VISCOSITY OF BINARY AND TERNARY LIQUID NONELECTROLYTE MIXTURES. COMPARISON OF CORRELATION EQUATIONS AND ANALYSIS OF VISCOSITY CURVES

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The derived equations for correlation of viscosity of binary and ternary mixtures were tested and compared with those of McAllister, Chandramouli and Laddha. Testing on a large number of experimental data revealed that the equations for correlation of viscosity of binary mixtures are approximately as accurate as the equation of McAllister. On adding another coefficient, the relative deviations are lowered by 26—40%. Equations for correlation of viscosity of ternary mixtures are about as accurate as the equation of Chandramouli and Laddha. Analysis of viscosity curves of binary mixtures revealed a correlation between their form and the chemical nature of the components of the mixture.

In our preceding work[1], we derived equations for the correlation of viscosity of binary and ternary mixtures of liquid nonelectrolytes (Eqs (*14*), (*16*), (*18*), (*31*), and (*33*)). The present work is devoted to their verifying and comparing with those of McAllister, Chandramouli and Laddha (Eqs (*1*) and (*2*)) on the basis of experimental data referring to viscosities of binary and ternary mixtures in the literature. The method of determination of these data, their accuracy, and their statistic weights were not taken into consideration. The coefficients of Eqs (*14*), (*16*), (*18*), (*31*), and (*33*) were calculated by the least squares method, namely by solving normal equations. The coefficients of Eqs (*1*) and (*2*) were calculated by the same method with the aid of the Taylor series and the relaxation method.

Binary Systems

Eqs (*14*), (*18*), and (*31*) were verified and compared with (*1*) for 547 different systems at different temperatures, representing 1267 isothermal systems or 8986 experimental points.

The studied systems consisted of the following compounds (roman numerals denote class according to Ewell[2], arabic in parentheses denote order numbers within the class):

Class I: (1) acetamide, (2) N-ethylacetamide, (3) ethylene glycol, (4) formamide, (5) 2-hydroxybenzoic acid, (6) 2-hydroxypropionic acid, (7) N-methylacetamide, (8) pyrocatechol, (9) resorcinol, (10) triethylene glycol, (11) heavy water, (12) water.

Class II: (1) acetaldoxime, (2) acetonitrile, (3) allyl alcohol, (4) aniline, (5) 1,2-benzenediamine, (6) benzyl alcohol, (7) benzylamine, (8) 1-butanol, (9) 2-butanol, (10) 1-butylamine, (11) 4-chloroaniline, (12) 2-chlorophenol, (13) cyclohexanol, (14) diethylamine, (15) diethylammonium picrate, (16) diphenylamine, (17) 2,4-dimethyl-3-ethylpyrrol, (18) 2,4-dimethylpyrrol, (19) 2,4-dinitrophenol, (20) ethanol, (21) N-ethylaniline, (22) phenol, (23) phenylhydrazine, (24) guaiacol, (25) 1-heptanol, (26) heptylammonium picrate, (27) 1-hexanol, (28) *o*-cresol, (29) *m*-cresol, (30) *p*-cresol, (31) benzoic acid, (32) chloroacetic acid, (33) dichloroacetic acid, (34) 2-phenylacrylic acid, (35) heptanoic acid, (36) hexanoic acid, (37) butyric acid, (38) formic acid, (39) acetic acid, (40) octadecanoic acid, (41) octanoic acid, (42) picric acid, (43) propionic acid, (44) trichloroacetic acid, (45) trifluoroacetic acid, (46) valeric acid, (47) menthol, (48) methanol, (49) N-methylaniline, (50) 3-methylbutane-1-ol, (51) 2-methylpropane-1-ol, (52) urea, (53) 1-naphthylamine, (54) nitroethane, (55) 2-nitrophenol, (56) 4-nitrophenol, (57) nitromethane, (58) 1-octanol, (59) 1-pentanol, (60) piperidine, (61) 1-propanol, (62) 2-propanol, (63) propionitrile, (64) pyrrol, (65) *o*-toluidine, (66) *m*-toluidine, (67) *p*-toluidine, (68) tripropylammonium picrate, (69) thymol.

Class III: (1) acetaldehyde, (2) acetanhydride, (3) acetophenone, (4) acetone, (5) allyl isothiocyanate, (6) n-amyl acetate, (7) n-amylbutyrate, (8) anisole, (9) *p*-azoxyanisole, (10) *p*-azoxyphenetole, (11) benzaldehyde, (12) benzonitrile, (13) benzyl acetate, (14) benzyl benzoate, (15) n-butyl acetate, (16) n-butyl methyl ketone, (17) N-t-butylpyrrolidone, (18) butyraldehyde, (19) 2-chloronitrobenzene, (20) 3-chloronitrobenzene, (21) quinoline, (22) cyclohexanone, (23) n,n-dibutyl ester of decanedioic acid, (24) n,n-dibutyl ether, (25) N,N-diethylacetamide, (26) N,N-diethylaniline, (27) diethylene glycol dimethyl ether, (28) diethyl ether, (29) N,N-diethylformamide, (30) diethyl ketone, (31) N,N-diethylpropionamide, (32) diphenyl ether, (33) diphenylmethylamine, (34) N,N-diisopropylacetamide, (35) N,N-dimethylacetamide, (36) N,N-dimethylaniline, (37) dimethyl sulphoxide, (38) *m*-dinitrobenzene, (39) 1,4-dioxane, (40) N,N-dipropylacetamide, (41) ethyl acetate, (42) ethylacetoacetate, (43) ethylbenzoate, (44) ethyl butyrate, (45) ethylene glycol dimethyl ether, (46) ethylene carbonate, (47) ethyl ester of 2,2-diethyl-3-hydroxybutyric acid, (48) ethyl ester of 2-ethyl-3-hydroxybutyric acid, (49) ethyl formate, (50) ethyl isobutyrate, (51) ethyl isovalerate, (52) ethyl methyl ketone, (53) ethyl nitrate, (54) ethyl propionate, (55) ethyl trichloroacetate, (56) phenetole, (57) phenyl isothiocyanate, (58) n-hexadecyl acetate, (59) isoamyl acetate, (60) isoamyl ether, (61) isobutyl acetate, (62) isobutylformate, (63) isobutyl methyl ketone, (64) D-camphor, (65) thioacetic acid, (66) 2,6-lutidine, (67) methyl acetate, (68) 2-methylquinoline, (69) 4-methylcyclohexanone, (70) N-methyl-ε-caprolactam, (71) methyl propyl ketone, (72) 2-methylpyridine, (73) 3-methylpyridine, (74) N-methyl-2-pyrrolidone, (75) nicotine, (76) nitrobenzene, (77) 1-nitronaphthalene, (78) *o*-nitrotoluene, (79) *p*-nitrotoluene, (80) *p*-nitroso-N,N-dimethylaniline, (81) paraldehyde, (82) n-propyl acetate, (83) pyridine, (84) tetrahydrofurane, (85) tetraisoamylammonium picrate, (86) tetranitromethane, (87) tetrapropylammonium picrate, (88) trichloroacetaldehyde, (89) triethylamine, (90) triethylene glycol dimethyl ether, (91) 1,3,5-trinitrobenzene.

Class IV: (1) bromoform, (2) chloroform, (3) 1,2-dibromoethane, (4) 1,2-dichloroethane, (5) dichlorofluoromethane, (6) dichloromethane, (7) pentachloroethane, (8) 1,1,2,2-tetrabromoethane, (9) 1,1,2,2-tetrachloroethane, (10) trichloroethylene.

Class V: (1) benzene, (2) bromobenzene, (3) 1-bromobutane, (4) 2-bromobutane, (5) 1-bromododecane, (6) chlorobenzene, (7) 4-chlorobromobenzene, (8) 1-chlorobutane, (9) 1-chlorodecane, (10) 1-chlorododecane, (11) 1-chlorohexadecane, (12) 1-chlorohexane, (13) antimony(III) chloride, (14) arsenum(III) chloride, (15) tin(IV) chloride, (16) titanium(IV) chloride, (17) 4-chloroiodobenzene, (18) 1-chlorooctadecane, (19) 1-chlorooctane, (20) 1-chloropentane, (21) 1-chlorotetradecane, (22) cyclohexane, (23) *cis*-decaline, (24) *trans*-decaline, (25) n-decane, (26) 1,4-dibromobenzene, (27) diphenyl, (28) diphenylmethane, (29) 2,2-dimethylpropane, (30) n-dodecane, (31) ethanethiol, (32) ethylbenzene, (33) ethyl iodide, (34) n-heptane, (35) n-hexadecane, (36) n-hexane, (37) iodobenzene, (38) cumene, (39) methylcyclohexane, (40) methyl iodide, (41) mesitylene, (42) naphthalene, (43) n-nonane, (44) n-octane, (45) n-pentane, (46) pyrene, (47) carbon disulphide, (48) *o*-terphenyl, (49) tetrachloroethylene, (50) tetrachloromethane, (51) n-tetradecane, (52) tetraline, (53) 2,2,4-tetramethylpentane, (54) toluene, (55) n-undecane, (56) *p*-xylene, (57) *m*-xylene, (58) *p*-xylene.

In further text, the compounds will be denoted by a fraction, whose numerator will indicate the class and denominator the order number (*e.g.*, III/67 denotes methyl acetate).

We shall consider the binary systems given in the following survey, where the temperature (°C), type of the viscosity curve in parentheses, and the reference are given after the symbols of both components.

II/2 + V/1 : 20(3a + 2a(3))[4]; II/2 + V/54 : 20(3a)[4]; II/2 + V/56 : 20(3a)[4];
II/2 + V/57 : 20 (3a)[4]; II/2 + V/58 : 20(3a)[4];
II/4 + V/1 : 9, 5(2b)[5]; 25(2b)[6]; 25[7]; 77(2a)[5]; II/4 + V/54 : 25(2b)[8]; 30(2b)[8]; 35(2b)[8]; 40(2b)[8];
II/6 + V/1 : 25(2b)[9]; 35[9]; 45[9]; 50(2ab)[9];
II/8 + V/1 : 20(2b)[9]; 25[10]; 30[9]; 35[10]; 35[9]; 45(2ab)[9]; II/8 + V/22 : 25(2cl)[11]; 35[11]; 45[11]; 55(2cl)[11];
II/8 + V/45 : 10(2b)[11]; 25(2b)[11];
II/8 + V/50 : 25(2a + 3a(4))[5]; 25[11]; 35[11]; 45[11]; 55[11];
II/9 + V/50 : 25(2cl)[11]; 35[11]; 45[11]; 55(2cl)[11];
II/10 + V/34 : 20(2c3)[12]; 30[12]; 40(2cl)[12];
II/11 + V/26 : 110(2c5)[13]; 115(2c5)[13]; 120(2c5)[13];
II/14 + V/1 : 20(2a)[9]; 30(2a)[9]; 35(2a)[9];
II/20 + V/1 : 15(2b)[14]; 15[15]; 20[14]; 25(2cl)[15]; 25[16]; 30[14]; 40[14]; 40(2c2)[15]; II/20 + V/22 : 20[17]; 25(2c3)[11]; 30[17]; 35[11]; 40[17]; 45[11]; 50[17]; 55(2c4)[11]; 60[17]; 80[17]; 100[17]; II/20 + V/31 : 25(2ab)[5]; II/20 + V/36 : 20(2a)[18]; 25(2a)[18]; 25[19]; 30(2a)[18]; 35(2a)[18]; 40(2a)[18]; II/20 + V/41 : 20(2cl)[5]; II/20 + V/47 : 25(2a)[20]; II/20 + V/50 : 20(2c2 + 3b9(6))[15]; 25[5]; 25[11]; 25[21]; 25[5]; 35[11]; 40(2c2 + 3b9(7))[15]; 45[11]; 55[11]; 60(2c5 + 3b9(8))[15]; II/20 + V/54 : 24(2b)[5];
II/22 + V/42 : 97, 5(2b)[5];
II/25 + V/50 : 25(2a + 3a(9))[5]; 25[11]; 35[11]; 45[11]; 55[11];
II/37 + V/50 : 25(2a + 3a(9))[5]; 25[11]; 35[11]; 45[11]; 55[11];
II/32 + V/13 : 50(3b5)[22]; 60(3b5)[22]; 70(3b5)[22]; II/32 + V/14 : 60(2ab)[23];
II/33 + V/15 : 50(2b)[24]; 60(2c2)[24]; 70(2c2)[24];
II/35 + V/1 : 25(2b)[5]; II/35 + V/50 : 25(2a)[5];
II/36 + V/1 : 25(2b)[5]; II/36 + V/50 : 25(2a)[5];
II/37 + V/1 : 25(2b)[5]; II/37 + V/50 : 25(2a)[5];
II/39 + V/1 : 25(2c2)[9]; 25[25]; II/39 + V/14 : 50(3b5)[23]; 60(3b5)[23]; 70(3b5)[23]; II/39 + V/50 : 25 (2c4)[5]; 25[26];
II/41 + V/50 : 25(2a)[5];
II/43 + V/1 : 20(2b)[9]; 30[9]; 35[9]; 45(2ab)[9];
II/44 + V/14 : 60(2b)[23]; II/44 + V/15 : 50(2b)[24]; 60(2b)[24]; 70(2b[24]);
II/46 + V/1 : 25(2b)[5];
II/47 + V/42 : 82,2(2b)[5]; 99(2a)[5];
II/48 + V/1 : 11(3a)[5]; 20(2c2 + 3b7(4))[5]; 25[5]; II/48 + V/41 : 20(3b6)[5]; II/48 + V/50 : 25(2a +
+ 3a(1))[5]; 25[11]; 35[11]; 45[11]; 55[11]; II/48 + V/57 : 20(3b6)[5];
II/49 + V/1 : 25(2ab)[7];
II/50 + V/1 : 18,4(2b)[5]; II/50 + V/36 : 18,7(2b)[5];
II/51 + V/50 : 25(2cl)[11]; 35[11]; 45[11]; 55(2c4)[11];
II/54 + V/50 : 30(2ab)[27]; 35(2ab)[27]; 45[27];
II/55 + V/15 : 40(2b)[28]; 60(2b)[28];

II/56 + V/42 : 121(2ab)[5];
II/57 + V/50 : 30(2a)[27]; 35(2a)[27]; 45(2a)[27];
II/58 + V/50 : 25(2a)[5]; 25[11]; 35[11]; 45[11]; 55[11];
II/59 + V/22 : 25(2b)[11]; 35[11]; 45[11]; 55(2a)[11]; II/59 + V/36 : 25(2b)[11]; 40[11]; 55(2ab)[11];
II/59 + V/50 : 25(2a + 3a(9))[11]; 35[11]; 45[11]; 55(2a)[11];
II/61 + V/1 : 20[29]; 25(2b)[30]; 35(2b)[30]; 40[29]; 60[29]; II/61 + V/22 : 20[29]; 25(2c1)[11]; 35[11]; 40[29]; 45[11]; 55(2c2)[11]; 60[29]; II/61 + V/34 : 20(2b)[29]; 40[29]; 60(2a)[29]; II/61 + V/36 : 20(2b)[29]; 40[29]; II/61 + V/41 : 20(2b)[5]; II/61 + V/50 : 25(2a + 3a(3))[5]; 25[11]; 35[11]; 45[11]; 55[11]; II/61 + V/54 : 20[29]; 25(2b)[31]; 40[29]; 60[29];
II/69 + V/42 : 97,5(2b)[5];
III/3 + IV/2 : 25(1)[32]; III/3 + IV/4 : 25(2a)[32]; III/3 + IV/7 : 25(3b3)[32]; III/3 + IV/9 : 10(3b5)[33]; 80(3b5)[33];
III/4 + IV/1 : 25(2a)[34]; III/4 + IV/2 : 0(3b2)[35]; 0[36]; 19[36]; 20(3b2)[37]; 20[35]; 25[38]; 25[39]; 25[37]; 30[37]; 39[36]; 40(3b1)[35]; III/4 + IV/5 : −80(3b3)[40]; III/4 + IV/6 : 0(3a)[32]; III/4 + IV/7 : 25(2a)[32];
III/4 + IV/9 : 0(2a)[32]; 25[28];
III/22 + IV/2 : 18(2a + 1(5))[5];
III/28 + IV/2 : 0(3a + 2a(6))[41]; 10[41]; 15[37]; 20[41]; 20(3a + 2a(6))[37]; 25[37]; 25[42]; 30[41]; III/28 + IV/7 : 0(2ab)[32];
III/32 + IV/7 : 25(1)[32];
III/39 + IV/2 : 20[37]; 25(3a)[37]; 25[38]; 30[37];
III/56 + IV/2 : 0(2a)[5]; III/56 + IV/7 : 25(2a)[32]; III/56 + IV/9 : 0(2a + 3a(3))[32];
III/63 + IV/2 : 25(3b4)[39];
III/75 + IV/4 : 25(2a)[43]; 50[43]; 75(3a)[43];
III/76 + IV/1 : 10(3b2)[33]; 76·5(1)[33]; III/76 + IV/9 : 5(3b5)[33]; 76,5(3b5)[33];
II/8 + IV/4 : 30(2c1)[44]; 40(2c1)[44]; 50(2c1)[44]; 60(2c1)[44];
II/20 + IV/2 : 0(3a + 2b(2))[5]; 5[5]; 10(3a + 2b(2))[5]; 15[5]; 20[5]; 25(3a + 2b(2))[5]; II/20 + IV/4 : 30(2c3)[44]; 40(2c3)[44]; 50(2c3)[44]; 60(2c3)[44];
II/39 + IV/2 : 25(2ab)[45];
II/48 + IV/4 : 30(2a + 3a(7))[44]; 40(2 + 3a(7))[44]; 50(2a)[44]; 60(2a)[44];
II/50 + IV/4 : 30(2b)[44]; 40(2b)[44]; 50(2b)[44]; 60(2b)[44];
II/57 + IV/2 : 20(3b5)[46]; 25[46]; 30[46]; 35(3b7)[46];
II/61 + IV/4 : 30(2c2)[44]; 40(2c2)[44]; 50(2c2)[44]; 60(2c2)[44];
II/64 + IV/2 : 20(2a)[5];
I/1 + I/5 : 161(3b1)[5]; I/1 + I/9 : 115(1)[5];
I/2 + I/12 : 25(3c5)[47];
I/3 + I/12 : 20[48]; 25(2b)[49]; 30[50];
I/6 + I/12 : 25(3c5)[51];
I/7 + I/12 : 30(3c5)[47];
I/8 + I/9 : 110(2a)[13]; 115(2a)[13]; 120(2a)[13];
I/10 + I/12 : 25(2b)[49];
I/11 + I/12 : 25(1)[52];
I/1 + II/31 : 122(3b4)[5]; 132(3b4)[5]; I/1 + II/34 : 135(3b1)[5]; 145(3b1)[5];
I/1 + II/40 : 80(3b6)[5]; 90(3b6)[5];
I/3 + II/8 : 25(2c1)[53]; I/3 + II/61 : 25(2b)[53];
I/4 + II/8 : 25(2c + 3b(3))[5]; I/4 + II/20 : 25(2a)[5]; 40(2a)[5]; I/4 + II/38 : 25(3a)[5]; 40(3a)[5]; I/4 + II/39 : 25(3b6)[5]; 40(3b7)[5]; I/4 + II/43 : 25(3b7)[5]; 40(3b7)[5]; I/4 + II/48 : 25(2a)[5]; 40(2a)[5]; I/4 + II/50 : 25(2c + 3b(3))[5]; I/4 + II/61 : 25(2a + 3a(4))[5];
I/8 + II/5 : 110(3b5)[13]; 115(3b5)[13]; 120(3b5)[13];

I/12 + II/2 : 25(2a + 3a(7))[54]; I/12 + II/3 : 25(3c5)[51]; I/12 + II/20 : 10[55]; 20(3c8)[15]; 40(3c7)[15]; 60(3c7)[15]; I/12 + II/22 : 70(3a)[56]; I/12 + II/39 : 10(3c5)[57]; 15[57]; 15[55]; 20[57]; 25[57]; 30[57]; 35[57]; 40[57]; 45[57]; 50[57]; 55[57]; 60[57]; I/12 + II/48 : 0[58]; 10(3c7)[58]; 10[14]; 20[15]; 20[14]; 25[59]; 30[59]; 30[14]; 35[59]; 40[15]; 40(3c6)[59]; 40[14]; 50[59]; 60[15]; I/12 + II/61 : 25(3c7)[19]; 25[51]; 25[60]; 30[60]; 35(3c7)[60]; 40[60]; 50(3c7)[60];

I/4 + III/29 : 25(3b2)[5]; 50[5]; 75(3b1)[5]; I/4 + III/39 : 25(2a + 3b9(7))[5]; 40(2a + 3b9(7))[5];

I/7 + III/37 : 40(2b)[61]; I/7 + III/46 : 40(2c2)[61];

I/12 + III/1 : 0(3c7)[62]; I/12 + III/2 : 0(2c + 3c7(3))[36]; 18(2c + 3c7(4))[36]; 25[63]; 40[63]; 73[36]; I/12 + III/4 : 0[58]; 10[58]; 20(2a + 3c8(4))[15]; 25[38]; 40(2a + 3c8(4))[15]; 60[15]; I/12 + III/17 : 25 (3c7)[47]; I/12 + III/25 : 25(3c7)[47]; I/12 + III/27 : 25(3c9)[64]; I/12 + III/29 : 25(3c7)[47]; I/12 + III/31 : 25(3c8)[47]; I/12 + III/34 : 25(3c7)[47]; I/12 + III/35 : 25(3c7)[47]; 25[65]; I/12 + III/37 : 25[66]; 25[38]; 25(3c7)[67]; 45[67]; 65[67]; I/12 + III/39 : 20(3c8)[68]; 25[68]; 25[63]; 30[68]; 40[68]; 40[63]; 60[68]; 80(3c7)[68]; I/12 + III/40 : 25(3c8)[47]; I/12 + III/45 : 25(3c9)[69]; I/12 + III/70 : 25(3c6)[47]; I/12 + III/72 : 25(3c7)[70]; I/12 + III/73 : 25(3c7)[70]; I/12 + III/74 : 25(3c7)[47]; 25[71]; I/12 + III/75 : 25(3c7)[72]; 35[72]; 50(3c7)[72]; I/12 + III/83 : 25(3c7)[70]; I/12 + III/84 : 25[73]; 25(3c8)[74]; I/12 + III/90 : 25(3c9)[75];

II/2 + II/20 : 20(2b)[5]; II/2 + II/48 : 25(2c8)[54];

II/4 + II/8 : 25(2c3)[8]; 30[8]; 35[8]; 40(2c4)[8]; II/4 + II/16 : 60(2a)[5]; 90(2a)[5]; II/4 + II/20 : 0(2a)[5]; 20(2a)[5]; II/4 + II/22 : 20(3b3)[76]; 30(3b3)[76]; 40[76]; 40[5]; 50[5]; 60[76]; 75[5]; 80(3b3)[76]; 100[5]; 125[76]; II/4 + II/24 : 30(3b6)[5]; II/4 + II/29 : 25(3a)[35]; II/4 + II/30 : 50(3b1)[5]; II/4 + II/55 : 30(2c7)[77]; 40[77]; 60[77]; 80(2c8)[77]; II/4 + II/64 : 20(3a)[5];

II/7 + II/18 : 20(3b3)[5]; II/7 + II/64 : 20(3b5)[5];

II/8 + II/10 : 20(2ab)[78]; II/8 + II/14 : 25(2ab)[79]; II/8 + II/50 : 25(2a)[80]; 35(2a)[81]; II/8 + II/51 : 20(2a)[82]; 25(2a)[80]; 40(2a)[82]; 60(2a)[82]; II/8 + II/61 : 25(2a)[80]; II/8 + II/62 : 25(2a)[80];

II/13 + II/22 : 25(2a)[9];

II/14 + II/64 : 15(3a + 2a(6))[83];

II/15 + II/68 : 150(3b7)[5];

II/16 + II/22 : 30(2a)[76]; 40[76]; 61[76]; 81(1)[76];

II/17 + II/60 : 20(3b8)[5];

II/18 + II/43 : 20(2a + 3a(5))[5]; II/18 + II/60 : 20(3b7)[5];

II/19 + II/42 : 130(2ab)[5];

II/20 + II/48 : 20(2a)[15]; 20[84]; 25[84]; 25[5]; 40(2a)[15]; 60(1)[15]; II/20 + II/61 : 25(1)[5]; II/20 + II/64 : 25(2c5)[5]; II/20 + II/65 : 20(2a)[5]; II/20 + II/66 : 20(2a)[5];

II/21 + II/22 : 40(3b2 + 2a(6))[5]; 50(3b2 + 2a(8))[5]; 75(3b1 + 2a(9))[5]; 100[5];

II/22 + II/28 : 25(3a)[5]; II/22 + II/29 : 25(2a)[5]; II/22 + II/30 : 25(2a)[5]; II/22 + II/32 : 60(2c + 3b(6))[5]; 75(2c + 3b(7))[5]; II/22 + II/38 : 50(2ab)[85]; 75(2a)[85]; II/22 + II/39 : 50(2a)[86]; 70(2a)[86]; 90(2a)[86]; II/22 + II/44 : 50[86]; 50(3b2)[5]; 60[5]; 75[5]; II/22 + II/49 : 40(3b7)[5]; 50(3b7)[5]; 75 (3b6)[5]; 100[5]; II/22 + II/53 : 50(3b1)[5]; II/22 + II/67 : 39,9(3b7)[76]; 59,9[76]; 79,8[76]; 99,9(3b7)[76]; 125[76]; 150[76]; 175[76];

II/23 + II/64 : 25(2b)[5];

II/24 + II/49 : 30(2a)[5]; II/24 + II/65 : 30(3b7)[5];

II/25 + II/48 : 25(2a)[87];

II/28 + II/29 : 25(2a)[5]; II/28 + II/30 : 25(2a)[5];

II/29 + II/30 : 25(2a)[5]; II/29 + II/38 : 20(2ab)[35]; II/29 + II/52 : 140(2a + 3a(4))[5]; II/29 + II/65 : 25(3b7)[35];

II/32 + II/45 : 50(2b)[88];

II/38 + II/39 : 25(3b2)[89]; 50(3b2)[89];

II/39 + II/44 : 25(3b2)[90]; II/39 + II/45 : 25(3b7)[88]; 50(3b7)[88]; II/39 + II/57 : 25(2ab)[79];

II/39 + II/61 : 25(2a + 3a(5))[79];

II/44 + II/45 : 50(2b)[88];
II/48 + II/69 : 20(3a)[5];
II/50 + II/51 : 20(2a)[82]; 40(1)[82]; 60(1)[82];
II/51 + II/62 : 20(2a)[82]; 40(2a)[82]; 60(2a)[82];
II/52 + II/56 : 140(3b6)[5];
II/57 + II/61 : 25(2c8)[79];
II/60 + II/64 : 20(3b5)[83]; 20[5];
II/61 + II/63 : 20(2b)[79];
II/1 + III/4 : 25(2b)[79];
II/2 + III/37 : 25(2a)[38]; II/2 + III/39 : 25(2ab)[91]; 35[91]; 45[91];
II/3 + III/4 : 25(2ab)[79];
II/4 + III/13 : 30(2a)[92]; II/4 + III/56 : 0(2b)[76]; 9,9[76]; 20,2[76]; 29,6[76]; 40[76]; 60[76]; 80(1)[76];
II/4 + III/76 : 25(2b)[5];
II/8 + III/15 : 25(2b)[10]; 35(2b)[10]; II/8 + III/39 : 25(2c3)[5]; II/8 + III/52 : 35(2b)[81]; II/8 + III/63 : 30(2b)[93]; 40[93]; 50[93]; 60(2a)[93];
II/12 + III/16 : 30(2a + 3a(5))[94]; II/12 + III/30 : 30(2a + 3a(6))[94]; II/12 + III/33 : 0[77]; 10(2a)[77]; 20[77]; 30[77]; 40(1)[77]; 60[77]; 80(1)[77]; II/12 + III/52 : 30(2a + 3a(5))[94]; II/12 + III/71 : 30(2a + 3a(5))[94];
II/14 + III/28 : 25(2a)[79]; II/14 + III/49 : 25(3b7)[79]; II/14 + III/52 : 25(2a)[79];
II/15 + III/87 : 150(3b8)[5];
II/16 + III/19 : 60(2ab)[5]; II/16 + III/21 : 60(3b7)[5]; 90(3b8)[5]; II/16 + III/77 : 97,5(2c3)[5];
II/17 + III/72 : 20(2a)[5]; II/17 + III/75 : 20(3b7)[5]; II/17 + III/83 : 20(2a)[5];
II/18 + III/75 : 20(3b7)[5]; II/18 + III/83 : 20(2a)[5];
II/20 + III/1 : 18(2a + 3b7(2))[5]; II/20 + III/4 : 25(2b)[16]; II/20 + III/8 : 25(2c5)[5]; II/20 + III/11 : 25(2c8)[16]; II/20 + III/12 : 30(2c6 + 3b10(8))[5]; II/20 + III/21 : 20(2a)[5]; II/20 + III/28 : 25(2b)[5]; II/20 + III/39 : 20(2c7)[5]; 25[5]; 25[91]; 35[91]; 40(2c7)[5]; 45[91]; 60(2c8)[5]; II/20 + III/41 : 20(2b)[84]; 25(2b)[84]; II/20 + III/42 : 25(2c8)[20]; II/20 + III/47 : 25(2a)[20]; II/20 + III/48 : 25(2c9)[20]; II/20 + III/52 : 20(2b)[84]; 25(2b)[84]; II/20 + III/56 : 25(2c5)[5]; II/20 + III/66 : 25(3b8)[70]; II/20 + III/83 : 25(2c3)[70];
II/21 + III/21 : 0(3b3)[5]; 20(3b4)[5]; 70(3b3)[5]; II/21 + III/83 : 0(3a)[5]; 20(3a)[5]; 70(1)[5];
II/22 + III/26 : 40(2a + 3b9(6))[5]; 50[5]; 75[5]; 100[5]; II/22 + III/33 : 9,8[76]; 20,1(2c1)[76]; 30[76]; 40[76]; 60[76]; 80(1)[76]; II/22 + III/36 : 40(2a + 3a(8))[5]; 50[5]; 75[5]; 100[5]; II/22 + III/80 : 97·5(2a)[5];
II/24 + III/4 : 30(2b)[5]; II/24 + III/36 : 30(2ab)[5];
II/26 + III/87 : 150(2ab)[5];
II/29 + III/13 : 40(2ab)[92];
II/32 + III/4 : 50(2b)[95];
II/37 + III/4 : 25(2a)[95]; 35[95]; 45[95];
II/38 + III/4 : 25(2b)[95]; 35[95]; 45(2ab)[95]; II/38 + III/28 : 0(2b)[96]; 25(2ab)[96]; II/38 + III/49 : 0(2b)[96]; 25(2ab)[96]; 50(2a)[96]; II/38 + III/52 : 25(2b)[97]; 35[97]; 45[97]; II/38 + III/71 : 25(2ab)[97]; 35[97]; 45[97]; II/38 + III/76 : 0(2c4 + 3b9(7))[89]; 25(2a + 3b9(8))[89]; 50(2a + 3b9(8))[89];
II/39 + III/2 : 15(2a)[5]; 76,5(2a)[5]; II/39 + III/3 : 25(3b2)[90]; II/39 + III/4 : 20(2a)[5]; 25[5]; 25[79]; 30[95]; 40[95]; II/39 + III/6 : 25(3b3 + 2a(8))[5]; 40(3b3 + 2a(8))[5]; 60[5]; II/39 + III/7 : 25(3b1 + 2c6(2))[5]; 40(2c6)[5]; 60(2c6)[5]; II/39 + III/15 : 25(3a + 2a(2))[5]; 40(3a + 2a(2))[5]; 60(3a + 2a(2))[5]; II/39 + III/21 : 20(3b7)[98]; II/39 + III/28 : 18(2b)[5]; 20[5]; 30[5]; 40(2ab)[5]; II/39 + III/37 : 25(3b6)[38]; II/39 + III/39 : 25(3b8)[99]; 40(3b8)[99]; II/39 + III/41 : 25(2a)[90]; 25[5]; 40(2a)[5]; 60(2a)[5]; II/39 + III/43 : 25(3a)[90]; II/39 + III/49 : 25(2a + 3a(9))[100]; 40(3a + 2a + 3a(2,8))[100]; II/39 + III/52 : 25(2a)[97]; 35[97]; 45[97]; II/39 + III/55 : 25(2c9)[100]; 40(2c9)[100]; 60(2c9)[100]; II/39 + III/58 : 40(3a)[5]; 50[5]; 60[5]; 70(3a)[5]; II/39 + III/71 : 25(2a)[97]; 35[97]; 45[97];
II/39 + III/76 : 20(2ab)[5]; 40[5]; 60[5]; 80(2a)[5]; II/39 + III/81 : 15(3b5)[5]; 20(3b5)[5];

II/44 + III/6 : 50[101]; 60(2ab)[101]; 70(2ab)[101]; II/44 + III/15 : 50(2b)[101]; 60(2ab)[101]; 70(2ab)[101]

II/44 + III/55 : 50(2b)[101]; 60(2ab)[101]; 70(2ab)[101];

II/48 + III/4 : 0(2a)[5]; 25(2a)[5]; 30[102]; II/48 + III/8 : 25(3a)[5]; II/48 + III/23 : 20(3a)[5]; II/48 + + III/28 : 25(2b)[5]; II/48 + III/37 : 25(2a)[38]; II/48 + III/39 : 20(2ab)[5]; 40(2a)[5]; II/48 + III/56 : : 25(3a)[5]; II/48 + III/76 : 25(2a)[5];

II/50 + III/26 : 0(2c3)[33]; 76,5(2a)[33]; II/50 + III/52 : 35(2b)[81]; II/50 + III/76 : 0(2c2)[33]; 80[33];

II/55 + III/21 : 30(3b5)[77]; 40[77]; 60[77]; 80(3b5)[77]; II/55 + III/53; : 40(2ab)[103]; II/55 + III/83 : : 30[77]; 40(2a + 3a(3))[77]; 60,1[77]; 80(2a + 3a(2))[77];

II/57 + III/4 : 20(2a)[79]; II/57 + III/41 : 20(2a)[104]; 25(2a)[104]; 30(2a)[104]; 35(2a)[104];

II/59 + III/81 : 0(2b)[33]; 76,5(2a)[33];

II/60 + III/41 : 30(2a)[5];

II/61 + III/82 : 25(2b)[30];

II/63 + III/4 : 20(2a)[79];

II/64 + III/5 : 20(2ab)[5]; II/64 + III/18 : 20(2c1)[5]; II/64 + III/21 : 20(3a)[83]; II/64 + III/36 : : 20(2a + 3a(5))[5]; 25(3a + 2a(5))[5]; II/64 + III/68 : 20(3b3)[5]; II/64 + III/72 : 20(3b6)[5]; II/64 + + III/76 : 20(2a)[5]; II/64 + III/83 : 15(3b2)[83]; 20(3b3)[5]; 30[5]; II/64 + III/89 : 20(2a + 3a(6))[5];

II/65 + III/76 : 11(2b)[5];

II/67 + III/80 : 97·5(2ab)[5];

II/68 + III/87 : 150(3a)[5];

III/1 + III/81 : 15(2a)[5];

III/2 + III/39 : 25(2b)[99]; 40(2ab)[99]; III/2 + III/81 : 10(3a)[33]; 76,5 (3b5)[33];

III/3 + III/75 : 25(3a + 2a(8))[105]; 50(3a + 2a(6))[105]; 75(3a + 1(5))[105];

III/4 + III/32 : 25(2a)[32]; III/4 + III/52 : 20(1)[82]; 40(1)[82]; III/4 + III/63 : 25(2a)[39]; III/4 + + III/65 : 25(3a + 2a(4))[9]; III/4 + III/71 : 20(1)[82]; 40(1)[82]; III/4 + III/75 : 25(2ab)[105]; 35 (2ab)[105]; 50(2a)[105]; III/4 + III/76 : 20(2a)[5]; III/4 + III/88 : 25(3a)[106]; 35(3a)[105]; 50(3a)[106];

III/5 + III/26 : 30(2a)[5]; III/5 + III/36 : 30(2a)[5];

III/9 + III/10 : 136(2b)[5];

III/13 + III/39 : 40(2a)[92];

III/14 + III/43 : 25(2a)[107];

III/15 + III/41 : 30(2a)[108]; 50(2a)[108]; 70(1)[108]; III/15 + III/59 : 20(1)[82]; 40(1)[82]; 60(1)[82];

III/15 + III/82 : 20(1)[82] : 40(1)[82]; 60(1)[82];

III/24 + III/42 : 25(2a)[79];

III/26 + III/76 : 11(2b)[5];

III/28 + III/49 : 25(1)[79]; III/28 + III/52 : 20(2a)[79]; III/28 + III/56 : 25(2a)[107];

III/32 + III/56 : 25(2a)[107];

III/36 + III/76 : 11(2a)[5]; 25(2c1)[5]; 77,5(1)[5];

III/37 + III/46 : 40(2c9)[61];

III/41 + III/43 : 25(2a)[107]; III/41 + III/55 : 11(2a)[5]; III/41 + III/60 : 11(2a)[5]; III/41 + III/67 : : 11(3a)[5]; 25(1)[5]; III/41 + III/76 : 25(2a)[109];

III/42 + III/83 : 25(2a)[20];

III/44 + III/50 : 20(1)[110]; 40(1)[110];

III/48 + III/83 : 25(2a)[20];

III/51 + III/61 : 20(1)[110]; 40(1)[110];

III/52 + III/71 : 20(1)[82]; 40(1)[82]; 60(1)[82]; III/52 + III/75 : 25(2ab)[105]; 50(2a)[105]; 75(2a)[105];

III/52 + III/88 : 25(3a)[106]; 50(3a)[106]; 75(3a)[106];

III/53 + III/77 : 60(2ab)[103]; 70(2ab)[103]; III/53 + III/79 : 60(2a)[103]; 70(2a)[103]; III/53 + III/83 : :20(2ab)[103]; 40(2a)[103]; 50(2a)[103];

III/54 + III/62 : 20(1)[110]; 40(1)[110]; III/54 + III/82 : 20(1)[110]; 40(1)[110];

III/57 + III/83 : 25(2a + 3a(8))[5];
III/71 + III/88 : 25(1)[106]; 50(1)[106]; 75(1)[106];
III/75 + III/76 : 25(1)[111]; 50(1)[111]; 75(1)[111]; III/75 + III/78 : 25(1)[111]; 50(1)[111]; 75(1)[111];
III/75 + III/79 : 50(1)[111]; 75(1)[111];
III/85 + III/87 : 150(1)[5];
III/3 + V/1 : 16(2a)[5]; III/3 + V/49 : 25(2a)[32];
III/4 + 1/V : 20(2a)[37]; 25(2a)[37]; 30(2a)[37]; III/4 + V/2 : 0(2a)[32]; III/4 + V/6 : 0(3a + 2a(5))[32];
III/4 + V/36 : 25(3b3 + 2c6 + 3b9(5,8))[112]; III/4 + V/47 : —13(2c6)[36]; —10[36]; 0(2c7)[36]; 10[5]; 15[5]; 15(2a)[36]; 20[5]; 25[5]; 35(2a)[36]; III/4 + V/50 : 0[32]; 20(2a)[15]; 40(2a)[15]; 60(2a)[15]; III/8 + V/1 : : 25(2a)[9]; 35(1)[9]; 45(1)[9]; 50(1)[9]; III/8 + V/15 : 20(2a + 3b9(6))[112]; 40(2c3 + 3b9(7))[112]; 60(2c4 + 3b8(7))[112]; 80(2c6 + 3b9(8))[112];
III/11 + V/1 : 25(2a)[9]; 35(2a)[9]; 45(2a)[9]; 50(2a)[9];
III/14 + V/1 : 15[113]; 25[113]; 25(2ab)[114]; 40[113]; 60[113];
III/15 + V/1 : 25(2ab)[10]; 35(2a)[10];
III/20 + V/7 : 110(2a)[13]; 115(2a)[13]; 120(2a)[13]; III/20 + V/26 : 110(2c5)[13]; 115(2c5)[13]; 120(2c5)[13];
III/21 + V/1 : 25(2a)[5];
III/22 + V/1 : 18(2a)[5];
III/26 + V/1 : 25(2a)[6];
III/28 + V/1 : 20(2a)[9]; 20[115]; 25[116]; 25[115]; 30[115]; III/28 + V/47; 12(2a)[5]; 20(2a)[5]; 25(2a)[5]; 25[109];
III/36 + V/1 : 25(1)[6]; III/36 + V/38 : 20(2a)[117]; 40(2a)[117]; 60(2a)[117]; III/36 + V/54 : 25(2a)[8]; 30(2a)[8]; 35(2a)[8]; 40(2a)[8];
III/38 + V/15 : 80(2a)[118]; III/38 + V/42 : 90(2a)[5];
III/39 + V/1 : 25(2a)[15];
III/41 + V/1 : 25[109]; 25(2a)[16]; III/41 + V/36 : 20(2ab)[79]; III/41 + V/47 : 25(2a)[109]; III/41 + + V/54 : 25(2ab)[109];
III/42 + V/1 : 25(2a)[20];
III/43 + V/1 : 25(2a)[114]; III/43 + V/54 : 25[109]; 25(2a)[114];
III/47 + V/1 : 25(2a)[20];
III/48 + V/1 : 25(2a)[20];
III/49 + V/1 : 20(2ab)[9]; 30(2a)[9]; 35(2a)[9];
III/52 + V/1 : 9,9[119]; 20(2a)[9]; 26[119]; 30(2a)[9]; 35(2a)[9]; 45(2a)[9]; III/52 + V/50 : 25(2a)[26];
III/53 + V/54 : 50(2c8)[103]; 60(2c8)[103]; 70(2c8)[103];
III/56 + V/33 : 0(2a)[32];
III/64 + V/46 : 179,8—182,7(2a)[120]; 189,9—190,2(2a)[120]; 209,0—210,8(2a)[120]; 229,7—230,3 (2a)[120];
III/65 + V/1 : 25(2c7)[9];
III/67 + V/1 : 25(2a)[5]; 35(2a)[5]; 45(2a + 3a(7))[5];
III/69 + V/35 : 25(2a)[121]; III/69 + V/36 : 25(2ab)[121]; III/69 + V/51 : 25(2a)[121];
III/71 + V/1 : 25(2a)[25];
III/72 + V/2 : 20(3a)[117]; 40(3a)[117]; 60(1)[117]; III/72 + V/54 : 20(2a)[117]; 40(2a)[117]; 60(2a)[117];
III/75 + V/2 : 25(2a)[43]; 50(2a + 3a(8))[43]; 75(1 + 3a(8))[43]- III/75 + V/6 : 25(2a)[43]; 50(2a)[43]; 75(1)[43]; III/75 + V/50 : 25(2a)[43]; 35(2a)[43]; 50(1)[43];
III/76 + V/1 : 20(2a)[5]; III/76 + V/2 : 25(2a)[5]; 30[5]; 35(2a)[5]; III/76 + V/15 : 20(3b8)[118]; 40(3b9)[118]; 60(3b9)[118]; 80(3a)[118]; III/76 + V/36: 15(2ab)[5]; III/76 + V/42 : 80(2a)[5]; III/76 + + V/54 : 25(2a)[5]; 30(2a)[5]; 35(2a)[5];
III/77 + V/42 : 97·5(2a)[5];
III/81 + V/1 : 20(2a)[5];
III/82 + V/1 : 25(1)[30]; III/82 + V/34 : 25(2ab)[79];

III/83 + V/1 : 20(1)[117]; 25(1)[70]; 40(1)[117]; 60(1)[117];
III/86 + V/1 : 20(2c1)[122];
III/91 + V/42 : 152(2b)[5];
IV/3 + IV/4 : 25(2a)[123];
IV/7 + IV/10 : 25(2a)[5];
IV/2 + V/1 : 25(2a + 3a(5))[38]; IV/2 + V/50 : 25(2a)[109]; 25,01[124];
IV/3 + V/1 : 25(2a)[5]; 45(2a)[5]; 65(2a)[5]; IV/3 + V/50 : 25(2a)[5];
IV/4 + V/1 : 0(2c3)[36]; 19,4(2c2)[36]; 25[38]; 50(2a)[36]; IV/6 + V/1 : 20(2a)[37]; 25[37]; 30(2ab)[37]; IV/6 + V/54 : 20(3a + 2a(5))[37]; 25[37]; 30(3a + 2a(5))[37]; IV/6 + V/56 : 20(1)[37]; 25[37]; 30(3a + 1(5))[37]; IV/6 + V/57 : 20(1)[37]; 25[37]; 30(3a + 1(8))[37]; IV/6 + V/58 : 20(3a + 1(6))[37]; 25[37]; 30(3a + 2a(6))[37];
IV/8 + V/5 : 25(2ab)[125]; 30(2ab)[125]; 35(2ab)[125]; 40(2ab)[125];
V/1 + V/2 : 25(1)[9]; 50(1)[9]; V/1 + V/3 : 25(2c5)[9]; 35(2c5)[9]; 45(2c5)[9]; 50(2c5)[9]; V/1 + V/6 : 25(1)[9]; 35[9]; 45[9]; 50(1)[9]; V/1 + V/8 : 25(2a)[9]; 50(2a)[9]; V/1 + V/22 : 6[5]; 9,8[126]; 20(2c7)[15]; 23,6[126]; 25[9]; 25[127]; 25[38]; 39·8[126]; 40(2c8)[15]; 60(2ab)[15]; V/1 + V/23 : 35(2a)[128]; V/1 + V/28 : 29,7(2a)[124]; V/1 + V/32 : 20[129]; 25(2c9)[9]; 25[5]; 30[129]; 35(2a)[9]; 40[129]; 45[9]; 45[5]; 50[9]; 50(1)[129]; 60[129]; 65[5]; 70[129]; V/1 + V/34 : 20(2b)[15]; 40(2ab)[15]; 60(2a)[79]; V/1 + V/35 : 25(2a)[121]; V/1 + V/36 : 20[15]; 25(2b)[121]; 25[9]; 25[127]; 40[15]; 60[15]; V/1 + V/37 : 25(2a)[9]; 50(2a)[9]; V/1 + V/38 : 10(2a)[129]; 20[129]; 30[129]; 40(1)[129]; 50[129]; 60[129]; 70(1)[129]; V/1 + V/47 : 20[5]; 25(1)[109]; 40[5]; V/1 + V/50 : 0[14]; 10[41]; 20[41]; 20[14]; 25(2a)[121]; 25[38]; 25[109]; 30[41]; 40[41]; 40[14]; 50[41]; 60[41]; 60[14]; V/1 + V/52 : 25(2a)[123]; V/1 + V/53 : 20(2c4)[84]; 25(2c3)[84]; V/1 + V/54 : 20(2c8)[15]; 20[129]; 25[109]; 30[129]; 40[129]; 50[129]; 60[129]; 60[15]; 70[129]; V/1 + V/56 : 10(2a)[129]; 20[129]; 30(2a)[129]; 40(2a)[129]; 50[129]; 60[129]; 70(2a + 3a(7))[129]; V/1 + V/58 : 20(2c5)[129]; 30[129]; 40[129]; 50(2a)[129]; 60[129]; 60(1)[129];
V/2 + V/6 : 20(2a)[5]; 40(2a)[5]; 60(1)[5]; V/2 + V/54 : 20(2a)[117]; 25[5]; 30[5]; 40(2a)[117]; 60(2a)[117];
V/4 + V/35 : 25(2a)[121]; V/4 + V/36 : 25(2a)[121]; V/4 + V/51 : 25(2a)[121];
V/6 + V/50 : 25(2ab)[38]; V/6 + V/54 : 20(1)[117]; 40(1)[117]; 60(1)[117];
V/7 + V/17 : 110(3a + 2c9(4))[13]; 115(3a + 2c9(4))[13]; 120(2c9)[13]; V/7 + V/26 : 110(2a)[13]; 115(2a)[13]; 120(2a)[13];
V/8 + V/35 : 25(2a)[130];
V/9 + V/35 : 25(2a)[130];
V/10 + V/35 : 25(2a)[130];
V/11 + V/35 : 25(2a)[130];
V/12 + V/35 : 25(2a)[130];
V/15 + V/16 : 20(2a)[117]; 40(2a)[117]; 60(2a)[117];
V/18 + V/35 : 25(2a)[130];
V/19 + V/35 : 25(2a)[130];
V/20 + V/35 : 25(2a)[130];
V/21 + V/35 : 25(2a)[130];
V/22 + V/24 : 25(2a)[123]; V/22 + V/25 : 25(2c5)[131]; V/22 + V/36 : 25[9]; 25(2b)[9]; 25[131]; V/22 + V/50 : 25(2c6 + 3b9(8))[132]; 25[21]; V/22 + V/52 : 25(2a)[123];
V/23 + V/24 : 35(2a)[128]; V/23 + V/50 : 35(2a)[128]; V/23 + V/52 : 35(2a)[128];
V/24 + V/28 : 24,8(2c7)[126]; 50[126]; V/24 + V/52 : 35(2a)[128];
V/25 + V/55 : —15(2a)[133]; 1[133]; 12[133]; 25(2a)[133];
V/27 + V/48 : 98,89(2a)[134];
V/29 + V/50 : 0(2a)[135];
V/30 + V/36 : 25(2a)[136]; V/30 + V/44 : 25(1)[137];
V/32 + V/54 : —20(3a + 2a(7))[129]; —10(3a + 2a(2))[129]; 0(2c1)[129]; 10(2a)[129]; 20[129]; 30[129]; 40(1)[129]; 50[129]; 60[129]; 70(3a)[129];

V/34 + V/35 : 25(2a)[136]; V/34 + V/36 : 25(1)[133]; 12[133]; 1[133]; —15(2a)[133]; V/34 + V/39 : : 20(2a)[84]; 25(2a)[84]; V/34 + V/44 : 20(1)[138]; 40(1)[138]; 60(1)[138]; V/34 + V/53 : 20(2a)[138]; 25[84]; 40(2a)[138]; 60(2a)[138]; V/34 + V/54 : 20(2ab)[84]; 25(2ab)[84];
V/35 + V/36 : 25(2a)[121]; 25[139]; V/35 + V/50 : 25(3a)[121]; V/35 + V/51 : 25(2a)[121];
V/36 + V/50 : 25[121]; 25(2b)[136]; 25[31]; V/36 + V/51 : 25(2a)[121]; V/36 + V/54 : 25(2ab)[31];
V/38 + V/54 : —20(1)[129]; —10[129]; 0[129]; 10(1)[129]; 20[129]; 30[129]; 40[129]; 50[129]; 60[129]; 70[129]; 80(1)[129];
V/40 + V/47 : 0(2a)[41]; 20(2a)[41];
V/41 + V/50 : 25(2a)[31];
V/43 + V/44 : —15(2a)[133]; 1[133]; 12[133]; 25(2a)[133];
V/44 + V/53 : 20(2a)[138]; 40(2a)[138]; 60(2a)[138];
V/47 + V/54 : 0(2c8)[5]; 20[5]; 25(1)[109]; 25[31]; 40[5];
V/49 + V/50 : 25(2c6)[5];
V/50 + V/54 : 20(2a)[15]; 40(2a)[15]; 60(2a)[15]; V/50 + V/58 : 25(2a)[31];
V/53 + V/54 : 20(2c7)[84]; 25(2c8)[84];
V/54 + V/56 : —20(1)[129]; —10[129]; 0[129]; 10(2a)[129]; 20[129]; 30[129]; 40[129]; 50[129]; 60[129]; 70[129]; 80(1)[129]; V/54 + V/58 : 20(2a)[129]; 25[31]; 30[129]; 40[129]; 50(1)[129]; 60[129]; 70[129]; 80(1)[129];

The coefficients of Eqs (*14*), (*18*) and (*31*) were calculated by two alternative methods: *a*) with the assumption that the volume change due to mixing the components is equal to zero (method I), and *b*) with the volume change taken into account (method R). The latter was applied only to 451 systems for which the dependence of the density on the composition was known.

To simplify the evaluation of results, the liquids were divided into five classes according to Ewell and coworkers[2]. All binary systems were then divided into 13 combinations of these classes and these in turn into five groups based on the ability to change the character of bonds by hydrogen bridges on mixing the components. The statistical evaluation was done for each isothermal system, for each combination of classes, and for each group of systems separately. The purpose of this procedure was to find out how is the accuracy of the correlation equations influenced by the change in the character of the bonds by hydrogen bridges due to mixing the components.

We calculated for each experimental result the relative deviation $\varepsilon = [(\eta_{\text{cal}} - \eta_{\text{exp}})/\eta_{\text{exp}}] \cdot 100\%$, further the mean relative deviation and standard deviation for each isothermal system, combination of classes, and group of systems:

$$\bar{\varepsilon} = \frac{1}{n} \sum_{i=1}^{n} |\varepsilon_i| \,, \quad \delta = \left(\frac{1}{n - k} \sum_{i=1}^{n} \varepsilon_i^2 \right)^{1/2} .$$

We denote η_{exp} and η_{cal} the measured and calculated viscosities of the mixture, n number of experimental data in a given set, and k number of coefficients in the correlation equation. Only systems with more than five experimental points were considered in the final evaluation for a given combination of classes or group of systems. Such systems were 332 in number, representing 725 isothermal systems or 6724 experimental points.

The results of the statistical evaluation and the coefficients of Eqs (*1*), (*14*), (*18*), and (*31*) for each isothermal system are given in ref.[3]. The results of statistical evaluation and comparison of the accuracy of the correlation equations within the individual combinations of classes are given in Table I. For the combination of classes IV–IV, only two systems were correlated (11 points), therefore no statistical evaluation is given. The value of $\bar{\varepsilon}$ was smaller than 1% in both systems for all four correlation equations.

TABLE I

Statistical Evaluation of Systems and System Groups (method R/method I)

Eq.[a]	No of systems	No of isoth. systems	No of exp. points	δ, %[b]	$\bar{\varepsilon}$, %	ε_{max}, %
			Systems II–V			
(*14*)	42/52	110/129	1 064/1 241	1·44/1·43	1·03/1·02	9·57/9·59
(*31*)	42/52	110/129	1 064/1 241	1·68/1·67	1·22/1·21	9·80/9·81
(*18*)	42/52	110/129	1 064/1 241	1·05/1·05	0·663/0·661	9·97/9·97
(*1*)	52	129	1 241	1·19	0·813	9·15
			Systems III–IV			
(*14*)	4/6	6/15	47/111	1·92/1·35	1·26/0·819	5·98/6·55
(*31*)	4/6	6/15	47/111	1·83/1·33	1·22/0·846	5·64/6·39
(*18*)	4/6	6/15	47/111	1·49/1·03	0·911/0·598	3·50/6·65
(*1*)	6	15	111	1·22	0·796	7·50
			Systems II–IV			
(*14*)	7/8	25/26	211/227	0·954/0·950	0·694/0·698	5·09/5·18
(*31*)	7/8	25/26	211/227	1·10/1·13	0·827/0·853	5·29/5·36
(*18*)	7/8	25/26	211/227	0·570/0·605	0·402/0·428	2·91/2·91
(*1*)	8	26	227	0·720	0·508	4·74
			Systems I–I			
(*14*)	6/8	7/11	57/87	2·88/4·99	2·15/2·68	7·83/27·5
(*31*)	6/8	7/11	57/87	2·01/3·28	1·20/1·73	10·2/19·3
(*18*)	6/8	7/11	57/87	1·59/2·95	0·864/1·40	5·39/12·8
(*1*)	8	11	87	4·66	3·00	21·9
			Systems I–II			
(*14*)	15/18	33/51	298/504	2·15/2·27	1·47/1·57	11·0/11·5
(*31*)	15/18	33/51	298/504	4·50/4·35	3·03/2·89	25·8/26·7
(*18*)	15/18	33/51	298/504	1·86/1·84	1·10/1·18	12·5/13·4
(*1*)	18	51	504	7·07	4·95	21·3
			Systems I–III			
(*14*)	22/24	41/46	454/492	10·1/10·5	6·45/6·57	73·9/76·2
(*31*)	22/24	41/46	454/492	15·9/16·7	10·6/10·9	89·0/94·9
(*18*)	22/24	41/46	454/492	6·39/7·28	3·65/4·03	44·6/49·1
(*1*)	24	46	492	15·3	10·0	100·1

TABLE I
(*Continued*)

Eq.^{a}	No of systems	No of isoth. systems	No of exp. points	δ, %^{b}	$\bar{\varepsilon}$, %	ε_{max}, %
Systems II–II						
(*14*)	33/42	77/87	766/862	2·09/1·98	1·31/1·21	12·1/12·1
(*31*)	33/42	77/87	766/862	2·04/1·92	1·28/1·18	10·7/11·6
(*18*)	33/42	77/87	766/862	1·27/1·21	0·76/0·715	7·66/7·70
(*1*)	42	87	862	1·65	1·02	10·0
Systems II–III						
(*14*)	62/72	146/161	1 388/1 540	1·83/2·12	1·06/1·20	31·0/32·9
(*31*)	62/72	146/161	1 388/1 540	2·10/2·34	1·26/1·38	16·0/17·0
(*18*)	62/72	146/161	1 388/1 540	1·28/1·50	0·595/0·679	28·5/30·1
(*1*)	72	161	1 540	2·84	1·26	30·5
Systems III–III						
(*14*)	20/22	37/39	312/328	1·44/1·38	0·973/0·919	9·29/9·20
(*31*)	20/22	37/39	312/328	1·35/1·33	0·930/0·896	5·48/5·48
(*18*)	20/22	37/39	312/328	1·05/1·02	0·738/0·706	4·63/4·63
(*1*)	22	39	328	1·26	0·868	5·66
Systems III–V						
(*14*)	19/31	43/68	348/531	1·08/1·01	0·711/0·649	5·87/6·19
(*31*)	19/31	43/68	348/531	1·07/1·01	0 701/0·649	6·14/6·32
(*18*)	19/31	43/68	348/531	0·848/0·807	0·516/0·496	5·57/5·71
(*1*)	31	68	531	0·971	0·622	7·57
Systems IV–V						
(*14*)	1/6	3/18	27/153	0·517/0·359	0·356/0·270	1·61/1·67
(*31*)	1/6	3/18	27/153	0·705/0·398	0·506/0·288	1·69/1·72
(*18*)	1/6	3/18	27/153	0·457/0·282	0·367/0·211	1·13/1·14
(*1*)	6	18	153	0·333	0·250	1·63
Systems V–V						
(*14*)	26/42	47/73	449/642	0·562/0·509	0·364/0·329	3·92/3·96
(*31*)	26/42	47/73	449/642	0·610/0·546	0·386/0·344	4·57/4·61
(*18*)	26/42	47/73	449/642	0·365/0·334	0·236/0·219	2·03/2·05
(*1*)	42	73	642	0·506	0·320	4·31

TABLE I

(*Continued*)

Eq.[a]	No of systems	No of isoth. systems	No of exp. points	δ, %[b]	$\bar{\varepsilon}$, %	ε_{max}, %
Group of systems I–I, I–II, I–III, II–II and II–III						
(*14*)	138/164	304/356	2 963/3 485	4·36/4·43	2·01/2·05	73·9/76·2
(*31*)	138/164	304/356	2 963/3 485	6·64/6·76	2·87/2·90	89·0/94·9
(*18*)	138/164	304/356	2 963/3 485	2·79/3·08	1·16/1·25	44·6/49·1
(*1*)	164	356	3 485	6·68	3·02	100·1
Group of systems III–III, III–V, IV–IV, IV–V and V–V						
(*14*)	67/102	131/199	1 142/1 660	1·02/0·899	0·635/0·542	9·29/9·20
(*31*)	67/102	131/199	1 142/1 660	1·00/0·895	0·633/0·545	6·14/6·32
(*18*)	67/102	131/199	1 142/1 660	0·756/0·679	0·461/0·403	5·57/5·71
(*1*)	102	199	1 660	0·849	0·518	7·57

[a] Numbers of equations refer to the preceding work[1]. [b] Standard deviation.

A comparison of Eqs (*14*) and (*31*) involving two coefficients with Eq. (*1*) leads to the following conclusions: *1*) Eq. (*1*) gives the smallest deviations for the group of systems whose formation is not accompanied by a change in the hydrogen bonds (III–III, III–V, IV–IV, IV–V, V–V), while Eqs (*14*) and (*31*) give only somewhat larger deviations, which, however, plays almost no role from the point of view of their accuracy (Table I). *2*) For the group of systems, in which splitting of the bonds by hydrogen bridges (II–V, II–IV) or the formation of new bonds (III–IV) prevails, Eq. (*1*) gives better results than (*14*) and (*31*), this improvement being more marked than in the preceding case (Table I). *3*) For the complicated group of systems in which the hydrogen bonds are at the same time formed and splitted (I–I, I–II, I–III, II–II, II–III), Eq. (*14*) gives the smallest deviations, while the accuracy of (*31*) is about the same as that of (*1*). The improvement in using Eq. (*14*) against the other two is considerable (Table I). *4*) Eq.(*14*) gives the smallest deviations for all binary systems, while (*31*) is about as good as (*1*). This conclusion remains unaltered if we omit from all binary systems the combination of classes I–III, which are most difficult to correlate (Table II). *5*) A comparison of the distribution functions for mean values of relative deviations for all isothermal systems (725) having more than 5 experimental points shows that Eq. (*1*) gives small relative deviations with a larger probability than (*14*) and (*31*) (Fig. 1). The intersection of the distribution curves for Eqs (*1*) and (*14*) corresponds to $\bar{\varepsilon} = 1{\cdot}7\%$ and for Eqs (*1*) and (*31*) to $\bar{\varepsilon} = 2{\cdot}6\%$. It follows that Eq. (*14*) is about as accurate as the equation of McAllister (*1*), moreover its coefficients are in a linear form and its form is simpler. Eq. (*31*) is less advantageous than (*14*).

An improvement resulting from the use of Eq. (*18*) (four-molecular model) as compared with (*14*) (three-molecular model) is obvious from Table I and II. The standard deviation becomes lowér by 24 – 36% and the mean relative deviation by 26 – 40%. The lower limits of these intervals refer to the systems where the hydrogen bonds do not change and the upper limits to systems where simultaneous splitting and formation of hydrogen bonds takes place.

TABLE II
Statistical Evaluation of All Systems (method R/method I)

Eq.[a]	No of systems	No of isoth. systems	No of exp. points	δ, %[b]	$\bar{\varepsilon}$, %	ε_{max}, %
All binary systems						
(*14*)	258/332	576/725	5 427/6 724	3·33/3.29	1·47/1·42	73·9/76·2
(*31*)	258/332	576/725	5 427/6 724	4·99/4·93	1·98/1·77	89,0/94·9
(*18*)	258/332	576/725	5 427/6 724	2·12/2·27	0·788/0·795	44·6/49·1
(*1*)	332	725	6 724	4·84	1·75	100·1
All binary systems except for combinations of classes I–III						
(*14*)	236/308	535/679	4 973/6 232	1·65/1·76	1·02/1·02	31·0/32·9
(*31*)	236/308	535/679	4 973/6 232	2·01/2·04	1·19/1·05	25·8/26·7
(*18*)	236/308	535/679	4 973/6 232	1·10/1·18	0·526/0·540	28·5/30·1
(*1*)	308	679	6 232	2·64	1·09	30·5
Ternary systems during whose formation the hydrogen bonds do not change						
(*16*)	12/13	14/15	175/210	0·618/0·628	0·427/0·442	3·78/3·52
(*33*)	12/13	14/15	175/210	0·621/0·639	0·421/0·440	3·64/3·59
(*2*)	13	15	210	0·686	0·515	2·56
All ternary systems						
(*16*)	17/18	21/22	342/377	2·06/1·93	1·19/1·11	10·9/11·0
(*33*)	17/18	21/22	342/377	3·10/2·96	1·63/1·53	18·1/18·0
(*2*)	18	22	377	2·13	1·18	14·2

[a] Numbers of equations from ref.[1]. [b] Standard deviation.

It is apparent from these results that Eq. (*18*) is most accurate of all the equations used, and this for each combination of classes, group of systems, and all binary systems (Tables I and II, Fig. 1).

A comparison of the accuracy of Eqs (*14*), (*18*), and (*31*) for the methods of calculation R and I reveals that the error in calculating their coefficients due to neglecting the volume changes is negligible for most of the binary systems (Tables I and II). A small but systematic drop in the accuracy was observed with systems I–III and some systems I–II (water–lower aliphatic alcohols). In these cases, however, the volume changes during mixing are large (up to 10%).

Ternary Systems

Eqs (*16*) and (*33*) (ref.[1]) were tested and compared with (*2*) on the basis of measured viscosities of ternary mixtures. 18 systems were correlated, corresponding to 22 isothermal systems or 377 experimental points. The systems under consideration were as follows:

I/12 + III/2 + III/39: 25[63]; 40[63];
II/8 + III/15 + V/1: 25[10]; 35[10];
II/39 + III/4 + V/1: 25[140]; II/39 + III/71 + V/1: 25[25];
II/61 + III/82 + V/1: 25[30]; III/69 + V/35 + V/36: 25[141];
III/69 + V/35 + V/51: 25[141]; III/69 + V/36 + V/51: 25[141];
V/1 + V/22 + V/36: 25[127]; V/1 + V/35 + V/36: 25[141];
V/1 + V/35 + V/50: 25[141]; V/1 + V/36 + V/50: 25[141];

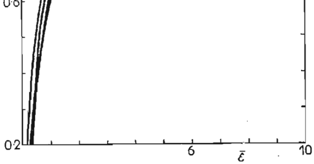

FIG. 1

Distribution Functions of Mean Values $\bar{\varepsilon}$ (%) from Absolute Values of Relative Deviations

1 Eq. (*31*); 2 Eq. (*14*); 3 Eq. (*1*); 4 Eq. (*18*); numbers of equations refer to the preceding work[1].

V/4 + V/35 + V/36: 25^{141}; V/4 + V/35 + V/51: 25^{141};
V/4 + V/36 + V/51; 25^{141}; V/34 + V/44 + V/53: 20^{138}; 40^{138}; 60^{138};
V/35 + V/36 + V/50: 25^{141}; V/35 + V/36 + V/51: 25^{141}.

Similarly to binary mixtures, also here the ternary coefficients and the viscosities from *(16)* and *(33)* were calculated by the methods R and I. The correlation equations *(2)*, *(16)*, and *(33)* were compared for systems in which the hydrogen bonds did not change during mixing the components, and also for all systems under consideration separately (Table II). It is seen from Table II that neglecting the excess volumes has no appreciable influence on the accuracy of Eqs *(16)* and *(33)*. These give approximately the same deviations for systems where the hydrogen bonds do not change, whereas Eq. *(2)* gives somewhat larger deviations. In comparing all systems, Eqs *(16)* and *(2)* are nearly equally accurate and *(33)* gives somewhat larger deviations. Eqs *(16)* and *(33)* are, however, simpler and linear with respect to their coeficients.

Analysis of Viscosity Curves

The simplest case would be if the form of the viscosity curve were determined uniquely by the correspondence of the binary system to a certain combination of classes, introduced by Ewell and coworkers[2]. In reality, however, the relations are more complex. For practical purposes, it would be desirable to know for which systems is the form of the viscosity curve given by the correspondence to a certain class combination, or to estimate the course of the viscosity curve for other systems.

To this end, we analysed the viscosity curves of all binary systems under study. Within a given class combination, we sought the relation between the form of the viscosity curve and the chemical nature of the compounds of the mixture, eventually a correlation between the sign of the viscosity deviations (deviations of the viscosity of the mixture from the straight line joining the points representing the viscosity of the components) and of the excess volumes. The latter were calculated from the densities of the binary mixtures published together with their viscosities. In most cases (so far as possible), the viscosity curves were compared at the laboratory temperature, in other cases at the temperature at which the viscosity was measured.

According to the form of the viscocity curve, the analysed binary systems were divided into four basic groups as follows: *1)* Quasiideal systems, *i.e.*, systems with curves which coincide within the range of experimental errors with the straight line joining the viscosities of pure components. *2)* Systems with negative deviations, *i.e.*, those whose viscosity is in the whole concentration range lower than would correspond to the mentioned straight line. These can be divided into three subgroups *a)* The negative deviations are small and the curve is practically symmetrical with respect to the straight line joining the viscosities of pure components. *b)* The negative deviations are large and the curve is unsymmetrical; it initially decreases abruptly toward the concentration axis, becomes rounded and often parallel to the concentration axis approaching the viscosity axis. *c)* The curve passes through a minimum. We introduce the symbols, *e.g.*, 2*c*6 denotes a curve of the type 2*c* with a minimum close to the point $x_1 = 0{\cdot}6$. Some systems have curves intermeditae between types 2*a* and 2*b*. Their deviations are usually larger than with the type 2*a*, but the curves remain symmetrical. They are denoted as 2*ab*. *3*) Systems with positive deviations, *i.e.*, those whose viscosity is in the whole concentration range higher than would correspond to the straight line joining the viscosity points of the components. These can be divided into three subgroups: *a*) All points of the curve lie between the values of the component viscosities, the curve has no maximum. *b*) Nonaqueous solutions whose dependence of the viscosity on composition shows constantly positive deviations and passes through a maximum. *c*) Aqueous solutions with a dependence of the viscosity as in the preceding case. The magnitude of the positive deviations, the sharpness of the maximum, and its shift toward pure water increase in the order of systems I–I, I–II, I–III. *4*) Systems with both negative and positive deviations: their viscosity curve intersects at least once the straight line joining the viscosities of pure components. In a certain concentration range they behave usually as systems of group *2*, while at other concentrations as systems of group *3*. We introduce symbols, *e.g.*, 2*c*2 + 3*b*9(*4*) denotes that the viscosity curve has in the concentration range from zero to $x_1 = 0{\cdot}4$ the form 2*c*, at $x_1 = 0{\cdot}2$ passes through a minimum, at $x_1 = 0{\cdot}4$ intersects the line joining the viscosity points of the components, and in the interval $x_1 = 0{\cdot}4 - 1{\cdot}0$ has the form 3*b* with a maximum at $x_1 = 0{\cdot}9$.

Symbols of the curve types for the analysed systems are given in parentheses in the survey of the binary systems under study.

Systems II–V. The systems belonging to the combination of classes II and V are expected to have positive deviations from the Raoult's law, since the hydrogen bonds between the molecules of substance II are splitting during the formation of the solution. By analogy, it can be expected that these systems will have viscosity curves of group *2*. This applies for 76·8% of the analysed systems. These are: Aliphatic carboxylic acids (C_2—C_7)-benzene, aliphatic alcohols (C_2, 1-C_3, 1-C_4, 1-C_5)-cyclohexane (all have positive excess volumes), and aliphatic alcohols (C_2, 1-C_3, 1-C_4)-benzene, toluene, or mesitylene. Their viscosity curves has the form 2*b*, which practically does not change with changing number of C atoms in the acid or alcohol. With the acid and alcohol C_2, sometimes also C_3, a small, flat minimum appears on this curve. The systems methanol-*m*-xylene or mesitylene, which have a 3*b* type curve, form an exception. In the region of the formation of a maximum, negative excess volumes were observed.

Mixtures of aliphatic carboxylic acids (C_2, C_4—C_7) with CCl_4 have curves mostly of the type 2*a*. The excess volumes are positive, only at higher concentrations of the acid beginning with C_4 they become negative. Aliphatic alcohols with a linear chain (C_1—C_8) with CCl_4 give curves of group *4*. Positive deviations occur in mixtures richer on alcohol and their interval decreases with increasing number of C atoms in the alcohol, reaching zero for n-octanol. With increasing temperature, the interval of the positive deviations diminishes until they become negative. The sign of the excess volume is positive for negative deviation of viscosity and *vice versa*, A small, flat minimum is observed also in mixtures of CCl_4 with and acid and alcohol C_2.

A pronounced change occurs if a normal aliphatic alcohol is replaced by an isomeric one with a branched chain. For example, with mixtures of an alcohol with CCl_4, replacing n-butanol with isobutanol or 2-methylpropane-1-ol causes a change of the viscosity curve from type *4* to *2c*, the negative deviations being very large. In mixtures of normal aliphatic alcohols with CCl_4, positive or small negative deviations occur thanks to the fact that the linear alcohol molecules and spherical CCl_4 molecules can form a structure with a higher space density than in the case where the alcohol molecules are branched. Since positive deviations occur in solutions richer in alcohol, the central molecules of the formed clusters will be CCl_4, the alcohol molecules forming their envelope. With branched alcohols molecules, a steric hindrance takes place in forming such clusters or they cannot be formed at all, whereby the change of the curve can be elucidated. In accord with this, the mentioned change did not occur on passing from the system 1-pentanol–n-hexane to 3-methylbutane-1-ol–n-hexane, since none of them has molecules of a shape enabling to form a mixtur of a higher density. The curves of both systems are of the type *2b*.

Large negative deviations of the viscosity and curves mostly of the type *2b*, sometimes *2ab*, are obtained with systems both components of which have an aromatic character.

Mixtures of acetonitrile with an aromatic hydrocarbon exert a rather anomalous behaviour with respect to the class combination II–V. For benzene, the curve has small negative and positive deviations, but for toluene and xylene only small positive ones. The sign of the excess volume is always opposite with respect to that of the viscosity deviations. The influence of dissociation of acetonitrile after dissolution is weaker than with alcohols and carboxylic acids and interactions between unequal molecules prevail.

Systems III–IV. The systems belonging to the combination of classes III and IV should exert negative deviations from Raoult's law owing to the formation of a hydrogen bond between the electronegative atom of component III and active hydrogen of component IV. Hence, viscosity curves of type *3* could be expected. This, however, is true only for 9 systems (7 of which being of type *3b*), whereas 8 systems have curves of type *2* (7 of which of type *2a*). A possible explanation is that the effect of the formation of weaker bonds by hydrogen bridges overlaps with the effect of disintegration of a more organized structure of compound III on forming a solution with compound IV. The latter effect, which causes negative deviations of the viscosity, prevails in systems, the viscosities of whose components differ much from each other, whereas the effect of the hydrogen bonds prevails in systems, the viscosities of whose components are similar. For all 8 systems with negative deviations, the ratio of viscosities of pure components was in the interval 2·0 – 12;0; two systems with positive deviations (curve *3a*) had these ratios equal to 1·4 and 2·2, and 7 systems with a maximum on the viscosity curve *3b* had them in the interval 1·0 – 1·9.

Systems II–IV. In the formation of a solution of the type II–IV, the effect of dissociation of component II usually prevails over that due to the formation of weaker hydrogen bonds between unequal molecules. These systems have therefore positive deviations from Raoult's law and they are expected to give viscosity curves of type *2*. This is true for 6 systems in the whole concentration range and two systems in 70 to 80% of the whole concentration range. Positive excess volumes were found with all of them in the region of negative viscosity deviations. Only the system nitromethane–chloroform exerted small positive deviations of the viscosity with a maximum on the curve. The hydrogen bonds in pure nitromethane are probably weaker than *e.g.*, in alcohols or in carboxylic acids, which constitute systems with negative viscosity deviations. On dissolving in chloroform, bonds between unequal molecules are formed which can play a significant role.

Systems I–I. These are very complicated and their viscosity deviations are difficult to predict. On the basis of an analysis of a rather small number of systems, it can only be concluded that all four systems with positive viscosity deviations have negative excess volumes.

Systems I–II. These are also complicated since the formation of a solution is accompanied by simultaneous splitting and formation of hydrogen bonds, making the estimation of the viscosity deviations difficult.

Positive deviations with a relatively sharp maximum are observed with systems water–aliphatic alcohols or aliphatic carboxylic acids. All these systems have rather large negative excess volumes (up to 2·0 – 5·8% of the total volume).

A comparison of the systems formamide–aliphatic alcohols (C_1, C_2, 1-C_3, 1-C_4, i-C_5) shows that negative viscosity deviations diminish in the order C_1(2a), C_2(2a), 1-C_3(2a + 3a), 1-C_4(2c + 3b), i-C_5(2c + 3b), some of them becoming positive, until a relatively sharp maximum and minimum is formed on the curve. The tendency of these systems to give negative viscosity deviation can be attributed to decomposition of a well organized structure of formamide. This is manifested mainly in mixture with lower alcohols: their basicity with respect to formamide increases in the order indicated above, hence also their ability to form hydrogen bonds with it, whereby the negative deviations are caused to diminish until eventually they become positive.

Positive deviations, often with maximums, are obtained with mixtures of carboxylic acids with amides. All these have negative excess volumes corresponding well to the positive viscosity deviations. For example, for systems formamide–carboxylic aliphatic acids, positive viscosity deviations increase in the order C_1, C_2, C_3, together with absolute values of (negative) excess volumes ($1\cdot2 < 2\cdot0 < 3\cdot1\%$ with respect to the total volume of the mixture).

Systems I–III. The discussion of systems I–III, where the component I is represented by water, is simpler. Large positive deviations of the viscosity are observed in all cases with a sharp maximum which is shifted to the side of pure water. Only with systems water–acetone and water–acetanhydride, the viscosity curve in the region close to the pure substance III drops so that small negative deviations appear. All aqueous systems have negative excess volumes reaching up to 10% of the molar volume of the mixture.

Systems II–II. These are complicated for the same reason as the systems I–II mentioned above. Mixtures of aliphatic alcohols give viscosity curves of a quasiideal form. Eventual deviations are always small and negative; they decrease with increasing temperature until they disappear. This is in accord with very small excess volumes.

A type *2a* curve is obtained with all combinations of cresols and phenol, except for mixtures of phenol with *o*-cresol (*3a*). The disintegration of clusters in more associated aliphatic alcohols forming solutions with nitriles is manifested by viscosity curves *2b* or *2c*.

Negative deviations are obtained also with mixtures of aliphatic alcohols with primary and secondary amines (diethylamine, aniline, toluidines, pyrrol, *etc.*), mixtures of halogenated carboxylic acids, and of phenols with carboxylic acids. In the latter case, the deviations can be changed gradually to positive ones by increasing the acidity of the acid, *i.e.*, the activity of its hydrogen atom. For example, the system acetic acid–phenol gives negative deviations of the viscosity, chloroacetic acid–phenol both negative and positive. and trichloroacetic acid–phenol only positive deviations with a maximum. The formation of associates in the latter case obviously cannot suppress the unfavourable effect of large substituents on the geometry of acetic acid molecule.

Primary or secondary aromatic amines (aniline, N-alkylanilines, toluidines, naphthylamines, *etc.*) give with phenols (phenol, cresols, guaiacol) almost in all cases positive deviations with a maximum, which is often shifted to the side of the component with a higher viscosity. Exceptional cases are those where very unfavourable geometry of one of the molecules prevents the formation of associates. For example, one component has a substituent in *o*-position with respect to the active group and the other has a decreased ability to form hydrogen bonds (guaiacol–N-alkylaniline), or a very large substituent in the *o*-position (*o*-nitrophenol–aniline). These systems give negative deviations.

The negative effect of a substituent (such as alkyl) on the formation of associates is caused by the fact that the substituent in the functional group replaces active hydrogen and at the same time can worsen the geometric characteristics of the molecule, or (on an aromatic ring) only the later effect takes place. For comparison, we can start from the system phenol–aniline, which gives large positive deviations with a maximum shifted to the side of phenol. Its position suggests that probably two phenol molecules and one aniline molecule participate on the formation of a complex. The positive deviations diminish on introducing a methyl group into the functional group in aniline, but the position of the maximum does not change, whereas no essential change of the curve takes place on introducing the methyl group into the *p*-position in aniline. In this case the substitution of the active H atom has an effect, whereas the change in the geometry is unimportant. Introducing an ethyl group into the functional group in aniline causes a further decrease of the positive deviations, which in the region close to the pure N-ethylaniline become even negative. In addition, the maximum is shifted to the side of phenol. It is apparent that the geometric properties of N-ethylaniline molecules become here unfavourable, since its proportion in its complex with phenol is smaller than, *e.g.*, with aniline or N-methylaniline. In this connection we mention the systems guaiacol–aniline (*3b4*), guaiacol–*o*-toluidine (*3b7*), and guaiacol–N-alkylaniline (*2a*).

Systems II–III. These are complicated since the component II dissociates in solution and, in addition, new bonds are formed by hydrogen bridges between unequal molecules. The viscosity curve is decisively influenced by the prevailing effect.

Most of these systems give curves of the type *2*, *e.g.*, mixtures of aliphatic alcohols with esters of carboxylic and hydroxycarboxylic acids. When the molecules of the alcohol and ester are about equal in size (ethanol–ethyl acetate, 1-propanol–propyl acetate, *etc.*), the viscosity curve is of the type *2b*. These mixtures have positive excess volumes. The space density of molecules increases with the ratio of diameters of the component molecules (d_1/d_2), which suppresses the effect of dissociation of component II. For example, by replacing ethyl acetate in its mixture with ethanol in turn with ethyl 3-hydroxybutyrate, ethyl 2-ethyl-3-hydroxybutyrate, and ethyl 2,2-diethyl-3-hydroxybutyrate the negative viscosity deviations decrease (d_1/d_2: $1{\cdot}18 < 1{\cdot}30 < 1{\cdot}41 < 1{\cdot}49$). In extreme cases, the effect of the high space density can prevail and the system shows positive deviations of viscosity (*e.g.*, methanol–n,n-dibutyl ester of decandioic acid, $d_1/d_2 = 2{\cdot}02$).

Also mixtures of aliphatic alcohols with aliphatic ketones give viscosity curves mostly of type *2b*. With a larger ratio of molecular diameters (methanol–acetone: $d_1/d_2 = 1{\cdot}22$), the negative deviations become much smaller and the viscosity curve acquires the form *2a*. Analogously, mixtures of aliphatic alcohols and ethers (including 1,4-dioxane) give negative deviations of viscosity except for those where the effect of a high space density of molecules prevails (methanol–anisol, methanol–phenetol); these give type *3a* curves.

Carboxylic and halogencarboxylic acids give with their esters negative viscosity deviations, which are usually smaller than with aliphatic alcohols–ester mixtures. With increasing size of the ester molecule, the deviations can become positive in the concentration region close to the pure ester. This applies for mixtures of acetic acid with n-butyl acetate ($d_1/d_2 = 1{\cdot}32$) and acetic acid–n-amylbutyrate ($d_1/d_2 = 1{\cdot}47$). In extreme cases, the viscosity deviations can be only positive, *e.g.*, with acetic acid–n-hexadecyl acetate ($d_1/d_2 = 1{\cdot}79$), or when the ester molecules have a suitable shape enabling to increase the space density of molecules (acetic acid–ethyl benzoate, $d_1/d_2 = 1{\cdot}36$).

Mixtures of carboxylic acids with aliphatic ketones give negative deviations of viscosity, which are smaller than with alcohol–ketone mixtures. Excess volumes are negative nearly in all cases. Mixtures of carboxylic acids with aliphatic ethers give also negative deviations of viscosity; the same applies to mixtures where the component II is a heterocyclic compound (pyrrol, piperidine) and III is arbitrary but not heterocyclic. Pyrrol with tertiary amines gives small deviations, both positive and negative.

Dissociation of component II is important with mixtures of aromatic compounds. When component II is an aromatic amine (aniline, toluidines, diphenylamine) and III another aromatic compound (nitro-, ester, ether, *etc.*), curves with negative deviations are observed (*2a*, *2ab*, *2b*), which decrease with increasing temperature until eventually they disappear. Excess volumes are positive or negative.

Similar viscosity curves are obtained with mixtures where component II is a phenol (phenol, cresols, guaiacol, *etc.*) and III either aromatic or aliphatic compound. In some cases, mainly at higher phenol concentrations, some deviations can become positive, as with mixtures of phenol with N,N-dialkylamines of of *o*-chlorophenol with ketones. Also these systems have positive or negative excess volumes.

With aromatic amines or phenols as component II and heterocyclic compounds (pyridine, quinoline, *etc.*) as component III, the viscosity has positive deviations which often pass through a maximum and diminish with increasing temperature. Only in one case (*o*-nitrophenol–pyridine) the viscosity curve has negative deviations at high concentrations of pyridine. The excess volumes are negative in all cases.

Mixtures of heterocyclic compounds (II: pyrrol and its derivatives, III: pyridine, picolines, quinoline, nicotine, *etc.*) give curves mostly of the type *3b*. Excess volumes are negative. An exceptional case is where the geometry of one molecule sort is unfavourable for the formation of an associate of unequal molecules. For example, the pyrrol–pyridine system gives a curve of type *3b*, whereas mixtures of 2,4-dimethylpyrrol or 2,4-dimethyl-3-ethylpyrrol with pyridine give curves of type *2a*. In these cases the excess volumes are positive.

Systems III–III. These give always positive deviations from Raoult's law or are quasiideal. It could hence be expected that their viscosity curves are either quasiideal or show negative deviations. Of 51 systems examined, 16 have a quasiideal course of the viscosity and 28 give negative deviations (mostly *2a* type), so that 86·3% of all systems behave as expected. The quasiideal behaviour show all mixtures of ketones and most of the mixtures of carboxylic acid esters. If one of the esters contains an aromatic ring or the acid is chlorated, the viscosity curve is of type *2a*. Also the systems ether–ether, ether–ester, and ether–ketone give usually this type of curve, only rarely a curve of type *1*.

Systems III–V. These behave in the same manner as the preceding ones. Of 65 systems examined, 54 show negative deviations of viscosity (mostly of type *2a*) and 7 behave in a quasiideal way. Hence, 93·8% of all systems behave as expected. Exceptions are mainly systems involving $SnCl_4$; their positive deviations are probably caused by the formation of clusters of molecules with $SnCl_4$ in the center.

Systems IV–IV, IV–V, *and* V–V. Also these systems are expected to behave either quasiideally or to show negative deviations of viscosity. In reality, both systems IV–IV examined give a viscosity curve of type *2a* and 6 systems IV–V out of 11 show negative deviations, the other 5 systems giving both positive and negative, but small deviations, so that they can be denoted as nearly quasiideal.

Of 79 systems of the class combination V–V, 65 have negative deviations from the straight line (mostly of type *2a*) and 11 behave quasiideally, hence 96·2% of the total number of systems. Only the system n-hexadecane–tetrachloromethane gives very small positive deviations owing to an increase of the space density of molecules in solution with extremely different geometrical properties.

It can be concluded that a very good estimate of the form of the viscosity curve can be made on the basis of the correspondence to a certain class combination according to Ewell if the hydrogen bonds do not change during formation of binary mixtures. This applies for systems III–III, III–V, IV–IV, IV–V, and V–V. A relatively good estimate can be made of the hydrogen bonds are splitted during formation of the solution (II–V). With other systems, it is necessary to observe the rules derived in the present work.

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