl or alkylaryl) siloxanes	Branched or cross-link poly(dialkyl or alkylaryl siloxanes
Schematic formula	$(CH_3RSiO)_n$ n=3, 4, 8	$XO(CH_3RSiO)_nX$, n>15	$HO(R_2SiO)_a(RSiO_{1.5})_b$ a : b=0.5–0.7
R (substituents) ^b	R=CH ₃ or (CH ₂) ₃ CH ₃ , CH(C ₂ H ₅)CH ₃ , C ₆ H ₅ , (CH ₂) ₃ Cl	X=H or Si(CH ₃) ₃ , R=CH ₃ or (CH ₂) ₃ CH ₃ , CH(C ₂ H ₅)CH ₃ , C ₆ H ₅ , (CH ₂) ₃ Cl, (CH ₂) ₂ CF ₃ , C ₆ H ₂ Cl ₃	R=CH ₃ or (CH ₂) ₃ CH ₃ , CH(C ₂ H ₅)CH ₃
Examples of compounds tested ^a	$[(CH_3)_2SiO]_4$ [CH ₃ (ClPr)SiO] ₄ (M=610)(CH ₃ PhSiO) ₄ (M=520)	H(BuSiO _{1.5}) _a [(CH ₃) ₂ SiO] _b OH, (M=950)HO[(CH ₃) ₂ SiO] _a [CH ₃ (ClPr)SiO] _b OH, (M=2600)	HO(BuSiO _{1.5}) _a (CH ₃ BuSiO) _b OH, (M=2550)

^a compounds either synthesized or purchased from commercial sources.

^b Bu=C₄H₉, Pr=C₃H₇, Ph=C₆H₅.

Table 3. Comparison of biodegradability of three siloxanes tested

Bacteria species symbol	Bacteria species tested	Percentage of the siloxane degraded ^a		
		Me ₃ SiO(Me ₂ SiO) _n SiMe ₃	HO(MeBuSiO) _n H	HO(BuSiO _{1.5}) _a (MeBuSiO) _b OH
	None (control)	0	0	0
a	<i>Bacillus subtilis</i>	50	90	90
b	<i>Proteus species</i>	0	0	80

Bacteria species symbol	Bacteria species tested	Percentage of the siloxane degraded ^a		
		$\text{Me}_3\text{SiO}(\text{Me}_2\text{SiO})_n\text{SiMe}_3$	$\text{HO}(\text{MeBuSiO})_n\text{H}$	$\text{HO}(\text{BuSiO}_{1.5})_a(\text{MeBuSiO})_b\text{H}$
c	<i>Pseudomonas aeruginosa</i>	20	0	100
d	<i>Staphylococcus aureus</i>	0	70	100

^a Me=CH₃; Bu=C₄H₉; Pr=C₃H₇; Ph=C₆H₅.

^b unidentified species.

Table 4. Bacterial strains tested and active in biodegradation process of certain examined siloxanes

Siloxane ^a	Type of polysiloxane ^b	Species of bacteria tested ^c	Evidence of microbial degradation for species ^c
$(\text{Me}_2\text{SiO})_3$	I	c	c
$(\text{Me}_2\text{SiO})_4$	I	a,b,c	a,b,c
$(\text{MeBuSiO})_4$	I	a,b,c,r	a,b,c,r
$[\text{Me}(\text{ClPr})\text{SiO}]_4$	I	a,b,c,r	
$(\text{MePhSiO})_4$	I	a,b,c	
$\text{Me}_3\text{SiO}(\text{Me}_2\text{SiO})_n\text{SiMe}_3$	II	a,b,c,d,e,f,g,h,i,j,l,m,n,o,p	a,c,e,g,l,m
$\text{HO}(\text{MeBuSiO})_n\text{H}$	II	a,b,c,d,e,f,g,h,i,j,l,m,n,o,p	a,d,e,g,i,j,l,m,o
$\text{HO}(\text{BuSiO}_{1.5})_a(\text{MeBuSiO})_b\text{H}$	III	a,b,c,d,e,f,g,h,i,j,l,m,n,o,p	a,b,c,d,e,f,g,h,i,j,l,m,n,o,p

Siloxane ^a	Type of polysiloxane ^b	Species of bacteria tested ^c	Evidence of microbial degradation for species ^c
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^a Me=CH₃, Bu=C₄H₉, Pr=C₃H₇, Ph=C₆H₅.

^b see Table 1.

^c for bacteria species symbols, see Table 3.

Table 5. Biodegradation degree of chosen siloxanes after 30-day incubation with *Pseudomonas aeruginosa*

No.	Siloxane	Degree of biodegradation (%)
1	Octamethylcyclotetrasiloxane	21
2	Cyclotetra(methyl- <i>n</i> -butyl)siloxane	22
3	Cyclotetra[methyl(isobutyl)]siloxane	18
4	Cyclotetra(methylphenyl)siloxane	2
5	Cyclotetra[methyl-(3-chloropropyl)]siloxane	1
6	Polydimethylsiloxane	2
7	Poly(methylbutyl)siloxane	5
8	Poly[methyl-(3-chloropropyl)]siloxane	3
9	Poly(methyloctyl)siloxane	68
10	Polyethoxysiloxane	91

The second stage of research focused on developing optimal conditions of biodegradation by using carefully selected microorganisms. For the purpose of testing, standardized strains

included in the National Committee for Testing Clinical Laboratory Standard listing were chosen, namely *Escherichia coli* ATCC 25922, *Pseudomonas aeruginosa* ATCC 27853, *Klebsiella pneumoniae* K 28, *Proteus mirabilis* 14 a, and *Bacillus subtilis* (strain for didactic purpose). The products of siloxane's degradation were analyzed by using the methods of ^1H -NMR, IR, ASA, and gel chromatography. The obtained results showed that all these species were able to biodegrade various siloxanes in a manner similar to that of the clinical isolates (Lukasiak et al., 1995b; Lukasiak and Dorosz, 1997; Rosciszewski et al., 1995b, 1998).

It was found that nearly all polysiloxanes tested are biodegradable and that the degree of biodegradation depends both on their structure and on the bacteria strain used (Rosciszewski et al., 1998).

According to the results, the degradation process under the influence of contact with microorganisms is slow and requires at least a 14-day incubation of samples in liquid culture, while the polymer-microorganisms colony contact surface (polymer concentration, bacteria concentration) has an impact on the rate of biodegradation process. *Pseudomonas aeruginosa*, *Proteus mirabilis*, *Klebsiella pneumoniae*, *Escherichia coli*, and *Bacillus subtilis* appear to be the most active strains in the process of biodegradation.

The susceptibility of silicones to biodegradation depends on the structure of organosiloxane:

siloxanes with long-chain aliphatic substituents are easier to degrade than compounds with methyl groups. The highest resistance to biodegradation is typical of polydimethylsilicone oil, while susceptibility to microbiological biodegradation of polymers containing butyl groups increases with the growth of the ratio of butyl groups to methyl groups in the polymer structure;

cross-linking in polymer associated with branching structure probably enhances the transformations triggered by bacterial enzymes;

presence of alkoxyl groups increases the susceptibility to biodegradation;

cyclic and low-molecular-weight siloxanes are easier to utilize by microorganisms;

presence of silanol groups (Si-OH) in the structure of a compound enhances the biodegradation process;

silicones whose structure incorporates halogen atoms belong to the ones that are hard to degrade;

among the tested compounds, polydiethoxysiloxane and polymethyloctylsiloxane are the easiest to biodegrade.

In compliance with the conducted research (Batich et al., 1996; Lehmann et al., 1995), one may state that the process of biodegradation of silicones produces polymer molecules of lower mass, low-molecular-weight silanols of the general formula $\text{HO}(\text{R}_2\text{SiO})_n\text{OH}$, low-molecular-weight cyclic siloxanes, and inorganic water-soluble silicon compounds.

A diagram of the suggested biodegradation pathway of organosilicon compounds is displayed in Figure 2.

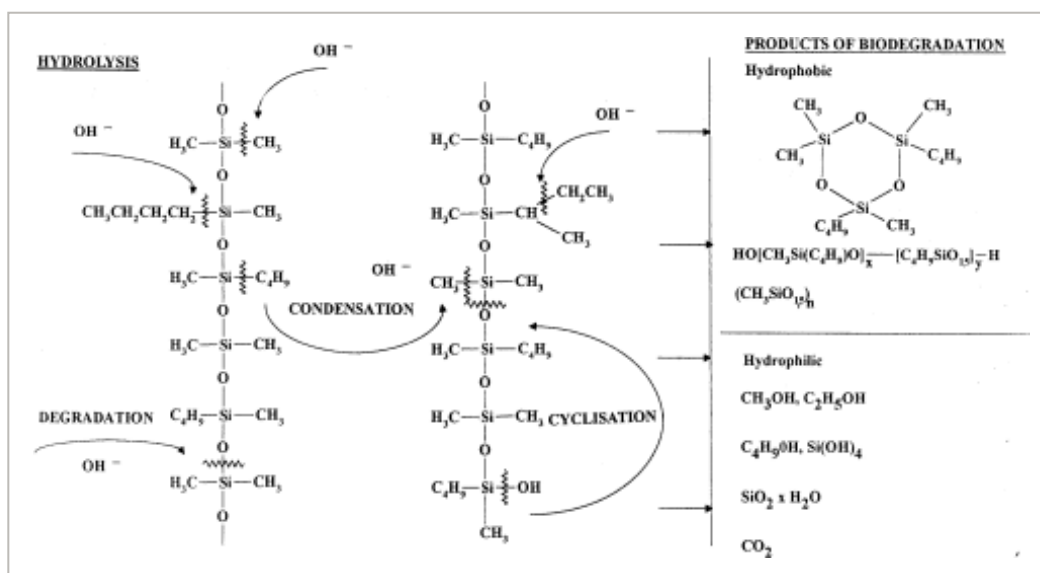


Figure 2

[Open in figure viewer](#) | [PowerPoint](#)

Probable biodegradation pathway of polysiloxanes.

In patent literature, one can find a method of biodegradation of polyorganosiloxanes, particularly polydimethylsiloxanes, with the use of one or more microscopic fungi (selected from the Corticiacea family, preferably from the genus *Phanaerochaete* or *Aspergillus*, especially *Aspergillus sydowii* BJS94, *Phanaerochaete chrysosporium*, or *Phanaerochaete sordida*), preferably in the presence of a co-substrate (Baud-Grasset and Palla, 1997). The biodegradation occurs at various pH levels, and the requested co-substrate is a source of carbon and energy, preferably a particular sugar (glucose or sucrose) (Baud-Grasset and Palla, 1997).

5.1.2.1 Silicone Rubbers and Other Solid Silicone Materials

Another research problem is the process of biodegradation of rubbers and other solid silicone plastics in laboratory conditions. Adhesion of microbes to silicone rubber is one of the first steps in the biodeterioration of silicone rubber prostheses. Silicone rubber prostheses are generally susceptible to adhesion of *Streptococcus* sp., *Staphylococcus* sp., *Candida* sp. (especially *Candida*

tropicalis), *Acinetobacter* sp., *Micrococcus* sp., *Pseudomonas* sp., *Bacillus* sp., *Chryseomonas* sp., and other kinds of bacteria, yeasts, and higher fungi, and, as a consequence, undergo biodegradation under their impact (Dolezel et al., 1989; Neu et al., 1993; Busscher et al., 1994 and references cited therein, 1997, 1998a,b; Slavicek et al., 1998, 2000; van der Mei et al., 1999, 2000). *Candida tropicalis* could be made to grow into silicone rubber under laboratory conditions (Busscher et al., 1994). Silicone prostheses may be deeply degraded by these microorganisms *in vivo* (Neu et al., 1993; Slavicek et al., 2000), and *Micrococcus* seems to be a typical vector that assimilates silicone (Slavicek et al., 2000). The formation of the biofilm on the silicone rubber implant surfaces may be inhibited by perfusion of the device with dairy products or probiotic bacteria (Busscher et al., 1998a; van der Mei et al., 1999, 2000). In addition, in dental materials it has been reported that silicone rubber denture liners are apt to colonization and degradation by yeasts (Gettleman et al., 1983).

5.1.2.2 Volatile Methylsiloxanes

The biodegradation of VMS is less important for its removal from the environment than is that of PDMS. In the atmosphere, volatile siloxanes undergo indirect photodegradation, mainly as a consequence of attacks of OH or NO₃ radicals and Cl atoms (Atkinson, 1991; Atkinson et al., 1995; Sommerlade et al., 1993; Markgraf and Wells, 1997; Stevens, 1998a; Tuazon et al., 2000). The half-life period of volatile methyl-silicon compounds in the atmosphere is estimated at approximately 5 to 30 days (Atkinson, 1991; Atkinson et al., 1995; Sommerlade et al., 1993; Markgraf and Wells, 1997; Stevens, 1998a; Tuazon et al., 2000). The products of photolysis are mainly carbon dioxide and various silanols, which are subsequently washed out with rain. In water solutions, particularly in the presence of nitrate ions, photodegradation of oligomeric dimethylsiloxanes is possible (Anderson et al., 1987). The final product of degradation is silica, while the half-life of oligomer mix is 4 to 9 days. In the same conditions, a much slower degradation of high-molecular-weight PDMS is also possible (Anderson et al., 1987). Some part of low-molecular-weight cyclic siloxanes also undergoes adsorption to the components of sewage (Mueller et al., 1995; Stevens, 1998a).

5.1.2.3 Octamethylcyclotetrasiloxane

Octamethylcyclotetrasiloxane (D₄) is a compound used in large quantities in the manufacture of cosmetics and personal hygiene agents; hence, a number of studies into its toxicity, environmental fate, and degradation were done (Hamelink et al., 1996; Kent et al., 1994, 1996; Mueller et al., 1995; Hobson, 1995; Hobson and Silberhorn, 1995; Parker et al., 1999; Sousa et al., 1995; Stevens, 1998a and references cited therein; Grümping et al., 1999). It seems that D₄ may be a model compound of various low-molecular-weight polydimethylsiloxanes. D₄ is characteristic of a rather high coefficient of adsorption to soil components, which may be used for treating a stream of gases from admixtures of the said compound and indirect products of its degradation (Lehmann and Xu, 1999). Under normal conditions, D₄ and its higher molecule counterparts, D₅ and dodecamethylcyclohexasiloxane (D₆), undergo degradation to DMSD in

soil, competing with volatilization (Xu, 1999; Xu and Chandra, 1999). The stage limiting the rate of degradation reaction is the ring-opening reaction of cyclosiloxane (Xu, 1999). In conditions of high humidity, volatilization prevails, while in conditions of low humidity, degradation prevails. As a result, siloxanes of this type are unlikely to persist in any soils (Xu and Chandra, 1999). Regardless of the place of hydrolysis of a siloxanes chain, DMSD is produced instead of longer short-chain siloxanols, since in water solutions of similar concentrations in a mixture with longer-chain siloxanols, DMSD is the main element of the equilibrium (Spivack and Dorn, 1994). In almost all the leachates from locations where silicone materials were deposited, the presence of DMSD was found in the low mg/L range, and in several of them, trimethylsilanol (TMS) was found in the low $\mu\text{g/L}$ range (Grümping and Hirner, 1999). However, the same group of researchers was not sufficiently successful in detecting products of the degradation of siloxanes (TMS) in sewage gases of municipal plants (Grümping et al., 1998). This was in all probability caused by the lack of volatile methylsiloxanes, previously stripped of during wastewater treatment, and by the relatively short time (approximately 20 days) of the retention of sludge in treatment plants, which was too short for a detectable degradation of siloxanes (Hobson et al., 1997; Grümping et al., 1998, 1999).

D₄ undergoes biodegradation in composted sewage sludge under anaerobic conditions (Grümping et al., 1999). This has been proven by *in vitro* examination of the presence of the main product of its degradation, DMSD, in these conditions, in a comparison between sterilized and non-sterilized samples incubated at 37°C (Grümping et al., 1999). The maximum concentration of DMSD was reached after approximately 100 days upon the commencement of the test; it corresponded to approximately 3% of the quantity of the added D₄, and subsequently dropped (Grümping et al., 1999), probably as a consequence of biodegradation or sorption of DMSD (Sabourin et al., 1996; Lehmann and Miller, 1996). In similar samples of sewage sludge, to which PDMS without D₄ was added, no presence of DMSD was detected after 3 months of incubation, which seems to once again verify the non-biodegradability of PDMS in these conditions (Grümping et al., 1999).

The oxidase-positive, gram-negative bacteria were able to grow with D₄ as the only carbon source, and this siloxane was eliminated from solution culture within several hours; biodegradation was oxygen-dependent, but its products were not identified (H. G. Rast, 1993, Bayer AG, Leverkusen, Germany, personal communication; according to ECETOC, 1994).

5.1.2.4 Silanols

DMSD (mainly), linear silanols of the general formula $\text{HO}(\text{CH}_3)_2\text{SiO}[\text{Si}(\text{CH}_3)_2\text{O}]_n\text{H}$ ($n < 8$) (in much smaller quantities), and TMS, being the products of PDMS degradation (Lehmann et al., 1994a, 1994b, 1995, 2000; Carpenter et al., 1995; Grümping et al., 1998; Grümping and Hirner, 1999), are subject to volatilization and escape to the atmosphere (the majority) or are subject to biodegradation (Griessbach and Lehmann, 1999; Lehmann et

al., 1994a, 1994b, 1998b; Lehmann and Miller, 1996). In the case of DMSD, volatilization is the main mechanism of the outlet of the compound from soil, which is proven by the release of [^{14}C]DMSD, whereas biodegradation (connected with the release of $^{14}\text{CO}_2$) is of smaller significance (Griessbach and Lehmann, 1999). In both cases, DMSD undergoes oxidation and complete mineralization (in the atmosphere or directly in soil), and has as final products CO_2 , SiO_2 , and H_2O . In laboratory conditions, where reaction proceeds in laboratory flasks, the bonding of DMSD by soil's humus is also observable (Lehmann and Miller, 1996; Lehmann et al., 1998b), whereas in the processes taking place in open conditions (i.e., natural ones) only a very small quantity of the introduced DMSD is bound by soil (Lehmann et al., 1996).

In the atmosphere or in a water environment, DMSD and other methylsilanols probably undergo radical reactions, mainly with hydroxyl radicals, produced as a consequence of sunlight operation in the presence of nitrate or nitrite ions (Sommerlade et al., 1993; Buch et al., 1984; Stevens, 1998b). Photolytic oxidative demethylation of DMSD results in the production of $\text{Si}(\text{OH})_4$ (silic acid) and CO_2 . Similarly, in the presence of nitrate ions, the mixtures of short-chain dimethylsiloxanols with dimethylcyclsiloxanes decompose, whereas high-molecular-weight PDMS exposed to the light does not react (Anderson et al., 1987).

The products of microorganisms affecting DMSD are methylsilanetriol ($\text{CH}_3\text{Si}(\text{OH})_3$, MST), which undergoes further transformation to SiO_2 and H_2O (Sabourin et al., 1996, 1999), $\text{Si}(\text{OH})_4$ and inorganic silicates, and CO_2 (Lehmann et al., 1994a, 1994b, 1998b; Lehmann and Miller, 1996; Stevens, 1998b; Sabourin et al., 1999; Griessbach and Lehmann, 1999). The rate of biodegradation of [^{14}C]DMSD, measured by releasing $^{14}\text{CO}_2$, approximates 0.4% to 1.6% weekly (depending on the type of soil) (Lehmann et al., 1998b), whereas the rate of volatilization approximates 1% to 7% weekly (Lehmann and Miller, 1996). Comparison of the values in particular conditions shows that degradation prevails in damp, heavy soils, whereas volatilization prevails in dry, sandy soils (Lehmann and Miller, 1996; Stevens, 1998b; Lehmann et al., 1998b). In the experiments done by Lehmann et al. (1998b), after 30 weeks of incubation of [^{14}C]DMSD (1 mg/kg) at 25°C in different soil types, the majority of the remaining activity of ^{14}C could be detected in acid- and base-extractable fractions, and not in water-extractable ones, such as DMSD (98% activeness ^{14}C at the beginning of the experiment). Finally, upon incubation of DMSD in soil with 2% plant litter added, one can observe an intense phenomenon of a transfer of the signal of the activated ^{14}C to the plant litter layer (Lehmann et al., 1998b).

The idea that the decomposition of [^{14}C]DMSD takes place in soil, and is catalyzed by microorganisms, is supported by the growth of the biodegradation rate accompanied by the growth of microbial biomass in most soil types, the hindering of the emission of $^{14}\text{CO}_2$ upon the fall in temperature to 4°C , and the decrease in the emission of $^{14}\text{CO}_2$ after the application of tetrahydrofuran as a bacteriostatic compound (Lehmann et al., 1994b, 1998b). On the other hand, Sabourin et al. (1996) isolated from soil and identified a bacterium (*Arthrobacter* sp.) and a fungus (*Fusarium oxysporum* Schlechtendahl) capable of co-metabolizing DMSD in soil and in

liquid culture (Sabourin et al., 1996, 1999). It is worth noting that although DMDS of liquid culture or soil and MST of liquid culture are metabolized by these microorganisms quite effectively (Sabourin et al., 1999), the degree of biodegradation of [^{14}C]MST to $^{14}\text{CO}_2$ in soil does not exceed 0.4% at the most, which is the level of impurities of ^{14}C in [^{14}C]MST. Therefore, one may not verify whether the biodegradation of MST in soil takes place at all (Sabourin et al., 1999). This is most probably a consequence of a much stronger bonding of MST to soil particles (Sabourin et al., 1999). It is supposed that the mechanism of biodegradation of methylsilanols by these microorganisms contains the stage of hydroxylation of the methyl group followed by oxidation of the hydroxymethyl derivative to a formyl or a carboxylic acid group (Sabourin et al., 1996). These acylsilanes are extremely sensitive to bases and under basic conditions are transformed to the corresponding silanol and aldehyde (Sabourin et al., 1996). Formyl silanes are particularly unstable and decompose at temperatures above 25°C (Ricci and Degl'Innocenti, 1989). The proposed mechanism involves addition of hydroxide to the carbonyl group and then a 1,2-migration of the Si from carbon to oxygen, forming a silyl-hemiacetal that spontaneously undergoes decomposition and gives the silanol and aldehyde (Sabourin et al., 1996).

From similar compounds, a possible degradation of trimethoxyphenylsilane by a *Pseudomonas* sp. was reported in 1967 (Fessenden and Fessenden, 1967).

5.2 Biodegradation *In vivo*

Siloxanes *in vivo* in higher organisms undergo biodegradation, although research into this is very scarce. Nonetheless, this is an issue of vital importance, considering the broad application of siloxanes in the production of silicone prostheses, the biodegradation and partial absorption of siloxanes taken in oral doses (e.g., anti-flatulent preparations) (Pilbrant and Strindberg, 1975; Calandra et al., 1976; Lukasiak and Falkiewicz, 1998b; Lukasiak et al., 1999b, 2001a), and in view of the fact that silicones are an intentional additive to food (Lukasiak and Falkiewicz, 2000). The first test verifying the degradation of the Si-O-Si bonds *in vivo* comes from the examination done by Fessenden and Ahlfors (1967). So far, little research has been done on biotransformation of siloxanes in animals (Pfleiderer et al., 1993c, 1995, 1999; Garrido et al., 1993; Dain and Nicoletti, 1995; Garrido, 1997) or in humans (Garrido et al., 1994; Dain and Nicoletti, 1995; Pfleiderer and Garrido, 1995; Pfleiderer et al., 1996; Garrido, 1997).

5.2.1 Silicone Implants and Their Components

In medicine, different types of silicone implants are widely applied. In many cases, macroscopic examination does not reveal any biodegradation of various types of silicone implants or their bacterial or fungal infestation (Kominek et al., 1998). Nevertheless, some types of silicone prostheses are frequently attacked and degraded by microorganisms, e.g., silicone intraocular lenses are more susceptible to adherence and formation of biofilm by clinical strains of *Pseudomonas aeruginosa* than are lenses made of other polymers (Gabriel et al., 1998).

Until recently, one of the most frequently applied silicone implants was breast prosthesis. Silicone-filled breast implants are most often made from a layer of silicone rubber, filled with siloxane gel, which is a mixture of liquid PDMS and copolymer of dimethylsiloxane and vinylmethylsiloxane cross-linked with PDMS (Table 1). It has been known since the 1970s that this type of prosthetic appliance demonstrates permanent gel bleeding (approximately 1 mg/day) (Batich et al., 1996). The elements of the gel bleed are mainly low-molecular-weight linear and cyclic siloxanes, like D₄–D₆ and higher-molecular-weight species, and small amounts of vinyl siloxane and other gel components (Batich et al., 1996).

In the first research on biodegradation of polysiloxanes in rats, water emulsion of polysiloxanes was injected into the animals' thighs twice at weekly intervals, and then after 3 days the extracts obtained from lymph nodes were subjected to ²⁹Si NMR examination (Garrido et al., 1993). In the examined samples, several resonance signals were observed within the range from –40 to –150 ppm, which corresponded to various functional groups containing Si, perhaps including polysiloxane, hydrolyzed silicones, and high-coordinated silicon complexes (Garrido et al., 1993). Similar results were obtained when water emulsions of linear and cyclic polysiloxanes (mainly D₄ and D₅) and extracts from silicone-filled breast implants were, only once or six times at weekly intervals, injected into the thigh and/or lymph nodes in the necks of rats (Pfleiderer et al., 1999). In the period of up to 72 days, the presence of siloxanes and products of their metabolism was examined in animals' lymph nodes with the use of the ²⁹Si NMR technique. It has been proven that despite the partially preserved signal of the injected polysiloxanes, with the passage of time the appearance and intensification of varied unidentified signals of partially hydrolyzed polysiloxanes and products of the reaction of siloxanes with other molecules, as well as of silica, were observed (Pfleiderer et al., 1999). Although all the examined siloxanes underwent biodegradation, its rate in the case of high-molecular-weight polymers was lower than that of oligomers (Pfleiderer et al., 1999).

In another study, silicone gel-filled implants were inserted into the lower back of rats, and after 9 or 12 months of implantation, liver, spleen, and the capsule formed around the implant samples were analyzed with ²⁹Si NMR. Resonance signals corresponding to both silicone and products of its metabolism were observed (Figure 3) (Garrido et al., 1993; Pfleiderer et al., 1993c, 1995). The number of Si-containing species found increased with an increase of implantation time; products of silicone metabolism were not observed in implants before implantation (Garrido et al., 1993). Similar results regarding the behavior of silicone implants and biodegradation of the siloxanes released from them were obtained in several tests done by Garrido and Pfleiderer et al. (Garrido et al., 1993, 1994, 1996, 1998, 1999; Pfleiderer et al., 1993c, 1995, 1996, 1999; Pfleiderer and Garrido, 1995; Garrido, 1997).

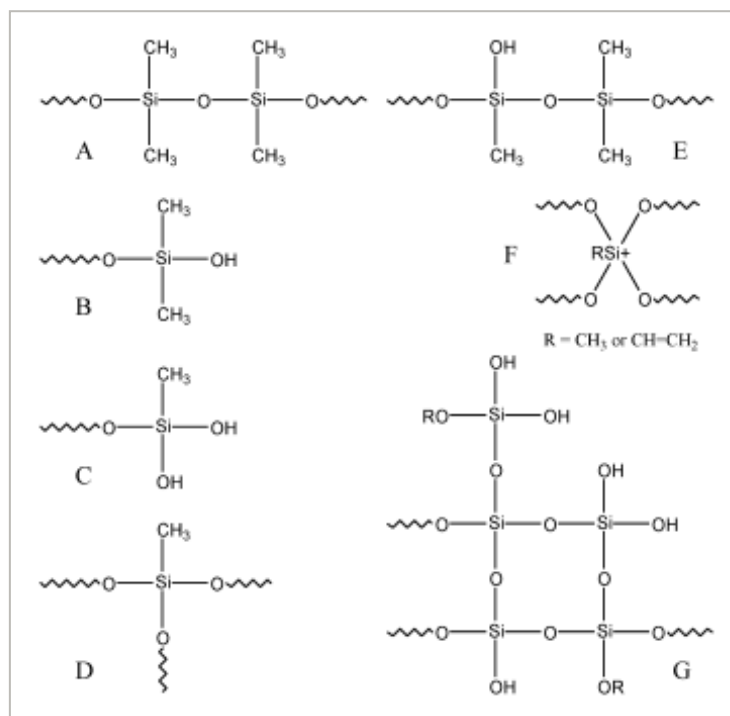


Figure 3

[Open in figure viewer](#) | [PowerPoint](#)

Organosiloxane groups originating from PDMS biodegradation observed by Garrido et al. A=PDMS; B–E=partially hydrolyzed PDMS; F=high coordinated silicon complex; G=silica.

Contradictory results pertain to chemical modifications of silicone oils used for retinal tamponade as a vitreous substitute *in vivo*. These oils often undergo emulsification after prolonged clinical use. In some examinations, after the removal of silicone oils from the human eye, no chemical modifications were revealed (Lakits et al., 1999); however, other studies prove that these oils are subject to chemical reactions in conditions *in vivo* and in modeled conditions *in vivo* (Lukasiak et al., 2001b).

[¹⁴C]Octamethylcyclotetrasiloxane ([¹⁴C]D₄) given to rats intravenously undergoes biodegradation, and its products were analyzed in animals' urine by Varaprath et al. (1999) (see Table 6). At least two main metabolites (DMSD and MST) were identified, together with at least five other metabolites (tetramethyldisiloxane-1,3-diol, hexamethyltrisiloxane-1,5-diol, trimethyldisiloxane-1,3,3-triol, dimethyldisiloxane-1,1,3,3-tetrol, and dimethyldisiloxane-1,3,3,3-tetrol); at the same time, no presence of D₄ was found in the urine (Varaprath et al., 1999). The production of MST explicitly indicates the demethylation process of D₄ that takes place *in vivo* (Varaprath et al., 1999).

Table 6. Metabolites of [¹⁴C]D₄ in rat urine, as identified and quantified by Varaprath et al. (1999)

Compound	Percent of total radioactivity of the urine
DMSD	30–50
MST	30–38
Dimethyldisiloxane-1,3,3,3-tetrol	8–10
Tetramethyldisiloxane-1,3-diol	2–10
Trimethyldisiloxane-1,3,3-triol	4–8
Dimethyldisiloxane-1,1,3,3-tetrol	2–6
Hexamethyltrisiloxane-1,5-diol	1

5.2.2 Other Silicones

Dain and Nicoletti (1995) assessed the metabolism of a silicon-containing xenobiotic, 4-[(3-methoxyphenyl)-methyl]-2,2,6,6-tetramethyl-1-oxa-4-aza-2,6-disilacyclohexane hydrochloride (MPSC), in rats, dogs, and humans. What is very interesting is that after a single oral dose, one could verify that the metabolism of MPSC proceeds differently in each of the species (Dain and Nicoletti, 1995).

Different enzymatic systems are responsible for biodegradation of siloxanes *in vivo*. In rats, upon repeated whole-body inhalation exposure to decamethylcyclopentasiloxane (D₅) or D₄, one observed significant induction of hepatic xenobiotic metabolizing enzymes, similar to that reported for phenobarbital (McKim et al., 1995, 1998, 1999). One must remember that both silicones (especially low-molecular-weight and cyclic ones) and indirect products of silicone biodegradation are not biologically inert (Fessenden and Fessenden, 1967; Garson and Kirchner, 1971; Bennet et al., 1972; Hayden and Barlow, 1972; Le Fevre et al., 1972; Le Vier and Jankowiak, 1972; Hobbs et al., 1972; Palazzolo et al., 1972; Alabanus et al., 1975; Le Vier et al., 1975; Le Vier and Boley, 1975; Le Vier and Jankowiak, 1975; Nicander, 1975; Pilbrant and Strindberg, 1975, and references cited therein; Anderson and Robertson, 1995; Nicholson et al., 1996; Sun et al., 1997; Lieberman et al., 1999). In particular, various types of silanols are inhibitors of proteolytic enzymes and regulators of many metabolic processes and of growth and

multiplication of cells, which indicates potentially strong pharmacological action (Gueyne et al., 1991; Sieburth and Mutahi, 1998).

6 Silicone Industry

The silicone industry is one of the most important sectors of the chemical industry. As reported by Chandra et al. (1997), over the period of 1991–1995, an annual growth rate of about 7% for silicone sales was observed in the major consuming nations, and the trend is expected to continue in these nations for several years to come.

6.1 Production of Silicones

The world's production of silicones was estimated at approximately 10 million tonnes in the year 2000 and demonstrates a continuously growing tendency (Prof. J. Chojnowski, personal communication, Center for Molecular and Macromolecular Research, Polish Academy of Science, Łódź, Poland). Annual sales of silicones (on a weight basis, excluding intra-industry sales and inter-plant transfers) was calculated as equaling approximately 613,000 metric tons in the western world in 1995 (Chandra et al., 1997). According to the Centre Européen des Silicones (CES), the total sales of silicones in Europe are estimated at 2.5 billion EUR per year, making it a significant economic sector that accounts for some 10,000 jobs across Europe. Notwithstanding, because silicones have so many different applications (Tomanek, 1991; see also Section 4 above), the overall employment generated by the industry is actually much higher. Domestic consumption of liquid linear or cyclic siloxanes (mainly PDMS) was estimated at 70 to 73 million kilograms in 1991 (SRI International, 1994).

6.2 Main Commercial Silicone Producers

The total sales of the silicone industry figured at about 5.7 billion USD in 1995, 94% of which was accounted for by eight major companies (Dow Corning, USA; General Electric, USA; Wacker Silicones, Germany; Shin-Etsu, Japan; Rhone-Poulenc, France; OSi Specialties, USA; Bayer, Germany; and Huels, Germany) (Chandra et al., 1997; Smart et al., 1996). Over the past few decades, the main global commercial silicone producers have formed three regional nonprofit organizations (The Silicones Environmental, Health, and Safety Council of North America [SEHSC]; Centre Européen des Silicones [CES]; and The Silicone Industry Association of Japan [SIAJ]), with the Global Silicones Council (GSC), representing companies that produce and sell silicone products globally.

The Global Silicones Council (GSC) was formed in 1993 as a nonprofit international organization whose mission is to promote the safe use and stewardship of silicones globally. To accomplish this mission, GSC anticipates and identifies environmental, health, and safety initiatives and coordinates the strategies and activities of the three regional silicone associations (CES, SEHSC, SIAJ) that undertake these initiatives. GSC also encourages industry communication

with international environmental, health, and safety organizations such as the World Health Organization, the Organization of Economic and Community Development, and the United Nations.

CES is a nonprofit trade organization representing all the major silicone and silane manufacturers in Europe, including:

Degussa AG,

Dow Corning Corp,

GE Bayer Silicones,

Goldschmidt AG,

OSi Specialties (formerly Union Carbide),

Rhodia Silicones,

Shin-Etsu Chemical Company, and

Wacker-Chemie GmbH.

CES provides information on silicones to downstream users, industry customers, and the public, mainly from a health, safety, and environmental perspective. CES was formed in 1968, and is a sector group of the European Chemical Industry Council (CEFIC). Contact: David Thomas, CES Secretary General, c/o CEFIC, Avenue van Nieuwenhuyse 4, Box 2, B-1160 Brussels, Belgium; Tel.: +32-2-6767247; Fax: +32-2-6767301; E-mail: dth@cefic.be; Website: <http://www.silicones-europe.com>.

SEHSC is a trade association comprised of North American silicone chemical producers and importers, who represent over 95% of the silicone chemical manufacturing capacity in North America. They include:

Clariant LSM (Florida), Inc.,

Degussa Corporation,

Dow Corning Corporation,

GE Silicones,

OSi Specialties, a Crompton business,

Rhodia Inc.,

Shin-Etsu Silicones of America, and

Wacker Silicones Corporation.

SEHSC operates to fund research and promote the safe and responsible use of silicones. The organization is also involved in legislative and regulatory issues relating to silicone materials. Originally formed in the mid-1970s as the Silicones Health Council (SHC), the organization became SEHSC in 1993. Contact: SEHSC, 11921 Freedom Drive, Suite 550, Reston, VA 20190; Tel.: +1-703-9044322; Fax: +1-703-9255955; E-mail: sehsc@sehsc.com; Website: <http://www.sehsc.com>.

SIAJ represents companies that produce and sell silicone products in Japan. SIAJ was founded in 1967 and includes:

Shin-Etsu Chemical Co., Ltd.,

GE Toshiba Silicone Co., Ltd.,

Dow Corning Toray Silicone Co., Ltd.,

Nippon Unicar Company Ltd.,

Dow Corning Asia Ltd.,

Wacker Asahikasei Silicone Co., Ltd., and

Chisso Corporation.

SIAJ initiatives are aimed at providing accurate information about silicone products and fostering product stewardship globally within the silicone industry. SIAJ also exchanges information and sponsors environmental, health, and safety initiatives related to silicones through cooperation with SEHSC and CES. Contact: SIAJ Secretariat, Shimbashi Yanagiya Building, 16–6 Shimbashi 1-Chome, Minato-ku, Tokyo 105, Japan; Website: <http://www.siaj.jp/> (in Japanese), <http://www.siaj.jp/en/index.html> (in English).

7 Patents

The authors are not aware of patents in the field of biodegradation of silicones besides the patent by Baud-Grasset and Palla (1997), as mentioned above. The patent describes a method of biodegradation of polyorganosiloxanes, particularly polydimethylsiloxanes, occurring at various pH levels, with the use of one or more microscopic fungi (selected from the Corticiacea family), preferably in the presence of a sugar co-substrate (glucose or sucrose) (Baud-Grasset and Palla, 1997).

8 Outlook and Perspectives

Twelve principles of “green chemistry” (Anastas and Warner, 1998) recommended by the American Chemical Society (<http://www.acs.org/portal/Chemistry?PID=acsdisplay.html&DOC=education\greenchem\principles.html>) state, “Chemical products should be designed so that at the end of their function they break down into innocuous degradation products and do not persist in the environment”.

In our opinion, every individual compound, especially polymeric materials, should follow this criterion whenever possible. It is especially noteworthy in the case of siloxanes because of their wide application, high-scale production (approximately 1,000,000 tons per year) with continuous growth, and “inherited” resistance to degradation. Therefore, the idea of a simplification (supporting) of the process of biodegradation in designing any new siloxane materials should be seriously taken into account.

We further propose that in the near future each polymeric material should be accompanied by a piece of information on its biodegradability (e.g., $\tau_{0.5}$ of biodegradation) and specific conditions required to shorten its time, as well as information on the products of its degradation or, in the case of *in vivo* application, $t_{1/2}$ of elimination.

However, our knowledge of the biodegradability of silicones is very poor, and more extensive research must be done in order to attain the necessary data related to this problem. It is an area nearly completely open for exploration, as we have tried to show above. On the other hand, it is urgent that the study of the products of siloxane biodegradation, the ones generated in the course of both *in vitro* and *in vivo* degradation, be intensified with respect to their toxicity. All the doubts about the processes occurring during the *in vivo* biodegradation of medical silicones should be explained and reported exhaustively.

cPDMS	polydimethylcyclsiloxane
cSt	centistokes
D₄	octamethylcyclotetrasiloxane
D₅	decamethylcyclopentasiloxane
D₆	dodecamethylcyclohexasiloxane
DMSD	dimethylsilanediol
MST	methylsilanetriol
OMCTS	octamethylcyclotetrasiloxane

PDMS	polydimethylsiloxane
PEMS	polyethermethoxysiloxane
TMS	trimethylsilanol
VMS	volatile methoxysiloxane

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