JOURNAL OF CHEMICAL & ENGINEERING DATA

SHORT ARTICLE

pubs.acs.org/jced

Densities and Viscosities of Binary and Ternary Mixtures of (Nitrobenzene + 1-Bromobutane), (1-Bromobutane + Methylcyclohexane), (Nitrobenzene + Methylcyclohexane), and (Methylcyclohexane + Nitrobenzene + 1-Bromobutane) from (293.15 to 308.15) K

Shahram Ranjbar* and Seyyed Hamid Momenian†

School of Chemistry, Razi University, Kermanshah, Iran

ABSTRACT: The viscosities and the densities of binary mixtures of methylcyclohexane + nitrobenzene, methylcyclohexane + 1-bromobutane, and 1-bromobutane + nitrobenzene and those of the constituent ternaries have been measured at (293.15 to 308.15) K with the whole entire composition range and atmospheric pressure. The excess molar volumes for the binary mixtures were fitted to the Redlich−Kister equation to determine the appropriate coefficients. The experimental viscosity data have been correlated with the Grunberg−Nissan equation too.

INTRODUCTION

Thermodynamic and transport properties are essential in process design and operation. Density and viscosity values of multicomponent mixtures are required in many chemical engineering calculations involving fluid flow, heat, and mass transfer.[1,2] It is well-known that the solutions of many engineering problems and the study of transport fluid phenomena require the knowledge of the dependence of kinematic viscosity on both temperature and composition of the systems. However, one cannot hope to have direct measurements for the multitude of complex systems of practical interest which justifies the theoretical and practical importance of prediction methods. Although a number of predictive equations[3] are available for estimating thermodynamic excess properties (excess volume, excess enthalpy, and excess free energy) of multicomponent systems, such methods are rarely used for viscosity. Excess molar volumes and viscosity deviation data are fitted to empirical equations.[4,5]

Multicomponent liquid mixtures have attracted the attention of many researchers in the past decades. Ansón et al.[6] have measured viscosities and densities of 1-bromobutane + 1-butanol, + 2-methyl-1-propanol, + 2-butanol, or + 2-methyl-2-propanol at temperatures between (288.15 and 318.15) K and correlated the data using the McAllister biparametric equation.

Oswal et al.[7] have measured the viscosities of 18 binary mixtures of ethanol, 1-propanol, 1-butanol, 1-pentanol, 1-hexanol, 1-heptanol, 1-octanol, 1-nonanol, 1-decanol, 1-dodecanol, 2-propanol, 2-butanol, 2-pentanol, and 2-octanol with cyclohexane and of 1-butanol, 1-pentanol, 1-hexanol, and 1-heptanol with methylcyclohexane at 303.15 K. They also analyzed the viscosity deviations and the excess Gibbs energy of activation ΔG^{*E} of viscous flow in terms of change in molecular interactions and the structure of pure component molecules.

The current study focuses on the viscosity and density of the binary and ternary mixtures of nitrobenzene + 1-bromobutane, 1-bromobutane + methylcyclohexane, nitrobenzene + methylcyclohexane, and methylcyclohexane + 1-bromobutane + nitrobenzene at the temperature range from (293.15 to 308.15) K. For industrial applications, it is more convenient to use models to obtain the viscosity of the fluids of interest, since it is impossible to measure the viscosity at all temperatures, pressures, and compositions. This kind of study is important because the viscosity of liquid mixtures provides information that is often required to solve chemical engineering problems related to heat transfer, mass transfer, and fluid flow. No work has been done to measure viscosity and density of the mixtures containing methylcyclohexane, 1-bromobutane, and nitrobenzene. In addition, the mentioned components are chosen to investigate and interpret the nature of

Table 1. Densities ρ and Viscosities η of Pure Components at Several Temperatures

		ρ/kg·m^{-3}		η/mPa·s	
component	T/K	this work	lit.	this work	lit.
methylcyclohexane	293.15	770.1	769.29[14]	0.7293	0.7367[14]
	298.15	766.7	764.97[14]	0.6827	0.6816[14]
	303.15	761.8	760.6[6]	0.6385	0.6390[6]
	308.15	757.6	756.3[15]	0.5997	0.590[15]
nitrobenzene	293.15	1202.2		1.9834	
	298.15	1197.2	1198.35[16]	1.8095	1.811[17]
	303.15	1191.5	1193.481[18]	1.6595	1.6461[19]
	308.15	1186.4	1188.222[18]	1.5286	
1-bromobutane	293.15	1279.1		0.6473	
	298.15	1272.2	1266.83[20]	0.6096	0.607[20]
	303.15	1265.2	1259.9[20]	0.5811	0.575[20]
	308.15	1257.2	1252.7[20]	0.5500	0.546[20]

Received: January 23, 2010
Accepted: September 7, 2011
Published: September 15, 2011

dx.doi.org/10.1021/je200350a | J. Chem. Eng. Data 2011, 56, 3949–3954

Table 2. Density ρ and Viscosity η for the Binary and Ternary Solutions of Nitrobenzene (1), Methylcyclohexane (2), and 1-Bromobutane (3) from T = (293.15 to 308.15) K as a Function of Mole Fractions x_1 and x_2

		T/K = 293.15 K		T/K = 298.15		T/K = 303.15		T/K = 308.15	
		$10^{-3}\,\rho$	η	$10^{-3}\,\rho$	η	$10^{-3}\,\rho$	η	$10^{-3}\,\rho$	η
x_1	x_2	kg·m^{-3}	mPa·s	kg·m^{-3}	mPa·s	kg·m^{-3}	mPa·s	kg·m^{-3}	mPa·s
					Binary				
0.1230	0.0000	1.2709	0.7256	1.2635	0.6851	1.2565	0.6486	1.2490	0.6150
0.2445	0.0000	1.2628	0.8243	1.2566	0.7761	1.2498	0.7321	1.2424	0.6920
0.3645	0.0000	1.2537	0.9397	1.2471	0.8811	1.2408	0.8280	1.2343	0.7802
0.4240	0.0000	1.2497	0.9936	1.2440	0.9308	1.2380	0.8743	1.2307	0.8225
0.5420	0.0000	1.2408	1.1310	1.2344	1.0546	1.2282	0.9860	1.2222	0.9250
0.6585	0.0000	1.2294	1.2813	1.2238	1.1897	1.2185	1.1091	1.2124	1.0369
0.7737	0.0000	1.2206	1.4768	1.2147	1.3639	1.2099	1.2660	1.2051	1.1796
0.8875	0.0000	1.2115	1.7034	1.2066	1.5656	1.2016	1.4466	1.1960	1.3404
0.9439	0.0000	1.2067	1.8355	1.2016	1.6816	1.1965	1.5479	1.1912	1.4305
0.1422	0.8578	0.8202	0.7850	0.8162	0.7350	0.8124	0.6901	0.8082	0.6490
0.2103	0.7897	0.8452	0.8282	0.8408	0.7723	0.8374	0.7233	0.8323	0.6777
0.3412	0.6588	0.9009	0.9436	0.8975	0.8765	0.8942	0.8177	0.8910	0.7653
0.4040	0.5960	0.9196	0.9971	0.9156	0.9244	0.9128	0.8618	0.9090	0.8054
0.5249	0.4751	0.9732	1.1250	0.9702	1.0424	0.9650	0.9677	0.9626	0.9044
0.6397	0.3603	1.0270	1.2662	1.0227	1.1716	1.0196	1.0893	1.0160	1.0155
0.7489	0.2511	1.0757	1.4157	1.0716	1.3122	1.0678	1.2183	1.0629	1.1356
0.8530	0.1470	1.1236	1.6129	1.1192	1.4836	1.1145	1.3705	1.1115	1.2725
0.9031	0.0969	1.1508	1.7185	1.1463	1.5798	1.1420	1.4595	1.1375	1.3535
0.0000	0.8639	0.8300	0.6993	0.8258	0.6558	0.8218	0.6166	0.8165	0.5806
0.0000	0.7979	0.8583	0.6877	0.8535	0.6457	0.8490	0.6074	0.8438	0.5721
0.0000	0.6080	0.9465	0.6622	0.9412	0.6235	0.9358	0.5882	0.9307	0.5561
0.0000	0.5473	0.9788	0.6581	0.9732	0.6200	0.9678	0.5854	0.9625	0.5540
0.0000	0.4293	1.0410	0.6517	1.0355	0.6152	1.0299	0.5826	1.0244	0.5516
0.0000	0.3720	1.0710	0.6465	1.0648	0.6104	1.0594	0.5780	1.0532	0.5479
0.0000	0.2607	1.1298	0.6441	1.1230	0.6066	1.1185	0.5762	1.1110	0.5462
0.0000	0.1535	1.1873	0.6402	1.1804	0.6034	1.1742	0.5745	1.1675	0.5458
0.0000	0.1014	1.2187	0.6398	1.2122	0.6033	1.2063	0.5754	1.1982	0.5463
					Ternary				
0.0184	0.1775	1.1714	0.6308	1.1648	0.5968	1.1580	0.5671	1.1514	0.5372
0.0223	0.2151	1.1470	0.6312	1.1409	0.5968	1.1346	0.5655	1.1270	0.5364
0.0255	0.2464	1.1339	0.6380	1.1273	0.6032	1.1212	0.5716	1.1144	0.5423
0.0283	0.2729	1.1182	0.6379	1.1119	0.6027	1.1061	0.5708	1.0995	0.5411
0.0337	0.3254	1.0992	0.6492	1.0930	0.6129	1.0872	0.5798	1.0801	0.5490
0.0387	0.3732	1.0722	0.6523	1.0661	0.6156	1.0604	0.5821	1.0539	0.5510
0.0453	0.4376	1.0358	0.6587	1.0301	0.6237	1.0241	0.5863	1.0188	0.5551
0.0532	0.1712	1.1816	0.6580	1.1750	0.6220	1.1681	0.5887	1.1619	0.5587
0.0611	0.5901	0.9571	0.6794	0.9518	0.6385	0.9464	0.6014	0.9411	0.5677
0.0640	0.2059	1.1618	0.6648	1.1553	0.6279	1.1492	0.5945	1.1420	0.5633
0.0692	0.6678	0.9200	0.7273	0.9154	0.6730	0.9103	0.6307	0.9050	0.5897
0.0729	0.2344	1.1451	0.6715	1.1387	0.6335	1.1323	0.5996	1.1258	0.5681
0.0796	0.7689	0.8655	0.7046	0.8610	0.6606	0.8563	0.6208	0.8517	0.5846
0.0803	0.2583	1.1308	0.6751	1.1246	0.6374	1.1188	0.6033	1.1121	0.5713
0.0947	0.3048	1.1042	0.6840	1.0979	0.6446	1.0919	0.6093	1.0855	0.5766
0.1010	0.1626	1.1863	0.6882	1.1799	0.6501	1.1734	0.6157	1.1665	0.5826
0.1077	0.3464	1.0802	0.6927	1.0741	0.6525	1.0682	0.6162	1.0624	0.5830
0.1203	0.1935	1.1677	0.7036	1.1610	0.6653	1.1547	0.6280	1.1485	0.5954
0.1247	0.4012	1.0484	0.7080	1.0425	0.6661	1.0368	0.6284	1.0311	0.5941

dx.doi.org/10.1021/je200350a |*J. Chem. Eng. Data* 2011, 56, 3949–3954

Table 2. Continued

		T/K = 293.15 K		T/K = 298.15		T/K = 303.15		T/K = 308.15	
		$10^{-3}\,\rho$	η	$10^{-3}\,\rho$	η	$10^{-3}\,\rho$	η	$10^{-3}\,\rho$	η
x_1	x_2	kg·m^{-3}	mPa·s	kg·m^{-3}	mPa·s	kg·m^{-3}	mPa·s	kg·m^{-3}	mPa·s
0.1303	0.1573	1.1868	0.7081	1.1805	0.6674	1.1741	0.6313	1.1675	0.5977
0.1358	0.2185	1.1517	0.7130	1.1453	0.6716	1.1393	0.6345	1.1332	0.6008
0.1486	0.2391	1.1386	0.7210	1.1326	0.6792	1.1264	0.6411	1.1202	0.6064
0.1542	0.1861	1.1678	0.7260	1.1616	0.6843	1.1555	0.6465	1.1497	0.6128
0.1634	0.5259	0.9792	0.7430	0.9739	0.6973	0.9686	0.6561	0.9636	0.6191
0.1731	0.2785	1.1152	0.7433	1.1092	0.6968	1.1009	0.6551	1.0975	0.6207
0.1732	0.2091	1.1530	0.7396	1.1469	0.6968	1.1407	0.6581	1.1350	0.6226
0.1823	0.5866	0.9472	0.7565	0.9424	0.7095	0.9372	0.6668	0.9322	0.6304
0.1835	0.1476	1.1916	0.7465	1.1853	0.7037	1.1792	0.6642	1.1730	0.6291
0.1888	0.2278	1.1410	0.7511	1.1346	0.7067	1.1286	0.6666	1.1221	0.6298
0.1944	0.3128	1.0940	0.7554	1.0883	0.7106	1.0823	0.6697	1.0762	0.6318
0.2061	0.6633	0.9077	0.7818	0.9035	0.7318	0.8987	0.6857	0.8940	0.6449
0.2147	0.1728	1.1742	0.7720	1.1680	0.7262	1.1621	0.6851	1.1559	0.6479
0.2182	0.2633	1.1176	0.7724	1.1119	0.7267	1.1060	0.6878	1.1002	0.6466
0.2193	0.1412	1.1970	0.7794	1.1907	0.7401	1.1845	0.6922	1.1783	0.6546
0.2217	0.3567	1.0668	0.7765	1.0613	0.7295	1.0555	0.6867	1.0504	0.6484
0.2391	0.1924	1.1599	0.7896	1.1542	0.7426	1.1481	0.6997	1.1426	0.6613
0.2434	0.2937	1.0985	0.7916	1.0925	0.7430	1.0867	0.6997	1.0806	0.6603
0.2521	0.1352	1.1976	0.8075	1.1915	0.7594	1.1855	0.7161	1.1797	0.6787
0.2547	0.1640	1.1816	0.8086	1.1752	0.7607	1.1690	0.7176	1.1633	0.6780
0.2588	0.2082	1.1481	0.8070	1.1422	0.7584	1.1364	0.7146	1.1311	0.6753
0.2725	0.1315	1.1867	0.8203	1.1806	0.7710	1.1751	0.7275	1.1684	0.6863
0.2753	0.3322	1.0728	0.8164	1.0676	0.7659	1.0621	0.7204	1.0569	0.6794
0.2809	0.4520	1.0100	0.8289	1.0050	0.7762	1.0000	0.7289	0.9945	0.6860
0.2821	0.1815	1.1682	0.8317	1.1623	0.7813	1.1564	0.7356	1.1512	0.6940
0.2909	0.1560	1.1824	0.8397	1.1760	0.7885	1.1702	0.7430	1.1647	0.7029
0.2918	0.1281	1.1919	0.7126	1.1853	0.7465	1.1797	0.7848	1.1733	0.8271
0.2951	0.2374	1.1276	0.8370	1.1220	0.7856	1.1163	0.7391	1.1101	0.6966
0.3038	0.1955	1.1575	0.8517	1.1518	0.7989	1.1459	0.7514	1.1406	0.7089
0.3084	0.4962	0.9838	0.8586	0.9790	0.8028	0.9741	0.7531	0.9694	0.7088
0.3131	0.1511	1.1720	0.8575	1.1660	0.8054	1.1602	0.7571	1.1548	0.7144
0.3189	0.1232	1.1910	0.8655	1.1848	0.8120	1.1797	0.7646	1.1737	0.7211
0.3204	0.1718	1.1707	0.8618	1.1649	0.8088	1.1584	0.7604	1.1515	0.7166
0.3255	0.2619	1.1109	0.8604	1.1054	0.8067	1.0999	0.7585	1.0946	0.7151
0.3339	0.1465	1.1777	0.8736	1.1717	0.8194	1.1658	0.7706	1.1600	0.7265
0.3418	0.5499	0.9545	0.8935	0.9500	0.8334	0.9453	0.7795	0.9407	0.7317
0.3425	0.4133	1.0208	0.8779	1.0156	0.8204	1.0107	0.7691	1.0055	0.7229
0.3435	0.2211	1.1384	0.8843	1.1328	0.8308	1.1273	0.7812	1.1221	0.7361
0.3437	0.1843	1.1572	0.8794	1.1522	0.8253	1.1472	0.7777	1.1415	0.7326
0.3438	0.1659	1.1607	0.8899	1.1552	0.8344	1.1492	0.7840	1.1436	0.7386
0.3629	0.2920	1.0909	0.8940	1.0862	0.8371	1.0799	0.7847	1.0747	0.7393
0.3629	0.1402	1.1773	0.9138	1.1710	0.8556	1.1654	0.8038	1.1597	0.7568
0.3656	0.1604	1.1667	0.9024	1.1608	0.8454	1.1549	0.7947	1.1493	0.7488
0.3678	0.1775	1.1521	0.9157	1.1464	0.8575	1.1406	0.8048	1.1352	0.7575
0.3728	0.4499	0.9980	0.9062	0.9930	0.8456	0.9881	0.7932	0.9840	0.7446
0.3762	0.2422	1.1220	0.9145	1.1167	0.8562	1.1115	0.8042	1.1062	0.7574
0.3857	0.2069	1.1406	0.9183	1.1352	0.8597	1.1297	0.8070	1.1234	0.7604
0.3902	0.1712	1.1573	0.9184	1.1522	0.8606	1.1467	0.8083	1.1400	0.7629
0.3957	0.1528	1.1668	0.9388	1.1616	0.8755	1.1557	0.8209	1.1498	0.7724
0.4091	0.4937	0.9728	0.9457	0.9679	0.8803	0.9636	0.8225	0.9590	0.7706
0.4109	0.1984	1.1368	0.9526	1.1314	0.8838	1.1257	0.8315	1.1207	0.7799

dx.doi.org/10.1021/je200350a |*J. Chem. Eng. Data* 2011, 56, 3949–3954

Table 2. Continued

		T/K = 293.15 K		T/K = 298.15		T/K = 303.15		T/K = 308.15	
		$10^{-3}\,\rho$	η	$10^{-3}\,\rho$	η	$10^{-3}\,\rho$	η	$10^{-3}\,\rho$	η
x_1	x_2	kg·m^{-3}	mPa·s	kg·m^{-3}	mPa·s	kg·m^{-3}	mPa·s	kg·m^{-3}	mPa·s
0.4159	0.2677	1.1011	0.9488	1.0960	0.8865	1.0907	0.8309	1.0869	0.7811
0.4199	0.2252	1.1259	0.9547	1.1206	0.8945	1.1149	0.8384	1.1093	0.7885
0.4210	0.1626	1.1589	0.9549	1.1533	0.8898	1.1476	0.8377	1.1423	0.7880
0.4342	0.1905	1.1410	0.9616	1.1355	0.8989	1.1303	0.8430	1.1251	0.7927
0.4386	0.3529	1.0482	0.9771	1.0433	0.9111	1.0384	0.8521	1.0330	0.7989
0.4458	0.2152	1.1240	0.9791	1.1188	0.9146	1.1134	0.8566	1.1083	0.8049
0.4608	0.2471	1.1085	0.9983	1.1035	0.9321	1.0982	0.8784	1.0928	0.8198
0.4658	0.1799	1.1450	0.9994	1.1396	0.9334	1.1340	0.8745	1.1285	0.8213
0.4694	0.2060	1.1293	1.0143	1.1231	0.9463	1.1180	0.8887	1.1123	0.8329
0.4714	0.3792	1.0306	1.0093	1.0254	0.9395	1.0206	0.8780	1.0159	0.8229
0.4870	0.2351	1.1087	1.0209	1.1037	0.9524	1.0983	0.8905	1.0933	0.8365
0.4941	0.3180	1.0639	1.0293	1.0587	0.9585	1.0539	0.8958	1.0492	0.8402
0.5013	0.1936	1.1335	1.0363	1.1281	0.9655	1.1229	0.9036	1.1176	0.8479
0.5094	0.4098	1.0098	1.0580	1.0050	0.9821	1.0004	0.9153	0.9963	0.8565
0.5109	0.2242	1.1133	1.0531	1.1081	0.9811	1.1028	0.9165	1.0975	0.8590
0.5271	0.3393	1.0475	1.0686	1.0426	0.9937	1.0378	0.9281	1.0334	0.8697
0.5396	0.2894	1.0751	1.0870	1.0720	1.0129	1.0648	0.9429	1.0604	0.8833
0.5427	0.2096	1.1199	1.0784	1.1149	1.0048	1.1099	0.9394	1.1050	0.8807
0.5648	0.3635	1.0289	1.1180	1.0242	1.0357	1.0203	0.9653	1.0158	0.9023
0.5656	0.2730	1.0799	1.1142	1.0751	1.0377	1.0703	0.9658	1.0655	0.9042
0.5722	0.3069	1.0615	1.1276	1.0568	1.0440	1.0518	0.9733	1.0474	0.9178
0.5889	0.2584	1.0864	1.1476	1.0826	1.0672	1.0765	0.9931	1.0721	0.9289
0.5978	0.2886	1.0684	1.1715	1.0636	1.0909	1.0591	1.0086	1.0540	0.9419
0.6090	0.3266	1.0457	1.1771	1.0410	1.0879	1.0366	1.0170	1.0316	0.9540
0.6194	0.2392	1.0956	1.2067	1.0905	1.1192	1.0854	1.0419	1.0802	0.9721
0.6205	0.2723	1.0751	1.1888	1.0706	1.1030	1.0655	1.0263	1.0603	0.9580
0.6338	0.3059	1.0552	1.2168	1.0505	1.1277	1.0458	1.0482	1.0414	0.9783
0.6501	0.2510	1.0848	1.2212	1.0804	1.1323	1.0755	1.0534	1.0708	0.9839
0.6556	0.2877	1.0628	1.2345	1.0578	1.1432	1.0532	1.0626	1.0488	0.9910
0.6839	0.2641	1.0736	1.2750	1.0697	1.1804	1.0644	1.0953	1.0596	1.0207

interaction between the molecules of the mixture by measurement of density or volume of the mixtures.

EXPERIMENTAL SECTION

Materials. Pure methylcyclohexane (mole fraction >0.99, Merck), 1-bromobutane (mole fraction >0.99, Fluka), and nitrobenzene (mole fraction >0.99, Fluka) after purification were used. The purities of all chemicals were checked by gas chromatography using a semicapillary methyl silicon column (outer diameter 530 μm). The solvents were degassed by ultrasound and dried over molecular sieves (Sigma Union Carbide, 0.4 nm).

Apparatus and Procedures. The density and viscosity of the mixtures were measured immediately after their preparation, and all of the properties were measured simultaneously. All binary and ternary mixtures were prepared by mixing known masses of pure liquids in airtight narrow mouth stoppered bottles, taking due precautions to minimize the evaporation losses. The mass measurements were made using an electronic balance (model: AB 204 N Mettler) accurate to ± 0.1 mg. The uncertainty in mole fractions is estimated to be lower than $\pm\, 2\cdot10^{-4}$.

Densities of the purified chemicals and their ternary mixture were measured with a precision of $\pm\, 1\cdot10^{-4}$ g·cm^{-3} using a vibrating tube density meter (model: DMA-58, Anton Paar), which measures oscillating periods, calculates densities, and controls temperature with a precision of ± 0.01 K. The uncertainty in the density measurements was within $\pm\, 2\cdot10^{-4}$ g·cm^{-3}. The density meter was calibrated with double-distilled water and dry air as per the manufacturer's instructions. Furthermore, the accuracies of the density measurements were ascertained by comparing the experimental values for pure solvents with the corresponding literature values (Table 1). After each measurement, the U-tube cell was cleaned with 5 mL of ethanol. Then, dry air was passed through the cell to evaporate any remaining liquid. This process continued until the oscillation period was that of the dry air obtained in the initial calibration. Each measurement was repeated at least twice to check reproducibility. All measurements described above were performed at least four times, and the results reported are the averages of these measurements.

The viscosity was measured by means of a suspended Ubbelohde-type (model: AVS 450, Schott−Gerate) viscometer, calibrated at the experimental temperatures with doubly distilled water,

dx.doi.org/10.1021/je200350a |J. Chem. Eng. Data 2011, 56, 3949–3954

Table 3. Coefficients of the Redlich−Kister (eq 2) Equation for Excess Molar Volume of Binary Solutions at Various Temperatures

T	$10^6 A_0$	$10^6 A_1$	$10^6 A_2$	$10^6 A_3$	
K	$m^3 \cdot mol^{-1}$	$m^3 \cdot mol^{-1}$	$m^3 \cdot mol^{-1}$	$m^3 \cdot mol^{-1}$	σ
		Nitrobenzene + 1-Bromobutane			
293.15	−0.6537	0.9282	0.5673	−1.0346	0.0398
298.15	−0.6282	1.3011	0.8188	−2.2484	0.0442
303.15	−0.7615	0.9918	0.4443	−2.8173	0.0399
308.15	−0.8149	0.6134	0.2862	−2.1227	0.0357
		Nitrobenzene + Methylcyclohexane			
293.15	−0.4703	−1.06647	2.0125	2.286103	0.2711
298.15	−0.7608	−1.1409	3.11413	1.059885	0.2909
303.15	−1.5989	−0.45702	1.918384	0.542864	0.3035
308.15	−2.3710	−0.24958	3.207356	−1.59987	0.3230
		1-Bromobutane + Methylcyclohexane			
293.15	−0.1168	4.5875	0.8349	−9.7737	0.0963
298.15	0.2489	4.7398	1.2673	−9.6082	0.1182
303.15	0.0343	5.6021	−0.4031	−12.1968	0.1244
308.15	−0.1807	5.2999	0.9794	−10.6824	0.1151

Table 4. Adjustable Parameters G_{12} (eq 4) and Standard Deviations of Binary Solutions for Various Temperatures

T	G_{12}	σ
K	mPa·s	mPa·s
	Nitrobenzene + 1-Bromobutane	
293.15	−0.2004	0.0065
298.15	−0.1671	0.0067
303.15	−0.1616	0.0052
308.15	−0.1345	0.0050
	Nitrobenzene + Methylcyclohexane	
293.15	−0.4124	0.0118
298.15	−0.3944	0.0102
303.15	−0.3683	0.0084
308.15	−0.3470	0.0083
	1-Bromobutane + Methylcyclohexane	
293.15	−0.2003	0.0033
298.15	−0.1857	0.0036
303.15	−0.1786	0.0029
308.15	−0.1601	0.0027

purified methylcyclohexane, and 1-bromobutane using density and viscosity values from the literature (Table 1). The Ubbelohde viscometer which depends of the Ubbelohde capillary number [(60 to 600) s, with water] was used to minimize the kinetic energy corrections. For each specified composition, five readings were taken for the flow time with variations not exceeding then ± 0.01 s. The kinematic viscosities, ν, of all mixtures were calculated from the average flow time, while the viscometer constants were determined using values from Marsh[8] for the water viscosity together with the corresponding flow times measured by the means of the viscometer. The viscometer was held in a Heidolph water bath. The temperature control of the capillary was provided by a Schott−Gerate CT1650 thermostatic bath maintained to ± 0.01 K. The uncertainty in the viscosity measurements was within $\pm 2 \cdot 10^{-4}$ mPa·s. The details of the methods and techniques of the measurements have been described earlier.[5−8]

RESULTS AND DISCUSSION

The densities, ρ, and viscosities, η, for the pure liquids and binary and ternary mixtures at different concentrations were determined over the temperature range (293.15 to 308.15) K at intervals of 5 K. The results are given in Tables 1 and 2. The excess molar volumes, V_m^E, can be calculated from

$$V_m^E = \sum \left(\frac{1}{\rho} - \frac{1}{\rho_i} \right) x_i M_i \tag{1}$$

where x_i, M_i, and ρ_i are mole fractions, molecular weights, and densities of pure components, respectively. V_m^E is fitted to a Redlich−Kister polynomial equation of type[9]

$$V_m^E = x_1 x_2 \sum_{n=0}^{p} A_n (x_1 - x_2)^n \tag{2}$$

where A_n are the fitting coefficients which can be obtained by the least-squares method. The A_n coefficients for the binary mixtures, at different temperatures are given in Table 3. In each temperature, the choice of the proper number of coefficients, p, was based on the F-test and the standard deviations,

$$\sigma = \left(\frac{1}{m-p} \sum_{i=1}^{m} (Y_{exp} - Y_{cal})^2 \right)^{1/2} \tag{3}$$

as a criterion of goodness with an error lower at 2 %. In eq 3, m is the number of experimental data, p is number of parameters, and Y_{exp} and Y_{cal} are experimental and calculated properties values, respectively.

The dependence of V_m^E on both composition and temperature for the present mixtures may be explained as a balance between positive and negative contributions. The positive contribution arises from dispersion forces or weak dipole−dipole interaction between unlike molecules. The negative contribution (which also means that the mixtures are less compressible than the corresponding ideal mixture[10]) comes from specific interactions, which include charge transfer, and the structural contributions of the geometrical fitting of one component into another due to a difference in molar volumes. For example, in methylcyclohexane and 1-bromobutane solutions, the positive value of V_m^E (approximately between 0.1 to 0.2 and 0.55 to 0.8 mole fraction of methylcyclohexane) as a result of the dispersion interaction between two components arises may be due to the breaking of cohesive force in like molecules and weak dipole−dipole interaction of 1-bromobutane. The negative values of V_m^E (approximately between 0.2 to 0.55 mole fraction of methylcyclohexane) may be attributed to the fact that adhesive forces between two mixing component are large and dipole−dipole interaction goes on increasing from high polarity; 1-bromobutane concentration increases, and also the difference in the dielectric constants of the liquid components of the binary mixtures increases.

Grunberg and Nissan[11] have suggested the following logarithmic relation between the viscosity of binary liquid mixtures and of pure components

$$\eta = \exp(x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1 x_2 G_{12}) \tag{4}$$

dx.doi.org/10.1021/je200350a | J. Chem. Eng. Data 2011, 56, 3949–3954

where G_{12} is a parameter proportional to interchange energy, which represents a measure of the strength of interaction between the component molecules in the mixture. It allows for the positive and negative deviations from the additive rule. As in presented in Table 4, G_{12} values decrease with increasing temperature in all binary mixtures which means that a measure of the strength of interaction between the component molecules in the mixture is decreasing. This may be attributed to the breaking of cohesive force in like molecules. The standard deviation and adjustable parameters of this equation for binary mixtures are given in Table 4.

According to Kauzman and Eyring,[12] the viscosity of a mixture strongly depends on the entropy of mixture, which is related to liquid structure and enthalpy (and consequently to the molecular interactions between the components of the mixture). Vogel and Weiss[13] affirm that mixtures with strong interactions between different molecules ($H^E < 0$ and negative deviations from Raoult's law) present positive viscosity deviations ($\eta > \Sigma x_i\eta_i$, where η is the viscosity of the mixture, η_i and x_i are the viscosity of pure component i and the mole fraction component i, respectively), whereas for mixtures with positive deviations of Raoult's law and without strong specific interactions, the viscosity deviations are negative ($\eta > \Sigma x_i\eta_i$). The negative values imply the presence of dispersion forces between the mixing components in the mixtures, and the positive values may be attributed to the presence of specific interactions between them.

CONCLUSION

In this study, the viscosity and the densities of binary and ternaries mixtures of nitrobenzene, 1-bromobutane, and methylcyclohexane have been measured. These experiments have been carried out at atmospheric pressure, the temperature range from (293.15 to 308.15) K, and the whole entire composition range. The corresponding excess molar volumes of the binary are fitted to a Redlich−Kister polynomial equation to determine the appropriate coefficients (Table 3). In addition, the experimental viscosity data have been correlated to the equation of Grunberg−Nissan to determine G_{12} (Table 4).

AUTHOR INFORMATION

Corresponding Author

*E-mail: shm.ranjbar@gmail.com. Fax: + 98-831-4274559.

Funding Sources

We would like to acknowledge Razi University for financial support.

Notes

†E-mail: hamid_momenian@yahoo.com.

REFERENCES

(1) Alvarez, E.; Sanjurjo, B.; Cancela, A.; Navaza, J. M. Mass Transfer and Influence of Physical Properties of Solutions in a Bubble Column. *Chem. Eng. Res. Des.* **2000**, *78*, 889–893.

(2) Kumar, S.; Kusakabe, K.; Fan, L. S. Heat Transfer in Three Phase Fluidization and Bubble Columns with High Gas Holdups. *AIChE J.* **1993**, *39*, 1399–1405.

(3) Acree, W. E. *Thermodynamic Properties of Nonelectrolyte Solutions*; Academic Press: New York, 1984; Chapter 56.

(4) Ranjbar, S.; Fakhri, K.; Ghasemi, J. B. Densities and Viscosities of (1-Propanol + 1,2-Dichloroethane), (1-Propanol + Benzaldehyde), (Benzaldehyde + 1,2-Dichloroethane), and (1-Propanol + 1, 2-Dichloroethane + Benzaldehyde) Mixtures from T = 288.15 to 313.15 K. *J. Chem. Eng. Data* **2009**, *54*, 3284–3290.

(5) Nagata, M.; Sakura, M. Refractive index and excess volume for binary liquid mixtures. Part 1. Analyses of new and old data for binary mixtures. *J. Chem. Soc., Faraday Trans.* **1987**, *83*, 2449–2457.

(6) Ansón, A.; Garriga, R.; Martínez, S.; Pérez, P.; Gracia, M. Densities and Viscosities of Binary Mixtures of 1-Bromobutane with Butanol Isomers at Several Temperatures. *J. Chem. Eng. Data* **2005**, *50*, 1478–1483.

(7) Oswal, S. L.; Prajapati, K. D.; Oswal, P.; Ghael, N. Y.; Ijardar, S. P. Viscosity of binary mixtures of 1-Alkanol + Cyclohexane, 2-Alkanol + Cyclohexane and 1-Alkanol + Methylcyclohexane at 303.15 K. *J. Mol. Liq.* **2005**, *116*, 73–82.

(8) Marsh, K. N., Ed. *Recommended Reference Materials for the Realization of Physico Chemical Properties*; Blackwell: Oxford, U.K., 1987.

(9) Redlich, O.; Kister, A. T. Algebraic Representation of Thermodynamic Properties and the Classification of Solutions. *Ind. Eng. Chem.* **1948**, *40*, 345–348.

(10) Nath Ray, M.; Sarkar, B. K.; Chanda, R. Viscosity, Density, and Speed of Sound for the Binary Mixtures of Formamide with 2-Methoxyethanol, Acetophenone, Acetonitrile, 1,2-Dimethoxyethane, and Dimethylsulfoxide at Different Temperatures. *J. Chem. Eng. Data* **2007**, *52*, 1630–1637.

(11) Grunberg, L.; Nissan, A. H. Mixture law for viscosity. *Nature* **1949**, *164*, 799–800.

(12) Kauzman, W.; Eyring, H. The Viscous Flow of large Molecules. *J. Am. Chem. Soc.* **1940**, *62*, 3113–3125.

(13) Vogel, H.; Weiss, A. Transport Properties of Liquids 3. Viscosities of Athermal Liquids Mixtures. *Phys. Chem. Chem. Phys.* **1982**, *86*, 193–198.

(14) Iloukhani, H.; Rezaei-Sameti, M.; Basiri-Parsa, J.; Azizian, S. Studies of Dynamic Viscosity and Gibbs Energy of Activation of Binary Mixtures of Methylcyclohexane with n-Alkanes (C_5−C_{10}) at Various Temperatures. *J. Mol. Liq.* **2006**, *126*, 117–123.

(15) Jagadish, G. B.; Mrityunjaya, I. A. Excess and Deviation Properties for the Binary Mixtures of Methylcyclohexane with Benzene, Toluene, P-xylene, Mesitylene, and Anisole at T = (298.15, 303.15, and 308.15) K. *J. Chem. Thermodyn.* **2006**, *38*, 1717–1724.

(16) Riddick, J. A.; Bunger, W. B.; Sakano, T. K. *Organic Solvents*; Wiley: New York, 1986; Vol. 2.

(17) Gill, D. S.; Bakshi, M. S. Transference Number Measurements of silver nitrate in pure and mixed solvents using the electromotive force method. *J. Chem. Soc., Faraday Trans.* **1989**, *85*, 2285–2295.

(18) Nandhibatla, V. S.; Rakesh, R. T.; Mitesh, C. P. Excess Molar Volumes, Viscosity Deviations, Excess Isentropic Compressibilities and Deviations in Relative Permittivities of (Alkyl acetates (Methyl, Ethyl, Butyl and Isoamyl) + *n*-Hexane, + Benzene, + Toluene, + (*o*-, *m*-, *p*-) Xylenes, + (Chloro-, Bromo-, Nitro-) Benzene at Temperatures from 298.15 to 313.15 K. *J. Mol. Liq.* **2009**, *144*, 13–22.

(19) Ali, A.; Tariq, M. Thermodynamic and Transport Behavior of Binary Liquid Mixtures of Benzyl Alcohol with Monocyclic Aromatics at 303.15 K. *J. Mol. Liq.* **2006**, *128*, 50–55.

(20) Ansón, A.; Garriga, R.; Martínes, S.; Pérez, P.; Gracia, M. Densities and Viscosities of Binary Mixtures of 1-Bromobutane with Butanol Isomers at Several Temperatures. *J. Chem. Eng. Data* **2005**, *50*, 1478–1483.