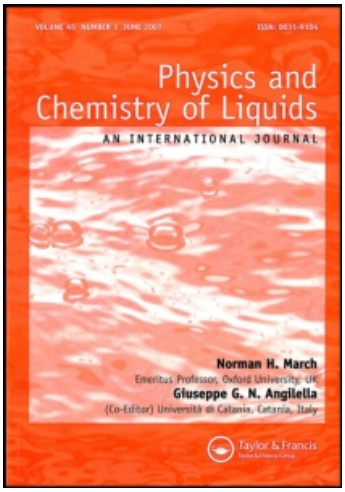

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Theoretical evaluation of thermal conductivity and diffusion coefficient of binary liquid mixtures

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Theoretical evaluation of thermal conductivity and diffusion coefficient of binary liquid mixtures

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Theoretical methods employed for gases have been extended to study thermal conductivity of five binary liquid mixtures (DEG-DBE + *n*-dodecane, DEG-DME + *n*-dodecane, MEG-DME+ *n*-dodecane, TEG-DME + *n*-dodecane and TRG-DME + *n*-dodecane) at 298.15 K and 323.15 K, and diffusion coefficients of five binary mixtures [2-(2-methoxy ethoxy) ethanol + propylene carbonate, carbon tetrachloride + cyclohexane, carbon tetrachloride + benzene, cyclohexane + benzene, and *n*-heptane + toluene] at 298.15 K. There is a reasonable agreement between the theoretical and the experimental findings.

Keywords: Polyethers; Diffusion coefficient; Empirical relation; Thermal conductivity

1. Introduction

The experimental data of thermal conductivity of gaseous mixtures (binary and higher order mixtures) have been analyzed on the basis of various models [1–4] using mathematical theories of non-uniform gases [5]. Very recently, an attempt has been made to extend some of these gaseous state models for computing the thermal conductivity of a few binary liquid mixtures [14]. Due to the scarcity of experimental data, no further extension could be made on the theoretical estimation of thermal conductivity of liquid mixtures. Moreover, as far as our knowledge goes, it appears that such theoretical studies of diffusion coefficient have not been done, mainly due to the lack of experimental data. In the year 1999, accurate measurements of thermal conductivities of five binary liquid mixtures had been made by Burgdorf *et al.* [6]. In the present work we are extending the theoretical methods for computing the thermal conductivities to the aforesaid binary liquid mixtures. For the computation of a diffusion coefficient, we have employed the viscosity data of binary liquid mixtures to obtain its

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values, using the recently proposed interrelation by Marcus [7]. In this way, the theoretical values of diffusion coefficient are calculated for five binary liquid mixtures, using the literature values of liquid viscosity and Marcus relation.

2. Theory

In the present work, the Sutherland–Wassiljewa equation [14] has been applied to evaluate the thermal conductivity and diffusion coefficient of binary liquid mixtures. The aforesaid equation for any transport property (say Z_m) can be expressed as

$$Z_m = \frac{Z_1}{1 + A_{12}(x_2/x_1)} + \frac{Z_2}{1 + A_{21}(x_1/x_2)} \tag{1}$$

where Z_1 and Z_2 are the transport properties of pure components of species 1 and 2. A_{12} and A_{21} are the Wassiljewa coefficients interpreted by Pandey *et al.* [3,4,8] and Grey *et al.* [1,2] as the ratio of efficiencies with which molecules '2' and '1' impede the transport of momentum by molecules '1'.

Equation (1) was independently obtained by Sutherland (1895), Wassiljewa (1904) and Hirshfelder (1958), using totally different approaches. Sutherland's expression was based on the predictions of a simple kinetic theory, whereas Wassiljewa derived it on the basis of a mean free path. Hirshfelder's approximation was based on Chapman and Enskog's kinetic theory. Sutherland–Wassiljewa equation (1) is highly nonlinear and flexible, and capable of explaining an even maxima or minima in any transport property–composition curve.

A new empirical form has been given to the Wassiljewa coefficient [8], A_{ij} to give the best possible results in case of liquids. It is expressed as

$$A_{ij} = \frac{1}{4}\left[1 + \left(\frac{Z_i}{Z_j}\right)^{1/2}\left(\frac{M_j}{M_i}\right)^{3/8}\right]^2 \tag{2}$$

where M_j and M_i represent molecular weights of components 'j' and 'i', respectively.

Wilke [15] proposed a modified expression for the coefficient which can be expressed as

$$A_{ij} = \frac{1}{4}\left[1 + \left(\frac{Z_i}{Z_j}\right)^{1/2}\left(\frac{M_j}{M_i}\right)^{1/4}\right]^2\left[\frac{2M_j}{M_i + M_j}\right]^{1/2} \tag{3}$$

Li [14] proposed an equation to predict thermal conductivity (λ) as follows

$$\lambda_m = \sum_i \sum_j \phi_i \phi_j \lambda_{ij} \tag{4}$$

where

$$\lambda_{ij} = 2(\lambda_i^{-1} + \lambda_j^{-1})^{-1}$$
$$\phi_i = \frac{x_i V_i}{\sum_{i=1}^{2} x_i V_i}$$
$$\phi_j = 1 - \phi_i$$

Here Φ_i and Φ_j are the volume fractions of the ith and jth components. V_i and V_j are the molar volume of ith and jth components, respectively.

The equation of Kendal and Monroe [9], which has no adjustable parameter, is expressed as

$$Z_m = \left[x_1 Z_1^{1/3} + x_2 Z_2^{1/3}\right]^3 \tag{5}$$

Hind *et al.* [10] proposed the following equation

$$Z_m = x_1^2 Z_1 + x_2^2 Z_2 + 2x_1 x_2 Z_{12} \tag{6}$$

Frenkel [16], with the help of Eyring's model, developed the logarithmic relation (9) for non-ideal binary liquid mixtures.

$$\log Z_m = x_1^2 \log Z_1 + x_2^2 \log Z_2 + 2x_1 x_2 \log Z_{12} \tag{7}$$

where

$$Z_{12} = \frac{Z_1 + Z_2}{2}$$

The above-mentioned empirical relations have been used to compute viscosity, now is applied to evaluate thermal conductivity and diffusion coefficient.

The self-diffusion coefficient were found to be strictly proportional to the fluidity of the liquids and to follow, at 298.15 K, the simple expression [7].

$$D\eta = (1.297 \pm 0.024) \times 10^{-12}\ \mathrm{m^2\ Pa} \tag{8}$$

Equation (8) has been used here to evaluate the diffusion coefficient (D) of pure components of liquid mixtures at 298.15 K, using the literature values of viscosity (η).

3. Results and discussion

The thermal conductivity (λ) of five different binary liquid mixtures of polyethers and *n*-dodecane at 298.15 and 323.15 K has been computed for different mole fractions, using Wassiljewa equation with two different Wassiljewa coefficients and the empirical relations. The values obtained theoretically are compared with the experimental findings and are found to be in fairly good agreement. Kendal's empirical relation gives better results than the others. The results of the comparative study of different methods are reported in table 1. Thermal conductivity increases with the size

Table 1. Thermal conductivity of binary liquid mixtures at 298.15 K and 323.15 K in Watt/mK.

x_1	λ (exp)	λ, equations (1) and (2)	λ, equations (1) and (3)	λ, equation (4)	λ, equation (5)	λ, equation (6)	λ, equation (7)
			DEG-DBE + *n*-dodecane at 298.15 K				
0.093	0.1367	0.1368	0.1370	0.1368	0.1368	0.1368	0.1368
0.193	0.1365	0.1367	0.1380	0.1378	0.1377	0.1377	0.1377
0.311	0.1350	0.1388	0.1391	0.1388	0.1387	0.1387	0.1387
0.409	0.1353	0.1396	0.1400	0.1397	0.1395	0.1396	0.1395
0.494	0.1360	0.1404	0.1408	0.1404	0.1403	0.1403	0.1403
0.603	0.1368	0.1413	0.1417	0.1414	0.1412	0.1412	0.1412
0.702	0.1383	0.1422	0.1425	0.1422	0.1421	0.1421	0.1421
0.792	0.1392	0.1429	0.1432	0.1430	0.1429	0.1429	0.1429
0.903	0.1421	0.1439	0.1440	0.1439	0.1438	0.1439	0.1438
			DEG-DME + *n*-dodecane at 298.15 K				
0.093	0.1363	0.1373	0.1374	0.1370	0.1376	0.1377	0.1376
0.147	0.1359	0.1381	0.1382	0.1376	0.1385	0.1386	0.1385
0.296	0.1363	0.1404	0.1406	0.1395	0.1411	0.1413	0.1412
0.420	0.1380	0.1424	0.1427	0.1413	0.1433	0.1435	0.1433
0.512	0.1396	0.1440	0.1443	0.1428	0.1449	0.1451	0.1450
0.605	0.1393	0.1457	0.1460	0.1444	0.1466	0.1468	0.1466
0.707	0.1416	0.1476	0.1479	0.1465	0.1484	0.1486	0.1485
0.790	0.1462	0.1493	0.1495	0.1483	0.1499	0.1501	0.1500
0.903	0.1475	0.1517	0.1518	0.1511	0.1520	0.1521	0.1520
			MEG-DME + *n*-dodecane at 298.15 K				
0.129	0.1366	0.1367	0.1375	0.1368	0.1376	0.1376	0.1376
0.250	0.1358	0.1376	0.1390	0.1376	0.1391	0.1391	0.1391
0.410	0.1360	0.1390	0.1410	0.1389	0.1410	0.1411	0.1411
0.496	0.1367	0.1399	0.1421	0.1398	0.1421	0.1422	0.1421
0.628	0.1370	0.1416	0.1438	0.1413	0.1438	0.1439	0.1438
0.711	0.1395	0.1428	0.1449	0.1425	0.1448	0.1449	0.1448
0.799	0.1395	0.1443	0.1460	0.1439	0.1459	0.1460	0.1459
0.896	0.1424	0.1462	0.1472	0.1459	0.1472	0.1472	0.1472
			TEG-DME + *n*-dodecane at 298.15 K				
0.103	0.1371	0.1387	0.1389	0.1384	0.1385	0.1386	0.1385
0.203	0.3930	0.1412	0.1416	0.1407	0.1409	0.1412	0.1410
0.744	0.1508	0.1550	0.1554	0.1544	0.1547	0.1550	0.1548
0.803	0.1525	0.1565	0.1569	0.1560	0.1562	0.1565	0.1563
0.910	0.1571	0.1592	0.1594	0.1580	0.1591	0.1590	0.1591

			TRG-DME + *n*-dodecane at 298.15 K				
0.122	0.1367	0.1384	0.1384	0.1379	0.1384	0.1385	0.1384
0.215	0.1358	0.1402	0.1402	0.1394	0.1402	0.1404	0.1403
0.325	0.1393	0.1424	0.1424	0.1414	0.1424	0.1426	0.1425
0.410	0.1412	0.1441	0.1441	0.1429	0.1441	0.144	0.1442
0.514	0.1427	0.1462	0.1462	0.1450	0.1462	0.1465	0.1463
0.602	0.1440	0.1480	0.1480	0.1468	0.1481	0.1483	0.1481
0.699	0.1448	0.1500	0.1500	0.1489	0.1501	0.1503	0.1501
0.787	0.1497	0.1518	0.1519	0.1510	0.1519	0.1521	0.1519
0.899	0.1536	0.1542	0.1542	0.1530	0.1543	0.1543	0.1543
			DEG-DBE + *n*-dodecane at 323.15 K				
0.205	0.1305	0.1332	0.1335	0.1332	0.1331	0.1331	0.1331
0.310	0.1316	0.1343	0.1346	0.1343	0.1342	0.1342	0.1342
0.405	0.1329	0.1353	0.1357	0.1353	0.1351	0.1352	0.1352
0.502	0.1338	0.1363	0.1367	0.1363	0.1362	0.1362	0.1362
0.606	0.1343	0.1374	0.1377	0.1374	0.1372	0.1373	0.1373
0.703	0.1354	0.1384	0.1387	0.1384	0.1383	0.1383	0.1383
0.799	0.1359	0.1394	0.1396	0.1394	0.1393	0.1393	0.1393
0.903	0.1368	0.1405	0.1406	0.1405	0.1404	0.1405	0.1404
			DEG-DBE + *n*-dodecane at 323.15 K				
0.060	0.1294	0.1318	0.1318	0.1316	0.1319	0.1320	0.1320
0.178	0.1288	0.1334	0.1335	0.1328	0.1338	0.1339	0.1338
0.247	0.1294	0.1343	0.1345	0.1336	0.1349	0.1350	0.1349
0.308	0.1297	0.1352	0.1354	0.1344	0.1359	0.1360	0.1359
0.404	0.1305	0.1367	0.1369	0.1357	0.1374	0.1376	0.1375
0.549	0.1325	0.1390	0.1392	0.1378	0.1398	0.1399	0.1398
0.619	0.1353	0.1401	0.1404	0.1390	0.1409	0.1411	0.1410
0.710	0.1364	0.1417	0.1420	0.1406	0.1424	0.1426	0.1425
0.800	0.1423	0.1434	0.1436	0.1425	0.1439	0.1440	0.1440
0.906	0.1423	0.1454	0.1455	0.1449	0.1457	0.1458	0.1457
			MEG-DME + *n*-dodecane at 323.15 K				
0.155	0.1293	0.1298	0.1307	0.1304	0.1299	0.1299	0.1299
0.302	0.1280	0.1286	0.1302	0.1298	0.1288	0.1288	0.1288
0.497	0.1288	0.1271	0.1292	0.1287	0.1274	0.1274	0.1274
0.730	0.1307	0.1255	0.1273	0.1270	0.1257	0.1257	0.1257
0.815	0.1340	0.1249	0.1264	0.1261	0.1251	0.1251	0.1251
			TEG-DME + *n*-dodecane at 323.15 K				
0.108	0.1297	0.1337	0.1384	0.1334	0.1335	0.1337	0.1336
0.209	0.1330	0.1362	0.1402	0.1357	0.1359	0.1362	0.1360

(*Continued*)

Table 1. Continued.

x_1	λ, (exp)	λ, equations (1) and (2)	λ, equations (1) and (3)	λ, equation (4)	λ, equation (5)	λ, equation (6)	λ, equation (7)
0.309	0.1341	0.1387	0.1424	0.1380	0.1383	0.1386	0.1384
0.395	0.1351	0.1408	0.1441	0.1401	0.1404	0.1408	0.1405
0.509	0.1400	0.1437	0.1462	0.1429	0.1432	0.1436	0.1433
0.607	0.1413	0.1461	0.1480	0.1453	0.1457	0.1460	0.1457
0.706	0.1434	0.1485	0.1500	0.1478	0.1481	0.1484	0.1482
0.805	0.1467	0.1509	0.1519	0.1504	0.1507	0.1509	0.1507
0.885	0.1500	0.1529	0.1542	0.1526	0.1527	0.1529	0.1528
			TRG-DME + *n*-dodecane at 323.15 K				
0.114	0.1291	0.1332	0.1332	0.1327	0.1332	0.1333	0.1332
0.193	0.1300	0.1347	0.1347	0.1340	0.1347	0.1349	0.1347
0.298	0.1313	0.1357	0.1367	0.1358	0.1368	0.1370	0.1368
0.404	0.1328	0.1388	0.1388	0.1377	0.1389	0.1391	0.1389
0.560	0.1365	0.1419	0.1419	0.1407	0.1420	0.1422	0.1420
0.702	0.1400	0.1448	0.1448	0.1437	0.1448	0.1450	0.1449
0.784	0.1410	0.1465	0.1465	0.1456	0.1465	0.1467	0.1466
0.895	0.1427	0.1488	0.1488	0.1483	0.1488	0.1489	0.1488

Table 2. Diffusion coefficient (D) (m^2s^{-1}) of binary liquid mixtures at 298.15 K 2-(2-methoxy ethoxy)ethanol + propylenecarbonate at 298.15 K.

x_1	D, (exp)	D, equations (1) and (2)	D, equations (1) and (3)	D, equation (5)	D, equation (6)	D, equation (7)
0.034	5.1756	5.1369	5.1370	5.1480	5.1532	5.1496
0.063	5.1571	5.0823	5.0825	5.1019	5.1113	5.1047
0.085	5.1387	5.0424	5.0427	5.0679	5.0802	5.0715
0.110	5.1123	4.9966	4.9970	5.0284	5.0439	5.0329
0.141	5.0903	4.9421	4.9427	4.9809	4.9999	4.9864
0.162	5.0684	4.9051	4.9057	4.9482	4.9695	4.943
0.174	5.0506	4.8839	4.8846	4.9493	4.9519	4.9358
0.219	4.9865	4.8074	4.8082	4.8604	4.8872	4.8679
0.269	4.9036	4.7238	4.7247	4.7835	4.8143	4.7919
0.323	4.8073	4.6375	4.6385	4.70223	4.7363	4.7114
0.370	4.7146	4.5659	4.5670	4.6336	4.6699	4.6431
0.381	4.6806	4.5487	4.5498	4.6168	4.6537	4.6464
0.442	4.5605	4.4576	4.4588	4.5271	4.5655	4.5368
0.472	4.4941	4.4150	4.4163	4.4843	4.5231	4.4941
0.537	4.3670	4.3224	4.3237	4.3897	4.4283	4.3991
0.569	4.3075	4.2794	4.2806	4.3449	4.3829	4.3540
0.619	4.2344	4.2128	4.2140	4.2745	4.3110	4.2831
0.658	4.1731	4.1622	4.1633	4.2202	4.2550	4.2282
0.750	4.0317	4.0461	4.0470	4.0925	4.1214	4.0989
0.776	3.9932	4.0148	4.0157	4.0575	4.0842	4.0633
0.800	3.9676	3.9868	3.9876	4.0257	4.0504	4.3031
0.814	3.9519	3.9695	3.9706	4.0061	4.0293	4.0111
0.845	3.9149	3.9333	3.9340	3.9646	3.9847	3.9688
0.906	3.8521	3.8639	3.8644	3.8837	3.8968	3.8864
0.941	3.8102	3.8249	3.8252	3.8376	3.8460	3.8393
0.980	3.7791	3.7882	3.7829	3.7872	3.7901	3.7878
			Carbontetrachloride + cyclohexane at 298.15 K			
0.2753	14.5681	14.3795	14.5690	14.4670	14.4670	14.4670
0.3845	14.5469	14.3588	14.5701	14.4575	14.4575	14.4575
0.4727	14.5273	14.3500	14.5620	14.4498	14.4490	14.4490
0.5800	14.5030	14.3475	14.5030	14.4405	14.4405	14.4405
0.6707	14.3835	14.3515	14.5206	14.4326	14.4326	14.4326
0.7701	14.4609	14.3617	14.4905	14.4220	14.4240	14.4240
0.8640	14.4400	14.3760	14.4577	14.4158	14.4158	14.4158
0.6805	14.4819	14.3523	14.5179	14.4318	14.4318	14.4318
0.4418	14.5338	14.3524	14.5656	14.4525	14.4526	14.4526

(*Continued*)

Table 2. Continued.

x_1	D, (exp)	D, equations (1) and (2)	D, equations (1) and (3)	D, equation (5)	D, equation (6)	D, equation (7)
			Carbontetrachloride + benzene at 298.15 K			
0.137	20.2656	19.7780	19.9401	20.4064	20.5194	20.4000
0.181	19.8622	19.3054	19.5005	20.0648	20.2063	20.1059
0.218	19.5331	18.9384	19.1549	19.7840	19.9463	19.8303
0.252	19.3006	18.6126	18.4884	19.5224	19.7019	19.5727
0.288	19.0735	18.2903	18.5347	19.2512	19.4462	19.3049
0.319	18.8517	18.0310	18.2826	19.0233	19.2294	19.0792
0.471	17.6463	16.9030	17.1558	17.9142	18.1492	17.9730
0.615	16.7355	16.0442	16.2573	16.9094	17.1312	16.9608
			Cyclohexane + benzene at 298.15 K			
0.2942	19.1298	18.9861	18.9822	19.2377	19.4296	19.2903
0.2613	19.3871	19.2435	19.2398	19.4811	19.6597	19.5309
0.2368	19.5038	19.4386	19.4351	19.6637	19.8311	19.7109
0.1692	19.6515	19.9926	19.9896	20.1734	20.3040	20.2115
0.1659	19.7412	20.0202	20.0173	20.1985	20.3271	20.2361
0.1226	19.8318	20.3883	20.3859	20.5299	20.6300	20.5598
0.0770	19.8318	20.7871	20.7854	20.8827	20.9490	20.9030
0.2951	19.2433	18.9791	18.9753	19.2310	19.4233	19.2837
0.6686	16.4803	18.3836	16.3807	16.6089	16.8104	16.6543
			n-heptane + *n*-toluene at 298.15 K			
0.6030	28.6998	28.1003	28.1237	28.3593	28.7920	28.4723
0.5790	28.3118	27.8008	27.8244	28.0612	28.5013	28.1745
0.5610	28.1040	27.5785	27.6021	27.8390	28.2833	27.9622
0.5500	27.9526	27.4435	27.4671	27.7038	28.1500	27.8168
0.5408	27.8625	27.3312	27.3547	27.5911	28.0386	27.7037
0.5328	27.7730	27.2739	27.2574	27.4933	27.9417	27.6056
0.5212	27.6546	27.0935	27.1169	27.3519	27.8012	27.4637
0.5171	27.5957	27.0536	27.0771	27.3117	27.7612	27.4234
0.5106	27.5372	26.9658	26.9892	27.2231	27.6728	27.3343
0.5045	27.4788	26.8926	26.9159	27.1492	27.5989	27.2600
0.4983	27.3918	26.8185	26.8417	27.0742	27.5238	27.1845
0.4953	27.3629	26.7827	26.8059	27.0380	27.4874	27.1481
0.4959	27.2471	26.7898	26.8130	27.0452	27.4947	27.1554
0.4771	27.1623	26.5665	26.5894	26.8188	27.2670	26.9274

of dimethyl ethers. A binary mixture of DEG-DBE + *n*-dodecane has the lowest thermal conductivity. Simple one-parameter equations have justified the experimental data successfully (6). The experimental results used for the comparative studies are discussed in terms of three body interactions and equation of state contributions to the mixing behaviour. Thermal conductivity decreases with an increase in temperature, as depicted in table 1. The required experimental data have been taken from the paper of Burgdorf *et al.* [6].

Diffusion coefficients (D) of the five binary mixtures (2-(2-methoxy ethoxy) ethanol + propylenecarbonate, carbontetrachloride + cyclohexane, carbontetrachloride + benzene, benzene + cyclohexane and *n*-heptane + toluene) have been computed at different compositions and constant temperature. A close perusal of table 2 shows that the smallest and the largest values of diffusion coefficients are obtained for 2-(2-methoxy ethoxy) ethanol propylene carbonate and *n*-heptane + toluene binary system, respectively. The diffusion coefficient, evaluated in conjunction with equations (1) and (3) predicts better results in most of the cases than others do. The close agreement between the theoretical and the experimental values indicates the success of the present approaches. Necessary data have been taken from earlier papers [11–13].

Transport properties are of great importance because of the well-established relationships between transport and thermodynamic properties [7] and industrial applications of liquids. Still the thermodynamic relations permit the predictions of viscosities, their activation energy self-diffusion coefficients and the thermal conductivities of liquids at ambient conditions, when certain generally available thermodynamic quantities of liquids are known.

4. Conclusions

Transport properties vary monotonically with compositions. The values of these properties decrease with an increase in temperature. The thermal conductivity and diffusion coefficient shows structural dependence behaviour of pure components of binary mixtures. The theoretical values evaluated using the present approaches are found in a fairly good agreement with the experimental findings.

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