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Binary Liquid Mixture Viscosities and Densities

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Liquid mixture viscosities and densities have been measured for 24 binary systems as a function of composition at 298.15 K and ambient pressure. Systems were chosen in order to provide a range of constituent types to test a newly developed predictive equation. The new calculation technique is based on a local-composition thermodynamic model and uses no adjustable viscosity parameters. Shear viscosities calculated with this model agreed with the experimental data within an average absolute deviation of 5% over the entire composition range; alternatively, a constant in the model can be treated as an adjustable parameter for a 2% average absolute deviation.

Introduction

The composition dependence of nonaqueous liquid mixture shear viscosity is not adequately correlated with equations that are composition averages of pure-component values (*1*) because of strong mixture effects that may even lead to minima or maxima in the viscosity vs. composition curves. Experimentalists often correlate their data with equations containing multiple adjustable parameters, but this procedure is of little efficacy when mixture data are unavailable. Accurate predictive techniques usable without any mixture viscosity data would therefore be valuable, particularly if applicable to multicomponent systems. We have recently developed a local-composition model which permits prediction of thermal conductivity (*2*, *3*) or shear viscosity (*1*) from equilibrium thermodynamic data without use of adjustable parameters. In this work, viscosity and density measurements were made on 24 binary systems to test this model. Although viscosity data have been reported in the literature for numerous systems, the 24 systems studied in this work were chosen because of the variety of interaction types; e.g., nonpolar-nonpolar, polar-nonpolar, polar-polar, hydrogen bonding, etc. Only data on a few of these systems have, to our knowledge, been previously reported.

Experimental Section

Pure components were used directly without further purification but they were purchased in their highest commercial purity. Pure-component densities and kinematic viscosities were measured and compared to average literature values to assure no significant effects due to impurities. Pure-component properties are shown and compared in Table I. Generally the measured pure-component property fell within the range of values obtained from the literature. Mixtures were gravimetrically prepared to the nearest 0.1 mg. Sample preparation flasks were stoppered to prevent evaporative composition changes; additions were made through a septum by displacing an equal volume of vapor into a syringe for the added liquid.

Densities were measured to ±0.0001 g/cm^3 with a previously calibrated 4$^1/_2$ digit, Anton Paar, Model DMA45, calculating digital density meter. Air and distilled, deionized water were used as reference fluids to determine the apparatus constants of the density meter. Absolute densities have a ±0.0001 g/cm^3 accuracy over the range 0.5–1.5 g/cm^3 when water and air are used as the calibration fluids, as suggested by the manufacturer, because of the known relationship between the oscillation period of the sample tube and the fluid density. The density cell was water jacketed and maintained isothermal to ±0.002 K by circulating water from a Tamson 45-L visibility bath. The viscosity apparatus was submerged in this same bath. Temperature monitoring was done with a Hewlett-Packard digital quartz thermometer of resolution ±0.0001 K and absolute accuracy of better than ±0.04 K, NBS traceable to IPTS-68.

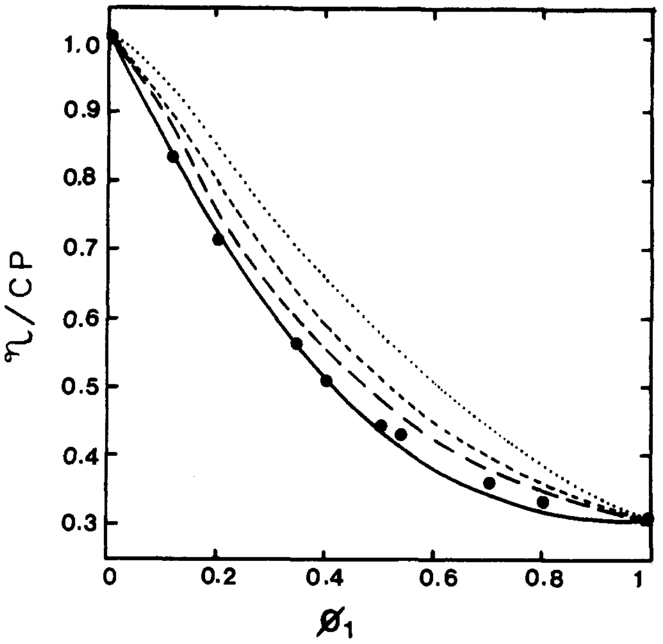

Figure 1. Comparison of local-composition and excess-enthalpic contributions to shear viscosity predictions for acetone–ethanol mixtures at 298.15 K: (●) experimental values; (—) predicted by the NRTL model; (- -) NRTL model with $\sigma = 0$; (---) model with $\xi = \sum \phi_i \xi_i^0 - \sigma H^E/RT$ and $\sigma = 0.25$; (···) same as previous model but $\sigma = 0$.

Kinematic viscosities were measured with a Schott KPG Ubbelohde capillary viscometer, previously calibrated by Schott-Gerate, and a Schott AVS-300 automatic viscosity apparatus consisting of a support stand submerged in the visibility bath and a control unit. The comparison of pure-component data to literature values in Table I indicates a capillary accuracy of better than 0.1%. The stand contains two optical sensors which initiate and stop a quartz digital timer as the meniscus passes through the respective light barriers. The controller has a time resolution of ±0.01 s. The capillary bore size was selected to provide an average residence time in the measuring portion of the capillary tube on the order of 500 s. Kinematic viscosities were computed from

$$\nu = k(t - \theta) \qquad (1)$$

where k is a well-known constant for each capillary, dependent upon the bore size, and θ is the Hagenbach–Couette error correction supplied with each calibrated capillary. Experiments were generally performed in six replicates for each composition. Results from the first run for each new composition were disregarded and treated as a purge of the capillary tube. The

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Table I. Densities and Kinematic Viscosities of Pure Components at 298.15 K[a]

compd	ρ, g/mL exptl	ρ, g/mL lit.	ν, cSt exptl	ν, cSt lit.
acetone	0.7844	0.784 (*5*, *8*)	0.3857	0.386 (*8*, *20*)
carbon tetrachloride	1.5839	1.584 (*5*, *7*, *8*)	0.5675	0.567 (*7*, *19*, *20*)
chlorobenzene	1.1003	1.101 (*5*)	0.6785	0.685 (*20*)
chloroform	1.4718	1.477 (*5*, *8*, *10–12*)	0.3623	0.357 (*10*, *18*, *20*)
cyclohexane	0.7736	0.774 (*5*, *8*, *15*, *16*)	1.1512	1.153 (*20*)
dimethyl sulfoxide	1.0951	1.096 (*5*, *6*, *18*)	1.8028	1.844 (*6*, *20*)
ethanol	0.7852	0.785 (*8*, *9*)	1.3770	1.409 (*8*, *17*)
hexane	0.6562	0.655 (*7*, *10*, *13*)	0.4470	0.451 (*7*, *8*, *10*, *13*)
2-propanol	0.7804	0.781 (*8*, *14*, *15*)	2.6086[b]	
methanol	0.7872	0.787 (*5*, *8*, *9*)	0.7020	0.694 (*8*, *9*, *17*, *20*)
methyl acetate	0.9268	0.928 (*8*)	0.3881	0.392 (*8*)
triethylamine	0.7254	0.725 (*8*, *18*)	0.4908	0.497 (*8*, *17*)

[a] Standard deviations for the viscosity measurements are 0.0001 unless otherwise noted. [b] Standard deviation was 0.0020.

remaining five runs were averaged to yield the reported value. All measurements were made at 298.15 ± 0.01 K. The capillary tube was thoroughly cleaned between mixtures and flushed several times with the new mixture before filling.

Results

Measured mixture viscosities are shown in Table II. The average standard deviation, determined from the replicate runs at each composition, was 0.05%. Table II results have an uncertainty of 0.07% at the 95% confidence level.

We have recently reported a local-composition model useful for predicting mixture thermal conductivity (*2*, *3*) and viscosity (*1*) from equilibrium thermodynamic data. The model is hereafter referred to as the nonrandom two-liquid (NRTL) model due to its derivation from the equilibrium-local-composition model of Renon and Prausnitz (*21*). This model has been evaluated with a combination of literature data and data obtained in this laboratory. The study reported here was performed to obtain test data involving nonideal mixtures. The local-composition model for liquid mixture shear viscosity has been derived elsewhere (*1*) but can be written in terms of volume fractions, ϕ_i, for an n-component system as

$$\eta = \exp(\xi)/V \quad (2)$$

$$\xi = \sum_i \phi_i \xi_i{}^0 + \sum_i \phi_i \left[\sum_j \phi_j G_{ji}(\xi_{ji} - \xi_i{}^0)/\left(\sum_l \phi_l G_{li}\right)\right] - \sigma H^E/RT \quad (3)$$

$$\xi_{ji} = \xi_{ij} = \sum_i (\phi_i{}^* \phi_{ii}{}^* \xi_i{}^0)/\sum_j (\phi_j{}^* \phi_{jj}{}^*) \quad (4)$$

where for the i, j ($i \neq j$) pair of interactions

$$G_{ji} = \exp(-\alpha A_{ji}/RT) \quad (5)$$

$$\phi_j{}^* = (1 + \Gamma_{ji})^{-1} \qquad \phi_{ii}{}^* = (1 + \phi_j{}^* G_{ji}/\phi_i{}^*)^{-1} \quad (6)$$

$$\Gamma_{ji} = (V_i/V_j)(G_{ij}/G_{ji})^{1/2} \exp[(\xi_i{}^0 - \xi_j{}^0)/2] \quad (7)$$

and $\xi_i{}^0$ represents the pure-component-i value given by

$$\xi_i{}^0 = \ln(\eta_i V_i) \quad (8)$$

In the above model, α, A_{ij}, and A_{ji} represent the NRTL parameters for each binary system. For this work, these parameters were obtained directly from the literature, primarily from ref *22* and *23*. However, they may be obtained by fitting the NRTL equation to vapor–liquid or liquid–liquid equilibrium data. Molar volumes, V, for both the pure components and the mixtures were obtained from our density measurements. Excess-enthalpy data, H^E, were obtained from Christensen's compilation (*4*); raw data were curve fitted as a function of composition where such a correlation was not reported.

The net effect of the above model is prediction of liquid mixture shear viscosities from readily available equilibrium thermodynamic data without recourse to adjustable parameters. The nonidealities of the system are fully taken into account by the NRTL data input. Although experimental binary mixture viscosities are relatively more easily obtained than are phase equilibrium data, we view this method as significant for the following reasons: (1) there is a growing tendency to maintain binary NRTL interaction parameters in industrial thermophysical property simulators and to compile extensive tables of these parameters; (2) multiple properties can be obtained from the same set of NRTL binary interaction parameters; and (3) it may be possible to use viscosity data and back out the NRTL parameters. Results of the comparison between calculated and experimental mixture viscosities are shown in Table III. It is important to note that the local-composition model was formulated in terms of the property ξ for which the average absolute deviation for the 24 systems is 1.3%. On the other hand, η is more often the property desired. While the model works extremely well for ξ, exponentiation tends to amplify small deviations in predicted results. Nevertheless, the results are quite good for an entirely predictive equation with no adjustable viscosity parameters. Figure 1 shows the type of compositional behavior that often occurs for nonideal systems. It also illustrates the relative importance of the excess-enthalpy term and the local-composition concept showing that both ideas must be incorporated to achieve acceptable results.

The factor σ in eq 3 was taken as identically 0.25 throughout this work to test the validity of the local-composition model as a nonparametric, predictive method. For correlation of experimental data, σ could be left as an adjustable parameter. Table III also contains a summary of a comparison between experimental and correlated values based upon a one-parameter fit of σ in the local-composition model. The results of this comparison indicate that if σ could be successfully correlated in terms of mixture properties, an even better nonparametric, predictive equation could be obtained.

Finally, it appears that associating or strongly hydrogen bonding systems are not predicted very well with a fixed value of $\sigma = 0.25$. The local-composition model with $\sigma = 0.25$ is currently not appropriate for aqueous systems but is being modified to include strong associations. The results of this work indicate substantial improvement in calculation of triethylamine–methanol and dimethyl sulfoxide–chloroform viscosities when σ is treated as an adjustable parameter. Further studies on highly nonideal and associating mixtures may provide additional insight into more effective a priori values for σ.

Table II. Binary Mixture Kinematic Viscosities Measured at 298.15 K and Ambient Pressure

ϕ_1	ρ, g/mL	ν, cSt
Ethanol–Cyclohexane		
0.0000	0.7736	1.1512
0.2172	0.7719	1.0811
0.3103	0.7724	1.1128
0.3891	0.7729	1.1306
0.4473	0.7735	1.1502
0.5902	0.7755	1.1964
0.6765	0.7771	1.2217
0.7739	0.7791	1.2595
0.8998	0.7827	1.3161
1.0000	0.7852	1.3770
Acetone–Cyclohexane		
0.0000	0.7736	1.1512
0.1900	0.7687	0.7793
0.2998	0.7681	0.6663
0.3939	0.7684	0.5957
0.5033	0.7698	0.5349
0.5979	0.7714	0.4884
0.7449	0.7750	0.4449
0.7874	0.7765	0.4265
0.8777	0.7793	0.4084
1.0000	0.7851	0.3878
Acetone–Ethanol		
0.0000	0.7852	1.3770
0.1163	0.7856	1.0575
0.2030	0.7857	0.8966
0.3396	0.7856	0.7107
0.4016	0.7858	0.6532
0.5058	0.7855	0.5711
0.5437	0.7855	0.5475
0.7084	0.7852	0.4617
0.8097	0.7849	0.4275
1.0000	0.7844	0.3857
Acetone–Methanol		
0.0000	0.7872	0.7020
0.2022	0.7904	0.5922
0.2837	0.7910	0.5525
0.3911	0.7913	0.5094
0.4928	0.7913	0.4748
0.5911	0.7907	0.4460
0.7150	0.7898	0.4163
0.7758	0.7891	0.4047
0.8916	0.7871	0.3885
1.0000	0.7844	0.3857
Methanol–Ethanol		
0.0000	0.7852	1.3770
0.2269	0.7860	1.1457
0.3477	0.7864	1.0482
0.4596	0.7868	0.9693
0.5484	0.7870	0.9155
0.6232	0.7874	0.8751
0.7067	0.7876	0.8320
0.7959	0.7880	0.7903
0.8961	0.7882	0.7459
1.0000	0.7872	0.7020
Acetone–Hexane		
0.0000	0.6562	0.4470
0.1926	0.6756	0.4128
0.3137	0.6897	0.4039
0.4004	0.6994	0.3991
0.5055	0.7157	0.3925
0.6039	0.7278	0.3883
0.6745	0.7373	0.3851
0.7667	0.7517	0.3824
0.8910	0.7651	0.3821
1.0000	0.7844	0.3857
Hexane–Ethanol		
0.0000	0.7852	1.3770
0.2024	0.7580	1.1197
0.2864	0.7468	1.0312
0.3942	0.7329	0.9182
0.4739	0.7223	0.8376
0.6082	0.7048	0.7131
0.7083	0.6914	0.6199
0.7796	0.6822	0.5590
0.8913	0.6684	0.4806
1.0000	0.6562	0.4470
Hexane–Cyclohexane		
0.0000	0.7736	1.1512
0.2086	0.7484	0.8643
0.3070	0.7367	0.7799
0.4143	0.7240	0.6965
0.5082	0.7129	0.6381
0.6118	0.7011	0.5861
0.6840	0.6930	0.5595
0.8116	0.6781	0.5082
0.8825	0.6702	0.4840
1.0000	0.6562	0.4470
Acetone–2-Propanol		
0.0000	0.7804	2.6086
0.2045	0.7795	1.1892
0.2916	0.7792	0.9547
0.4172	0.7792	0.7339
0.4961	0.7795	0.6411
0.5902	0.7799	0.5585
0.6881	0.7806	0.4928
0.7886	0.7814	0.4472
0.9135	0.7830	0.4049
1.0000	0.7844	0.3857
Methanol–2-Propanol		
0.0000	0.7804	2.6086
0.1975	0.7822	1.8044
0.3013	0.7828	1.5682
0.3971	0.7835	1.3698
0.5149	0.7844	1.1732
0.5928	0.7851	1.0688
0.7064	0.7860	0.9393
0.7895	0.7865	0.8593
0.9021	0.7877	0.7673
1.0000	0.7872	0.7020
Ethanol–2-Propanol		
0.0000	0.7804	2.6086
0.2054	0.7816	2.2504
0.3303	0.7824	2.0875
0.4088	0.7827	1.9807
0.4903	0.7831	1.8769
0.5843	0.7834	1.7659
0.6803	0.7838	1.6574
0.8148	0.7843	1.5343
0.8943	0.7847	1.4664
1.0000	0.7852	1.3770
Acetone–Carbon Tetrachloride		
0.0000	1.5839	0.5675
0.2164	1.4184	0.4808
0.3132	1.3357	0.4526
0.4050	1.2601	0.4574
0.5146	1.1727	0.4124
0.5823	1.1188	0.4035
0.6717	1.0523	0.3948
0.8005	0.9461	0.3859
0.9073	0.8602	0.3831
1.0000	0.7844	0.3857
Carbon Tetrachloride–Cyclohexane		
0.0000	0.7736	1.1512
0.2047	0.9435	0.9179
0.3317	1.0402	0.8224
0.4330	1.1218	0.7599
0.5333	1.2033	0.7115
0.6298	1.2811	0.6698
0.7166	1.3542	0.6398
0.7959	1.4170	0.6160
1.0000	1.5839	0.5675
Carbon Tetrachloride–2-Propanol		
0.0000	0.7804	2.6086
0.1997	0.9417	1.6898
0.3717	1.0749	1.2206
0.3706	1.0749	1.2127
0.4912	1.1710	0.9800
0.5807	1.2422	0.8480
0.6908	1.3344	0.7180
0.7920	1.4133	0.6447
0.8682	1.4743	0.6070
1.0000	1.5839	0.5675
Cyclohexane–2-Propanol		
0.0000	0.7804	2.6086
0.2335	0.7765	1.8834
0.3071	0.7756	1.7601
0.4113	0.7744	1.5623
0.5001	0.7730	1.4070
0.5772	0.7732	1.3165
0.6445	0.7717	1.2354
0.7950	0.7711	1.1238
0.9350	0.7721	1.1093
1.0000	0.7736	1.1512
Triethylamine–Chloroform		
0.0000	1.4718	0.3623
0.2271	1.3147	0.4476
0.3346	1.2349	0.5009
0.3966	1.1898	0.5326
0.5222	1.0996	0.5913
0.5981	1.0427	0.6167
0.6761	0.9900	0.6242
0.8110	0.8792	0.5867
0.8725	0.8294	0.5556
1.0000	0.7254	0.4908
Dimethyl Sulfoxide–Chloroform		
0.0000	1.4718	0.3623
0.2115	1.3949	0.6433
0.3054	1.3570	0.8024
0.3688	1.3313	0.9047
0.5432	1.2669	1.1431
0.5842	1.2516	1.2067
0.7003	1.2057	1.3585
0.7970	1.1694	1.4866
0.8938	1.1343	1.6141
1.0000	1.0951	1.8028
Dimethyl Sulfoxide–Methanol		
0.0000	0.7872	0.7020
0.1936	0.8557	0.7450
0.3063	0.8954	0.7862
0.3934	0.9200	0.8343
0.4784	0.9463	0.8920
0.6075	0.9852	1.0111
0.6796	1.0079	1.1108
0.8033	1.0437	1.3163
0.8781	1.0625	1.4625
1.0000	1.0951	1.8028
Triethylamine–Methanol		
0.0000	0.7872	0.7020
0.1940	0.7898	0.7769
0.3028	0.7846	0.8025
0.3965	0.7824	0.8127
0.4830	0.7792	0.8058
0.6167	0.7716	0.7610
0.6746	0.7670	0.7257
0.8009	0.7542	0.6349
0.8837	0.7449	0.5792
1.0000	0.7254	0.4908
Triethylamine–Chlorobenzene		
0.0000	1.1003	0.6785
0.1954	1.0350	0.6466
0.2977	0.9949	0.6272
0.4088	0.9539	0.6068
0.4753	0.9290	0.5946
0.5783	0.8889	0.5680
0.6952	0.8446	0.5440
0.8265	0.7925	0.5150
0.8710	0.7753	0.5051
1.0000	0.7254	0.4908
Hexane–Chloroform		
0.0000	1.4718	0.3623
0.2064	1.3068	0.3631
0.3256	1.2038	0.3666
0.4125	1.1308	0.3705
0.5313	1.0358	0.3823
0.5693	1.0103	0.3849
0.6802	0.9198	0.3943
0.7820	0.8404	0.4060
0.9290	0.7127	0.4347
1.0000	0.6562	0.4470
Methyl Acetate–Hexane		
0.0000	0.6562	0.4470
0.1971	0.7047	0.4083
0.2709	0.7211	0.3979
0.4314	0.7645	0.3861
0.4933	0.7807	0.3839
0.5902	0.8075	0.3806
0.7210	0.8437	0.3795
0.7924	0.8673	0.3809
0.8502	0.8818	0.3810
1.0000	0.9268	0.3881
Chloroform–Methanol		
0.0000	0.7872	0.7020
0.1962	0.9317	0.6591
0.3340	1.0181	0.6273
0.4441	1.0932	0.5899
0.5340	1.1553	0.5514
0.6117	1.2085	0.5147
0.6609	1.2415	0.4881
0.7958	1.3368	0.4207
0.8844	1.3979	0.3840
1.0000	1.4718	0.3623
Methyl Acetate–Chloroform		
0.0000	1.4718	0.3623
0.1807	1.3679	0.4007
0.2953	1.3097	0.4104
0.3790	1.2637	0.4146
0.5358	1.1767	0.4124
0.5976	1.1439	0.4091
0.7016	1.0861	0.4024
0.7808	1.0417	0.3966
0.8762	0.9957	0.3919
1.0000	0.9268	0.3881

Table III. Average Absolute Deviation of Experimental and Calculated Viscosities

	AAD, %				
	for σ = 0.25		for adjustable σ		
system	η	ξ	η	ξ	σ
acetone–cyclohexane	1.67	0.50	1.41	0.38	0.21
ethanol–cyclohexane	7.80	1.76	0.53	0.13	0.66
acetone–ethanol	2.24	0.68	1.79	0.49	0.16
acetone–methanol	6.38	2.07	1.72	0.57	0.50
methanol–ethanol	1.20	0.33	0.46	0.13	8.33
acetone–hexane	2.10	0.64	1.56	0.48	0.22
hexane–ethanol	4.85	1.30	2.72	0.71	0.00
hexane–cyclohexane	3.96	1.02	1.05	0.26	−0.36
acetone–2-propanol	5.71	1.68	4.88	1.22	0.11
methanol–2-propanol	2.30	0.56	1.79	0.43	0.00
ethanol–2-propanol	0.72	0.16	0.29	0.06	0.68
acetone–carbon tetrachloride	4.83	1.35	4.83	1.35	0.25
carbon tetrachloride–cyclohexane	3.10	0.68	0.11	0.03	1.01
carbon tetrachloride–2-propanol	6.86	1.44	4.51	0.95	0.37
cyclohexane–2-propanol	9.65	1.99	2.09	0.45	0.55
triethylamine–chloroform	4.48	1.06	1.25	0.30	0.22
dimethyl sulfoxide–chloroform	13.36	3.12	4.02	0.87	0.45
dimethyl sulfoxide–methanol	9.90	2.41	1.30	0.33	−0.60
triethylamine–methanol	15.63	3.89	2.97	0.83	0.00
triethylamine–chlorobenzene	4.42	1.10	1.25	0.31	1.64
hexane–chloroform	6.04	1.69	0.52	0.14	0.00
methyl acetate–hexane	3.07	0.91	1.17	0.34	0.21
methyl acetate–chloroform	1.38	0.38	1.38	0.38	0.25
chloroform–methanol	1.64	0.46	1.64	0.46	0.25
AAD for 24 systems, 191 points, %	5.15	1.30	1.64	0.50	

Glossary

A_{ij}	NRTL parameters
AAD	average absolute deviation
G_{ij}	NRTL nonrandomness factor
H^E	excess enthalpy
k	capillary viscometer constant
R	gas constant
T	absolute temperature
t	time
V	molar volume

Greek Letters

α	nonrandomness parameter
Γ_{ji}	ratio defined by eq 7
η	shear viscosity
ν	kinematic viscosity
ρ	density
ξ	ln (ηV)
σ	free energy of activation parameter
θ	Hagenbach–Couette error correction factor
ϕ	volume fraction
ϕ_i^*	volume fraction at specific composition defined by eq 6
ϕ_{ji}	local volume fraction of j around component i

Registry No. Acetone, 67-64-1; carbon tetrachloride, 56-23-5; chlorobenzene, 108-90-7; chloroform, 67-66-3; cyclohexane, 110-82-7; dimethyl sulfoxide, 67-68-5; ethanol, 64-17-5; hexane, 110-54-3; 2-propanol, 67-63-0; methanol, 67-56-1; methyl acetate, 79-20-9; triethylamine, 121-44-8.

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