Application of Neural Networks to Modeling and Estimating Temperature-Dependent Liquid Viscosity of Organic Compounds

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Back-propagation neural network models for correlating and predicting the viscosity−temperature behavior of a large variety of organic liquids were developed. Experimental values for the liquid viscosity for 1229 data points from 440 compounds containing C, H, N, O, S, and all halogens have been collected from the literature. The data ranges covered are from −120 to 160 °C for temperature and from 0.164 (*trans*-2-pentene at 20 °C) to 1.34×10^5 (glycerol at −20 °C) mPa·s for viscosity value. After dividing the total database of 440 compounds into training (237 with 673 data points), validation (124 with 423 data points), and test (79 with 133 data points) sets, the modeling performance of two separate neural network models with different architectures, one based on a compound-specific temperature dependence and the second based on a compound-independent one, has been examined. The resulting former model showed somewhat better modeling performance than latter, and the model gave squared correlation coefficients of 0.956, 0.932, and 0.884 and root mean-squares errors of 0.122, 0.134, and 0.148 log units for the training, validation, and test sets, respectively. The input descriptors include molar refraction, critical temperature, molar magnetic susceptibility, cohesive energy, temperatures, and five kinds of indicator variables for functionalities, alcohols/phenols, nitriles, amines, amides, and aliphatic ring. The reliability of the proposed model was assessed by comparing the results against calculated viscosities by two existing group-contribution approaches, the method of van Velzen et al. and the Joback and Reid method.

INTRODUCTION

The viscosity of organic liquids is a measure of an internal resistance of a liquid to flow. This property is one of the significant transport properties indispensable to many scientific studies and practical applications in the fields such as chemical engineering or mechanical engineering to calculate the heat- and mass-transfer of liquids, pharmaceutics in drug formulation or administration,[1] and environmental chemistry to assess the environmental behavior of chemicals. Because of the large number of chemicals in active use, and the lack of experimentally determined data, there is an incentive and growing need of a reliable estimation method. Unfortunately, differing from the case of gas viscosity, there is so far no theory of the viscosity of liquids. In liquids, the momentum transfer is dominated by collisions and interacting force fields between the packed molecules. The theoretical description of the intermolecular forces, which consist of short-range effects such as repulsion and hydrogen bonding, wide range effects such as electrostatic effects, and long-range effects such as attractions,[2] is a very difficult task. In this context, recently, QSPR (quantitative structure−property relationships) models for liquid viscosity have been developed.[3−6]

In our previous paper, estimation schemes for liquid viscosities of 361 various organic compounds at a standard temperature of 20 °C based on the QSPR techniques using both multiple linear regression (MLR) and two-layer neural network (NN) were reported.[4] The pool of compounds was partitioned into a training set of 237 compounds, and a prediction set containing 124 compounds; the partition was made with an algorithm that assured a similar structural variety in the two subsets. The nine input descriptors were used for both MLR and NN models. The best results were found by a 9:3:1 NN with 34 adjustable parameters, compared with 10 in the MLR model. For the MLR and NN models derived from the training set compounds, squared correlation coefficients of 0.92 and 0.93 as well as root-mean-squares of 0.17 and 0.16 log units were achieved for the prediction set compounds, reflecting a reasonable accuracy for a wide range of chemical structures and viscosity values. However, on occasion, it is important to know not only the property itself at a standard temperature but also its temperature dependence. The viscosity of liquids depends strongly on temperature. Although as presented in the earlier study,[3] the Lewis-Squires chart[7] can be made use of estimating approximate viscosities at other temperatures from the viscosity value at 20 °C. The "compound-free" chart is based on the empirical fact that the sensitivity of viscosity to temperature variations appears to depend primarily upon the value of the viscosity. The chart is for only approximate and more reliable approach is needed for estimating viscosities at any temperature. The present study is concerned with the development of a new model which is capable of estimating liquid viscosity at any given temperatures with an acceptable accuracy.

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Table 1. Experimental and Calculated Viscosity Values for the Training Set Compounds

no.	compound [CAS no.]	T (°C): log η_L obsd (log η_L calcd)	ref
1	2-methylbutane [78-78-4]	0: −0.564(−0.555); 20: −0.648(−0.636); 25:-0.668(−0.654)	13−15
2	2-methylpentane [107-83-5]	0: −0.425(−0.435); 20: −0.509(−0.530); 40:-0.595(−0.609)	13, 15
3	3-methylpentane [96-14-0]	20: −0.523(−0.503); 30: −0.534(−0.546)	8, 15
4	2,3-dimethylbutane [79-29-8]	20: −0.425(−0.531); 30: −0.466(−0.573)	8, 15
5	heptane [142-82-5]	0: −0.281(−0.257); 20: −0.388(−0.371); 40: −0.467(−0.467); 70: −0.582(−0.584)	8, 13
6	2-methylhexane [591-76-4]	0: −0.318(−0.311); 20: −0.423(−0.419); 40: −0.502(−0.511)	13, 15
7	3-methylhexane [589-34-4]	20: −0.429(−0.401)	15
8	2,2,3-trimethylbutane [464-06-2]	20: −0.237(−0.443)	15
9	octane [111-65-9]	0: −0.151(−0.131); 20: −0.266(−0.257); 40:-0.364(−0.366)	13
10	2,2,3-trimethylpentane [564-02-3]	20: −0.223(−0.317)	15
11	2,2,4-trimethylpentane [540-84-1]	20: −0.299(−0.403)	15
12	nonane [111-84-2]	0: −0.007(−0.003); 20: −0.148(−0.142); 40: −0.254(−0.262); 60: −0.354(−0.365);80: −0.444(−0.452); 100: −0.523(−0.524); 130: −0.629(−0.610)	8, 13
13	dodecane [112-40-3]	20: 0.178(0.183); 40: 0.024(0.030); 60: −0.094(−0.100); 80: −0.200(−0.216); 100: −0.293(−0.314); 124: −0.391(−0.409); 152: −0.496(−0.498)	15, 17
14	1-pentene [109-67-1]	−50: −0.379(−0.347); 0: −0.620(−0.600); 20: −0.706(−0.674)	8
15	2-methyl-1-butene [563-46-2]	20: −0.650(−0.673)	8
16	1-hexene [592-41-6]	−30: −0.337(−0.300); 0: −0.499(−0.466); 20: −0.585(−0.556); 35: −0.648(−0.613); 50: −0.701(−0.663)	8, 15
17	1-heptene [592-76-7]	10: −0.418(−0.415); 20: −0.456(−0.464); 35: −0.524(−0.529); 50: −0.580(−0.586); 80: −0.676(−0.677)	8, 15
18	1-decene [872-05-9]	0: 0.032(−0.081); 20: −0.094(−0.212); 50: −0.261(−0.374); 100: −0.476(−0.565); 125: −0.564(−0.631)	8, 14
19	cyclopentene [142-29-0]	20: −0.461(−0.425)	8
20	cyclopentane [287-92-3]	−50: 0.079(0.202); 0: −0.254(−0.219); 13.5: −0.307(−0.298); 20: −0.358(−0.333); 50: −0.484(−0.462); 100: −0.658(−0.604)	8, 13, 15
21	cyclohexene [110-83-8]	20: −0.187(−0.166)	15
22	methylcyclopentane [96-37-7]	−20: −0.072(−0.128); 0: −0.199(−0.261); 20: −0.295(−0.367); 50: −0.449(−0.488); 80: −0.564(−0.575)	8, 15
23	methylcyclohexane [108-87-2]	−20: 0.142(0.145); 0: −0.011(−0.023); 20: −0.134(−0.158); 50: −0.310(−0.315); 110: −0.565(−0.512)	8, 15
24	*trans*-decalin [493-02-7]	20: 0.328(0.304)	15
25	benzene [71-43-2]	0: −0.040(−0.219); 10: −0.120(−0.279); 20: −0.186(−0.334); 30: −0.249(−0.384); 40: −0.298(−0.430); 60: −0.407(−0.510); 80: −0.483(−0.576)	13
26	toluene [108-88-3]	0: −0.112(−0.100); 20: −0.229(−0.226); 40: −0.327(−0.334); 80: −0.451(−0.499)	13
27	*o*-xylene [95-47-6]	0: 0.043(0.068); 20: −0.092(−0.075); 40: −0.203(−0.198)	13
28	*p*-xylene [106-42-3]	20: −0.188(−0.138); 40:-0.290(−0.255)	13
29	1,3,5-trimethylbenzene [108-67-8]	20: 0.062(−0.013)	15
30	propylbenzene [103-65-1]	−25: 0.260(0.351); 0: 0.066(0.135); 20: −0.068(−0.015); 50: −0.233(−0.201); 100: −0.452(−0.424); 160: −0.648(−0.583)	8, 16
31	butylbenzene [104-51-8]	−25: 0.367(0.480); 0: 0.159(0.253); 20: 0.015(0.093); 50: −0.161(−0.109); 100: −0.394(−0.353); 160: −0.604(−0.530)	8, 15
32	1-isopropyl-4-methylbenzene [99-87-6]	20: 0.532(0.052)	15
33	1-methylnaphthalene [90-12-0]	20: 0.560(0.472); 40: 0.318(0.303); 60: 0.158(0.155); 80: 0.037(0.029); 100: −0.063(−0.077)	8, 13
34	methanol [67-56-1]	−22: 0.086(0.705); 0: −0.086(0.368); 20: −0.224(0140); 30: −0.292(0.046); 50: −0.395(−0.110)	13
35	1-propanol [71-23-8]	0: 0.589(0.540); 20: 0.349(0.294); 40: 0.148(0.100); 70: −0.119(−0.119)	13, 16
36	1-butanol [71-36-3]	−51: 1.558(1.709); −22: 1.045(1.070); 0: 0.715(0.694); 20: 0.470(0.431); 50: 0.150(0.132); 100:-0.268(−0.192)	13
37	2-methyl-1-propanol [78-83-1]	20: 0.606(0.351); 30: 0.459(0.245); 50: 0.171(0.065); 90: −0.418(−0.195)	8, 15, 16
38	1-pentanol [71-41-0]	0: 0.923(0.834); 20: 0.602(0.555); 50: 0.270(0.236); 100: −0.208(−0.114); 140: −0.506(−0.286)	8, 16
39	2-methyl-2-butanol [75-85-4]	0: 1.039(0.611); 20: 0.666(0.360); 50: 0.188(0.072); 100: −0.435(−0.242)	8
40	benzyl alcohol [100-51-6]	20: 0.747(0.905); 45: 0.479(0.587); 60: 0.303(0.431)	8, 15
41	*m*-cresol [108-39-4]	10: 1.642(1.157); 20: 1.265(0.995); 40: 0.791(0.717)	13, 15
42	tetrahydrofuran [109-99-9]	20: −0.260(−0.248)	15
43	ethyl propyl ether [628-32-0]	0: −0.410(−0.399); 20: −0.491(−0.497); 40: −0.597(−0.579); 60: −0.648(−0.648)	8, 15
44	dibutyl ether [142-96-1]	20: −0.161(−0.183); 30: −0.220(−0.244)	8, 15
45	propionaldehyde [123-38-6]	10: −0.328(−0.372); 20: −0.394(−0.422); 40: −0.481(−0.510)	8, 13
46	benzaldehyde [100-52-7]	20: 0.161(0.270)	8
47	2-pentanone [107-87-9]	0: −0.199(−0.094); 20: −0.296(−0.222); 50: −0.446(−0.378) 100: −0.629(−0.561)	8, 16
48	4-methyl-2-pentanone [108-10-1]	20: −0.233(−0.183); 30: −0.282(−0.242); 45: −0.350(−0.322)	13, 15

Table 1 (Continued)

no.	compound [CAS no.]	T (°C): log η_L obsd (log η_L calcd)	ref
49	acetophenone [98-86-2]	15: 0.304(0.319); 20: 0.258(0.278); 50: 0.096(0.059); 80: −0.134(−0.114)	8, 13
50	propionic acid [79-09-4]	10: 0.110(0.155); 20: 0.042(0.079); 30: −0.020(0.009); 40: −0.073(−0.056)	13, 15
51	ethyl acetate [141-78-6]	0: −0.235(−0.225); 20: −0.342(−0.340); 50: −0.462(−0.481); 75: −0.548(−0.572)	13
52	2-methylpropyl acetate [110-19-0]	20: −0.157(−0.180); 50: −0.321(−0.347); 78: −0.437(−0.468)	8, 14, 15
53	ethyl butyrate [105-54-4]	20: −0.173(−0.146); 25: −0.213(−0.177)	14, 15
54	ethyl benzoate [93-89-0]	20: 0.350(0.362); 50: 0.105(0.129); 80: −0.092(−0.060)	8, 13
55	acetic anhydride [108-24-7]	0: 0.093(0.039); 20: −0.042(−0.101); 40: −0.156(−0.221); 100: −0.310(−0.479)	13, 15
56	acetonitrile [75-05-8]	0: −0.355(−0.274); 20: −0.426(−0.384); 30: −0.483(−0.432)	13−15
57	butylamine [109-73-9]	20: −0.167(−0.309)	15
58	diethylamine [109-89-7]	10: −0.411(−0.427); 20: −0.450(−0.480); 38: −0.564(−0.565)	8, 15
59	piperidine [110-89-4]	20: 0.172(0.093); 30: 0.088(0.007)	8, 15
60	dipropylamine [142-84-7]	20: −0.272(−0.232); 37:-0.370(−0.335)	15
61	dibutylamine [111-92-2]	20: −0.022(−0.016)	14
62	pyridine [110-86-1]	10: 0.053(0.164); 20: −0.011(0.082); 30: −0.081(0.004)	13, 15
63	*m*-toluidine [108-44-1]	20: 0.581(0.472); 30: 0.438(0.379)	13, 15
64	nitromethane [75-52-5]	20: −0.188(−0.095); 30: −0.225(−0.155)	8, 15
65	thiophene [110-02-1]	0: −0.061(0.036); 20: −0.179(−0.103); 82: −0.452(−0.386)	8, 15
66	1-chloropropane [540-54-5]	0: −0.361(−0.320); 20: −0.453(−0.426); 40: −0.536(−0.516)	13
67	1-chlorobutane [109-69-3]	20: −0.339(−0.288)	8
68	2-chloro-2-methylpropane [507-20-0]	15:-0.265(−0.416); 20:-0.291(−0.441)	8, 15
69	bromoethane [74-96-4]	−120: 0.748(0.843); −80: 0.258(0.352); 0: −0.312(−0.288); 20: −0.401(−0.398); 30: −0.458(−0.446)	13, 15
70	trichloroethylene [79-01-6]	20: −0.247(−0.165); 25: −0.274(−0.196)	15
71	iodomethane [74-88-4]	0: −0.218(−0.210); 20: −0.301(−0.328); 40: −0.373(−0.428)	13, 15
72	carbon tetrachloride [56-23-5]	0: 0.124(−0.171); 20: −0.014(−0.292); 40: −0.131(−0.395); 60: −0.233(−0.481); 100: −0.416(−0.612)	13
73	chlorobenzene [108-90-7]	20: −0.097(0.007); 40: −0.200(−0.123); 80: −0.366(−0.326); 100: −0.435(−0.403)	13
74	1,3-dichlorobenzene [541-73-1]	20: 0.036(0.289); 33: −0.020(0.186)	8, 15
75	iodobenzene [591-50-4]	20: 0.223(0.345); 149: −0.312(−0.348)	8, 13
76	morpholine [110-91-8]	20: 0.348(0.191); 30: 0.253(0.099)	14, 15
77	1,4-dioxane [123-91-1]	20: 0.079(−0.100); 30: 0.036(−0.159)	14, 15
78	2,3-dimethylpentane [565-59-3]	20: −0.391(−0.401)	15
79	3,3-dimethylhexane [563-16-6]	20: −0.301(−0.311)	8
80	3-ethylhexane [619-99-8]	20: −0.343(−0.278)	8
81	undecane [1120-21-4]	−20: 0.406(0.419); 20: 0.068(0.083); 50: −0.112(−0.123); 100: −0.364(−0.379)	8, 13
82	tridecane [629-50-5]	0: 0.431(0.460); 20: 0.275(0.286); 50: 0.055(0.054); 100: −0.220(−0.244)	8, 15
83	tetradecane [629-59-4]	20: 0.338(0.393); 40.6: 0.191(0.222); 60.1: 0.056(0.077); 99.4: −0.162(−0.166); 150.8: −0.382(−0.386)	13, 17
84	pentadecane [629-62-9]	10: 0.518(0.569); 20: 0.457(0.479); 50: 0.204(0.227); 100: −0.094(−0.110)	8, 16
85	hexadecane [544-76-3]	20: 0.524(0.553); 50: 0.268(0.295); 100: −0.038(−0.057)	8, 13
86	heptadecane [629-78-7]	20: 0.572(0.660); 50: 0.329(0.394); 100: 0.015(0.022)	8, 16
87	*trans*-3-hexene [13269-52-8]	20: −0.564(−0.527)	8
88	*trans*-2-octene [13389-42-9]	20: −0.320(−0.308)	8
89	1-nonene [124-11-8]	0: −0.094(−0.116); 20: −0.208(−0.243); 50: −0.362(−0.400);100: −0.556(−0.584)	8, 15
90	*cis*-decalin [493-01-6]	20: 0.529(0.326)	15
91	1-hexanol [111-27-3]	20: 0.702(0.676); 40: 0.444(0.440); 60: 0.218(0.247)	8
92	1-heptanol [111-70-6]	20: 0.846(0.796); 50: 0.427(0.441); 100: −0.107(0.040)	8, 15
93	1-nonanol [143-08-8]	20: 1.155(0.884)	15
94	1,2-propanediol [57-55-6]	20: 1.659(1.399); 40: 1.255(0.965); 50: 1.054(0.789); 100: 0.472(0.200)	8, 15
95	1,3-propanediol [504-63-2]	20: 1.748(1.689); 40: 1.255(1.253)	15
96	glycerol [56-81-5]	−20: 5.127(4.694); 0: 4.083(4.199); 20: 3.173(3.534); 30: 2.799(3.164); 50: 2.127(2.428)	13
97	diethylene glycol [111-46-6]	20: 1.553(1.824)	14
98	triethylene glycol [112-27-6]	20: 1.690(2.001); 60: 0.929(1.220)	15
99	propylene oxide [75-56-9]	20: −0.485(−0.447)	15
100	1,2-epoxybutane [106-88-7]	20: −0.387(−0.341); 25: −0.398(−0.366)	15
101	diisopropyl ether [108-20-3]	20: −0.467(−0.518)	8
102	ethoxybenzene [103-73-1]	0: 0.255(0.187); 20: 0.093(0.033); 30: 0.017(−0.036); 50: −0.112(−0.159); 90: −0.333(−0.350)	8, 15
103	diisopentyl ether [544-01-4]	11: 0.146(0.000); 20: 0.005(−0.063)	15
104	2-propenal [107-02-8]	20: −0.463(−0.482)	8
105	butyraldehyde [123-72-8]	20: −0.342(−0.315); 39: −0.435(−0.409)	15

Table 1 (Continued)

no.	compound [CAS no.]	T (°C): log η_L obsd (log η_L calcd)	ref
106	3-pentanone [96-22-0]	0: −0.233(−0.088); 20: −0.321(−0.216); 50: −0.474(−0.373); 100: −0.650(−0.557)	8, 14
107	cyclohexanone [108-94-1]	20: 0.347(0.117); 30: 0.256(0.047)	8, 15
108	isobutyric acid [79-31-2]	10: 0.187(0.166); 20: 0.118(0.090); 30: 0.053(0.018); 50: −0.070(−0.109)	8, 13
109	methyl formate [107-31-3]	20: −0.458(−0.480); 29: −0.496(−0.519)	15, 16
110	ethyl formate [109-94-4]	0: −0.305(−0.301); 20: −0.396(−0.407); 50: −0.532(−0.536); 70: −0.604(−0.604)	8, 13 15
111	vinyl acetate [108-05-4]	20: −0.376(−0.423)	15
112	methyl propionate [554-12-1]	0: −0.243(−0.188); 20: −0.321(−0.307); 50: −0.485(−0.452); 80: −0.599(−0.563)	8, 14
113	methyl butyrate [623-42-7]	0: −0.127(−0.077); 20: −0.237(−0.207); 40: −0.338(−0.319); 60: −0.442(−0.413); 100: −0.597(−0.557)	8, 15
114	methyl isobutyrate [547-63-7]	0: −0.182(−0.157); 20: −0.281(−0.279); 40: −0.378(−0.384); 60: −0.479(−0.472); 100: −0.625(−0.606)	8, 15
115	ethyl propionate [105-37-3]	0: −0.166(−0.107); 20: −0.270(−0.234); 50: −0.428(−0.392); 100: −0.622(−0.576)	8, 16
116	butyl acetate [123-86-4]	0: 0.002(0.040); 20: −0.135(−0.101); 50: −0.311(−0.278)	8, 13
117	1,2-ethanediol diacetate [111-55-7]	20: 0.496(0.329)	15
118	diethyl malonate [105-53-3]	20: 0.332(0.369); 25: 0.288(0.327)	15
119	dibutyl sebacate [109-43-3]	20: 0.956(1.239)	14
120	bis(2-ethylhexyl)-*o*-phthalate [117-81-7]	20: 1.910(1.467); 35: 1.527(1.334); 45: 1.330(1.244)	14, 15
121	maleic anhydride [108-31-6]	20: 0.647(0.273)	8
122	methacrylonitrile [126-98-7]	20: −0.407(−0.436)	15
123	butyronitrile [109-74-0]	20: −0.210(−0.241); 30:-0.288(−0.298)	8, 15
124	pyrrole [109-97-7]	20: 0.131(0.214); 25: 0.091(0.166)	15
125	cyclohexylamine [108-91-8]	20: 0.221(0.174); 49: 0.065(−0.061)	15
126	methylphenylamine [100-61-8]	0: 0.600(0.627); 20: 0.373(0.427); 40: 0.172(0.247); 80: −0.159(−0.055)	8
127	*N*,*N*-diethylaniline [91-66-7]	0.5: 0.584(0.461); 20: 0.338(0.285); 30: 0.061(0.200); 75: −0.125(−0.131)	13, 15
128	nitrobenzene [98-95-3]	3: 0.464(0.433); 20: 0.307(0.286); 35: 0.190(0.170)	13
129	3-nitrotoluene [99-08-1]	20: 0.367(0.302); 40: 0.204(0.149); 60: 0.072(0.018)	13
130	*N*-methylpropionamide [1187-58-2]	20: 0.782(0.618); 40: 0.551(0.445)	15
131	*N*,*N*-dimethylacetamide [127-19-5]	20: 0.331(0.655)	15
132	ethanethiol [75-08-1]	20: −0.602(−0.489)	8
133	trimethylene sulfide [287-27-4]	20: −0.195(−0.088); 25: −0.217(−0.118)	15
134	1-butanethiol [109-79-5]	20: −0.300(−0.249); 30: −0.347(−0.305)	15
135	allyl chloride [107-05-1]	0: −0.399(−0.372); 20: −0.492(−0.471); 50: −0.607(−0.589)	8
136	1-chloropentane [543-59-9]	20: −0.237(−0.226)	15
137	1,1,2-trichloroethane [79-00-5]	20: 0.044(0.102)	8
138	1-bromobutane [109-65-9]	0: −0.089(0.031); 20: −0.199(−0.111); 50: −0.332(−0.287); 100: −0.509(−0.497)	8, 15
139	2-bromobutane [78-76-2]	20: 0.157(−0.185)	15
140	1-bromo-2-methylpropane [78-77-3]	0: −0.090(−0.043); 20: −0.192(−0.178); 40: −0.286(−0.294); 60: −0.387(−0.392); 100: −0.533(−0.542)	8, 15
141	iodoethane [75-03-6]	0: −0.138(−0.088); 20: −0.228(−0.218); 40: −0.305(−0.330); 70: −0.408(−0.466)	13
142	tetrachloroethylene [127-18-4]	0: 0.057(−0.066); 20: −0.053(−0.197); 50: −0.176(−0.356); 100: −0.347(−0.542)	8
143	dibromomethane [74-95-3]	20: 0.008(0.060); 40: −0.087(−0.080)	8
144	1-iodo-2-methylpropane [513-38-2]	0: 0.055(0.148); 20: −0.058(−0.005); 40: −0.157(−0.138); 60: −0.256(−0.252); 120: −0.472(−0.495)	8, 15
145	1,2-dibromoethane [106-93-4]	0: 0.372(0.212); 20: 0.236(0.055); 40: 0.109(−0.083); 60: 0.000(−0.202); 100: −0.182(−0.389); 130: −0.294(−0.490)	8, 15
146	1,1,2.2-tetrabromoethane [79-27-6]	11: 1.145(1.226); 20: 0.991(1.124)	15
147	2-fluorotoluene [95-52-3]	20: −0.167(−0.145); 30: −0.221(−0.206)	15
148	4-fluorotoluene [352-32-9]	20: −0.206(−0.133); 30: −0.282(−0.195)	15
149	benzyl chloride [100-44-7]	20: 0.146(0.191); 25: 0.111(0.152)	15
150	dimethoxymethane [109-87-5]	15: −0.469(−0.484); 20: −0.488(−0.507)	15
151	2,4-pentanedione [123-54-6]	20: −0.222(−0.026)	14
152	trifluoroacetic acid [76-05-1]	20: −0.033(−0.265); 40: −0.185(−0.372)	15
153	4-hydroxy-4-methyl-2-pentanone [123-42-2]	20: 0.462(0.736)	14
154	2,2′-dichloroethyl ether [111-44-4]	20: 0.382(0.333); 25: 0.315(0.291)	15
155	benzoyl bromide [618-32-6]	20: 0.291(0.534); 25: 0.255(0.489)	15
156	*trans*-2-pentene [646-04-8]	20: −0.785(−0.652)	8
157	1,5-hexadiene [592-42-7]	0: −0.469(−0.528); 20: −0.561(−0.609); 36: −0.613(−0.664); 50: −0.666(−0.705)	8, 15
158	*cis*-2-hexene [7688-21-3]	20: −0.564(−0.525)	8
159	1-dodecene [112-41-4]	0: 0.269(0.231); 20: 0.114(0.073); 50: −0.080(−0.130); 100: −0.335(−0.381); 125: −0.439(−0.470)	8, 16
160	1-tridecene [2437-56-1]	0: 0.377(0.332); 20: 0.212(0.166); 50: 0.004(−0.050); 100: −0.269(−0.320); 125: −0.380(−0.417)	8, 16

Table 1 (Continued)

no.	compound [CAS no.]	T (°C): log η_L obsd (log η_L calcd)	ref
161	1-hexadecene [629-73-2]	5: 0.614(0.572); 20: 0.476(0.436); 50: 0.230(0.190); 100: −0.088(−0.137); 125: −0.217(−0.260)	8, 16
162	1-heptadecene [6765-39-5]	20: 0.556(0.537); 50: 0.296(0.281); 100: −0.032(−0.066); 125: −0.164(−0.198)	8, 16
163	1-octadecene [112-88-9]	20: 0.634(0.601); 50: 0.360(0.340); 100: 0.022(−0.020); 125: −0.116(−0.159)	8, 16
164	butylcyclopentane [2040-95-1]	−20: 0.222(0.375); 0: 0.073(0.183); 20: −0.052(0.024); 50: −0.218(−0.162); 120: −0.500(−0.430)	8, 16
165	propylcyclohexane [1678-92-8]	−25: 0.344(0.545); 0: 0.142(0.281); 20: 0.001(0.112); 50: −0.169(−0.087); 125: −0.489(−0.385)	8, 16
166	*trans*-1,3,5-trimethylcyclohexane [1839-63-0]	20: −0.146(−0.030); 30: −0.205(−0.095)	15
167	butylcyclohexane [1678-93-9]	−20: 0.443(0.675); 0: 0.270(0.451); 20: 0.117(0.264); 50: −0.070(0.041); 125: −0.418(−0.299)	8, 16
168	*n*-amylcyclohexane [4292-92-6]	−10: 0.499(0.520); 20: 0.235(0.234); 40: 0.085(0.082); 80: −0.161(−0.150); 120: −0.358(−0.307)	8, 16
169	*n*-hexylcyclohexane [4292-75-5]	−10: 0.631(0.622); 20: 0.344(0.324); 40: 0.183(0.162); 80: −0.083(−0.087); 120: −0.296(−0.256)	8, 16
170	*n*-heptylcyclohexane [5617-41-4]	−10: 0.751(0.720); 20: 0.447(0.410); 40: 0.273(0.240); 80: −0.012(−0.024); 120: −0.239(−0.207)	8, 16
171	*n*-nonylcyclopentane [2882-98-6]	−20: 0.849(0.944); 0: 0.637(0.707); 20: 0.550(0.501); 50: 0.220(0.244); 120: −0.185(−0.154)	8, 16
172	*n*-decylcyclopentane [1795-21-7]	−20: 0.953(1.032); 0: 0.730(0.792); 20: 0.550(0.581); 50: 0.293(0.314); 120: −0.133(−0.108)	8, 16
173	*n*-undecylcyclopentane [6785-23-5]	−10: 0.938(0.973); 20: 0.631(0.643); 40: 0.442(0.454); 80: 0.146(0.147); 120: −0.089(−0.073)	8, 16
174	*n*-undecylcyclohexane [54105-66-7]	10: 0.904(0.843); 20: 0.801(0.737); 40: 0.580(0.541); 80: 0.233(0.220); 120: −0.043(−0.016)	8, 16
175	*n*-tridecylcyclopentane [6006-34-4]	5: 0.935(0.983); 20: 0.781(0.818); 40: 0.577(0.618); 80: 0.254(0.283); 120: −0.003(0.034)	8, 16
176	*n*-tetradecylcyclopentane [1795-22-8]	20: 0.852(0.895); 40: 0.639(0.691); 80: 0.304(0.345); 120: 0.038(0.082)	8, 16
177	*n*-pentadecylcyclopentane [4669-01-6]	20: 0.919(0.959); 40: 0.696(0.753); 80: 0.352(0.398); 120: 0.078(0.124)	8, 16
178	*n*-hexadecylcyclopentane [6812-39-1]	20: 0.982(1.036); 40: 0.751(0.828); 80: 0.397(0.463); 120: 0.116(0.176)	8, 16
179	1-methyl-4-ethylbenzene [622-96-8]	20: −0.160(−0.013); 50: −0.306(−0.200); 90: −0.464(−0.386)	8
180	amylbenzene [538-68-1]	−20: 0.489(0.565); 0: 0.305(0.376); 20: 0.124(0.207); 50: −0.056 (−0.010); 100: −0.319(−0.279); 160: −0.554(−0.475)	8, 16
181	1-phenylhexane [1077-16-3]	−20: 0.616(0.684); 0: 0.419(0.490); 20: 0.223(0.313); 50: 0.031(0.081); 100: −0.253(−0.209); 160: −0.506(−0.425)	8, 16
182	1-phenyloctane [2189-60-8]	−20: 0.861(0.867); 0: 0.633(0.667); 20: 0.408(0.480); 50: 0.187(0.230); 100: −0.140(−0.096); 140: −0.345(−0.278)	8, 16
183	1-phenylnonane [1081-77-2]	−20: 0.976(0.979); 0: 0.735(0.776); 20: 0.496(0.584); 50: 0.262(0.324); 100: −0.085(−0.023); 140: −0.302(−0.219)	8, 16
184	1-phenylundecane [6742-54-7]	−10: 1.010(1.025); 20: 0.662(0.731); 50: 0.395(0.459); 100: 0.033(0.082); 160: −0.291(−0.224)	8, 16
185	1-phenyldodecane [123-01-3]	0: 0.955(0.995); 20: 0.736(0.800); 50: 0.452(0.524); 100: 0.084(0.134); 160: −0.246(−0.188)	8, 16
186	1-phenyltetradecane [1459-10-5]	20: 0.884(0.975); 50: 0.560(0.691); 100: 0.173(0.272); 160: −0.173(−0.091)	8, 16
187	1-phenylpentadecane [2131-18-2]	20: 0.954(1.054); 50: 0.613(0.768); 100: 0.215(0.338); 160: −0.140(−0.045)	8, 16
188	allyl alcohol [107-18-6]	0: 0.331(0.481); 20: 0.134(0.241); 40: −0.039(0.054); 70: −0.257(−0.154)	13
189	2-methyl-1-butanol [34713-94-5]	0: 1.011(0.745); 20: 0.740(0.478); 40: 0.422(0.262); 60: 0.158(0.089); 100: −0.225(−0.165); 140: −0.551(−0.331)	8, 15
190	3-ethyl-3-pentanol [597-49-9]	20: 0.829(0.752)	13
191	2-ethyl-1-hexanol [104-76-7]	20: 0.991(0.937)	15
192	eugenol [97-53-0]	0: 1.476(1.448); 20: 0.965(1.119); 40: 0.625(0.844)	13
193	2-methyl-2,4-pentanediol [107-41-5]	20: 1.536(1.539)	15
194	tripropylene glycol [1638-16-0]	20: 1.749(1.889)	14
195	tetrahydropyran [142-68-7]	20: −0.083(−0.136); 25: −0.117(−0.166)	15
196	butyl vinyl ether [111-34-2]	20: −0.301(−0.435)	15
197	dihexyl ether [112-58-3]	20: 0.232(0.226)	8
198	isobutyraldehyde [78-84-2]	20: −0.246(−0.361)	8
199	2,6-dimethyl-4-heptanone [108-83-8]	20: 0.013(0.032)	15
200	acrylic acid [79-10-7]	20: 0.114(−0.009); 25: 0.064(−0.042)	15
201	pentanoic acid [109-52-4]	20: 0.350(0.287); 30: 0.249(0.207); 50: 0.119(0.063); 100: −0.183(−0.213)	8, 15, 16
202	hexanoic acid [142-62-1]	20: 0.505(0.359); 30: 0.400(0.276); 50: 0.248(0.125); 100: −0.081(−0.167)	8, 15, 16
203	2-ethylbutyric acid [88-09-5]	20: 0.519(0.313)	15
204	octanoic acid [124-07-2]	20: 0.766(0.534); 50: 0.433(0.280); 100: 0.027(−0.049)	8, 15

Table 1 (Continued)

no.	compound [CAS no.]	T (°C): log η_L obsd (log η_L calcd)	ref
205	2-ethylhexanoic acid [149-57-5]	20: 0.886(0.480)	15
206	oleic acid [2027-47-6]	20: 1.589(1.264); 30: 1.408(1.167)	13, 14
207	methyl methacrylate [80-62-6]	20: −0.199(−0.298)	15
208	ethyl acrylate [140-88-5]	20: −0.210(−0.303)	8
209	methyl pentanoate [624-24-8]	20: −0.147(−0.123)	15
210	2-methylbutyl acetate [624-41-9]	20: −0.059(−0.053)	15
211	isoamyl acetate [123-92-2]	9: 0.013(0.020); 20: −0.059(−0.058); 25: −0.102(−0.091)	13, 15
212	ethyl pentanoate [539-82-2]	20: −0.072(0.045); 40: −0.220(−0.092); 60: −0.335(−0.210)	8, 15
213	butyl benzoate [136-60-7]	20: 0.493(0.515)	8
214	dimethyl maleate [624-48-6]	20: 0.549(0.270); 25: 0.507(0.230)	15
215	dibutyl maleate [105-76-0]	20: 0.751(0.663); 25: 0.678(0.617)	14, 15
216	diisobutyl *o*-phthalate [84-69-5]	20: 1.477(1.151)	15
217	butyl decyl *o*-phthalate [89-19-0]	20: 1.740(1.612)	15
218	4-methylpentanenitrile [542-54-1]	20: −0.009(−0.190)	15
219	isopropylamine [75-31-0]	20: −0.419(−0.495); 25: −0.444(−0.519)	8, 15
220	4-methylpyridine [108-89-4]	20: −0.045(0.322)	8
221	4-*tert*-butylpyridine [3978-81-2]	20: 0.175(0.595)	15
222	*N*-butylaniline [1126-78-9]	20: 0.536(0.567)	8
223	ethyl methyl sulfide [624-89-5]	20: −0.428(−0.389); 25: −0.451(−0.413)	15
224	tetrahydrothiophene [110-01-0]	20: 0.018(−0.062); 25: −0.013(−0.094)	15
225	1,1-dichloroethylene [75-35-4]	0: −0.355(−0.363); 20: −0.446(−0.464)	15
226	1-bromopropane [106-94-5]	0: −0.186(−0.155); 20: −0.281(−0.279); 50: −0.419(−0.430); 80: −0.523(−0.547)	8,13
227	bromochloromethane [74-97-5]	20: −0.174(−0.166)	15
228	1,2,3-trichloropropane [96-18-4]	20: 0.406(0.380)	8
229	1,1,2-trichlorotrifluoroethane [76-13-1]	20: −0.148(−0.058); 30: −0.203(−0.125)	15
230	2-methoxyethanol [109-86-4]	20: 0.236(0.452); 25: 0.204(0.394)	15
231	2-mercaptoethanol [60-24-2]	20: 0.531(0.732)	15
232	2-ethoxyethanol [110-80-5]	20: 0.312(0.494); 25: 0.267(0.435)	14, 15
233	2-hydroxybenzaldehyde [90-02-8]	20: 0.462(0.979); 45: 0.223(0.674)	8, 15
234	2,2-dimethyl-1,3-dioxolane-4-methanol [100-79-8]	20: 1.041(1.096)	15
235	bis(2-methoxyethyl)ether [111-96-6]	20: 0.299(−0.029)	15
236	*o*-phenetidine [94-70-2]	0: 1.217(0.977); 20: 0.784(0.768); 30: 0.625(0.668)	13
237	*p*-phenetidine [156-43-4]	20: 1.111(0.818); 30: 0.919(0.719)	13

The viscosities of most gases at low density increase with temperature as accounted based on theoretical expressions; however, for most liquids this trend is just reversed, for which the viscosities decrease rapidly with increasing temperature.[8] The theory for the temperature dependence of liquid viscosity, on the other hand, is not yet firmly established. This liquid behavior has been correlated by the Arrhenius type equation or the simple linear relation between logarithmic values of the liquid viscosities and reciprocal absolute temperature, which is commonly referred to as the Andrade equation.[2,9,10] Based on the Andrade equation, some group contribution methods such as Orrick and Erbar,[8] van Velzen et al.,[11] and Joback and Reid[12] have been developed. However, such methods cannot treat compounds containing sulfur and/or nitrogen, and their range of the applicability for viscosities is up to about 15 mPa·s.

In the NN modeling approach the nonlinear temperature effect could be treated with ease by employing temperature as one of the input parameters into the architecture. The accuracy of the trained NN model for temperature-dependent estimation of liquid viscosity is compared with the performance of the existing temperature-dependent viscosity estimation schemes based solely on molecular structure input.

MATERIALS AND METHODS

Data Sets. Experimental liquid viscosities or dynamic viscosities (η_L) at 20 °C of 361 organic compounds were taken from the previous works.[3,4] The viscosity data at other temperatures for the compounds were received from a collection.[8,13−17] This set is structurally sufficient diverse and includes compounds containing C, H, O, N, S, and all halogens; alkanes, alkenes, cycloalkanes, cycloalkenes, aromatic hydrocarbons, halogenated hydrocarbons, alcohols, polyhydroxy alcohols, phenols, aldehydes, ketones, acides, esters (including furanes), anhydrides, amines (including pyridines, anilines, azols, and azines), amides, nitriles, nitro and sulfur compounds, and polyfunctional compounds.

The data set, 361 compounds with 1096 data points, was split into two parts based on the previous partitioning;[4] 237 compounds with 673 data points for the training set (Table 1) and 124 compounds with 423 data points for the validation set (Table 2). The partitioning gave a balanced distribution of the chemical classes among the training and validation sets.[4] Besides an additional data of 79 compounds with 133 data points were collected from literature[13,15] and used as a test set (Table 3) for finally checking the overall performance of the neural nets.

Descriptors. Considering a broader range of structural variety of compounds in this study, neither the viscosity nor its logarithm is a constitutionally additive property. Most application of the group contribution approach has been based on additive parameters that allow viscosity estimations in combination with other experimental data such as density or vapor pressure.[10]

The following ten physicochemical and structural descriptors were used for neural network modeling: (1) molar refraction at 20 °C (*MR*, 10^{-6} m^3 mol^{-1}); (2) critical temperature (*Tc*, K); (3) absolute value of molar magnetic susceptibility (χ_m, 10^{-12} m^3 mol^{-1}); (4) cohesive energy (energy of vaporization) at 298 K (E_{coh}, kJ mol^{-1}); indicator

Table 2. Experimental and Calculated Viscosity Values for the Validation Set Compounds

no.	compound [CAS no.]	T (°C): log η_L obsd (log η_L calcd)	ref
1	pentane [109-66-0]	0: −0.539(−0519); 15: −0.625(−0.584); 20: −0.620(−0.604); 25: −0.668(−0.623)	13, 15
2	hexane [110-54-3]	0: −0.397(−0.384); 20: −0.487(−0.484); 25: −0.532(−0.507); 40: −0.567(−0.568); 50: −0.606(−0.605)	13
3	2, 2-dimethylbutane [75-83-2]	20: −0.438(−0.568); 25: −0.455(−0.588); 30: −0.481(−0.607)	8, 15
4	2,4-dimethylpentane [108-08-7]	20: −0.442(−0.462)	15
5	decane [124-18-5]	20: −0.032(−0.031); 39: −0.155(−0.156); 60.1: −0.264(−0.276); 78.9: −0.351(−0.367); 101.1: −0.443(−0.457); 123.3: −0.527(−0.530); 150: −0.621(−0.599)	15, 17
6	1-octene [111-66-0]	0: −0.229(−0.234); 20: −0.328(−0.349); 35: −0.403(−0.424); 50: −0.466(−0.489); 100: −0.640(−0.652); 125: −0.710(−0.708)	8, 15
7	cyclohexane [110-82-7]	15.5: 0.024(−0.179); 20: −0.009(−0.205); 25.2: −0.051(−0.235); 35.9: −0.125(−0.290); 50.6: −0.219(−0.356); 60.7: −0.278(−0.396)	15, 17
8	ethylcyclohexane [1678-91-7]	−25: 0.237(0.388); 0: 0.050(0.145); 20: −0.074(−0.009); 50: −0.237(−0.189); 100:-0.446(−0.389); 125: −0.532(−0.456)	8, 15
9	styrene [100-42-5]	−30: 0.221(0.247); 0: 0.005(0.001); 20: −0.124(−0.134); 25: −0.157(−0.165); 50: −0.274(−0.298); 100: −0.488(−0.488)	8, 15
10	ethylbenzene [100-41-4]	20: −0.169(−0.122); 40: −0.275(−0.239)	14, 15
11	*m*-xylene [108-38-3]	0: −0.094(0.006); 15: −0.187(−0.098); 20: −0.208(−0.131); 40: −0.304(−0.248)	13
12	1,2,4-trimethylbenzene [95-63-6]	20: 0.048(0.035)	8
13	isopropylbenzene [98-82-8]	−20: 0.200(0.252); 0: 0.038(0.086); 20: −0.102(−0.059); 50: −0.264(−0.240); 100: −0.471(−0.455); 150: −0.622(−0.588)	8, 15
14	tetralin [119-64-2]	20: 0.343(0.349); 30: 0.255(0.266); 40: 0.176(0.188); 60: 0.045(0.049); 80: −0.066(−0.069); 100: −0.167(−0.169)	13, 15
15	*sec*-butylbenzene [135-98-8]	20: 0.019(0.092)	8
16	ethanol [64-17-5]	−59.4: 0.925(1.543); −32.0: 0.584(0.911); −17.6: 0.428(0.656); 0: 0.249(0.401); 10: 0.166(0.279); 20: 0.079(0.172); 30: 0.001(0.077); 50: −0.154(−0.082); 60: −0.228(−0.148); 70: −0.298(−0.208)	13
17	2-butanol [78-92-2]	15: 0.624(0.361); 20: 0.592(0.305); 100: −0.278(−0.272)	13, 15
18	1-octanol [111-87-5]	15: 1.025(1.019); 20: 0.951(0.946); 30: 0.787(0.809); 50: 0.514(0.570); 100: −0.031(0.137)	8, 14, 15
19	ethylene glycol [107-21-1]	20: 1.299(1.609); 40: 0.960(1.176); 60: 0.695(0.838); 80: 0.480(0.576); 100: 0.299(0.374)	13
20	furan [110-00-9]	20: −0.420(−0.431); 25: −0.442(−0.453)	15
21	diethyl ether [60-29-7]	−100: 0.228(0.160); −60: −0.196(−0.180); −20: −0.441(−0.431); 0: −0.546(−0.530); 20: −0.611(−0.613); 40: −0.706(−0.682); 60: −0.780(−0.739); 80: −0.851(−0.785); 100: −0.928(−0.823)	13, 14
22	butyl ethyl ether [628-81-9]	20: −0.376(−0.389); 25: −0.401(−0.415)	15
23	acetaldehyde [75-07-0]	0: −0.553(−0.451); 10: −0.592(−0.498); 20: −0.613(−0.540)	13, 14
24	acetone [67-64-1]	−92.5: 0.332(0.429); −59.6: −0.031(0.101); −30: −0.240(−0.134); −13: −0.328(−0.248); 0: −0.399(−0.324); 20: −0.498(−0.427); 30: −0.530(−0.473); 41: −0.553(−0.518)	13, 15
25	2-butanone [78-93-3]	0: −0.278(−0.215); 20: −0.369(−0.330); 40: −0.457(−0.427); 60: −0.558(−0.508)	8, 15
26	acetic acid [64-19-7]	15: 0.117(0.016); 20: 0.091(−0.019); 30: 0.017(−0.084); 59: −0.155(−0.245); 100: −0.367(−0.411)	13, 15
27	butyric acid [107-92-6]	0: 0.359(0.328); 20: 0.188(0.163); 40: 0.049(0.020); 70: −0.119(−0.158); 100: −0.259(−0.295)	13
28	methyl acetate [79-20-9]	0: −0.315(−0.307); 20: −0.419(−0.413); 40: −0.495(−0.502); 70: −0.633(−0.608)	8, 13
29	propyl acetate [109-60-4]	10: −0.180(−0.225); 20: −0.233(−0.283); 40: −0.357(−0.387)	13, 15
30	acrylonitrile [107-13-1]	20: −0.456(−0.502); 25: −0.469(−0.523)	14, 15
31	propionitrile [107-12-0]	15: −0.343(−0.292); 20: −0.372(−0.320); 30: −0.410(−0.372)	8, 15
32	aniline [62-53-3]	−6: 1.140(0.671); 5: 0.906(0.555); 20: 0.643(0.408); 30: 0.500(0.316); 50: 0.267(0.147); 70: 0.104(−0.001); 100: −0.084(−0.187)	13
33	dimethyl sulfide [75-18-3]	20: −0.539(−0.493); 36: −0.577(−0.559)	15
34	diethyl sulfide [352-93-2]	0: −0.249(−0.172); 20: −0.351(−0.293); 25: −0.375(−0.320); 50: −0.474(−0.440); 80: −0.578(−0.553)	8, 15
35	dichloromethane [75-09-2]	15: −0.348(−0.343); 20: −0.371(−0.369); 30: −0.406(−0.419)	8, 13
36	chloroform [67-66-3]	−13: −0.068(−0.102); 0: −0.155(−0.190); 8.1: −0.192(−0.241); 20: −0.237(−0.310); 30: −0.289(−0.363); 39: −0.301(−0.407)	13
37	fluorobenzene [426-06-6]	15: −0.208(−0.230); 20: −0.223(−0.259); 40: −0.321(−0.364); 60: −0.410(−0.452); 80: −0.483(−0.525); 100: −0.561(−0.585)	13, 15
38	bromobenzene [108-86-1]	0: 0.180(0.349); 15: 0.078(0.221); 20: 0.054(0.181); 50: −0.108(−0.030); 100: −0.319(−0.285)	8, 13
39	1, 2-ethanediamine [107-15-3]	20: 0.188(0.174); 30: 0.088(0.094)	15
40	2, 2-dimethylpentane [590-35-2]	20: −0.420(−0.469)	8
41	2-methylheptane [592-27-8]	20: −0.283(−0.313)	8
42	2-methyl-1,3-butadiene [78-79-5]	0.3: −0.575(−0.634); 20: −0.652(−0.701)	15
43	*cis*-3-hexene [7642-09-3]	20: −0.564(−0.533)	8
44	3-methyl-1-butanol [123-51-3]	10: 0.792(0.625); 15: 0.682(0.559); 20: 0.708(0.496); 30: 0.471(0.381)	13, 15, 16
45	cyclohexanol [108-93-0]	20: 1.833(0.987); 30: 1.614(0.807); 45: 1.235(0.581)	13, 15
46	1-decanol [112-30-1]	20: 1.158(1.158)	8
47	1,5-pentanediol [111-29-5]	20: 2.107(1.803)	15

Table 2 (Continued)

no.	compound [CAS no.]	T (°C): log η_L obsd (log η_L calcd)	ref
48	ethyl vinyl ether [109-92-2]	20: −0.699(−0.642)	14
49	methoxybenzene [100-66-3]	0: 0.250(0.140); 20: 0.121(−0.010); 40: 0049(−0.138); 70: −0.060(−0.294)	8, 13
50	pentanal [110-62-3]	20: −0.290(−0.198)	8
51	cyclopentanone [120-92-3]	20: 0.067(0.093)	8
52	4-heptanone [123-19-3]	20: −0.133(−0.043); 25: −0.161(−0.076)	8, 15
53	methacrylic acid [79-41-4]	20: 0.121(0.129)	15
54	propyl formate [110-74-7]	0: −0.171(−0.171); 20: −0.241(−0.291); 40: −0.380(−0.394): 50: −0.439(−0.439); 80: −0.558(−0.551)	8, 15
55	2-methylpropyl formate [542-55-2]	20: −0.167(−0.249)	15
56	propyl propionate [106-36-5]	20: −0.172(−0.101)	15
57	pentyl acetate [628-63-7]	11: 0.199(0.050); 20: −0.034(−0.015); 25: −0.064(−0.050); 45: −0.094(−0.176)	13, 15
58	methyl benzoate [93-58-3]	15: 0.361(0.349); 20: 0.310(0.307); 30: 0.223(0.227)	8, 15
59	ethyl cinnamate [103-36-6]	20: 0.940(0.363)	15
60	dibutyl *o*-phthalate [84-74-2]	20: 1.299(1.072); 45: 0.895(0.831)	15
61	*trans*-3-butenenitrile [109-75-1]	20: −0.289(−0.324)	8
62	triethylamine [121-44-8]	15: −0.405(−0.338); 20: −0.444(−0.367); 30: −0.440(−0.422)	8, 15
63	tributylamine [102-82-9]	20: 0.189(0.251); 25: 0.130(0.211)	8, 15
64	2,4,6-trimethylpyridine [108-75-8]	20: 0.176(0.504)	15
65	*o*-toluidine [95-53-4]	0: 0.929(0.625); 15: 0.716(0.476); 20: 0.642(0.429); 50: 0.314(0.168); 100: −0.135(−0.173)	8, 13,15
66	*N*,*N*-dimethylaniline [121-69-7]	10: 0.228(0.360); 20: 0.149(0.269); 30: 0.068(0.183); 40: 0.017(0.101); 50: −0.041(0.024)	13
67	nitroethane [79-24-3]	20: −0.169(−0.021); 35: −0.201(−0.116)	15
68	2-nitrotoluene [88-72-2]	0: 0.583(0.427); 20: 0.375(0.256); 40: 0.212(0.107); 60: 0.083(−0.020)	13
69	thiophenol [108-98-5]	20: 0.093(0.113); 25: 0.058(0.077)	15
70	2-chloropropane [75-29-6]	0: −0.321(−0.413); 20: −0.398(−0.510); 30: −0.524(−0.552); 40: −0.465(−0.570)	8, 15
71	2-chlorobutane [78-86-4]	15: −0.358(−0.351); 20: −0.385(−0.377)	8, 15
72	1-chloro-2-methylpropane [513-36-0]	0: −0.240(−0.235); 20: −0.335(−0.351); 40: −0.428(−0.449); 60: −0.529(−0.531); 80: −0.604(−0.599)	8, 15
73	*cis*-1,2-dichloroethylene [156-59-2]	20: −0.331(−0.362); 25: −0.353(−0.387)	15
74	1,1-dichloroethane [75-34-3]	10: −0.268(−0.273); 20: −0.318(−0.329); 30: −0.367(−0.381); 40: −0.408(−0.429); 60: −0.487(−0.513)	8, 14
75	1,2-dichloropropane [78-87-5]	20: −0.063(−0.061); 25: −0.155(−0.095)	15
76	1,2-dichlorobenzene [95-50-1]	20: 0.155(0.423); 25: 0.122(0.379)	8, 15
77	pentachloroethane [76-01-7]	15: 0.438(0.323); 20: 0.389(0.281); 30: 0.316(0.200)	14, 15
78	3-fluorotoluene [352-70-5]	20: −0.216(−0.134); 30: −0.272(−0.195)	15
79	ethanolamine [141-43-5]	20: 1.367(1.161)	8
80	2,2,2-trifluoroethanol [75-89-8]	20: 0.300(0.224)	15
81	1,2-diethoxyethane [629-14-1]	20: −0.187(−0.258)	15
82	3-bromoaniline [591-19-5]	20: 0.833(1.043); 40: 0.568(0.830); 80: 0.230(0.449)	13
83	*cis*-2-pentene [627-20-3]	20: −0.695(−0.654)	8
84	2-methyl-2-butene [513-35-9]	0: −0.607(−0.585); 10: −0.648(−0.625); 20: −0.686(−0.661) 30: −0.724(−0.694); 40: −0.757(−0.724)	8
85	*trans*-2-hexene [4050-45-7]	20: −0.564(−0.535)	8
86	1-undecene [821-95-4]	0: 0.153(0.114); 20: 0.013(−0.035); 25: −0.018(−0.069); 50: −0.168(−0.223); 100: −0.402(−0.451); 125: −0.498(−0.531)	8, 16
87	1-tetradecene [1120-39-1]	0: 0.478(0.440); 20: 0.301(0.267); 25: 0.262(0.226); 50: 0.083(0.039); 100: −0.206(−0.253); 125: −0.323(−0.359)	8, 16
88	1-pentadecene [13360-61-7]	0: 0.579(0.517); 20: 0.391(0.340); 25: 0.350(0.298); 50: 0.160(0.104); 100: −0.146(−0.203); 125: −0.271(−0.317)	8, 16
89	ethylcyclopentane [1640-89-7]	−20: −0.024(0.146); 0: −0.149(−0.021); 20: −0.248(−0.157); 25: −0.276(−0.187); 50: −0.395(−0.315); 110: −0.606(−0.513)	8, 16
90	propylcyclopentane [2040-96-2]	−20: 0.075(0.317); 0: −0.057(0.130); 20: −0.167(−0.022); 25: −0.194(−0.056); 50: −0.313(−0.201); 120: −0.564(−0.451)	8, 16
91	*n*-amylcyclopentane [3741-00-2]	−20: 0.362(0.494); 0: 0.199(0.291); 20: 0.061(0.121); 25: 0.025(0.083); 50: −0.121(−0.081); 120: −0.433(−0.374)	8, 16
92	*n*-hexylcyclopentane [4457-00-5]	−20: 0.501(0.641); 0: 0.323(0.424); 20: 0.274(0.241); 25: 0.134(0.200); 50: −0.027(0.021); 120: −0.368(−0.303)	8, 16
93	*n*-heptylcyclopentane [5617-42-5]	−20: 0.621(0.752); 0: 0.432(0.526); 20: 0.373(0.334); 25: 0.236(0.291); 50: 0.061(0.100); 120: −0.300(−0.250)	8, 16
94	*n*-octylcyclopentane [1795-20-6]	−20: 0.738(0.847); 0: 0.537(0.615); 20: 0.464(0.416); 25: 0.332(0.371); 50: 0.143(0.171); 120: −0.240(−0.203)	8, 16
95	*n*-octylcyclohexane [1795-15-9]	−10: 0.869(0.810); 20: 0.544(0.492); 25: 0.491(0.445); 40: 0.359(0.314); 80: 0.055(0.035); 120: −0.187(−0.160)	8, 16
96	*n*-nonylcyclohexane [2883-02-5]	−10: 0.977(0.901); 20: 0.634(0.575); 25: 0.580(0.526); 40: 0.439(0.391); 80: 0.118(0.097); 120: −0.138(−0.112)	8, 16
97	*n*-decylcyclohexane [1795-16-0]	0: 0.945(0.920); 20: 0.719(0.700); 25: 0.663(0.650); 40: 0.512(0.506); 80: 0.177(0.192); 120: −0.090(0.035)	8, 16
98	*n*-dodecylcyclopentane [5634-30-0]	−5: 0.969(1.020); 20: 0.708(0.743); 25: 0.659(0.691); 40: 0.512(0.547); 80: 0.203(0.224); 120: −0.043(−0.012)	8, 16
99	*n*-dodecylcyclohexane [1795-17-1]	15: 0.919(0.869); 20: 0.876(0.816); 25: 0.813(0.764); 40: 0.644(0.616); 80: 0.285(0.281); 120: −0.001(0.032)	8, 16

Table 2 (Continued)

no.	compound [CAS no.]	T (°C): log η_L obsd (log η_L calcd)	ref
100	*n*-tridecylcyclohexane [6006-33-3]	20: 0.949(0.894); 25: 0.885(0.841); 40: 0.705(0.690); 80: 0.334(0.344); 120: 0.039(0.082)	8, 16
101	α-methylstyrene [98-83-9]	20: −0.099(−0.071)	8
102	1-phenylheptane [1078-71-3]	−20: 0.735(0.712); 0: 0.525(0.518); 20: 0.316(0.340); 50: 0.113(0.105); 100: −0.188(−0.194); 160: −0.457(−0.420)	8, 16
103	1-phenyldecane [104-72-3]	−15: 0.990(0.984); 0: 0.810(0.834); 20: 0.579(0.641); 50: 0.332(0.376); 100: −0.018(0.016); 160: −0.332(−0.269)	8, 16
104	1-phenyltridecane [123-02-4]	5: 0.967(1.121); 20: 0.812(0.975); 25: 0.751(0.926); 50: 0.509(0.690); 100: 0.129(0.271); 160: −0.210(−0.090)	8, 16
105	2-propyn-1-ol [107-19-7]	20: 0.225(0.457)	15
106	1, 3-butanediol [107-88-0]	20: 2.115(1.617); 25: 1.949(1.505)	15
107	dibenzyl ether [103-50-4]	0: 1.021(0.930); 20: 0.727(0.728); 25: 0.569(0.680); 40: 0.507(0.542)	13, 15
108	3-methylbutyric acid [503-74-2]	15: 0.436(0.272); 20: 0.382(0.231)	15
109	heptanoic acid [111-14-8]	20: 0.639(0.457); 30: 0.531(0.370); 50: 0.359(0.211); 100: 0.003(−0.101)	8, 16
110	nonanoic acid [112-05-0]	20: 0.920(0.680); 50: 0.572(0.412); 100: 0.146(0.056)	8, 16
111	vinyl formate [692-45-5]	20: −0.444(−0.563)	8
112	isopropyl acetate [108-21-4]	20: −0.245(−0.297)	15
113	propyl butyrate [105-66-8]	20: −0.080(0.067); 30: −0.151(−0.005); 40: −0.210(−0.071); 60: −0.318(−0.190)	8, 15
114	propyl isobutyrate [644-49-5]	20: −0.080(0.012)	15
115	2-ethylhexyl acetate [103-09-3]	20: 0.176(0.214)	15
116	diethyl maleate [141-05-9]	20: 0.553(0.381); 25: 0.497(0.339)	15
117	pentylamine [110-58-7]	20: 0.008(−0.189)	15
118	2-methylpyridine [109-06-8]	20: −0.094(0.220); 30: −0.149(0.138)	15
119	*trans*-1, 2-dichloroethylene [156-60-5]	15: −0.374(−0.350); 20: −0.394(−0.376)	15
120	methyl cyanoacetate [105-34-0]	20: 0.446(0.296)	14
121	methyl acetoacetate [105-45-3]	20: 0.231(0.190)	15
122	tetrahydropyran-2-methanol [100-72-1]	20: 1.041(1.181)	15
123	2, 2′-thiodiethanol [111-48-8]	20: 1.814(2.093)	15
124	1, 2-bis(methoxyethoxy)ethane [112-49-2]	20: 0.575(0.240)	15

variables for (5) alcohols/phenols (I_{OH}), (6) nitriles (I_{CN}), (7) amines (I_{amine}), (8) amides (I_{amide}), and (9) aliphatic ring structures (including O-, N-, and S-containing heterocycles) (I_{ring}); and (10) temperature (T, °C). For the indicator variables, values of 1 or 0 are assigned for the presence or absence of the relevant functional group except for polyols, where I_{OH} was set to 1.5 and 2 for dihydroxy and trihydroxy alcohols, respectively. The first nine descriptors have been used in our previous studies to predict liquid viscosity of organic compounds at 20 °C.[3,4] The four property descriptors, *MR*, *Tc*, χ_m, and E_{coh}, were selected from a set of 18 property descriptors which had been used as several empirical equations giving a functional relationship between viscosity and some other physical properties.[3] The properties discarded in the model derivation process using PLS were as follows: molecular weight; melting point; boiling point; density; molar volume; dipole moment; critical pressure; acentric factor; enthalpy of vaporization at the boiling point; logarithm of the vapor pressure at 20 °C; logarithm of the partition coefficient in the 1-octanol/water system at 25 °C; surface tension; cohesive energy density; and solubility parameter. The additional indicator variables, I_{OH}, I_{CN}, I_{amine}, I_{amide}, and I_{ring}, were introduced to explain extra contributions. The descriptors I_{OH}, I_{amine}, and I_{amide} could be due to the effect of hydrogen bonding and the assignment of the values of I_{OH} for diols and triols were performed empirically (the observed contributions by OH groups are less than that calculated simple additivity of OH contributions).

If the experimental physicochemical descriptors, *MR*, T_c, χ_m, or E_{coh}, are not known, the appropriate estimation schemes[8,18,19] can be used for their calculations with reasonable accuracy. The ranges of values for above descriptors and liquid viscosities for the training set compounds are presented in Table 4.

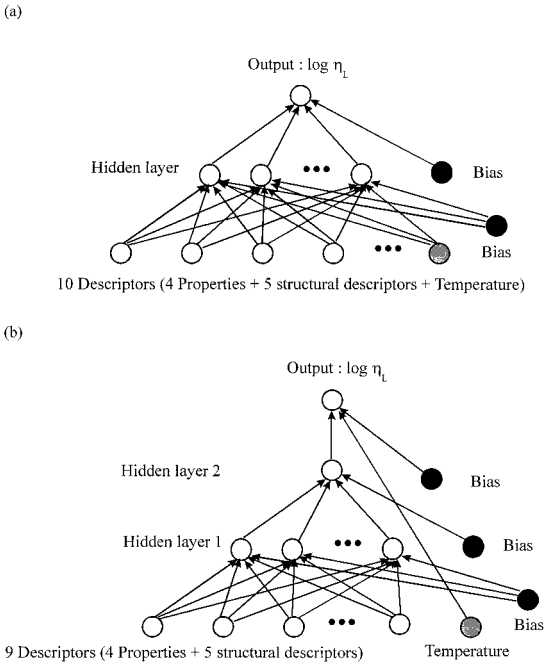


Figure 1. Two architectures of neural network model A (a) full connected two layers model of (9+T+1):variable hidden neurons + 1: 1 and B (b) pruned three layers model of (9+1):variable first hidden neurons +1: 1+T+1 second hidden neurons: 1 model, for temperature-dependent viscosity estimation.

Neural Network Calculations. Figure 1 illustrates the two architectures of neural networks, A and B, examined in the present study. The network A is composed of fully connected

Table 3. Experimental and Calculated Viscosity Values for the Testing Set Compounds

no.	compound [CAS no.]	T (°C): log η_L obsd (log η_L calcd)	ref
1	cycloheptane [291-64-5]	13.5: 0.215(0.065)	13
2	cyclohexylbenzene [827-52-1]	0: 0.566(0.682)	15
3	2-propanol [67-63-0]	15: 0.456(0.204); 30: 0.247(0.060)	15
4	2-pentanol [6032-29-7]	15: 0.710(0.475); 30: 0.444(0.308)	15
5	3-pentanol [584-02-1]	15: 0.866(0.419); 30: 0.519(0.257)	15
6	3-methyl-2-butanol [598-75-4]	25: 0.545(0.280)	15
7	2-ethyl-1-butanol [97-95-0]	15: 0.904(0.598); 25: 0.770(0.477)	15
8	4-methyl-2-pentanol [108-11-2]	25: 0.610(0.391)	15
9	*cis*-2-methylcyclohexanol [7443-70-1]	25: 1.257(0.829); 30: 1.134(0.747)	15
10	3,5,5-trimethyl-1-hexanol [3452-97-9]	25: 1.044(0.743)	15
11	phenol [108-95-2]	50: 0.543(0.551); 60: 0.417(0.447); 70: 0.307(0.354) 90: 0.100(0.196)	13
12	tri(propylene glycol) [24800-44-0]	25: 1.749(1.789)	15
13	propyl ether [111-43-3]	15: −0.349(−0.366); 30: −0.425(−0.441)	15
14	pentyl ether [693-65-2]	15: 0.075(0.062); 30: −0.035(−0.046)	15
15	heptaldehyde [111-71-7]	15: −0.010(0.037)	15
16	2-hexanone [591-78-6]	25: −0.234(−0.136)	15
17	4-methyl-3-penten-2-one [141-79-7]	25: −0.056(−0.177)	15
18	2-heptanone [110-43-0]	15: −0.069(0.171); 30: −0.164(0.057)	15
19	2-undecanone [112-12-9]	30: 0.207(0.287)	15
20	formic acid [64-18-6]	10: 0.354(0.210); 20: 0.256(0.133); 30: 0.166(0.062); 40: 0.086(−0.005); 70: −0.108(−0.176)	13
21	isopropyl formate [625-55-8]	20: −0.291(−0.346)	15
22	allyl acetate [591-87-7]	30: −0.684(−0.328)	15
23	butyl formate [592-84-7]	0: −0.027(−0.116); 20: −0.162(−0.242)	13
24	butyl butyrate [109-21-7]	25: −0.076(−0.004)	15
25	benzyl acetate [140-11-4]	45: 0.146(0.036)	15
26	diethyl carbonate [105-58-8]	15: −0.061(−0.169); 25: −0.126(−0.230)	15
27	diethyl oxalate [95-92-1]	15: 0.364(0.349); 30: 0.209(0.226)	15
28	propionic anhydride [123-62-6]	20: 0.058(0.062); 25: 0.026(0.027)	15
29	butyric anhydride [106-31-0]	20: 0.208(0.213); 25: 0.172(0.174)	15
30	isobutyronitrile [78-82-0]	15: −0.259(−0.301); 30: −0.341(−0.381)	15
31	pentanenitrile [110-59-8]	15: −0.108(−0.108); 30: −0.196(−0.217)	15
32	hexanenitrile [628-73-9]	15: 0.017(−0.052); 30: −0.081(−0.151)	15
33	benzonitrile [100-47-0]	25: 0.093(0.172)	13
34	phenylacetonitrile [140-29-4]	25: 0.286(0.354)	15
35	propylamine [107-10-8]	25: −0.465(−0.414)	15
36	isobutylamine [78-81-9]	25: −0.257(−0.388)	13
37	isoamylamine [107-85-7]	25: −0.140(−0.245)	13
38	diisopropylamine [108-18-9]	25: −0.398(−0.399)	15
39	*N*-methylaniline [100-61-8]	25: 0.305(0.413); 30: 0.190(0.367); 55: 0.035(0.153)	13, 15
40	*N*-ethylaniline [103-69-5]	25: 0.310(0.355); 55: 0.033(0.104)	15
41	1-nitropropane [108-03-2]	25: −0.098(−0.005); 35: −0.155(−0.070)	15
42	2-nitropropane [79-46-9]	25: −0.125(−0.058)	15
43	dimethyl sulfoxide [67-68-5]	20: 0.393(0.354); 55: 0.076(0.097)	15
44	bromoform [75-25-2]	15: 0.333(0.487); 25: 0.276(0.397)	13
45	diiodomethane [75-11-6]	15: 0.483(0.662); 30: 0.379(0.516)	15
46	1,1,1-trichloroethane [71-55-6]	15: −0.044(−0.203); 30: −0.140(−0.291)	15
47	1,1,2,2-tetrachloroethane [79-34-5]	15: 0.266(0.422); 30: 0.163(0.292)	15
48	2-bromopropane [75-26-3]	15: −0.271(−0.205); 30: −0.360(−0.294)	15
49	1-iodopropane [107-08-4]	15: −0.077(−0.005); 30: −0.174(−0.108)	15
50	2-iodopropane [75-30-9]	15: −0.135(−0.138); 30: −0.208(−0.231)	15
51	1,2-dibromopropane [78-75-1]	25: 0.176(0.128)	15
52	2,2-dichloropropane [594-20-7]	15: −0.114(−0.227); −0.208(−0.313)	15
53	2-chloro-2-methylpropane [507-20-0]	15: −0.265(−0.414)	13
54	1-bromopentane [110-53-2]	25: −0.121(−0.182); 50: −0.240(−0.323); 75: −0.331(−0.438); 100: −0.423(−0.529)	13
55	cyclohexyl bromide [108-85-0]	25: 0.297(0.294); 50: 0.130(0.106); 75: −0.015(−0.041)	13
56	cyclohexyl chloride [542-18-7]	25: 0.194(0.131); 50: 0.019(−0.036); 75: −0.126(−0.165)	13
57	2-bromotoluene [95-46-5]	25: 0.114(0.337)	15
58	benzotrichloride [98-07-7]	10: 0.487(0.759); 17: 0.407(0.689)	15
59	1-bromooctane [111-83-1]	25: 0.174(0.275); 50: 0.012(0.082); 75: −0.122(−0.080)	13
60	1-bromonaphthalene [90-11-9]	15: 0.777(0.906); 40: 0.505(0.658)	15
61	1-chloronaphthalene [90-13-1]	25: 0.468(0.628)	15
62	1-bromododecane [143-15-7]	25: 0.526(0.616); 50: 0.310(0.394)	13
63	2-chloroethanol [107-07-3]	15: 0.593(0.684)	15
64	2-aminoethanol [141-43-5]	15: 1.489(1.405); 25: 1.287(1.207)	15
65	ethylenediamine [107-15-3]	18: 0.188(0.185)	15
66	lactonitrile [78-97-7]	30: 0.303(0.605)	15
67	1,2-dimethoxyethane [110-71-4]	10: −0.276(−0.344); 25: −0.342(−0.421)	15
68	2-(ethylamino)ethanol [110-73-6]	25: 1.093(1.029)	15
69	2-furaldehyde [98-01-1]	0: 0.394(0.311); 25: 0.174(0.113)	15
70	furfuryl alcohol [98-00-0]	25: 0.665(0.575)	15
71	ethyl cyanoacetate [105-56-6]	15: 0.513(0.331); 30: 0.332(0.210)	15
72	1-ethoxy-2-propanol [1569-02-4]	25: 0.225(0.528)	15
73	ethyl lactate [687-47-8]	25: 0.387(0.630)	15

Table 3 (Continued)

no.	compound [CAS no.]	T (°C): log η_L obsd (log η_L calcd)	ref
74	2-(2-methoxyethoxy)ethanol [111-77-3]	25: 0.542(0.756); 60: 0.207(0.364)	15
75	ethyl acetoacetate [141-97-9]	20: 0.152(0.241)	15
76	2-ethoxyethyl acetate [111-15-9]	25: 0.011(0.006)	15
77	2-butoxyethanol [111-76-2]	25: 0.498(0.604); 60: 0.179(0.245)	15
78	tetraethylene glycol [112-60-7]	25: 1.652(1.988)	15
79	safrole [94-59-7]	25: 0.361(0.279)	15

Table 4. Ranges of the Target Property and Molecular Descriptors Covered by the Compounds in the Training Set of 237 Compounds with 673 Viscosity Data Points

variables	minimum	maximum
	Target Property	
log η_L	−0.928[a]	5.127
	Descriptors	
MR	8.2	113.9
T_c	460.4	861.4
χ_m	265	3360
E_{coh}	16.43	143.02
I_{OH}	0	2
I_{CN}	0	1
I_{amine}	0	1
I_{amide}	0	1
I_{ring}	0	1
T	−120	160

[a] The lowest viscosity value is in the validation set instead of the lowest viscosity value of −0.785 in the training set.

two layers: 10 input nodes plus a bias, a varying number of hidden-layer neurons (between two and five plus one bias), and a single output neuron corresponding to a compound's log η_L, the logarithmic value of the liquid viscosity in mPa·s. In contrast, the network B is a three-layer network and includes 10 input nodes, first hidden layer with a varying number of neurons (between two to five plus one bias) to receive the nine inputs except for T, second hidden layer with a neuron plus a bias, and a single output. The output node receives the output value from the second hidden-layer neuron and one input node (temperature (T, °C)) together with one bias. This network architecture can be suggested from the process of the estimation scheme using the Lewis-Squires chart[7] and supposed as a "compound-free" temperature dependence model for viscosity prediction. On the other hand, the model A can be classified as a "compound-specific" temperature dependence model.

All input and output data were normalized between 0.05 and 0.95, using the following scaling equation

$$x_i' = 0.9\,(x_i - x_{i,min})/(x_{i,max} - x_{i,min}) + 0.05 \qquad (1)$$

where x_i is the value of the ith input or output value and $x_{i,min}$ and $x_{i,max}$ are its minimum and maximum data over the data set. A sigmoid function was selected as the transfer function for each neuron. The training and validation data sets were submitted to analysis by two networks, using the same set of descriptors, to produce the best results with the fewest adjustable parameters. After finishing the learning process, the nets A and B were finally checked with the third data set, the test set.

The quality of the fitting of the modeling results for training, validation and test sets was evaluated based on the following statistical indices:[4] squared correlation coefficient r^2, root-mean-square error RMSE, average absolute error AAE, and bias error BIAS. Although such parameters are determined using experimental and calculated log η_L values, the η_L values in original unit may be more useful from practical use of this property in most chemical and mechanical engineering problems. Therefore, the following average percent error APE was added

$$\text{APE}\ (\%) = (100/n)\sum|(\eta_L\ (\text{calc}) - \eta_L\ (\text{exp}))/\eta_L\ (\text{exp})| \qquad (2)$$

where n is a number of data points to be evaluated. The network was optimized by classical back-propagation with patternwise (immediate) correction, i.e., weights are adapted after considering each individual pattern. Target function was the RMSE of the validation set only (weighted error of training and validation set is possible, however, the former gave better results). The learning was stopped in the minimum of the validation set RMSE.

RESULTS AND DISCUSSION

Comparison of Two NN Models. The modeling performances of two networks A and B trained using back-propagation are summarized in Table 5. The best architectures were determined to be (9+T+1):(3+1):1 (10 input neurons for the 10 descriptors (including T) plus a bias, three hidden-layer neurons plus a bias, and one output layer neuron for a total of 37 adjustable parameters) for the model A and (9+1):(4+1):(1+T+1):1 (9 input neurons plus a bias, four first hidden-layer neurons plus a bias, two second hidden-layer neurons (including T) plus a bias, and one output neuron for a total of 48 adjustable parameters) for the model B, respectively, using both training and validation data sets.

The number of training data points (=673) is about 14−18 times greater than the number of adjustable parameters for both models. As can be seen from Table 5, the statistical results, the values of r^2, RMSE, AAE, BIAS, and APE, clearly show that the best neural network model A found performs a somewhat better fit and prediction than the model B. Many alternative models with different architectures might be considered. One of such models, a two layer model having direct connection of T to output neuron, (9+1):(3+T+1):1, gave moderate modeling performance with compared to the performance of models A and B.

Thus, the results advocate the use of neural network model A for temperature-dependent viscosity estimation and the connection weights between the input and hidden-layers and between the hidden and output-layers, the bias elements for the hidden nodes and the output node are listed in Table 6 for convenient use of this model. The model performance for the training and validation sets, the relationship between the RMSE, and the number of training epochs are shown in Figure 2. The initial learning rate was 0.95 and momentum term was 0.75. Minimum of RMSE of validation data was

Table 5. Comparison of Modeling Statistics of the Trained Neural Network Models A and B

	net A (9+T+1): (3+1): 1 $n_{descr} = 10$, $n_{param} = 37$			net B (9+1): (4+1): (1+T+1): 1 $n_{descr} = 10$, $n_{param} = 48$		
	training	validation	test	training	validation	test
no. of compounds	237	124	79	237	124	79
no. of data points	673	423	133	673	423	133
r^2	0.956	0.932	0.884	0.940	0.922	0.835
RMSE	0.122	0.134	0.148	0.142	0.144	0.172
AAE	0.081	0.085	0.114	0.089	0.090	0.127
BIAS	0.003	−0.004	−0.007	0.005	0.011	0.003
error range in log units	−0.49 to +0.62	−0.85 to +0.62	−0.45 to +0.36	−1.00 to 1.26	−0.50 to 0.92	−0.35 to +0.63
APE (%)	19.41	19.48	24.59	21.42	20.39	28.97

Table 6. Weights of an Optimum Neural Network Model A with Three Hidden-Layer Neurons To Estimate Liquid Viscosity at Various Temperatures (−120 to 160 °C) of Organic Compounds

	hidden-layer neuron			
neuron	1	2	3	bias[a]
		Input Layer		
1	1.6698	4.0565	3.1916	na
2	−2.0432	−2.5681	−0.2127	na
3	−1.4937	−1.6493	−3.2384	na
4	1.9890	−2.2786	−1.6957	na
5	−5.0189	−0.9030	0.0558	na
6	0.3882	0.4835	−0.0501	na
7	−1.2170	0.8652	−1.2894	na
8	−0.1711	−0.9789	−0.1561	na
9	−0.9265	−0.5165	0.3987	na
10	6.0819	0.2802	3.5731	na
bias	2.9990	−0.1486	−0.8965	na
		Output Layer		
1	−4.8701	−2.7183	−2.1322	5.3614

[a] na denotes "not applicable".

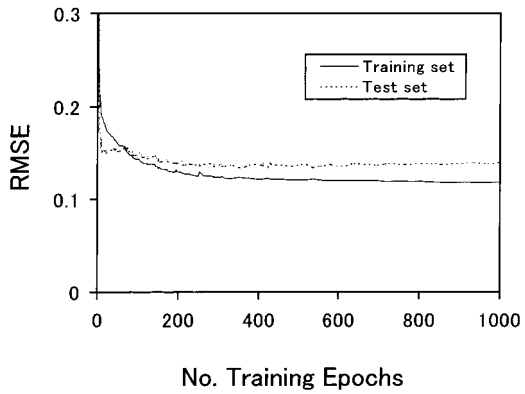


Figure 2. Effects of the number of training epochs on the performance of the neural model A with three hidden neurons.

achieved when the network was trained for 536 epochs. The more detailed discussions on the model A will be presented in the subsequent sections.

The calculated log η_L values by the model A for the compounds in training, validation, and test sets are shown in parentheses in Tables 1−3. The results are also shown in Figures 3−5.

Example Calculations. Application of the neural network model A ((9+T+1):(3+1):1) to 4-methyl-2-pentanone is demonstrated in Figure 6 to show how to estimate η_L at 35 °C. The calculated value is 0.537 mPa·s, compared to the value of 0.494 mPa·s found in the literature.[20] According to Reinhard and Drefahl,[10] the Joback and Reid method[12] gives

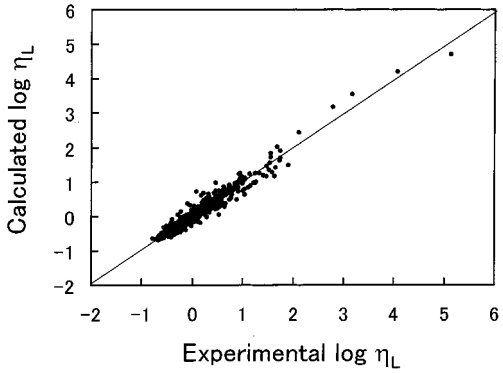


Figure 3. Scatter plot of the calculated versus experimental log η_L values for training set data points.

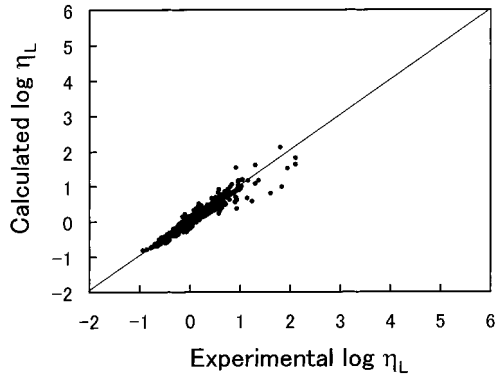


Figure 4. Scatter plot of the calculated versus experimental log η_L values for validation set data points.

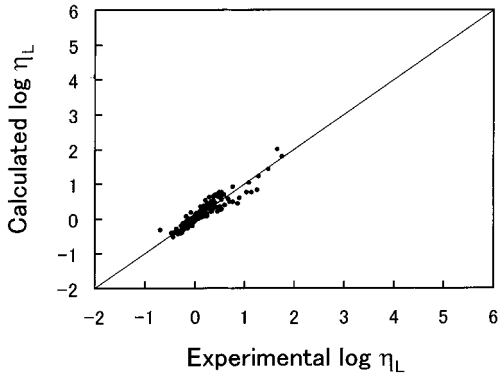


Figure 5. Scatter plot of the calculated versus experimental log η_L values for test set data points.

the value of 0.641 mPa·s and a compound-specific viscosity−temperature correlation proposed by Yaws et al.[21] gives the value of 0.541 mPa·s, which is just equivalent to ours. This method is easy to use compared with the use of the

Table 7. Comparison of the Statistics of the Neural Network Model A with the Previous Neural Network Model for Calculating Liquid Viscosity at 20 °C

	previous NN n_{descr}=9, n_{param}= 34		net A n_{descr}=10, n_{param}= 37	
	training	test	training	test
no. of compounds (= data points)	237	124	237	124
r^2	0.958	0.926	0.939	0.928
RMSE	0.118	0.161	0.142	0.158
AAE	0.084	0.105	0.097	0.101
BIAS	0.000	−0.008	−0.007	−0.021
error range in log units	−0.47 to +0.43	−0.86 to +0.35	−0.48 to +0.52	−0.85 to 0.33
APE (%)	19.7	23.1	22.7	22.0

Table 8. Evaluation Results for Calculating Liquid Viscosities of the Entire Data Set of 439 Compounds

chemical class	no. comp.	no. data, n	data range in log η_L	temp range °C	this work AAE[a]	this work BIAS[b]	van Velzen et al. n	van Velzen et al. AAE	van Velzen et al. BIAS	Joback-Reid n	Joback-Reid AAE	Joback-Reid BIAS
alkanes	29	86	−0.67−0.57	−20−152.3	0.028	−0.006	86	0.016	−0.000	86	0.040	0.013
cycloalkanes	35	171	−0.66−0.98	−50−125	0.064	0.029	171	0.057	−0.048	171	0.040	−0.024
alkenes/cycloalkenes/dienes	27	90	−0.78−0.63	−50−125	0.039	−0.022	90	0.021	−0.007	82	0.030	0.004
aromatic hydrocarbons	30	126	−0.65−1.01	−30−160	0.064	0.035	126	0.065	−0.051	125	0.055	−0.046
halogenated alkanes	43	121	−0.60−1.14	−120−130	0.074	−0.023	121	0.062	−0.022	119	0.076	−0.043
halogenated alkenes	6	15	−0.61−0.06	0−100	0.067	−0.031	15	0.143	0.143	7	0.095	−0.095
halogenated aromatics	14	35	−0.56−0.78	0−149	0.109	0.095	35	0.085	−0.056	23	0.085	−0.038
alcohols	29	90	−0.55−1.83	−59.4−140	0.188	−0.039	89	0.145	−0.007	89	0.155	−0.115
polyalcohols	7	20	0.30−5.13	−20−100	0.241	−0.042	20	0.431	−0.358	20	0.470	−0.443
phenols	2	7	0.10−1.64	10−90	0.145	−0.093	7	0.233	0.128	7	0.168	−0.168
ethers (incl. furan or dioxin)	26	58	−0.93−1.02	−100−100	0.065	−0.042	56	0.236	0.064	58	0.109	−0.087
aldehydes	8	13	−0.61−0.16	0−40	0.062	0.026	13	0.066	−0.043	13	0.078	−0.065
ketones	15	39	−0.65−0.35	−92.5−100	0.088	0.057	39	0.066	−0.042	38	0.080	0.045
acids	16	50	−0.37−1.59	0−100	0.107	−0.101	40	0.042	0.004	49	0.073	−0.051
esters	48	106	−0.68−1.91	0−100	0.072	−0.017	106	0.076	−0.025	105	0.079	−0.011
carbonates/anhydrides	5	11	−0.31−0.65	0−100	0.086	−0.084	2	0.005	0.005	11	0.157	0.040
polyfunctional O-compounds	21	30	0.01−1.75	0−60	0.181	0.161	27	0.480	0.462	30	0.232	0.199
halogenated O-compounds	5	8	−0.18−0.59	15−40	0.142	0.000	4	0.203	−0.031	5	0.290	−0.116
amines	25	59	−0.56−1.14	−6−100	0.100	−0.035	59	0.227	0.046			
nitriles	12	21	−0.48−0.29	0−30	0.053	−0.010						
pyridines (incl. pyrrole)	6	10	−0.15−0.18	10−30	0.216	0.216						
polyfunctional N-compounds	10	18	0.23−1.49	0−80	0.166	−0.051	14	0.284	−0.058			
amides	2	3	0.33−0.78	20−40	0.198	0.018						
nitro compounds	7	17	−0.22−0.58	0−60	0.081	−0.005	17	0.177	0.172			
S-compounds	12	25	−0.60−1.81	0−82	0.071	0.055						
total	440	1229	−0.93−5.13	−120−160	0.086	0.000	1137	0.103	−0.008	1038	0.198	−0.151

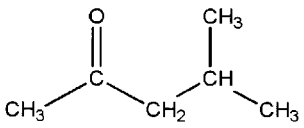
[a] AAE = average absolute error, $\Sigma|(\eta_L(\text{calcd}) - \eta_L(\text{exp}))|$/number of data points. [b] BIAS = bias error, $\Sigma(\eta_L(\text{calcd}) - \eta_L(\text{exp}))$/number of data points.

method of van Velzen et al., which has been used widely as the most reliable estimation scheme.

Comparison with the Previous Model for η_L at 20 °C. To check the performance of the present model to estimate η_L at 20 °C, statistical comparison with previous neural network model is shown in Table 7. As can be identified from this table, the values of RMSE, AAE, BIAS, and APE for the present model are slightly inferior than those for the previous neural network model for the training set. However, the predictive power of the present model is somewhat superior, and the results supports the model's eventual use as a tool to provide reasonable estimations at any temperature including 20 °C.

Comparison with Existing Approaches. Joback and Reid,[12] using their group-contribution approach, found that the liquid viscosity of 36 compounds at 3 to 5 temperatures was predicted with an average absolute error of 18%, which was comparable to predictions using methods developed by other workers, 20% for Thomas,[23] 16% for Orrick,[24] 15% for van Velzen et al.,[11] and 15% for Morris.[25] For representative 20 diverse liquids with 22 data points at various temperatures, a limited comparison[18] for the three methods gave average absolute errors of 19% (excluded the data points whose viscosities of larger than 2 mPa·s) for Lewis and Squires,[7] 22% for van Velzen et al.,[11] and 19% for Grain.[18] Reid et al.[8] discussed three methods of Orrick and Erbar,[8] van Velzen et al.,[11] and Przezdziecki and Sridhar[26] as the three best methods available against experimental 119 data points for 35 randomly selected simple compounds. Predicted results were not given for compounds which could not be treated or were not recommended, and the average absolute errors were not mentioned. They concluded that it is recommended that, in general, the method of van Velzen et al. be used to estimate low-temperature (somewhat above the freezing point) liquid viscosities except for first member of homologous series. For the method of van Velzen et al. in which log η_L is linear in T^{-1}, they originally tested their method on 314 different liquids with nearly 4500 data points, and 86.7% of the compounds tested had average errors of 15% or less.

Under the circumstances, a direct comparison of the present approach with other methods would be difficult and



1. Descriptors:

(1) *MR*: *MW*= 100.16, ρ (density) = 0.801 and n_D= 1.3958 [13-15]

$MR = (MW/\rho)\ \{ (n_D^2 - 1) / (n_D^2 + 2) \} = 30.0$

(2) *Tc*: Reported value [8] = 571.0 (K)

(3) χ_M: Experimental value [13] = 880 (10^{-12} $m^3 \cdot mol^{-1}$)

(4) *Ecoh*: Based on Fedor's group contribution method; [22]

$Ecoh = 3\ [CH_3] + [CH_2] + [CH] + [\text{-CO-}] = 3\ (4.71) + \text{- - -}$

$= 39.87\ (10^{-12}\ m^3 \cdot mol^{-1})$

(5) $I_{OH} = I_{CN} = I\ amine = I\ amide = I\ ring = 0$

2. Using neural network model:

(1) Normalization of inputs by using eq 1 and Table 4;

$x_1' = 0.2356$, $x_2' = 0.2982$, $x_3' = 0.2288$, $x_4' = 0.2166$,

$x_5' = x_6' = x_7' = x_8' = x_9' = 0.05$, $x_{10}' = 0.5482$

(2) Calculation of outputs from the hidden layer with Table 6;

$H_1 = 1 / [1+\exp \{- (x_1' \cdot w_{11} + x_2' \cdot w_{21} + \text{- - -})\}]$

$= 1 / [\ 1 + \exp \{- (0.2356)\ (1.6698) + (0.2982)\ (-2.0432) + \text{- - -}\}]$

$= 0.9972,$

$H_2 = 0.3255,\ H_3 = 0.643$

(3) Calculation of output from the output layer with Table 6;

$O = 1 / [1 + \exp \{- (H_1 \cdot v_1 + H_2 \cdot v_2 + H_3 \cdot v_3 + v_4)\ ]$

$= 1 / [1 + \exp \{- \{(0.9972)\ (-4.8701) + \text{- - -} + 5.3614\}\ ] = 0.1478$

(4) Rescaling the output value using eq 1;

$\log \eta_L = (0.1478 - 0.05)\{5.127 - (-0.928)\} / 0.9 + (-0.928) = -0.270$

η_L = antilog (-0.270) = 0.537 mPa·s at 35°C

Figure 6. Example estimation of η_L at 35 °C for 4-methyl-2-pentanone using (9+T+1): (3+1):1 neural net model.

depend heavily on the test data or compounds employed. For this reason the entire data set of 440 liquids employed in this study was used for the statistical evaluation. In Table 8 the evaluation results are compared with those by two conventional group-contribution methods of van Velzen et al.[11] and Joback and Reid.[12] The software system, ChemProp,[27] was used to get the estimated viscosity values by the two methods. Average absolute (AAE) and bias errors (BIAS) are shown for individual classes of compounds. The missing values found in the results by the two existing methods show the out of applicability range (compound type) of the methods. In addition, the original developers of such methods were based on rather simple structures and did not consider the application to more complex structures. In such cases ChemProp program generates an error message if the correct fragmentation is not clear rather than to calculate a dubious result. Therefore, the numbers of calculated data points by the two methods are sometimes less than those by this method. For example, 4 and 5 data points were calculated by van Velzen and Joback-Reid, respectively, against 8 data points by this method for halogenated oxygen-containing compounds as found in Table 8.

Table 9. Error Distribution for 1229 Data Points from 440 Compounds Studied in This Study

absolute error	no. of data points	cumulative percent
0−5%	271	22.1
5−10%	242	41.7
10−15%	176	56.1
15−20%	117	65.6
20−30%	179	80.1
30−50%	151	92.4
50−100%	72	98.3
>100%	21	100.0

According to the overall AAEs, the present method has the lowest value of 0.086 as compared with 0.103 by van-Velzen method and 0.198 by Joback-Reid method. Besides both BIAS and applicability range of our method are superior than those by the two methods. The performance of the present model for calculating the viscosity−temperature behavior has been also checked by examining the absolute error distribution as shown in Table 9. The resulting overall APE is 20.0% and about 80% of the data points could be calculated within a precision of 30% deviation. However, 7.6% of the data points calculated, 93 points from 62 compounds, had APEs of more than 50%. The list of the 62 compounds is shown in Table 10. An asterisk is attached to the compounds whose APEs of some data points at any

Table 10. List of 62 Compounds Whose Calculated Viscosities Showed Large Absolute Percent Errors More than 50%

compound class	compound
hydrocarbons	propylcyclohexane,[a] butylcyclohexane,[a] 1-isopropyl-4-methylbenzene, propylcyclopentane,[a] 1-phenyltridecane[a]
alcohols/phenols	methanol, ethanol,[a] 2-methyl-1-propanol,[a] 2-methyl-1-butanol,[a] 1-pentanol,[a] 3-pentanol,[a] 2-ethyl-1-butanol,[a] 2-methyl-2-butanol,[a] 2-propyn-1-ol, cyclohexanol, *cis*-2-methylcyclohexanol, *m*-cresol[a]
polyols	1,5-pentanediol, 1,3-butanediol,[a] ethylene glycol,[a] glycerol[a]
ketones	2-heptanone
acides	2-ethylhexanoic acid, oleic acid[a]
esters	ethyl cinnamate, allyl acetate,
diesters	dibutyl sebacate, bis(2-ethylhexyl)-*o*-phthalate,[a] diisobutyl *o*-phthalate
anhydrides	maleic anhydride
amines/anilines	aniline,[a] *o*-toluidine,[a] 2,4,6-trimethylpyridine, 2-methylpyridine, *N*-methylaniline,[a] 4-methylpyridine, 4-*tert*-butylpyridine
amides	*N*,*N*-dimethylacetamide
halogenated hydrocarbons	1,2-dichlorobenzene, 2-bromotoluene, benzotrichloride, 1,3-dichlorobenzene, 2-bromobutane, diiodomethane[a]
multifunctional compounds	3-bromoaniline, 2,2′-thiodiethanol, 1,2-bis(methoxyethoxy)ethane, lactonitrile, ethyl lactate, 1-ethoxy-2-propanol, 2-(2-methoxyethoxy)ethanol,[a] 4-hydroxy-4-methyl-2-pentanone, benzoyl bromide, eugenol,[a] 2-methoxyethanol, 2-mercaptoethanol, 2-ethoxyethanol,[a] 2-hydroxybenzaldehyde, bis(2-methoxyethyl)ether, diethylene glycol, triethylene glycol, tetraethylene glycol

[a] Not invalid for all data points.

temperatures are less than 50%. It is interesting to note that about 47% (29 compounds) of the compounds listed are OH containing ones, which can be classified into alcohols, phenols, polyols, and multifunctional compounds. The most of other remaining compounds are also N- and/or O-containing molecules which could participate in hydrogen boding. This observation is attributed to the insufficient description of the effects of hydrogen bonding which can form between two neighboring molecules or among small clusters of molecules, although the present model employs the indicator variables I_{OH} for the functionality. The polar interactions between hydrogen bonding and polar groups may play a prominent role in liquid viscosity process. On this point further refinement would be required. Other recent studies on liquid viscosity have also revealed that hydrogen bonding is a key factor to control liquid viscosity.[5,6]

In addition, an alternative NN model using a completely different set of descriptors which can be easily available either directly from the molecular structure or simple group-contributions is under development and some comparable analysis with the present model would be performed by us. The descriptors showing interactions between OH and O or C═O would be included in the new descriptor set.

CONCLUSIONS

The results obtained in this paper demonstrate that it is possible to generate a robust neural network model for temperature dependence of liquid viscosity for wide range of chemical structures and viscosity values. The agreement between observed and calculated values is generally excellent. For predictive use of this approach, however, erroneous results may be expected for some OH containing and/or multifunctional compounds, for which a new additional parameter on hydrogen bonding or the interactions of polar groups should be included. The success of this study shows the potential of applying the neural computing technique for estimating many other physical properties as a function of temperature.

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