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Viscosity of Alcohol–Ethene Mixtures at Pressures up to 195 MPa

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Viscosities of several alcohol–ethene and vinyl acetate–ethene mixtures were measured with a rolling-ball viscometer. The viscosity measurements were performed at temperatures from 298 to 413 K and pressures up to 195 MPa with an estimated uncertainty of ±2%. The appearance of hydrogen bonds forming alcohol associates has a notable effect on the viscosity of alcohol–ethene mixtures above an alcohol mole fraction of about 0.2. This could be noticed from the deviation of the viscosity from the Arrhenius law, which occurs when alcohol associates are formed. An effect of pressure on the association of alcohol molecules through hydrogen bonding was not observable from the viscosity data. A new method for the description of the temperature- and pressure-dependent parameters in the McAllister and Dizechi models is proposed.

KEY WORDS: alcohols; density; ethene; high pressure; hydrogen bond; mixtures; vinyl acetate; viscosity.

1. INTRODUCTION

Alcohol–ethene mixtures are used in several chemical processes. One important example is supercritical extraction, in which the alcohol is used as an entraining agent. For the design of a process one needs the viscosity, in addition to other data. Therefore viscosities of alcohol–ethene mixtures were measured in this work.

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2. MEASUREMENTS

2.1. Materials

The alcohols were supplied by Aldrich-Chemie GmbH & Co. KG, Steinheim. The samples of 1-pentanol and 2-methyl-2-butanol had a purity of better than 99%. 3-Pentanol, 1-heptanol, 3-ethyl-3-pentanol, 1-nonanol, and 5-nonanol had a nominal purity of 98%. The vinyl acetate and the *n*-decane used for calibration were supplied by Merck KGaA, Darmstadt, at a purity of 99% or better. Ethene was supplied by Erdölchemie, Köln, with a purity of 99.9996%.

2.2. Procedure

The viscosities were measured with a rolling-ball viscometer, constructed by Stanislawski and Luft [1–3]. It consists of a Schott KPG glass tube, which is closed at one end, with a steel ball inside this tube. In addition, an electromagnet is placed on the open end of the glass tube. This arrangement is installed in a high-pressure autoclave. To achieve a wide measuring range, the inclination from a horizontal line can be varied in six steps from 3.5 to 80°. The rolling time is detected inductively. For this purpose two pairs of measuring coils form the boundary of the measuring distance. When starting the experiment, the viscometer is tilted in measuring position, the electromagnet is released, and the steel ball begins to roll. After a short distance, when a constant rolling velocity is reached, the ball passes the first pair of coils and the timer starts. At the end of the measuring distance, the timer is stopped by a signal from the second pair of coils. Because the rolling-ball viscometer is not a fundamental method, it must be calibrated. For this purpose *n*-decane was used, whose viscosity values are known in the entire range of pressure and temperature [4].

The temperature in the autoclave was measured at two positions by NiCr/Ni thermocouples with an accuracy of ± 0.2 K. The pressure was determined by a calibrated strain gauge pressure sensor with an accuracy of $\pm 0.3\%$. Each measuring point was determined about 10 times. The deviation of the rolling times was 0.3%.

The preparation of the mixtures was performed within an autoclave. The liquid components were metered by a high-pressure hand pump. The ethene was condensed and fed into two high-pressure vessels using a two-stage diaphragm compressor. From there the ethene was fed into the autoclave. It was possible to determine the amount of ethene injected from the difference in density before and after the addition and the known volume of the two high-pressure vessels. The densities of ethene were computed

using an equation from Angus et al. [5], recommended by the IUPAC. The components inside the autoclave were mixed for several hours to achieve homogeneity before commencing the viscosity measurements.

Evaluation of the viscosities was carried out by the method of Hubbard and Brown [6]. This method is based on the analysis of the dimensions of all parameters influencing the rolling time. For this purpose a resistence factor was defined which depends on the diameter of the ball and the tube, the ball velocity, the angle of incline, the density of the steel ball, and the density of the substance. At Reynold's numbers smaller than 50, a plot of the logarithm of the resistance factors against the logarithm of the Reynolds numbers shows a slope of -1. The calibration constant, which depends only on the temperature and the pressure, was received by the axis segment.

Densities and mixture compositions were determined by the weight of components placed in the autoclave and the autoclave volume as function of temperature and pressure measured during calibration with n-decane. The estimated error in density is about 1.7%. Knowledge of the densities was necessary for calculation of the kinematic viscosities.

3. RESULTS AND DISCUSSION

Tables I to VIII give the viscosities and densities of the liquid alcohol–ethene mixtures at several temperatures and pressures. The measuring error was estimated to be about $\pm 2\%$, taking into account all individual errors inclusive the accuracy of the literature values used in calibration.

The viscosity data of the pure liquids have been measured by Sülzner and Luft [3] and the viscosities of the ethene were taken from Stanislawski and Luft [1].

3.1. Temperature Dependence of the Viscosities

The viscosities of alcohol–ethene mixtures are influenced by hydrogen bonding just as those of pure alcohols [2, 3]. The addition of ethene reduces this influence by the entropic effect, which reduces the size of the associated alcohol clusters.

The existence of associated alcohol groups formed by hydrogen bonding causes, especially at low temperatures, a greater viscosity temperature dependence than exists for nonassociated substances. Moreover, the viscosity temperature dependence shows a distinct deviation from the Andrade equation,

$$\eta = \eta_0 \exp\left(\frac{E_{\text{Vis}}}{RT}\right) \tag{1}$$

Table I. Viscosities and Densities of 1-Pentanol–Ethene Mixtures

T (K)	P (MPa)	x	ρ (kg · cm^{-3})	η (mPa · s)
298.15	50	0.796	0.802	2.840
298.15	80	0.795	0.818	3.602
298.15	120	0.794	0.836	4.663
298.15	160	0.792	0.851	5.907
298.15	195	0.789	0.862	6.889
323.15	50	0.792	0.784	1.550
323.15	80	0.792	0.802	1.964
323.15	120	0.791	0.822	2.513
323.15	160	0.790	0.838	3.099
323.15	195	0.787	0.851	3.677
373.15	50	0.791	0.748	0.671
373.15	80	0.790	0.769	0.824
373.15	120	0.790	0.792	0.998
373.15	160	0.789	0.810	1.230
373.15	195	0.786	0.824	1.461
298.15	50	0.623	0.760	1.555
298.15	80	0.622	0.778	1.989
298.15	120	0.620	0.798	2.557
298.15	160	0.617	0.813	3.113
298.15	195	0.608	0.824	3.727
323.15	50	0.621	0.737	0.896
323.15	80	0.620	0.757	1.111
323.15	120	0.619	0.779	1.425
323.15	160	0.617	0.796	1.726
323.15	195	0.615	0.810	2.048
373.15	50	0.622	0.708	0.428
373.15	80	0.622	0.733	0.522
373.15	120	0.621	0.758	0.647
373.15	160	0.620	0.778	0.835
373.15	195	0.615	0.786	0.997
298.15	50	0.409	0.689	0.586
298.15	80	0.408	0.711	0.744
298.15	120	0.407	0.734	0.936
298.15	160	0.405	0.753	1.139
298.15	195	0.401	0.766	1.324
323.15	50	0.413	0.660	0.384
323.15	80	0.412	0.685	0.478
323.15	120	0.412	0.711	0.608

Table I. (*Continued*)

T (K)	P (MPa)	x	ρ (kg · cm^{-3})	η (mPa · s)
323.15	160	0.410	0.730	0.734
323.15	195	0.406	0.746	0.847
373.15	50	0.400	0.616	0.214
373.15	80	0.398	0.646	0.280
373.15	120	0.403	0.676	0.343
373.15	160	0.402	0.699	0.419
373.15	195	0.400	0.716	0.491
298.15	50	0.206	0.573	0.232
298.15	80	0.204	0.602	0.284
298.15	120	0.203	0.631	0.355
298.15	160	0.201	0.654	0.428
298.15	195	0.199	0.669	0.485
323.15	50	0.207	0.560	0.180
323.15	80	0.206	0.593	0.219
323.15	120	0.206	0.623	0.272
323.15	160	0.205	0.647	0.326
323.15	195	0.203	0.664	0.378
373.15	50	0.206	0.510	0.123
373.15	80	0.204	0.550	0.146
373.15	120	0.203	0.587	0.177
373.15	160	0.202	0.614	0.212
373.15	195	0.200	0.632	0.242
298.15	50	0.103	0.534	0.137
298.15	80	0.103	0.563	0.166
298.15	120	0.102	0.597	0.202
298.15	160	0.102	0.624	0.232
298.15	195	0.101	0.639	0.254
323.15	50	0.109	0.509	0.123
323.15	80	0.108	0.546	0.143
323.15	120	0.108	0.580	0.170
323.15	160	0.107	0.605	0.196
323.15	195	0.106	0.623	0.226
373.15	50	0.102	0.453	0.087
373.15	80	0.101	0.501	0.112
373.15	120	0.101	0.540	0.133
373.15	160	0.100	0.568	0.158
373.15	195	0.099	0.588	0.184

Table II. Viscosities and Densities of 3-Pentanol–Ethene Mixtures

T (K)	P (MPa)	x	ρ (kg · cm^{-3})	η (mPa · s)
298.15	50	0.810	0.808	3.132
298.15	80	0.806	0.826	4.323
298.15	120	0.805	0.845	6.037
298.15	160	0.802	0.860	8.304
298.15	195	0.797	0.872	11.215
323.15	50	0.796	0.784	1.310
323.15	80	0.796	0.802	1.675
323.15	120	0.795	0.822	2.264
323.15	160	0.794	0.839	2.999
323.15	195	0.791	0.852	3.764
373.15	50	0.793	0.745	0.501
373.15	80	0.793	0.767	0.629
373.15	120	0.793	0.791	0.812
373.15	160	0.792	0.810	1.010
373.15	195	0.790	0.825	1.223
298.15	50	0.616	0.760	1.157
298.15	80	0.615	0.779	1.528
298.15	120	0.614	0.800	2.043
298.15	160	0.610	0.817	2.678
298.15	195	0.601	0.828	3.351
323.15	50	0.594	0.731	0.617
323.15	80	0.594	0.752	0.774
323.15	120	0.594	0.775	1.003
323.15	160	0.593	0.794	1.271
323.15	195	0.592	0.808	1.566
373.15	50	0.604	0.694	0.322
373.15	80	0.604	0.720	0.399
373.15	120	0.603	0.746	0.512
373.15	160	0.601	0.767	0.630
373.15	195	0.597	0.781	0.739
298.15	50	0.425	0.694	0.468
298.15	80	0.424	0.716	0.603
298.15	120	0.423	0.742	0.774
298.15	160	0.421	0.761	0.967
298.15	195	0.416	0.774	1.156
323.15	50	0.398	0.658	0.303
323.15	80	0.397	0.685	0.373
323.15	120	0.397	0.711	0.467

Table II. (*Continued*)

T (K)	P (MPa)	x	ρ (kg · cm^{-3})	η (mPa · s)
323.15	160	0.396	0.731	0.573
323.15	195	0.394	0.746	0.667
373.15	50	0.408	0.620	0.196
373.15	80	0.407	0.651	0.243
373.15	120	0.406	0.682	0.306
373.15	160	0.405	0.704	0.367
373.15	195	0.401	0.721	0.431
298.15	50	0.198	0.555	0.185
298.15	80	0.198	0.584	0.227
298.15	120	0.198	0.613	0.277
298.15	160	0.198	0.635	0.334
298.15	195	0.196	0.651	0.380
323.15	50	0.198	0.528	0.152
323.15	80	0.198	0.561	0.181
323.15	120	0.198	0.592	0.226
323.15	160	0.198	0.615	0.271
323.15	195	0.196	0.632	0.310
373.15	50	0.196	0.479	0.113
373.15	80	0.196	0.519	0.137
373.15	120	0.197	0.556	0.170
373.15	160	0.196	0.582	0.203
373.15	195	0.195	0.601	0.234
298.15	50	0.109	0.508	0.136
298.15	80	0.110	0.539	0.165
298.15	120	0.110	0.569	0.200
298.15	160	0.110	0.592	0.232
298.15	195	0.109	0.609	0.261
323.15	50	0.107	0.480	0.118
323.15	80	0.108	0.516	0.137
323.15	120	0.109	0.548	0.166
323.15	160	0.109	0.572	0.197
323.15	195	0.108	0.590	0.222
373.15	50	0.102	0.432	0.088
373.15	80	0.104	0.477	0.111
373.15	120	0.105	0.516	0.136
373.15	160	0.105	0.544	0.162
373.15	195	0.105	0.563	0.186

Table III. Viscosities and Densities of 2-Methyl-2-Butanol–Ethene Mixtures

T (K)	P (MPa)	x	ρ (kg · cm^{-3})	η (mPa · s)
298.15	50	0.790	0.808	2.466
298.15	80	0.790	0.825	3.617
298.15	120	0.788	0.844	5.638
298.15	160	0.785	0.860	8.378
298.15	195	0.779	0.871	12.366
323.15	50	0.782	0.786	1.234
323.15	80	0.781	0.805	1.648
323.15	120	0.780	0.826	2.381
323.15	160	0.778	0.844	3.358
323.15	195	0.774	0.856	4.411
373.15	50	0.777	0.749	0.484
373.15	80	0.777	0.773	0.626
373.15	120	0.776	0.797	0.839
373.15	160	0.773	0.817	1.093
373.15	195	0.769	0.831	1.372
298.15	50	0.583	0.735	0.830
298.15	80	0.582	0.756	1.117
298.15	120	0.581	0.778	1.529
298.15	160	0.580	0.796	2.033
298.15	195	0.577	0.809	2.564
323.15	50	0.596	0.711	0.578
323.15	80	0.595	0.733	0.743
323.15	120	0.594	0.758	1.000
323.15	160	0.593	0.779	1.318
323.15	195	0.588	0.793	1.648
373.15	50	0.606	0.673	0.311
373.15	80	0.606	0.700	0.393
373.15	120	0.605	0.727	0.518
373.15	160	0.602	0.749	0.650
373.15	195	0.598	0.763	0.775
298.15	50	0.408	0.661	0.369
298.15	80	0.407	0.686	0.476
298.15	120	0.406	0.711	0.619
298.15	160	0.404	0.731	0.769
298.15	195	0.400	0.745	0.897
323.15	50	0.407	0.635	0.276
323.15	80	0.406	0.662	0.345
323.15	120	0.405	0.688	0.442

Table III. (*Continued*)

T (K)	P (MPa)	x	ρ (kg · cm^{-3})	η (mPa · s)
323.15	160	0.403	0.710	0.550
323.15	195	0.400	0.725	0.650
373.15	50	0.415	0.593	0.179
373.15	80	0.414	0.627	0.225
373.15	120	0.412	0.658	0.287
373.15	160	0.411	0.682	0.352
373.15	195	0.408	0.699	0.418
298.15	50	0.211	0.581	0.170
298.15	80	0.210	0.611	0.211
298.15	120	0.209	0.640	0.263
298.15	160	0.207	0.662	0.312
298.15	195	0.205	0.678	0.357
323.15	50	0.212	0.556	0.146
323.15	80	0.211	0.589	0.178
323.15	120	0.210	0.620	0.219
323.15	160	0.210	0.644	0.266
323.15	195	0.207	0.661	0.301
373.15	50	0.212	0.506	0.111
373.15	80	0.210	0.547	0.137
373.15	120	0.208	0.584	0.169
373.15	160	0.208	0.610	0.202
373.15	195	0.206	0.629	0.230
298.15	50	0.103	0.526	0.126
298.15	80	0.102	0.555	0.152
298.15	120	0.102	0.589	0.180
298.15	160	0.101	0.617	0.207
298.15	195	0.100	0.632	0.231
323.15	50	0.111	0.504	0.111
323.15	80	0.110	0.542	0.129
323.15	120	0.109	0.575	0.156
323.15	160	0.109	0.600	0.183
323.15	195	0.107	0.618	0.209
373.15	50	0.121	0.460	0.091
373.15	80	0.119	0.506	0.109
373.15	120	0.118	0.546	0.133
373.15	160	0.117	0.574	0.157
373.15	195	0.116	0.594	0.183

Table IV. Viscosities and Densities of 1-Heptanol–Ethene Mixtures

T (K)	P (MPa)	x	ρ (kg · cm^{-3})	η (mPa · s)
298.15	50	0.804	0.820	5.441
298.15	80	0.803	0.835	7.072
298.15	120	0.802	0.852	9.495
298.15	160	0.799	0.867	12.642
298.15	195	0.793	0.877	15.682
323.15	50	0.799	0.801	2.742
323.15	80	0.799	0.817	3.425
323.15	120	0.798	0.835	4.531
323.15	160	0.796	0.851	5.884
323.15	195	0.786	0.862	7.205
373.15	50	0.798	0.749	1.020
373.15	80	0.797	0.768	1.267
373.15	120	0.797	0.789	1.630
373.15	160	0.795	0.806	2.058
373.15	195	0.792	0.819	2.483
298.15	50	0.611	0.762	2.661
298.15	80	0.610	0.780	3.436
298.15	120	0.608	0.798	4.500
298.15	160	0.605	0.814	5.755
298.15	195	0.598	0.825	7.030
323.15	50	0.606	0.742	1.444
323.15	80	0.605	0.761	1.789
323.15	120	0.604	0.781	2.326
323.15	160	0.603	0.798	2.953
323.15	195	0.600	0.810	3.560
373.15	50	0.609	0.708	0.629
373.15	80	0.608	0.730	0.782
373.15	120	0.607	0.754	1.000
373.15	160	0.606	0.772	1.237
373.15	195	0.603	0.786	1.493
298.15	50	0.404	0.696	0.920
298.15	80	0.403	0.717	1.171
298.15	120	0.402	0.738	1.488
298.15	160	0.401	0.757	1.846
298.15	195	0.399	0.770	2.161
323.15	50	0.391	0.668	0.573
323.15	80	0.391	0.692	0.709
323.15	120	0.390	0.715	0.911

Table IV. (*Continued*)

T (K)	P (MPa)	x	ρ (kg · cm^{-3})	η (mPa · s)
323.15	160	0.388	0.734	1.122
323.15	195	0.386	0.748	1.323
373.15	50	0.393	0.630	0.316
373.15	80	0.393	0.659	0.391
373.15	120	0.392	0.686	0.499
373.15	160	0.391	0.708	0.608
373.15	195	0.389	0.724	0.712
298.15	50	0.207	0.620	0.291
298.15	80	0.207	0.647	0.366
298.15	120	0.206	0.674	0.451
298.15	160	0.205	0.695	0.540
298.15	195	0.202	0.710	0.617
323.15	50	0.221	0.604	0.237
323.15	80	0.219	0.632	0.292
323.15	120	0.218	0.660	0.368
323.15	160	0.217	0.682	0.446
323.15	195	0.214	0.698	0.512
373.15	50	0.218	0.558	0.155
373.15	80	0.216	0.595	0.194
373.15	120	0.215	0.628	0.246
373.15	160	0.214	0.653	0.297
373.15	195	0.211	0.670	0.344
298.15	50	0.097	0.545	0.154
298.15	80	0.097	0.576	0.188
298.15	120	0.096	0.606	0.234
298.15	160	0.096	0.629	0.277
298.15	195	0.095	0.645	0.312
323.15	50	0.104	0.525	0.131
323.15	80	0.103	0.560	0.161
323.15	120	0.103	0.592	0.202
323.15	160	0.102	0.617	0.236
323.15	195	0.101	0.634	0.261
373.15	50	0.107	0.483	0.100
373.15	80	0.106	0.527	0.126
373.15	120	0.106	0.565	0.156
373.15	160	0.105	0.593	0.186
373.15	195	0.104	0.612	0.213

Table V. Viscosities and Densities of 3-Ethyl-3-Pentanol–Ethene Mixtures

T (K)	P (MPa)	x	ρ (kg · cm^{-3})	η (mPa · s)
298.15	50	0.783	0.833	4.009
298.15	80	0.782	0.849	5.969
298.15	120	0.780	0.866	9.969
298.15	160	0.776	0.880	15.999
298.15	195	0.770	0.891	24.250
323.15	50	0.783	0.811	1.504
323.15	80	0.782	0.829	2.050
323.15	120	0.781	0.848	3.027
323.15	160	0.780	0.864	4.411
323.15	195	0.777	0.876	6.158
373.15	50	0.783	0.764	0.540
373.15	80	0.782	0.785	0.700
373.15	120	0.782	0.808	0.942
373.15	160	0.780	0.827	1.236
373.15	195	0.777	0.840	1.575
413.15	50	0.783	0.729	0.341
413.15	80	0.783	0.754	0.435
413.15	120	0.781	0.780	0.569
413.15	160	0.780	0.801	0.720
413.15	195	0.776	0.816	0.877
298.15	50	0.601	0.791	1.519
298.15	80	0.600	0.809	2.105
298.15	120	0.599	0.828	3.154
298.15	160	0.596	0.843	4.429
298.15	195	0.590	0.854	6.041
323.15	50	0.601	0.766	0.808
323.15	80	0.600	0.786	1.080
323.15	120	0.599	0.807	1.420
323.15	160	0.596	0.825	1.895
323.15	195	0.592	0.838	2.474
373.15	50	0.600	0.717	0.368
373.15	80	0.599	0.741	0.469
373.15	120	0.597	0.767	0.623
373.15	160	0.594	0.786	0.798
373.15	195	0.590	0.801	0.982
413.15	50	0.599	0.676	0.239
413.15	80	0.598	0.705	0.309
413.15	120	0.596	0.735	0.395
413.15	160	0.594	0.757	0.489
413.15	195	0.590	0.774	0.586
298.15	50	0.422	0.725	0.647
298.15	80	0.421	0.746	0.832
298.15	120	0.419	0.768	1.121
298.15	160	0.417	0.786	1.458
298.15	195	0.413	0.798	1.821
323.15	50	0.420	0.697	0.411
323.15	80	0.418	0.720	0.515
323.15	120	0.417	0.744	0.676
323.15	160	0.415	0.764	0.852
323.15	195	0.413	0.779	1.034

Table V. (*Continued*)

T (K)	P (MPa)	x	ρ (kg · cm^{-3})	η (mPa · s)
373.15	50	0.403	0.639	0.209
373.15	80	0.402	0.669	0.271
373.15	120	0.400	0.698	0.353
373.15	160	0.398	0.720	0.443
373.15	195	0.395	0.737	0.542
413.15	50	0.406	0.594	0.146
413.15	80	0.403	0.630	0.189
413.15	120	0.402	0.663	0.242
413.15	160	0.400	0.689	0.298
413.15	195	0.397	0.707	0.357
298.15	50	0.211	0.618	0.243
298.15	80	0.210	0.647	0.300
298.15	120	0.208	0.670	0.382
298.15	160	0.207	0.688	0.461
298.15	195	0.205	0.709	0.544
323.15	50	0.216	0.590	0.186
323.15	80	0.214	0.622	0.228
323.15	120	0.213	0.653	0.286
323.15	160	0.211	0.676	0.346
323.15	195	0.210	0.697	0.407
373.15	50	0.221	0.550	0.138
373.15	80	0.218	0.579	0.165
373.15	120	0.217	0.620	0.209
373.15	160	0.215	0.648	0.250
373.15	195	0.216	0.671	0.298
413.15	50	0.243	0.507	0.094
413.15	80	0.239	0.555	0.121
413.15	120	0.236	0.600	0.157
413.15	160	0.234	0.624	0.188
413.15	195	0.232	0.650	0.231
298.15	50	0.151	0.581	0.194
298.15	80	0.150	0.609	0.238
298.15	120	0.149	0.642	0.298
298.15	160	0.147	0.656	0.350
298.15	195	0.146	0.677	0.405
323.15	50	0.137	0.542	0.148
323.15	80	0.135	0.579	0.181
323.15	120	0.134	0.609	0.223
323.15	160	0.133	0.629	0.265
323.15	195	0.131	0.657	0.307
373.15	50	0.123	0.489	0.110
373.15	80	0.120	0.525	0.137
373.15	120	0.118	0.565	0.169
373.15	160	0.117	0.603	0.201
373.15	195	0.115	0.621	0.238
413.15	50	0.122	0.438	0.073
413.15	80	0.118	0.487	0.093
413.15	120	0.115	0.530	0.114
413.15	160	0.114	0.564	0.136
413.15	195	0.113	0.589	0.159

Table VI. Viscosities and Densities of 1-Nonanol–Ethene Mixtures

T (K)	P (MPa)	x	ρ (kg · cm^{-3})	η (mPa · s)
298.15	50	0.799	0.848	8.283
298.15	80	0.798	0.862	10.879
298.15	120	0.796	0.877	15.306
298.15	160	0.793	0.891	20.374
298.15	195	0.787	0.901	25.852
323.15	50	0.797	0.828	3.884
323.15	80	0.797	0.844	5.000
323.15	120	0.796	0.861	6.811
323.15	160	0.794	0.876	8.928
323.15	195	0.791	0.888	11.224
373.15	50	0.802	0.781	1.350
373.15	80	0.802	0.800	1.713
373.15	120	0.802	0.820	2.266
373.15	160	0.801	0.836	2.898
373.15	195	0.800	0.849	3.603
413.15	50	0.806	0.751	0.747
413.15	80	0.806	0.773	0.944
413.15	120	0.806	0.796	1.212
413.15	160	0.806	0.815	1.522
413.15	195	0.805	0.830	1.854
298.15	50	0.592	0.793	3.673
298.15	80	0.591	0.809	4.719
298.15	120	0.589	0.827	6.465
298.15	160	0.585	0.842	8.362
298.15	195	0.580	0.853	10.502
323.15	50	0.588	0.773	1.868
323.15	80	0.588	0.791	2.369
323.15	120	0.587	0.810	3.138
323.15	160	0.585	0.826	3.984
323.15	195	0.581	0.838	4.852
373.15	50	0.589	0.732	0.777
373.15	80	0.588	0.753	0.983
373.15	120	0.587	0.776	1.261
373.15	160	0.585	0.794	1.589
373.15	195	0.582	0.808	1.944
413.15	50	0.587	0.698	0.465
413.15	80	0.586	0.723	0.588
413.15	120	0.584	0.749	0.754
413.15	160	0.582	0.770	0.931
413.15	195	0.579	0.785	1.128
298.15	50	0.410	0.740	1.541
298.15	80	0.409	0.759	1.960
298.15	120	0.407	0.779	2.590
298.15	160	0.403	0.796	3.252
298.15	195	0.397	0.807	3.936
323.15	50	0.408	0.718	0.892
323.15	80	0.407	0.740	1.116
323.15	120	0.406	0.762	1.446
323.15	160	0.405	0.780	1.790
323.15	195	0.402	0.793	2.087

Table VI. (*Continued*)

T (K)	P (MPa)	x	ρ (kg · cm^{-3})	η (mPa · s)
373.15	50	0.403	0.675	0.440
373.15	80	0.402	0.701	0.559
373.15	120	0.401	0.726	0.721
373.15	160	0.399	0.746	0.892
373.15	195	0.396	0.761	1.074
413.15	50	0.406	0.650	0.295
413.15	80	0.406	0.680	0.375
413.15	120	0.405	0.709	0.480
413.15	160	0.403	0.732	0.591
413.15	195	0.401	0.748	0.708
298.15	50	0.202	0.632	0.372
298.15	80	0.200	0.664	0.465
298.15	120	0.199	0.693	0.597
298.15	160	0.197	0.715	0.712
298.15	195	0.193	0.731	0.841
323.15	50	0.201	0.606	0.269
323.15	80	0.199	0.643	0.338
323.15	120	0.197	0.675	0.427
323.15	160	0.194	0.699	0.521
323.15	195	0.191	0.716	0.617
373.15	50	0.206	0.554	0.168
373.15	80	0.202	0.602	0.214
373.15	120	0.199	0.641	0.275
373.15	160	0.197	0.668	0.334
373.15	195	0.194	0.688	0.399
413.15	50	0.210	0.515	0.122
413.15	80	0.204	0.571	0.154
413.15	120	0.200	0.615	0.196
413.15	160	0.196	0.645	0.241
413.15	195	0.192	0.666	0.286
298.15	50	0.107	0.562	0.199
298.15	80	0.106	0.592	0.245
298.15	120	0.106	0.621	0.307
298.15	160	0.105	0.643	0.361
298.15	195	0.104	0.659	0.415
323.15	50	0.102	0.539	0.150
323.15	80	0.101	0.573	0.189
323.15	120	0.101	0.605	0.239
323.15	160	0.100	0.629	0.283
323.15	195	0.099	0.646	0.326
373.15	50	0.097	0.479	0.109
373.15	80	0.096	0.522	0.136
373.15	120	0.095	0.560	0.171
373.15	160	0.095	0.587	0.205
373.15	195	0.093	0.606	0.241
413.15	50	0.105	0.441	0.089
413.15	80	0.103	0.491	0.113
413.15	120	0.103	0.533	0.142
413.15	160	0.102	0.563	0.173
413.15	195	0.101	0.584	0.205

Table VII. Viscosities and Densities of 5-Nonanol–Ethene Mixtures

T (K)	P (MPa)	x	ρ (kg · cm^{-3})	η (mPa · s)
298.15	50	0.797	0.822	7.877
298.15	80	0.795	0.837	11.343
298.15	120	0.794	0.854	17.723
298.15	160	0.790	0.868	25.714
298.15	195	0.784	0.878	35.373
323.15	50	0.805	0.805	3.308
323.15	80	0.803	0.822	4.523
323.15	120	0.802	0.840	6.611
323.15	160	0.798	0.855	9.294
323.15	195	0.791	0.865	12.419
373.15	50	0.801	0.763	0.981
373.15	80	0.800	0.782	1.282
373.15	120	0.799	0.803	1.751
373.15	160	0.797	0.821	2.310
373.15	195	0.793	0.834	2.969
413.15	50	0.801	0.732	0.550
413.15	80	0.800	0.755	0.710
413.15	120	0.798	0.779	0.933
413.15	160	0.794	0.799	1.195
413.15	195	0.788	0.813	1.480
298.15	50	0.591	0.783	2.645
298.15	80	0.590	0.800	3.578
298.15	120	0.589	0.819	5.207
298.15	160	0.587	0.834	7.005
298.15	195	0.583	0.846	9.060
323.15	50	0.595	0.764	1.375
323.15	80	0.593	0.783	1.804
323.15	120	0.592	0.803	2.488
323.15	160	0.589	0.819	3.269
323.15	195	0.585	0.831	4.141
373.15	50	0.587	0.720	0.577
373.15	80	0.586	0.742	0.745
373.15	120	0.585	0.765	0.994
373.15	160	0.583	0.784	1.271
373.15	195	0.580	0.798	1.572
413.15	50	0.593	0.683	0.359
413.15	80	0.592	0.710	0.461
413.15	120	0.590	0.737	0.597
413.15	160	0.588	0.758	0.750
413.15	195	0.584	0.774	0.919
298.15	50	0.406	0.727	0.958
298.15	80	0.405	0.747	1.246
298.15	120	0.404	0.769	1.689
298.15	160	0.401	0.785	2.140
298.15	195	0.397	0.797	2.628
323.15	50	0.409	0.706	0.621
323.15	80	0.408	0.729	0.789
323.15	120	0.407	0.751	1.047
323.15	160	0.405	0.770	1.316
323.15	195	0.402	0.784	1.591

Table VII. (*Continued*)

T (K)	P (MPa)	x	ρ (kg · cm^{-3})	η (mPa · s)
373.15	50	0.402	0.659	0.323
373.15	80	0.400	0.686	0.412
373.15	120	0.398	0.713	0.535
373.15	160	0.396	0.734	0.668
373.15	195	0.394	0.749	0.812
413.15	50	0.405	0.622	0.226
413.15	80	0.403	0.654	0.290
413.15	120	0.401	0.685	0.375
413.15	160	0.399	0.709	0.460
413.15	195	0.397	0.726	0.551
298.15	50	0.174	0.615	0.327
298.15	80	0.173	0.647	0.393
298.15	120	0.172	0.677	0.475
298.15	160	0.171	0.699	0.554
298.15	195	0.169	0.715	0.617
323.15	50	0.181	0.593	0.254
323.15	80	0.180	0.630	0.311
323.15	120	0.179	0.663	0.382
323.15	160	0.177	0.686	0.450
323.15	195	0.176	0.703	0.507
373.15	50	0.191	0.548	0.173
373.15	80	0.189	0.594	0.224
373.15	120	0.188	0.633	0.289
373.15	160	0.186	0.660	0.354
373.15	195	0.185	0.679	0.412
413.15	50	0.183	0.502	0.123
413.15	80	0.180	0.557	0.161
413.15	120	0.178	0.601	0.209
413.15	160	0.176	0.631	0.257
413.15	195	0.175	0.652	0.300
298.15	50	0.109	0.563	0.194
298.15	80	0.109	0.593	0.236
298.15	120	0.109	0.622	0.297
298.15	160	0.108	0.643	0.352
298.15	195	0.106	0.659	0.411
323.15	50	0.102	0.525	0.156
323.15	80	0.101	0.559	0.186
323.15	120	0.100	0.591	0.228
323.15	160	0.100	0.615	0.271
323.15	195	0.098	0.634	0.313
373.15	50	0.104	0.475	0.114
373.15	80	0.103	0.518	0.144
373.15	120	0.102	0.555	0.179
373.15	160	0.101	0.583	0.217
373.15	195	0.100	0.602	0.256
413.15	50	0.102	0.427	0.083
413.15	80	0.100	0.478	0.111
413.15	120	0.099	0.521	0.140
413.15	160	0.099	0.551	0.167
413.15	195	0.097	0.573	0.196

Table VIII. Viscosities and Densities of Vinyl Acetate–Ethene Mixtures

T (K)	P (MPa)	x	ρ (kg · cm^{-3})	η (mPa · s)
298.15	50	0.805	0.873	0.433
298.15	80	0.804	0.890	0.515
298.15	120	0.803	0.924	0.617
298.15	160	0.805	0.962	0.720
298.15	195	0.802	0.992	0.810
323.15	50	0.803	0.849	0.346
323.15	80	0.803	0.873	0.401
323.15	120	0.803	0.899	0.483
323.15	160	0.803	0.920	0.560
323.15	195	0.801	0.936	0.635
373.15	50	0.805	0.807	0.248
373.15	80	0.805	0.836	0.296
373.15	120	0.805	0.866	0.353
373.15	160	0.805	0.897	0.421
373.15	195	0.803	0.910	0.489
298.15	50	0.587	0.809	0.328
298.15	80	0.587	0.833	0.384
298.15	120	0.587	0.857	0.456
298.15	160	0.586	0.877	0.538
298.15	195	0.585	0.892	0.599
323.15	50	0.598	0.800	0.271
323.15	80	0.599	0.826	0.312
323.15	120	0.599	0.853	0.375
323.15	160	0.599	0.875	0.431
323.15	195	0.598	0.892	0.491
373.15	50	0.592	0.747	0.201
373.15	80	0.593	0.778	0.238
373.15	120	0.594	0.810	0.283
373.15	160	0.594	0.834	0.340
373.15	195	0.593	0.853	0.390
298.15	50	0.416	0.739	0.241
298.15	80	0.417	0.763	0.289
298.15	120	0.417	0.787	0.348
298.15	160	0.416	0.807	0.400
298.15	195	0.415	0.822	0.449
323.15	50	0.388	0.699	0.187
323.15	80	0.386	0.729	0.218
323.15	120	0.385	0.759	0.268

Table VIII. (*Continued*)

T (K)	P (MPa)	x	ρ (kg · cm^{-3})	η (mPa · s)
323.15	160	0.384	0.783	0.305
323.15	195	0.383	0.800	0.348
373.15	50	0.416	0.639	0.153
373.15	80	0.415	0.676	0.182
373.15	120	0.414	0.713	0.215
373.15	160	0.413	0.739	0.259
373.15	195	0.411	0.757	0.297
298.15	50	0.204	0.632	0.149
298.15	80	0.204	0.663	0.175
298.15	120	0.204	0.694	0.204
298.15	160	0.203	0.717	0.236
298.15	195	0.201	0.734	0.258
323.15	50	0.186	0.580	0.122
323.15	80	0.187	0.616	0.145
323.15	120	0.188	0.649	0.172
323.15	160	0.188	0.675	0.198
323.15	195	0.188	0.694	0.226
373.15	50	0.192	0.528	0.103
373.15	80	0.195	0.573	0.123
373.15	120	0.196	0.614	0.147
373.15	160	0.196	0.643	0.173
373.15	195	0.196	0.663	0.198
298.15	50	0.102	0.530	0.117
298.15	80	0.102	0.565	0.140
298.15	120	0.101	0.597	0.168
298.15	160	0.101	0.622	0.192
298.15	195	0.101	0.640	0.213
323.15	50	0.103	0.507	0.097
323.15	80	0.102	0.545	0.111
323.15	120	0.102	0.576	0.134
323.15	160	0.101	0.603	0.159
323.15	195	0.101	0.622	0.184
373.15	50	0.103	0.451	0.080
373.15	80	0.101	0.501	0.096
373.15	120	0.100	0.542	0.114
373.15	160	0.100	0.571	0.133
373.15	195	0.099	0.592	0.153

where η is viscosity, η_0 is a preexponential parameter, E_{Vis} is the activation energy of viscous flow, R is the ideal-gas constant, and T is the absolute temperature.

Above a temperature of 370 K the branched alcohols do not form associates. The temperature dependence of the viscosity of substances, which do not form associates, follows the Andrade equation, Eq. (1). For this reason the viscosity of mixtures with hypothetical monomer alcohols can be approximated by extrapolating the viscosities at temperatures above 370 K to lower temperatures (dashed lines in Fig. 1). The effect of association on the viscosity is the difference between the measured data points and the dashed lines shown in Fig. 1.

It can be seen that the temperature dependence of the viscosity of pure ethene and mixtures with alcohols up to a mole fraction of 0.2 follows the law of Arrhenius. Above this mole fraction, significant amounts of associated alcohols exist, which leads to increasing deviations from Eq. (1). The pure alcohols show an increase in viscosity by association of more than 150% (difference between dashed and solid lines in Fig. 1). In

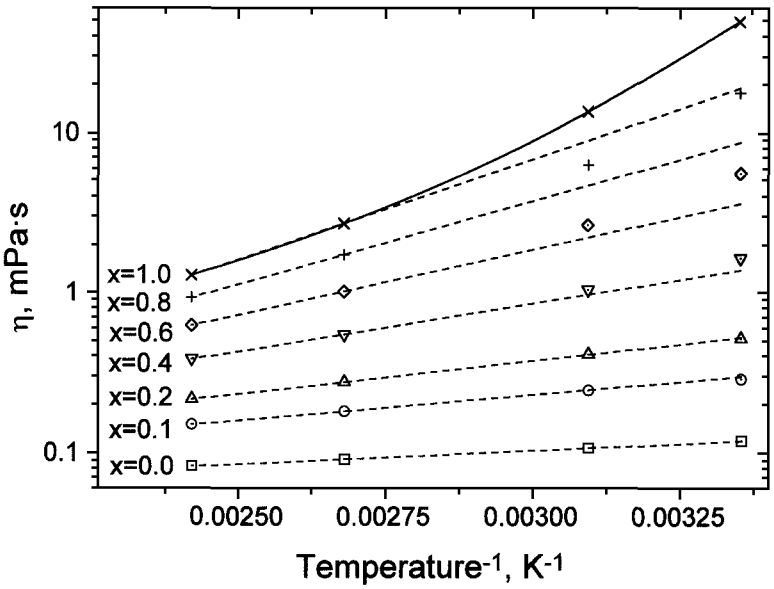

Fig. 1. The viscosities of 5-nonanol–ethene mixtures as a function of temperature at 120 MPa. x is the mole fraction of the alcohol. The dashed lines represent ideal behavior, if no alcohol-clusters are formed. They were obtained by extrapolating the viscosity data at the two highest temperatures (373 and 413 K) assuming an Arrhenius-like behavior. The solid line represents the actual course of viscosity of the pure 5-nonanol.

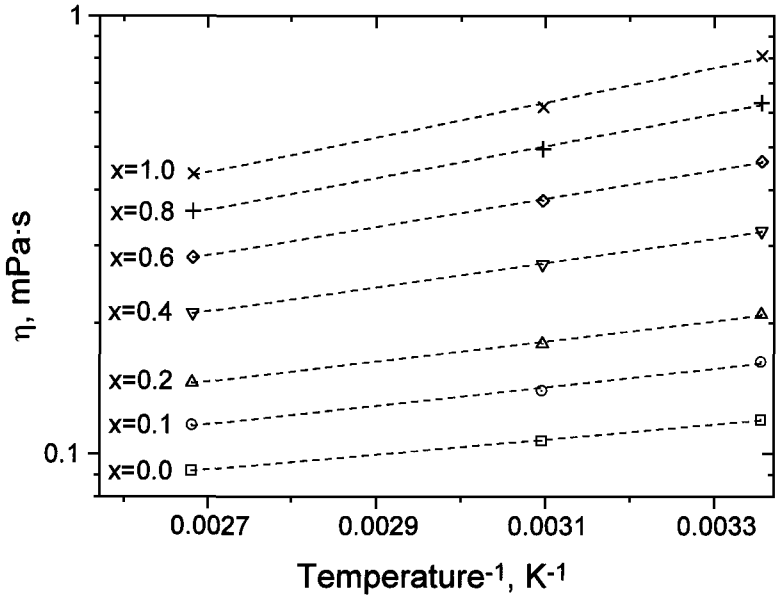

Fig. 2. The viscosities of vinyl acetate–ethene mixtures as a function of temperature at 120 MPa. x is the mole fraction of the alcohol. The dashed lines show the results of a linear regression of the logarithm of viscosity against the reciprocal temperature.

contrast to this behavior, vinyl acetate–ethene mixtures show a viscosity temperature dependence in agreement with the Arrhenius equation over the entire range of compositions (Fig. 2).

3.2. Pressure Dependence of the Viscosities

The viscosities of liquid alcohol–ethene mixtures at high densities increase with pressure, showing the viscosity–pressure behavior of a liquid. The pressure dependence of the viscosity of high-density substances can be approximately described by an equation analogous to Eq. (1) which defines an activation volume $\Delta V^{\#}_{\text{vis}}$

$$\Delta V^{\#}_{\text{vis}} = RT \left(\frac{\partial \ln \eta}{\partial P} \right)_T \tag{2}$$

where P is the pressure. Therefore, it is advantageous to plot the logarithm of viscosity against pressure. From this plot, one can see that the viscosity pressure dependence of alcohols shows the same behavior as substances which do not form hydrogen bonds. No influence of pressure on the extent

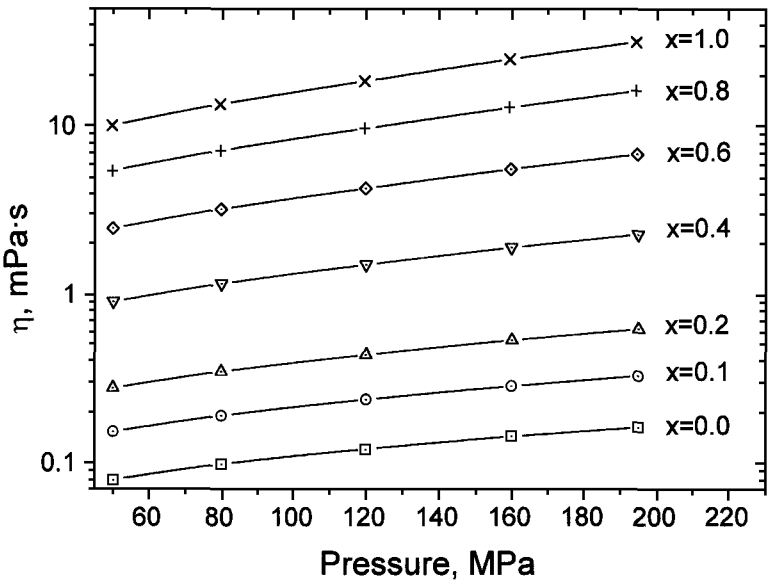

Fig. 3. The pressure dependence of the viscosity of 1-heptanol–ethene mixtures at 298 K. x is the mole fraction of the alcohol.

of hydrogen bonding has been observed [2, 3]. This is consistent with our observations for alcohol–ethene mixtures. As an example, the viscosities of 1-heptanol–ethene mixtures are presented in Fig. 3. The viscosities of ethene, pure alcohols, and their mixtures increase first more steeply with pressure and then less steeply.

4. THE VISCOSITY MODEL OF McALLISTER

The viscosities of mixtures are often calculated from the viscosities of the pure components. Numerous viscosity models for mixtures exist. One of the most widely used models is the model of McAllister and its variations [7–9] such as the Dizechi model. The McAllister model is based on Eyring's absolute rate model for viscosity [10] in which three- or four-body interactions are assumed and kinematic viscosities of the pure components, v_1 and v_2, are used for the correlation. The three- and four-body interaction models also need two or three adjustable parameters, respectively. These parameters are dependent on temperature and pressure and can be understood as kinematic viscosities at a given composition ($x = 0.33$ and $x = 0.66$ for the three-body model; $x = 0.25$, $x = 0.5$, and $x = 0.75$ for the four-body model). Instead of describing these parameters as a function

of temperature and pressure, they can also be given as a function of the viscosities of the components at the same pressure and temperature. The deviation from the ideal Kendall's behavior can be taken into consideration by a small variation of composition (Δx_{12}), which must be fitted to experimental viscosity data. For binary mixtures, McAllister developed the following expression from the three-body model:

$$\ln v = x_1^3 \ln v_1 + 3x_1^2 x_2 \ln v_{12} + 3x_1 x_2^2 \ln v_{21} + x_2^3 \ln v_2 - \ln\left[x_1 + x_2 \frac{M_2}{M_1}\right] + 3x_1^2 x_2 \ln\left[\frac{2 + M_2/M_1}{3}\right] + 3x_1 x_2^2 \ln\left[\frac{1 + 2M_2/M_1}{3}\right] + x_2^3 \ln\left[\frac{M_2}{M_1}\right] \tag{3}$$

in which x is the mole fraction and M is the molecular mass. Using the new method, we can take the parameters v_{12} and v_{21} from the following equations, which were derived from Kendall's equation [7, 11]:

$$\ln v_{12} = (\tfrac{2}{3} + \Delta x_{12}) \ln v_1 + (\tfrac{1}{3} - \Delta x_{12}) \ln v_2 \tag{4}$$

$$\ln v_{21} = (\tfrac{1}{3} + \Delta x_{21}) \ln v_1 + (\tfrac{2}{3} - \Delta x_{21}) \ln v_2 \tag{5}$$

where Δx_{12} and Δx_{21} are small variations in composition, which consider the deviation from Kendall's ideal behavior. For the four-body McAllister model of binary mixtures,

$$\ln v = x_1^4 \ln v_1 + 4x_1^3 x_2 \ln v_{1112} + 6x_1^2 x_2^2 \ln v_{1122} + 4x_1 x_2^3 \ln v_{2221} + x_2^4 \ln v_2 - \ln\left[x_1 + x_2 \frac{M_2}{M_1}\right] + 4x_1^3 x_2 \ln\left[\frac{3 + M_2/M_1}{4}\right] + 6x_1^2 x_2^2 \ln\left[\frac{1 + M_2/M_1}{2}\right] + 4x_1 x_2^3 \ln\left[\frac{1 + 3M_2/M_1}{4}\right] + x_2^4 \ln\left[\frac{M_2}{M_1}\right] \tag{6}$$

the parameters v_{1122}, v_{1112}, and v_{2221} can be defined in the same way.

$$\ln v_{1112} = (0.75 - \Delta x_{1112}) \ln v_1 + (0.25 + \Delta x_{1112}) \ln v_2 \tag{7}$$

$$\ln v_{1122} = (0.5 - \Delta x_{1122}) \ln v_1 + (0.5 + \Delta x_{1122}) \ln v_2 \tag{8}$$

$$\ln v_{2221} = (0.25 - \Delta x_{2221}) \ln v_1 + (0.75 + \Delta x_{2221}) \ln v_2 \tag{9}$$

The parameters of the three- and four-body McAllister model are listed in Table IX. Figure 4 shows the viscosity data for mixtures of a linear

Table IX. Parameters of the Three- and Four-Body McAllister Model [$T = 298.15$–373.15 K (413.15 K), $P = 50$–195 MPa]

	Three-body McAllister model			Four-body McAllister model			
Ethene mixture with	x_{12}	Δx_{21}	Average deviation (%)	Δx_{1112}	Δx_{1122}	Δx_{2221}	Average deviation (%)
1-Pentanol	−0.09728	−0.07595	2.4	−0.0808011	−0.0660377	−0.0568781	2.5
3-Pentanol	0.069182	4.733E-3	6.3	−0.0469881	0.202314	−0.064687	5.5
2-Methyl-2-butanol	0.14716	0.10099	7.2	0.0688228	0.195429	0.0500076	7.0
1-Heptanol	−0.16466	−0.14169	2.3	−0.10384	−0.176434	−0.0814957	2.1
3-Ethyl-3-pentanol	0.109868	−0.038987	9.5	0.0150506	0.151194	−0.100061	9.7
1-Nonanol	−0.10515	−2.59573	3.2	−0.0651849	−0.193907	−0.171298	3.0
5-Nonanol	−6.253E-3	−0.20195	10.2	−0.147024	0.128526	−0.24123	8.4
Vinyl acetate	−0.13541	−0.27383	2.2	−0.1005058	−0.1843159	−0.189488	2.1

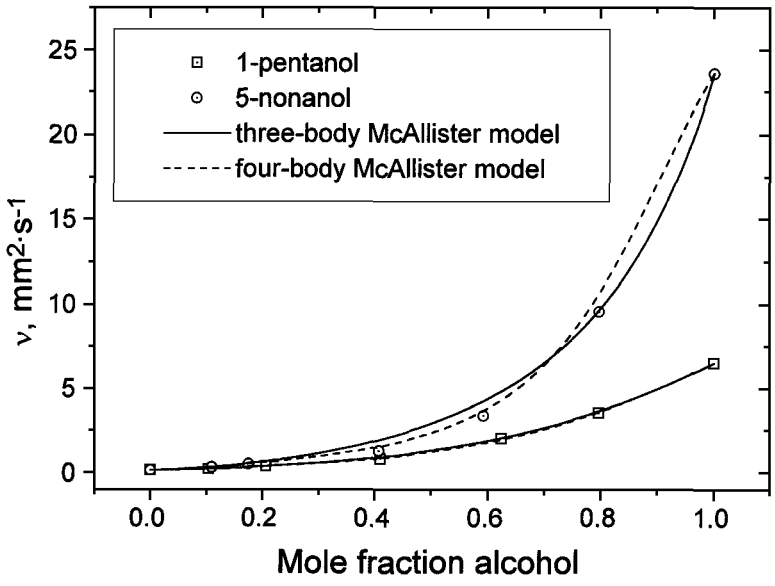

Fig. 4. Comparison of viscosity data of 1-pentanol–ethene and 5-nonanol–ethene mixtures with curves computed from the three- and four-body McAllister model at 298 K and 50 MPa.

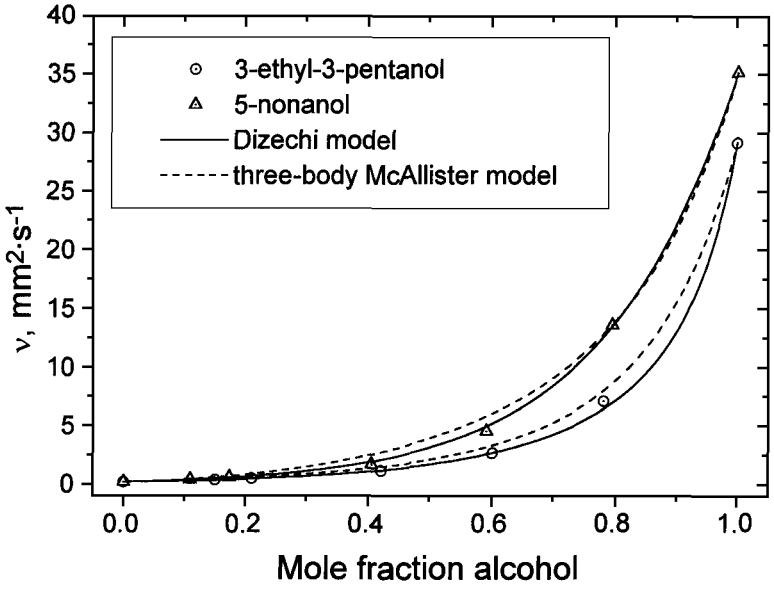

Fig. 5. Comparison of the three-body McAllister model and the Dizechi model for the viscosity of 5-nonanol–ethene and 3-ethyl-3-pentanol–ethene mixtures at 298 K and 80 MPa.

Table X. Parameters of the Dizechi Model
[$T = 298.15$–373.15 K (413.15 K), $P = 50$–195 MPa]

Ethene mixture with	Δx_{12}	C_{12}	Δx_{21}	C_{21}	Average deviation (%)
1-Pentanol	−0.08986	−293.88	−0.0612	−294.45	2.3
3-Pentanol	0.24188	−1116.2	0.28759	−438.31	3.0
2-Methyl-2-butanol	−0.01043	−286.84	0.03828	−263.32	3.9
1-Heptanol	−0.09823	−2461.8	0.10715	−1092.2	1.5
3-Ethyl-3-pentanol	−0.05615	−290.74	−1.314	−266.25	5.5
1-Nonanol	−0.09122	−567.43	−0.18995	−513.11	2.5
5-Nonanol	0.09637	−2454.1	0.25796	−577.5	4.3
Vinyl acetate	−0.23102	15.008	−0.48803	87.599	1.9

and a branched alcohol with ethene. The curves computed from the three- and four-body McAllister model are given as well.

It can be seen that both the three- and the four-body McAllister model describe the viscosity data of linear alcohol–ethene mixtures very well. In contrast, the viscosity data for branched alcohol–ethene mixtures cannot be described by either the three- or the four-body model with good accuracy. This is not surprising, because in the investigated range the temperature dependence of branched alcohols cannot be obtained by the Arrhenius-like Eyring equation, whereas for the temperature dependence of linear alcohols and ethene, there is approximate agreement with Arrhenius' law [2, 3]. Since Eyring's equation is the basis of the McAllister models, another model for describing the temperature dependence of the viscosity data for branched alcohols has to be inserted. One possible model is the model of Vogel [12], which leads to a variation of the three-body McAllister model, namely, the Dizechi model [8].

$$\ln \nu = \frac{T + C_1}{T + C_{av}} x_1^3 \ln(\nu_1 M_1) + \frac{T + C_2}{T + C_{av}} x_2^3 \ln(\nu_2 M_2) - \ln M_{av} + \frac{T + C_{12}}{T + C_{av}} x_1^2 x_2 \ln(\nu_{12} M_{12}) + \frac{T + C_{21}}{T + C_{av}} x_1 x_2^2 \ln(\nu_{21} M_{21}) \tag{10}$$

with

$$M_{av} = \sum x_i M_i, \qquad C_{av} = \sum x_i C_i$$

$$M_{ij} = \frac{(2M_i + M_j)}{3}, \qquad C_{ij} = \frac{(2C_i + C_j)}{3}$$

where C_1 and C_2 are adjustable parameters. The parameters v_{12} and v_{21} can be described in the same way as by the three-body McAllister model.

The parameters of the Dizechi model are listed in Table X. Figure 5 shows the measured viscosities of 5-nonanol–ethene and 3-ethyl-3-pentanol–ethene mixtures together with the curves computed using the Dizechi and the three-body McAllister model. It is obvious that the viscosities calculated from the Dizechi model fit the experimental data better than the values from the three-body McAllister model.

5. CONCLUSION

The viscosities and densities of several mixtures of alcohols and vinyl acetate with ethene were measured with a rolling-ball viscometer at high pressures. Whereas at a high mole fraction the temperature dependence of the viscosity is strongly influenced by the association, no influence of the alcohol associates on the viscosity of the mixtures can be observed below a mole fraction of about 0.2. A pressure effect on alcohol association cannot be evaluated from our viscosity data of alcohol–ethene mixtures. The viscosities of linear alcohol–ethene mixtures can be described by both the three- and the four-body McAllister model. The Dizechi model is a better model for describing the viscosity data of the ethene mixtures with branched alcohols. The parameters of both the McAllister model and the Dizechi model can be described by simple equations.

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