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Vapor Pressures, Refractive Index at 20.0 °C, and Vapor–Liquid Equilibrium at 101.325 kPa in the Methyl *tert*-Butyl Ether–Methanol System

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The vapor pressures of methyl *tert*-butyl ether (MTBE) at roughly 3–55 °C and of methanol (MeOH) at roughly 26–65 °C were determined by comparative ebulliometry, the refractive index over the entire composition range of the MTBE–MeOH system was measured at 20.0 °C by dipping refractometry, and the isobaric vapor–liquid equilibrium in the system at 101.325 kPa was determined by dynamic method using a recirculation still. The estimated uncertainties in the measured temperature, pressure, refractive index, and compositions of the equilibrium phases are respectively ±0.005 °C, ±0.02%, ±0.000 03 refractive index units, and ±0.0005 to ±0.005 in mole fraction. The vapor–liquid equilibrium data were fitted to the Wilson equation. At 101.325 kPa, the MTBE–MeOH system exhibits a minimum-boiling azeotrope of composition 68.5 mol % (85.7 wt %) MTBE at 51.27 °C.

Introduction

MTBE, well-known for its excellent antiknock quality, has been extensively tested as an octane booster for lead-free or low-leaded gasoline, and the use of pure MTBE (*1*, *2*) and of its mixtures with C_4 alcohols (*3*) for high-octane gasoline blending was recommended. Recently the use of MTBE as a gasoline component at concentrations of 7% or less by volume was approved by the Environmental Protection Agency (*4*). Produced by catalytic reaction of methanol with isobutylene, MTBE appears as a welcome outlet for isobutylene formed in C_4 streams of ethylene production plants as well as for the conventional refinery C_4 streams. It is also increasingly valued as a means of making pure isobutylene.

Although most of the processes of the abruptly growing MTBE production involve distillation at atmospheric pressure as a final stage, a search of the available literature failed to reveal reliable data on vapor–liquid equilibrium in the MTBE–MeOH system. For this system we report here the isobaric vapor–liquid equilibrium data at 101.325 kPa along with refractive indices at 20.0 °C useful for analysis. Vapor pressures of pure components were determined ebulliometrically over the approximate pressure range 15–100 kPa for comparison with the most accurate recent results for methanol obtained by Gibbard and Creek (*5*) using a static technique and with the data of Ambrose et al. (*6*) for MTBE.

Experimental Section

Materials. MTBE from two sources was used for the study. The first batch was the product of Chemische-Werke Hüls, FRG, with an indicated purity of at least 98%, and the second batch was synthesized in our laboratory from methanol and *tert*-butyl alcohol (both A.R. grade) in diluted sulfuric acid at ca. 52 °C by a procedure analogous to that described in literature (*7*, *8*) for the preparation of ethyl *tert*-butyl ether. In both cases, the MTBE product was repeatedly washed with water, shaken with the saturated potassium permanganate aqueous solution containing ca. 120 g/L potassium hydroxide (until the aqueous layer remained green), washed with water again, and twice rectified on a 60-plate bubble-cup column at 20:1 reflux ratio. The 40 and 60% heart-cut was taken at the first and second distillation stage, respectively. The MTBE was then dried by staying in contact with freshly cut metal sodium for several days and finally being distilled from sodium on a column (30 mm i.d., 65 cm length) packed with Pyrex-glass helices (3 mm diameter) at a reflux ratio of 7:1. The middle 50% fraction was used for the experiments.

A.R. grade methanol (Lachema, Brno, Czechoslovakia) was twice rectified in a 60-plate bubble-cup column at 20:1 reflux ratio, the middle 50% distillate portion being retained in both stages. It was then dried by the procedure of Lund and Bjerrum (*9*) which, briefly, involved a separate conversion of a portion (corresponding stoichiometrically to 5 g of magnesium/L) of the methanol batch to magnesium methoxide and refluxing the methanol with dissolved methoxide for 3 h. With the exclusion of moisture from the system, methanol was then given the final distillation in the packed column under the same conditions as described for MTBE.

The water used for pycnometric calibrations, refractometric adjustments, and indirect pressure determinations by means of the water boiling point was a sample redistilled on addition of potassium permanganate in a quartz apparatus and deareated by boiling off one-third of its volume.

No impurities in the MTBE and methanol employed for experiments were detected by gas chromatographic analysis. Karl

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Table I. Density at 20.0 °C, Refractive Index at 20.0 °C, and Boiling Point at 101.325 kPa for MTBE and Methanol

solvent	density, g/cm^3	n_D	bp, °C	ref
MTBE	0.74054	1.36892	55.061	this work
			55.143	6
			55.0	19
	0.7405	1.3689	55.2	13, 14, 15
	0.7406	1.3690	55.1	16, 17
	0.7406	1.3690	55.2	18
	0.7418			20
methanol	0.79120	1.32846	64.515	this work
			64.514	5
			64.546	23
			64.50	21, 22
	0.79105			24
	0.79128	1.3286	64.52	25
	0.79122	1.32857	64.51	18
	0.79129	1.32840	64.70	26
		1.3287	64.7	17
	0.79144	1.3288	64.96	15
	0.7913-0.7916	1.3287	64.501-64.75	20

Fischer titration (*10*) revealed that in both organic reagents the water content was less than 0.01 mol %. Physical constants of the pure substances obtained are summarized in Table I.

Apparatus and Procedure. Modified 25-mL two-arm Sprengel-Ostwald pycnometers with mercury-calibrated capillaries were used for the determination of liquid densities at 20.0 ± 0.004 °C. Appropriate buoyancy corrections were applied to all experimental weights. Duplicate measurements with two pycnometers confirmed that the uncertainty in the densities obtained was less than ±0.02 kg/m^3.

Refractive indices were measured by means of a dipping refractometer (VEB Carl Zeiss, Jena) using the L1 and T2 thermoprisms, whose temperature was maintained at 20.0 °C, better than to ±0.01 °C, by means of an ethanol circulating bath unit. The refractometer yielded replicate refractive index results which had a standard deviation of less than 0.000 02 refractive index units. For absolute measurements the instrument was adjusted according to the reading for the water sample. The accuracy of the refractive index determinations was believed to be within ±0.000 03 refractive index units. For measurements on mixtures, the solutions of known composition in the amount of ca. 10 mL were prepared by weighing on an analytical balance so that the uncertainty in composition was negligible as compared to that in refractive index.

Saturated vapor pressures of the pure solvents were measured by means of the standard Swietoslawski ebulliometer connected, in parallel with the second ebulliometer filled with water, to a buffer reservoir of the pressure-controlling assembly. The boiling points of the organic substance and water, respectively, were measured simultaneously by using a quartz thermometer, Model HP-2801A (Hewlett-Packard), with two 2850D probes. The thermometer was calibrated against a Leeds & Northrup standard platinum-resistance thermometer 8163-B provided with the National Bureau of Standards certificate and connected to a Mueller bridge 8069-B. We believe that the measured temperatures were within ±0.005 °C of the true temperatures on the International Practical Temperature Scale of 1968. From the boiling point of water, the corresponding pressure in the system (read from the water vapor pressure tables of Bridgeman and Aldrich (*11*)) was determined with an accuracy of approximately ±0.02% of the measured value. The run of vapor pressure measurements was conducted in order from the highest pressure downward. At the start of each run, a check was made for the absence of any significant elevation of boiling temperature on boiling off ca. 20% of the volume of the investigated substance into a cold trap.

Table II. Vapor Pressures of MTBE and Methanol

MTBE			methanol		
t, °C	P, kPa	$10^3 \times (P_{calcd} - P)$, kPa	t, °C	P, kPa	$10^3 \times (P_{calcd} - P)$, kPa
3.898^a	12.952	10	26.028	17.900	−1
4.022^a	13.027	15	29.431	21.286	0
6.217	14.530	−19	33.261	25.736	0
7.723^a	15.577	18	36.316	29.830	2
10.652	17.912	−18	39.963	35.429	3
11.838^a	18.876	25	43.371	41.441	5
14.426	21.271	−12	43.392	41.494	−8
14.456	21.302	−15	47.402	49.644	6
18.749	25.740	−13	47.425	49.708	−7
22.184	29.806	−10	52.229	61.245	−11
26.373	35.453	−5	52.230	61.228	8
26.400	35.491	−3	57.705	77.032	8
30.258	41.429	3	57.737	77.151	−10
30.299	41.496	4	57.857	77.512	11
34.919	49.639	16	63.855	98.729	−5
34.949	49.697	15	64.497	101.265	−10
40.536	61.207	35	64.528	101.367	11
40.570	61.326	−8			
46.998	77.160	−33			
54.288	98.773	2			
54.413	99.181	3			
55.048	101.289	−8			

^{a} Points measured by the static method.

A modified Gillespie recirculation equilibrium still as described by Dvořák and Boublík (*12*) was used for vapor–liquid equilibrium measurements in the binary system. The total amount of liquid in the apparatus was ca. 160 mL, the respective volumes of the liquid-phase and vapor-phase receivers being ca. 40 and ca. 30 mL. The still was connected to the assembly maintaining pressure at 101.325 kPa. The equilibrium temperature and pressure were determined as described for the vapor pressure measurements on pure components. Indicated by constancy of temperature at a given pressure, steady state in the apparatus was usually attained in ca. 20 min on adjusting the composition of the binary system. Samples of equilibrium phases were then taken for refractometric analysis after another ca. 15 min of still operation in the steady-state mode. The composition as a function of refractive index was read from the interpolation tables computed from eq 2 (see below). Estimated uncertainty in the determined mole fraction varied from ±0.0005 in the methanol-rich region to ±0.005 in the MTBE-rich region due to the shape of the refractive index concentration dependence for the binary. A set of data over the entire composition range was a result of two experimental runs starting, respectively, from pure components and overlapping in the middle region.

Results and Discussion

The obtained average densities and refractive indices at 20.0 °C and boiling points at 101.325 kPa (calculated from eq 1) for MTBE and methanol are listed and compared with previously published values in Table I. It is noted that the data for both of the MTBE batches were coincident within the limits of experimental uncertainty.

Our results on vapor pressure measurements for MTBE and methanol are shown in Table II. The data set for MTBE was gathered from three experimental runs, namely, two ebulliometric measurements on the two above-mentioned MTBE batches, respectively, and four points as indicated in Table II obtained by static technique, a procedure described in detail in our earlier paper (*27*). The data set for methanol is a result of two ebulliometric runs. In Table II the first column is the temperature and the second is the measured vapor pressure. The third column gives the difference between the vapor pressure calculated from the Antoine equation of the form

$$\log P/\text{kPa} = A - B/(C + t/{}^\circ\text{C}) \qquad (1)$$

Table III. Coefficients of Equation 1 and Standard Deviation

solvent	A	B	C	$10^3 S$/kPa
MTBE	6.01607	1126.594	225.860	17
methanol	7.19367	1575.503	239.170	8

Table IV. Refractive Index of the MTBE (1)-Methanol (2) Solutions at 20.0 °C

x_1	n_D	$10^5 \times (n_{D(calcd)} - n_D)$
0.0364	1.333 13	2
0.0376	1.333 34	-5
0.0395	1.333 52	0
0.0585	1.335 67	2
0.0775	1.337 73	-1
0.0938	1.339 33	2
0.0964	1.339 60	-1
0.1368	1.343 18	2
0.1719	1.345 87	6
0.1906	1.347 24	2
0.2152	1.348 79	9
0.2278	1.349 60	6
0.2610	1.351 59	-1
0.2719	1.352 23	-6
0.3258	1.355 05	-23
0.3443	1.355 69	-5
0.4290	1.358 95	-2
0.4572	1.359 91	-2
0.5481	1.362 55	-1
0.5645	1.362 89	6
0.6940	1.365 57	8
0.7137	1.365 91	6
0.7664	1.366 73	-2
0.8161	1.367 34	-5
0.8855	1.367 94	0
0.9246	1.368 26	0

Table V. Vapor-Liquid Equilibrium Data for the System MTBE (1)-Methanol (2) at 101.325 kPa

exptl			calcd	
t, °C	x_1	y_1	$10^2 \times (y_{1(calcd)} - y_1)$	$t_{calcd} - t$, °C
63.762	0.0095	0.0368	0.07	-0.011
62.886	0.0206	0.0776	-0.03	0.037
62.361	0.0285	0.1023	0.10	0.011
61.845	0.0360	0.1257	0.07	0.032
60.964	0.0495	0.1654	-0.08	0.082
60.970	0.0505	0.1665	0.07	0.018
60.418	0.0595	0.1900	0.02	0.060
59.559	0.0763	0.2283	0.09	0.045
59.091	0.0851	0.2488	-0.11	0.092
58.406	0.1016	0.2792	0.03	0.047
57.596	0.1214	0.3144	-0.11	0.074
57.200	0.1331	0.3320	-0.06	0.049
55.970	0.1743	0.3845	0.14	0.008
55.336	0.2006	0.4133	0.15	-0.028
54.685	0.2313	0.4408	0.34	-0.047
54.369	0.2470	0.4530	0.47	-0.033
53.703	0.2896	0.4860	0.43	-0.074
53.102	0.3388	0.5153	0.69	-0.122
52.770	0.3678	0.5320	0.69	-0.106
52.248	0.4280	0.5620	0.80	-0.112
51.990	0.4610	0.5790	0.66	-0.084
51.460	0.5651	0.6270	0.46	-0.060
51.232	0.6781	0.6804	0.23	-0.048
51.280	0.7140	0.6955	0.54	-0.085
51.412	0.7970	0.7432	0.69	0.014
51.618	0.8330	0.7650	1.14	0.033
52.306	0.9248	0.8485	2.02	0.516
53.517	0.9790	0.9310	2.39	0.730

and the experimental value. The coefficients A, B, and C of eq 1 were evaluated from our data by the weighted least-squares procedure accepted by Boublík et al. (*28*) and are listed together with the corresponding standard deviation of the fit in Table III. It can be seen that the Antoine equation represents the data within standard deviations favorably comparable to the precision of our measurements.

The only reliable source of data to be compared with our vapor pressure results for MTBE appears to be the study of Ambrose et al. (*6*). To make the comparison more adequate, we excerpted from their data set the vapor pressures covering the range examined here and fitted them to eq 1 by using the identical procedure, thus receiving A = 6.145 31, B = 1200.465, and C = 234.853 at a standard deviation of 0.036 kPa. With these coefficients eq 1 yields a somewhat higher normal boiling point than with those from our data (see Table I). The deviation decreases with decreasing temperature and at about 27 °C the equations with the two triads of coefficients intersect. Ambrose et al. reported mole fraction of impurity present in their MTBE to be 0.001. We believe that the purity of our sample was higher, this being supported also by substantially lower scatter of our vapor pressure points.

The vapor pressure of methanol has been extensively studied (*5*, *23*). Most reliable and accurate seem to be the recently published data of Gibbard and Creek (*5*), obtained by means of a static vapor pressure apparatus. With these data as represented by eq 1 of their paper, our ebulliometric results were found to be in excellent agreement over the entire temperature range investigated.

The refractive indices at 20.0 °C as a function of composition of the MTBE-MeOH solutions are listed in Table IV. The first column is the MTBE mole fraction, the second is the observed refractive index, and the third presents the difference between the refractive index calculated from the equation

$$n_D = x_1 n_{D1} + x_2 n_{D2} + x_1 x_2 \sum_{k=1}^{4} A_k x_1^{k-1} \qquad (2)$$

and the experimental value. In eq 2, x_1 and x_2 are mole fractions and n_{D1} and n_{D2} are refractive indices of pure MTBE and methanol, respectively, and n_D is the refractive index of the mixture. The empirical coefficients A_k, evaluated from our measurements by the least-squares method, are A_1 = 0.097 60, A_2 = -0.168 25, A_3 = 0.191 71, and A_4 = -0.090 60, representing the experimental data with a mean absolute deviation of 0.000 04 refractive index units. Notably, the four constants were necessary to arrive at a good fit of refractive index data in this system.

The vapor-liquid equilibrium data for the system MTBE-MeOH at 101.325 kPa are presented in Table V. The data set contains two subsets obtained over the entire composition range for the two batches of MTBE, respectively. For each point the equilibrium total pressure, temperature, liquid composition, and vapor composition were determined; the measured values of the latter three variables are given in the first three columns of Table V, the compositions being expressed in MTBE mole fraction.

By minimization of the sum of square deviations in total pressure by having each point being assigned the same weight, the data were smoothed by using the Wilson equation (selected as yielding the best fit) to represent the composition dependence of liquid-phase activity coefficients:

$$\ln \gamma_1 = -\ln (x_1 + \Lambda_{12} x_2) + x_2 \left[\frac{\Lambda_{12}}{x_1 + \Lambda_{12} x_2} - \frac{\Lambda_{21}}{\Lambda_{21} x_1 + x_2} \right]$$

$$\ln \gamma_2 = -\ln (x_2 + \Lambda_{21} x_1) - x_1 \left[\frac{\Lambda_{12}}{x_1 + \Lambda_{12} x_2} - \frac{\Lambda_{21}}{\Lambda_{21} x_1 + x_2} \right] \qquad (3)$$

Table VI. Second Virial Coefficients in the MTBE (1)-Methanol (2) System, Estimated by the Method of Hayden and O'Connell

t, °C	B_{11}, mL/mol	B_{22}, mL/mol	B_{12}, mL/mol
50	−1236	−1598	−1291
55	−1187	−1446	−1210
60	−1141	−1313	−1137
65	−1098	−1197	−1070

The equation of vapor–liquid equilibrium for a binary system was written in the form

$$y_iP = x_i\gamma_iP_i^{\rm s}\exp\{[(B_{ii} - V_i)(P_i^{\rm s} - P) - P(2\sum_{j=1}^{2} y_jB_{ij} - B_{ii} - B)]/RT\} \quad (4)$$

derived for the standard state of pure component at temperature T and pressure P of the system, assuming that the pure component molar volumes V_i are pressure independent and that the vapor phase obeys the volume explicit virial expansion truncated after the second term. In eq 4, B_{ii} and $P_i^{\rm s}$ are the second virial coefficient and saturated vapor pressure of pure ith component, respectively, B_{ij} is the second virial cross coefficient, and R is the universal gas constant. The virial coefficients requisite to the calculations, estimated by the method of Hayden and O'Connell (*29*) for four temperature levels covering the studied range, are listed in Table VI. The second virial coefficients for methanol are in excellent agreement with the data given by Dymond and Smith (*30*). No literature values could be located for the second virial coefficients of MTBE and for the second virial cross coefficients.

The resulting parameters of the Wilson equation for the MTBE–MeOH system, regarded over the measured region as temperature independent, are $\Lambda_{12} = 0.5524$ and $\Lambda_{21} = 0.4557$. Calculated from eq 3 and 4 by using these constants, the differences between the smoothed and observed values of the MTBE mole fraction in vapor phase and of the equilibrium temperature, respectively, are given in the fourth and fifth columns of Table V. The fit characterized by the mean absolute deviation between calculated and experimental values, for the vapor-phase mole fraction equal to 0.0045 and for the temperature equal to 0.095 °C, is slightly inferior to the estimated precision of experiments.

The MTBE–MeOH system exhibits large positive deviations from ideality with a minimum-boiling azeotrope. At 101.325 kPa, the coordinates of the azeotropic point evaluated from our measurements are 68.5 mol % (85.7 wt %) MTBE and 51.27 °C. This is in fair agreement with the azeotropic data reported by Evans and Edlund (*13*), viz., 85 wt % MTBE and 51.6 °C, the only dependable information on vapor–liquid equilibrium in the MTBE–MeOH system discovered in literature.

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Glossary

A_k	coefficients in eq 2
A, B, C	coefficients in eq 1
B	second virial coefficient of a mixture (in eq 4), mL/mol
B_{ii}	second virial coefficient of pure component i, mL/mol
B_{ij}	second virial cross coefficient, mL/mol
$n_{\rm D}$	refractive index of a solution
$n_{{\rm D}i}$	refractive index of pure liquid i
P	(total) pressure, kPa
$P_i^{\rm s}$	saturated vapor pressure of pure component i, kPa
R	gas constant, mL kPa/(mol K)
S	standard deviation = $[\sum^{\text{no. of points}}(\text{calcd value} - \text{exptl value})^2/(\text{no. of points} - \text{no. of coeffs})]^{1/2}$
t	temperature, °C
T	absolute temperature, K
V_i	volume of pure liquid i, mL/mol
x_i	mole fraction of component i in the liquid phase
y_i	mole fraction of component i in the vapor phase
γ_i	liquid-phase activity coefficient of component i
Λ_{ij}	Wilson parameter

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