Excess Properties for Propyl Propanoate + Hexane + Benzene at 298.15 K

Herminio Casas, Luisa Segade, Carlos Franjo, and Eulogio Jiménez*

Departamento de Física, Facultade de Ciencias, Universidade da Coruña, 15071 A Coruña, Spain

M. Inmaculada Paz Andrade

Departamento de Física Aplicada, Facultade de Física, Universidade de Santiago de Compostela, 15706 Santiago de Compostela, Spain

Densities, viscosities, and refractive indexes of the ternary system {x_1 propyl propanoate + x_2 hexane + $(1 - x_1 - x_2)$ benzene} and of the corresponding binary mixtures {x_1 propyl propanoate + x_2 hexane}, {x_1 propyl propanoate + x_2 benzene}, and {x_1 hexane + x_2 benzene} have been measured at 298.15 K and atmospheric pressure, over the whole composition range. The excess molar volumes, viscosity deviations, and changes of refractive index on mixing were calculated from experimental measurements. These results were further fitted to the polynomial relation to estimate the coefficients and standard errors. All the experimental values were compared with the results obtained with empirical expressions for estimating ternary properties from binary data.

Introduction

In previous papers (Souza et al., 1992; Franjo et al., 1994, 1995), we have reported experimental excess volumes of nonelectrolyte ternary mixtures with six carbon atoms {propyl propanoate + 1-chlorohexane + hexane}, {propyl propanoate + 1-chlorohexane + cyclohexane}, {propyl propanoate + cyclohexane + hexane}, {propyl propanoate + 1-hexanol + hexane}, {propyl propanoate + 1-hexanol + cyclohexane}, {propyl propanoate + 1-hexanol + 1-chlorohexane}, and {propyl propanoate + 2-hexanone + 1-chlorohexane} at 298.15 K. In continuation of this way, we report here the excess molar volumes of {propyl propanoate + hexane + benzene} and of the binary mixtures: {propyl propanoate + benzene} and {hexane + benzene} at 298.15 K. In addition, we include viscosity deviations and changes of refractive index on mixing of this ternary system, together with the corresponding values for the binary mixtures. The Cibulka (1982) equation has been used to correlate the experimental values of ternary mixtures. The experimental values obtained were used to test the empirical methods of Kohler (1960), Jacob and Fitzner (1977), Colinet (Colinet, 1967; Souza et al., 1992), Tsao and Smith (1953), Toop (1965), Scatchard et al. (1952), and Hillert (1980). These methods predict excess properties of the ternary mixtures from those of the involved binary mixtures. The results obtained for viscosities of binary mixtures were used to test the semiempirical relations of Grunberg and Nissan (1949), McAllister (McAllister, 1960; Eyring, 1936), Auslander (1964), and Teja and Rice (1981a,b). The experimental refractive indexes were compared with the predicted results for the Lorentz−Lorenz, Gladstone−Dale, Wiener, Heller, and Arago−Biot equations, which were compiled by Tasic et al. (1992).

Experimental Section

The chemicals employed were supplied by Fluka and Sigma. Their mole-fraction purities were propyl propanoate (Fluka >0.99), hexane (Sigma >0.99), and benzene (Fluka >0.995). The substances were degassed by ultrasound and dried over molecular sieves (Sigma Union

* To whom correspondence should be addressed.

Table 1. Data for Pure Liquids at 298.15 K

	ρ/g·cm^{-3}		η/mPa·s		n	
substance	exptl	lit.	exptl	lit.	exptl	lit
propyl propanoate	0.875 53	0.875 52[a] 0.875 54[b]	0.641	0.6409[c]	1.3909	1.3920[c]
hexane	0.655 28	0.655 2[d]	0.291	0.2861[e] 0.2968[f]	1.3727	1.3724[g] 1.3732[h]
benzene	0.873 45	0.873 47[i]	0.592	0.5977[j]	1.4979	1.4979[i,k]

[a] Franjo et al. (1995). [b] Souza et al. (1996). [c] Daubert et al. (1985). [d] Eduljee and Boyes (1980). [e] Franjo et al. (1995). [f] TRC Tables. [g] Savini et al. (1965). [h] Aucejo et al. (1995). [i] Ortega and Paz Andrade (1986). [j] Petrino et al. (1995). [k] Tasic et al. (1992). [l] Riddich et al. (1986).

Table 2. Densities and Excess Molar Volumes for Binary Mixtures at the Temperature 298.15 K

x_1	ρ/g·cm^{-3}	V_m^E/cm^3 mol^{-1}	x_1	ρ/g·cm^{-3}	V_m^E/cm^3 mol^{-1}
	x_1 Propyl Propanoate + $(1 - x_1)$ Benzene				
0.0477	0.873 63	−0.0018	0.5475	0.875 12	−0.0443
0.1296	0.873 91	−0.0066	0.5945	0.875 20	−0.0433
0.1696	0.874 06	−0.0123	0.6593	0.875 30	−0.0422
0.2673	0.874 36	−0.0222	0.7675	0.875 41	−0.0329
0.2794	0.874 42	−0.0252	0.8008	0.875 44	−0.0292
0.3559	0.874 64	−0.0325	0.8329	0.875 48	−0.0271
0.3878	0.874 74	−0.0362	0.9327	0.875 53	−0.0140
0.4247	0.874 84	−0.0381	0.9374	0.875 52	−0.0124
	x_1 Hexane + $(1 - x_1)$ Benzene				
0.0496	0.857 12	0.0818	0.5001	0.740 90	0.3987
0.0965	0.842 51	0.1464	0.5432	0.732 13	0.3898
0.1282	0.833 01	0.1869	0.5956	0.721 81	0.3805
0.1695	0.821 03	0.2419	0.6455	0.712 42	0.3588
0.2296	0.804 64	0.2873	0.7010	0.702 44	0.3216
0.2696	0.794 12	0.3250	0.7710	0.690 41	0.2715
0.3354	0.777 82	0.3591	0.8378	0.679 54	0.2073
0.3805	0.767 18	0.3742	0.9378	0.664 95	0.0855
0.4247	0.757 17	0.3861			

S0021-9568(98)00060-0 CCC: $15.00
Published on Web 07/21/1998

Table 3. Densities, Viscosities, and Viscosity Deviations for Binary Mixtures at the Temperature 298.15 K

x_1	ρ/g·cm^{-3}	η/mPa·s	$\Delta\eta$/mPa·s
	x_1 Propyl Propanoate + $(1 - x_1)$ Hexane		
0.0581	0.667 58	0.299	−0.012
0.1227	0.681 39	0.310	−0.024
0.2151	0.701 33	0.330	−0.037
0.3111	0.722 26	0.355	−0.045
0.3983	0.741 43	0.379	−0.051
0.4343	0.749 39	0.389	−0.054
0.4898	0.761 66	0.401	−0.061
0.5466	0.774 24	0.421	−0.061
0.5860	0.783 00	0.433	−0.063
0.6800	0.803 92	0.465	−0.064
0.8894	0.850 69	0.557	−0.045
0.9133	0.856 06	0.573	−0.038
	x_1 Propyl Propanoate + x_2 Benzene		
0.0701	0.873 69	0.622	−0.009
0.1478	0.873 97	0.583	−0.017
0.2542	0.874 34	0.583	−0.022
0.3452	0.874 62	0.586	−0.023
0.4049	0.874 79	0.588	−0.024
0.6160	0.875 24	0.604	−0.018
0.7421	0.875 40	0.616	−0.012
0.8096	0.875 45	0.622	−0.009
0.8780	0.875 49	0.630	−0.005
0.9369	0.875 51	0.635	−0.003
	x_1 Hexane + x_2 Benzene		
0.0488	0.857 37	0.550	−0.027
0.1676	0.821 63	0.466	−0.076
0.2486	0.799 61	0.434	−0.083
0.3321	0.778 62	0.397	−0.095
0.3638	0.771 05	0.386	−0.096
0.4168	0.758 91	0.373	−0.094
0.4682	0.747 66	0.358	−0.093
0.5206	0.736 69	0.347	−0.089
0.6260	0.716 03	0.328	−0.076
0.7434	0.695 06	0.312	−0.056
0.8753	0.673 69	0.299	−0.029
0.9498	0.662 51	0.294	−0.012

Table 4. Refractive Indexes and Changes of Refractive Index on Mixing for Binary Mixtures at the Temperature 298.15 K

x_1	n	Δn	x_1	n	Δn
	x_1 Propyl Propanoate + $(1 - x_1)$ Hexane				
0.0613	1.3733	−0.0003	0.5189	1.3810	−0.0010
0.1321	1.3743	−0.0006	0.5736	1.3821	−0.0009
0.1791	1.3750	−0.0008	0.6253	1.3831	−0.0009
0.2305	1.3758	−0.0009	0.7155	1.3849	−0.0007
0.2968	1.3769	−0.0010	0.7665	1.3860	−0.0006
0.3464	1.3778	−0.0010	0.8161	1.3870	−0.0005
0.4187	1.3791	−0.0011	0.8599	1.3879	−0.0004
0.4729	1.3801	−0.0011	0.9337	1.3894	−0.0002
	x_1 Propyl Propanoate + x_2 Benzene				
0.0032	1.4975	−0.0001	0.5615	1.4265	−0.0112
0.0102	1.4961	−0.0007	0.5945	1.4230	−0.0112
0.0141	1.4956	−0.0008	0.6593	1.4177	−0.0095
0.0191	1.4948	−0.0011	0.7089	1.4130	−0.0089
0.0477	1.4902	−0.0026	0.7107	1.4130	−0.0087
0.0974	1.4825	−0.0050	0.7675	1.4084	−0.0072
0.1296	1.4775	−0.0065	0.8008	1.4057	−0.0064
0.1696	1.4723	−0.0074	0.8329	1.4030	−0.0056
0.2794	1.4573	−0.0107	0.8342	1.4029	−0.0056
0.4247	1.4405	−0.0119	0.9327	1.3955	−0.0024
0.4858	1.4338	−0.0120	0.9374	1.3953	−0.0021
0.5475	1.4279	−0.0113			
	x_1 Hexane + x_2 Benzene				
0.0496	1.4889	−0.0028	0.5001	1.4207	−0.0145
0.0965	1.4810	−0.0048	0.5432	1.4158	−0.0140
0.1282	1.4756	−0.0062	0.5956	1.4097	−0.0136
0.1695	1.4675	−0.0092	0.6455	1.4045	−0.0125
0.2296	1.4589	−0.0102	0.7010	1.3989	−0.0112
0.2696	1.4417	−0.0124	0.7710	1.3921	−0.0092
0.3354	1.4396	−0.0134	0.8378	1.3859	−0.0070
0.3805	1.4360	−0.0142	0.9378	1.3777	−0.0032
0.4247	1.4304	−0.0143			

Table 5. Coefficients A_K and B_K of Eqs 4 and 5 and Standard Deviations s

	A_0	A_1	A_2	A_3	s
	x_1 Propyl Propanoate + $(1 - x_1)$ Hexane				
V_m^E/cm^3·mol^{-1} [a]	1.3658	−0.6515	0.3004		0.0045
$\Delta\eta$/mPa·s	−0.2348	−0.1344	−0.1720		0.0016
Δn	−0.00416	0.00152			0.00003
	x_1 Propyl Propanoate + $(1 - x_1)$ Benzene				
V_m^E/cm^3·mol^{-1}	−0.1697	−0.0797	0.0714		0.0011
$\Delta\eta$/mPa·s	−0.0891	0.0540			0.0005
Δn	−0.0473	0.0115			0.0002
	x_1 Hexane + $(1 - x_1)$ Benzene				
V_m^E/cm^3·mol^{-1}	1.5846	−0.0576		−0.1570	0.0035
$\Delta\eta$/mPa·s	−0.3601	0.1760	−0.0796		0.0022
Δn	−0.0572	0.0070			0.0004

	B_1	B_2	B_3	s
	x_1 Propyl Propanoate + x_2 Hexane + $(1 - x_1 - x_2)$ Benzene			
V_m^E/cm^3·mol^{-1}	−1.0828	−5.2292	−3.5274	0.0092
$\Delta\eta$/mPa·s	1.2523	−1.5191	−2.0432	0.0065
Δn	0.0305	0.0128	−0.0039	0.0002

[a] From Souza et al. (1992).

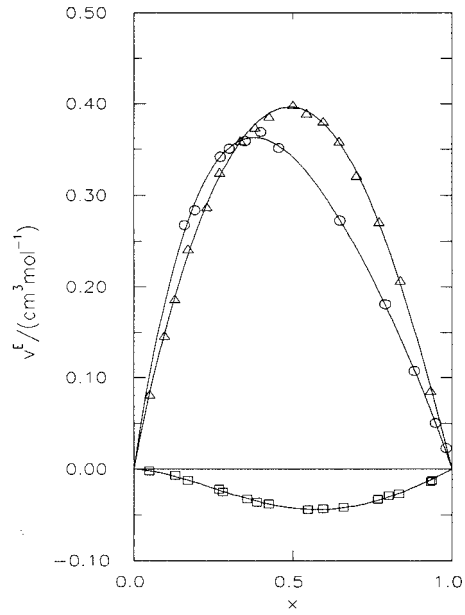

Figure 1. Excess molar volumes V_m^E at the temperature 298.15 K of ○, {x $C_2H_5CO_2(CH_2)_2CH_3$ + (1 − x) $CH_3(CH_2)_4CH_3$}; □,{x $C_2H_5CO_2(CH_2)_2CH_3$ + (1 − x) C_6H_6}; △, {x $CH_3(CH_2)_4CH_3$ + (1 − x) C_6H_6}.

Carbide, type 0.4 nm). The measured densities, viscosities, and refractive indexes of the pure liquids are listed in Table 1 together with published values.

Excess molar volumes were determined from the densities of the pure liquids and mixtures measured with a Anton-Paar DMA 60/602 densimeter thermostated at T = (298.15 ± 0.01) K in a Haake F3 circulating-water bath. Immediately prior to each series of measurements, distilled water and heptane were used to calibrate the densimeter. Kinematic viscosities were measured by means of Schott-Geräte automatic system with a accuracy of ±5 × 10^{-4} mm^2·s^{-1}. Refractive indexes were measured with a thermostated automatic refractometer Atago RX-1000 with a reproducibility in the refractive index data of 1 × 10^{-4}. Mixtures were prepared in all cases by mass using a Mettler AT 201. The precision of the mole fraction is estimated to better than ±1 × 10^{-4}.

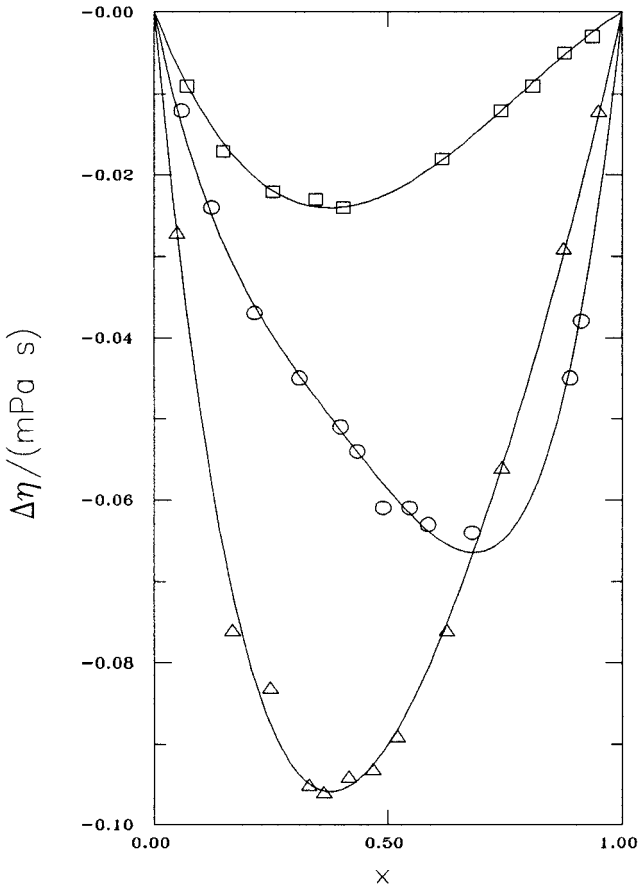

Figure 2. Experimental $\Delta\eta$ at 298.15 K of ○, {x $C_2H_5CO_2(CH_2)_2CH_3$ + (1 − x) $CH_3(CH_2)_4CH_3$}; □, {x $C_2H_5CO_2(CH_2)_2CH_3$ + (1 − x) C_6H_6}; △, {x $CH_3(CH_2)_4CH_3$ + (1 − x) C_6H_6}.

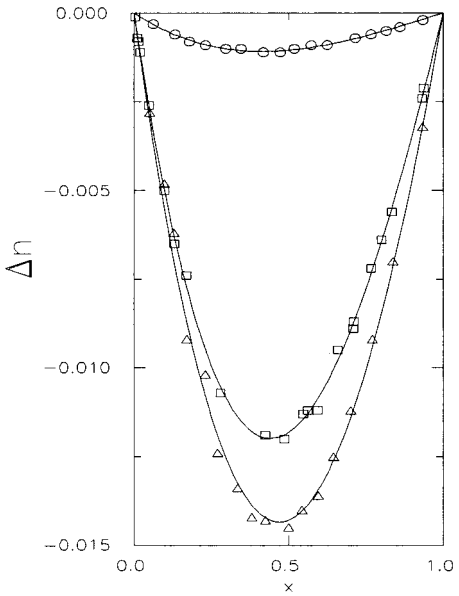

Figure 3. Experimental Δn at 298.15 K of ○, {x $C_2H_5CO_2(CH_2)_2CH_3$ + (1 − x) $CH_3(CH_2)_4CH_3$}; □, {x $C_2H_5CO_2(CH_2)_2CH_3$ + (1 − x) C_6H_6}; Δ, {x $CH_3(CH_2)_4CH_3$ + (1 − x) C_6H_6}.

Results and Discussion

Experimental densities, viscosities and refractive indexes for binary mixtures are listed in Tables 2−4. Excess molar volumes of the binary mixture x_1 propyl propanoate + x_2 hexane were taken from Souza et al. (1992).

The excess molar volumes (V_m^E), viscosity deviations ($\Delta\eta$), and changes in the refractive indexes on mixing (Δn) were computed using eqs 1−3 respectively

$$V_m^E \sum_{i=1}^{N} x_i M_i(\rho^{-1} - \rho_i^{-1}) \quad (1)$$

$$\Delta\eta = \eta - \sum_{i=1}^{N} x_i \eta_i \quad (2)$$

$$\Delta n = n - \sum_{i=1}^{N} x_i n_i \quad (3)$$

In these equations ρ, η, and n are the density, dynamic viscosity, and refractive index, respectively, in the mixture. The ρ_i, η_i, and n_i are the properties of the pure components, and N is the number of components in the mixture. The derived excess functions of the binary systems can be represented by a Redlich−Kister type equation

$$Q^E = x_i x_j \sum_{K=0}^{n} A_K (2x_i - 1)^K \quad (4)$$

where Q^E represents any of the following properties, V_m^E, $\Delta\eta$, or Δn; x_i and x_j are the mole fractions of components i and j, respectively; and A_K denotes the polynomial coefficients. The degree of the polynomial was optimized by applying the F-test (Bevington and Robinson, 1994). The coefficients A_K and the standard deviations s are given in Table 5. Figures 1, 2, and 3 show the experimental V_m^E, $\Delta\eta$, and Δn plotted against x together with the fitted curves. The V_m^E is negative for the mixture propyl propanoate + benzene and positive for the other systems. The results for $\Delta\eta$, and Δn show negative curves for all the mixtures.

Table 6. Parameters for the Semiempirical Relations of Grunberg−Nissan (13), McAllister (14, 15), Auslander (16), and Teja (17, 18) and standard deviations s

	system			s/mPa·s
	x_1 Propyl Propanoate + (1 − x_1) Hexane			
Grunberg−Nissan	$dw = -0.2754$			0.006
McAllister	$n_{12} = 0.4250$	$n_{21} = 0.3706$		0.004
Auslander	$B_{12} = 34.4216$	$B_{21} = 0.0678$	$A_{21} = 68.1872$	0.002
Teja	$\alpha_{12} = 0.7209$			0.007
	x_1 Propyl Propanoate + (1 − x_1) Benzene			
Grunberg−Nissan	$dw = -0.2754$			0.016
McAllister	$n_{12} = 0.4250$	$n_{21} = 0.3706$		0.016
Auslander	$B_{12} = 34.4216$	$B_{21} = 0.0678$	$A_{21} = 68.1872$	0.016
Teja	$\alpha_{12} = 0.7209$			0.016
	x_1 Hexane + (1 − x_1) Benzene			
Grunberg−Nissan	$dw = -0.6979$			0.005
McAllister	$n_{12} = 0.3139$	$n_{21} = 0.3515$		0.002
Auslander	$B_{12} = 1.2423$	$B_{21} = 0.4513$	$A_{21} = 0.3884$	0.002
Teja	$\alpha_{12} = 0.4390$			0.003

Table 7. Standard Deviations of the Experimental Results from the Predicted for the Lorentz−Lorenz (L−L), Gladstone−Dale (G−D), Arago−Biot (A−B), Heller (H), and Wiener (W) Eqs

system	L−L	G−D	A−B	H	W
x_1 propyl propanoate + (1 − x_1) hexane	0.0011	0.0001	0.0019	0.0010	0.0010
x_1 propyl propanoate + (1 − x_1) benzene	0.0029	0.0027	0.0077	0.0007	0.0011
x_1 hexane + (1 − x_1) benzene	0.0010	0.0011	0.0019	0.0015	0.0014

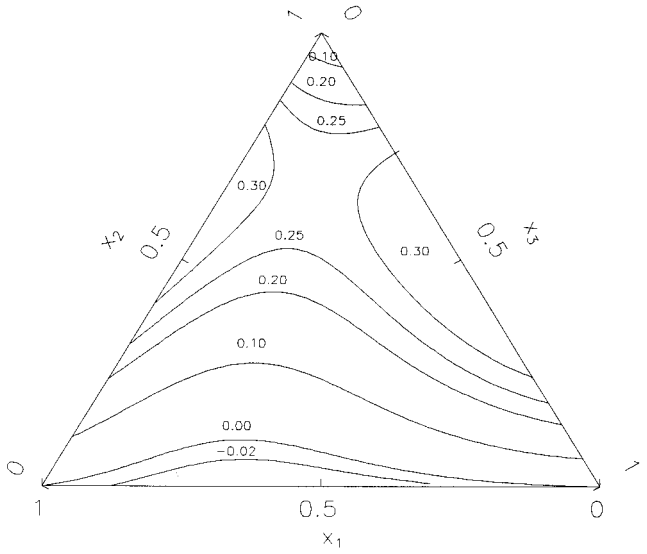

Figure 4. Curves of constant V_m^E/cm^3·mol^{-1} for {x $C_2H_5CO_2(CH_2)_2CH_3$ + x_2 $CH_3(CH_2)_4CH_3$ + (1 − x_1 − x_2) C_6H_6}.

Table 8. Densities (ρ) and Excess Molar Volumes ($V^E_{m,123}$) at 298.15 K for the Ternary Mixture x_1 Propyl Propanoate + x_2 Hexane + (1 − x_1 − x_2) Benzene

x_1	x_2	ρ/g·cm^{-3}	$V^E_{m,123}$/cm^3 mol^{-1}
0.0098	0.0553	0.855 44	0.0892
0.0141	0.0801	0.847 80	0.1197
0.0266	0.1510	0.827 06	0.2071
0.0430	0.2438	0.802 43	0.2859
0.0602	0.3414	0.779 17	0.3245
0.0674	0.3824	0.770 08	0.3337
0.0789	0.4473	0.756 44	0.3341
0.1006	0.5704	0.732 70	0.3255
0.1249	0.7082	0.709 17	0.2673
0.0194	0.0389	0.860 86	0.0541
0.0308	0.0616	0.853 81	0.0853
0.0620	0.1241	0.835 72	0.1554
0.0964	0.1931	0.817 59	0.2072
0.1331	0.2667	0.800 09	0.2410
0.1507	0.3018	0.792 30	0.2530
0.1780	0.3566	0.780 87	0.2573
0.2232	0.4471	0.763 54	0.2514
0.0294	0.0362	0.861 83	0.0467
0.0425	0.0524	0.856 85	0.0712
0.0846	0.1043	0.841 89	0.1265
0.1309	0.1614	0.826 97	0.1692
0.1792	0.2209	0.812 84	0.2023
0.2009	0.2477	0.806 95	0.2065
0.3730	0.4598	0.767 25	0.2268
0.0339	0.0220	0.866 49	0.0214
0.0584	0.0379	0.861 64	0.0404
0.1120	0.0728	0.851 68	0.0748
0.1738	0.1128	0.841 26	0.1007
0.2427	0.1576	0.830 67	0.1245
0.2741	0.1780	0.826 18	0.1339
0.3241	0.2105	0.819 55	0.1266
0.5054	0.3282	0.798 38	0.1425
0.0414	0.0146	0.868 88	0.0144
0.0704	0.0249	0.865 81	0.0244
0.1360	0.0481	0.859 26	0.0459
0.2129	0.0753	0.852 29	0.0633
0.2960	0.1046	0.845 51	0.0712
0.3347	0.1183	0.842 58	0.0708
0.6166	0.2179	0.824 33	0.0862
0.0563	0.0067	0.871 49	0.0066
0.0833	0.0099	0.870 65	0.0030
0.1690	0.0200	0.867 99	0.0077
0.2600	0.0308	0.865 46	0.0045
0.3544	0.0419	0.863 06	0.0011
0.4017	0.0475	0.861 97	−0.0040
0.4752	0.0562	0.860 30	−0.0059
0.6004	0.0710	0.857 64	−0.0024
0.7455	0.0882	0.854 90	−0.0015

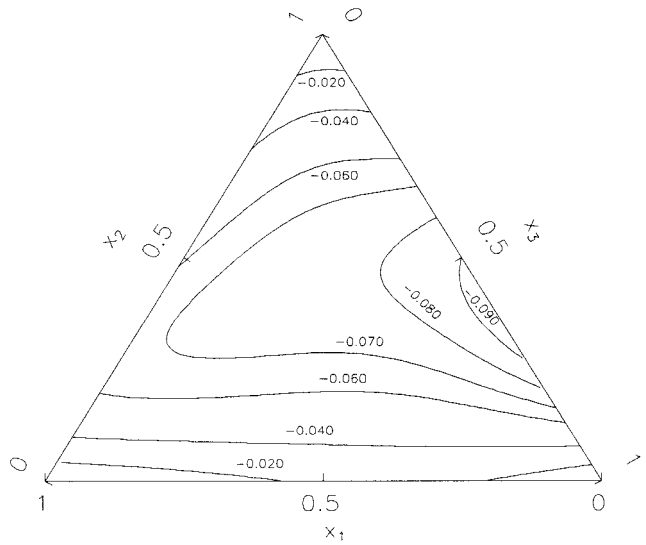

Figure 5. Curves of constant $\Delta\eta_{123}$/mPa·s for {x $C_2H_5CO_2(CH_2)_2CH_3$ + x_2 $CH_3(CH_2)_4CH_3$ + (1 − x_1 − x_2) C_6H_6}.

Table 9. Densities (ρ) and Viscosity Increments ($\Delta\eta_{123}$) at 298.15 K for the Ternary Mixture x_1 Propyl Propanoate + x_2 Hexane + (1 − x_1 − x_2) Benzene

x_1	x_2	ρ/cm^3·mol^{-1}	$\Delta\eta_{123}$/mPa s
0.0527	0.9074	0.672 50	−0.014
0.0481	0.7853	0.691 39	−0.046
0.1368	0.8253	0.690 64	−0.032
0.0900	0.8086	0.690 37	−0.039
0.0938	0.6929	0.710 02	−0.053
0.0874	0.6000	0.726 45	−0.062
0.1654	0.6197	0.727 34	−0.064
0.3146	0.5851	0.741 15	−0.063
0.0813	0.4185	0.762 80	−0.090
0.1525	0.4408	0.761 60	−0.086
0.2314	0.4527	0.762 88	−0.075
0.3127	0.4728	0.762 42	−0.074
0.4113	0.4860	0.763 67	−0.065
0.0808	0.3401	0.780 43	−0.093
0.1521	0.3538	0.780 45	−0.087
0.2121	0.3667	0.780 17	−0.083
0.3848	0.3937	0.781 07	−0.075
0.0712	0.2673	0.797 59	−0.090
0.1622	0.2666	0.801 16	−0.081
0.2121	0.2875	0.798 02	−0.084
0.2849	0.3023	0.797 20	−0.082
0.3781	0.3097	0.798 61	−0.076
0.4632	0.3066	0.801 71	−0.072
0.5629	0.3375	0.797 74	−0.067
0.0727	0.1827	0.819 59	−0.067
0.2747	0.2274	0.814 24	−0.077
0.4468	0.2290	0.818 36	−0.064
0.5482	0.2321	0.819 82	−0.064
0.6522	0.2452	0.818 84	−0.058
0.5170	0.1620	0.835 09	−0.062
0.6213	0.1574	0.837 70	−0.056
0.5038	0.0741	0.856 05	−0.044
0.5959	0.0776	0.856 03	−0.036
0.6984	0.0796	0.856 34	−0.043
0.8120	0.0815	0.856 56	−0.039

Table 6 shows the parameters calculated and the standard deviations between experimental values obtained for dynamic viscosity and the predicted results using the semiempirical relations of Grunberg and Nissan, McAllister, Auslander, and Teja and Rice. The values of critical temperature and critical volume for pure components were obtained from Riddick et al. (1986).

Table 7 compares the experimental refractive indexes for binary mixtures with the predicted results for the Lorentz−Lorenz, Gladstone−Dale, Arago−Biot, Heller, and Wiener equations, which were compiled by Tasic et al.

Table 10. Refractive Indexes and Changes of Refractive Index on Mixing at 298.15 K for the Ternary Mixture x_1 Propyl Propanoate + x_2 Hexane + $(1 - x_1 - x_2)$ Benzene

x_1	x_2	n (exptl)	Δn_{123} (exptl)
0.0098	0.0553	1.4858	−0.0041
0.0141	0.0801	1.4811	−0.0052
0.0430	0.2438	1.4508	−0.0119
0.0602	0.3414	1.4346	−0.0141
0.0674	0.3824	1.4286	−0.0142
0.0789	0.4473	1.4197	−0.0137
0.1006	0.5704	1.4040	−0.0116
0.1249	0.7082	1.3885	−0.0073
0.0194	0.0389	1.4873	−0.0035
0.0308	0.0616	1.4814	−0.0054
0.0620	0.1241	1.4669	−0.0087
0.0964	0.1931	1.4512	−0.0121
0.1331	0.2667	1.4366	−0.0135
0.1507	0.3018	1.4301	−0.0137
0.1780	0.3566	1.4207	−0.0133
0.2232	0.4471	1.4066	−0.0112
0.2766	0.5539	1.3916	−0.0071
0.0294	0.0362	1.4864	−0.0039
0.0425	0.0524	1.4816	−0.0053
0.0846	0.1043	1.4666	−0.0093
0.1309	0.1614	1.4518	−0.0120
0.1792	0.2209	1.4378	−0.0133
0.2009	0.2477	1.7320	−0.0134
0.2994	0.3691	1.4087	−0.0110
0.3730	0.4598	1.3938	−0.0066
0.0339	0.0220	1.4858	−0.0031
0.0584	0.0379	1.4818	−0.0051
0.1120	0.0728	1.4680	−0.0088
0.1738	0.1128	1.4539	−0.0113
0.2427	0.1576	1.4395	−0.0127
0.2741	0.1780	1.4335	−0.0128
0.3241	0.2105	1.4243	−0.0126
0.4100	0.2663	1.4104	−0.0103
0.5054	0.3282	1.3961	−0.0066
0.0414	0.0146	1.4886	−0.0030
0.0704	0.0249	1.4823	−0.0049
0.1360	0.0481	1.4689	−0.0084
0.2129	0.0753	1.4547	−0.0110
0.2960	0.1046	1.4409	−0.0122
0.3347	0.1183	1.4348	−0.0124
0.3967	0.1402	1.4259	−0.0119
0.4944	0.1747	1.4129	−0.0101
0.6166	0.2179	1.3985	−0.0060
0.0563	0.0067	1.4876	−0.0034
0.0833	0.0099	1.4830	−0.0047
0.1690	0.0200	1.4689	−0.0084
0.2600	0.0308	1.4556	−0.0106
0.3544	0.0419	1.4429	−0.0117
0.4017	0.0475	1.4371	−0.0118
0.4752	0.0562	1.4285	−0.0114
0.6004	0.0710	1.4150	−0.0096
0.7455	0.0882	1.4013	−0.0056

The experimental excess molar volumes (V^E_m), viscosity deviations ($\Delta\eta_{123}$), and changes of refractive index on mixing (Δn_{123}) for ternary mixtures are shown in Tables 8−10. The Cibulka equation has been used to correlate the experimental properties of the ternary mixtures

$$Q_{123} = Q_{bin} + x_1x_2(1 - x_1 - x_2)(B_1 + B_2x_1 + B_3x_2) \quad (5)$$

where

$$Q_{bin} = Q_{12} + Q_{13} + Q_{23} \quad (6)$$

The symbol $Q_{123} = V^E_{m,123}$, $\Delta\eta_{123}$, or Δn_{123} and Q_{ij} are given by eq 4. The parameter B_K for eq 5 and corresponding standard deviations are given in Table 5. The lines of constant ternary excess properties calculated by eqs 5 and 6 are shown in Figures 4−6. Figures 7−9 shows lines of constant "ternary contribution", which represent the dif-

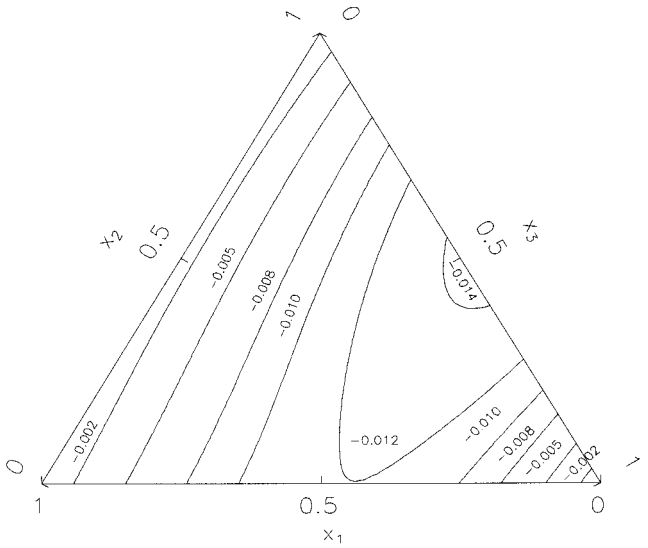

Figure 6. Curves of constant Δn_{123} for $\{x\ C_2H_5CO_2(CH_2)_2CH_3 + x_2\ CH_3(CH_2)_4CH_3 + (1 - x_1 - x_2)\ C_6H_6\}$.

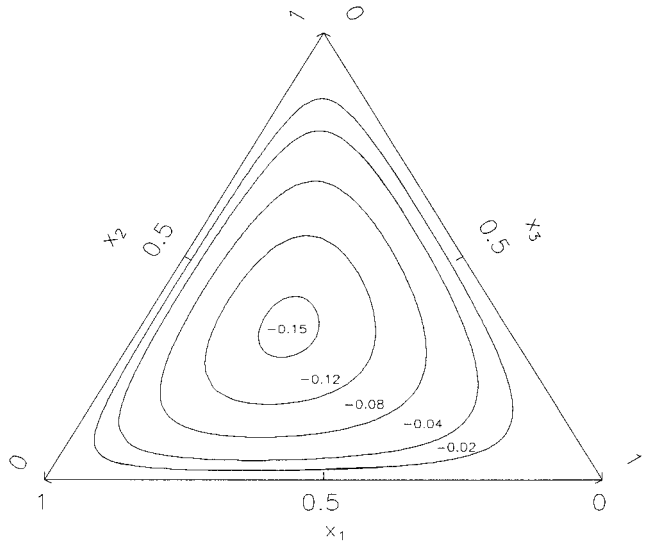

Figure 7. Curves of ternary contribution $V^E_{m,123} - V^E_{m,bin}$/ $cm^3 \cdot mol^{-1}$ for $\{x\ C_2H_5CO_2(CH_2)_2CH_3 + x_2\ CH_3(CH_2)_4CH_3 + (1 - x_1 - x_2)\ C_6H_6\}$.

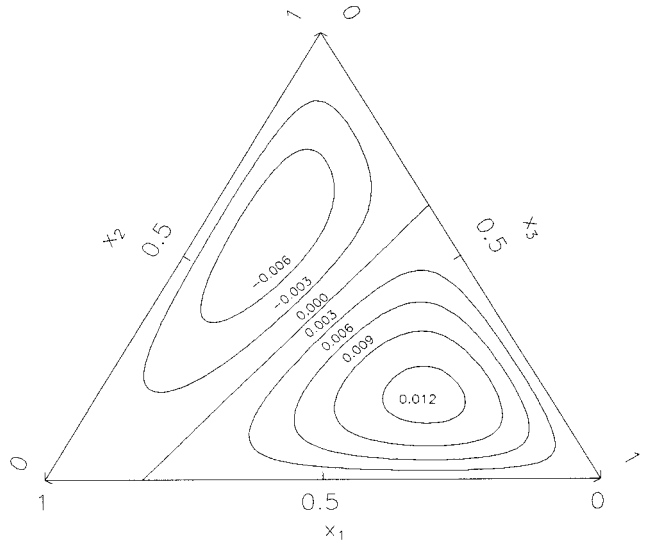

Figure 8. Curves of ternary contribution $\Delta\eta_{123} - \Delta\eta_{bin}$/mPa·s for $\{x\ C_2H_5CO_2(CH_2)_2CH_3 + x_2\ CH_3(CH_2)_4CH_3 + (1 - x_1 - x_2)\ C_6H_6\}$.

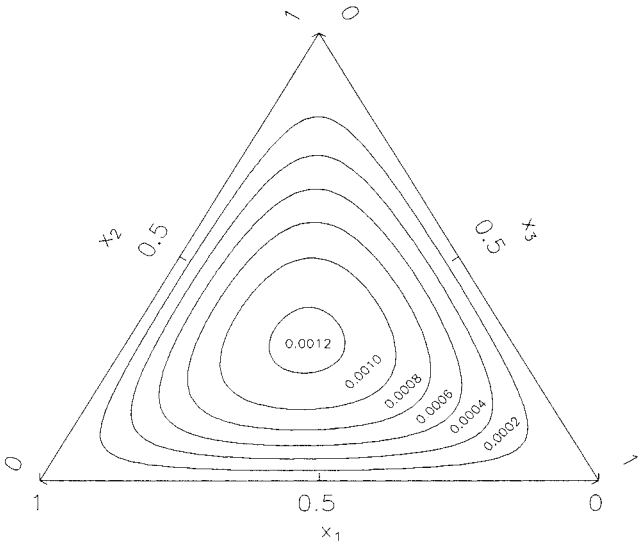

Figure 9. Curves of ternary contribution $\Delta n_{123} - \Delta n_{bin}$ for $\{x\ C_2H_5CO_2(CH_2)_2CH_3 + x_2\ CH_3(CH_2)_4CH_3 + (1 - x_1 - x_2)\ C_6H_6\}$.

Table 11. Maxima and Minima for Ternary Contribution of the Mixture x_1 Propyl Propanoate + x_2 Hexane + (1 − x_1 − x_2) Benzene

	value	coordinates	
$V^E_{m,123} - V^E_{bin}$/cm³·mol⁻¹	minimum −0.155	$x_1 = 0.39$	$x_2 = 0.34$
$\Delta\eta_{123} - \Delta\eta_{bin}$/mPa·s	maximum 0.013	$x_1 = 0.22$	$x_2 = 0.18$
	minimum −0.008	$x_1 = 0.31$	$x_2 = 0.56$
$\Delta n_{123} - \Delta n_{bin}$	maximum 0.0012	$x_1 = 0.37$	$x_2 = 0.31$

Table 12. Standard Deviations *s* of Models for (a) x_1 Propyl Propanoate + x_2 Hexane + (1 − x_1 − x_2) Benzene, (b) x_1 Hexane + x_2 Propyl Propanoate + (1 − x_1 − x_2) Benzene, and (C) x_1 Benzene + x_2 Hexane + (1 − x_1 − x_2) Propyl Propanoate

	a	*b*	*c*
	$V^E_{m,123}$, *s*/cm³·mol⁻¹		
Jacob and Fitzner	0.0546	0.0546	0.0546
Kohler	0.0572	0.0572	0.0572
Colinet	0.0570	0.0570	0.0570
Tsao and Smith	0.1075	0.0460 (*)	0.0998
Toop	0.0693	0.0492	0.0532
Scatchard	0.0687	0.0486	0.0517
Hillert	0.0689	0.0489	0.0536
	$\Delta\eta_{123}$, *s*/mPa·s		
Jacob and Fitzner	0.0038 (*)	0.0038	0.0038
Kohler	0.0048	0.0048	0.0048
Colinet	0.0042	0.0042	0.0042
Tsao and Smith	0.0144	0.0131	0.0073
Toop	0.0049	0.0100	0.0059
Scatchard	0.0048	0.0099	0.0062
Hillert	0.0046	0.0099	0.0052
	Δn_{123}, *s*		
Jacob and Fitzner	0.0006	0.0006	0.0006
Kohler	0.0006	0.0006	0.0006
Colinet	0.0006	0.0006	0.006
Tsao and Smith	0.0025	0.0020	0.0004
Toop	0.0008	0.0007	0.0003 (*)
Scatchard	0.0008	0.0007	0.0003 (*)
Hillert	0.0008	0.0007	0.0003 (*)

ference between the experimental value and that predicted from the binary mixtures ($Q^E_{123} - Q^E_{bin}$). Inside the triangular diagrams exist maxima and minima, whose coordinates are presented in Table 11.

Values of $V^E_{m,123}$, $\Delta\eta_{123}$, and Δn_{123} have been also calculated using the empirical equations proposed by Kohler, Jacob and Fitzner, Colinet, Tsao and Smith, Toop, Scatchard, and Hillert, which take only the binary contribution in account. For the asymmetric methods (Toop, Scatchard, Tsao-Smith, and Hillert), we must indicate the order of components in the mixtures. Table 12 shows the standard deviations between experimental and predicted values; we appoint with (*) the most accurate predictions in each case.

Literature Cited

Aucejo, A.; Burguet, M. C.; Muñoz, R.; Marques, J. L. Densities, Viscosities and Refractive Indices of Some *n*-Alkane Binary Systems at 298.15 K. *J. Chem. Eng. Data* **1995**, *40*, 141−147.

Auslander, G. *Br. Chem. Eng.* **1964**, *9*, 610.

Bevington, P. R.; Robinson, D. K. *Data Reduction and Error Analysis for the Physical Sciences*; McGraw-Hill: Singapore, 1994.

Cibulka, I. Estimation of Excess Volume and Density of Ternary Mixtures of Nonelectrolytes from Binary Data. *Collect. Czech. Commun.* **1982**, *47*, 1414−1419.

Colinet, C. Thesis University of Grenoble, France. 1967.

Daubert, T. E.; Danner, R. P. *Physical and Thermodynamic Properties of Pure Chemicals: Data Compilation*; Hemisphere Publishing Corporation: Bristol, PA, 1989.

Eduljee, G. H.; Boyes, A. P. Viscosity of Some Binary Liquid Mixtures of Oleic Acid and Triolein with Selected Solvents. *J. Chem. Eng. Data* **1980,** *25*, 249−252.

Eyring, H. *J. Chem. Phys.* **1936**, *4*, 283.

Franjo, C.; Jiménez, E.; Iglesias, T. P.; Legido, J. L.; Paz Andrade, M. I. Viscosities and Densities of Hexane + Butan-1-ol, + Hexan-1-ol, and + Octan-1-ol at 298.15 K. *J. Chem. Eng. Data* **1995**, *40*, 68−70.

Franjo, C.; Lorenzana, M. T.; Legido, J. L.; Paz Andrade, M. I.; Jiménez, E. Excess Molar Volumes of $[x_1\ CH_3CH_2CO_2(CH_2)_2CH_3 + x_2\ CH_3(CH_2)_5OH + (1 - x_1 - x_2)\ \{CH_3(CH_2)_4CH_3$ or *c*-$C_6H_{12}\}]$ at the Temperature 298.15 K. *J. Chem. Thermodyn.* **1994**, *26*, 1025−1030.

Franjo, C.; Lorenzana, M. T.; Segade, L.; Jiménez, E.; Legido, J. L.; Paz Andrade, M. I. Excess Molar Volumes of $\{x_1\ CH_3CH_2COO(CH_2)_2$-$CH_3 + x_2\ CH_3(CH_2)_5OH + (1 - x_1 - x_2)\ Cl(CH_2)_5CH_3\}$ at the Temperature 298.15 K. *J. Chem. Thermodyn.* **1995**, *27*, 1197−1204.

Grunberg, L.; Nissan, A. H. Mixture Law for Viscosity. *Nature* **1949**, *164*, 799−800.

Hillert, M. Empirical Methods of Predicting and Representing Thermodynamic Properties of Ternary Solution Phases. *Calphad* **1980**, *4*, 1−12.

Iglesias, T. P.; Legido, J. L.; Romaní, L.; Peleteiro, J.; Franjo, C. Relative Permittivities, Densities, and Excess Molar Volumes of $\{xCH_3(CH_2)_4CH_3 + (1 - x)CH_3(CH_2)_vOH\}$ ($v = 2$, 4, and 6) at the Temperature 298.15 K. *J. Chem. Thermodyn.* **1994**, *26*, 797−802.

Jacob, K. T.; Fitzner, K. The Estimation of the Thermodynamic Properties of Ternary Alloys from Binary Data Using the Shortest Distance Composition Path. *Thermochim. Acta* **1977,** *18*, 197−206.

Kohler, F. Estimation of the Thermodynamic Data for a Ternary System from the Corresponding Binary Systems. *Monatsh. Chem.* **1960**, *91*, 738−740.

McAllister, L. The Viscosity of Liquid Mixtures. *AIChE J.* **1960**, *6*, 427−431.

Ortega, J.; Paz-Andrade, M. I. Expansivities of the Binary Mixtures Benzene + Pentan-1-ol and Benzene + Pentan-2-ol between 298.15 and 323.15 K. *J. Chem. Eng. Data* **1986,** *31*, 231−235.

Petrino, P. J.; Gaston-Bonhomme, Y. H.; Chevalier, J. L. E. Viscosity and Density of Binary Liquid Mixtures of Hydrocarbons, Esters, Ketones, and Normal Chloroalkanes. *J. Chem. Eng. Data* **1995,** *40*, 136−140.

Riddick, J. A.; Bunger, W. B.; Sakano, T. K. *Organic Solvents. Physical Properties and Methods of Purification*; Wiley: New York, 1986.

Savini, C. G.; Winterthaler, D. R.; Van Ness, H. C. Heats of Mixing of Some Alcohol Hydrocarbon Systems. *J. Chem. Eng. Data* **1965**, *10*, 168−171.

Scatchard, G.; Ticknor, L. B.; Goates J. R.; McCartney, E. R. Heats of Mixing in Some Nonelectrolyte Solutions. *J. Am. Chem. Soc.* **1952**, *74*, 3721−3724.

Souza, M. J.; Jiménez, E.; Legido, J. L.; Fernández, J.; Pérez-Martell, E.; Paz Andrade, M. I. Excess Molar Volumes at the Temperature 298.15 K of $\{x_1\ C_2H_5CO_2(CH_2)_2CH_3 + x_2\ Cl(CH_2)_5CH_3 + (1 - x_1 - x_2)$ c−$C_6H_{12}\}$, of $\{x_1\ C_2H_5CO_2(CH_2)_2CH_3 + x_2\ Cl(CH_2)_5CH_3 + (1 - x_1 - x_2)\ CH_3(CH_2)_4CH_3\}$, and $\{x_1\ C_2H_5CO_2(CH_2)_2CH_3 + x_2$ c−$C_6H_{12} + (1 - x_1 - x_2)\ CH_3(CH_2)_4CH_3\}$. *J. Chem. Thermodyn.* **1992**, *24*, 119−128.

Souza, M. J.; Vijande, J.; Jiménez, E.; Franjo, C.; Segade, L.; Legido, J. L.; Paz Andrade, M. I. Excess Molar Volumes at 298.15 K of the Ternary Mixture: Propyl Propanoate + 2-Hexanone + 1-Chlorohexane. *Fluid Phase Equilib.* **1996**, *126*, 225−231.

Tasic, A. Z.; Djordjevic, B. D.; Grozdanic, D. K. Use of Mixing Rules in Predicting Refractive Indices and Specific Refractivities for Some Binary Mixtures. *J. Chem. Eng. Data* **1992**, *37*, 310−313.

Teja, A. S.; Rice, P. *Chem. Eng. Sci.* **1981a**, *36*, 7.
Teja, A. S.; Rice, P. *Ind. Eng. Chem. Fundam.* **1981b**, *20*, 70.
Toop, G. W. Predicting Ternary Activities Using Binary Data. *Trans. TMS-AIME* **1965**, *223*, 850−855.
TRC Thermodynamic Tables. Hydrocarbons. Thermodynamics Research Center, The Texas A&M University System: College Station, Texas.
Tsao, C. C.; Smith, J. M. Heats of Mixing of Liquids. "Applied Thermodynamics". *Chem. Eng. Prog. Symp. Ser.* **1953**, *49*, 107−117.

Received for review March 4, 1998. Accepted May 18, 1998.

JE9800609