CI&CEQ
Chemical Industry & Chemical Engineering Quarterly

Available on line at
Association of the Chemical Engineers of Serbia *AChE*
www.ache.org.rs/CICEQ

Chem. Ind. Chem. Eng. Q. 20 (3) 441–455 (2014) CI&CEQ

EMILA M. ŽIVKOVIĆ
MIRJANA LJ. KIJEVČANIN
IVONA R. RADOVIĆ
SLOBODAN P. ŠERBANOVIĆ

Faculty of Technology and Metallurgy, University of Belgrade, Belgrade, Serbia

SCIENTIFIC PAPER

UDC 547.284.3:544:66

DOI 10.2298/CICEQ130305026Z

VISCOSITIES AND REFRACTIVE INDICES OF BINARY SYSTEMS ACETONE+1-PROPANOL, ACETONE+1,2-PROPANEDIOL AND ACETONE+1,3-PROPANEDIOL

Article Highlights

- Binary mixtures of acetone with 1-propanol, 1,2-propanediol and 1,3-propanediol were analyzed
- Viscosity and refractive index were measured
- Deviations in viscosity and refractive index were calculated
- Modeling of viscosity and refractive index was performed

Abstract

Viscosities and refractive indices of three binary systems, acetone+1-propanol, acetone+1,2-propanediol and acetone+1,3-propanediol, were measured at eight temperatures (288.15, 293.15, 298.15, 303.15, 308.15, 313.15, 318.15 and 323.15 K) and at atmospheric pressure. From these data, viscosity deviations and deviations in refractive index were calculated and fitted to the Redlich-Kister equation. The viscosity modelling was done by two types of models: predictive UNIFAC-VISCO and ASOG VISCO and correlative Teja--Rice and McAlister equations. The refractive indices of binary mixtures were predicted by various mixing rules and compared with experimental data.

Keywords: viscosity, refractive index, experimental measurements, modelling.

Thermodynamic investigation of acetone and alcohols (or diols) binary mixtures is of great interest due to the complex molecular interactions [1] present in these mixtures and their diverse industrial application. Acetone has a wide range of practical applications in the pharmaceutical, cosmetic and food industry. In the chemical industry, acetone is used as a solvent for most plastics and synthetic fibers, including those used in laboratory bottles made of polystyrene, polycarbonate and some types of polypropylene. In the laboratory, acetone is used as a polar solvent in organic chemical reactions but also for rinsing laboratory glassware and instruments. Acetone is fluorescent under ultraviolet light, and its vapor may be used as a tracer in fluid flow experiments [2].

Correspondence: M.Lj. Kijevčanin, Faculty of Technology and Metallurgy, University of Belgrade, Karnegijeva 4, 11120 Belgrade, Serbia.
E-mail: mirjana@tmf.bg.ac.rs
Paper received: 5 March, 2013
Paper revised: 23 April, 2013
Paper accepted: 18 July, 2013

Alcohols, including 1-propanol, alone or in combination with other solvents, are widely used in the pharmaceutical and chemical industry [3-6], for pesticides, fats, oils, rubber, paints, varnishes, waxes, plastics, explosives, drugs, detergents, perfumes and cosmetics. Some alcohols (especially ethanol and 1-buthanol) are used as biofuels derived from biomass sources [7].

Alkanediols such as 1,2-propanediol or 1,3-propanediol have a variety of applications in automotive, aviation, explosives, textile, surface coatings, food, cosmetic, pharmaceutical, tobacco, petroleum and other industries [8]. They are also basic structural units for polyhydroxy compounds, which find widespread application as solvents, coolants, antifreezes, plasticizers, chemical intermediates, food additives and heat transfer fluids [9].

Both 1,2-propanediol and 1,3-propanediol are environmentally friendly, biodegradable and of very low human toxicity [10-12]. Due to the similarity of its properties, 1,2-propanediol is recommended as a replacement for more toxic ethylene glycol. 1,2-Propanediol finds application as a solvent in the pharma-

ceutical industry in many oral, injectable and topical formulations and in the cosmetic industry. In the food industry, 1,2-propanediol is used as a solvent for food colors and flavorings and as a food additive [10]. In the chemical industry, 1,3 propanediol is mainly used in the production of polymers, composites, adhesives, laminates and coatings [13]. It is also a solvent and used as antifreeze and wood paint.

In this work experimental dynamic viscosities and refractive indices are reported for three binary mixtures (acetone+1-propanol, acetone+1,2-propanediol and acetone+1,3-propanediol) at eight temperatures (288.15, 293.15, 298.15, 303.15, 308.15, 313.15, 318.15 and 323.15 K) and at atmospheric pressure. From these experimental data, deviations in viscosity ($\Delta\eta$) and deviations in refractive index (Δn_D) were calculated and correlated by the Redlich-Kister equation [14]. The predictive UNIFAC-VISCO [15,16] and ASOG-VISCO [17] models were used for modelling the viscosity of the investigated binary mixtures. The viscosity data were also correlated by Teja-Rice [18, 19] and McAlister [20] correlation equations. The refractive indices of binary mixtures were calculated by different mixing rules (Lorentz-Lorenz, Dale-Gladstone, Eykman, Arago-Biot, Newton, and Oster [21]) and compared with experimental data.

EXPERIMENTAL

Chemicals

Acetone (99.8 mass%) and 1-propanol (99.5 mass%) were supplied by Merck, 1,2-propanediol (99.5 mass%) and 1,3-propanediol (98.0 mass%) were supplied by Fluka. Chemicals were kept in dark bottles, in an inert atmosphere and ultrasonically degassed before a sample preparation. In Table 1, dynamic viscosities and refractive indices of pure substances are compared with literature values at 298.15 K [4,22-24]. The agreement between literature and our experimental values is very good with differences within 2×10^{-2} mPa·s for viscosity measurements of less viscous fluids and in most cases within 6×10^{-4} for refractive indices. The differences between experimental and literature values of viscosity are somewhat higher by absolute value for more viscous fluids, especially 1,3-propanediol, but still below 2% deviation.

Apparatus and procedures

Viscosity measurements were performed using a digital Stabinger viscometer (model SVM 3000/G2). The instrument contains two measuring cells; one of them is used for measuring the density of the sample, while the other one measures dynamic viscosity. The kinematic viscosity is calculated from the measured density and dynamic viscosity. The stated reproducibility of the dynamic viscosity and density measurements is 0.35% and 0.5 kg m^{-3} in the temperature interval 288.15–378.15 K. During this procedure temperature in the cell was regulated to ±0.01 K with a built in solid-state thermostat. The relative uncertainty in dynamic viscosity measurements was estimated to be within ±0.40%.

Refractive index measurements were performed using an automatic Anton Paar RXA-156 refractometer, which works with the wavelength of 589 nm. Throughout this procedure temperature of the sample was kept constant with a built-in thermostat within an accuracy of ±0.03 K. The refractive index data have the uncertainty of ±0.00005 units.

A Mettler AG 204 balance, with a precision 1×10^{-4}, was used for precise measuring of mass composition for all binary mixtures, using the cell and the procedure described previously [25]. The uncertainty of the mole fraction calculation was less than $\pm1\times10^{-4}$.

RESULTS AND DISCUSSION

The experimental data of viscosity, η, refractive index, n_D, as well as the calculated values of viscosity deviation, $\Delta\eta$, and deviations in refractive index, Δn_D, for three binary systems (acetone+1-propanol, acetone+1,2-propanediol and acetone+1,3-propanediol) at eight temperatures (288.15, 293.15, 298.15, 303.15, 308.15, 313.15, 318.15 and 323.15 K) and at atmospheric pressure are reported in Table 2.

Table 1. Dynamic viscosities η *and refractive indices* n_D *of pure components at 298.15 K*

Component	η/mPa· s		n_D	
	Exp.	Literature	Exp.	Literature
Acetone	0.31858	0.302[22]	1.35622	1.35596[4]
		0.304[4]		
1-Propanol	1.9503	1.943 [23]	1.38369	1.3837 [4]
1,2-Propanediol	43.437	43.428 [24]	1.43081	1.4314 [4]
1,3-Propanediol	41.041	40.067 [24]	1.43805	1.4386 [4]

Table 2. Viscosity, η, refractive index, n_D, viscosity deviation, $\Delta\eta$, and deviations in refractive index, Δn_D, for the binary mixtures acetone+1-propanol, acetone+1,2-propanediol and acetone+1,3-propanediol

x_1	η /mPas	$\Delta\eta$ /mPas	n_D	Δn_D
Acetone (1) + 1-propanol (2)				
		288.15 K		
0.0000	2.5139	-	1.38780	-
0.0998	1.7806	-0.5160	1.38547	0.0003
0.1998	1.2992	-0.7796	1.38312	0.0006
0.3000	0.99045	-0.8701	1.38071	0.0008
0.4001	0.79067	-0.8519	1.37815	0.0008
0.4999	0.65812	-0.7671	1.37549	0.0008
0.6000	0.54172	-0.6655	1.37271	0.0006
0.6998	0.47064	-0.5192	1.36988	0.0004
0.8002	0.42313	-0.3481	1.36703	0.0002
0.9000	0.38431	-0.1696	1.36428	0.00003
1.0000	0.33611	-	1.36163	-
		293.15 K		
0.0000	2.2085	-	1.38576	-
0.0998	1.5942	-0.4266	1.38339	0.0003
0.1998	1.1800	-0.6527	1.38089	0.0005
0.3000	0.91131	-0.7329	1.37833	0.0006
0.4001	0.73501	-0.7210	1.37570	0.0007
0.4999	0.61748	-0.6508	1.37301	0.0007
0.6000	0.51373	-0.5663	1.37023	0.0006
0.6998	0.44932	-0.4430	1.36735	0.0004
0.8002	0.40695	-0.2965	1.36442	0.0001
0.9000	0.37277	-0.1430	1.36155	-0.0001
1.0000	0.32765	-	1.35894	-
		298.15 K		
0.0000	1.9503	-	1.38369	-
0.0998	1.4327	-0.3548	1.38119	0.0002
0.1998	1.0750	-0.5493	1.37862	0.0004
0.3000	0.84069	-0.6201	1.37599	0.0005
0.4001	0.68440	-0.6130	1.37331	0.0006
0.4999	0.58006	-0.5545	1.37055	0.0006
0.6000	0.48754	-0.4837	1.36771	0.0005
0.6998	0.42897	-0.3795	1.36476	0.0003
0.8002	0.39103	-0.2536	1.36175	0.00004
0.9000	0.35307	-0.1287	1.35883	-0.0001
1.0000	0.31858	-	1.35622	-
		303.15 K		
0.0000	1.7270	-	1.38162	-
0.0998	1.2920	-0.2935	1.37906	0.0003
0.1998	0.98217	-0.4615	1.37638	0.0004
0.3000	0.77739	-0.5242	1.37368	0.0005
0.4001	0.63794	-0.5218	1.37091	0.0006
0.4999	0.54561	-0.4726	1.36810	0.0006
0.6000	0.46286	-0.4134	1.36520	0.0005
0.6998	0.40941	-0.3253	1.36217	0.0003
0.8002	0.37514	-0.2173	1.35907	-0.00002
0.9000	0.33997	-0.1109	1.35609	-0.0002

Table 2. Continued

x_1	η /mPas	$\Delta\eta$ /mPas	n_D	Δn_D
Acetone (1) + 1-propanol (2)				
		303.15 K		
1.0000	0.30910	-	1.35346	-
		308.15 K		
0.0000	1.5419	-	1.37918	-
0.0998	1.1689	-0.2490	1.37656	0.0002
0.1998	0.89942	-0.3942	1.37384	0.0003
0.3000	0.72028	-0.4488	1.37111	0.0005
0.4001	0.59495	-0.4498	1.36833	0.0005
0.4999	0.51297	-0.4077	1.36548	0.0005
0.6000	0.43889	-0.3574	1.36255	0.0005
0.6998	0.39107	-0.2812	1.35949	0.0002
0.8002	0.35995	-0.1876	1.35634	-0.0001
0.9001	0.32207	-0.1014	1.35332	-0.0002
1.0000	0.29925	-	1.35072	-
		313.15 K		
0.0000	1.3783	-	1.37695	-
0.0998	1.0605	-0.2091	1.37423	0.0002
0.1998	0.82501	-0.3357	1.37147	0.0003
0.3000	0.66839	-0.3831	1.36870	0.0005
0.4001	0.55377	-0.3887	1.36588	0.0005
0.4999	0.48153	-0.3522	1.36300	0.0006
0.6000	0.41576	-0.3090	1.36001	0.0005
0.6998	0.37335	-0.2427	1.35687	0.0002
0.8002	0.34485	-0.1618	1.35364	-0.0001
0.9000	0.30095	-0.0970	1.35055	-0.0003
1.0000	0.28903	-	1.34795	-
		318.15 K		
0.0000	1.2365	-	1.37469	-
0.0998	0.9672	-0.1737	1.37186	0.0001
0.1998	0.75829	-0.2868	1.36902	0.0003
0.3000	0.62057	-0.3285	1.36620	0.0004
0.4001	0.51467	-0.3385	1.36332	0.0005
0.4999	0.44951	-0.3080	1.36038	0.0005
0.6000	0.39197	-0.2697	1.35730	0.0004
0.6998	0.35427	-0.2118	1.35407	0.0002
0.8002	0.32858	-0.1413	1.35079	-0.0001
0.9000	0.28746	-0.0868	1.34765	-0.0003
1.0000	0.27842	-	1.34499	-
		323.15 K		
0.0000	1.1241	-	1.37231	-
0.0998	0.88788	-0.1507	1.36939	0.0001
0.1998	0.69722	-0.2557	1.36650	0.0002
0.3000	0.57604	-0.2910	1.36363	0.0004
0.4001	0.47693	-0.3044	1.36072	0.0005
0.4999	0.41949	-0.2763	1.35772	0.0005
0.6000	0.36606	-0.2440	1.35459	0.0004
0.6998	0.33173	-0.1928	1.35131	0.0001

Table 2. Continued

x_1	η /mPas	$\Delta\eta$ /mPas	n_D	Δn_D
Acetone (1) + 1-propanol (2)				
		323.15 K		
0.8002	0.31556	-0.1230	1.34797	-0.0002
0.9000	0.27902	-0.0740	1.34479	-0.0003
1.0000	0.26733	-	1.34210	-
Acetone (1) + 1,2 propanediol (2)				
		288.15 K		
0.0000	81.206	-	1.43411	-
0.1000	39.825	-33.294	1.42939	0.0025
0.2000	18.409	-46.623	1.42332	0.0037
0.2998	8.7428	-48.219	1.41695	0.0046
0.3999	4.6465	-44.220	1.41012	0.0050
0.5004	2.7258	-38.013	1.40294	0.0051
0.5999	1.7303	-30.962	1.39558	0.0050
0.6999	1.0193	-23.586	1.38811	0.0047
0.8000	0.63096	-15.879	1.38008	0.0040
0.9001	0.49360	-7.9214	1.37142	0.0025
1.0000	0.33611	-	1.36163	-
		293.15 K		
0.0000	58.753	-	1.43247	-
0.1000	29.944	-22.967	1.42765	0.0025
0.2000	14.459	-32.609	1.42147	0.0037
0.2998	7.1453	-34.092	1.41499	0.0046
0.3999	3.8748	-31.514	1.40815	0.0051
0.5004	2.3634	-27.154	1.40081	0.0051
0.5999	1.5324	-22.171	1.39337	0.0050
0.6999	0.93400	-16.927	1.38580	0.0048
0.8000	0.60104	-11.412	1.37772	0.0041
0.9001	0.46710	-5.6972	1.36890	0.0026
1.0000	0.32765	-	1.35894	-
		298.15 K		
0.0000	43.437	-	1.43081	-
0.1000	22.953	-16.172	1.42589	0.0025
0.2000	11.537	-23.276	1.41960	0.0037
0.2998	5.9181	-24.592	1.41304	0.0046
0.3999	3.3029	-22.891	1.40617	0.0052
0.5004	2.0662	-19.794	1.39872	0.0052
0.5999	1.3556	-16.215	1.39117	0.0051
0.6999	0.85967	-12.399	1.38345	0.0048
0.8000	0.56884	-8.3734	1.37528	0.0041
0.9001	0.44630	-4.1798	1.36628	0.0026
1.0000	0.31858	-	1.35622	-
		303.15 K		
0.0000	32.754	-	1.42915	-
0.1000	17.903	-11.607	1.42410	0.0025
0.2000	9.3403	-16.925	1.41772	0.0037
0.2998	4.9645	-18.063	1.41104	0.0046
0.3999	2.8822	-16.897	1.40412	0.0052

Table 2. Continued

x_1	η /mPas	$\Delta\eta$ /mPas	n_D	Δn_D
Acetone (1) + 1,2 propanediol (2)				
		303.15 K		
0.5004	1.8207	-14.698	1.39663	0.0053
0.5999	1.2013	-12.089	1.38895	0.0052
0.6999	0.79430	-9.2515	1.38115	0.0050
0.8000	0.53792	-6.2602	1.37283	0.0042
0.9001	0.42092	-3.1294	1.36368	0.0027
1.0000	0.30910	-	1.35346	-
		308.15 K		
0.0000	25.156	-	1.42751	-
0.1000	14.187	-8.4833	1.42230	0.0025
0.2000	7.6623	-12.522	1.41578	0.0036
0.2998	4.2078	-13.496	1.40905	0.0046
0.3999	2.5499	-12.666	1.40197	0.0052
0.5004	1.6137	-11.104	1.39440	0.0053
0.5999	1.1033	-9.1411	1.38666	0.0052
0.6999	0.73563	-7.0231	1.37885	0.0051
0.8000	0.52083	-4.7498	1.37043	0.0044
0.9001	0.39528	-2.3872	1.36117	0.0028
1.0000	0.29925	-	1.35072	-
		313.15 K		
0.0000	19.646	-	1.42581	-
0.1000	11.408	-6.3023	1.42051	0.0025
0.2000	6.3619	-9.4127	1.41388	0.0036
0.2998	3.6019	-10.241	1.40702	0.0046
0.3999	2.1892	-9.7159	1.39978	0.0051
0.5004	1.4396	-8.5202	1.39222	0.0054
0.5999	0.99244	-7.0413	1.38437	0.0053
0.6999	0.68217	-5.4159	1.37648	0.0052
0.8000	0.49449	-3.6659	1.36796	0.0044
0.9001	0.37701	-1.8458	1.35854	0.0028
1.0000	0.28903	-	1.34795	-
		318.15 K		
0.0000	15.581	-	1.42407	-
0.1000	9.2973	-4.7534	1.41870	0.0025
0.2000	5.3408	-7.1797	1.41194	0.0037
0.2998	3.1092	-7.8841	1.40496	0.0046
0.3999	1.9707	-7.4908	1.39752	0.0051
0.5004	1.2979	-6.6257	1.38979	0.0053
0.5999	0.89437	-5.5066	1.38206	0.0054
0.6999	0.63173	-4.2390	1.37396	0.0052
0.8000	0.47309	-2.8659	1.36537	0.0046
0.9001	0.36908	-1.4381	1.35585	0.0030
1.0000	0.27842	-	1.34499	-
		323.15 K		
0.0000	12.535	-	1.42230	-
0.1000	7.6715	-3.6367	1.41687	0.0026
0.2000	4.5290	-5.5525	1.40995	0.0037

Table 2. Continued

x_1	η /mPas	$\Delta\eta$ /mPas	n_D	Δn_D
Acetone (1) + 1,2 propanediol (2)				
		323.15 K		
0.2998	2.7102	-6.1470	1.40296	0.0047
0.3999	1.7173	-5.9119	1.39520	0.0050
0.5004	1.1749	-5.2214	1.38743	0.0053
0.5999	0.81498	-4.3606	1.37958	0.0054
0.6999	0.58126	-3.3676	1.37143	0.0053
0.8000	0.45332	-2.2675	1.36278	0.0046
0.9001	0.34767	-1.1452	1.35315	0.0030
1.0000	0.26733	-	1.34210	-
Acetone (1) + 1,3 propanediol (2)				
		288.15 K		
0.0000	66.527	-	1.44089	-
0.1008	36.589	-23.266	1.43524	0.0023
0.1914	21.190	-32.668	1.42960	0.0039
0.3021	10.972	-35.559	1.42281	0.0059
0.4009	6.1318	-33.859	1.41595	0.0068
0.5100	3.3985	-29.371	1.40744	0.0070
0.6004	1.9768	-24.809	1.39989	0.0066
0.7000	1.5738	-18.620	1.39053	0.0051
0.7983	1.3466	-12.340	1.38113	0.0035
0.9002	1.1853	-5.7556	1.37114	0.0016
1.0000	0.33611	-	1.36163	-
		293.15 K		
0.0000	51.913	-	1.43947	-
0.1008	29.203	-17.511	1.43367	0.0023
0.1914	17.284	-24.756	1.42795	0.0039
0.3021	9.1621	-27.167	1.42101	0.0059
0.4009	5.2293	-26.003	1.41403	0.0068
0.5100	2.9423	-22.662	1.40542	0.0070
0.6004	1.7643	-19.177	1.39775	0.0066
0.7000	1.3209	-14.482	1.38837	0.0053
0.7983	1.1797	-9.5528	1.37869	0.0035
0.9002	1.0405	-4.4353	1.36861	0.0016
1.0000	0.32765	-	1.35894	-
		298.15 K		
0.0000	41.041	-	1.43805	-
0.1008	23.553	-13.383	1.43206	0.0023
0.1914	14.219	-19.028	1.42629	0.0039
0.3021	7.7206	-21.018	1.41919	0.0059
0.4009	4.4957	-20.220	1.41210	0.0069
0.5100	2.5672	-17.705	1.40341	0.0071
0.6004	1.5834	-15.008	1.39556	0.0066
0.7000	1.1396	-11.396	1.38609	0.0053
0.7983	1.0380	-7.4943	1.37629	0.0036
0.9002	0.91743	-3.4652	1.36605	0.0017
1.0000	0.31858	-	1.35622	-

Table 2. Continued

x_1	η /mPas	$\Delta\eta$ /mPas	n_D	Δn_D
Acetone (1) + 1,3 propanediol (2)				
		303.15 K		
0.0000	32.843	-	1.43665	-
0.1008	19.212	-10.351	1.43047	0.0022
0.1914	11.815	-14.801	1.42463	0.0039
0.3021	6.5535	-16.461	1.41735	0.0058
0.4009	3.8938	-15.906	1.41013	0.0068
0.5100	2.2566	-13.994	1.40137	0.0071
0.6004	1.4281	-11.882	1.39333	0.0066
0.7000	1.0313	-9.0380	1.38367	0.0053
0.7983	0.91728	-5.9539	1.37396	0.0037
0.9002	0.81225	-2.7437	1.36348	0.0017
1.0000	0.30910	-	1.35346	-
		308.15 K		
0.0000	26.580	-	1.43526	-
0.1008	15.863	-8.0683	1.42892	0.0022
0.1914	9.9219	-11.628	1.42296	0.0039
0.3021	5.6090	-13.032	1.41552	0.0058
0.4009	3.3955	-12.649	1.40816	0.0068
0.5100	1.9968	-11.180	1.39929	0.0071
0.6004	1.2934	-9.5076	1.39126	0.0068
0.7000	0.93314	-7.2503	1.38142	0.0053
0.7983	0.81384	-4.7862	1.37164	0.0039
0.9002	0.72199	-2.2001	1.36098	0.0018
1.0000	0.29925	-	1.35072	-
		313.15 K		
0.0000	21.737	-	1.43387	-
0.1008	13.224	-6.3514	1.42740	0.0022
0.1914	8.4161	-9.2158	1.42126	0.0038
0.3021	4.8498	-10.408	1.41364	0.0057
0.4009	2.9793	-10.159	1.40612	0.0067
0.5100	1.7800	-9.0185	1.39709	0.0070
0.6004	1.1752	-7.6844	1.38908	0.0068
0.7000	0.84417	-5.8793	1.37914	0.0054
0.7983	0.72482	-3.8903	1.36916	0.0039
0.9002	0.64420	-1.7853	1.35843	0.0019
1.0000	0.28903	-	1.34795	-
		318.15 K		
0.0000	17.950	-	1.43244	-
0.1008	11.128	-5.0408	1.42585	0.0022
0.1914	7.1910	-7.3767	1.41960	0.0039
0.3021	4.2151	-8.3964	1.41170	0.0057
0.4009	2.6333	-8.2322	1.40413	0.0068
0.5100	1.5966	-7.3409	1.39497	0.0071
0.6004	1.0699	-6.2701	1.38672	0.0068
0.7000	0.76316	-4.8167	1.37673	0.0055
0.7983	0.64789	-3.1949	1.36664	0.0040
0.9002	0.57680	-1.4652	1.35570	0.0020

Table 2. Continued

x_1	η /mPas	$\Delta\eta$ /mPas	n_D	Δn_D
Acetone (1) + 1,3 propanediol (2)				
		318.15 K		
1.0000	0.27842	-	1.34499	-
		323.15 K		
0.0000	14.958	-	1.43099	-
0.1008	9.4598	-4.0174	1.42424	0.0022
0.1914	6.1981	-5.9481	1.41787	0.0039
0.3021	3.6787	-6.8413	1.40988	0.0057
0.4009	2.3430	-6.7255	1.40211	0.0068
0.5100	1.4330	-6.0328	1.39276	0.0071
0.6004	0.97369	-5.1640	1.38442	0.0068
0.7000	0.69461	-3.9799	1.37423	0.0055
0.7983	0.58114	-2.6493	1.36439	0.0044
0.9002	0.51825	-1.2152	1.35310	0.0021
1.0000	0.26733	-	1.34210	-

The viscosity deviations, $\Delta\eta$, were calculated from the viscosity of the mixture, η, and pure component *i*, η_i, according to the equation:

$$\Delta\eta = \eta - \sum_{i=1}^{n} x_i \eta_i \tag{1}$$

where n is the number of components and x_i is the mole fraction of the component *i*.

The deviations in refractive index were calculated from the equation:

$$\Delta n_D = n_D - \sum_{i=1}^{n} x_i n_{Di} \tag{2}$$

where n_D and n_{Di} refers to the refractive index of the mixture and the pure component *i*, respectively. In all the above given equations n denotes the number of components.

Viscosity deviations, $\Delta\eta$, and deviations in refractive index, Δn_D, were correlated with the Redlich-Kister (RK) equation [14]:

$$Y = x_i x_j \sum_{p=0}^{k} A_p (2x_i - 1)^p \tag{3}$$

where Y represents the binary viscosity deviation, $\Delta\eta$, or deviation in refractive index Δn_D of the mixture, A_p are fitting parameters, and k+1 is the number of parameters, which was optimized using the *F*-test. The quality of correlation was assessed by calculating the root-mean-square deviations (rmsd), σ, defined as:

$$\sigma = \left(\sum_{i=1}^{m} \left(Y_{exp} - Y_{cal} \right)^2 / m \right)^{1/2} \tag{4}$$

where m is the number of experimental data points.

The viscosity deviations obtained from experimental viscosity data, together with the curves calculated from the Redlich-Kister equation are shown in Figure 1. Although values of $\Delta\eta$ are negative over the entire temperature and composition range for all investigated binary mixtures, the systems with diols are characterized with much higher negative viscosity deviations than system with 1-propanol. Increase in temperature decreases the viscosity deviation negative values for all investigated binary mixtures.

The deviation of refractive indices for acetone+1-propanol, acetone+1,2-propanediol and acetone+1,3-propanediol binary systems, together with the curves calculated from the Redlich-Kister equation are displayed in Figure 2.

In mixtures with diols, the Δn_D values are positive over the whole range of mixture compositions and at all temperatures. For acetone+1-propanol binary system, the deviation of refractive index is positive for lower mole fraction of acetone but turns negative in the acetone rich region. Comparison with literature data [26] of Δn_D values for acetone+1-propanol binary system shows that two sets of data follow the same trend line with positive maximum between 0.3 and 0.4 and negative minimum around 0.9 mole fractions of acetone. The temperature effect on refractive indices deviations is relatively weak, particularly in the solutions with 1,3-propandiol.

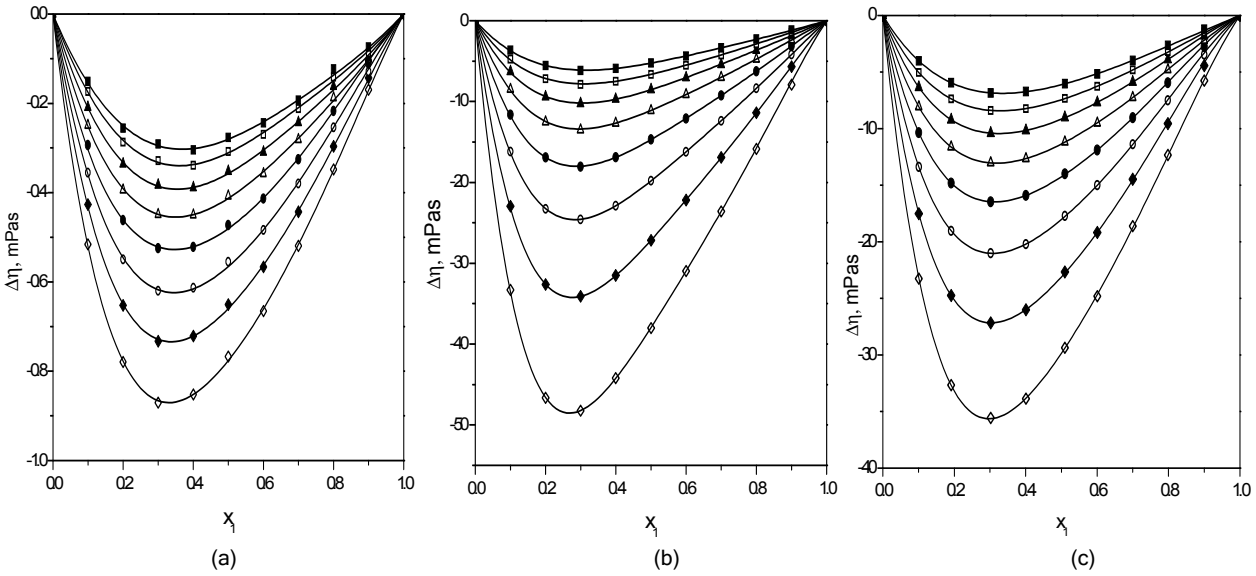

Figure 1. Experimental values of viscosity deviation Δη as a function of acetone molar fraction x_1 for the systems: a) acetone+1-propanol; b) acetone+1,2-propanediol; c) acetone+1,3-propanediol; at following temperatures: (◊) 288.15, (♦) 293.15, (○) 298.15, (●) 303.15, (Δ) 308.15, (▲) 313.15, (□) 318.15 and (■) 323.15 K; (——) RK equation.

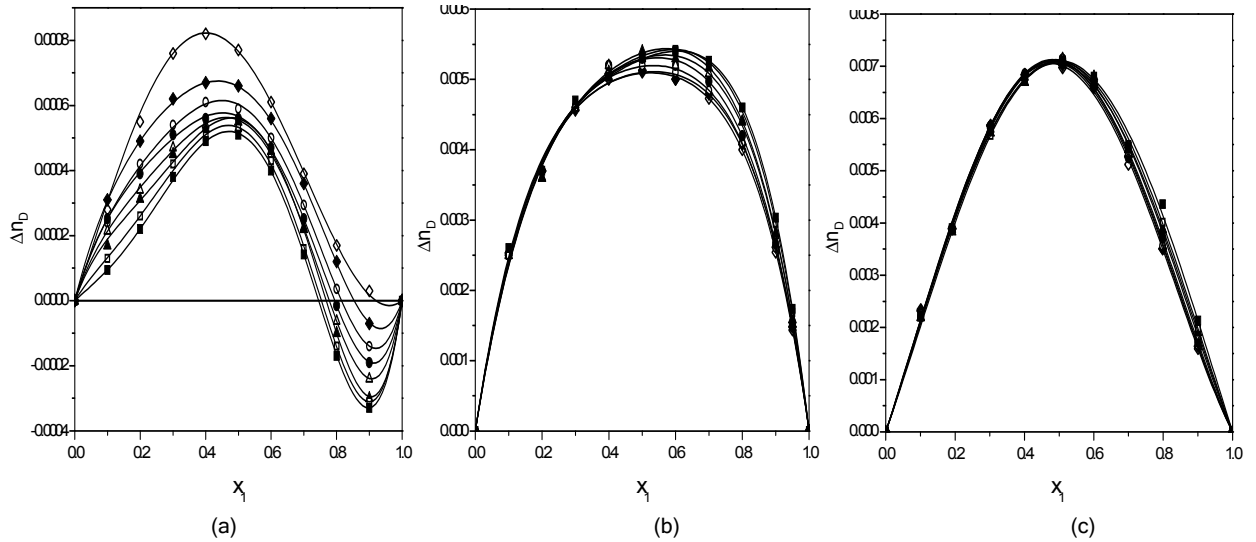

Figure 2. Experimental values of deviation in refractive index Δn_D as a function of acetone molar fraction x_1 for the systems a) acetone+1-propanol; b) acetone+1,2-propanediol; c) acetone+1,3-propanediol;at following temperatures: (◊) 288.15, (♦) 293.15, (○) 298.15, (●) 303.15, (Δ) 308.15, (▲) 313.15, (□) 318.15 K and (■) 323.15 K; (——) RK equation.

It has been stated earlier [1,22,27] that excess thermodynamic properties of mixtures are the consequence of three main types of contributions:

a) physical interaction, such as dispersion forces or week dipole-dipole interactions, resulting in negative contributions to excess molar volume and viscosity deviation,

b) chemical interaction like hydrogen bonds or other complex formation leading to negative contributions to V^E and positive to $\Delta\eta$ and

c) structural interactions due to interstitial accommodation and changes in free volume.

Although straightforward relation between viscosity deviation and molecular interactions was established for a number of solutions [22], excess molar volume is usually a more convenient property for molecular interactions analysis. It has already been pointed out in our previous work [1] that for the mixture of acetone with 1-propanol excess molar volumes are mostly negative, although very small by absolute value (Figure 3a). For this solution, values of V^E were explained as a balance between positive and negative contributions. Positive contributions which are dominant for higher acetone concentrations occur

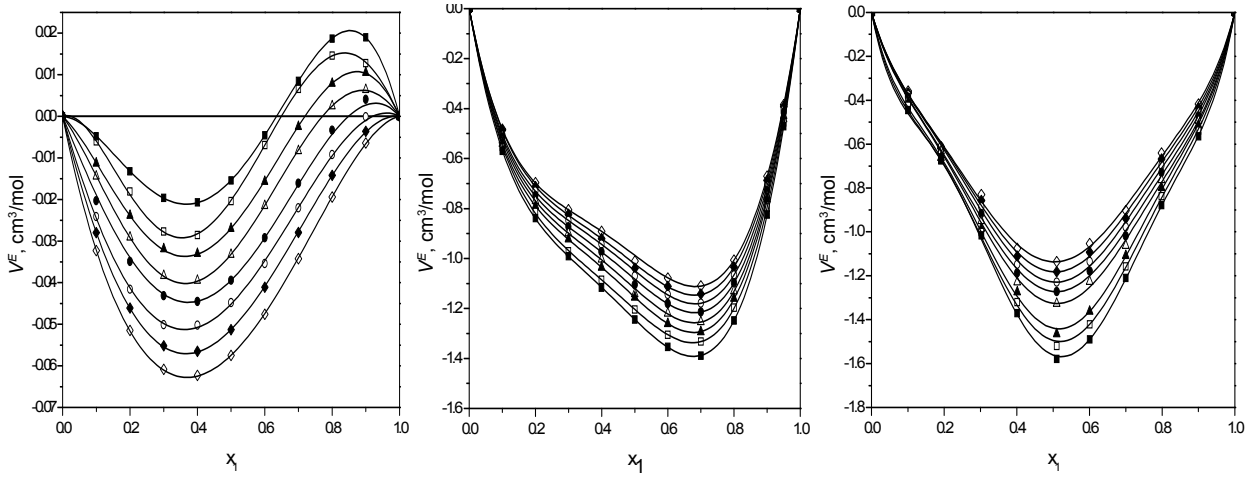

Figure 3. Experimental values of excess molar volume V^E as a function of acetone molar fraction x_1 for the systems: a) acetone+1-propanol; b) acetone+1,2-propanediol; c) acetone+1,3-propanediol; at following temperatures: (◊) 288.15, (♦) 293.15, (○) 298.15, (●) 303.15, (Δ) 308.15, (▲) 313.15, (□) 318.15 and (■) 323.15 K; (——) RK equation.

due to a H-bond rupture of 1-propanol associates and considerable difference in molecular volume between 1-propanol and acetone that leads to important steric hindrance. Negative contributions are a result of intermolecular OH and C=O interactions and possible geometrical fitting between unlike molecules leading to slightly negative values of V^E in the alcohol rich region. Values of V^E for the mixtures of acetone with 1,2-propanediol and 1,3-propanediol are large and negative (Figures 3b and 3c) thus indicating interaction by hydrogen bonding between C=O group of acetone and OH groups of diols. The minimum of the curve for the mixture with 1,2-propanediol is located near 0.7 acetone mole fraction while the curve with for the mixture is almost symmetrical. This phenomenon could be attributed to structural factors and the presence of vicinal hydroxyl group in the molecule of 1,2-propanediol [1].

According to [22] predominance of chemical interactions leading to H-bond formation is expected to result in positive viscosity deviation. Negative deviations obtained for all investigated mixtures indicate that the strength of the hydrogen bonds is not the only factor influencing viscosity deviation or excess molar volume negative values. It was stated earlier [28] that molecular size and shape of the components are important factors and negative values of $\Delta\eta$ could be expected for systems of different molecular size in which the dispersion forces are dominant. Explanation for negative excess molar volumes and negative viscosity deviations could be attributed to the structural factors, inclusion of acetone molecules in the aggregates interstices, thus resulting in fewer surfaces available for friction and reduction of mixture viscosity [29]. For all analyzed binary mixtures, viscosity deviation negative values decrease with temperature rise, probably due to higher intermolecular vibrations and thermal motion.

The refractive indices and their deviations are intrinsically related to dispersion attractive interactions [30,31]. Since the deviations obtained within this work are mostly positive for all of the studied solutions, it can be concluded that the dispersion interactions between the unlike molecules are higher than those in the pure components.

Viscosity modeling was performed with two types of models: predictive and correlative. Predictive approach for viscosity determination is mostly based on group contribution models. In this paper UNIFAC--VISCO [15,16] and ASOG-VISCO [17] predictive models were used for calculating dynamic viscosities of the selected binary mixtures. In addition, viscosity data have been correlated with one-parameter Teja and Rice [18,19], and McAllister [20] two-parameter Three-body and three-parameter Four-body models. More details about the models used in this work can be found in our previous papers [32,33].

The ability of these models to predict dynamic viscosities of selected binary mixtures and to correlate experimental viscosity data was tested by calculating the percentage deviations ($PD_{\max}$) between the experimental and the calculated viscosities from the following equation:

$$PD_{\max}(Y) = \frac{100}{n}\sum_{i=1}^{m}\left|\frac{Y_{\exp}-Y_{\mathrm{cal}}}{\left(Y_{\exp}\right)_{\max}}\right| \tag{5}$$

where $\left(Y_{\text{exp}}\right)_{\text{max}}$ is the maximum of experimental η values.

The results obtained by UNIFAC-VISCO, ASOG-VISCO, Teja-Rice and McAlister models for the binary mixtures of acetone + 1-propanol, +1,2-propanediol and+1,3-propanediol over the investigated temperature range are given in Table 3.

The predictive UNIFAC-VISCO model gives satisfactory results for all analyzed systems with maximum percentage deviation (PD_{max}), in almost all investigated cases less than 10%. The best results are obtained for the acetone+1-propanol system with PD_{max} below 4%. Results obtained with the ASOG-VISCO predictive model are even better, with maximum percentage deviation in most cases less than 5% and for acetone+1,3-propanediol binary mixture less than 1.3% at all investigated temperatures.

Two parameter McAlister-3 and three parameter McAlister-4 correlative models give very good results for all analyzed systems and at all temperatures with maximum percentage deviation (PD_{max}) bellow 1%. Results obtained with the one-parameter Teja-Rice model are characterized with slightly higher values of PD_{max} but still below 2% for all analyzed binary mixtures. The interaction parameters for correlative models, presented in Table 4, were determined from experimental data using the Marquardt [34] optimization technique.

The results for refractive indices obtained by Lorentz-Lorenz (L-L), Dale-Gladstone (D-G), Eykman (Eyk), Arago-Biot (A-B), Newton (New), and Oster (Ost) [21] mixing rules for acetone+1-propanol, acetone+ +1,2-propanediol and acetone+1,3-propanediol binary mixtures over the investigated temperature range are summarized in Table 5. The predictive ability of these models was tested by calculating the percentage deviations (PD_{max}) from Eq. (5), where Y_{exp} and Y_{cal} denotes experimental and calculated values of refractive index and $\left(Y_{\text{exp}}\right)_{\text{max}}$ is the maximum of experimental n_{D} values. From the obtained results it can be con-

Table 3. Results of viscosity prediction and correlation (PD_{max} / %) for the binary mixtures acetone+1-propanol, acetone+1,2-propanediol and acetone+1,3-propanediol at temperatures 288.15-323.15 K and atmospheric pressure

T / K	Predictive approach		Correlative models		
	UNIFAC-VISCO	ASOG-VISCO	Teja-Rice	McAlister-3	McAlister-4
			Acetone (1) + propanol (2)		
288.15	1.36	5.28	1.90	0.36	0.31
293.15	1.52	4.51	1.81	0.37	0.31
298.15	1.98	3.90	1.80	0.36	0.30
303.15	2.55	3.40	1.77	0.37	0.30
308.15	2.99	2.88	1.57	0.26	0.24
313.15	3.45	2.71	1.58	0.38	0.37
318.15	3.91	2.62	1.39	0.47	0.44
323.15	3.91	3.00	1.11	0.75	0.77
			Acetone (1) + 1,2-propandiol (2)		
288.15	3.72	5.09	1.41	0.75	0.26
293.15	4.04	4.42	1.32	0.75	0.26
298.15	4.49	3.87	1.21	0.75	0.27
303.15	5.02	3.40	1.08	0.75	0.28
308.15	5.69	2.82	0.99	0.76	0.30
313.15	6.02	2.74	0.88	0.75	0.27
318.15	6.80	2.23	0.78	0.73	0.29
323.15	7.25	2.01	0.70	0.72	0.24
			Acetone (1) + 1,3-propandiol (2)		
288.15	8.88	1.29	0.86	0.76	0.72
293.15	9.16	0.93	0.88	0.77	0.72
298.15	9.42	0.76	0.80	0.72	0.69
303.15	9.73	0.89	0.79	0.74	0.71
308.15	9.95	0.97	0.66	0.59	0.59
313.15	10.27	1.11	0.65	0.56	0.55
318.15	10.49	1.11	0.58	0.40	0.38
323.15	10.73	1.10	0.66	0.26	0.25

Table 4. Binary interaction parameters in correlative models for viscosity determination

T / K	Teja-Rice	McAlister-3		McAlister-4		
	ψ_{12}	υ_{12}	υ_{21}	υ_{1112}	υ_{2221}	υ_{1122}
Acetone (1) + propanol (2)						
288.15	-0.0360	0.5834	0.8844	0.5694	1.2370	0.6754
293.15	-0.0027	0.5585	0.8339	0.5523	1.1518	0.6356
298.15	0.0238	0.5359	0.7899	0.5333	1.0757	0.6096
303.15	0.0538	0.5159	0.7433	0.5103	1.0003	0.5794
308.15	0.0786	0.4898	0.7048	0.4735	0.9230	0.5701
313.15	0.1000	0.4654	0.6677	0.4465	0.8576	0.5503
318.15	0.1002	0.4379	0.6353	0.4295	0.8114	0.5110
323.15	0.1012	0.3836	0.6231	0.3668	0.7600	0.5060
Acetone (1) + 1,2-propandiol (2)						
288.15	-0.5381	0.4552	7.7193	1.8013	15.6106	0.9294
293.15	-0.4999	0.4600	6.3186	1.5069	12.3293	0.8947
298.15	-0.4597	0.4668	5.2357	1.2993	9.9038	0.8590
303.15	-0.4160	0.4759	4.3919	1.1246	8.0510	0.8391
308.15	-0.3507	0.4902	3.7142	1.0200	6.6262	0.8155
313.15	-0.3227	0.4752	3.2125	0.9014	5.5638	0.7718
318.15	-0.2688	0.4739	2.7785	0.8221	4.6976	0.7486
323.15	-0.2294	0.4553	2.4481	0.7445	4.0342	0.7065
Acetone (1) + 1,3-propandiol (2)						
288.15	0.2219	1.2911	9.1834	1.7702	15.8994	2.9980
293.15	0.2164	1.1817	7.7151	1.5548	13.0811	2.1976
298.15	0.2068	1.0774	6.5584	1.3336	10.8603	2.0085
303.15	0.2012	0.9873	5.6245	1.2099	9.1674	1.7910
308.15	0.1880	0.8897	4.8974	1.0094	7.7517	1.6798
313.15	0.1824	0.8127	4.2960	0.9095	6.6764	1.5225
318.15	0.1592	0.7308	3.8177	0.7528	5.7421	1.4558
323.15	0.1303	0.6524	3.4365	0.6387	5.0193	1.3680

Table 5. Results for refractive index n_D prediction (PD_{max} / %) for binary mixtures acetone+1-propanol, acetone+1,2-propanediol and acetone+1,3-propanediol at temperatures 288.15-323.15 K and atmospheric pressure

T / K	L-L	D-G	Eyk	A-B	New	Ost
Acetone (1) + propanol (2)						
288.15	0.03	0.03	0.03	0.03	0.03	0.03
293.15	0.03	0.03	0.03	0.03	0.02	0.02
298.15	0.03	0.02	0.02	0.02	0.02	0.02
303.15	0.03	0.02	0.02	0.02	0.02	0.02
308.15	0.02	0.02	0.02	0.02	0.02	0.02
313.15	0.02	0.02	0.02	0.02	0.02	0.02
318.15	0.03	0.02	0.03	0.02	0.02	0.02
323.15	0.02	0.02	0.02	0.02	0.02	0.02
Acetone (1) + 1,2-propandiol (2)						
288.15	0.29	0.27	0.27	0.27	0.24	0.25
293.15	0.29	0.27	0.28	0.27	0.25	0.26
298.15	0.30	0.28	0.28	0.28	0.25	0.26
303.15	0,31	0.28	0.29	0.28	0.26	0.27
308.15	0.32	0.29	0.30	0.29	0.27	0.28
313.15	0.33	0.30	0.31	0.30	0.28	0.29

Table 5. Continued

T / K	L-L	D-G	Eyk	A-B	New	Ost
Acetone (1) + 1,2-propandiol (2)						
318.15	0.34	0.31	0.32	0.31	0.28	0.30
323.15	0.35	0.32	0.33	0.32	0.29	0.31
Acetone (1) + 1,3-propandiol (2)						
288.15	0.37	0.34	0.35	0.34	0.31	0.33
293.15	0.38	0.35	0.36	0.35	0.32	0.33
298.15	0.39	0.36	0.37	0.36	0.33	0.34
303.15	0.40	0.36	0.37	0.36	0.33	0.35
308.15	0.40	0.37	0.38	0.37	0.34	0.35
313.15	0.41	0.38	0.39	0.38	0.34	0.36
318.15	0.42	0.39	0.40	0.39	0.35	0.37
323.15	0.43	0.40	0.41	0.40	0.36	0.38

cluded that all the studied mixing rules predict the experimental refarctive index data satisfactorily, with the values of maximum percentage deviation less than 0.35%. Temperature effects seem to be negligible for all analyzed systems and mixing rules.

CONCLUSION

This paper reports experimental data of viscosity η and refractive index n_D for acetone+1-propanol, acetone+1,2-propanediol and acetone+1,3-propanediol binary systems at eight temperatures (288.15, 293.15, 298.15, 303.15, 308.15, 313.15, 318.15 and 323.15 K) and at atmospheric pressure. From these data, viscosity deviations $\Delta\eta$ and deviations in refractive indices Δn_D were calculated and fitted to the Redlich-Kister polynomial equation.

The values of $\Delta\eta$ are negative over the entire temperature and composition range for all analyzed binary mixtures. Increase in temperature decrease viscosity deviation negative values for all investigated systems. Viscosity modeling was performed with Teja-Rice and McAlister correlative models. Also, UNIFAC-VISCO and ASOG-VISCO group contribution models were used to predict dynamic viscosities of investigated binary mixtures.

The values of Δn_D are positive for acetone+1,2--propanediol and acetone+1,3-propanediol binary mixtures over the entire temperature and composition range. For acetone +1-propanol binary mixture, the deviation in refractive index changes sign from positive to negative for higher acetone mole fractions. In addition, refractive indices were predicted by different mixing rules (Lorentz-Lorenz, Dale-Gladstone, Eykman, Arago-Biot, Newton, and Oster) and compared with experimental results.

Acknowledgements

The authors gratefully acknowledge the financial support received from the Research Fund of Ministry of Science and Environmental Protection, Serbia and the Faculty of Technology and Metallurgy, University of Belgrade (project No. 172063).

REFERENCES

[1] M.Lj. Kijevčanin, B.D. Djordjević, I.R. Radović, E.M. Živković, A.Ž. Tasić, S.P. Šerbanović, Modeling of Volumetric Properties of Organic Mixtures Based on Molecular Interactions, Molecular Interaction, Intech, 2012

[2] A. Lozano, B. Yip, R.K. Hanson, Exp. Fluids **13** (1992) 369-376

[3] J. Swarbrik, J.C. Boyland, Encyclopedia of Pharmaceutical Technology, Marcel Dekker, Inc., New York, 1993

[4] J.A. Riddick, W.B. Bunger, T.K. Sakano, Physical properties and methods of purification, Techniques of Chemistry, Organic solvents, Vol. II, John Wiley & Sons, New York, 1986

[5] M.Lj. Kijevčanin, M.M. Djuriš, I.R. Radović, B.D. Djordjević, S.P. Šerbanović, J. Chem. Eng. Data **52** (2007) 1136-1140

[6] S.P. Šerbanović, M.Lj. Kijevčanin, I.R. Radović, B.D. Djordjević, Fluid Phase Equilib. **239** (2006) 69-82

[7] I.R. Radović, M.Lj. Kijevčanin, S.P. Šerbanović, B.D. Djordjević, Fluid Phase Equilib. **298** (2010) 117-130

[8] C. Yang, P. Ma, Q. Zhou, J. Chem. Eng. Data **49** (2004) 582-587

[9] S.K. Mehta, G.Ram, C. Mani, K.K. Bhasin, J. Chem. Thermodyn. **38** (2006) 836-848

[10] J.-Y. Jung, H.S. Yun, J. Lee, M.-K. Oh, J. Microbiol. Biotechnol. **21** (2011) 846-853

[11] R. Gingell, J.B. Kirkpatrick, D.R. Steup, Int. J. Toxicol. **19** (2000) 27-32

[12] A. Behr, J.Elting, K. Irawadi, J.Leschinski, F.Lindner, Green Chem. **10** (2008) 13-30

[13] Z.-L. Xiu, A.-P. Zeng, A.-P. Appl. Microbiol. Biotechnol. **78** (2008) 917-926

[14] O. Redlich, A. Kister, Ind. Eng. Chem. **40** (1948) 345-348

[15] J.L. Chevalier, P. Petrino, Y. Gaston-Bonhomme, Chem. Eng. Sci. **43** (1988) 1303-1309

[16] Y. Gaston-Bonhomme, P. Petrino, J. L. Chevalier, Chem. Eng. Sci. **49** (1994) 1799-1806

[17] K. Tochigi, K. Yoshino, V.K. Rattan, Int. J. Thermophys. **26** (2005) 413-419

[18] A.S. Teja, P. Rice, Ind. Eng. Chem. Fundam. **20** (1981) 77-81

[19] A.S. Teja, P. Rice, Chem. Eng. Sci. **36** (1981) 7-10

[20] R.A. McAllister, AIChE J. **6** (1960) 427-431

[21] A. Pineiro, P. Brocos, A. Amigo, M. Pintos, R. Bravo, J. Chem.Thermodyn. **31** (1999) 931-942

[22] R.J. Fort, W.R. Moore, Trans. Faraday Soc. **62** (1966) 1112-1119

[23] Selected Values of Properties of Chemical Compounds, Data Project, Thermodynamics Research Center, Texas A&M University, College Station, TX, 1980

[24] J. George, N.V.Sastry, J. Chem. Eng. Data **48** (2003) 1529-1539

[25] A.Ž. Tasić, D.K. Grozdanić, B.D. Djordjević, S.P. Šerbanović, N. Radojković, J. Chem. Eng. Data **40** (1995) 586--588

[26] M. Iglesias, B. Orge, J. Tojo, J. Chem. Eng. Data **41** (1996) 218-221

[27] V.K. Misra, I. Vibhu, R. Singh, M. Gupta, J.P. Shukla, J. Mol. Liq. **135** (2007) 166-169

[28] S.L. Oswal, H.S. Desai, Fluid Phase Equilib. **149** (1998) 359-376

[29] M.A. Chowdhury, M.A. Majid, M.A. Saleh, J. Chem. Thermodynamics **33** (2001) 347-360

[30] J.M. Prausnitz, R.N. Lichtenthaler, E. Gomes de Azevedo, Molecular Thermodynamics of Fluid Phase Equilibria, Prentice Hall, New York, 1986

[31] Á. Piñeiro, P. Brocos, A. Amigo, M. Pintos, R. Bravo, Phys. Chem. Liq. **38** (2000) 251-260

[32] M.Lj. Kijevčanin, V.Z. Kostić, I.R. Radović, B.D. Djordjević, S.P. Šerbanović, Chem. Ind. Chem. Eng. Q. **14** (2008) 223-226

[33] E.M. Živković, M.Lj. Kijevčanin, I.R. Radović, S.P. Šerbanović, B.D. Djordjević, Fluid Phase Equilib. **299** (2010) 191-197

[34] D.W. Marquardt, J. Soc. Ind. Appl. Math. **11** (1963) 431--441.

ЕМИЛА М. ЖИВКОВИЋ
МИРЈАНА Љ. КИЈЕВЧАНИН
ИВОНА Р. РАДОВИЋ
СЛОБОДАН П. ШЕРБАНОВИЋ

Технолошко-металуршки факултет,
Универзитет у Београду,
Карнегијева 4, 11120 Београд,
Србија

НАУЧНИ РАД

ВИСКОЗНОСТИ И ИНДЕКСИ ПРЕЛАМАЊА БИНАРНИХ СИСТЕМА АЦЕТОН+1-ПРОПАНОЛ, АЦЕТОН+1,2-ПРОПАНДИОЛ И АЦЕТОН+ 1,3-ПРОПАНДИОЛ

Вискозности и индекси преламања три бинарна система ацетон+1-пропанол, ацетон +1,2-пропандиол и ацетон+1,3-пропандиол измерени су на осам температура (288,15, 293,15, 298,15, 303,15, 308,15, 313,15, 318,15 и 323,15 K) и на атмосферском притиску. Из измерених података израчунате су промене вискозности и промене индекса преламања које су корелисане једначином Redlich-Kister. Подаци за вискозност су моделовани помоћу два типа модела: предиктивних UNIFAC-VISCO и ASOG VISCO и корелативних Teja-Rice и McAlister. Индекси преламања добијени предвиђањем различитим правилима мешања поређени су са експерименталним подацима.

Кључне речи: вискозност, индекс преламања, експериментално мерење, моделовање.