Calculation of the Viscosity of Binary Liquids at Various Temperatures Using Jouyban–Acree Model

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Applicability of the Jouyban–Acree model for calculating absolute viscosity of binary liquid mixtures with respect to temperature and mixture composition is proposed. The correlation ability of the model is evaluated by employing viscosity data of 143 various aqueous and non-aqueous liquid mixtures at various temperatures collected from the literature. The results show that the model is able to correlate the data with an overall percentage deviation (PD) of 1.9±2.5%. In order to test the prediction capability of the model, three experimental viscosities from the highest and lowest temperatures along with the viscosities of neat liquids at all temperatures have been employed to train the model, then the viscosity values at other mixture compositions and temperatures were predicted and the overall PD obtained is 2.6±4.0%.

Key words prediction; absolute viscosity; modelling; Jouyban–Acree model; binary liquid mixture

Viscosity is an expression of the resistance of a fluid to flow, the higher the viscosity, the greater the resistance. Fluidity is defined as the reciprocal value of viscosity. Knowledge of viscosity of liquid mixtures at ambient and higher/lower temperatures is required in several industrial computations and also in analytical sciences. Determination of flow in pipelines and capillaries, heat-transfer and mass-transfer operations, liquid and semi-solid pharmaceutical formulation processes, developing separation methods like HPLC and capillary electrophoresis could be considered as the examples of the applications of viscosity information. In the pharmaceutical area, the sedimentation rate of suspensions, creaming of emulsions and drug release from aqueous solutions, are the important processes which could be affected by the viscosity.[1] Despite the reported experimental viscosity data for pure and mixed solvents at ambient and different temperatures in the chemical literature, a number of models have been presented to calculate the viscosity at a fixed temperature. The Arrhenius model[1] correlates the absolute viscosity (mPa s) of liquid mixtures for the ideal mixtures showing a linear relationship between viscosity and mixture composition. Grunberg and Nissan[2] proposed an extended form of the Arrhenius model for better representation of experimental data. Redlich–Kister extension[3] has been used in many papers[4—12] for expressing the non-ideality of the mixtures in arithmetic and logarithmic scales at a fixed temperature. To the best of our knowledge, there is no published model for calculating the viscosity of liquid mixtures at various temperatures and the aim of this work is to present such a model. The correlation ability of the proposed model was evaluated employing reported viscosity data from the literature using percentage deviation (PD) as an accuracy criterion. The prediction capability of the proposed model was also assessed using trained models by a minimum number of experimental data.

Theoretical Treatment

An equation analogous to the Arrhenius equation of chemical kinetics correlates temperature dependence of viscosity of a liquid:

$$\eta = B \exp\left(\frac{E_v}{RT}\right) \tag{1}$$

In which η is viscosity, B is a constant depending on molar volume and molecular weight of the liquid, E_v denotes an activation energy required to initiate flow between molecules, R represents the molar gas constant and T is absolute temperature.[1]

Equation 1 can be rewritten as:

$$\eta = \frac{N_0 h}{V} \exp\left(\frac{-\Delta G^*}{RT}\right) \tag{2}$$

Where N_0 denotes Avogadro's number, h is Plank's constant, V represents molar volume of the liquid and ΔG^* is the free energy of activation. For ideal binary liquid mixtures and by using the addition rule, Eq. 2 can be rearranged as[1]:

$$\eta_m = \frac{N_0 h}{X_1 V_1 + X_2 V_2} \exp\left(\frac{-\Delta G_m}{RT}\right) \tag{3}$$

Where X_1 and X_2 are the mole fraction of liquids 1 and 2, ΔG_m is free energy of activation for a flow of the mixture. Arrhenius proposed the following equation for calculating viscosity of the ideal liquid mixtures[1]:

$$\ln \eta_m = X_1 \ln \eta_1 + X_2 \ln \eta_2 \tag{4}$$

Where η_m is viscosity of the mixture, η_1 and η_2 are viscosity of neat liquids 1 and 2, respectively. The viscosity of real mixtures shows deviations from ideal values predicted by Eq. 4. In order to cover the deviations, Grunberg and Nissan[2] added one more variable to the Eq. 4 and obtained:

$$\ln \eta_m = X_1 \ln \eta_1 + X_2 \ln \eta_2 + dX_1X_2 \tag{5}$$

In which d is a curve-fit parameter. They found that d is positive for liquid mixtures showing negative deviation from Raoult's law, and is negative for systems exhibiting positive deviation from Raoult's law.[2]

A solution model (*i.e.* the Jouyban–Acree model) presented by Acree[13] was used to correlate different physico-chemical properties in mixed solvent systems; including the solubility of drugs in water-cosolvent mixtures,[14] electrophoretic mobility of analytes in mixed solvent electrolyte systems,[15—17] the instability rate constants in binary solvent systems,[18] the acid dissociation constants in water-organic solvent mixtures,[19] the dielectric constants of solvent mixtures[20] and the surface tensions of liquid mixtures.[21] Theoretical basis of the Jouyban–Acree model for describing the chemical potential of solutes dissolved in mixed solvents[13] and acid dissociation constants in aqueous-organic mixtures[19] have been provided in earlier papers. The constants of the Jouyban–Acree model represent differences in the various solute-solvent and solvent–solvent interactions in the mixture.[13] Therefore, it could be employed for mathematical representation of other physicochemical properties of the liquid mixtures like viscosity. The proposed

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Table 1. Details of Binary Absolute Viscosity Data, Temperature Range, Number of Data Points (*N*) and Percentage Deviations (PD) for Correlative and Predictive Analyses

No.	Liquid 1	Liquid 2	*T* range (K)	*N*	PD correlative	*N*	PD predictive	Ref.
1	Benzene	Chlorobenzene	298—313	44	0.3	30	0.4	25
2	Benzonitrile	Butan-1-ol	303—313	33	0.3	21	0.9	26
3	Benzonitrile	Methanol	303—308	22	0.1	12	0.1	26
4	Benzonitrile	2-Methylpropan-2-ol	303—313	33	2.7	21	3.1	26
5	Benzonitrile	Pentan-1-ol	303—313	33	0.7	21	0.9	26
6	Benzonitrile	Propan-1-ol	303—313	33	0.4	21	0.5	26
7	Benzonitrile	Ethanol	303—313	33	0.4	21	0.6	26
8	2,3-Butandiol	Water	308—318	27	2.7	15	4.6	27
9	1,2-Butanediol	Water	298—313	44	3.4	30	4.8	28
10	1,3-Butanediol	Water	298—318	55	3.3	39	4.2	28
11	1,4-Butanediol	Water	298—318	55	4.6	39	6.5	28
12	2,3-Butanediol	Water	298—318	55	3.9	39	6.9	28
13	2-Butanol	Decane	293—303	39	1.0	27	1.3	29
14	2-Butanol	Dodecane	293—303	39	1.0	27	1.4	29
15	2-Butanol	Octane	293—303	39	1.0	27	1.6	29
16	Butan-1-ol	1-Chlorobutane	288—308	39	0.4	27	0.5	30
17	Butanol	Dibutylamine	303—313	22	0.6	12	0.8	7
18	Butanol	Dipropylamine	303—313	22	0.3	12	0.3	7
19	1-Butanol	Tri-*n*-butylamine	303—313	22	1.3	12	1.4	8
20	Butanone	Isobutanol	288—318	44	2.8	30	2.9	31
21	Butanone	1-Butanol	288—318	44	1.4	30	1.5	31
22	Butanone	2-Butanol	288—318	44	4.2	30	4.4	31
23	Butanone	*tert*-Butanol	298—318	55	4.6	39	5.0	31
24	Butyl acetate	Propylene glycol	298—308	26	2.7	16	2.9	32
25	Butyl acetate	Triethylene glycol	298—308	26	5.7	16	6.0	32
26	Butylamine	Butanol	303—313	22	0.4	12	0.4	6
27	Butylamine	Decanol	303—313	22	2.0	12	2.3	6
28	Butylamine	Ethanol	303—313	22	1.3	12	1.5	6
29	Butylamine	Heptanol	303—313	22	2.0	12	2.2	6
30	Butylamine	Hexanol	303—313	22	0.7	12	0.8	6
31	Butylamine	Octanol	303—313	22	2.2	12	2.4	6
32	Butylamine	Pentanol	303—313	22	0.8	12	1.0	6
33	Butylamine	Propanol	303—313	22	0.5	12	0.7	6
34	*m*-Cresol	*o*-Cresol	313—333	33	0.3	21	0.4	33
35	*m*-Cresol	*p*-Cresol	313—333	33	0.4	21	0.4	33
36	*o*-Cresol	*p*-Cresol	313—333	33	0.2	21	0.2	33
37	Cyclohexane	2-Butanol	313—333	22	1.3	12	1.5	34
38	Cyclohexane	Chlorocyclohexane	313—333	22	0.0	12	0.1	9
39	Cyclohexane	Tetrahydrofuran	313—333	22	0.3	12	0.6	9
40	Decanol	Dibutylamine	298—313	22	2.3	12	2.5	7
41	Decanol	Dipropylamine	298—313	22	1.5	12	1.8	7
42	1-Decanol	Tri-*n*-butylamine	298—313	22	1.0	12	1.0	8
43	*N*,*N*-Dimethylformamide	Acetophenone	303—313	44	0.5	30	0.5	35
44	*N*,*N*-Dimethylformamide	Benzene	303—313	44	0.2	30	0.4	25
45	*N*,*N*-Dimethylformamide	Benzyl alcohol	303—313	44	0.4	30	0.6	35
46	*N*,*N*-Dimethylformamide	Chlorobenzene	298—313	44	0.2	30	0.3	25
47	1,4-Dioxane	Diethyl oxalate	298—313	33	0.3	21	0.4	36
48	1,4-Dioxane	Diethyl phthalate	298—313	33	1.6	21	2.3	36
49	1,4-Dioxane	Ethanediol	298—313	33	1.4	21	1.9	37
50	1,4-Dioxane	Ethyle acetoacetate	298—308	33	0.4	21	0.5	36
51	1,4-Dioxane	Ethyle acetoacetate	298—308	33	4.9	21	22.6	36
52	1,4-Dioxane	Hexane	298—308	33	0.4	21	0.6	37
53	1,4-Dioxane	Tributylamine	298—308	33	0.4	21	0.7	37
54	1,4-Dioxane	Triethylamine	298—303	33	0.2	21	0.3	37
55	1,4-Dioxane	1-Propanol	298—308	64	0.9	50	1.1	38
56	1,4-Dioxane	2-Propanol	298—308	56	1.3	42	1.6	38
57	1,3-Dioxolane	2-Butanol	298—308	22	2.7	12	3.0	34
58	Ethanol	C6	298—313	63	0.5	51	0.7	39
59	Ethanol	1-Chlorobutane	298—313	39	0.4	28	0.8	30
60	Ethanol	Dipropylamine	298—313	22	2.1	12	2.4	7
61	Ethanol	Dibutylamine	298—318	22	1.7	12	1.8	7
62	Ethanolamine	Water	288—308	40	2.2	26	2.6	11
63	Ethyl acetate	Decan-1-ol	303—313	33	2.0	21	3.2	40
64	Ethyl acetate	Diethylene glycol	303—313	26	8.3	16	8.8	32
65	Ethyl acetate	Heptan-1-ol	303—318	33	1.8	21	2.5	40
66	Ethyl acetate	Hexan-1-ol	298—308	33	1.3	21	1.7	40
67	Ethyl acetate	Octan-1-ol	298—308	33	0.9	21	1.4	40
68	Ethyl acetate	Pentan-1-ol	298—308	33	2.0	21	3.0	40
69	Ethyl acetate	Propylene glycol	298—308	26	8.3	16	8.8	32
70	Ethyl acetate	Triethylene glycol	298—308	26	2.7	16	2.9	32

Table 1. (Continued)

71	Ethyl acetate	3,5,5-Trimethylhexan-1-ol	298—308	33	4.1	21	5.1	40
72	Ethyl chloroacetate	Decane	298—308	33	0.3	21	0.5	41
73	Ethyl chloroacetate	Dodecane	298—308	33	0.4	21	0.5	41
74	Ethyl chloroacetate	Heptane	298—308	33	0.2	21	0.8	41
75	Ethyl chloroacetate	Hexane	298—308	33	0.3	21	0.4	41
76	Ethyl chloroacetate	Nonane	298—308	33	0.4	21	0.5	41
77	Ethyl chloroacetate	Octane	298—308	33	0.3	21	0.5	41
78	Ethyl valerate	1-Hexanol	298—308	33	0.8	21	0.8	42
79	Ethyl valerate	1-Hexanol	298—308	33	0.3	21	0.5	42
80	Ethyl valerate	1-Pentanol	298—308	33	0.5	21	0.5	42
81	Ethyl valerate	1-Pentanol	293—313	33	0.3	21	0.3	42
82	Ethylene glycol	Water	293—313	77	2.8	57	5.5	43
83	Ethylene carbonate	Dimethyl carbonate	293—313	22	0.0	12	0.0	44
84	Heptanol	Dibutylamine	293—313	22	0.9	12	1.0	7
85	Heptanol	Dipropylamine	293—353	22	0.9	12	1.3	7
86	1-Heptanol	Tri-*n*-butylamine	298—313	22	1.2	11	1.5	8
87	Hexanol	Dibutylamine	303—313	22	0.6	12	0.7	7
88	Isoamyl acetate	Propylene glycol	303—313	26	13.7	16	10.3	32
89	2-Isopropoxyethanol	Propylamine	303—313	51	1.8	39	1.7	45
90	Methanol	C6	303—313	52	0.6	40	1.2	39
91	Methanol	1-Chlorobutane	298—308	39	0.3	27	0.4	30
92	2-Methoxyethanol	Acetone	298—318	63	1.2	51	2.2	46
93	Methyl acetate	Diethylene glycol	298—318	26	13.3	16	14.2	32
94	Methyl acetate	Ethylene glycol	288—308	26	3.8	16	4.0	32
95	Methyl acetate	Propylene glycol	293—303	26	3.4	16	3.6	32
96	Methyl acetate	Triethylene glycol	298—308	26	5.7	16	6.1	32
97	4-Methyl-2-pentanone	Ethyl benzoate	298—308	42	1.1	30	1.7	47
98	2-Methylpropan-2-ol	C6	298—308	63	5.5	51	5.7	39
99	*n*-Octane	Di-*n*-butyl ether	298—308	59	0.6	45	0.7	4
100	*n*-Octane	1-Propanol	283—303	46	0.9	32	1.0	4
101	Octanol	Dibutylamine	298—318	22	11.9	12	16.0	7
102	Octanol	Dipropylamine	293—308	22	0.7	12	0.9	7
103	1-Octanol	Tri-*n*-butylamine	293—308	22	1.1	12	1.3	8
104	Pentanol	Dibutylamine	303—313	22	0.3	12	0.3	7
105	Pentanol	Dipropylamine	303—313	22	0.5	12	0.7	7
106	1-Pentanol	Tri-*n*-butylamine	303—313	22	1.9	12	2.1	8
107	Propane-1,2 diol	Water	303—313	44	2.0	30	3.8	10
108	Propan-2-ol	C6	303—313	63	1.1	51	1.6	39
109	Propan-1-ol	1-Chlorobutane	303—313	39	0.3	28	0.5	30
110	Propanol	Dibutylamine	303—318	22	0.7	12	0.9	7
111	1-Propanol	Di-*n*-butyl ether	298—318	73	0.8	57	0.9	4
112	Propanol	Dipropylamine	288—308	22	0.3	12	0.4	7
113	1-Propanol	Tri-*n*-butylamine	303—313	22	1.2	12	1.3	8
114	2-Propoxyethanol	Dipropylamine	293—308	51	2.1	39	2.2	45
115	2-Isopropoxyethanol	Dipropylamine	303—313	51	1.5	39	1.6	45
116	2-Propoxyethanol	Propylamine	303—313	54	2.0	46	2.7	45
117	Propylamine	Butanol	298—318	22	0.5	12	0.9	5
118	Propylamine	Decanol	298—318	22	2.7	12	2.8	5
119	Propylamine	Ethanol	298—318	22	0.4	12	0.5	5
120	Propylamine	Heptanol	303—313	22	1.7	12	1.8	5
121	Propylamine	Octanol	303—313	22	1.6	12	1.7	5
122	Propylamine	Pentanol	303—313	22	0.9	12	0.9	5
123	Propylamine	Propanol	303—313	22	0.3	12	0.6	5
124	Toluene	Butan-1-ol	303—313	33	0.7	21	1.3	48
125	Toluene	Ethanol	303—313	22	0.4	12	0.6	48
126	Toluene	Methanol	303—313	22	0.2	12	0.3	48
127	Toluene	2-Methylpropan-2-ol	303—313	33	2.6	21	2.5	48
128	Toluene	Pentan-1-ol	303—308	33	0.3	21	0.5	48
129	Toluene	Propan-1-ol	303—308	33	0.7	21	1.2	48
130	Toluene	Acetic acid	303—313	68	0.8	54	1.1	49
131	Toluene	Propionic acid	303—313	68	1.3	54	1.5	49
132	Tri-*n*-butylamine	Pyridine	303—313	33	0.3	21	0.4	50
133	Tri-*n*-butylamine	Tetrachloroethylene	293—323	33	0.3	21	0.4	50
134	Tri-*n*-butylamine	Tetradecane	293—323	33	1.1	21	1.7	50
135	Tri-*n*-butylamine	Tetrahydrofurane	298—308	33	0.2	21	0.3	50
136	Tri-*n*-butylamine	Trichloroethylene	298—308	33	0.4	21	0.5	50
137	Tri-*n*-butylamine	Triethylamine	298—308	33	0.3	21	0.4	50
138	Water	*N*,*N*-Dimethylenediamine	298—308	70	8.2	56	9.2	51
139	Water	Ethylenediamine	298—308	70	6.6	56	31.0	51
140	Water	1-Methyl-2-pyrrolidinone	298—308	105	5.4	89	6.1	52
141	Water	2-Pyrrolidinone	303—323	95	5.3	89	5.1	52
142	Water	Trimethylenediamine	303—323	65	7.2	51	8.2	51
143	Water	1-Vinyl-2-pyrrolidinone	298—338	95	9.5	89	13.5	52

model for representing the viscosity of binary mixtures is:

$$RT\cdot\ln \eta_m = RT\cdot X_1 \ln \eta_1 + RT\cdot X_2 \ln \eta_2 + W_0X_1X_2 + W_1X_1X_2(X_1-X_2) + W_2X_1X_2(X_1-X_2)^2 \quad (6)$$

Where W_0—W_2 are the model constants. Since RT is a constant figure at a fixed temperature, Eq. 6 could be rearranged as:

$$\ln \eta_m = X_1 \ln \eta_1 + X_2 \ln \eta_2 + K_0X_1X_2 + K_1X_1X_2(X_1-X_2) + K_2X_1X_2(X_1-X_2)^2 \quad (7)$$

in which K_0, K_1 and K_2 are W_0/RT, W_1/RT and W_2/RT, respectively. Equation 7 has been employed in a series of papers published by Oswal and Desai and produced accurate correlation for the viscosity of binary solvents.[5—8] The applications of the model could be extended for calculating the viscosity of binary solvents at different temperatures as:

$$\ln \eta_m = X_1 \ln \eta_{1,T} + X_2 \ln \eta_{2,T} + A_0\left[\frac{X_1X_2}{T}\right] + A_1\left[\frac{X_1X_2(X_1-X_2)}{T}\right] + A_2\left[\frac{X_1X_2(X_1-X_2)^2}{T}\right] \quad (8)$$

Where $\eta_{1,T}$ and $\eta_{2,T}$ are viscosity of pure liquids at temperature T and A_0—A_2 are the model constants. These constants are computed by a least squares analysis. The other versions of Eq. 8 have been successfully applied for calculation of solute solubilities in mixed solvent at various temperatures,[22] apparent acid dissociation constants of analytes,[23] dielectric constant[20] and also surface tension[24] of liquid mixtures at different temperatures.

Experimental Data and Computational Methods

Available viscosity data of binary liquid mixtures from the literature[5—11,25—52] were collected and used to build a database. All experimental data were used as reported in the literature and only one datum corrected as there was a typographical error. The viscosity of butanone (X_1=0.9037)+ *tert*-butanol (X_2=0.0961) at 313.15 K was reported as 0.212.[31] When it was compared with the corresponding viscosities at lower temperatures and also viscosity of pure butanone at the same temperature, it is simply justified that the correct value should be 1.212.

To assess the accuracy of the proposed model for correlating experimental viscosity data at different temperatures, the experimental viscosity data was fitted to the model and the percentage deviation (PD) was calculated as:

$$\mathrm{PD} = \frac{100}{N}\sum_{1}^{N}\frac{|\text{calculated} - \text{observed}|}{\text{observed}}$$

Where N is the number of experimental viscosity data points in each set. Individual percentage deviation (IPD) was also computed by:

$$\mathrm{IPD} = 100\left(\frac{|\text{calculated} - \text{observed}|}{\text{observed}}\right)$$

Results and Discussion

Available viscosity data of binary liquids at various temperatures collected from the literature has been fitted to Eq. 8 and the back-calculated viscosities have been employed to compute the corresponding PD and IPD values. This analysis was called correlative analysis and the obtained results along with details of the data sets were listed in Table 1. The viscosity data of isoamyl acetate+propylene glycol produced the highest PD (*i.e.* 13.7%) and cyclohexane+chlorocyclohexane produced the lowest PD (<0.05%). The overall PD (±S.D.) of 143 sets studied was 1.9±2.5%. The relative frequency of IPD values sorted in three subgroups for correlative analysis was shown in Fig. 1. The probability of IPD ≤4% is 0.86 whereas the corresponding value for IPD >10% is 0.04. These findings show the accuracy of the proposed model for calculating the viscosity of binary mixtures at various temperatures and the model could be used for screening the experimentally obtained viscosity data to detect the possible outliers for re-determination.

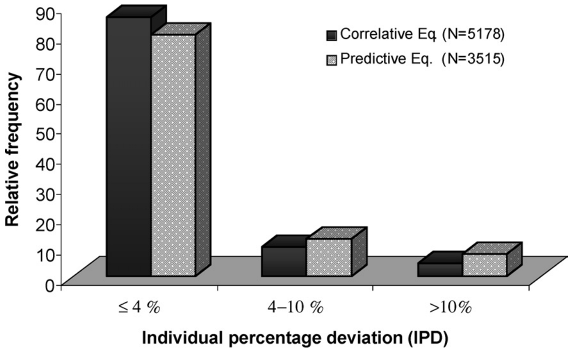

Fig. 1. Relative Frequency of IPD for Correlative and Predictive Analyses

To test the prediction capability of Eq. 8, a minimum number of five viscosity data points (*i.e.* X_1≡0, 0.3, 0.5, 0.7, 1.0) at the highest and lowest temperatures of interest and the viscosity data of pure liquids at each temperature have been used as training set. After training the proposed model, it has been used to predict viscosity of other liquid compositions and temperatures by using the interpolation technique. Then PD values were calculated using predicted viscosities and listed in Table 1. The minimum and maximum PD values were observed for cyclohexane+chlorocyclohexane (0.1%) and water+ethylenediamine (31.0%), respectively. The overall PD for predictive analysis was 2.6±4.0%. As shown in Fig. 1, the probability of viscosity prediction with IPD ≤4% is 0.80 and this means that one can use a minimum number of experimental data and to predict the viscosity of the rest of solvent compositions and temperatures of interest. This is obviously help the researchers to design the process using minimum experimental efforts.

In conclusion, the proposed model provided reasonably accurate calculations for the absolute viscosity of binary liquid mixtures at various temperatures and the model could be used in viscosity data modelling. The model could be used in practice for screening the experimentally collected viscosity data to detect possible outliers in order to re-determination and also predicting unmeasured viscosities at different binary mixtures at various temperatures after successful training of the model using a minimum number of experimental data.

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