# A VISCOSITY FORMULA FOR BINARY MIXTURES, THE ASSOCIATION DEGREES OF CONSTITUENTS BEING TAKEN INTO CONSIDERATION. XII.

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**Mixtures showing minima in the viscosity versus concentration curves.** Mixtures of alcohols with benzene have been treated by several investigators with recent views to the molecular complexity of liquids. Despite that the mixture of ethyl alcohol with benzene has an eutectic point on the freezing point curve, the molecular weight of the alcohol in benzene as determined by the freezing point method has been found abnormally high for concentrations above 0.278 mol. % alcohol according to Gibbons⁽¹⁾ or 0.383 mol. % alcohol according to Peterson and Rodebush.⁽²⁾ The mean molecular weight calculated from Gibbons's data if it is taken at the concentration range from 1.8 to 35.5 mol. % is 170 or 3.7 times the formal molecular weight. Such an anomalous phenomenon of association as observed in the case of the alcohol in benzene has been quite recently criticized by Müller and Mortier.⁽³⁾ The van Arkel association⁽⁴⁾ which signifies that the molecular polarization $P$ of dipole substances dissolved in a solvent polar or non-polar is simply related to the concentration by van Arkel's formula

$$P=\frac{4\pi}{3}n\left(\alpha+\frac{\mu^2}{3kT+cn\mu^2}\right)$$

($\alpha$ is the optical polarization, $n$ the number of dipole per c.c., $\mu$ the dipole moment, $k$ the Boltzmann constant, and $c$ a constant) does not fit for the case in question. Substances with van Arkel's association scarcely indicate molecular complex cryoscopically and ebullioscopically as it is a well known fact that alcohols give normal molecular weights in dilute aqueous solutions. They point out that although propyl alcohol appears to obey the above rule at very low concentration, yet the constant $c$ thereby gives about 60, whereas the normal value lies between 1.1 and 1.8, and conclude that even at extremely high dilution the reciprocal action of the alcohol molecules seems to

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(1) W. A. Gibbons, *J. Phys. Chem.*, **21** (1917), 48.

(2) J. M. Peterson and W. H. Rodebush, *J. Phys. Chem.*, **32** (1928), 709.

(3) F. H. Müller and P. Mortier, *Physik. Z.*, **10** (1935), 371.

(4) A. E. v. Arkel and J. L. Snoek, *Trans. Faraday Soc.*, **30** (1934), 707.

become greater than the separating force of the solvent and such effect is attributed to the anomalous position of the dipole within the alcohol molecules.

Gundermann(5) observes that the pairs of butyl alcohol-benzene and especially ethyl alcohol-benzene do not obey the theory of Debye which tells that the dielectric constant of liquids decreases as the square of the applied voltage. As to the former pair the decrease of dielectric constant with voltage is qualitatively understood from the Debye theory, but the latter pair behaves anomalously in such a manner that the effect indicates relatively high value at low concentration of the alcohol, but too low at high concentration, the pure alcohol, however, showing little effect against field intensity. He attributes this to the association of the alcohol molecules in benzene solution.

The anomalous behaviour of benzene towards alcohols has also been known in the measurement of infra-red absorption spectra. Freymann(6) notices that in the mixtures of butyl alcohol and ethyl alcohol with benzene the characteristic frequency band of OH-group at 0.9631 $\mu$ shifts intensively to 0.9778 $\mu$, and that the same band of alcohols also shifts to longer wave in the dilution of the alcohols with benzene, but that such phenomenon is scarcely observed with other solvents.

The viscosity of mixtures of benzene with alcohols, especially with ethyl and propyl alcohols studied by Dunstan(7), indicates a distinct minimum against the composition in each system. No adequate reason has been given for binary liquid mixtures which exhibit minima in the viscosity versus concentration curves. If we take an analogy to the phenomena of some inorganic salts such as $NH_4$-, K-, Rb-, and Cs-halides in consideration,(8) the viscosity minimum would be elucidated by the dissociation of alcohol molecules as Dunstan supposes, but such opinion is not only opposite to the recent view above cited, but also gives unsatisfactory results in actual calculation, since midway of the curves cannot be expressible by such method of calculation in which two solutional viscosities of both components must be postulated at the same time. Therefore, so far as the present knowledge of alcohols-benzene mixtures is concerned, we are reduced to assume that the anomalous change of association of alcohol molecules occurs in benzene solution, a change tends probably to greater association.

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(5) H. Gundermann, *Ann. Physik.*, [5], **6** (1930), 545.

(6) R. Freymann, *Compt. rend.*, **193** (1932), 923.

(7) A. E. Dunstan, *J. Chem. Soc.*, **85** (1904), 817; **87** (1905), 11; *Z. physik. Chem.*, **49** (1904), 590.

(8) T. Ishikawa and T. Baba, this Bulletin, **10** (1935), 153.

In this series of papers⁽⁹⁾ the authors have developed the Ishikawa formula for the viscosity of binary mixtures standing on the fundamental hypothesis that the associations of both components are invariant throughout the concentration when no chemical change on their mixing. A reflection shall be given on this point.

If there happens some anomalous (increasing or decreasing) change of association in one component, say, component 2 in its greatest dilution with component 1 where the quantity of component 2 is quite enough to change its original association $a_2$ into $a'_2$ statistically, component 2 in the opposite case in quantity may also suffer change in association more or less, the degree of which, however, is not always the same in both the extremities, say, $a_2$ tends statistically to $a''_2$ in this case. If such is the case, the viscosity change with composition between these extremities may be expressible by the following formula:

$$\eta=\frac{\eta_1 k_1 a_1(1-z_m)}{k_1 a_1(1-z_m)+k_2 a'_2 z_m}+\frac{\eta_2 k_2 a''_2 z_m}{k_1 a_1(1-z_m)+k_2 a''_2 z_m}$$

$$=\frac{\eta_1}{1+K'\dfrac{z_m}{1-z_m}}+\frac{\eta_2}{1+K''\dfrac{1-z_m}{z_m}}$$

in which $K'=\dfrac{k_2 a'_2}{k_1 a_1}$ and $K''=\dfrac{k_1 a_1}{k_2 a_2''}$.

It is easily understood by simple calculation that the necessary condition under which $\eta$ as expressed by the above formula takes a minimum is $K'K''>1$.

Provided that the anomalous behaviour of benzene towards alcohols attributes wholly to the change of association of alcohols as defined above and to no further interactions between both the component molecules, viscosity curves between the two extremities must be completely expressible by this formula.

The viscosity data for these mixtures are adopted from the studies of Dunstan, Fischler, and Bhide and Watson.⁽¹⁰⁾ The calculated values are listed in the third column in each table from Tables 1 to 4, and how they are compared with observed values is clearly seen from Fig. 1, in which the curves are all traced by use of the calculated values, putting the viscosities of benzene at given temperatures as unity.

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(9) T. Ishikawa, this Bulletin, **4** (1929), 5, 25, 149, 288; **5** (1930), 47, 117; **8** (1933), 280, 293; **9** (1934), 155; **10** (1935), 153, 248.

(10) A. E. Dunstan, *loc. cit.* J. Fischler, *Z. Elektrochem.*, **19** (1913), 126. B. W. Bhide and H. E. Watson, *J. Chem. Soc.*, **1927**, 2101.

For the system ethyl alcohol (2)-benzene (1) the $K'$ (1.754) thus obtained is, as recent views demand, much greater than the normal $K$ (0.466) as calculated on the assumption that no change of association occurs on mixing the two liquids, their ratio giving 3.76. This value is quite coincident with the value 3.7 as the abnormal value of the association of the alcohol already explained at the begining of this paper.

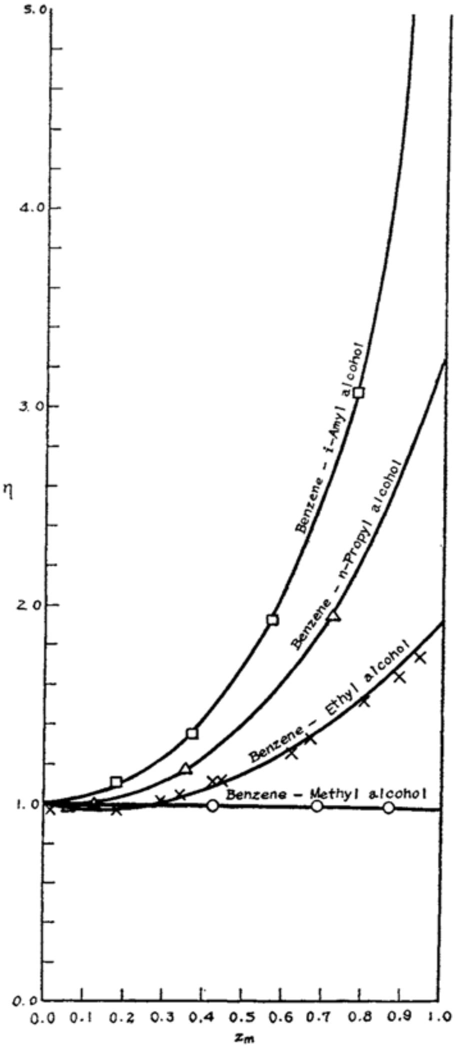

Fig. 1.

Table 1. Benzene—Methyl alcohol, 25°C. (Fischler).

| $z_m$ | $\eta$ obs. | $\eta$ calc. |
|---|---|---|
| 0.0000 | 0.005758 | — |
| 0.4235 | 0.005723 | 0.005720 |
| 0.6879 | 0.005684 | 0.005681 |
| 0.8684 | 0.005645 | 0.005643 |
| 1.0000 | 0.005608 | — |

$K' = 0.619$, $K'' = 1.603$. $K'K'' = 0.99$.

Table 2. Benzene—Ethyl alcohol, 25°C. (Dunstan).

| $z_m$ | $\eta$ obs. | $\eta$ calc. |
|---|---|---|
| 0.0000 | 0.005821 | — |
| 0.0192 | 0.005668 | 0.005778 |
| 0.1874 | 0.005669 | 0.005669 |
| 0.2927 | 0.005885 | 0.005840 |
| 0.3421 | 0.006071 | 0.005977 |
| 0.4267 | 0.006465 | 0.006287 |
| 0.4485 | 0.006514 | 0.006395 |
| 0.6295 | 0.007282 | 0.007398 |
| 0.6692 | 0.007758 | 0.007758 |
| 0.8067 | 0.008837 | 0.008956 |
| 0.8923 | 0.009545 | 0.009848 |
| 0.9475 | 0.01013 | 0.01048 |
| 1.0000 | 0.01113 | — |

$K' = 1.754$, $K'' = 1.453$. $K'K'' = 2.55$.

Table 3. Benzene—$n$-Propyl alcohol, 25°C. (Dunstan).

| $z_m$ | $\eta$ obs. | $\eta$ calc. |
|---|---|---|
| 0.0000 | 0.005978 | — |
| 0.0631 | 0.005917 | 0.005934 |
| 0.1291 | 0.005989 | 0.005990 |
| 0.3554 | 0.007028 | 0.006952 |
| 0.7243 | 0.01167 | 0.01167 |
| 1.0000 | 0.01962 | — |

$K' = 1.701$, $K'' = 2.246$. $K'K'' = 3.82$.

Table 4. Benzene—$i$-Amyl alcohol, 30°C. (Bhide and Watson).

| $z_m$ | $\eta$ obs. | $\eta$ calc. |
|---|---|---|
| 0.0000 | 0.00562 | — |
| 0.1813 | 0.00621 | 0.00607 |
| 0.3714 | 0.00760 | 0.00760 |
| 0.5707 | 0.0108 | 0.0108 |
| 0.7802 | 0.0173 | 0.0173 |
| 1.0000 | 0.0321 | — |

$K' = 1.712$, $K'' = 3.355$. $K'K'' = 5.74$.

For other systems the anomalies of the association of alcohols are not so great as that of ethyl alcohol, but their products $K'K''$'s still take greater values than unity, although $K''$'s seem to approach to their normal $K$'s.

The anomalous change of molecular complexity of ethyl alcohol is also observed in viscosity determination with other solvents such as nitromethane, cyanobenzene, and allyl mustard oil.(11) With respect to these mixtures other physical properties to be needly explained are little known except that nitromethane has van Arkel's association.

Tables 5, 6, and 7 and Fig. 2 show the coincidence between the observed and calculated viscosities.

Table 5. Ethyl alcohol—Nitromethane (Wagner).

| $z_m$ | $\eta$ obs. | $\eta$ calc. |
|---|---|---|
| 0.0000 | 1.0000 | — |
| 0.0579 | 0.9015 | 0.9043 |
| 0.1153 | 0.8210 | 0.8249 |
| 0.2290 | 0.7037 | 0.7042 |
| 0.4505 | 0.5704 | 0.5652 |
| 0.6518 | 0.5143 | 0.5143 |
| 0.8288 | 0.5003 | 0.5114 |
| 1.0000 | 0.5385 | — |

$K' = 2.231$, $K'' = 1.268$. $K'K'' = 2.83$.

Table 6. Ethyl alcohol—Cyanobenzene (Wagner).

| $z_m$ | $\eta$ obs. | $\eta$ calc. |
|---|---|---|
| 0.0000 | 1.0000 | — |
| 0.0073 | 0.9958 | 0.9941 |
| 0.0147 | 0.9883 | 0.9880 |
| 0.0297 | 0.9784 | 0.9771 |
| 0.0607 | 0.9539 | 0.9560 |
| 0.1273 | 0.9210 | 0.9206 |
| 0.2703 | 0.8949 | 0.8823 |
| 0.3718 | 0.8785 | 0.8780 |
| 0.6346 | 0.9091 | 0.9264 |
| 1.0000 | 1.0768 | — |

$K' = 2.000$, $K'' = 0.925$. $K'K'' = 1.85$.

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(11) J. Wagner, *Z. physik. Chem.*, **46** (1903), 867.

Table 7. Ethyl alcohol—Allyl mustard oil (Wagner).

| $z_m$ | $\eta$ obs. | $\eta$ calc. |
|---|---|---|
| 0.0000 | 1.0000 | — |
| 0.0611 | 0.9237 | 0.9279 |
| 0.1275 | 0.8523 | 0.8551 |
| 0.2790 | 0.7331 | 0.7331 |
| 0.4810 | 0.6512 | 0.6349 |
| 0.6902 | 0.5995 | 0.5917 |
| 0.7769 | 0.5879 | 0.5879 |
| 1.0000 | 0.6100 | — |

$K' = 1.823$, $K'' = 1.221$. $K'K'' = 2.23$.

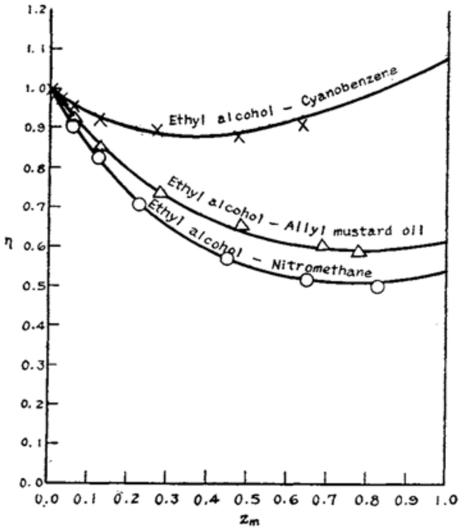

Fig. 2.

The mixture of acetic acid and benzene has been a subject in recent years for the same object of study as in alcohols-benzene mixtures. Bury and Jenkins⁽¹²⁾ observe with a cryoscopic method that the molecular form of acetic acid in benzene is completely dimeric at all concentrations greater than ca 3 mol. % acetic acid. Beckmann and Liesche⁽¹³⁾ obtain with an ebullioscopic determination that acetic acid in benzene is not completely dimeric, but partially suffers molecular dissociation to give 0.87 times the dimolecular weight for the mean value between 0.9 and 9.4 mol. % acetic acid. Dadieu and Kohlrausch⁽¹⁴⁾ study the Raman effect of this mixture and of mixtures of acetic acid in other solvents such as ether, alcohol, and water, and point out that the deepest frequency 440 of acetic acid is markedly weakened in benzene solution, and moreover that

Table 8. Benzene—Acetic acid, 25°C. (Dunstan).

| $z_m$ | $\eta$ obs. | $\eta$ calc. |
|---|---|---|
| 0.0000 | 0.005978 | — |
| 0.0355 | 0.005941 | 0.005965 |
| 0.1295 | 0.005907 | 0.005942 |
| 0.2289 | 0.005962 | 0.005960 |
| 0.2769 | 0.005969 | 0.005988 |
| 0.5822 | 0.006658 | 0.006637 |
| 0.7081 | 0.007341 | 0.007344 |
| 0.8666 | 0.008932 | 0.009007 |
| 1.0000 | 0.01174 | — |

$K' = 0.711$, $K'' = 3.108$. $K'K'' = 2.21$.

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(12) C. R. Bury and H. O. Jenkins, *J. Chem. Soc.*, **1934**, 688.

(13) E. Beckmann and O. Liesche, *Z. physik. Chem.*, **92** (1917), 421.

(14) A. Dadieu and K. W. F. Kohlrausch, *Physik. Z.*, **31** (1930), 514.

the powerful frequency 1432 is replaced by two weak frequencies, these phenomena being not observed in other solvents.

Dunstan(15) who makes viscosity determinations on this mixture confirms a distinct minimum existing at 16.7 mol. % acetic acid at 25°C.

Opposite to alcohols-benzene mixtures, acetic acid in benzene suffers molecular dissociation, and the ratio of the anomalous $K'$ (0.711) to the normal $K$ (0.820) is 0.867, which is quite equivalent to the 0.87 calculated from Beckmann and Liesche's data above explained.

**The variation of the product $K'K''$ with temperature.** Substances which behave anomalous association in non-polar or polar liquids with van Arkel's association are, in most cases, hydroxylated, carboxylated, and some chlorinated compounds. Acetone-carbon bisulphide(16) is an example of the second case, and ethylene dichloride-benzene and tetrachloroethane-benzene(17) are good examples of the last case.

For one more example of hydroxylated compounds phenol-diphenylmethylamine, and for chlorinated compounds ethylene dichloride-benzene will be considered below, since the measurements done by Bramley(18) and by Faust(19) respectively on these mixtures are convenient for the consideration

Table 9. Phenol—Diphenylmethylamine (Bramley).

| $z_m$ | $\eta_{9.8°}$ | | $\eta_{20.1°}$ | |
|---|---|---|---|---|
| | obs. | calc. | obs. | calc. |
| 0.0000 | 0.2010 | — | 0.1104 | — |
| 0.0580 | 0.1877 | 0.1884 | 0.1048 | 0.1050 |
| 0.1214 | 0.1752 | 0.1755 | 0.0995 | 0.0996 |
| 0.2011 | 0.1614 | 0.1618 | 0.0935 | 0.0937 |
| 0.2803 | 0.1502 | 0.1502 | 0.0885 | 0.0885 |
| 0.3536 | 0.1411 | 0.1410 | 0.0845 | 0.0844 |
| 0.4933 | 0.1306 | 0.1286 | 0.0798 | 0.0789 |
| 0.5728 | 0.1226 | 0.1216 | 0.0764 | 0.0758 |
| 0.7106 | 0.1145 | 0.1144 | 0.0730 | 0.0729 |
| 0.8307 | 0.1097 | 0.1109 | 0.0715 | 0.0718 |
| 0.9084 | 0.1090 | 0.1098 | 0.0708 | 0.0717 |
| 1.0000 | 0.1096 | — | 0.0722 | — |
| | $K' = 1.648$, $K'' = 1.150$.<br>$K'K'' = 1.895$. | | $K' = 1.440$, $K'' = 1.190$.<br>$K'K'' = 1.714$. | |

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(15) A. E. Dunstan, *J. Chem. Soc.*, **87** (1905), 11.
(16) O. Faust, *Z. physik. Chem.*, **79** (1912), 97.
(17) W. Herz and W. Rathmann, *Z. Elektrochem.*, **19** (1913), 589.
(18) A. Bramley, *J. Chem. Soc.*, **109** (1916), 10.
(19) O. Faust, *loc. cit.*

Table 9 (*Concluded*).

| $z_m$ | $\eta_{30°}$ | | $\eta_{40°}$ | | $\eta_{60°}$ | | $\eta_{80°}$ | |
|---|---|---|---|---|---|---|---|---|
| | obs. | calc. | obs. | calc. | obs. | calc. | obs. | calc. |
| 0.0000 | 0.0709 | — | 0.0474 | — | 0.0253 | — | 0.01585 | — |
| 0.0537 | 0.0673 | 0.0676 | 0.0451 | 0.0458 | 0.02495 | 0.0250 | 0.01586 | 0.01586 |
| 0.1000 | 0.0650 | 0.0653 | 0.0440 | 0.0444 | 0.0247 | 0.0247 | 0.01585 | 0.01587 |
| 0.1557 | 0.0626 | 0.0628 | 0.0428 | 0.0431 | 0.02445 | 0.0245 | 0.01589 | 0.01590 |
| 0.2384 | 0.0597 | 0.0597 | 0.0415 | 0.0415 | 0.02415 | 0.0242 | 0.01594 | 0.01594 |
| 0.3403 | 0.0570 | 0.0567 | 0.0402 | 0.0399 | 0.02395 | 0.0239 | 0.01603 | 0.01603 |
| 0.4843 | 0.0542 | 0.0539 | 0.0388 | 0.0385 | 0.02375 | 0.0236 | 0.01624 | 0.01619 |
| 0.6720 | 0.0519 | 0.0519 | 0.0377 | 0.0377 | 0.0237 | 0.0237 | 0.01650 | 0.01649 |
| 0.8187 | 0.0513 | 0.0513 | 0.0376 | 0.0377 | 0.0240 | 0.0240 | 0.01682 | 0.01683 |
| 0.9074 | 0.0510 | 0.0512 | 0.03785 | 0.0380 | 0.02435 | 0.0244 | 0.01708 | 0.01706 |
| 1.0000 | 0.0513 | — | 0.03835 | — | 0.02480 | — | 0.01735 | — |

$K'=1.791$, $K''=0.812$. $K'=1.557$, $K''=0.916$. $K'=1.081$, $K''=1.163$. $K'=0.908$, $K''=1.197$.
$K'K''=1.455$. $K'K''=1.585$. $K'K''=1.257$. $K'K''=1.086$.

Table 10. Benzene—Ethylene dichloride (Faust).

| $z_m$ | $\eta_{0°}$ | | $\eta_{19.4°}$ | | $\eta_{50°}$ | |
|---|---|---|---|---|---|---|
| | obs. | calc. | obs. | calc. | obs. | calc. |
| 0.0000 | 0.00850 | — | 0.00619 | — | 0.00418 | — |
| 0.1000 | 0.00843 | 0.00840 | 0.00615 | 0.00613 | 0.00422 | 0.00422 |
| 0.2000 | 0.00837 | 0.00837 | 0.00615 | 0.00615 | 0.00428 | 0.00423 |
| 0.3000 | 0.00835 | 0.00840 | 0.00620 | 0.00621 | 0.00435 | 0.00435 |
| 0.5000 | 0.00868 | 0.00868 | 0.00645 | 0.00645 | 0.00455 | 0.00455 |
| 0.7000 | 0.00933 | 0.00926 | 0.00690 | 0.00689 | 0.00492 | 0.00486 |
| 1.0000 | 0.01077 | — | 0.00800 | — | 0.00565 | — |

$K' = 1.132$, $K'' = 1.295$. $K' = 0.997$, $K'' = 1.370$. $K' = 0.721$, $K'' = 1.664$.
$K'K'' = 1.466$. $K'K'' = 1.366$. $K'K'' = 1.200$.

of the temperature variation of the product $K'K''$. Figs. 3 and 4 show the points of the observed values lying on the calculated curves.

As seen from the footnotes of Tables 9 and 10, the decrease of the product $K'K''$ with rising temperature is a strong indication that the anomalous molecular complexity decreases with rise of temperature. This tendency expects that at sufficiently high temperature where the anomaly disappears or where they are truely chemically indifferent with each other the product $K'K''$ tends to unity.

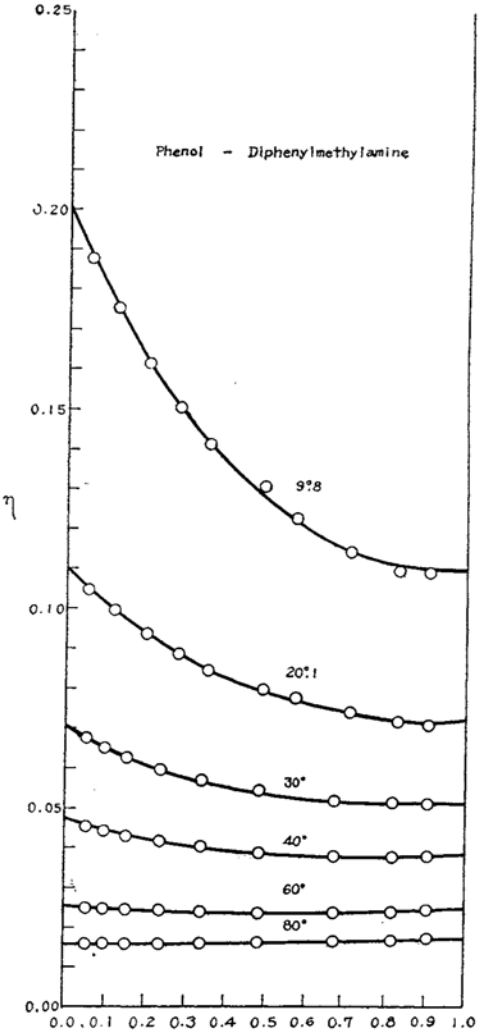

Fig. 3.

It is also noticed from these examples that the condition $K'K''>1$ under which anomaly of molecular complexity occurs is not always followed by a minimum in the viscosity versus concentration curves.

**A further comparison of the product $K'K''$ in the formula newly proposed for liquid mixtures with the corresponding term in the Thiesen formula for gaseous mixtures.** In Part II⁽²⁰⁾ we have made a little comparison of the Ishikawa formula with the Thiesen formula for gaseous mixtures. The type of the Thiesen formula has been rigorously supported by the classical kinetic theory of gases,⁽²¹⁾ the general form of which runs:

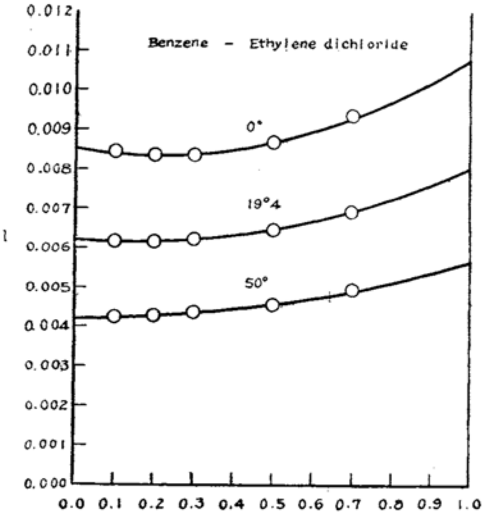

Fig. 4.

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(20) T. Ishikawa, this Bulletin, **4** (1929), 5.

(21) J. P. Kuenen, "Eigenschaften der Gase," Leipzig 1919. H. Schmick, *Physik. Z.*, **29** (1928), 633.

$$\eta=\frac{\eta_1}{1+A_1\dfrac{z_m}{1-z_m}}+\frac{\eta_2}{1+A_2\dfrac{1-z_m}{z_m}}\,,$$

$$A_1=\left(\frac{\sigma}{S_1}\right)^2\frac{1-\dfrac{m_1}{m_1+m_2}\Theta_1}{0.797}\sqrt{\frac{m_1+m_2}{2m_2}}\,\frac{T+C_{12}}{T+C_1}\,,$$

$$\Theta_1=\frac{m_1}{2(m_1+m_2)}+\frac{1}{4}\,\frac{m_1^2}{m_1^{1/2}(m_1+m_2)^{3/2}}\,\ln\frac{\sqrt{m_1+m_2}+\sqrt{m_2}}{\sqrt{m_1+m_2}-\sqrt{m_2}}\,.$$

$A_2$, $\Theta_2$ take the similar forms as $A_1$ and $\Theta_1$ respectively, in which $m_1$, $m_2$ are the molecular weights of components 1 and 2; $S_1$, $S_2$ the molecular diameters of components 1 and 2; $\sigma=\frac{1}{2}(S_1+S_2)$; $C_1$, $C_2$ the Sutherland constants of components 1 and 2; $C_{12}$ the corresponding constant due to the attraction between a molecule of kind 1 and a molecule of kind 2; and $T$ the absolute temperature.

Judging from the form of the formula newly proposed, one may imagine that no apparent difference is observed between these two formulae. It is of great interest, however, to note here that for gaseous mixtures whether or not the viscosity indicates a maximum the product $A_I\,A_2$, the corresponding term to the product $K'K''$ in our case, is always less than or equal to unity (see Table 11), whilst for liquid mixtures $K'K''$ is always greater than or equal to unity.

Table 11.

| Mixtures showing a maximum. | | |
|---|---|---|
| Mixture | $A_1\,A_2$ | Observers |
| $H_2-CH_4$ | 0.624 | Azumi, Graham, Trautz and Sorg. |
| $H_2-C_2H_2$ | 0.528 | Azumi. |
| $H_2-C_2H_4$ | 0.517 | Thomsen, Trautz and Stauf. |
| $H_2-C_2H_6$ | 0.508 | Azumi. |
| $H_2-C_3H_6$ | 0.462 | Azumi. |
| $H_2-NH_3$ | 0.597 | Thomsen, Trautz and Heberling. |
| $H_2-CO_2$ | 0.404 | Graham, Puluj, Thomsen, Breitenbach, Trautz and Kurz. |
| $H_2-SO_2$ | 0.339 | Trautz and Weizel. |
| $H_2-NO$ | 0.467 | Graham. |
| $H_2-N_2O$ | 0.401 | Graham, Trautz and Kurz. |
| $H_2-HCl$ | 0.430 | Trautz and Narath. |
| $H_2-Cl_2$ | 0.341 | Thomsen. |
| $He-Ar$ | 0.575 | Tänzler, Trautz and Kipphan, Trautz and Binkele. |
| $He-Kr$ | 0.454 | Nasini and Rossi. |
| $CH_4-NH_3$ | 0.977 | Jung and Schmick. |
| $NH_3-C_2H_4$ | 0.997 | Thomsen, Trautz and Heberling. |

Table 11 (*Concluded*).

| Mixtures showing no maximum. | | |
|---|---|---|
| Mixture | $A_1 A_2$ | Observers |
| $H_2-CO$ | 0.491 | Graham, Trautz and Baumann. |
| $H_2-O_2$ | 0.457 | Kleint, Graham, Trantz and Melster. |
| $H_2-N_2$ | 0.496 | Graham, Kleint, Trautz and Baumann. |
| $H_2-$Air | 0.485 | Graham. |
| $H_2-He$ | 0.806 | Gille, Trautz and Kipphan, Trautz and Baumann. |
| $H_2-Ar$ | 0.411 | Trautz and Ludewigs, Trautz and Binkele. |
| $H_2-Ne$ | 0.547 | Trautz and Binkele. |
| $He-Ne$ | 0.739 | Trautz and Kipphan. |
| $Ne-Ar$ | 0.968 | Trautz and Kipphan, Trautz and Binkele. |
| $NH_3-N_2$ | 0.940 | Thomsen, Trautz and Heberling. |
| $NH_3-O_2$ | 0.901 | Thomsen, Trautz and Heberling. |
| $NH_3-$Air | 0.973 | Jung and Schmick. |
| $NH_3-CO_2$ | 0.901 | Thomsen. |
| $NH_3-Cl_2$ | 0.704 | Thomsen. |
| Air $-CO_2$ | 0.985 | Jung and Schmick. |
| Air $-HCl$ | 0.966 | Jung and Schmick. |
| Air $-H_2S$ | 0.978 | Jung and Schmick. |
| $CO-N_2$ | 1.00 | Trautz and Melster. |
| $CO_2-N_2O$ | 1.00 | Trautz and Kurz. |
| $CO_2-SO_2$ | 0.989 | Jung and Schmick. |
| $O_2-CO_2$ | 0.996 | Graham. |
| $CO-O_2$ | 1.00 | Graham, Trautz and Melster. |
| $N_2-O_2$ | 0.973 | Graham, Kleint, Trautz and Melster. |
| $CH_4-O_2$ | 0.923 | Graham. |
| $CH_4-C_2H_2$ | 0.983 | Azumi. |
| $CH_4-C_2H_6$ | 0.967 | Trautz and Sorg. |
| $CH_4-C_3H_8$ | 0.898 | Trautz and Sorg. |
| $C_2H_2-C_3H_6$ | 0.985 | Azumi. |
| $N_2-C_2H_4$ | 1.01 | Trautz and Melster. |
| $CO-C_2H_4$ | 1.01 | Trautz and Melster. |
| $C_2H_4-O_2$ | 1.06 | Trautz and Melster. |
| $C_2H_4-$Air | 1.00 | Breitenbach. |
| $C_2H_6-C_3H_8$ | 1.01 | Trautz and Sorg. |
| $C_3H_6-C_3H_8$ | 0.982 | Azumi. |
| $C_3H_8-N_2O$ | 1.05 | Trautz and Kurz. |
| $C_3H_8-CO_2$ | 1.04 | Trautz and Kurz. |

Although the formula for gaseous mixtures stands on a sound theoretical basis, a closer investigation(22) informs us that in the case where a maximum occurs the calculated viscosities are not quite coincident with the observed viscosities, giving some more or less functional divergence. On the other hand, our formula affords no such divergencies as clearly recognized from the figures above given.

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(22) H. Azumi, *Bull. Inst. Phys. Chem. Research*, **11** (1932), 1103.