Isobaric vapour–liquid equilibria of the ternary system toluene + *p*-xylene + 1,2-dichloroethane

Helle Kirss, Mati Kuus, Enn Siimer[*], and Ludmilla Kudryavtseva

Department of Material Science, Tallinn Technical University, Akadeemia tee 15, 12618 Tallinn, Estonia

Abstract. Isobaric vapour–liquid equilibrium (VLE) was measured for the system toluene + *p*-xylene + 1,2-dichloroethane and for constituent binaries at pressures 26.66, 53.33, 79.99, and 101.32 kPa. A modified Wilson equation with a linear temperature dependence of the main parameters $(\lambda_{ij} - \lambda_{ii})$ was used to correlate the binary VLE data and to predict those of the ternary system. Excess functions of the ternary system at 298.15 K were compared.

Key words: vapour–liquid equilibrium, ternary system, toluene, *p*-xylene, 1,2-dichloroethane.

INTRODUCTION

Knowledge of multicomponent vapour–liquid equilibrium (VLE) data is important in the design of equipment for separation processes. In addition, such experimental information can be used to test and develop prediction correlations. The purpose of this study was to determine VLE for the ternary system toluene + *p*-xylene + 1,2-dichloroethane and for three constituent binaries at four constant pressures, 26.66, 53.33, 79.99, and 101.32 kPa. A Wilson equation [1] whose main parameters $(\lambda_{ij} - \lambda_{ii})$ were assumed to be a linear function of temperature was used to obtain a fit of binary VLE data. As a continuation of our previous investigation [2], the possibility of predicting the behaviour of the ternary system from three binaries was checked. It appears that the ternary system toluene + *p*-xylene + 1,2-dichloroethane has not been studied earlier.

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[*] Corresponding author, siimer@chemnet.ee

EXPERIMENTAL

All the substances used in this work were the same as in our earlier work [3]. Toluene and *p*-xylene ("purum" grade) were twice distilled. 1,2-Dichloroethane ("puriss" grade) was used without further purification. Densities and refractive indices of pure liquids at 298.15 K agree well with the literature values, as seen from Table 1.

The boiling temperature (*T*)–liquid phase composition (*x*) equilibrium was measured in a semi-micro ebulliometer [6]. Uncertainties of the boiling temperatures were estimated to be less than 0.05 K, and those of the mole fractions composition of the liquid mixture less than 5×10^{-4}.

The *T*–*x* results for the binary systems toluene + *p*-xylene, toluene + 1,2-dichloroethane, and *p*-xylene + 1,2-dichloroethane obtained in this work are listed in Table 2.

The *T*–*x* data in binary systems were fitted with the Wilson model [1] in the form

$$\ln \gamma_i = -\ln(x_i - \Lambda_{ik} x_k) + x_k \left[\frac{\Lambda_{ik}}{x_i + \Lambda_{ik} x_k} - \frac{\Lambda_{ki}}{x_k + \Lambda_{ki} x_i} \right], \qquad (1)$$

where γ_i is the activity coefficient of component i in the liquid phase and Λ_{ik} and Λ_{ki} are expressed as

$$\Lambda_{ik} = \exp\left[a_{ik} + \frac{b_{ik}}{T} \right], \qquad (2)$$

$$\Lambda_{ki} = \exp\left[a_{ki} + \frac{b_{ki}}{T} \right], \qquad (3)$$

including in the parameters a_{ik} and b_{ik} molar volumes of components and the gas constant [2].

The vapour pressures of pure components P_i^0 needed for VLE calculation were calculated by the Antoine equation

$$\ln(P_i/\text{kPa}) = A_i - \frac{B_i}{T/\text{K} + C_i}. \qquad (4)$$

Table 1. Densities (d) and refractive indices (n_D) of pure components at 298.15 K

Component	d, kg m^{-3}		n_D	
	Exp.	Lit.	Exp.	Lit.
Toluene	862.2	862.19[a]	1.4940	1.49413[a]
p-Xylene	856.7	856.6[b]	1.4930	1.4931[b]
1,2-Dichloroethane	1246.3	1246.37[a]	1.4419	1.4421[a]

[a] – from [4]
[b] – from [5]

Table 2. Isobaric vapour–liquid equilibria data: liquid phase mole fraction (x_1) and boiling temperature (T) in binary systems

P = 26.66 kPa		P = 53.33 kPa		P = 79.99 kPa		P = 101.32 kPa	
x_1	T, K	x_1	T, K	x_1	T, K	x_1	T, K
Toluene (1) + p-xylene (2)							
0.000	367.78	0.000	389.00	0.000	402.85	0.000	411.49
0.195	361.80	0.195	382.69	0.195	396.49	0.195	404.97
0.227	360.66	0.227	381.63	0.227	395.40	0.227	404.09
0.282	358.86	0.282	379.89	0.282	393.57	0.282	402.06
0.390	355.95	0.390	376.85	0.390	390.46	0.390	398.91
0.396	355.41	0.396	376.24	0.396	389.90	0.396	398.32
0.499	353.01	0.499	373.67	0.499	387.27	0.499	395.64
0.589	350.84	0.589	371.46	0.589	384.90	0.589	393.26
0.680	348.90	0.680	369.33	0.680	382.68	0.680	390.97
0.785	346.66	0.785	367.06	0.785	380.27	0.785	388.53
0.867	345.36	0.867	365.47	0.867	378.61	0.867	386.77
0.903	344.29	0.903	364.35	0.903	377.56	0.903	385.73
1.000	342.65	1.000	362.63	1.000	375.66	1.000	383.76
Toluene (1) + 1,2-dichloroethane (2)							
0.000	319.22	0.000	337.45	0.000	349.29	0.000	356.63
0.110	320.79	0.108	339.03	0.106	350.96	0.104	358.38
0.205	322.45	0.206	340.95	0.207	352.98	0.208	360.49
0.284	323.67	0.279	342.06	0.274	354.16	0.269	361.65
0.391	325.86	0.392	344.74	0.393	357.03	0.394	364.58
0.483	327.54	0.481	346.42	0.478	358.79	0.475	366.49
0.596	330.81	0.602	350.14	0.608	362.63	0.615	370.42
0.695	333.22	0.697	352.65	0.699	365.29	0.701	373.17
0.781	336.01	0.787	355.55	0.793	368.31	0.799	376.27
1.000	342.65	1.000	362.63	1.000	375.66	1.000	383.76
p-Xylene (1) + 1,2-dichloroethane (2)							
0.000	319.22	0.000	337.45	0.000	349.29	0.000	356.63
0.100	321.40	0.098	339.62	0.095	351.54	0.093	358.95
0.199	323.46	0.194	342.11	0.189	354.23	0.184	361.79
0.292	325.89	0.291	344.86	0.290	357.32	0.289	365.11
0.326	327.29	0.322	346.15	0.318	358.61	0.314	366.32
0.377	329.72	0.382	349.03	0.387	361.63	0.392	369.48
0.464	332.78	0.468	352.33	0.472	365.22	0.476	373.17
0.555	337.01	0.558	356.49	0.561	369.68	0.564	377.73
0.677	343.76	0.612	359.44	0.615	372.69	0.618	380.75
0.767	349.02	0.676	363.52	0.675	376.75	0.674	384.98
1.000	367.78	0.718	364.54	0.702	377.19	0.686	384.06
		0.767	368.98	0.767	382.79	0.767	391.10
		1.000	389.00	1.000	402.85	1.000	411.49

The values of the constants A_i, B_i, and C_i are given in Table 3.

The values of the coefficients a_{ik} and b_{ik} of Eqs. 2 and 3, calculated by the Newton iteration method, together with the mean absolute relative error of pressure (ΔP) are listed in Table 4.

Table 3. Constants for the Antoine vapour pressure equation (Eq. 4)

Component	Temperature region, K	Constants		
		A_i	B_i	C_i
Toluene	343–384	14.08406	3148.177	−51.172
p-Xylene	368–411	13.82595	3178.599	−66.279
1,2-Dichloroethane	319–357	14.5197	3117.876	−41.736

Table 4. Fitted coefficients of the modified Wilson equation (Eqs. 2, 3) and calculated absolute mean error (ΔP) of pressure for binary systems

System (1)+(2)	a_{12}	b_{12}	a_{21}	b_{21}	ΔP, %
Toluene + *p*-xylene	−0.35434	118.040	0.43197	−124.937	0.32
Toluene + 1,2-dichloroethane	0.04946	−15.449	0.17245	−67.618	0.63
p-Xylene + 1,2-dichloroethane	−0.10223	−44.134	0.71791	−168.002	1.35

VLE data for the system toluene + *p*-xylene were reported at a temperature of 363.15 K by Wichterle [7], at normal pressure by Schmelzer & Wolf [8] and by Sartakova et al. [9], and at 101.59 kPa by Kutsarov et al. [10]. It can be seen from Fig. 1 that the data presented here are in the best agreement with those obtained by Schmelzer & Wolf [8].

Experimental data for the system toluene + 1,2-dichloroethane have been obtained by several investigators, most of them are published also in the data collection [11, p. 109]. At normal pressure VLE data have been obtained by Jones et al. [12], Alpert & Elving [13], Rollet et al. [14], and at barostatic pressure by Sharma & Singh [15]. Ellis reported VLE data at 26.66 kPa [16], and Rivenq at 26.66, 39.99, 53.33, 79.99, and 101.32 kPa (cited in [11, p. 109]). Deviations from our results are the smallest for those of Rollet et al. [14] and the greatest for those of Jones et al. [12]. Figure 2 compares the above-mentioned experimental results with the data of this work. Rivenq's data (measured at the same pressures used in our work) deviate from our results just in the temperature determination, especially at higher 1,2-dichloroethane concentrations. This may be caused by deviations in 1,2-dichloroethane boiling temperatures (see Table 5 for values measured by different authors). Table 5 presents also the boiling point of toluene.

The boiling temperatures of 1,2-dichloroethane at four pressures measured in this work agree well with those obtained by Gutsche & Knapp [17] and Dohnal et al. [18]. The sets of Antoine constants from these publications represent boiling temperatures which differ from those of our work with an overall average absolute deviation of 0.02 K.

VLE data for *p*-xylene + 1,2-dichloroethane have been given in several publications [11, 15, 19, 20] at conditions which differ from those of this study. Our *P* vs. *T* data for *p*-xylene measured at four pressures agree well with those of Gupta & Rawat [21] with an overall average absolute deviation of 0.02 K.
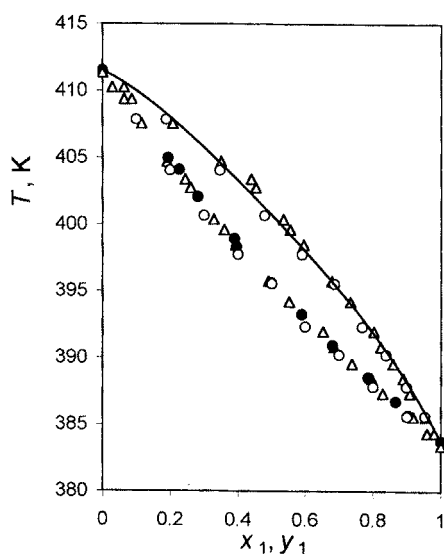
Fig. 1. $T–x–y$ diagram for toluene (1) + p-xylene (2) at 101.32 kPa. ● experiment of this work; — calculated by the modified Wilson equation on the basis of data from this work; △ experiment of Schmelzer & Wolf [8]; ○ experiment of Sartakova et al. [9].
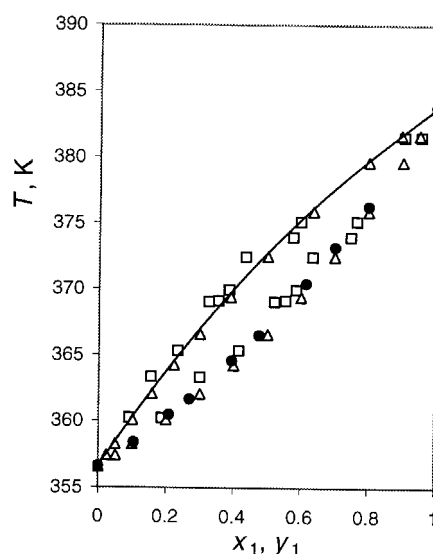
Fig. 2. $T–x–y$ diagram for toluene (1) + 1,2-dichloroethane (2) at 101.32 kPa. ● experiment of this work; — calculated by the modified Wilson equation on the basis of data from this work; △ experiment of Rollet et al. [14]; □ experiment of Jones et al. [12].

Table 5. Boiling temperature (T) of toluene and 1,2-dichloroethane at different pressures

Pressure, kPa	T, K	
	Toluene	1,2-Dichloroethane
101.32	383.76[a], 383.75[b], 383.76[c]	356.63[a], 355.75[b], 356.60[c]
79.99	375.66[a], 375.65[b]	349.29[a], 348.55[b]
53.33	362.63[a], 362.65[b]	337.45[a], 337.15[b]
26.66	342.65[a], 342.65[b], 342.65[d]	319.22[a], 318.85[b], 318.85[d]

[a] – this work
[b] – Rivenq (cited in [11, p. 109])
[c] – Rollet et al. [13]
[d] – Ellis [15]

All present binary systems are nonazeotropic and weakly nonideal. As shown by us earlier [3], these systems are close to ideal also at 298.15 K, because excess molar enthalpies are only slightly negative.

The isobaric VLE data for the ternary system studied are given in Table 6.

The prediction results obtained by the modified Wilson equation are presented in Table 7. By assuming that the ideal gas law holds, calculations of VLE were made using the values of coefficients for three constituent binaries given in Table 4 and constants of the Antoine equation from Table 3. The mean absolute error for boiling temperatures at all four pressures was 0.90 K.

Table 6. Isobaric vapour–liquid equilibrium data, liquid phase mole fraction (x_i), and boiling temperature (T) in the ternary system toluene $(1) + p$-xylene $(2) + 1,2$-dichloroethane (3) at different pressures (P, kPa)

x_1	x_2	x_3	T, K			
			$P = 26.66$	$P = 53.33$	$P = 79.99$	$P = 101.32$
0.289	0.289	0.422	333.92	353.18	366.05	373.94
0.413	0.413	0.174	344.30	364.26	377.35	385.65
0.177	0.177	0.646	326.94	345.88	–	–
0.553	0.277	0.170	343.22	362.95	375.90	384.05
0.432	0.216	0.352	335.65	354.72	367.31	375.20
0.286	0.143	0.571	328.07	346.62	358.68	366.15
0.222	0.444	0.334	338.80	358.01	370.73	377.72
0.163	0.326	0.511	331.99	351.39	364.09	371.94
0.272	0.544	0.184	347.58	–	–	–
0.280	0.560	0.160	349.58	369.25	381.84	390.20
0.332	0.336	0.332	336.95	355.74	367.82	376.39
0.133	0.734	0.133	353.36	373.60	386.65	394.83

– not determined

Table 7. Isobaric vapour–liquid equilibrium data for the ternary system toluene $(1) + p$-xylene (2) + 1,2-dichloroethane (3), predicted by the modified Wilson equation [2]

Experimental data			Predicted data						
x_1	x_2	T, K	ΔT	y_1	y_2	γ_1	γ_2	γ_3	P^*
$P = 26.66$ kPa									
0.289	0.289	333.92	−0.76	0.200	0.073	0.9946	0.9811	1.0076	205.8
0.413	0.413	344.30	−1.16	0.416	0.160	0.9892	0.9813	1.0022	208.6
0.177	0.177	326.94	−0.25	0.095	0.033	1.0041	0.9896	1.0049	201.9
0.553	0.277	343.22	−2.28	0.517	0.097	0.9958	0.9703	1.0075	217.4
0.432	0.216	335.65	−1.46	0.313	0.056	0.9983	0.9706	1.0097	211.3
0.286	0.143	328.07	−0.12	0.162	0.028	1.0047	0.9792	1.0068	201.0
0.222	0.444	338.80	−0.92	0.183	0.138	0.9844	0.9881	1.0028	206.9
0.163	0.326	331.99	−0.71	0.104	0.076	0.9914	0.9895	1.0051	205.5
0.272	0.544	347.58	−2.40	0.293	0.232	0.9800	0.9895	0.9937	218.0
0.280	0.560	349.58	−3.03	0.316	0.253	0.9793	0.9899	0.9911	222.9
0.332	0.336	336.95	−0.57	0.260	0.097	0.9921	0.9806	1.0072	204.3
0.133	0.734	353.36	−1.76	0.177	0.405	0.9641	0.9967	0.9778	212.9
$P = 53.33$ kPa									
0.289	0.289	353.18	−0.27	0.208	0.084	0.9945	0.9716	0.9986	403.6
0.413	0.413	364.26	−0.64	0.422	0.175	0.9898	0.9783	0.9851	408.3
0.177	0.177	345.88	−0.10	0.099	0.038	1.0008	0.9705	1.0013	401.4
0.553	0.277	362.95	−1.78	0.526	0.107	0.9963	0.9663	0.9928	423.6
0.432	0.216	354.72	−0.77	0.323	0.063	0.9983	0.9619	1.0005	410.3
0.286	0.143	346.62	0.51	0.169	0.032	1.0021	0.9623	1.0027	393.1
0.222	0.444	358.01	0.10	0.188	0.154	0.9851	0.9828	0.9898	398.8
0.163	0.326	351.39	−0.49	0.109	0.086	0.9909	0.9781	0.9974	406.6
0.280	0.560	369.25	−1.93	0.319	0.274	0.9797	0.9880	0.9721	425.0
0.332	0.336	355.74	0.66	0.268	0.108	0.9926	0.9741	0.9959	391.4
0.133	0.734	373.60	−0.88	0.176	0.434	0.9637	0.9958	0.9556	411.1

Table 7. *Continued*

Experimental data			Predicted data						
x_1	x_2	T, K	ΔT	y_1	y_2	γ_1	γ_2	γ_3	P^*
				$P = 79.99$ kPa					
0.289	0.289	366.05	−0.30	0.212	0.087	0.9945	0.9657	0.9932	605.5
0.413	0.413	377.35	−0.41	0.425	0.184	0.9902	0.9764	0.9750	607.2
0.553	0.277	375.90	−1.57	0.530	0.112	0.9966	0.9639	0.9840	628.6
0.432	0.216	367.31	−0.50	0.329	0.067	0.9983	0.9566	0.9949	609.3
0.286	0.143	358.68	0.92	0.172	0.034	1.0006	0.9523	1.0002	582.9
0.222	0.444	370.73	0.54	0.192	0.163	0.9855	0.9795	0.9819	590.4
0.163	0.326	364.09	−0.43	0.111	0.091	0.9905	0.9713	0.9927	608.1
0.280	0.560	381.84	−1.00	0.320	0.286	0.9798	0.9869	0.9611	617.7
0.332	0.336	367.82	1.60	0.272	0.114	0.9929	0.9703	0.9892	571.5
0.133	0.734	386.65	−0.18	0.175	0.451	0.9633	0.9953	0.9427	603.2
				$P = 101.32$ kPa					
0.289	0.289	373.94	−0.20	0.214	0.090	0.9945	0.9624	0.9900	764.4
0.413	0.413	385.65	−0.41	0.427	0.189	0.9904	0.9753	0.9690	768.8
0.553	0.277	384.05	−1.52	0.532	0.115	0.9968	0.9624	0.9788	793.5
0.432	0.216	375.20	−0.40	0.332	0.069	0.9983	0.9535	0.9917	768.8
0.286	0.143	366.15	1.20	0.175	0.035	0.9998	0.9464	0.9987	733.2
0.222	0.444	377.72	1.72	0.193	0.168	0.9856	0.9778	0.9778	723.3
0.163	0.326	371.94	−0.35	0.113	0.095	0.9903	0.9673	0.9900	767.9
0.280	0.560	390.20	−0.94	0.320	0.294	0.9799	0.9862	0.9544	780.0
0.332	0.336	376.39	1.15	0.275	0.119	0.9931	0.9677	0.9848	735.2
0.133	0.734	394.83	0.19	0.175	0.460	0.9631	0.9950	0.9352	756.0

* pressures calculated at experimental temperatures

By assuming that the main parameters of the Wilson equation $(\lambda_{ij} - \lambda_{ii})$ vary linearly with the temperature up to 298.15 K, the ternary excess Gibbs energies G^E vs. x_i at $x_j/x_k = 1$ are shown in Fig. 3. In the same figure the experimental H^E data at 298.15 K obtained by us earlier [3] and the entropic term TS^E calculated from the difference $H^E - G^E$ are also plotted.

In Fig. 3a H^E and TS^E are s-shaped changing from positive to negative and to positive again with increasing mole fraction of 1,2-dichloroethane, x_3. Up to x_3 about 0.2, TS^E is larger than H^E, which results in a negative G^E. With increasing x_3, G^E becomes positive when $|TS^E| > |H^E|$. This phenomenon is valid also when aromatic hydrocarbons are added as seen in Fig. 3b and c. Presumably, the formation of charge transfer (or donor–acceptor) complexes between the aromatic hydrocarbon and 1,2-dichloroethane and their decomposition are the most important factors influencing the signs and values of excess functions in this ternary system.
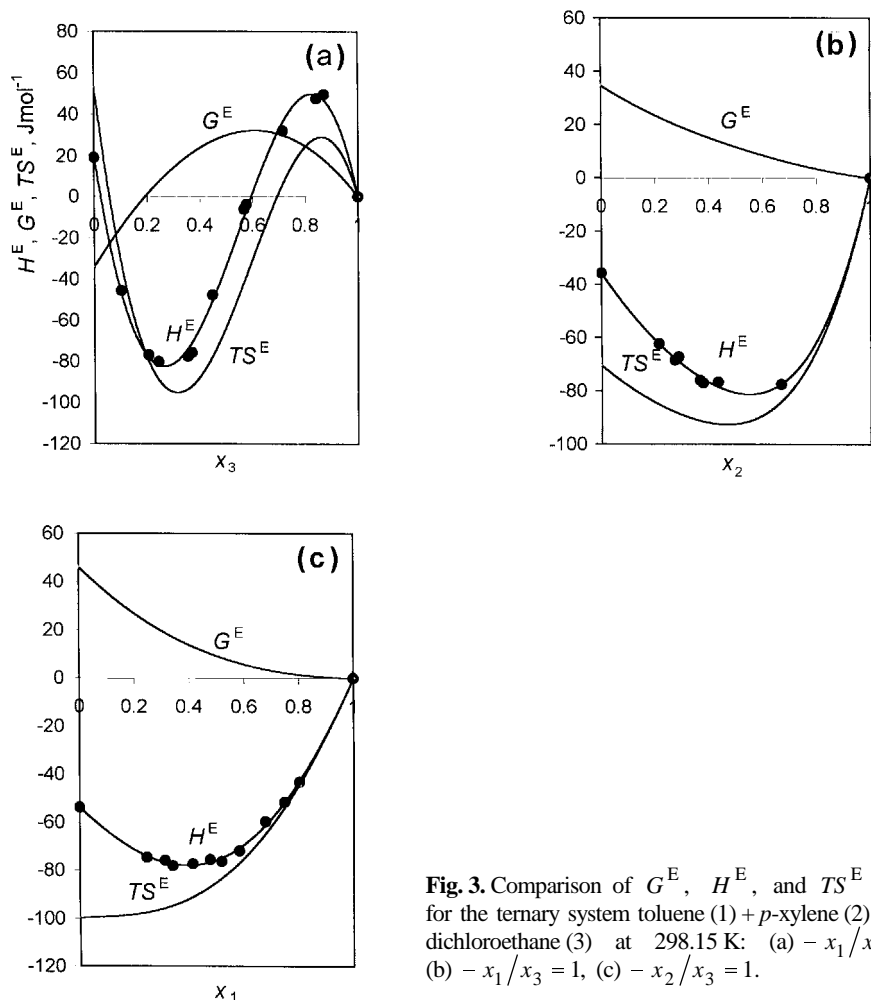
Fig. 3. Comparison of G^E, H^E, and TS^E values for the ternary system toluene (1) + p-xylene (2) + 1,2-dichloroethane (3) at 298.15 K: (a) – $x_1/x_2 = 1$, (b) – $x_1/x_3 = 1$, (c) – $x_2/x_3 = 1$.

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Kolmiksüsteemi tolueen + p-ksüleen + 1,2-dikloroetaan isobaariline auru ja vedeliku tasakaal

Helle Kirss, Mati Kuus, Enn Siimer ja Ludmilla Kudrjavtseva

Isobaarilist auru ja vedeliku tasakaalu mõõdeti süsteemi tolueen + p-ksüleen + 1,2-dikloroetaan ja selles sisalduvate binaarsete süsteemide puhul rõhkudel 26,66; 53,33; 79,99 ja 101,32 kPa. Kaksiksüsteemide auru ja vedeliku tasakaalu korreleerimiseks ja kolmiksüsteemi oma ennustamiseks kasutati modifitseeritud Wilsoni võrrandit, arvestades selle põhiparameetrite $(\lambda_{ij} - \lambda_{ii})$ lineaarset sõltuvust temperatuurist. Võrreldi kolmiksüsteemi liigfunktsioonide väärtusi 298,15 K juures.