J. Chem. Eng. Data 1994, 39, 673–678

Viscosity of Mixtures of Liquid Hydrocarbons with Ethene in the Temperature Range 298–453 K at Pressures up to 200 MPa

Ulrich G. Krahn and Gerhard Luft*

Institut für Chemische Technologie, Technische Hochschule Darmstadt,
Petersenstrasse 20, 64287 Darmstadt, Germany

The viscosity of mixtures of 2,3,4-trimethylpentane, squalene, 2,2,4-trimethylpentane, 2,2,4,4,6,8,8-heptamethylnonane, polyisobutylene (M = 500 g/mol), ethylbenzene, and 1,2-diphenylethane with ethene at elevated pressures was determined with a rolling ball viscometer. Viscosity measurements were made at 298, 353, and 453 K and pressures up to 200 MPa with an accuracy of ±2%. The viscosities of the binary mixtures are described as a function of mass fraction and the viscosities of the pure compounds by an empirical equation. The results are compared with literature data of binary nonassociating systems.

Introduction

Experimental viscosities of ethene (*1*) and of pure hydrocarbons (*2*, *3*) at elevated pressures have been reported previously. This paper extends the investigation to binary mixtures of liquid hydrocarbons with ethene. There are many equations which are able to describe the concentration dependence of the viscosity of nonassociating binary systems, if the viscosities of the pure components are similar. Such models are poor when there are large differences between the viscosities of the pure components. The viscosity results reported here were correlated well by an empirical equation based on the mass fraction and the viscosities of the pure compounds. It is shown that the number of variables influencing the viscosity of binary mixtures of nonassociating compounds can be reduced by the presented method.

Experimental Section

The rolling ball viscometer used for this investigation has been described previously (*1*, *2*).

The liquid hydrocarbons were compressed and fed to the autoclave by a hand pump. At the beginning of a series of measurements the viscometer was calibrated with decane in the total range of temperature and pressure, using the viscosity values of decane published by Naake (*3*, *4*).

After condensing the ethene in a two-stage diaphragm–compressor, it was fed to two high-pressure vessels. The knowledge of the volume and of the pressure dependence of the density (*5*) made it possible to calculate the pressure difference corresponding to a mass of ethene, so that a known amount of ethene could be metered into the autoclave.

The temperature in the autoclave was measured radially and axially by two NiCr/Ni thermocouples with an accuracy of ±0.2 K. The pressure could be determined by a calibrated strain gauge pressure sensor with an accuracy of ±0.3%. The rolling times were detected inductively and then evaluated by the method of Hubbard and Brown (*6*). The resulting calibration curves only depended on temperature and pressure.

The scattering of the measured viscosities under constant conditions of temperature, pressure, and concentration was less than ±1%. The total uncertainty of the viscosity values was estimated to be ±2% (*1*, *2*), taking into account the accuracy of the literature values used in the calibration. The measurements of mass, volume, and pressure gave an uncertainty in mass fraction, which was dependent on concentration. The mass fraction (w) has an estimated accuracy between ±1% at w = 0.8 and ±3% at w = 0.2 (*2*).

Results and Discussion

The viscosity of the mixtures was determined as a function of temperature, pressure, and concentration. The hydrocarbons measured were 2,3,4-trimethylpentane, squalane, 2,2,4-trimethylpentane, 2,2,4,4,6,8,8-heptamethylnonane, polyisobutylene (M = 500g/mol), ethylbenzene, and 1,2-diphenylethane. The measurements were made at 298, 353, and 453 K and pressures up to 200 MPa with an estimated accuracy of ±2%. Approximately 90 measurements for each binary system were made. The viscosity of ethene is reported by Stanislawski and Luft (*1*). The values of the pure hydrocarbons are reported in refs 2 and 3. The experimental viscosity values of the mixtures of liquid hydrocarbons with ethene are presented in Table 1.

The viscosity of the binary mixtures of decane with methane (*4*, *7*) and decane with carbon dioxide (*8*) was taken from the literature.

The calculation of the empirical parameters of binary n-alkane mixtures was based on the literature values of hexane + dodecane at atmospheric pressure (*9*–*11*), as well as of mixtures of hexane and octane with dodecane (*12*), of hexane + hexadecane (*13*), and of decane + heptane and decane + hexadecane (*14*) under pressure. Also the data of binary mixtures of n-alkanes with components from octane to pentadecane at atmospheric pressure (*15*) were used.

The mixtures of n-alkanes with aromatic hydrocarbons with dodecane + benzene (*11*) and binary systems containing toluene or ethylbenzene and n-alkanes from octane to hexadecane (*16*, *17*).

The mass fraction dependence of the viscosity for the above systems could be fitted by the equation

$$\ln(\eta/\eta_1) = Aw_2 + \{\ln(\eta_2/\eta_1) - Aw_2\}w_2^{B} \quad \text{with} \quad \eta_2 > \eta_1 \tag{1}$$

where w_2 is the mass fraction of component 2 (the component with the higher pure-liquid viscosity), η is the viscosity of the binary mixture, and η_1 and η_2 are the viscosities of the pure components 1 and 2.

The empirical parameters A and B were correlated to the logarithm of the relative viscosity of the pure compo-

0021-9568/94/1739-0673$04.50/0

Table 1. Viscosity η of Mixtures of Liquid Hydrocarbons with Ethene as a Function of Mass Fraction of the Liquid Hydrocarbon w_2, Temperature, and Pressure

w_2	T/K	P/MPa	η/(mPa·s)	w_2	T/K	P/MPa	η/(mPa·s)	w_2	T/K	P/MPa	η/(mPa·s)
2,3,4-Trimethylpentane + Ethene											
0.2106	298.15	50.0	0.1158	0.7324	353.15	50.0	0.2697	0.8316	453.15	50.0	0.1701
0.2121	298.15	80.0	0.1411	0.7349	353.15	80.0	0.3313	0.8344	453.15	80.0	0.2215
0.2121	298.15	120.0	0.1717	0.7365	353.15	120.0	0.4264	0.8332	453.15	120.0	0.2837
0.2121	298.15	160.0	0.1940	0.7381	353.15	160.0	0.5326	0.8388	453.15	160.0	0.3377
0.2121	298.15	195.0	0.2229	0.7381	353.15	195.0	0.6326	0.8399	453.15	195.0	0.4026
0.2248	353.15	50.0	0.0894	0.7308	453.15	50.0	0.1357	0.9432	298.15	50.0	0.7265
0.2248	353.15	80.0	0.1092	0.7365	453.15	80.0	0.1739	0.9432	298.15	80.0	0.9377
0.2243	353.15	120.0	0.1342	0.7.97	453.15	120.0	0.2227	0.9435	298.15	120.0	1.1900
0.2243	353.15	160.0	0.1573	0.7428	453.15	160.0	0.2639	0.9435	298.15	160.0	1.4587
0.2240	353.15	195.0	0.1798	0.7436	453.15	195.0	0.3113	0.9398	298.15	195.0	1.7955
0.5198	453.15	50.0	0.0950	0.8355	298.15	50.0	0.50.0	0.9411	353.15	50.0	0.4620
0.5389	453.15	80.0	0.1224	0.8360	298.15	80.0	0.6487	0.9415	353.15	80.0	0.5856
0.5446	453.15	120.0	0.1565	0.8366	298.15	120.0	0.8321	0.9418	353.15	120.0	0.7653
0.5474	453.15	160.0	0.1851	0.8366	298.15	160.0	1.0390	0.9418	353.15	160.0	0.9766
0.5488	453.15	195.0	0.2157	0.8355	298.15	195.0	1.2525	0.9418	353.15	195.0	1.1991
0.7267	298.15	50.0	0.3764	0.8264	353.15	50.0	0.3285	0.9349	453.15	50.0	0.2349
0.7275	298.15	80.0	0.4801	0.8281	353.15	80.0	0.4092	0.9356	453.15	80.0	0.3024
0.7291	298.15	120.0	0.5967	0.8293	353.15	120.0	0.5321	0.9363	453.15	120.0	0.3960
0.7300	298.15	160.0	0.7521	0.8299	353.15	160.0	0.6739	0.9363	453.15	160.0	0.4671
0.7308	298.15	195.0	0.9220	0.8293	353.15	195.0	0.8387	0.9356	453.15	195.0	0.5623
2,2,4-Trimethylpentane + Ethene											
0.1856	298.15	50.0	0.1167	0.4720	453.15	120.0	0.1262	0.8628	353.15	120.0	0.4767
0.1832	298.15	80.0	0.1465	0.4737	453.15	160.0	0.1543	0.8613	353.15	160.0	0.5956
0.1838	298.15	120.0	0.1825	0.4737	453.15	195.0	0.1787	0.8598	353.15	195.0	0.7113
0.1823	298.15	160.0	0.2143	0.7291	298.15	50.0	0.2989	0.8588	453.15	50.0	0.1737
0.1838	298.15	195.0	0.2423	0.7300	298.15	80.0	0.3678	0.8603	453.15	80.0	0.2321
0.2234	353.15	50.0	0.0912	0.7308	298.15	120.0	0.4770	0.8613	453.15	120.0	0.2910
0.2203	353.15	80.0	0.1139	0.7316	298.15	160.0	0.5666	0.8623	453.15	160.0	0.3565
0.2172	353.15	120.0	0.1475	0.7316	298.15	195.0	0.6503	0.8618	453.15	195.0	0.4172
0.2178	353.15	160.0	0.1786	0.7267	353.15	50.0	0.2072	0.9475	298.15	50.0	0.6630
0.2178	353.15	195.0	0.2056	0.7291	353.15	80.0	0.2580	0.9462	298.15	80.0	0.8485
0.2005	453.15	120.0	0.856	0.7316	353.15	120.0	0.3332	0.9449	298.15	120.0	1.1511
0.1975	453.15	160.0	0.1028	0.7324	353.15	160.0	0.4115	0.9439	298.15	160.0	1.4476
0.1916	453.15	195.0	0.1200	0.7324	353.15	195.0	0.4784	0.9425	298.15	195.0	1.7699
0.4884	298.15	50.0	0.1609	0.7225	453.15	50.0	0.1268	0.9485	353.15	50.0	0.4092
0.4950	298.15	80.0	0.2030	0.7283	453.15	80.0	0.1638	0.9479	353.15	80.0	0.5195
0.4934	298.15	120.0	0.2574	0.7316	453.15	120.0	0.2151	0.9472	353.15	120.0	0.7013
0.4934	298.15	160.0	0.3005	0.7332	453.15	160.0	0.2636	0.9465	353.15	160.0	0.9075
0.4918	298.15	195.0	0.3386	0.7332	453.15	195.0	0.3060	0.9465	353.15	195.0	1.0999
0.4669	353.15	50.0	0.1166	0.8638	298.15	50.0	0.4428	0.9492	453.15	50.0	0.2242
0.4686	353.15	80.0	0.1447	0.8633	298.15	80.0	0.5569	0.9485	453.15	80.0	0.2870
0.4703	353.15	120.0	0.1843	0.8628	298.15	120.0	0.7297	0.9482	453.15	120.0	0.3749
0.4703	353.15	160.0	0.2243	0.8618	298.15	160.0	0.8863	0.9479	453.15	160.0	0.4633
0.4703	353.15	195.0	0.2582	0.8603	298.15	195.0	1.0463	0.9472	453.15	195.0	0.5525
0.4617	453.15	50.0	0.0849	0.8613	353.15	50.0	0.2909				
0.4686	453.15	80.0	0.0972	0.8623	353.15	80.0	0.3650				
2,2,4,4,6,8,8-Heptamethylnonane + Ethene											
0.2138	298.15	50.0	0.1397	0.6198	298.15	195.0	0.9992	0.9324	298.15	160.0	15.7210
0.2176	298.15	80.0	0.1724	0.6329	353.15	50.0	0.2737	0.9333	298.15	195.0	23.6874
0.2165	298.15	120.0	0.2149	0.6329	353.15	80.0	0.3542	0.9372	353.15	50.0	1.6729
0.2149	298.15	160.0	0.2608	0.6248	353.15	120.0	0.4551	0.9369	353.15	80.0	2.3790
0.2207	298.15	195.0	0.2805	0.6281	353.15	160.0	0.5868	0.9349	353.15	120.0	3.5006
0.2249	353.15	50.0	0.1244	0.6297	353.15	195.0	0.7157	0.9359	353.15	160.0	5.2006
0.2228	353.15	80.0	0.1335	0.6714	453.15	50.0	0.1652	0.9367	353.15	195.0	7.5243
0.2186	353.15	120.0	0.1730	0.6687	453.15	80.0	0.2212	0.9475	453.15	50.0	0.6397
0.2160	353.15	160.0	0.1925	0.6361	453.15	120.0	0.2652	0.9473	453.15	80.0	0.8677
0.2223	353.15	195.0	0.2327	0.6544	453.15	160.0	0.3465	0.9449	453.15	120.0	1.2036
0.1881	453.15	50.0	0.0647	0.6659	453.15	195.0	0.4383	0.9463	453.15	160.0	1.6014
0.1801	453.15	80.0	0.0826	0.8033	298.15	50.0	1.1241	0.9475	453.15	195.0	2.0839
0.1755	453.15	120.0	0.1043	0.8082	298.15	80.0	1.5147	0.9812	298.15	50.0	6.8145
0.1721	453.15	160.0	0.1253	0.8143	298.15	120.0	2.2232	0.9796	298.15	80.0	11.3945
0.2228	453.15	195.0	0.1447	0.8143	298.15	160.0	3.1544	0.9801	298.15	120.0	21.2017
0.3491	298.15	50.0	0.1796	0.8176	298.15	195.0	4.0932	0.9810	298.15	160.0	39.1449
0.3514	298.15	80.0	0.2318	0.8342	353.15	50.0	0.7074	0.9825	298.15	195.0	66.7288
0.3420	298.15	120.0	0.2821	0.8336	353.15	80.0	0.9528	0.9823	353.15	50.0	2.4507
0.3416	298.15	160.0	0.3344	0.8221	353.15	120.0	1.2757	0.9823	353.15	80.0	3.6099
0.3424	298.15	195.0	0.3741	0.8295	353.15	160.0	1.7444	0.9813	353.15	120.0	5.6804
0.3703	353.15	50.0	0.1512	0.8324	353.15	195.0	2.2748	0.9818	353.15	160.0	9.0695
0.3499	353.15	80.0	0.1608	0.8487	453.15	50.0	0.3445	0.9820	353.15	195.0	13.3070
0.3575	353.15	120.0	0.2104	0.8482	453.15	80.0	0.4577	0.9851	453.15	50.0	0.8290
0.3572	353.15	160.0	0.2594	0.8383	453.15	120.0	0.5928	0.9851	453.15	80.0	1.1389
0.3610	353.15	195.0	0.3447	0.8433	453.15	160.0	0.7853	0.9846	453.15	120.0	1.6230
0.6164	298.15	50.0	0.4067	0.8471	453.15	195.0	0.9826	0.9851	453.15	160.0	2.2432
0.6181	298.15	80.0	0.5194	0.9283	298.15	50.0	3.7157	0.9855	453.15	195.0	2.9525
0.6198	298.15	120.0	0.6785	0.9297	298.15	80.0	5.6227				
0.6214	298.15	160.0	0.8547	0.9311	298.15	120.0	9.4870				

Table 1 (Continued)

w_2	T/K	P/MPa	η/(mPa·s)	w_2	T/K	P/MPa	η/(mPa·s)	w_2	T/K	P/MPa	η/(mPa·s)
					Squalane + Ethene						
0.1968	298.15	50.0	0.1832	0.5958	453.15	120.0	0.3532	0.9549	353.15	120.0	12.1948
0.1968	298.15	80.0	0.2261	0.5952	453.15	160.0	0.4412	0.9558	353.15	160.0	18.0819
0.1948	298.15	120.0	0.2950	0.5919	453.15	195.0	0.5150	0.9564	353.15	195.0	26.1123
0.1948	298.15	160.0	0.3551	0.7861	298.15	50.0	2.5874	0.9583	453.15	50.0	1.4710
0.1958	298.15	195.0	0.3963	0.7850	298.15	80.0	3.5219	0.9583	453.15	80.0	1.9918
0.1988	353.15	50.0	0.1333	0.7850	298.15	120.0	5.0988	0.9583	453.15	120.0	2.6907
0.1968	353.15	80.0	0.1713	0.7861	298.15	160.0	7.1249	0.9575	453.15	160.0	3.4845
0.1968	353.15	120.0	0.2199	0.7850	298.15	195.0	9.2338	0.9583	453.15	195.0	4.4308
0.1978	353.15	160.0	0.2661	0.7871	353.15	50.0	1.1882	0.9711	298.15	50.0	40.6662
0.1968	353.15	195.0	0.3150	0.7871	353.15	80.0	1.5482	0.9695	298.15	80.0	73.7114
0.1773	453.15	120.0	0.1132	0.7882	353.15	120.0	2.1436	0.9701	298.15	120.0	134.1358
0.1752	453.15	160.0	0.1380	0.7882	353.15	160.0	2.8644	0.9705	298.15	160.0	241.8542
0.1752	453.15	195.0	0.1583	0.7861	353.15	195.0	3.6633	0.9709	298.15	195.0	406.4055
0.3979	298.15	50.0	0.3480	0.8012	453.15	50.0	0.5092	0.9726	353.15	50.0	7.0615
0.3973	298.15	80.0	0.4404	0.8012	453.15	80.0	0.6755	0.9725	353.15	80.0	10.3977
0.4032	298.15	120.0	0.6030	0.7882	453.15	120.0	0.8329	0.9713	353.15	120.0	16.4367
0.3985	298.15	160.0	0.7243	0.7963	453.15	160.0	1.0960	0.9718	353.15	160.0	25.3361
0.3961	298.15	195.0	0.8364	0.8022	453.15	195.0	1.3343	0.9722	353.15	195.0	36.4977
0.3894	353.15	50.0	0.2249	0.9105	298.15	50.0	11.7958	0.9736	453.15	50.0	1.6212
0.3985	353.15	80.0	0.3066	0.9042	298.15	80.0	17.8932	0.9736	453.15	80.0	2.2045
0.3979	353.15	120.0	0.4040	0.9063	298.15	120.0	29.9066	0.9736	453.15	120.0	0.328
0.3888	353.15	160.0	0.4998	0.9084	298.15	160.0	48.8899	0.9731	453.15	160.0	4.0208
0.3882	353.15	195.0	0.6091	0.9098	298.15	195.0	72.7941	0.9735	453.15	195.0	5.1279
0.4020	453.15	50.0	0.1028	0.9141	353.15	50.0	3.4971	0.9898	298.15	50.0	61.1989
0.4020	453.15	80.0	0.1434	0.9112	353.15	80.0	4.6848	0.9893	298.15	80.0	102.9056
0.4014	453.15	120.0	0.1877	0.9132	353.15	120.0	6.9986	0.9895	298.15	120.0	205.8783
0.4008	453.15	160.0	0.2304	0.9125	353.15	160.0	10.2677	0.9897	298.15	160.0	388.5530
0.3997	453.15	195.0	0.2667	0.9135	353.15	195.0	14.0371	0.9898	298.15	195.0	658.9055
0.5919	298.15	50.0	0.7232	0.9186	453.15	50.0	1.1032	0.9902	353.15	50.0	8.9005
0.5916	298.15	80.0	0.9196	0.9186	453.15	80.0	1.4722	0.9903	353.15	80.0	13.4929
0.5967	298.15	120.0	1.3216	0.9145	453.15	120.0	1.9169	0.9899	353.15	120.0	21.6895
0.5913	298.15	160.0	1.6248	0.9170	453.15	160.0	2.5273	0.9901	353.15	160.0	33.9146
0.5895	298.15	195.0	1.9073	0.9186	453.15	195.0	3.1530	0.9903	353.15	195.0	50.0966
0.5979	353.15	50.0	0.4292	0.9543	298.15	50.0	24.7428	0.9906	453.15	50.0	1.8434
0.5923	353.15	80.0	0.5476	0.9521	298.15	80.0	38.6061	0.9906	453.15	80.0	2.5221
0.5979	353.15	120.0	0.7560	0.9531	298.15	120.0	72.0723	0.9902	453.15	120.0	3.5087
0.5923	353.15	160.0	0.9417	0.9538	298.15	160.0	129.0831	0.9904	453.15	160.0	4.7355
0.5961	353.15	195.0	1.1623	0.9543	298.15	195.0	205.0252	0.9906	453.15	195.0	6.0780
0.5967	453.15	50.0	0.1959	0.9564	353.15	50.0	5.5481				
0.5961	453.15	80.0	0.2699	0.9566	353.15	80.0	7.9115				
					Polyisobutylene (M = 500 g/mol) + Ethene						
0.2101	298.15	50.0	0.2007	0.6019	353.15	120.0	0.9144	0.9250	353.15	160.0	65.8792
0.2101	298.15	80.0	0.2118	0.6231	353.15	160.0	1.1959	0.9241	353.15	195.0	113.9747
0.2101	298.15	120.0	0.2565	0.6231	353.15	195.0	1.3945	0.9261	453.15	50.0	2.2446
0.2112	298.15	160.0	0.2896	0.6059	453.15	120.0	0.4374	0.9259	453.15	80.0	3.2436
0.2089	298.15	195.0	0.3309	0.6059	453.15	160.0	0.5407	0.9259	453.15	120.0	4.9435
0.2345	453.15	50.0	0.1166	0.6059	453.15	195.0	0.6297	0.9253	453.15	160.0	7.3282
0.2345	453.15	80.0	0.1799	0.8396	298.15	50.0	9.1044	0.9244	453.15	195.0	9.9405
0.2345	453.15	120.0	0.1767	0.8388	298.15	80.0	15.1441	0.9665	353.15	50.0	36.1290
0.2345	453.15	160.0	0.2131	0.8426	298.15	120.0	31.5380	0.9662	353.15	80.0	67.3917
0.3848	353.15	50.0	0.2499	0.8357	298.15	160.0	47.0396	0.9659	353.15	120.0	152.0095
0.3912	353.15	80.0	0.3372	0.8349	353.15	50.0	2.6309	0.9649	353.15	160.0	314.6278
0.3982	353.15	120.0	0.4308	0.8349	353.15	80.0	3.7943	0.9657	353.15	195.0	610.2154
0.4092	353.15	160.0	0.5457	0.8349	353.15	120.0	6.0362	0.9661	453.15	50.0	4.1290
0.4065	353.15	195.0	0.6704	0.8349	353.15	160.0	9.3434	0.9659	453.15	80.0	6.2575
0.4318	453.15	50.0	0.1407	0.8341	353.15	195.0	13.5578	0.9658	453.15	120.0	10.0775
0.4318	453.15	80.0	0.2307	0.8357	453.15	50.0	0.8730	0.9655	453.15	160.0	15.9109
0.4318	453.15	120.0	0.3224	0.8357	453.15	80.0	1.2037	0.9651	453.15	195.0	23.3309
0.4318	453.15	160.0	0.3995	0.8357	453.15	120.0	1.7021	0.9873	353.15	50.0	78.2999
0.4318	453.15	195.0	0.4460	0.8357	453.15	160.0	2.2657	0.9870	353.15	80.0	171.7092
0.6151	298.15	50.0	0.7728	0.8349	453.15	195.0	2.8445	0.9866	353.15	120.0	440.2056
0.6142	298.15	80.0	1.0487	0.9253	298.15	50.0	74.1514	0.9858	353.15	160.0	990.0068
0.6129	298.15	120.0	1.4159	0.9247	298.15	80.0	152.8098	0.9873	453.15	50.0	5.9095
0.6114	298.15	160.0	1.7762	0.9238	298.15	120.0	397.2708	0.9872	453.15	80.0	9.0743
0.6065	298.15	195.0	2.2110	0.9259	353.15	50.0	11.4582	0.9870	453.15	120.0	15.6051
0.6231	353.15	50.0	0.6400	0.9256	353.15	80.0	18.9814	0.9866	453.15	160.0	26.3388
0.6231	353.15	80.0	0.8574	0.9256	353.15	120.0	35.3967	0.9858	453.15	195.0	39.8289
					Ethylbenzene + Ethene						
0.3075	298.15	50.0	0.1297	0.3151	353.15	120.0	0.1382	0.4756	298.15	195.0	0.3292
0.3029	298.15	80.0	0.1513	0.3093	353.15	160.0	0.1595	0.4965	353.15	50.0	0.1340
0.2981	298.15	120.0	0.1820	0.3050	353.15	195.0	0.1833	0.4902	353.15	80.0	0.1580
0.2949	298.15	160.0	0.2016	0.4874	298.15	50.0	0.1874	0.4860	353.15	120.0	0.1880
0.2921	298.15	195.0	0.2266	0.4843	298.15	80.0	0.2182	0.4819	353.15	160.0	0.2240
0.3331	353.15	50.0	0.1041	0.4813	298.15	120.0	0.2600	0.4783	353.15	195.0	0.2580
0.3231	353.15	80.0	0.1200	0.4786	298.15	160.0	0.2930	0.7235	298.15	50.0	0.2733

Table 1 (Continued)

w_2	T/K	P/MPa	η/(mPa·s)	w_2	T/K	P/MPa	η/(mPa·s)	w_2	T/K	P/MPa	η/(mPa·s)
				Ethylbenzene + Ethene							
0.7218	298.15	80.0	0.3298	0.8555	298.15	80.0	0.5450	0.9434	298.15	80.0	0.8900
0.7197	298.15	120.0	0.4205	0.8546	298.15	120.0	0.6700	0.9427	298.15	120.0	1.1090
0.7176	298.15	160.0	0.5272	0.8535	298.15	160.0	0.8730	0.9418	298.15	160.0	1.3250
0.7144	298.15	195.0	0.6982	0.8517	298.15	195.0	1.0259	0.9398	298.15	195.0	1.5140
0.7318	353.15	50.0	0.1918	0.8545	353.15	50.0	0.2772	0.9409	353.15	50.0	0.3925
0.7294	353.15	80.0	0.2196	0.8537	353.15	80.0	0.3382	0.9406	353.15	80.0	0.4635
0.7271	353.15	120.0	0.2780	0.8529	353.15	120.0	0.4099	0.9402	353.15	120.0	0.5900
0.7249	353.15	160.0	0.3368	0.8518	353.15	160.0	0.4953	0.9395	353.15	160.0	0.7108
0.7224	353.15	195.0	0.3954	0.8509	353.15	195.0	0.6551	0.9384	353.15	195.0	0.8580
0.7339	453.15	50.0	0.1210	0.8540	453.15	50.0	0.1607	0.9408	453.15	50.0	0.2252
0.7265	453.15	80.0	0.1440	0.8529	453.15	80.0	0.1946	0.9405	453.15	80.0	0.2746
0.7211	453.15	120.0	0.1705	0.8516	453.15	120.0	0.2313	0.9400	453.15	120.0	0.3333
0.7159	453.15	160.0	0.1959	0.8503	453.15	160.0	0.2716	0.9393	453.15	160.0	0.3944
0.7121	453.15	195.0	0.2381	0.8485	453.15	195.0	0.3162	0.9382	453.15	195.0	0.4285
0.8561	298.15	50.0	0.4375	0.9438	298.15	50.0	0.7355				
				1,2-Diphenylethane + Ethene							
0.3001	353.15	50.0	0.1166	0.6275	353.15	50.0	0.2534	0.8288	453.15	195.0	0.5997
0.2946	353.15	80.0	0.1406	0.6255	353.15	80.0	0.3060	0.9127	353.15	50.0	0.9602
0.2902	353.15	120.0	0.1680	0.6599	353.15	120.0	0.3838	0.9116	353.15	80.0	1.2040
0.2880	353.15	160.0	0.1981	0.6211	353.15	160.0	0.4728	0.9098	353.15	120.0	1.5824
0.2827	353.15	195.0	0.2304	0.6160	353.15	195.0	0.5633	0.9050	353.15	160.0	2.1057
0.2924	453.15	80.0	0.0946	0.6425	453.15	50.0	0.1537	0.9057	453.15	50.0	0.4186
0.2831	453.15	120.0	0.1177	0.6351	453.15	80.0	0.1867	0.9053	453.15	80.0	0.5097
0.2774	453.15	160.0	0.1358	0.6289	453.15	120.0	0.2341	0.9048	453.15	120.0	0.6336
0.2721	453.15	195.0	0.1523	0.6255	453.15	160.0	0.2848	0.9038	453.15	160.0	0.7633
0.4367	353.15	50.0	0.1584	0.6208	453.15	195.0	0.3340	0.9016	453.15	195.0	0.9093
0.4333	353.15	80.0	0.1891	0.8334	353.15	50.0	0.5531	0.9676	353.15	50.0	1.6718
0.4304	353.15	120.0	0.2304	0.8325	353.15	80.0	0.6710	0.9673	353.15	80.0	2.1196
0.4275	353.15	160.0	0.2782	0.8314	353.15	120.0	0.8559	0.9656	353.15	120.0	2.8663
0.4233	353.15	195.0	0.3387	0.8295	353.15	160.0	1.0784	0.9679	453.15	50.0	0.6028
0.4551	453.15	50.0	0.0982	0.8251	353.15	195.0	1.3409	0.9676	453.15	80.0	0.7504
0.4454	453.15	80.0	0.1215	0.8372	453.15	50.0	0.2741	0.9670	453.15	120.0	0.9516
0.4400	453.15	120.0	0.1502	0.8365	453.15	80.0	0.3367	0.9661	453.15	160.0	1.1559
0.4344	453.15	160.0	0.1792	0.8351	453.15	120.0	0.4199	0.9638	453.15	195.0	1.3762
0.4302	453.15	195.0	0.2088	0.8333	453.15	160.0	0.5075				

nents by polynomials. The parameter B also depends on mass fraction. The resulting equations are

$$A = A_1y + A_2y^2 + A_3y^3 \qquad (2)$$

$$B = B_1 + B_2w_2 + B_3y + B_4y^2 \qquad (3)$$

where $y = \ln(\eta_2/\eta_1)$.

The average deviation of experimental data was minimized by the simplex method of Nelder and Mead (*18*). The resulting values of the adjustable parameters are listed in Table 2. Some of the corresponding planes of the logarithm of relative mixture viscosities as a function of mass fraction and the logarithm of relative viscosity of the pure components are shown in Figures 1–3.

In all cases no additive temperature or pressure dependence of the mixture viscosities could be seen. Mixtures with similar components such as the aromatic hydrocarbons ethylbenzene and 1,2-diphenylethane and mixtures with components of the same homologous series like n-alkanes or oligomeric isobutenes (2,2,4-trimethylpentane, 2,2,4,4,6,8,8-heptamethylnonane, polyisobutylene (M = 500g/mol)) could be described by the same set of empirical parameters.

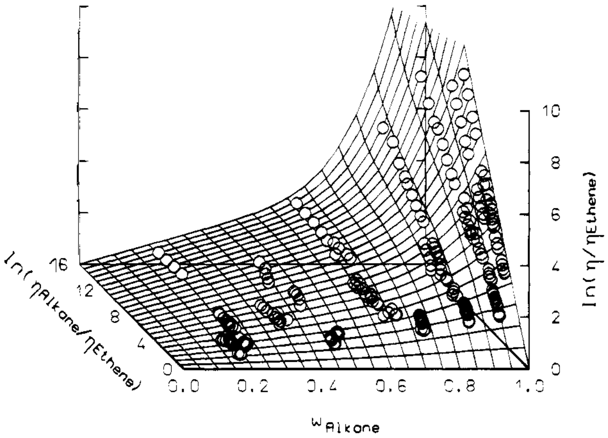

Figure 1. Relative viscosity of mixtures of oligomeric isobutenes (2,2,4-trimethylpentane, 2,2,4,4,6,8,8-heptamethylnonane, polyisobutylene (M = 500 g/mol)) with ethene as a function of mass fraction and of the viscosity of the pure components.

The viscosity increases monotonically with increasing relative viscosity of the pure components and with mass fraction of the pure liquid component having the higher viscosity. The logarithmic plot of the relative viscosity of the binary mixtures shows a linear concentration dependence in the range from $w_2 = 0$ to $w_2 = 0.6$. At higher values of w_2 the slope increases with increasing mass fraction. But the slope of the straight lines and the increase with the relative viscosity of the pure components depend on the type of the binary system. Both increase starting from mixtures of oligomeric isobutenes with ethene, to squalane + ethene, to binary mixtures of n-alkanes and of decane + methane, up to decane + carbon dioxide, which show an almost linear dependence over the whole range of concentration. So, as the relative viscosity of the pure components increases, progressively pronounced differences can be observed between the systems of binary mixtures.

The viscosity values of 2,3,4-trimethylpentane + ethene are similar to those of binary mixtures of n-alkanes and can be described by the empirical parameters of n-alkane mixtures (δ = 0.048).

The plane of mixtures of oligomeric isobutenes with ethene (Figure 1) is frequently observed in binary mixtures

Table 2. Parameters in the Polynomials 2 and 3 of Eq 1 of the Viscosity of Binary Mixtures

mixture	A_1	A_2	A_3
oligomeric isobutenes + ethene	0.7177	−0.0407	0.000 66
2,3,4-trimethylpentane + ethene	−0.2888	0.3237	−0.005 20
squalane + ethene	0.3668	0.1002	−0.010 39
aromatic hydrocarbons + ethene	0.5229	−0.1338	0.037 80
decane + methane	1.4804	−0.4266	0.065 67
decane + carbon dioxide	0.9102	−0.0469	0.026 14
n-alkanes + *n*-alkanes	0.6960	0.1816	−0.083 53
n-alkanes + aromatic hydrocarbons	−0.7538	0.2866	0.007 00

mixture	B_1	B_2	B_3	B_4
oligomeric isobutenes + ethene	5.352	−2.967	1.243	−0.0622
2,3,4-trimethylpentane + ethene	12.622	1.040	−11.324	2.8366
squalene + ethene	−9.506	0.780	5.420	−0.4086
aromatic hydrocarbons + ethene	7.683	2.918	−6.013	1.4090
decane + methane	8.209	1.911	1.202	−0.0018
decane + carbon dioxide	−77.991	10.872	38.135	0.0307
n-alkanes + *n*-alkanes	1.210	2.381	2.398	−0.7908
n-alkanes + aromatic hydrocarbons	1.404	0.248	−0.836	0.3701

mixture	δ^a	y_{min}	y_{max}	lit.
oligomeric isobutenes + ethene	0.065	0.0	16.0	2
2,3,4-trimethylpentane + ethene	0.031	1.6	2.8	2
squalane + ethene	0.055	3.0	9.0	2
aromatic hydrocarbons + ethene	0.045	1.5	4.0	2
decane + methane	0.035	2.0	4.5	4, 7
decane + carbon dioxide	0.030	2.2	3.2	8
n-alkanes + *n*-alkanes	0.010	0.0	3.0	9–15
n-alkanes + aromatic hydrocarbons	0.012	0.0	2.0	11, 16, 17

[a] δ = average absolute deviation of experimental values of $\ln(\eta/\eta_1)$. The experimental values are in the range from $\ln(\eta_2/\eta_1) = y_{min}$ to $\ln(\eta_2/\eta_1) = y_{max}$.

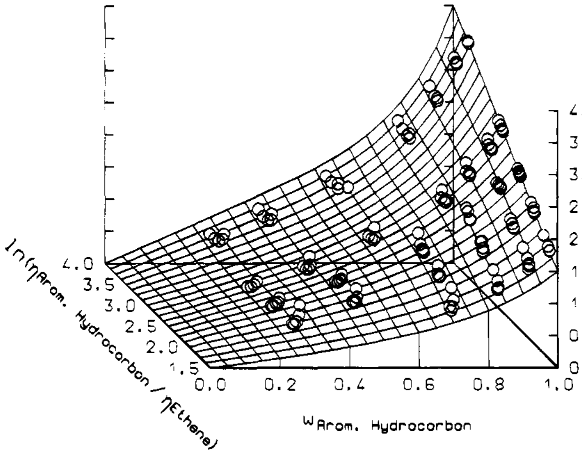

Figure 2. Relative viscosity of mixtures of aromatic hydrocarbons (ethylbenzene 1,2-diphenylethane) with ethene.

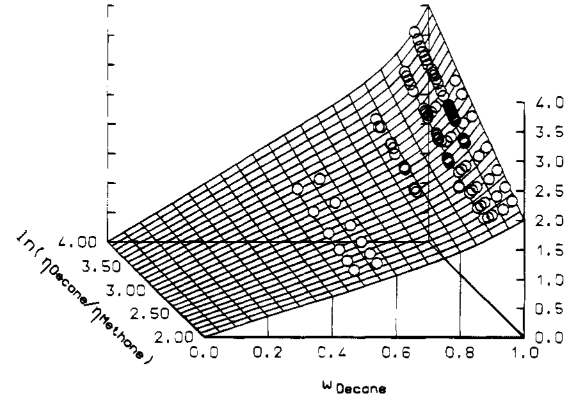

Figure 3. Relative viscosity of decane + methane.

of nonassociating compounds. Examples of binary systems which can be described by the empirical parameters of mixtures of oligomeric isobutenes with ethene (Table 2) are

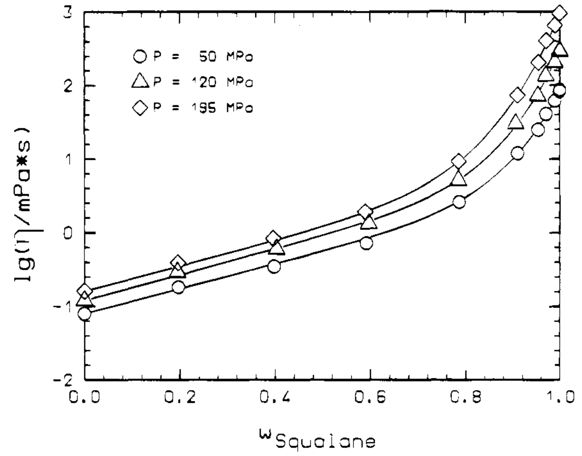

Figure 4. Logarithmic plot of the viscosity of squalane + ethene as a function of mass fraction at 298.15 K.

the mixtures of aromatic hydrocarbons with ethene (δ = 0.065), squalane + ethane (*19*) (δ = 0.053), mixtures of cyclohexane with hexane (*20*) (δ = 0.023) and with dodecane (*11*) (δ = 0.011), and mixtures of *n*-alkanes with aromatic hydrocarbons (δ = 0.030).

The viscosity of pure components can be described as a function of pressure and temperature by empirical equations (*1–3*). These equations were combined with eq 1. Using the resulting equation and the parameters of Table 2, it is possible to describe the viscosity of the binary systems as a function of pressure, temperature, and mass fraction.

Figures 4 and 5 show some of the calculated curves together with experimental viscosities measured by our group (*1–3*). Under constant conditions of temperature and pressure the logarithm of the viscosity increases with increasing mass fraction of the hydrocarbon (Figure 4), and the isobars are almost parallel at constant temperature.

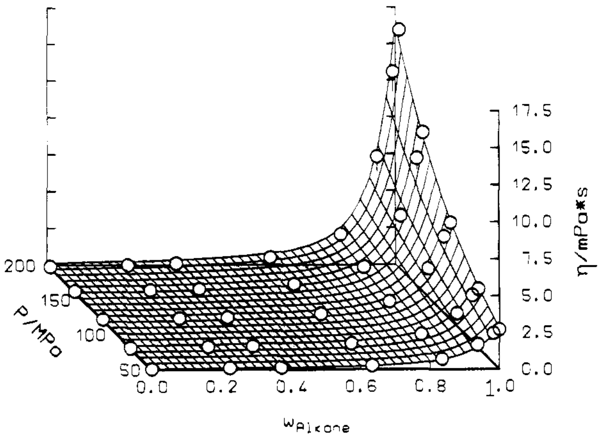

Figure 5. Viscosity of 2,2,4,4,6,8,8-heptamethylnonane + ethene as a function of mass fraction and of pressure at 353.15 K.

Figure 5 illustrates the typical concentration and pressure dependence of the viscosity of hydrocarbon + ethene.

The measured viscosities were also fitted by equations which describe the mixture viscosity as a function of mole fraction (*2*), like the equation of Grunberg and Nissan (*12*, *13*) or the models of McAllister (*21*). The best results were obtained with the four-body model of McAllister (*21*) and with the equation of Soliman (*22*). In both cases the temperature and pressure dependence of the adjustable parameters could be described by quadratic equations of $\ln(\nu_1\nu_2)$, where ν_1 and ν_2 are the kinematic viscosities of the pure components. The number of adjustable parameters was therefore nine for the modified McAllister model and six for the modified equation of Soliman. The deviation of measured viscosities from data calculated by these equations is substantially higher than the deviation from eq 1, which is based on mass fraction.

Literature Cited

(1) Stanislawski, U.; Luft, G. *Ber. Bunsen-Ges. Phys. Chem.* **1987**, *91*, 756.
(2) Krahn, U. G. Dissertation, TH Darmstadt, Darmstadt, Germany, 1993.
(3) Krahn, U. G.; Luft, G. *J. Chem. Eng. Data*, preceding paper in this issue.
(4) Naake, L. D.; Dissertation, University of Karlsruhe, Karlsruhe, Germany, 1984.
(5) Angus, S.; Armstrong, B.; de Reuck, K. M. *International Thermodynamic Tables of the Fluid State, Ethylene 1972*; Butterworths: London, 1974.
(6) Hubbard, R. M.; Brown, G. G. *Ind. Eng. Chem.* **1943**, *15*, 212.
(7) Lee, A. L.; Gonzalez, M. H.; Eakin, B. E. *J. Chem. Eng. Data* **1966**, *11*, 281.
(8) Cullick, A. S.; Mathis, M. L. *J. Chem. Eng. Data* **1984**, *29*, 393.
(9) Irving, J. B. Viscosities of Binary Liquid Mixtures: A Survey of Mixture Equations. NEL Report 630; Glasgow National Engineering Laboratory: East Kilbride, 1977.
(10) Reissmann, B.; Schuberth, H. *Z. Phys. Chem. (Leipzig)* **1977**, *258*, 983.
(11) Knapstad, B.; Skjølsvik, P. A.; Øye, H. A. *J. Chem. Eng. Data* **1991**, *36*, 84.
(12) Dymond, J. H.; Robertson, J.; Isdale, J. D. *Int. J. Thermophys.* **1981**, *2*, 133.
(13) Dymond, J. H.; Young, K. J.; Isdale, J. D. *Int. J. Thermophys.* **1980**, *1*, 345.
(14) Ducoulombier, D.; Zhou, H.; Boned, C.; Peyrelasse, J.; Saint-Guirons, H.; Xans, P. *J. Phys. Chem.* **1986**, *90*, 1692.
(15) Asfour, A.-F. A.; Cooper, E. F.; Wu, J.; Zahran, R. R. *Ind. Eng. Chem. Res.* **1991**, *30*, 1666.
(16) Asfour, A.-F. A.; Siddique, M. H.; Vavanellos, T. D. *J. Chem. Eng. Data* **1990**, *35*, 192.
(17) Asfour, A.-F. A.; Siddique, M. H.; Vavanellos, T. D. *J. Chem. Eng. Data* **1990**, *35*, 199.
(18) Nelder, J. A.; Mead, R. *Comput. J.* **1965**, *7*, 308.
(19) Kuss, E.; Killesreiter, H. BMFT-Forschungsbericht T 81−225; Bundesministerium für Forschung und Technologie: Bonn, 1981.
(20) Dymond, J. H.; Young, K. J.; Isdale, J. D. *J. Chem. Thermodyn.* **1979**, *11*, 887.
(21) McAllister, R. A. *AIChE J.* **1960**, *6*, 427.
(22) Soliman, K.; Marschall, E. *J. Chem. Eng. Data* **1990**, *35*, 375.

Received for review September 13, 1993. Revised May 13, 1994. Accepted June 13, 1994.®

® Abstract published in *Advance ACS Abstracts,* September 1, 1994.