these hydrocarbon systems. If one only considers the molecular packing of anisole and the methylanisole isomers, carbon dioxide would be more soluble in methylanisole isomers than in anisole since methylanisole isomers have more free volume and hence a lower cohesive energy density than those of anisole. On the other hand, if one considers only the entropic effect associated with the sizes of anisole and methylanisole isomers, the solubility of carbon dioxide would be more soluble in anisole than in methylanisole isomers because the size difference between the carbon dioxide and methylanisole isomers is greater than that between carbon dioxide and anisole molecules. It is believed that carbon dioxide is slightly more soluble in anisole than 2-methylanisole due to the competing effects of molecular structure and molecular size differences. Similar results are observed at 333 and 363 K.

Shown in Figure 6 is a comparison of vapor-phase compositions of aromatic hydrocarbons at 393 K. Since anisole has the highest vapor pressure, it has the highest concentration in the vapor phase. However, there is not much solubility difference among 2-methylanisole, 3-methylanisole, and 4-methylanisole, because they have similar vapor pressures.

Conclusions

The newly built equilibrium apparatus was tested by comparing the measured vapor-liquid equilibrium data for the system carbon dioxide-toluene at 393 K with those of Ng and Robinson (*1*). The comparison showed quite good agreement.

Phase equilibrium measurements were made on binary systems containing carbon dioxide as one of the components and anisole, 2-methylanisole, 3-methylanisole, or 4-methylanisole as the other component. When the solubilities of anisole, 2-methylanisole, 3-methylanisole, and 4-methylanisole in dense carbon dioxide at 393 K are compared, anisole is the most soluble and methylanisole isomers show little differences.

The comparison of liquid-phase compositions of carbon dioxide showed that the solubility of carbon dioxide is on the order of 4-methylanisole (most soluble) > 3-methylanisole > anisole > 2-methylanisole (least soluble).

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Viscosity of Three Binary Hydrocarbon Mixtures

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Viscosity measurements have been performed on three binary hydrocarbon mixtures at atmospheric pressure in the temperature range 15-70 °C. The binary mixtures reported are *n*-dodecane-*n*-hexane, *n*-dodecane-benzene, and *n*-dodecane-cyclohexane at *n*-dodecane mole fractions of 1.0, 0.75, 0.50, 0.25, and 0. The viscosities of the pure liquids are described by a modified Arrhenius equation and the mixtures by a modified Grundberg and Nissan equation with a standard deviation in fit of 0.4% or less. The accuracy is estimated to be 0.6% for the lowest viscosities reported, decreasing to 0.4% for the higher values reported. Literature data deviate by -2.4% to +3.0% from our results.

1. Introduction

A high-precision viscometer for studies of low-viscosity liquids has been utilized for studies of binary hydrocarbon mixtures. These measurements have been performed at atmospheric pressure. The present measurements of binary mixtures are part of a program on viscosity of oil reservoir related fluids, together with measurements of pure hydrocarbons, reported by Knapstad et al. (*1*) and Aasen et al. (*2*), and fluids pressurized up to 400 bar, reported by Knapstad et al. (*3*).

The aim of the study is to produce accurate viscosity data for pure hydrocarbons and their mixtures over a wide range in temperature and pressure for computer modeling of viscosity. The hydrocarbons and the pressure and temperature range in the study are chosen to be relevant to oil reservoir characterization and production, where viscosity is an important parameter.

2. Experimental Section

The oscillation viscometer employed measures the damping of a right cylinder in contact with the liquid under consideration. The cylinder may be a solid cylinder immersed in the liquid, or a hollow cup containing the liquid. In the present measurements the viscometer was equipped with a hollow cylinder that was completely filled with sample. Absolute viscosities are computed from the measured damping and the oscillating time of the cylinder. The working equations used are described by Brockner, Tørklep, and Øye (*4*).

The experimental setup to maintain a completely filled hollow cylinder during the measurements was described in a previous publication of viscosities of some pure hydrocarbons reported by Knapstad et al. (*1*). The hollow cylinder containing the sample has a capillary tube connected to its bottom part. In the opposite end the capillary is inserted into a sample reservoir at room temperature. The sample was thus sucked into the hollow cylinder when the temperature was decreased or transported from the hollow cylinder to the reservoir when the temperature was increased.

The temperature was controlled by means of a water bath circulator, and the temperature was measured with a calibrated Ni-NiCr thermocouple, as described by Knapstad et al. (*1*). The fully automated viscometer under control of a host computer

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Table I. Purity of Hydrocarbons

hydrocarbon	manufacturer	manufacturer claimed purity, %	GC analysis in this study, wt %: before possible distilln	GC analysis in this study, wt %: after distilln
n-hexane	E. Merck	>99	99.30[a]	99.40[a]
benzene	E. Merck	>99.5	100.00	
cyclohexane	Fluka AG	>99.5	99.98	
n-dodecane	Fluka AG	>98	99.66	100.00

[a] The impurity content of *n*-hexane is seen as a shoulder of the main *n*-hexane peak on the gas chromatogram. The shoulder is probably caused by an isomer of *n*-hexane, which may give a viscosity change of +0.09% to −0.04% at 20 °C, as measured by Eicher and Zwolinski (*7*).

has been thoroughly described in two previous papers by Tørklep and Øye (*5*) and by Berstad et al. (*6*).

The chemicals used are given in Table I. Gas chromatography analysis of the pure hydrocarbons as received from the manufacturer showed that *n*-dodecane and *n*-hexane had to be purified. The purification procedure and the chromatograph analysis employed were described by Knapstad et al. (*1*). The impurity of *n*-hexane (Table I) is probably an isomer of *n*-hexane, which may give a small change in viscosity as measured by Eicher and Zwolinsky (*7*).

The binary mixtures were prepared by weighing in a glass bottle closed with a stopper. Afterward, the mixture was stirred to give a homogeneous sample before it was transferred into the cup and sample reservoir, as described by Knapstad et al. (*1*).

3. Results

The viscosities of the binary mixtures of *n*-dodecane–*n*-hexane, *n*-dodecane–benzene, and *n*-dodecane–cyclohexane are reported. Measurements were performed with *n*-dodecane mole fractions of approximately 1, 0.75, 0.50, 0.25, and 0 at each mixture. All measurements were performed at atmospheric pressure in a temperature range from 15 to 70 °C.

The viscosities of pure *n*-hexane, benzene, and cyclohexane are reported by Knapstad et al. (*1*), while the viscosity of pure *n*-dodecane was remeasured. The remeasurements of *n*-dodecane were within 0.2% of the viscosities given by Knapstad et al. (*1*).

The densities used in the calculation of viscosity were taken from the literature [TRC—Thermodynamic Tables (*8*), Trevoy and Drickamer (*9*), Gomes-Ibanes and Liu (*10*, *11*), Schmidt and Randall (*12*), and Meeussen et al. (*13*)], and they were in good agreement with each other; see below. The densities tabulated in the literature were fitted by us using an empirical model equation. The *n*-dodecane–*n*-hexane system has contributions from refs 8, 11, and 13, and the standard deviation in fit is 0.01%. The *n*-dodecane–benzene system has contributions from refs 8, 9, and 12, and the standard deviation in fit is 0.02%. The *n*-dodecane–cyclohexane system has contributions from refs 8 and 10, and the standard deviation in fit is 0.09%. A relative change in density will give the same relative change in viscosity but with opposite sign (Table IV).

The experimental viscosities and computed densities are summarized in Table II, in chronological order by listing the average viscosity of four parallel runs.

The following modified Arrhenius equation was fitted to the experimental data of the pure hydrocarbons.

$$\eta = A \exp(B(1/T - 1/T_m) + C((1000/T - 1000/T_u)^2 - (1000/T_m - 1000/T_u)^2)) \quad (1)$$

T is the temperature (K) and T_m and T_u are the mean and upper temperatures of the measurement series introduced to improve orthogonality of the fitted parameters A, B, and C. A simpler equation is obtained by direct transformation of eq 1.

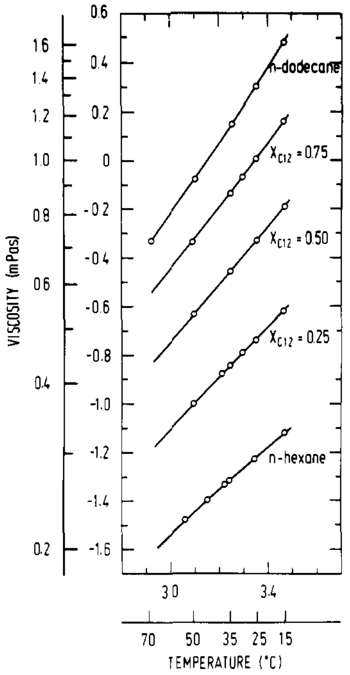

Figure 1. Arrhenius plot (ln η versus $1000/T$) of the mean experimental viscosities of *n*-dodecane–*n*-hexane mixtures.

$$\ln (\eta/(\text{mPa·s})) = a + b/T + c/T^2 \quad (2)$$

The transformed parameters a, b, and c are given in Table III. A modified Grundberg and Nissan equation was used to fit the experimental data of the mixtures.

$$\ln (\eta/(\text{mPa·s})) = X_1 \ln \eta_1 + X_2 \ln \eta_2 + X_1 X_2 G \quad (3)$$

where the characteristic parameter G is given as

$$G = K + L(T - T_m) + (2X_1 - 1)(M + N(T - T_m)) \quad (4)$$

which also is simplified by direct transformation to

$$G = \alpha + \beta T + (2X_1 - 1)(\gamma + \delta T) \quad (5)$$

The viscosities and mole fractions of the pure hydrocarbons are denoted by η and X, and the transformed parameters α, β, γ, and δ are given in Table III.

The parameters were fitted by a model parameter estimation program using the simplex method of Nelder and Mead as applied by Hertzberg (*14*). The entire experimental data set of 364 individual runs was used for the correlation.

Curves calculated from eqs 2, 3, and 5 for the three binary mixtures are shown in Figures 1–3 as Arrhenius plots (ln η versus $1000/T$), together with experimental data of Table II. Viscosities of the three binary mixtures at 25 °C are plotted versus *n*-dodecane mole fraction in Figure 4. Experimental viscosities are corrected to nominal temperature by piecemeal use of eqs 2, 3, and 5.

As seen from Table III, the standard deviation in fit was between 0.17% and 0.36%. An attempt to fit the two-parameter McAllistar (*15*) relation with a temperature coefficient (four parameters) gave a standard deviation of 6.3%.

4. Discussion

4.1. Error Analysis. Estimated standard deviations of relevant viscometer parameters and their influence on the damping viscosity are given in Table IV for a typical mea-

Table II. Experimental Viscosities[a]

temp, °C	*n*-dodecane mole fracn	density, kg/m^3	viscosity, mPa·s	std dev, %	temp, °C	*n*-dodecane mole fracn	density, kg/m^3	viscosity, mPa·s	std dev, %
				n-dodecane–*n*-hexane					
15.65	1	751.8	1.6149	0.05	15.55	0.2499	697.2	0.5381	0.04
25.07	1	745.2	1.3528	0.15	25.32	0.2499	689.8	0.4774	0.04
25.24	1	745.0	1.3503	0.03	30.43	0.2499	685.8	0.4512	0.07
34.37	1	738.5	1.1589	0.12	35.08	0.2499	682.2	0.4292	0.07
34.42	1	738.5	1.1590	0.03	38.27	0.2499	679.7	0.4144	0.02
49.45	1	727.7	0.9255	0.04	49.93	0.2499	670.4	0.3672	0.05
49.45	1	727.7	0.9257	0.06	15.47	0	663.3	0.3243	0.06
69.49	1	712.9	0.7157	0.08	25.63	0	654.1	0.2929	0.10
15.57	0.7481	738.2	1.1695	0.02	35.34	0	645.2	0.2671	0.07
25.32	0.7481	731.3	1.0016	0.03	36.97	0	643.7	0.2629	0.08
30.42	0.7481	727.7	0.9319	0.03	44.15	0	637.0	0.2465	0.08
34.97	0.7481	724.5	0.8716	0.02	53.39	0	628.3	0.2267	0.13
50.57	0.7481	713.1	0.7124	0.08	53.42	0	628.3	0.2268	0.09
50.57	0.7481	713.1	0.7130	0.18					
15.28	0.4998	720.8	0.8240	0.04					
25.12	0.4998	713.8	0.7168	0.07					
35.03	0.4998	706.5	0.6313	0.05					
49.53	0.4998	695.6	0.5301	0.09					
				n-dodecane–benzene					
15.25	0.7496	764.8	1.3052	0.04	15.44	0.2502	817.3	0.8194	0.05
15.54	0.7496	764.6	1.2973	0.04	25.37	0.2502	808.6	0.7103	0.06
25.61	0.7496	757.2	1.0965	0.04	35.36	0.2502	799.8	0.6242	0.03
25.62	0.7496	757.2	1.0959	0.04	49.79	0.2502	786.8	0.5262	0.08
25.64	0.7496	757.1	1.0950	0.04	67.55	0.2502	770.7	0.4343	0.09
35.75	0.7496	749.6	0.9482	0.03	15.40	0	883.8	0.6942	0.07
50.22	0.7496	738.7	0.7775	0.05	25.41	0	873.2	0.5984	0.10
68.84	0.7496	724.4	0.6228	0.08	25.41	0	873.2	0.5985	0.07
15.45	0.5002	784.4	1.0245	0.03	35.51	0	862.4	0.5220	0.19
25.23	0.5002	776.7	0.8885	0.06	39.57	0	858.1	0.4948	0.02
26.34	0.5002	775.8	0.8746	0.08	49.32	0	847.5	0.4402	0.16
34.93	0.5002	768.9	0.7750	0.06	59.72	0	836.2	0.3922	0.05
49.77	0.5002	757.0	0.6451	0.06					
66.95	0.5002	742.9	0.5289	0.09					
				n-dodecane–cyclohexane					
15.50	0.7489	755.2	1.4746	0.03	15.12	0.2501	767.4	1.1752	0.09
24.98	0.7489	748.3	1.2469	0.05	25.23	0.2501	759.0	0.9923	0.02
25.46	0.7489	747.9	1.2319	0.02	25.28	0.2501	759.0	0.9883	0.05
25.47	0.7489	747.9	1.2321	0.08	51.50	0.2501	736.8	0.6762	0.09
50.60	0.7489	729.1	0.8447	0.01	15.54	0	782.7	1.0568	0.05
16.00	0.5001	759.1	1.3101	0.03	15.55	0	782.7	1.0536	0.04
25.16	0.5001	752.1	1.1213	0.02	15.57	0	782.7	1.0540	0.04
25.20	0.5001	752.0	1.1163	0.05	25.08	0	773.8	0.8938	0.15
35.61	0.5001	743.9	0.9475	0.02	25.22	0	773.7	0.8899	0.06
35.71	0.5001	743.8	0.9480	0.02	25.26	0	773.6	0.8894	0.08
64.53	0.5001	720.9	0.6440	0.09	25.32	0	773.6	0.8889	0.03
64.54	0.5001	720.9	0.6442	0.03	25.34	0	773.6	0.8903	0.04
15.22	0.4996	759.8	1.3266	0.07	35.84	0	763.7	0.7494	0.03
24.63	0.4996	752.5	1.1307	0.09	35.92	0	763.6	0.7493	0.09
25.28	0.4996	752.0	1.1141	0.05	50.60	0	749.6	0.6033	0.05
51.05	0.4996	731.7	0.7657	0.07	60.68	0	739.9	0.5277	0.13
					60.70	0	739.9	0.5278	0.05

[a] Average values of four parallel runs.

surement. The reported viscosities were measured with the same viscometer as the determination of viscosity of pure hydrocarbons reported by Knapstad et al. (*1*). Hence, the error analysis and the estimated standard deviation of the viscometer parameters are equal, except for the error originated from the uncertainty in the sample composition.

The uncertainty in the sample composition can be divided into two error sources: one due to the accuracy of the weighing of the samples and one due to evaporation from the sample reservoir. The accuracy of the weighing of the mixture was estimated to be 0.01 g, which gives an uncertainty in mole fraction of 0.0003.

Due to different partial pressures of *n*-dodecane and the C-6 hydrocarbons, the sample reservoir will be enriched on *n*-dodecane with time. The viscosity was measured at 15 °C, both at the start and at the end of some measurement series, and no significant deviations were observed. However, the uncertainty in the composition was assumed to be 0.001, as given in the error table, Table IV.

The resulting total standard deviation of Table IV is 0.29% for the viscosity of a mixture of *n*-dodecane–benzene at 35.75 °C with a *n*-dodecane mole fraction of 0.7496. The total standard deviation can be computed to be within the range 0.25–0.41% for the mixtures reported.

The direct viscosity change due to the estimated uncertainty in measured temperature and mole fraction can be found by use of eqs 2, 3, and 5. The temperature change will give a relative deviation in viscosity of 0.2% and the mole fraction 0.1%. For comparison, the relative standard deviations in fit of the model equation for the mixtures (eq 3) are 0.17–0.36%.

A final accuracy of 0.4–0.6% is estimated for the reported results. The lowest viscosities have the highest uncertainty. The estimated uncertainties can be recognized to be at the 68% confidence level.

Table III. Parameters in Equations 2, 3, and 5 of the Viscosity of the Binary Mixtures[a]

parameter	*n*-dodecane–*n*-hexane	*n*-dodecane–benzene	*n*-dodecane–cyclohexane
a_1	−2.434 89	−2.434 89	−2.434 89
b_1, K	69.291	69.291	69.291
c_1, K^2	2.229×10^5	2.229×10^5	2.229×10^5
a_2	−5.336 44	−3.374 75	−4.159 91
b_2, K	1585.81	442.97	911.47
c_2, K^2	-1.07×10^5	1.228×10^5	8.807×10^4
α	0.1664	−1.3879	−0.7748
β, K^{-1}	1.165×10^{-3}	4.407×10^{-3}	2.817×10^{-3}
γ	−0.7072	0.6154	0.7089
δ, K^{-1}	2.019×10^{-3}	-1.474×10^{-3}	-2.109×10^{-3}
std dev in fit, %	0.17	0.36	0.21

[a] The indexes 1 and 2 denote *n*-dodecane and C-6 hydrocarbon, respectively.

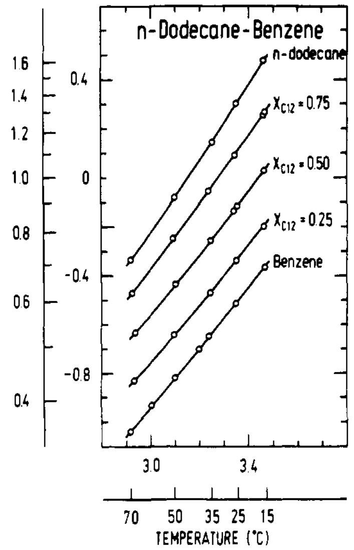

Figure 2. Arrhenius plot (ln η versus $1000/T$) of the mean experimental viscosities of *n*-dodecane–benzene mixtures.

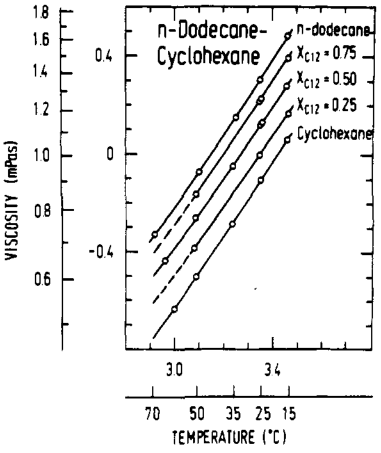

Figure 3. Arrhenius plot (ln η versus $1000/T$) of the mean experimental viscosities of *n*-dodecane–cyclohexane mixtures.

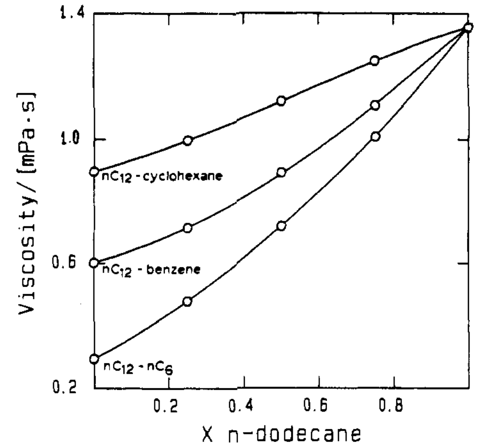

Figure 4. Plot of viscosities of the three binary mixtures at 25 °C versus *n*-dodecane mole fraction. Experimental viscosities are corrected to nominal temperature by piecemeal use of eqs 2, 3, and 5.

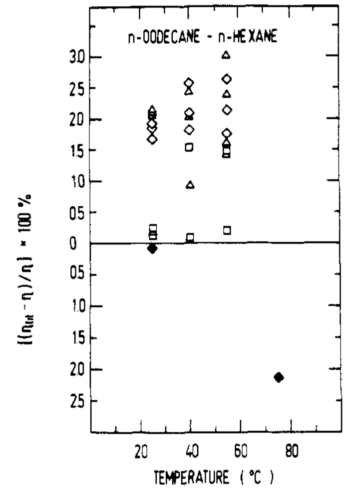

Figure 5. Comparison of literature viscosity data to our reported results of the binary mixtures of *n*-docane–*n*-hexane. Filled symbols represent viscosity data of Dymond et al. (*18*), and open symbols represent viscosity data of Meeussen et al. (*13*): (□) X_{C12} = 0.75 (◇, ◆) X_{C12} = 0.5, (△) X_{C12} = 0.25.

Table IV. Error Table[a]

exptl param	estd error	relative std dev (%)	relative viscosity change (%)
inertia of oscillating system	10^{-7} kg·m^2	0.11	0.22
net damping constant	7×10^{-6}	0.07	0.13
cup radius	2 μm	0.02	−0.11
cup height	20 μm	0.02	−0.04
density	0.3 kg/m^3	0.04	−0.04
temperature (indirect)	0.15 °C		−0.02
mole fraction *n*-C_{12} (indirect)	10^{-3}	0.13	−0.02
period gas	40 μs	0.002	0
period liquid	40 μs	0.002	0
total std dev $[(\sum(\Delta\eta)^2)^{1/2}]$			0.29

[a] All changes are positive. The table shows computer-calculated effects on damping viscosity of *n*-dodecane–benzene, X_{C12} = 0.7496 at 35.75 °C.

4.2. Comparison with Other Results. A comparison of the viscosities of pure hydrocarbons to those in literature was reported earlier by Knapstad et al. (*1*). Results published by Dymond et al. (*16*, *17*) usually fit our results within our estimated accuracy of the pure hydrocarbons. Viscosities reported by Meeussen et al. (*13*) of the binary mixture *n*-dodecane–*n*-hexane deviate from 0.1% to 3.0% of our correlated data. Mole fractions of *n*-dodecane above 0.7 were closest to our results. Two viscosity values at atmospheric pressure and at a *n*-dodecane mole fraction of 0.5 reported by Dymond et al. (*18*) are 0.1% and 2.1% lower than our results. A deviation plot of literature values compared to our reported values of the

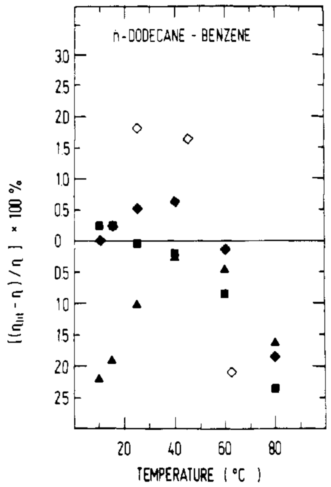

Figure 6. Comparison of literature viscosity data to our reported results of the binary mixtures of *n*-dodecane–benzene. Filled symbols represent viscosity data of Dymond and Young (*19*), and open symbols represent viscosity data of Trevoy and Drickamer (*9*): (■) $X_{C12} = 0.75$, (◇, ◆) $X_{C12} = 0.5$, (▲) $X_{C12} = 0.25$.

binary mixture *n*-dodecane–*n*-hexane is given in Figure 5.

Trevoy and Drickamer (*9*) have measured a binary mixture of *n*-dodecane–benzene with a mole fraction of *n*-dodecane of 0.5 at 20, 40, and 60 °C. Those viscosties are 1.8% and 1.6% higher and 2.1% lower, respectively, than our results. Mixtures of *n*-dodecane–benzene reported by Dymond and Young (*19*) fit our results within our estimated accuracy, except *n*-dodecane mole fractions of 0.25 below 30 °C and all viscosities above 70 °C, which are 1–2.4% lower than our reported results. A deviation plot of the literature values compared to our reported results of the binary mixtures of *n*-dodecane–benzene is given in Figure 6.

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Glossary

η	viscosity, mPa·s
A, B, C	model parameters in eq 1
a, b, c	model parameters in eq 2
T	temperature, K
T_m, T_u	mean and upper temperatures in a measurement series, K
X	mole fraction
G	characteristic parameter in the Grundberg and Nissan equation
K, L, M, N	model parameters in eq 4
α, β, γ, δ	model parameters in eq 5

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