

**Thermophysical Properties of  
Organosilicon Compounds**

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**A HANDBOOK**

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## **A Handbook**

**P.G. Alekseev  
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P.I. Povarnin**

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## Foreword

A substantial scientific achievement of last 50 years was the creation of new silicon-based compounds, organosilicons [118]. Progress in synthesis theory, as well as the discovery of unique properties of organosilicons, have made large-scale production of new compounds possible. There were obstacles that posed considerable problems both for consumers and for investigators of properties of substances. Starting in the Sixties, commercial-scale production of organosilicons, which only started about 40 years ago, has doubled in capacity approximately every five years.

Because of their unique properties, organosilicons have found wide application in different areas of science and technology. This gives us a powerful incentive to investigate their thermophysical properties, as this knowledge provides new possibilities for the more efficient use of new substances, and for the development of new technologies and equipment.

Presently, however, a significant, still growing gap exists between the need for and the availability of thermophysical data in science and industry. There are two reasons for this gap: we lack a theory of liquid state, and have to deal with expanding nomenclature of new organosilicons [89, 118]. Efforts to minimize this gap, based solely on experimental data and empirical correlations, do not, and can not give positive results.

What is urgently needed in this situation, is to develop semiempirical methods of estimating such properties. Such methods would be based on the analysis and generalization of the available body of experimental data [51, 89]. In the literature, both in Russia and abroad, there are almost no published papers concerning the systematization and generalization of thermophysical properties of organosilicons; this is understandable in view of the paucity of pertinent experimental evidence [51, 89].

The aim of the authors of the present handbook is to apply existing techniques, developed for investigation of other substances, to estimate the properties of organosilicons in a wide range of parameters.

The computational results summarized in the handbook were obtained using methods based on both the principle of thermodynamic similarity and on the additive principle [17, 99].

Usage of the thermodynamic similarity principle was appropriate where we had available the experimental information required to determine both the reference scales and the temperature dependence of the property.

The additive principle was used to estimate the properties of organosilicons in so called "standard" state ( $T = 293.15$  K,  $p = 1.03 \cdot 10^5$  Pa). The numerical values thus obtained subsequently enabled us to use the thermodynamic similarity principle.

The numerical values presented in the handbook qualify as "informational" in accordance with GOST 8.310-78.

We wish to express particular gratitude to Professor A.A.Gukhman for his helpful discussions and suggestions, which are reflected in the material appearing in the Section 1 of the handbook.

We would be thankful for feedback on this handbook. The readers are kindly requested to use the following address: The Begell House Inc. Publishers, 79 Madison Av., New York, NY 10016-7892.



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## Introduction

Organosilicons are most extensively used in such modern technologies as rocket building, the airspace industry, and radio electronics [56, 76, 79, 81, 88]. Despite comparatively high cost, organosilicons recently started to oust traditional materials in the shipbuilding and fabric industries [56], the construction industry, medicine, and other branches of science and industry [31, 47, 77, 78, 80]. Organosilicons are widely in use as coatings [79, 81], lubricants [76, 88], and heat-transport mediums [19, 118]. Their unique properties give them advantages over existing substances.

In the literature, both in Russia and abroad, papers concerned with systematization and generalization of thermophysical properties of organosilicons are almost completely lacking [70, 92]. This is because we lack pertinent experimental data.

While there is rather extensive literature available on the chemical structure and physical, chemical [9, 12, 24, 28, 35, 49, 82, 119], and thermodynamic properties [46, 66, 75, 76, 93, 95], the thermophysical property-data are very scarce and remain practically unclassified. Sufficiently detailed information is available only for a few compounds [84–87, 118]; for most organosilicons experimental data are available only for a single state, e.g. at  $T = 293.15$  K [51, 89].

The knowledge of various properties of organosilicons in a wide range of thermodynamic parameters is necessary both for engineers—to predict the parameters of technological processes [9, 19, 31, 77, 89] and the ranges of application of substances—and for the scientist—to investigate the relation between a substance's chemical structure and its properties.

Presently, due to the lack of a sufficiently rigorous theory of the liquid state, the properties of a new substance cannot be predicted from existing theory. At the same time, in most cases, experimental data that are of interest for designers are scarce or completely lacking.

For the reason listed previously, there is a need to develop semiempirical methods for estimating properties of substances. There are many methods presently available for estimating thermophysical properties, which fall into two groups. The first group is estimations based on thermodynamic similarity [97–107]. The second group includes those methods that are based on the additivity of the contributions made to properties by molecular structural elements [17, 100, 102–110]. Each of these has advantages and disadvantages, and in some cases are supplementary to one another.

Calculations based on the thermodynamic similarity principle imply that a number of scaling factors are known, which are suitable for estimating a wide variety of properties and states of a substance in a sufficiently wide region of reference parameters.

The additive principle can be used for calculations of a number of properties, such as critical temperature pressure and volume, latent heat of evaporation, boiling temperature at ambient pressure, etc.

In developing the additive calculating procedure to predict the properties of substances, the authors of refs. [17, 100, 102–110] have confined themselves to a few separate examples.

By applying existing methods for estimating a number of thermophysical properties of particular chemical compounds, organosilicons, the authors of the present handbook hope to generate property-tables for wide regions of thermodynamic parameters. These should be sufficiently accurate for practical usage.

In most cases, a straight forward application of both estimation methods was impossible and we had to introduce a number of modifications into these methods to allow for the peculiarities of organosilicons.

One of the primary goals of correlating experimental data is selecting a method of classification of the objects under investigation. A wide variety of silicon compounds, in particular, create some difficulties in the development of a unified nomenclature system [48, 65]. At present, there are several different nomenclatures, and one of the primary goals of the International Union of Pure and Applied Chemistry (IUPAC) [65] is to bring together the nomenclatures and terminology that exist in different languages.

Nomenclature of organosilicon compounds, according to IUPAC rules, is based on the precedence of atoms and atom groups and is outlined in refs. [64, 65, 68]. Chemical names, along with a short description of organosilicons commercially available in Russia, are summarized in a number of sources [27, 31, 35, 40, 77].

A nomenclature system most widely used for silicones is a substitutive one, where a hypothetical carbon open-chain, or carbon cycle compound, serves as a parent compound, and silicon heteroatoms substitute in part or completely for carbon heteroatoms. Among groups with the same central atom, the group with the higher degree of oxidation of this atom receives precedence [64, 65, 68].

Of a wide variety of organosilicons available, only two main types—silicon hydrides and their substituted siloxanes with a  $-\text{Si}-\text{O}-\text{Si}-\text{O}-\text{Si}-$  skeleton have been selected for discussion in this handbook.

A typical feature of silicones is the presence of a central silicon atom, to which a variety of groups and atoms, such as  $\text{SiH}_4$ ;  $(\text{CH}_3)_3\text{SiCl}$ , can be attached. Compounds of the general formula  $\text{H}_3\text{Si}-\text{SiH}_3-$  (disilanes) or  $(\text{SiH}_2)_n-$  (cyclosilanes) have not been included.

Compounds containing a  $-\text{Si}-\text{O}-\text{Si}-$  skeleton with their remaining silicon bonds occupied by organic radicals, are commonly termed oligoorganosiloxanes or polyorganosiloxanes [9, 35, 89]. Such compounds, holding no functional groups for subsequent polymerization and containing closed end-groups, remain liquids in a wide temperature region—regardless of their molecular mass which can range from several hundred to a few thousand. Such substances, commonly termed organosilicon liquids, have found wide applications owing to their thermal stability, unique physical properties, and good capacity for lubrication and insulation, etc.

The rules of classification of oligoorganosiloxanes cannot be considered as completely established [24, 35, 39, 47, 68, 80]. Indeed, there are different accepted classifications—based on the chemical composition and molecular structure, on properties and areas of application (as lubricants, electrical insulators, heat transfer agents) and on other keys [56, 76, 95, 116].

The properties of liquid oligomer systems are primarily affected by the composition and structure of the end groups. In principle, the basic properties of oligoorganosiloxanes can be modified solely by substituting end groups and by changing their composition.

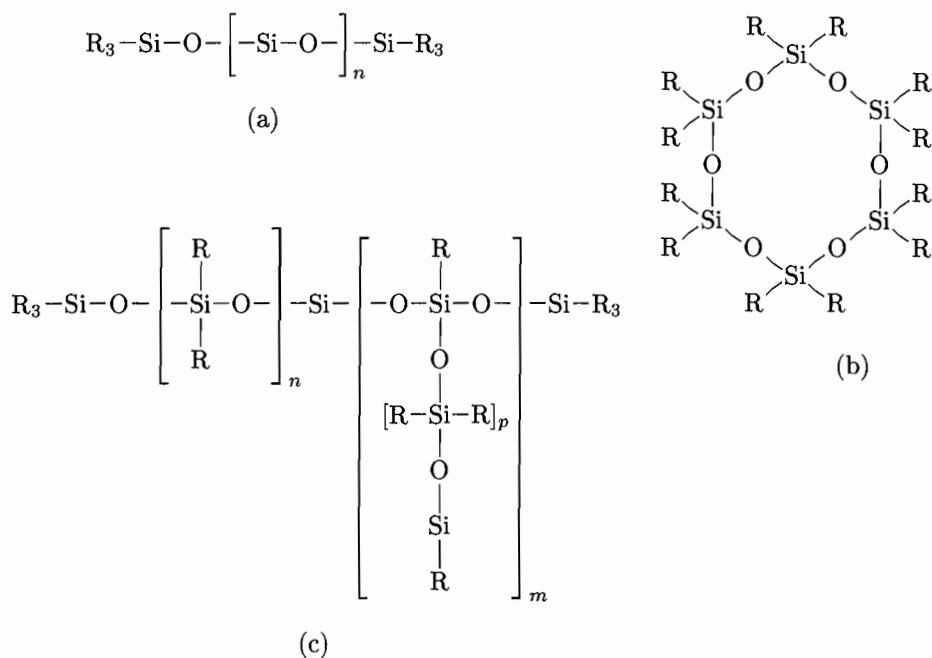


Figure I.1: Structure of organosiloxane molecules.  
a—linear, b—cyclic, c—branched

Structurally, oligoorganosiloxane molecules can be linear, cyclic or branched (Fig. I.1). Linear molecule compounds have lower viscosities than cyclic compounds and relatively weak physical properties-temperature dependencies, and branched-molecule compounds occupy intermediate positions [9, 89]. Below, organosilicons with linear, cyclic, and branched molecules are termed linear, cyclic, and branched, respectively.

The handbook includes thermophysical properties of the following oligoorganosiloxanes:

1. Oligodimethylsiloxanes or polydimethylsiloxanes can be linear, cyclic, or branched. Technical-grade mixtures of such compounds are labeled according to their kinematic viscosity values at 293.15 K, given in centistokes [84, 89]. Polyorganosiloxanes have lower intermolecular forces compared to other oligoorganosiloxanes as well as lower vitrification temperatures and melting temperatures [69, 84, 89, 117].

2. Oligodiethylsiloxanes or polydiethylsiloxanes have a looser structure than polydimethylsiloxanes. This is because of the larger volume occupied by the radicals  $C_2H_5$  compared to that occupied by  $CH_3$  [9]. They can be either linear or cyclic. The main peculiarity of polydiethylsiloxanes is their good compatibility with mineral oils, which enables them to be used widely as a component of compound lubricants [69, 76, 84, 88].

3. Combined compounds such as polydimethylsiloxane-polydiethylsiloxane, where in the radicals  $CH_3$  and  $C_2H_5$  alternate, have properties similar either to those of polydimethylsiloxanes or to those of polydiethylsiloxanes, according to which of the radicals prevails in a molecule. Such compounds can be either linear or cyclic [51].

4. Oligomethylphenylsiloxanes or polymethylphenylsiloxanes can be derived by the introduction of phenyl groups into the dimethylsiloxane chain [9, 89]. Such compounds offer elevated thermal stability, low saturated vapor pressure, low evaporativity, and

high ignition temperature [77]. The properties of polymethylphenylsiloxanes are dependent on the number of  $n$  ( $\text{CH}_3$ ) and  $m$  ( $\text{C}_6\text{H}_5$ ) ( $\epsilon$ ) radicals in a molecule [42, 43, 86, 117].

5. Organosiloxanes with halogen-containing organic radicals have more pronounced polar properties than their parent liquids [9, 89]. The presence of halogens in these compounds provides improved lubricating properties [88], alters both their solubility and compatibility with organic media, and reduces inflammability [20, 87]. Such compounds can be linear, cyclic, or branched. The ratio of the number of halides  $n$  (Cl) to the total number of atoms in a molecule.

Halogen-containing organosiloxanes are complex mixtures of molecules, differing from one another by degree of both polymerization and composition. Depending on the type of halogen present, these fall into three groups: oligomethylfluoroorganosiloxanes, oligomethylchloroorganosiloxanes, and oligomethylfluorochloroorganosiloxanes.

The thermophysical properties of such compounds depend both on the degree of polymerization of constituent molecules and on their composition and structure.

An inherent feature of such compounds is a stronger dependency of viscosity on temperature. This is explained by the increase of intermolecular interaction intensity due to the additional attraction of polar carbon halide bonds and to a certain correlation between the halogen and silicon atoms of adjacent molecules.

6. Oligoorganoalkoxysiloxanes are derived by introducing alkoxi-groups into oligoorganosiloxane molecules. This enhances their lubricating ability and compatibility with other organic compounds. Their thermal stability is dependent on the nature of the substituents of both silicon atoms and alkoxi-groups. These organic radicals, their volume and nature, have a major effect on the properties of the resulting oligomers.

The thermophysical properties of oligoorganoalkoxysiloxanes are related to the elasticity and rigidity of molecule chains as well as to their conformational mobility. This may result in unwinding the molecular ball due to a temperature rise.

7. With a hydrogen atom present in an oligomer molecule, polydimethylsiloxane hydride- and polydiethylsiloxane hydride-liquids will be formed. Such compounds are widely used as water-repellent coatings for various surfaces and objects. The protective coatings thus obtained can withstand temperatures as high as 580 K and do not mix with water or organic solvents [77, 81].

8. Bis(organo-cyclosiloxi)polydimethylsiloxanes form a separate group of "cellular" polymers with dumbbell-like molecules. The results of experimental studies of such compounds are summarized in refs. [7, 9, 36, 38].

The analysis, systemization, and generalization of available experimental data enabled us to put necessary improvements into the estimation procedures offered in refs. [17, 72, 99–107], to allow for the structure and composition of organosilicon molecules. Based upon such analysis, the correlation equations and numerical values of constants appearing in these equations were deduced and the thermophysical property tables were calculated. The errors of the data estimations do not exceed 10 percent. The correlations thus obtained can be used to predict thermophysical properties of newly synthesized compounds.

## Notation

$T$	temperature, K
$p$	pressure, Pa
$v$	specific volume, $\text{m}^3/\text{kg}$
$V_\mu$	molal volume, $\text{m}^3/\text{kmol}$
$M$	molecular mass
$c_p$	specific isobaric heat capacity, $\text{kJ}/(\text{kg}\cdot\text{K})$
$c_v$	specific isohoric heat capacity, $\text{kJ}/(\text{kg}\cdot\text{K})$
$\lambda$	thermal conductivity, $\text{W}/(\text{m}\cdot\text{K})$
$\eta$	viscosity, dynamic, $\text{Pa}\cdot\text{c}$
$\nu$	viscosity kinematic, $\text{m}^2/\text{c}$
$\rho$	density, $\text{kg}/\text{m}^3$
$\Delta H_{\text{vap}}$	enthalpy change during vaporization (heat of vaporization), $\text{kJ}/\text{kmol}$
$\sigma$	surface tension, $\text{N}/\text{m}$
$a$	thermal diffusivity, $\text{m}^2/\text{c}$
$n_D^{20}$	refraction index at $T = 293.15 \text{ K}$
$z$	compressibility factor
$R$	gas constant, $\text{kJ}/(\text{kg}\cdot\text{K})$
$R_\mu$	universal gas constant
$\tau = T/T_{\text{cr}}$	reduced temperature
$\pi = p/p_{\text{cr}}$	reduced pressure
$\varphi = v/v_{\text{cr}}$	reducer volume
$x$	property to be found
$\omega_i$	contribution of a chemical structure element to the property to be found
$\varepsilon$	ratio of a number of methyl radicals ( $\text{CH}_3$ ) to a number of phenyl radicals ( $\text{C}_6\text{H}_5$ ) in an organosilicon molecule
$R_D$	mole refraction

## Subscripts

liq	liquid
v	vapor
gl	glass transition
cr	critical state
s	saturation
id	ideal-gas state
melt	melting
boil	boiling
vap	vaporization
td	thermal decomposition

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# Chapter 1

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## Methods of Estimating Thermophysical Properties of Substances Based on the Thermodynamic Similarity Principle

There is a marked gap between the potentials of the theory and the requirements that arise during the development of analytical methods for estimating the thermophysical properties of substances. Existing theoretical models can only offer a possibility to estimate the properties. In actual practice, however, they hold well only for the simplest individual gases [17, 26, 72].

This gap is felt most keenly during the investigation of liquids, for which no sufficiently complete and exact theory is available. For liquids, in most cases, the main source of the evidence concerning the thermophysical properties is an experiment and its respective correlating procedures.

### 1.1 Basic Concept of Thermodynamic Similarity

Though we lack a theory of liquid state and, consequently, analytical techniques of estimating thermophysical properties, a large volume of experimental data is being accumulated, which needs to be subsequently analyzed and correlated. Presently there are no authoritative guidelines prevailing opinion concerning the system to outline the parameters that would comprehensively describe a property of the substance.

As a consequence, the objectives pursued by experimental investigation are often confined solely to the solution of particular problems. This does nothing to develop a general system for estimating the thermophysical properties of the liquid state [72, 86].

Such a system could allow us to concentrate on the investigation of thermophysical properties in major directions. The data thus obtained could serve as a basis for formulating extensive correlations, for creating theory of the liquid state, and for developing methods for predicting thermophysical properties. Among liquids there are compounds for which this problem is very acute, because of the paucity of pertinent thermophysical data. Organosilicons are among them.

Generally, a substance property can be expressed in terms of various parameters, as follows [86]

$$x = x(p, T, d, k, l, m, n \dots), \quad (1.1)$$

where  $d, k, l, m, n \dots$  are the parameters that characterize molecular structure.

Strictly speaking, eq. (1.1) defines the property of a single substance, provided all the parameters involved are assigned numerical values.

The above implicit form of eq. (1.1) is of little value; however, it is inconceivable that an explicit form of this equation will be developed on the basis of the theory. The available empirical correlations of experimental data for a single substance cannot be used to predict thermophysical properties for a wide range of substances [52]. This problem is too involved; but one of the tools that can be used to solve it is the theory of similarity [32]. This allows us to reduce a number of variables, by substituting dimensionless quantities composed of the parameters appearing in eq. (1.1). Written in dimensionless form, eq. (1.1) relates generalized variables, each numerical value of which corresponds to a set of real parameters appearing in eq. (1.1).

The particular combination of dimensionless quantities can be selected on the basis of detailed analysis of the structure and the state of the object under study. Generalized expressions, relating quantities of different physical nature within a single formula, can be generated from the theory of similarity combined with the experiment.

The theory of similarity is usually applied to a boundary problem of mathematical physics [32] where a set of differential equations, describing a physical phenomenon, is added by the boundary conditions. The phenomena that obey the same differential equations and identical boundary and initial conditions will be similar. In addition, numerical values of a number of dimensionless quantities (dimensionless numbers) obtained by the simple procedures outlined in the theory of similarity should be identical.

## 1.2 Calculation of Critical State Parameters and Thermodynamic Properties of Substances Using the Thermodynamic Similarity Principle

In practice, problems often occur where the similarity concept reduces itself to the determination of dimensionless relations linking macroscopic quantities which characterize the properties of the body, with the dimensionless parameters, which are the functions of geometry, dynamic properties of interaction between structural elements, and external (thermodynamic) parameters. In this case, we may speak about thermodynamic similarity by extrapolating this definition to the calorific equation of state (theory of corresponding states) and to kinetic properties [99, 103].

A group of similar substances has to obey the dimensionless equation of state (the law of corresponding states) that takes the form

$$f(\pi, \tau, \varphi) = 0, \quad (1.2)$$

and is plotted for the particular case of carbon compounds in Fig. 1.1.

From the point of view of the molecular theory, the validity of the corresponding states principle follows from the mechanical similarity of the system of molecules that obeys the same dimensionless intermolecular potential equation. In fact, the interaction-potential profiles define a group of thermodynamically similar substances. Thus, for example, a two-parameter Lennard-Jones potential satisfactorily describes a group of thermodynamically similar substances such as noble gases. By choosing another form of the two-parameter potential we can describe another group of thermodynamically similar substances.

However, no generalized representation of thermophysical properties of a wide range of substances can be obtained using a two-parameter potential function. Toward this end, one must use a more complex, three- or more- parameter potential function. As was shown by Arutyunov and Kazancheva [12, 13], a three-parameter interaction

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