



Silanes and Siloxanes Thermal Conductivity in the Liquid Phase: A Critical Review and an Improved Prediction Method

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ABSTRACT

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The thermal conductivity λ of the silanes and siloxanes families in the liquid phase at atmospheric pressure or along the saturation line is investigated as function of the reduced temperature. Because of the large scarcity or even of the lack of accurate experimental λ data an empirical equation is proposed as a generalization based on investigations presented in previous works [1, 2]. The families of silanes and siloxanes (21 chlorosilanes, 5 cyclosiloxanes, 10 linear siloxanes, 10 silanes and 19 other silanes) are taken into consideration using a large database [3] in order to extend the use of a general formula valid for organic compounds (alcohols, alkanes, ketones,...) and to improve preceding results obtained in the case of the cited silanes and siloxanes, for which experimental thermal conductivity data at atmospheric pressure or along the saturation line in the liquid phase are available in very few cases. The equation is proposed as acceptable for engineering purposes and comparable with the existing prediction methods [3]. The database DIPPR [3] in the version 2020 containing a linear correlation with various parameters is taken into account, also considering the results of 7 other prediction methods existing in the technical and scientific literature. An extensive and critical comparison points out that the method proposed in this work can be considered valid with absolute errors usually not greater than 5%.

1. INTRODUCTION

As well known the Fourier's law [4], for example in the mono-dimensional case, contains the thermal conductivity λ :

$$dq = -\lambda dA \frac{dT}{dx} \quad (1)$$

where, dq is the heat flow along the x axis, dA is an infinitesimal area normal to the direction x and T is the temperature. The SI units are used, unless differently stated.

The thermal conductivity λ of the substances in the liquid phase is closely related to temperature, pressure and chemical structure, but rigorous theoretical investigations, based on classical or quantum mechanics, lead to general results usually not acceptable for engineering purposes because of large errors; the theory of the liquid state is far away from producing acceptable results when, for example, the saturation line is investigated. In the liquid phase reasonable approximations are available only for some compounds characterized by simple chemical structure and in any case when several *ad hoc* constants are introduced. For gases and solids on the contrary models based on the similarities with the characteristics of the perfect gas and on the crystalline symmetries respectively are valid instruments for general theories. Moreover, accurate experimental thermal conductivity data for compounds in the liquid state are very often not available at all, and due to the lack of a general theory, empirical or semiempirical equations can provide reasonably accurate thermal conductivity data valid for engineering purposes. In this work, on the basis of

investigations presented in previous papers [1, 2] the families of silanes and siloxanes (21 chlorosilanes, 5 cyclosiloxanes, 10 linear siloxanes, 11 silanes and 20 other silanes) are taken into consideration using a larger database [3] in order to extend the use of a general formula valid for organic compounds (alcohols, alkanes, ketones,...) and to improve preceding results obtained in the case of the cited silanes and siloxanes, for which experimental thermal conductivity data at atmospheric pressure or along the saturation line in the liquid phase are available in very few cases. Substantially two parameters characterize the thermal conductivity of the investigated compounds, the first is an exponent a , the same for the all the compounds of the family, and the second is $\lambda_{0.618}$, that is the thermal conductivity estimated at the value of the reduced temperature equal to $(\Phi - 1)$ the mantissa of the "golden ratio" $\Phi = (1 + \sqrt{5})/2 = 1.618033...$ The proposed method is acceptable for engineering purposes and comparable with the method proposed by DIPPR [3] (a linear correlation with various and different parameters). An extensive and critical comparison between very different methods points out that the method proposed in this work can be considered valid with absolute errors not greater than 5%.

Because the dependence of the thermal conductivity λ upon the pressure P for the compounds in the liquid phase is negligible up to 40-50 bar, only the temperature is taken into account in this work concerning λ at atmospheric pressure or along the saturation line. On the other hand, as above pointed-out, very few accurate experimental λ values for liquids as function of temperature are available in technical and scientific literature. Moreover, with very limited exceptions,

from the normal melting point to the normal boiling point and over, the liquid thermal conductivity decreases almost linearly, and the critical point represents a singularity.

Some interesting models [5] present the liquid thermal conductivity as function of temperature and density by the sum of three contributions ("dilute gas contribution", "excess contribution" and "critical enhancement contribution"), but in this work, because the explored range is near or along the saturation line, the density dependence, as for the pressure dependence, is neglected.

2. THE PREVIOUS EQUATIONS

The large review contained in [1, 2] presents a summary of the empirical or semi-empirical methods appeared in the literature and presented as suggested by Govender [6]: general correlation methods, family methods, group contribution methods and corresponding state methods. The collection due to Horvath [7] and the classical book of Poling, Prausnitz and O'Connell [8] were also taken into account.

The work of Latini and his group [9-11] began in the 80's with the equation, reported also by Poling, Prausnitz and O'Connell [8]:

$$\lambda = A \frac{(1 - T_r)^{0.38}}{T_r^{\frac{1}{6}}} \quad (2)$$

$T_r = T/T_c$ is the reduced temperature, the factor A depends upon molecular weight M , critical temperature T_c , normal freezing point T_f and normal boiling point T_b .

A deepest exploration of the experimental λ data and a suitable mathematical apparatus [1, 2] lead to the expression:

$$\lambda = A \left[\sqrt{5} \frac{(\Phi - T_r)^2}{(\Phi + T_r)} \right]^a \quad (3)$$

Φ is the "golden ratio", the limit of the Fibonacci's sequence:

$$\Phi = \lim_{n \rightarrow \infty} \frac{F_{n-1}}{F_n} = \frac{1 + \sqrt{5}}{2} = 1.618033989 \dots \quad (4)$$

Otherwise given by: $\Phi = 1 + 1/\Phi$. Φ can be considered as characteristic of the liquid phase, the exponent a of the investigated family and A (which has the units Watt/(m K) of λ) is characteristic of the single compound and is equal to the value of the thermal conductivity when $T_r = 0.618033989 \dots$, the mantissa of the "golden ratio", usually a value near the reduced normal boiling point.

The first attempt for an expression for A was as follows for the organic family of alcohols [1] and also for alkanes, ketones and silanes [2]:

$$A = h \frac{T_c^{\frac{5}{4}}}{M^{\frac{1}{2}}} \quad (5)$$

where, h is a value characteristic of the single family.

3. THE PROPOSED EQUATION

In this work the Eq. (3) is examined again with respect to A

which is given by expression (5); this procedure changes the problem from A to h and, while A has a precise meaning (it is the thermal conductivity at the reduced temperature $T_r=0.618 \dots$), h derives from a process of optimization and has units of measure without any particular meaning. Substantially the factor h and the exponent a are coupled to give the minimum mean absolute deviation between experimental (when available) and predicted λ data. Finally, at the end of the mathematical examination, the value of the factor h determines the value of the exponent a and vice versa; moreover, the exponent a is adimensional and is linked to the slope of the function (3) at every value of the reduced temperature. What is proposed is to return to the physical meaning of A , that is:

$$A = \lambda_{0.618033989 \dots} \quad (6)$$

At this point the value of A has to be found in the scientific and technical literature, rendering the choice of the exponent a entirely independent. In any case the liquid thermal conductivity, which is quite linear as function of the temperature, depends upon two parameters and what is proposed is to accept expression (6) in order to search for the value of a . The procedure adopted in this work is based on the following points:

- the value of (6) is taken from the Database DIPPR (3);
- the families taken into account are Silanes and Siloxanes.

The choice of the Database DIPPR has the following reason:

- the available liquid experimental or predicted λ data in technical and scientific literature are by different apparatuses and mathematical procedures with very different accuracies;
- the test of Eq. (5) where A is given by expression (6) has to be developed by taking into account the large, updated and reliable collection of thermal conductivity data contained in the DIPPR database: the λ data are clearly reported as experimental, predicted and smoothed; in any case the accuracy of the thermal conductivity data is clearly indicated.

4. THE TEST OF EQUATION (5) WHEN A IS GIVEN BY EXPRESSION (6)

Silanes and siloxanes are silicon derived compounds and the monomers that form the semi-inorganic polymers based on silicon derive from nomenclature of silane SiH_4 in analogy to the organic polymers whose terminology is compared to the methane. The fluids can be grouped in four different sub-groups: chlorosilanes (21 compounds), cyclosiloxanes (5 compound), linear siloxanes (10 compounds), silanes (10 compounds) and "other" silanes (19 Compounds).

The explored reduced temperature range was usually from 0.30 to 0.80 (practically from the normal melting point to the normal boiling point) at atmospheric pressure or along the saturation line.

Tables 1-5 contain for the compounds of each family: name of the compound, structure of the compound, molecular weight M , critical temperature T_c , reduced temperature ranges of the investigation, the claimed error by DIPPR, value of a , value of $\lambda_{0.618}$ and the general results (average absolute deviations $\Delta_m\%$ and maximum absolute deviations $\Delta_M\%$ between the thermal conductivity values calculated by Eqns. (5) and (6) and the corresponding experimental or experimental & predicted values by DIPPR.

Table 1. Chlorosilanes

COMPOUND	Structure	M [kg/kmol]	T _c [K]	ΔTr	λ (DIPPR claimed error)	a	λ0.618	Δm%ΔM%
MONOCHLORO SILANE	H3SiCl	66.56230	399.200	0.39-0.61	<10%	0.500.162584	0.2	0.5
METHYL CHLOROSILANE	CH3SiH2Cl	80.58890	442.000	0.31-0.64	<25%	0.500.140696	0.8	2.3
DIMETHYLCHLOROSILANE	(CH3)2SiHCl	94.61548	472.000	0.34-0.65	<25%	0.500.117486	0.4	1.4
DICHLORO SILANE	H2SiCl2	101.00738	451.500	0.33-0.80	<10%	0.500.167208	1.6	4.9
TRIMETHYL CHLOROSILANE	(CH3)3SiCl	108.64206	497.750	0.43-0.66	<25%	0.500.124639	0.2	0.4
METHYL DICHLOROSILANE	CH3SiHCl2	115.03396	483.000	0.38-0.65	<25%	0.500.123587	0.2	0.8
DIMETHYL DICHLOROSILANE	(CH3)2SiCl2	129.06054	520.350	0.38-0.66	<25%	0.500.124665	0.2	0.9
TRICHLORO SILANE	SiHCl3	135.45244	479.000	0.30-0.79	<10%	0.500.117347	0.7	3.1
METHYL VINYL DICHLOROSILANE	CH2CHSiCH3Cl2	141.07124	544.100	0.33-0.67	<25%	0.500.092083	0.4	1.5
METHYL TRICHLOROSILANE	CH3SiCl3	149.47902	517.000	0.38-0.66	<10%	0.500.137143	0.2	0.9
DICHLORODIETHYL SILANE	(CH3CH2)SiCl2 (CH2HC3)	157.11370	595.750	0.30-0.80	<10%	0.500.117754	0.8	3.4
VINYLTRICHLORO SILANE	CH2CHSiCl3	161.48972	543.150	0.33-0.80	<10%	0.500.120556	0.8	3.3
ETHYLTRICHLORO SILANE	CH3CH2SiCl3	163.50600	559.950	0.30-0.66	<25%	0.500.118027	0.8	2.8
TETRACHLORO SILANE	SiCl4	169.89750	507.000	0.56-0.65	<25%	0.500.097490	1.5	3.4
3-CHLOROPROPYL DIMETHYLCHLOROSILANE	ClCH2CH2CH2Si (CH3)2(Cl)	171.14000	640.700	0.30-0.80	<10%	0.500.109399	0.8	3.4
PHENYLMETHYL DICHLOROSILANE	(C6H5)Si(CH3)Cl2	191.12992	689.000	0.33-0.69	<25%	0.500.096997	0.3	1.1
PHENYLTRICHLOROSILANE	(C6H5)SiCl3	211.54840	688.000	0.34-0.69	<10%	0.500.102081	0.3	1.0
3-CHLOROPROPYL TRICHLOROSILANE	SiCl3(CH2)3Cl	211.97700	661.100	0.34-0.69	<10%	0.500.114226	0.3	0.7
DIPHENYL DICHLOROSILANE	(C6H5)2SiCl2	253.19930	814.000	0.31-0.71	<10%	0.500.086738	0.8	2.4
(3-METHYLACRYLOYL) TRICHLOROSILANE	CH2=C(CH3)COO (CH2)3SiCl3	261.60600	708.100	0.31-0.74	<25%	0.500.088924	1.0	3.4
HEXACHLORO SILANE	CH2=C(CH3)COO (CH2)3SiCl3	268.889	599.7	0.45-0.70	<25%	0.500.083807	0.8	2.3

Table 2. Cyclosiloxanes

COMPOUND	Structure	M [kg/kmol]	T _c [K]	ΔTr	λ (DIPPR claimed error)	a	λ0.618	Δm%ΔM%
HEXAMETHYL CYCLO TRISILOXANE	-((CH3)2SiO)3-	222.4620	554.200	0.61-0.73	<10%	0.500.10272	1.2	2.6
OCTAMETHYL CYCLO TETRASILOXANE	-((CH3)2SiO)4-	296.6160	586.500	0.50-0.76	<10%	0.500.10079	1.1	2.1
DECAMETHYL CYCLOPENTA SILOXANE	-((CH3)2SiO)5-	370.7700	617.400	0.37-0.78	<10%	0.500.11089	2.3	5.5
DODECAMETHYL CYCLOHEXA SILOXANE	-((CH3)2SiO)6-	444.9236	645.800	0.42-0.80	<10%	0.500.11436	1.9	4.1
HEXADECA METHYL CYCLO OCTASILOXANE	-[Si(CH3)2O]8-	593.2320	689.200	0.44-0.84	<10%	0.500.11791	1.5	3.1

Table 3. Linear siloxanes

COMPOUND	Structure	M [kg/kmol]	T _c [K]	ΔTr	λ (DIPPR claimed error)	a	λ0.618	Δm%ΔM%
HEXAMETHYL DISILOXANE	(CH3)3SiOSi(CH3)3	162.3775	519.000	0.39-0.90	<10%	0.350.09904	2.2	4.2
OCTAMETHYL TRISILOXANE	(CH3)3SiOSi(CH3)2 OSi(CH3)3	236.53100	564.400	0.34-0.90	<10%	0.350.09824	1.4	3.0
HEXACHLORO	Cl[Si](Cl)(Cl)O[Si]	284.89000	594.7	0.41-0.55	<10%	0.350.08139	3.3	6.9

DISILOXANE	(Cl)(Cl)Cl								
DECAMETHYL TETRASILOXANE	(CH ₃) ₃ SiO(Si(CH ₃) ₂ O) ₂ Si(CH ₃) ₃	310.68500	599.400.34-0.90	<5%	0.350.09800	0.4	1.1		
DODECAMETHYL PENTASILOXANE	(CH ₃) ₃ SiO(Si(CH ₃) ₂ O) ₃ Si(CH ₃) ₃	384.83900	628.400.31-0.80	<10%	0.350.10228	0.1	0.5		
TETRADECA METHYL HEXASILOXANE	(CH ₃) ₃ SiO(Si(CH ₃) ₂ O) ₄ Si(CH ₃) ₃	458.99300	653.200.33-0.88	<10%	0.350.10662	0.4	1.0		
HEXADECA METHYL HEPTASILOXANE	(CH ₃) ₃ SiO(Si(CH ₃) ₂ O) ₅ Si(CH ₃) ₃	533.14700	671.800.29-0.83	<10%	0.350.11192	0.1	0.5		
OCTADECA METHYL OCTASILOXANE	(CH ₃) ₃ SiO(Si(CH ₃) ₂ O) ₆ Si(CH ₃) ₃	607.30100	688.900.31-0.85	<25%	0.350.11725	0.2	0.7		
EICOSAMETHYL NONASILOXANE	Si(CH ₃) ₃ O(Si(CH ₃) ₂ O) ₇ Si(CH ₃) ₃	681.45500	707.200.29-0.86	<25%	0.350.12247	0.4	1.2		
HEXACOSA METHYL DODECASILOXANE	Si(CH ₃) ₃ O(Si(CH ₃) ₂ O) ₁₀ Si(CH ₃) ₃	903.91700	751.300.27-0.89	<25%	0.350.13910	1.1	2.7		

Table 4. Silanes

COMPOUND	Structure	M [kg/kmol]	T _c [K]	ΔTr	λ (DIPPR claimed error)	a	λ(0.618)Δm%ΔM%		
SILANE	SiH ₄	32.11726	269.700.33-0.60	<25%	0.80	0.1120	1.8	2.5	
METHYL SILANE	CH ₃ SiH ₃	46.14384	352.500.33-0.61	<25%	0.50	0.1584	0.8	2.2	
DIMETHYL SILANE	(CH ₃) ₂ SiH ₂	60.17042	402.000.31-0.63	<25%	0.50	0.1458	0.9	2.6	
TRIMETHYL SILANE	(CH ₃) ₃ SiH	74.19700	432.000.32-0.65	<25%	0.50	0.1215	0.7	2.1	
TETRAMETHYL SILANE	Si(CH ₃) ₄	88.22358	450.400.39-0.67	<25%	0.50	0.1215	0.2	0.7	
TRISILANE	[SiH ₃][SiH ₂][SiH ₃]	92.32000	495.900.31-0.80	<10%	0.50	0.1384	0.7	3.3	
n-TETRASILANE	SiH ₃ SiH ₂ SiH ₂ SiH ₃	122.421	570.600.31-0.80	<10%	0.5	0.1417	0.7	3.2	
TETRAETHYL SILANE	Si(C ₂ H ₅) ₄	144.32990	606.000.31-0.70	<25%	0.50	0.1089	0.4	1.4	
n-PENTASILANE	SiH ₃ SiH ₂ SiH ₂ SiH ₂ SiH ₃	152.523	628.600.32-0.80	<10%	0.5	0.1429	0.7	3.2	
(3,3,3-TRIFLUOROPROPYL)METHYLDICHLORO SILANE	C(F ₃)CH ₂ CH ₂ Si(Cl) ₂ CH ₃	211.085	561.700.36-0.80	<10%	0.5	0.09634	0.7	3.1	

Table 5. Other silanes

COMPOUND	Structure	M [kg/kmol]	T _c [K]	ΔTr	λ (DIPPR claimed error)	a	λ(0.618)Δm%ΔM%		
TRIMETHYL SILANOL	(CH ₃) ₃ SiOH	90.1964	562.500.54-0.71	<10%	0.50	0.1147	0.3	0.8	
DIMETHYLDIMETOXY SILANE	(CH ₃) ₂ Si(OCH ₃) ₂	120.2224	524.000.37-0.68	<25%	0.50	0.1067	0.2	0.7	
TRIMETHOXY SILANE	(CH ₃ O) ₃ SiH	122.1952	525.000.30-0.80	<10%	0.50	0.1331	0.8	3.5	
VINYLTRIMETHOXY SILANE	CH ₂ CHSi(OCH ₃) ₃	148.2320	553.700.32-0.80	<10%	0.50	0.1360	0.8	3.3	
METHYL SILICATE	Si(OCH ₃) ₄	152.2210	562.800.49-0.70	<3%	0.50	0.1451	0.8	2.0	
HEXAMETHYL DISILAZANE	(CH ₃) ₃ SiNHSi(CH ₃) ₃	161.3928	544.000.54-0.73	<10%	0.50	0.0860	1.0	2.6	
3-(TRIMETHOXYSILYL)-1-PROPANETHIOL	(OCH ₃) ₃ SiCH ₂ CH ₂ CH ₂ SH	196.3400	657.600.30-0.74	<10%	0.50	0.1468	0.3	1.1	
3-CHLOROPROPYL TRIMETHOXY SILANE	(CH ₃ O) ₃ Si(CH ₂) ₃ Cl	198.7200	647.300.22-0.73	<10%	0.50	0.1408	0.7	2.1	
TETRAETHOXY SILANE	Si(OCH ₂ CH ₃) ₄	208.3280	592.200.32-0.74	<5%	0.50	0.1190	0.8	2.8	
3-(TRIETHOXYSILYL) PROPIONITRILE	NCCH ₂ CH ₂ Si(OCH ₂ CH ₃) ₃	217.3380	672.100.30-0.80	<10%	0.50	0.1475	0.8	3.4	
METHYLTRIACTOXY SILANE	CH ₃ Si(OC(=O)CH ₃) ₃	220.2520	663.500.47-0.80	<10%	0.50	0.1406	0.9	3.2	
gamma-AMINOPROPYL TRIETHOXY SILANE	Si(OCH ₂ CH ₃) ₃ CH ₂ CH ₂ CH ₂ NH ₂	221.3690	634.600.22-0.78	<25%	0.50	0.1526	1.7	5.5	
ETHYLTRIACTOXY SILANE	CH ₃ CH ₂ Si(OC(=O)CH ₃) ₃	234.2790	672.100.42-0.80	<10%	0.50	0.1379	0.8	3.2	

[3-(2,3-EPOXYPROXY) PROPYL] TRIMETHOXY SILANE	(CH ₃ O) ₃ Si(CH ₂) ₃ OCH ₂ -CHCH ₂ O-	236.3380	701.400.23-0.76	<10%	0.50	0.1481	0.7	2.8
[3-(MERCAPTO) PROPYL] TRIETHOXY SILANE	Si(OCH ₂ CH ₃) ₃ CH ₂ CH ₂ CH ₂ SH	238.4200	647.500.32-0.77	<10%	0.50	0.1374	0.5	2.2
(3-GLICIDOXYPROPYL) METHYL DIETOXY SILANE	(CH ₃)Si (OCH ₂ CH ₃) ₂ CH ₂ CH ₂ CH ₂ OCH ₂ -CHCH ₂ O-	248.3910	698.700.30-0.80	<10%	0.50	0.1462	0.7	3.3
TRIS(2-METHOXY ETHOXY) VINYL SILANE	CH ₂ =CHSi (OCH ₂ CH ₂ OCH ₃) ₃	280.3900	723.900.20-0.76	<10%	0.50	0.1398	1.0	3.2
BIS[3-(TRIMETHOXY Silyl) DISULFIDE]	Si(OCH ₃) ₃ CH ₂ CH ₂ CH ₂ SSCH ₂ CH ₂ CH ₂ Si(OCH ₃) ₃	390.6640	736.200.34-0.81	<10%	0.50	0.1311	1.0	4.8
BIS[3-(TRIETHOXY Silyl) PROPYL] DISULFIDE	Si(OCH ₂ CH ₃) ₃ CH ₂ CH ₂ CH ₂ SSCH ₂ CH ₂ CH ₂ Si(OCH ₂ CH ₃) ₃	474.8230	900.300.30-0.81	<10%	0.50	0.1458	0.9	4.3

5. RESULTS AND DISCUSSION

In the discussion about the results, it is very important to outline the need of a large and accurate database containing reliable data with a clear claimed accuracy in large temperature ranges for a number of compounds as large as possible.

The DIPPR database contains in a common reduced temperature range (usually from 0.3 to 0.8) for each compound whether experimental thermal conductivity data with their sources or λ data obtained from a linear regression obtained by the available experimental data and some other predicted data (when this is possible).

Finally, without a reliable rigorous theory of the liquid phase, it has to be again pointed out that an acceptable matching has to be reached between accuracy and generality of the equations minimizing the number of parameters characterizing the single compound.

Substantially, given the very different and complicate structures of the investigated silanes and siloxanes, the obtained results are generally satisfying as follows:

- on the basis of the data collected from the DIADEM database, the use of the Eq. (5), already given for different organic families, is generalized, taking into account the value of the thermal conductivity at a specific value of the reduced temperature as shown by the expression (6) to cover silanes and siloxanes;

- the families differ from each other for the value of the exponent α (the same for the compounds of each family, with the only exception of silane characterized by small value of the molar mass M);

- factor A has a clear physical meaning expressed by Eq. (6);

- the five investigated families are characterized by very different and complex structures;

- the method can be easily extended to other families.

- the method proposed in the present paper generally leads to low average absolute deviations, being less than 5%.

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NOMENCLATURE

M	molecular weight
	kg/kmole
T	absolute temperature
	K
T _c	critical temperature
	K
T _r	reduced temperature, T/T _c

$\Delta_m\%$ average absolute deviations
 $\Delta_M\%$ maximum absolute deviations

λ thermal conductivity
W/(m K)