

PREDICTION OF VISCOSITY OF ORGANIC LIQUIDS

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Received February 17th, 1984

A method was proposed for predicting the viscosity of organic liquids which is based on known saturated vapour pressures and the chemical structure. For 199 compounds (4 144 measurements), the method gives the mean relative deviation of 3.35% and the maximum deviation of 59.0%. A lower mean value of the relative deviation than 2% was obtained for 44.3% compounds, 76% compounds possess a lower deviation than 5%.

An abundant amount of literature on the determination of viscosity of liquids has been written as well as many attempts have been made to find a correlation between the viscosity and the structure of chemical compounds. So far, theoretical works have not met with much success and, simultaneously, no theory makes it possible to calculate the viscosity of a liquid from simple relations. Due to this fact, we must often resort to empirical methods. Only those methods are of practical importance which enable to predict the viscosity of liquids in the whole temperature range from the melting point up to the boiling point temperature. The temperature appears in these relations either directly or indirectly through the specific mass or the saturated vapour pressure. A concise review of the most often employed methods is given in Table I.

Porter¹² was the first who drew attention to the relation between the viscosity and the vapour pressure, when he showed that the logarithm of the viscosity or the fluidity of mercury and water depends linearly on the logarithm of the vapour pressure. An analytic formulation of this relation can be found in the paper by Drucker¹³:

$$\log \eta = a + b \log P, \quad (1)$$

where a , b are parameters of the given liquid. According to this author, larger deviations from Eq. (1) are observed for strongly associating liquids. Eq. (1) was tested by Mitra¹⁴ and Baum¹⁵, in their theoretical considerations and derivations of correlation equations it was used by Grunberg¹⁶, Thomas^{17,18} and Mukherjee¹⁹. Mitra and Chakravarty²⁰ showed that for strongly associating liquids the parameter b can be a function of temperature and so they recommend to employ the

equation:

$$\log \eta = a + b \log P - c(\log \eta)^2, \quad (2)$$

where c is the parameter of the liquid.

Sastri and Rao¹ assumed that the parameter b in Eq. (1) is a constant for most liquids and they proposed a modified form of Eq. (1) for predicting the viscosity of liquids at temperatures lower than the normal boiling point:

$$\eta \xi = 0.0272P^{-0.2}, \quad (3)$$

with

$$\xi = T_k^{1/6}/M^{1/2}P_k^{2/3}, \quad (4)$$

where T_k , P_k is the critical temperature and pressure, resp., and M is the molar mass.

It was the aim of this work to find a method of prediction of the viscosity of organic compounds from their saturated vapour pressures and chemical structures.

TABLE I

The review of methods of prediction of the viscosity of liquids

Author	It is necessary to know:		
	compound properties	structural contributions	constants in homological series
Sastri, Rao ¹	T_c, P_c, M, P	no	no
Jossi, Stiel, Thodos ²	$T_c, P_c, M, Z_c,$ ρ, ρ_c, T	no	no ^a
Kreps, Druin ³	ρ, M, T_c, T	yes	yes
Pachaiyappan and coworkers ^{4, b}	M	no	yes
Jagannathan and coworkers ⁵	T_v, T	no	yes
Das, Kuloor ^{6 b}	T_v	no	yes
van Velzen and coworkers ⁷	T	yes	yes
Thomas ⁸	ρ, T_c, T	yes	no
Souders ⁹	ρ, M	yes	no
Orrick, Erbar ¹⁰	ρ^c, M, T	yes	no
Morris ¹¹	T_c, T	yes	no
this work	P	yes	yes

^a Only for hydrogen, ammonia, and water; ^b only at 20°C; ^c at 20°C.

TABLE II

Structural contributions and functions for the determination of parameters a , b in Eq. (1)

Functional group	Δa^a	Functional group	Δb^a
n-Alkyl:		n-alkyl:	
$n_c^b = 1$	-0.1202	$n_c = 1$	-0.1532
$n_c = 2-20$	-0.18873 - 0.01049 n_c	$n_c = 2-16$	-0.12341 + 0.0016 n_c
		$n_c = 17, 18$	-0.1045
		$n_c = 19, 20$	-0.1007
1-Alkenyl:		1-alkenyl:	
$n_c = 2-4$	-0.01532 - 0.07325 n_c	$n_c = 2$	-0.1534
$n_c = 5-7$	-0.13897 - 0.0263 n_c	$n_c = 3$	-0.1640
$n_c = 8, 9$	-0.3181	$n_c = 4-11$	-0.13755 + 0.00473 n_c
$n_c = 10-20$	-0.2508 - 0.00723 n_c	$n_c = 12-20$	-0.0912
Cyclohexyl:		cyclohexyl:	
-C ₆ H ₁₁	0.1378	-C ₆ H ₁₁	-0.1305
Cyclopentyl:		cyclopentyl:	
-C ₅ H ₉	0.1052	-C ₅ H ₉	-0.0995
Phenyl:		phenyl:	
-C ₆ H ₅	0.0432	-C ₆ H ₅	-0.0970
Hydrogen:		hydrogen:	
in n-alkanes and		in n-alkanes and	
in 1-olefins:		in 1-olefins:	
$n_c = 1-4$	-0.3060 + 0.3150	$n_c = 1$	-0.0980
	$\log n_c + 0.2908 \log^2 n_c$	$n_c = 2-10$	-0.04911 - 0.00677 n_c
$n_c = 5-20$	$(0.7709n_c - 4.7391)/$ $(4.9159n_c - 5.5997)$	$n_c = 11-20$	-0.16006 + 0.00374 n_c
in cyclohexane	0.2421	in cyclohexane	-0.2529
in cyclopentane	-0.0623	in cyclopentane	-0.1619
in benzene	0.0800	in benzene	-0.2135
in formic acid and		in formic acid and	
in formamide	0.2415	in formamide	-0.2674
Hydroxyl group		hydroxyl group	
in primary alcohols:		in primary alcohols:	
$n_c = 1-3$	0.2531 n_c - 0.0564	$n_c = 1-4$	-0.0524 n_c - 0.0728
$n_c = 4-8$	-0.0480 n_c + 0.8257	$n_c = 5-8$	0.0340 n_c - 0.4771
Carboxyl group		carboxyl group	
in acids:		in acids:	
$n_c = 1, 2$	0.2672	$n_c = 1, 2$	-0.1259
$n_c = 3-12$	0.1008 + 0.01831 n_c	$n_c = 3-18$	-0.1066
$n_c = 14-18$	0.6872 - 0.02158 n_c		
Esters ^a :		esters ^a :	
Formate, $n_c = 2-5$	-0.0533	formate, $n_c = 2-5$	-0.2323
Acetate, $n_c = 3-6$	-0.1142	acetate, $n_c = 3-6$	-0.2323

TABLE II
 (Continued)

Functional group	Δa^a	Functional group	Δb^a
Propionate, $n_c = 4-7$	-0.1481	propionate, $n_c = 4-7$	-0.2323
Butyrate and Isobutyrate		butyrate and isobutyrate	
$n_c = 5-8$	-0.1356	$n_c = 5-8$	-0.2323
Amine group ($-\text{NH}_2$) in primary amines:		amine group ($-\text{NH}_2$) in primary amines:	
$n_c = 1$	0.0761	$n_c = 1$	-0.1246
$n_c = 3-8$	$0.3766 - 0.0209n_c$	$n_c = 3-8$	-0.1782
Imine group ($=\text{NH}$) in secondary amines:		imine group ($=\text{NH}$) in secondary amines:	
$n_c = 2-14$	$0.2632 + 0.00517n_c -$ $- 0.000178n_c^2$	$n_c = 2-14$	$-0.0560 + 0.00194$ $\log n_c + 0.0437 \log^2 n_c$
Amides of carboxylic acids, group $-\text{CONH}_2$:		amides of carboxylic acids, group $-\text{CONH}_2$:	
$n_c = 1-4$	$(0.6059n_c - 0.6950)/$ $(1.1225n_c - 0.8688)$	$n_c = 1-4$	$(-1.0160 + 1.0986n_c)/$ $(0.5953 - 3.666n_c)$
Nitriles of carboxylic acids, group $-\text{CN}$:		nitriles of carboxylic acids, group $-\text{CN}$:	
$n_c = 2-6$	$(0.6163n_c - 1.804)/$ $(3.402n_c - 1.734)$	$n_c = 2-6$	$(1.6426n_c - 2.788)/$ $(14.452 - 11.791n_c)$
n-Alkyl ethers, group $-\text{O}-$:		n-alkyl ethers, group $-\text{O}-$:	
$n_c = 4$	0.1478	$n_c = 4-10$	0.0030
$n_c = 5-10$	0.2798		
n-Alkanethiols, group $-\text{SH}$:		n-alkanethiols, group $-\text{SH}$:	
$n_c = 3-8$	$0.4404 - 0.7104$ $\log n_c + 0.2662 \log^2 n_c$	$n_c = 3-8$	$-0.0959 - 0.0024n_c$
1-Bromoalkanes, group $-\text{Br}$:		1-bromoalkanes, group $-\text{Br}$:	
$n_c = 2, 3$	0.1988	$n_c = 2-10$	-0.1051
$n_c = 4-10$	0.1693		
Aromates:		aromates:	
$-\text{NH}_2$	0.1514	$-\text{NH}_2$	-0.2870
$-\text{OH}$	0.3219	$-\text{OH}$	-0.3093
$-\text{NO}_2$	-0.0921	$-\text{NO}_2$	-0.1333
$-\text{F}$	0.0185	$-\text{F}$	-0.2432
$-\text{Cl}$	-0.1194	$-\text{Cl}$	-0.1227
$-\text{Br}$	-0.0718	$-\text{Br}$	-0.1100
$-\text{J}$	-0.0346	$-\text{J}$	-0.1123

TABLE II
(Continued)

Functional group	Δa^a	Functional group	Δb^a
Position:		position:	
<i>o</i> -(1,2-)	0.1614	<i>o</i> -(1,2-)	-0.0642
<i>m</i> -(1,3-)	0.1008	<i>m</i> -(1,3-)	-0.0461
<i>p</i> -(1,4-)	0.1228	<i>p</i> -(1,4-)	-0.0526

^a For esters, values of the contributions Δa , Δb are equal directly to the parameters a , b ; ^b n_c is the total number of carbon atoms in the molecule, only for n-alkyls and 1-alkenyls it denotes the number of carbon atoms in the corresponding chain.

THEORETICAL

We started from Eq. (1) and, by the least squares method, we calculated values of parameters a , b for 199 compounds, *i.e.*, for 4 144 measurements. All compounds tested are members of homological series such as n-alkanes, 1-olefins, n-alkylcyclopentanes, n-alkylcyclohexanes, n-alkylbenzenes, 1-bromoalkanes, primary alcohols, carboxylic acids, amides, nitriles and esters of carboxylic acids, 1-aminoalkanes, dialkylamines, n-alkyl ethers, n-alkanethiols, and different aromates. The vapour pressure in kPa was calculated from the Antoine equation, for 1-olefins, some n-alkylcyclohexanes, and n-alkylbenzenes it was calculated from the Frost-Kalkwarf equation.

The parameters a , b were divided into structural contributions Δa , Δb and these were expressed in the given homological series as a function of the number of carbon atoms. The separation into the structural contributions was achieved from the condition of the minimum of the sum of squared deviations between all values of Δa , Δb employed for the correlations (depending on the number of carbon atoms) and those calculated from these equations. Values of structural contributions Δa and Δb , resp. functions necessary to compute them, are given in Table II. Parameters a , b in Eq. (1) for a given compound may then be easily calculated by adding the corresponding structural contributions Δa and Δb .

Examples of the Calculation of Viscosity

Example 1. Compute the viscosity of butyric amide at $t = 190.1^\circ\text{C}$; values of constants in the Antoine equation are equal to $A = 6.64669$, $B = 1\,847.6$, $C = 167.50$.

$$a = \Delta a(-\text{C}_3\text{H}_7) + \Delta a(-\text{CONH}_2)$$

TABLE III

The comparison of the accuracy of predicted viscosities for different methods

Method	Groups of compounds ^a														
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
Sastri, Rao¹:															
No of compounds	7	2	—	—	3	4	2	2	1	1	—	1	5	1	29
No of points	29	6	—	—	13	13	8	7	4	3	—	4	17	5	109
$\bar{\epsilon}$, %	12	22	—	—	13	49	22	7.7	12	7.3	—	7.9	17	3.7	17.5
ϵ_{\max} , %	42	30	—	—	34	80	49	13	21	11	—	14	62	6.6	80
Jossi, Stiel, Thodos²:															
No of compounds	7	2	—	—	3	4	1	2	1	—	—	1	5	1	27
No of points	19	5	—	—	12	12	4	7	4	—	—	4	15	3	85
$\bar{\epsilon}$, %	7.7	30	—	—	9.8	39	30	19	52	—	—	19	28	32	23
ϵ_{\max} , %	26	49	—	—	24	76	36	34	89	—	—	22	73	69	89
Kreps, Druin³:															
No of compounds	16	16	—	—	15	14	—	—	—	—	—	—	—	—	61
No of points	378	177	—	—	177	234	—	—	—	—	—	—	—	—	966
$\bar{\epsilon}$, %	1.78	2.0	—	—	2.4	3.5	—	—	—	—	—	—	—	—	2.3
ϵ_{\max} , %	15	6.3	—	—	20	17	—	—	—	—	—	—	—	—	20
Pachaiyappan⁴:															
No of compounds	13	13	17	14	16	7	9	15	—	—	—	—	4	5	113
No of points	13	13	17	14	16	7	9	15	—	—	—	—	4	5	113
$\bar{\epsilon}$, %	9.9	5.7	10	11	12	3.3	18	4.8	—	—	—	—	12	2.9	7.7
ϵ_{\max} , %	16	9.9	57	69	62	12	77	16	—	—	—	—	30	7.4	77
Jagannathan⁵:															
No of compounds	20	—	—	—	—	7	13	19	—	—	—	4	—	5	68
No of points	20	—	—	—	—	7	13	20	—	—	—	4	—	5	69
$\bar{\epsilon}$, %	15	—	—	—	—	6.8	24	10	—	—	—	2.0	—	2.9	13
ϵ_{\max} , %	94	—	—	—	—	12	68	47	—	—	—	4.1	—	3.9	94
Das, Kuloor⁶:															
$\bar{\epsilon}$, %	3.3	3.4	3.5	3.5	4.1	—	2.8	4.9	—	—	—	—	—	7.1	4.1
ϵ_{\max} , %	12	8.6	13	7.1	14	—	6.9	15	—	—	—	—	—	24	24
van Velzen⁷:															
No of compounds	20	19	17	17	17	8	14	28	9	4	2	5	21	10	191
No of points	898	403	405	382	562	118	121	165	70	4	5	54	304	61	3 552
$\bar{\epsilon}$, %	3.4	5.1	4.3	4.3	5.7	58	16	4.5	3.3	19	13	6.3	9.3	3.7	7.0
ϵ_{\max} , %	119	88	39	49	40	611	70	37	15	35	38	14	46	7.7	611
Thomas⁸:															
No of compounds	7	2	—	—	3	4	1	2	1	—	—	1	4	1	26
No of points	27	6	—	—	14	13	4	7	4	—	—	4	15	5	99
$\bar{\epsilon}$, %	12	20	—	—	12	66	43	5.8	13	—	—	9.9	18	5.1	21
ϵ_{\max} , %	54	27	—	—	41	89	51	19	26	—	—	16	76	15	89

TABLE III
(Continued)

Method	Groups of compounds ^a														
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
Souders ⁹ :															
No of compounds	7	2	--	--	3	4	1	2	1	--	--	1	5	1	27
No of points	27	6	--	--	14	13	4	7	4	--	--	4	19	5	103
$\bar{\epsilon}$, %	21	41	--	--	4.8	37	35	10	16	--	--	9.1	48	4.5	25
ϵ_{\max} , %	94	76	--	--	26	92	41	30	23	--	--	25	270	18	270
Orrick, Erbar ¹⁰ :															
No of compounds	7	2	--	--	3	4	1	2	1	--	--	1	4	1	26
No of points	27	6	--	--	14	13	4	7	4	--	--	4	14	5	98
$\bar{\epsilon}$, %	13	22	--	--	16	13	13	13	38	--	--	3.3	4.6	12	13.4
ϵ_{\max} , %	60	27	--	--	46	59	22	27	58	--	--	11	37	25	60
Morris ¹¹ :															
No of compounds	6	2	--	--	3	4	1	1	1	--	--	1	5	1	25
No of points	25	6	--	--	14	13	4	4	4	--	--	4	18	5	97
$\bar{\epsilon}$, %	7.9	17	--	--	8	28	4.2	8	27	--	--	10	16	6.9	13.3
ϵ_{\max} , %	49	41	--	--	22	94	6.1	24	50	--	--	22	53	14	94
This work ^b :															
No of compounds	20	19	17	17	17	8	14	20	9	7	6	6	24	--	199
No of points	778	403	411	383	516	152	211	290	70	103	74	133	503	--	4 144
$\bar{\epsilon}$, %	2.5	1.3	2.7	2.4	2.3	7.8	5.4	3.1	1.7	2.8	4.6	6.8	6.1	--	3.4
ϵ_{\max} , %	13	15	14	10	11	31	56	16	8.9	11	25	16	59	--	59

^a 1 n-Alkanes, 2 1-olefins, 3 n-alkylcyclopentanes, 4 n-alkylcyclohexanes, 5 n-alkylbenzenes, 6- primary alcohols, 7 carboxylic acids, 8 esters, 9 1-bromoalkanes, 10 1-aminoalkanes, 11 dialkylamines, 12 n-alkyl ethers, 13 aromates, 14 ketones, 15 all groups of compounds; ^b for amides of carboxylic acids (4 compounds, 35 points), this method gives $\bar{\epsilon} = 3.2\%$ and $\epsilon_{\max} = 13\%$, for nitriles of carboxylic acids (5 compounds, 48 points), it gives $\bar{\epsilon} = 1.7\%$ and $\epsilon_{\max} = 4.5\%$, for n-alkanethiols (6 compounds, 34 points), it gives $\bar{\epsilon} = 1.0\%$ and $\epsilon_{\max} = 2.7\%$.

$$\Delta a(-C_3H_7) = -0.18873 - 0.01049 \cdot 3 = -0.2202$$

$$\Delta a(-CONH_2) = (0.6059 \cdot 4 - 0.6950)/(1.1225 \cdot 4 - 0.8688) = 0.4774$$

$$a = -0.2202 + 0.4774 = 0.2572$$

$$b = \Delta b(-C_3H_7) + \Delta b(-CONH_2)$$

$$\Delta b(-C_3H_7) = -0.12341 + 0.0016 \cdot 3 = -0.1186$$

$$\Delta b(-CONH_2) = (-1.0160 + 1.0986 \cdot 4)/(0.5953 - 3.666 \cdot 4) = -0.2401$$

$$b = -0.1186 - 0.2401 = -0.3587$$

$$\log P = 6.64669 - (1\,847.6 / (190.1 + 167.50)) = 1.48002$$

$$\log \eta = 0.2572 - 0.3587 \cdot 1.48002$$

$$\eta = 0.5325 \text{ mPa s}$$

The experimental value is 0.5242 mPa s and the relative deviation is 1.58%.

Example 2. Compute the viscosity of *o*-bromotoluene at $t = 60^\circ\text{C}$. Constants in the Antoine equation are $A = 5.41883$, $B = 1\,149.51$, $C = 154.630$.

$$a = \Delta a(-\text{C}_6\text{H}_5) - \Delta a(-\text{H}) + \Delta a(-\text{CH}_3) + \Delta a(-\text{Br}) + \Delta a(1,2-)$$

$$a = 0.0432 - 0.0800 - 0.1202 - 0.0718 + 0.1614$$

$$a = -0.0674$$

$$b = \Delta b(-\text{C}_6\text{H}_5) - \Delta b(-\text{H}) + \Delta b(-\text{CH}_3) + \Delta b(-\text{Br}) + \Delta b(1,2-)$$

$$b = -0.0970 + 0.2135 - 0.1532 - 0.1100 - 0.0642$$

$$b = -0.2109$$

$$\log P = 5.41883 - (1\,149.51 / (60 + 154.630)) = 0.06305$$

$$\log \eta = -0.0674 - 0.2109 \cdot 0.06305$$

$$\eta = 0.83 \text{ mPa s}$$

The experimental value is 0.87 mPa s and the relative deviation is 4.60%.

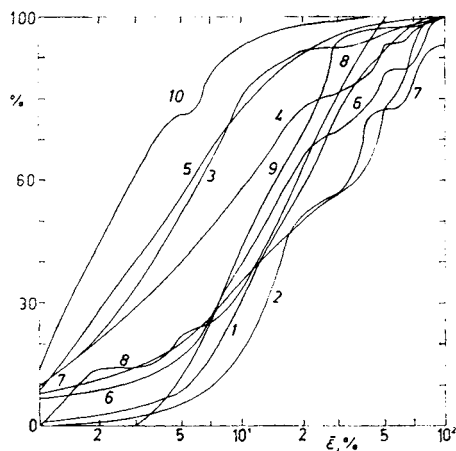


FIG. 1

The integral distribution of mean relative deviations for different methods. 1 Sastri, Rao¹ (the number of compounds for which the integral distribution was calculated — 49/ the total number of isothermal points — 171); 2 Jossi, Stiel, Thodos² (39/117); 3 Pachaiyappan⁴ (108/108); 4 Jagannathan⁵ (75/110); 5 van Velzen⁵⁴ (314/4 500); 6 Thomas⁸ (36/128); 7 Souders⁹ (39/137); 8 Orrick, Erbar¹⁰ (36/130); 9 Morris¹¹ (35/129); 10 this work (199/4 144)

DISCUSSION

In this manner we calculated viscosities of all compounds within corresponding temperature intervals, for which this method of prediction had been derived. For our calculations we employed data on the saturated vapour pressures from the literature²¹⁻²⁵. Our calculated values were compared with data on viscosities published in the literature^{8,15,25-51}. For 199 compounds (4 144 points), the method gives the mean relative deviation equal to 3.35% and the maximum deviation of 59.0%.

We compared the accuracy of our method of prediction of the viscosity of liquids with the accuracy of other methods from Table I. Results of this comparison for individual groups of compounds as well as for all compounds investigated can be found in Table III. For the methods by Souders, Thomas and Jossi, Stiel, Thodos we employed data published by Reid and Sherwood⁵². For the methods by Orrick, Erbar and Morris we employed data published by Reid, Prausnitz and Sherwood⁵³. It follows from Table III that our method of prediction of the viscosity of liquids yields for all groups of compounds investigated in almost all cases the lowest deviation.

The integral distribution of mean relative deviations for individual compounds and for different methods of prediction is illustrated in Fig. 1. It can be seen that our method yields a lower value of the mean relative deviation than 2% for 44.3% compounds and 76% compounds possess a lower value of the mean relative deviation than 5%.

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Translated by K. Hlavatý.