Liquid Viscosity and Surface Tension of *n*-Hexane, *n*-Octane, *n*-Decane, and *n*-Hexadecane up to 573 K by Surface Light Scattering (SLS)

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ABSTRACT: In the present study, the simultaneous and accurate determination of liquid viscosity and surface tension of the *n*-alkanes *n*-hexane (*n*-C₆H₁₄), *n*-octane (*n*-C₈H₁₈), *n*-decane (*n*-C₁₀H₂₂), and *n*-hexadecane (*n*-C₁₆H₃₄) by surface light scattering (SLS) in thermodynamic equilibrium is demonstrated. Measurements have been performed over a wide temperature range from 283.15 K up to 473.15 K for *n*-C₆H₁₄, 523.15 K for *n*-C₈H₁₈, and 573.15 K for *n*-C₁₀H₂₂ as well as *n*-C₁₆H₃₄. The liquid dynamic viscosity and surface tension data with average total measurement uncertainties (*k* = 2) of (2.0 and 1.7) % agree with the available literature and contribute to a new database at high temperatures. Over the entire temperature range, a Vogel-type equation for the dynamic viscosity and a modified van der Waals equation for the surface tension represent the measured data for the four *n*-alkanes within experimental uncertainties. By also considering our former SLS data for *n*-dodecane (*n*-C₁₂H₂₆) and *n*-octacosane (*n*-C₂₈H₅₈), empirical models for the liquid viscosity and surface tension for the surface tension of temperature and carbon number covering values between 6 and 28. Agreement between these models and reference correlations for further selected *n*-alkanes which were not included in the development procedure was found.

INTRODUCTION

Linear alkanes with carbon numbers between 6 and 16 are important working fluids in many fields of process and energy engineering. Examples are the catalytic synthesis of *n*-alkanes with varying chain length during the Fischer-Tropsch process,¹ their subsequent separation by distillation,^{2,3} and utilization in combustion processes.⁴⁻⁶ For the design and modeling of these processes, accurate thermophysical properties of the involved chemicals including viscosity and surface tension are required, in particular at process-relevant conditions above 400 K. In this regard, standard databases for thermophysical properties of pure components and mixtures have been established, for example the Dortmund database⁷ and the ThermoData Engine (TDE) of the National Institute of Standards and Technology (NIST).⁸ For the subsequent development of reference correlations of fluids as they are implemented in, e.g., the NIST reference database REFPROP,⁹ primary experimental data with low measurement uncertainties are ideally required. Data related to this category should be obtained from quasi-primary methods based on a physically sound working equation that connects the experimentally measured parameters with the property of interest.¹⁰ For the viscosity and surface tension of n-alkanes with carbon numbers of 10 and larger, there is still a lack of reliable, direct experimental data in the open literature, especially at temperatures above 400 K.¹¹⁻¹⁴

A quasi-primary method for the determination of viscosity of liquids in the medium viscosity range¹⁵ is the surface light scattering (SLS) technique. It probes the dynamics of thermal fluctuations at phase boundaries in a contactless way via the temporal analysis of the scattered light intensity and is based on rigorous working equations according to the linearized hydrodynamic description of surface fluctuations.^{16,17} By taking necessary precautions against instrumental line broadening effects, the technique can be used without any need for a calibration procedure. For systems consisting of a vapor and a liquid phase as well as showing relatively low viscosities, which is true for the systems in this study, the quantities of the surface

fluctuations with a defined wavelength directly accessible by SLS are their characteristic dynamics, *i.e.*, damping and frequency. Besides the latter two properties, only information on the liquid and vapor densities as well as the vapor viscosity is necessary for a simultaneous determination of the liquid viscosity and surface tension in macroscopic thermodynamic equilibrium.¹⁸⁻²⁰

During the last two decades, research activities at the Department of Chemical and Biological Engineering (CBI) and at the Erlangen Graduate School in Advanced Optical Technologies (SAOT) of the Friedrich-Alexander-University Erlangen-Nürnberg (FAU) developed SLS as a reliable and accurate instrument in thermophysical property research of fluids covering broad ranges of viscosity and surface or interfacial tension. For various systems at vapor-liquid equilibrium, e.g., reference fluids (toluene,²¹ diisodecyl phthalate,²² carbon dioxide,¹⁸ and *n*-pentane²³), refrigerants,^{20,24,25} hydrofluoroethers,²⁶ ionic liquids,²⁷⁻³⁰ linear³¹ and branched³² alkanes as well as mixtures of *n*-alkanes with other *n*-alkanes^{31,33} or oxygenated derivatives,³³ viscosity and surface or interfacial tension have been determined with typical measurement uncertainties (k = 2) below 2 % at temperatures between (233 and 573) K. For binary model systems, the simultaneous analysis of fluctuations at the surface and in the bulk of the fluid allowed for the determination of interfacial tension, viscosity, thermal diffusivity, and mutual diffusivity of the very same sample within one single SLS setup.³⁴ Recently, we demonstrated the applicability of SLS for the investigation of multiphase systems in vaporliquid-liquid equilibrium, simultaneously measuring the viscosities of the two liquid phases as well as the vapor-liquid and liquid-liquid interfacial tensions.³⁵

In the present study, the simultaneous determination of liquid viscosity and surface tension of the linear alkanes *n*-hexane (*n*-C₆H₁₄), *n*-octane (*n*-C₈H₁₈), *n*-decane (*n*-C₁₀H₂₂), and *n*hexadecane (*n*-C₁₆H₃₄) by SLS at saturation conditions over a broad temperature range between (283 and 573) K with low measurement uncertainties is shown. Besides a check of the existing literature database for the studied fluids, our measurement data contribute to a new database for the liquid viscosity and surface tension, in particular for n-C₁₀H₂₂ and n-C₁₆H₃₄ at temperatures above 400 K. In the following, first, the theory, experimental procedure, and data evaluation for SLS is described. Then, the experimental results for liquid dynamic viscosity and surface tension of the four studied *n*-alkanes are correlated as a function of temperature by appropriate fit equations and are discussed in comparison with experimental data and reference correlations given in the literature. Taking also into account our former SLS results for *n*-dodecane (n-C₁₂H₂₆) and *n*-octacosane (n-C₂₈H₅₈), empirical models for the liquid viscosity and surface tension of *n*-alkanes were developed as a function of temperature and carbon number ranging between 6 and 28. Finally, the transferability of these models to the representation of viscosity and surface tension of further selected *n*-alkanes with carbon numbers within the specified range of validity is tested against literature data.

SURFACE LIGHT SCATTERING

Theoretical background. Liquid viscosity and surface tension of the *n*-alkanes under saturation conditions were measured simultaneously by surface light scattering (SLS). With this non-invasive technique, the dynamics of microscopic thermal fluctuations at phase boundaries are probed in macroscopic thermodynamic equilibrium by analyzing the intensity of the scattered light originating from the interaction between the incident light and the fluctuating surface structure. For the studied *n*-alkanes with relatively small liquid viscosities, surface fluctuations at the phase boundary between a liquid (') and a vapor ('') phase propagate and show an oscillatory behavior. In this case, the measured data for the dynamics of surface fluctuations, *i.e.* frequency ω_q and damping Γ are, in a first-order approximation, governed by the surface tension σ and the liquid kinematic viscosity v'. For an accurate determination of liquid viscosity and surface tension by SLS, the exact numerical solution of the dispersion

relation for surface fluctuations at the vapor-liquid phase boundary was used in its complete form.^{16,17} For this, data for ω_q and Γ at a defined wave number of surface fluctuations q measured by SLS are combined with reference data for the dynamic viscosity of the vapor phase η'' and density data ρ' and ρ'' for the liquid and vapor phases. More details on the SLS method and its application for the simultaneous determination of viscosity and surface tension can be found in refs 16, 18, and 19.

Materials and sample preparation According to the specifications of the suppliers³⁶, all *n*-alkane samples studied in this work had a purity of 0.99 or higher in terms of mass fraction. Since the samples did not show any particle-like impurities, they could be used without any further purification. Helium, which served as an inert gas in all the experiments, was provided by Linde AG with a mole fraction purity of more than 0.99999. A summary of the investigated chemicals and their properties is given in Table 1.

substance	source	molar mass $M / (g \cdot mol^{-1})$	specified purity	critical temperature $T_{\rm C}$ / K
<i>n</i> -C ₆ H ₁₄ (<i>n</i> -hexane, CAS 110-54-3)	Merck	86.18	mass fraction $\geq 0.99^{a}$	507.82°
<i>n</i> -C ₈ H ₁₈ (<i>n</i> -octane, CAS 111-65-9)	Merck	114.23	mass fraction 0.99999ª	568.74 ^d
<i>n</i> -C ₁₀ H ₂₂ (<i>n</i> -decane, CAS 124-18-5)	Alfa Aesar	142.28	mass fraction 0.995 ^a	617.70 ^e
<i>n</i> -C ₁₆ H ₃₄ (<i>n</i> -hexadecane, CAS 544-76- 3)	Alfa Aesar	226.45	mass fraction 0.991 ^a	722.10 ^f
Helium (CAS 7440-59-7)	Linde AG	4.0026	volume fraction $\geq 0.99999^{b}$	5.1953 ^g

Table 1. Specification of the Investigated Chemicals

^{*a*}Purity as specified in the Certificate of Analysis of the supplier. ^{*b*}Reference 37. ^{*c*}Reference 38. ^{*d*}Reference 39. ^{*e*}Reference 40. ^{*f*}Reference 41. ^{*g*}Reference 42.

The experimental SLS setup including the sample cell used in the present study is identical to that employed in our former investigations of vapor-liquid systems.^{20,21,31-34} For details, refs 18, 20, and 31 are recommended. In the following, only the experimental conditions and procedure relevant for the present study are summarized.

The clean sample cell was filled with about 50 ml of the *n*-alkane sample under atmospheric conditions and flushed by helium several times to avoid any contamination of the sample with air or other gases. After that, the cell was closed and an initial partial pressure of about 0.1 MPa of helium was adjusted. Owing to the negligible effect of helium on the liquid density, liquid viscosity, and surface tension of *n*-alkanes at such small pressures,⁹ all measurements can be considered to be performed at saturation conditions.

For the sample cell placed inside an insulated housing, the temperature *T* was regulated through resistance heating and measured by calibrated 100 Ω platinum resistance probes with expanded uncertainties (k = 2) of U(T) = 20 mK. During each experimental run, the temperature stability was better than ± 2 mK. For each temperature point, six measurements at different *q* values corresponding to different external angles of incidence $\Theta_{\rm E} = (3.0, 3.1, \text{ and } 3.2)^{\circ}$ were performed. Misalignment was avoided by irradiating the laser from either side with respect to the axis of observation. Temperatures between (283.15 and 473.15) K for *n*-C₆H₁₄, (283.15 and 523.15) K for *n*-C₈H₁₈, (283.15 and 573.15) K for *n*-C₁₀H₂₂ as well as (293.15 and 573.15) K for *n*-C₁₆H₃₄ were investigated, where the temperature was stepwise increased. Here, the temperature step was 10 K between the lowest temperature and 323.15 K, and 25 K between 323.15 K and the largest temperature.

After finishing the measurements up to 423.15 K for each sample, the measurement cell was emptied, cleaned, and refilled with a new sample for the subsequent measurements at larger temperatures. As an orange coloration of the samples was observed via the laser beam in all cases, the measurement time was reduced to a minimum in order to minimize thermal degradation. After the measurements at the largest temperature of (523.15 or 573.15) K, replicates were performed for all samples at a specific temperature in the lower temperature range with reduced laser power. All results from these repeated measurements agreed with those

from the first measurements within combined uncertainties, which is an indication for the stability of the systems.

Measurement example and data evaluation. Assuming heterodyne conditions and negligible line broadening effects (consistent with our experiments), the normalized intensity correlation function for the analysis of surface fluctuations of a defined q number with an oscillatory behavior is described by¹⁶

$$g^{(2)}(\tau) = a + b \cos\left(\omega_{q} |\tau| - \phi\right) \exp\left(-|\tau| / \tau_{c}\right).$$
(1)

In eq 1, *a* and *b* are parameters that depend on the experimental boundary conditions, while the phase term ϕ mainly accounts for the deviations of the spectrum from the Lorentzian form. The correlation time $\tau_{\rm C}$ and the frequency $\omega_{\rm q}$ represent the mean lifetime or the reciprocal of the damping Γ (= 1/ $\tau_{\rm C}$) of the surface fluctuations and their frequency of propagation.

Figure 1 shows four example correlation functions obtained from the investigation of the vapor-liquid interface of n-C₆H₁₄ and n-C₁₆H₃₄ at saturation conditions at temperatures of T = (303.15 and 473.15) K and an incident angle of $\Theta_E = 3^\circ$. From a nonlinear regression based on a Levenberg-Marquardt algorithm in which the squared sum of residuals is minimized, the values and expanded uncertainties (k = 2) for τ_C and ω_q are calculated which are given in Figure 1. Within the entire fit range, no systematic deviations of the measured correlation function from eq 1 can be observed, as can be seen from the individual residual plots in Figure 1. For a given modulus of the wave vector q with a relative expanded uncertainties $U_r(q) = 0.2$ %, the measured values for ω_q and τ_C have average relative expanded uncertainties $U_r(\omega_q) = 0.3$ % and $U_r(\tau_C) = 1.5$ %. At T = 303.15 K in Fig. 1a, n-C₁₆H₃₄ shows a smaller decay time or larger damping than n-C₆H₁₆, which can be related to the larger viscosity of n-C₁₆H₃₄. At the increased temperature of 473.15 K shown in Fig. 1b, the viscosities of the n-alkanes decrease, which results in a larger decay times of the surface fluctuations. Here, the frequency related to n-C₆H₁₆

is more than a factor of two smaller that related to $n-C_{16}H_{34}$ because of the smaller surface tension of $n-C_6H_{16}$.



Figure 1. Measurement examples of normalized correlation functions including their residuals for n-C₆H₁₆ (upper part) and n-C₁₆H₃₄ (lower part) at 303.15 K (a) and 473.15 K (b) as a function of the lag time τ using an incident angle of 3°: \Box , measurement data; —, fit according to eq 1.

In addition to the measured data for ω_q and τ_c , reference data for the *n*-alkanes at saturation conditions in the form of the liquid density ρ' , the vapor density ρ'' , and the vapor dynamic viscosity η'' with relative expanded uncertainties $U_r(\rho')$ between (0.2 and 2) %, $U_r(\rho'')$ between (0.5 and 2) %, and $U_r(\eta'') = 10$ % were required to solve the dispersion relation.^{16,17} For this, the composition of the vapor phase as a function of temperature was determined in an ideal approach from the vapor pressure of the corresponding pure *n*-alkane specified by the REFPROP database^{9,38-41} and the partial pressure of helium used as inert gas. For helium, the data for the gas density and viscosity were adopted from a Helmholtz equation of state published by Ortiz-Vega⁴² and the pure fluid model of Arp *et al.*,⁴³ respectively. For the liquid and vapor densities, the equations of state³⁸⁻⁴¹ implemented in the REFPROP database⁹ were used. To describe the vapor density of the binary mixtures consisting of *n*-alkane and helium, a linear mole-based mixing rule using the densities of the pure substances was applied. For the vapor viscosity η'' , the corresponding-states model of Lucas⁴⁴ allowing predictions of the vapor viscosity of gas mixtures at low pressures within 5 %⁴⁵ was employed where the required critical pressures of the pure *n*-alkanes were taken from the REFPROP database.^{9,38-41}

By an exact numerical solution of the dispersion equation for surface fluctuations,^{16,17} the liquid dynamic viscosity $\eta' (=v'/\rho')$ and surface tension σ were determined with total expanded measurement uncertainties (k = 2) of $U_r(\eta') = 2.0$ % and $U_r(\sigma) = 1.7$ %, respectively, averaged over all investigated systems and temperatures. For the calculation of the uncertainties, an error propagation scheme^{20,31} taking into consideration the uncertainties induced by the primary measured variables and by the adopted reference data was applied.

RESULTS AND DISCUSSION

First, the measurement results for the liquid dynamic viscosity and surface tension of the four investigated *n*-alkanes are presented and correlated as a function of temperature. Then, the experimental data are compared to available literature in the form of experimental data and reference correlations. Based on the SLS data presented in this work including former results for two further *n*-alkanes, empirical correlations for the liquid viscosity and surface tension of *n*-alkanes as function of temperature and carbon number ranging between 6 and 28 are developed and applied to further selected *n*-alkanes for testing their transferability.

Summary and correlation of liquid viscosity and surface tension data. The results for the liquid dynamic viscosity η' and surface tension σ of n-C₆H₁₄, n-C₈H₁₈, n-C₁₀H₂₂, and n-C₁₆H₃₄ as well as their expanded uncertainties (k = 2) obtained from SLS at saturation conditions between (283.15 and 573.15) K are summarized in Table 2 and are shown in Figure 2. In this figure, our measurement results³¹ for n-dodecane (n-C₁₂H₂₆) and n-octacosane (n-C₂₈H₅₈) are given for comparison. The data listed in Table 2 are average values of six independent measurements with different angles of incidence Θ_E . Furthermore, Table 2 includes the used input data for the liquid density ρ' , vapor density ρ'' , and vapor viscosity η'' required for the evaluation of η' and σ .

Table 2. Liquid Dynamic Viscosity η' and Surface Tension σ of *n*-C₆H₁₄, *n*-C₈H₁₈, *n*-C₁₀H₂₂, and *n*-C₁₆H₃₄ Obtained by Surface Light Scattering at Saturation Conditions as Function of Temperature *T* Using Literature Data for the Liquid Density ρ' , Vapor Density ρ'' , and Vapor Dynamic Viscosity η''^a

T / K	ho' /	$ ho^{\prime\prime}$ /	$\eta^{\prime\prime}$ /	η' /	100U(m')	σ /	$100U(\pi^{1})$
	$(\text{kg} \cdot \text{m}^{-3})$	$(\text{kg} \cdot \text{m}^{-3})$	(µPa·s)	(mPa·s)	$100U_{\rm r}(\eta)$	$(mN \cdot m^{-1})$	$100 U_{\rm r}(O)$
			<i>n-</i> (C_6H_{14}			
283.15	668.28	0.81	17.93	0.3495	2.1	19.28	1.0
293.15	659.29	0.87	17.79	0.3127	1.2	18.22	0.8
303.15	650.15	1.06	17.47	0.2809	1.6	17.34	1.0
313.15	642.24	1.39	17.00	0.2546	1.4	16.22	1.2
323.15	631.37	1.89	16.43	0.2340	1.5	15.31	0.8
348.15	606.75	4.00	14.81	0.1905	1.2	12.94	0.8
373.15	580.55	7.72	13.45	0.1550	1.8	10.53	0.8
398.15	552.18	13.71	12.65	0.1268	1.7	8.21	0.9
423.15	520.69	23.10	12.43	0.1026	1.9	5.98	0.9
448.15	484.16	38.04	12.74	0.0837	2.2	3.75	1.9
473.15	437.84	63.64	13.76	0.0655	3.7	1.89	1.7
			<i>n-</i> (C ₈ H ₁₈			
283.15	710.56	0.18	19.32	0.6247	1.3	22.25	0.7
293.15	702.53	0.20	19.36	0.5523	1.2	21.31	1.0
303.15	694.43	0.25	19.40	0.4828	1.3	20.53	1.0
313.15	686.26	0.32	19.45	0.4320	1.3	19.45	1.0
323.15	678.00	0.42	19.32	0.3870	1.4	18.64	1.0
348.15	656.89	0.92	18.18	0.3079	1.2	16.37	1.2
373.15	634.92	1.92	16.15	0.2487	1.2	14.00	0.9
398.15	611.81	3.77	14.05	0.2028	1.8	11.90	0.9
423.15	587.18	6.91	12.50	0.1665	1.6	9.81	1.4
448.15	560.47	11.93	11.62	0.1393	1.2	7.88	0.8

473.15	530.83	19.71	11.33	0.1168	2.0	5.82	1.6
498.15	496.74	31.76	11.58	0.0947	3.7	4.00	3.1
523.15	455.09	51.27	12.51	0.0755	3.5	2.32	2.5
			<i>n-</i> 0	$C_{10}H_{22}$			
283.15	738.06	0.17	19.16	1.077	1.6	23.94	2.1
293.15	730.28	0.17	19.62	0.9005	1.2	23.05	2.4
303.15	722.55	0.18	20.01	0.7900	0.9	22.39	2.1
313.15	714.71	0.19	20.40	0.6924	1.6	21.43	2.1
323.15	706.98	0.21	20.75	0.6091	1.2	20.61	2.1
348.15	687.34	0.32	21.34	0.4636	0.9	18.75	2.0
373.15	667.36	0.61	21.20	0.3614	1.6	16.52	2.1
398.15	646.81	1.22	20.06	0.2895	1.1	14.38	2.1
423.15	625.48	2.38	18.15	0.2403	1.9	12.40	2.5
448.15	603.09	4.39	16.03	0.1978	1.5	10.29	2.1
473.15	579.21	7.68	14.24	0.1677	2.1	8.52	2.7
498.15	553.26	12.81	13.03	0.1366	2.9	6.58	2.4
523.15	524.29	20.68	12.38	0.1138	3.1	4.97	2.3
548.15	490.60	32.86	12.28	0.0974	4.4	3.43	3.0
573.15	448.50	52.75	12.90	0.0762	6.0	1.94	4.4
			n-($C_{16}H_{34}$			
293.15	773.70	0.16	19.56	3.486	1.8	27.38	2.3
303.15	766.65	0.16	20.10	2.772	2.4	26.60	2.6
313.15	759.63	0.16	20.58	2.261	2.6	25.56	1.1
323.15	752.63	0.16	21.02	1.840	2.1	24.28	1.6
348.15	735.17	0.16	22.07	1.236	2.6	22.19	1.3
373.15	717.69	0.17	23.12	0.8927	1.6	20.31	1.0
398.15	700.11	0.19	24.14	0.6788	1.3	18.54	0.6
423.15	682.35	0.25	24.95	0.5351	1.4	16.69	1.4
448.15	664.29	0.41	25.36	0.4256	0.9	15.09	0.8
473.15	645.82	0.73	25.17	0.3440	1.8	13.26	1.5
498.15	626.80	1.36	24.28	0.2862	1.6	11.46	1.6
523.15	607.01	2.47	22.74	0.2360	2.4	9.72	2.8
548.15	586.21	4.30	20.83	0.1937	3.0	8.25	2.6
573.15	564.04	7.20	19.11	0.1705	3.7	7.00	4.1

^{*a*}Directly measured values for frequency ω_q and damping Γ at a defined wave vector q of surface fluctuations were combined with reference data for ρ' , ρ'' , and η'' described in the text to determine η' and σ by an exact numerical solution of the dispersion relation.^{16,17} The relative expanded uncertainties (k = 2) for the employed properties are $U_r(\rho')$ between (0.2 and 2) %, $U_r(\rho'')$ between (0.5 and 2) %, and $U_r(\eta'') = 10$ % (level of confidence = 0.95). The expanded uncertainty for the temperature is U(T) = 0.02K. For the liquid dynamic viscosity and surface tension, the relative expanded uncertainties $U_r(\eta')$ and $U_r(\sigma)$ are given in the table.



Figure 2. Liquid dynamic viscosity and surface tension of investigated *n*-alkanes at saturation conditions from surface light scattering. \blacklozenge , *n*-C₆H₁₄, SLS, this work; \blacksquare , *n*-C₈H₁₈, SLS, this work; \blacktriangle , *n*-C₁₀H₂₂, SLS, this work; +, *n*-C₁₂H₂₆, SLS, previous work;³¹ \bigcirc , *n*-C₁₆H₃₄, SLS, this work; \times , *n*-C₂₈H₅₈, SLS, previous work;³¹ —, fits of the experimental data for viscosity and surface tension data according to eqs 2 and 3.

The upper part of Figure 2 shows that our experimental data for the liquid viscosity of the *n*-alkanes decrease with decreasing alkyl chain length and increasing temperature. For the representation of the saturated liquid dynamic viscosity of the four investigated *n*-alkanes as a function of temperature, a Vogel-type equation according to

$$\eta'_{\text{calc}} = \exp\left(\sum_{i=0}^{4} \frac{\eta'_i}{T^i}\right)$$
(2)

was used where all experimental data have the same statistical weight in the fitting. In eq 2, T is the temperature in K and η'_0 , η'_1 , η'_2 , η'_3 , and η'_4 are fit coefficients listed in Table 3 for the

four studied *n*-alkanes. As well, the absolute average deviation (AAD) of the experimental data from those calculated with eq 2 is given and between (0.36 and 0.94) %. For all systems and thermodynamic states, the relative deviations of the experimental η' data from the fits are smaller than the experimental uncertainties of the measurements, as it can be seen in Figure 3.

Table 3. Coefficients of Equation 2 for the Liquid Dynamic Viscosity $\eta'_{calc}(T)$ of *n*-C₆H₁₄, *n*-C₈H₁₈, *n*-C₁₀H₂₂, and *n*-C₁₆H₃₄ at Saturation Conditions.

	<i>n</i> -C ₆ H ₁₄	<i>n</i> -C ₈ H ₁₈	<i>n</i> -C ₁₀ H ₂₂	<i>n</i> -C ₁₆ H ₃₄
η'_0	$-2.27934 \cdot 10^{1}$	$-2.36227 \cdot 10^{1}$	$-1.94144 \cdot 10^{1}$	-9.05711
η'_1 / K	$2.24935 \cdot 10^4$	$2.65713 \cdot 10^4$	$2.18398 \cdot 10^4$	$7.13979 \cdot 10^3$
η'_2 /·K ²	$-9.37662 \cdot 10^{6}$	$-1.25743 \cdot 10^{7}$	$-1.04993 \cdot 10^{7}$	$-2.27994 \cdot 10^{6}$
η'_3 / K^3	$1.79808 \cdot 10^9$	2.74913·10 ⁹	$2.36279 \cdot 10^9$	$3.47185 \cdot 10^8$
η^{\prime_4} / K^4	$-1.28243 \cdot 10^{11}$	$-2.24664 \cdot 10^{11}$	$-1.97840 \cdot 10^{11}$	$-9.56343 \cdot 10^9$
$AAD^a / \%$	0.36	0.66	0.94	0.71
<i>T</i> range / K	283.15-473.15	283.15-523.15	283.15-573.15	293.15-573.15

^{*a*}Absolute average deviation of η' to the fit

For the surface tensions of the studied *n*-alkanes, decreasing values with decreasing alkyl chain length and increasing temperature are found, as it can be seen in the lower part of Figure 2. The experimental surface tension data are represented by a modified van der Waals equation according to

$$\sigma_{\text{calc}} = \sigma_0 \left(1 - T_{\text{R}} \right)^{1.26} \left[1 + \sigma_1 \left(1 - T_{\text{R}} \right)^{0.5} + \sigma_2 \left(1 - T_{\text{R}} \right) \right], \tag{3}$$

where all data have the same statistical weight in the fitting. In eq 3, $T_R (= T/T_C)$ is the reduced temperature and requires information on the critical temperature T_C . For the latter property, the data given in Table 1 were employed. Table 4 lists the fit parameters σ_0 , σ_1 , and σ_2 as well as the AAD of the measured σ data from eq 3 ranging between (0.36 and 0.95) %. The relative deviations of the experimental data from the fits are shown in Figure 4 and are in most cases smaller than the experimental uncertainties of the measured data.

	<i>n</i> -C ₆ H ₁₄	<i>n</i> -C ₈ H ₁₈	<i>n</i> -C ₁₀ H ₂₂	<i>n</i> -C ₁₆ H ₃₄
$\sigma_0 /(\mathrm{mN}\cdot\mathrm{m}^{-1})$	51.43	56.49	44.14	68.79
$\sigma_{ m l}$	0.5378	-0.0088	0.7697	-0.9615
σ_2	-0.7079	-0.1077	-0.7197	0.8507
$AAD^a / \%$	0.50	0.36	0.95	0.91
T range / K	283.15-473.15	283.15-523.15	283.15-573.15	293.15-573.15

Table 4. Coefficients of Equation 3 for the Surface Tension $\sigma_{calc}(T)$ of *n*-C₆H₁₄, *n*-C₈H₁₈, *n*-C₁₀H₂₂, and *n*-C₁₆H₃₄ at Saturation Conditions.

^{*a*}Absolute average deviation of η' to the fit

Comparison with literature. In the following, our experimental data for the saturated liquid dynamic viscosity and surface tension of n-C₆H₁₄, n-C₈H₁₈, n-C₁₀H₂₂, and n-C₁₆H₃₄ are compared with literature data in the form of the corresponding reference correlations implemented in the REFPROP database^{9,38-41} as well as experimental data measured in the temperature range of interest in this study. For the viscosity, only the primary experimental data sets used in the development of the respective reference correlations were considered. For the surface tension, all available sources collected by the ThermoData Engine (TDE), version 10.3,8 were employed. This was necessary because it is not clear which surface tension data were used as the basis of the reference correlations of $n-C_6H_{14}$, $n-C_8H_{18}$, and $n-C_{10}H_{22}$ developed by Mulero et al.¹¹ The relevant details on the literature data considered in this study including the experimental methods used, the specified uncertainties, the number of data points, and the values for the AADs from the fits of our experimental data according to eqs 2 and 3 are summarized in Tables 5 and 6. In Figures 3 and 4, the relative deviations of our SLS results and of the literature data from eqs 2 and 3 are shown as a function of temperature. For visibility purposes, only exemplary error bars representing the total uncertainties of the individual SLS measurements are shown, while error bars related to the literature data are omitted. For the representation of the uncertainty of the fits illustrated by the dotted lines in Figures 3 and 4, the mean uncertainty of our experimental SLS data averaged over the various temperatures was used.

Liquid viscosity. For the viscosity of n-C₆H₁₄ shown in Figure 3a, the reference correlation developed by Michailidou *et al.*⁴⁶ together with nine primary data sets⁴⁷⁻⁵⁵ are compared to eq 2. Agreement between our measurements and the reference correlation, which is specified with an uncertainty of (2 and 6) % below and above 450 K, is found within the experimental uncertainties. All primary data sets agree with our fit within the combined expanded uncertainties and show an AAD of 1.7 %. As can be seen from Figure 3a, our data follow the reference correlation at temperatures above 370 K better than the available literature data.



Figure 3. Relative deviations for liquid dynamic viscosity of investigated *n*-alkanes at saturation conditions from the fits of our experimental data (eq 2, —) as a function of temperature. (a) *n*-C₆H₁₄: \blacklozenge , this work; -----, reference correlation from Michailiou *et al.*;⁴⁶ \diamondsuit , Oliveira and Wakeham;⁴⁷ \diamondsuit , Assael *et al.*;⁴⁸ \diamondsuit , Berstad;⁴⁹ \diamondsuit , Knapstad *et al.*;⁵⁰ \diamondsuit , Grigor'ev *et al.*;⁵¹ \diamondsuit , Bauer and Meerlender;⁵² \diamondsuit , Dymond and Young;⁵³ \diamondsuit , Dymond *et al.*;⁵⁴ \diamondsuit , Agaev and Golubev.⁵⁵ (b) *n*-C₈H₁₈: \blacksquare , this work; ------, reference correlation from Huber *et al.*;¹³ \square , Dymond and Young;⁵³ \blacksquare , Badalyan and Rodchenko;⁵⁶ \blacksquare , Knapstad *et al.*;⁵⁰ \blacksquare , Keramidi and Badalyan;⁵⁷ \blacksquare , Oliveira and Wakeham;⁴⁷ \blacksquare , Harris *et al.*;⁵⁸ \blacksquare , Caudwell *et al.*;⁵⁹ (c) *n*-C₁₀H₂₂: \blacktriangle , this work; ------, reference correlation from Huber *et al.*;⁶⁰ \bigstar , Knapstad *et al.*;⁵⁰ \bigstar , Assael *et al.*;⁶¹ \bigstar , Oliveira and Wakeham;⁴⁷ \blacksquare , Assael *et al.*;⁵⁸ \blacksquare , Caudwell *et al.*;⁵⁹ (d) *n*-C₁₆H₃₄: \textcircled this work; ------, reference correlation from Huber *et al.*;⁵⁹ \bigstar , Numond and Young;⁶⁰ \bigstar , Knapstad *et al.*;⁵⁰ \bigstar , Assael *et al.*;⁶¹ \bigstar , Oliveira and Wakeham;⁴⁷ \bigstar , Caudwell *et al.*;⁵⁹ (d) *n*-C₁₆H₃₄: \textcircled this work; ------, reference correlation from Huber *et al.*;¹⁴ \circlearrowright , Nederbragt and

Boelhouwer;⁶² \bullet , Dymond and Young;⁵³ \bullet , Dymond *et al.*;⁵⁴ \bullet , Wakefield and Marsh;⁶³ \bullet , Wakefield;⁶⁴ \bullet , Tanaka *et al.*;⁶⁵ \bullet , Wu *et al.*;⁶⁶ \bullet , Mohammed *et al.*;⁶⁷ \bullet , Sanchez-Vicente *et al.*⁶⁸ Dotted lines (.....) represent the uncertainties of the fits based on the average uncertainty of our experimental data.

For the liquid viscosity of n-C₈H₁₈, Figure 3b shows that the reference correlation developed by Huber *et al.*¹³ and specified with an expanded uncertainty of 0.6 % along the saturated liquid line agrees with our experimental data within combined uncertainties over the complete temperature range up to 523.15 K. For the seven primary data sets, agreement with our fit within combined uncertainties is found up to temperatures of 448.15 K. At larger temperatures, the experimental data reported by Badalyan and Rodchenko⁵⁶ as well as Keramidi and Badalyan⁵⁷ show increasing negative deviations from eq 2 with maximum values up to -5.3 % at 523.15 K.

Values for the liquid viscosity of n-C₁₀H₂₂ at saturation conditions are compared in Figure 3c with the reference correlation from Huber *et al.*¹³ specified with an uncertainty of 1 % and with the five underlying primary data sets^{47,50,53,56-59} obtained at temperatures up to 423.15 K. Within the temperature range between (283.15 and 423.15) K, agreement between our data, the reference correlation, and the primary experimental data within combined uncertainties is found. Above 423.15 K, the reference correlation yields slightly larger viscosity values than our fit with maximum relative deviations up to 4.6 %. This trend seems to be caused by the extrapolative character of the reference correlation which is based on primary data for the saturated liquid viscosity measured only up to 423.15 K.

Table 5. Reference Correlations and Primary Data Sets for the Saturated Liquid Dynamic
Viscosity of <i>n</i> -C ₆ H ₁₄ , <i>n</i> -C ₈ H ₁₈ , <i>n</i> -C ₁₀ H ₂₂ , and <i>n</i> -C ₁₆ H ₃₄ Used for Comparison with the SLS
Measurements

1 st author	year of publication	technique employed ^a	$100U_{\rm r}$	number of data points	AAD ^b / %	
<i>n</i> -C ₆ H ₁₄						
Reference correlation						

Michailidou ⁴⁶	2013				0.5
		Prima	ry data		
Oliveira ⁴⁷	1992	VW	0.5–2.4	3	0.2
Assael ⁴⁸	1991	VW	0.5	1	0.7
Berstad ⁴⁹	1989	OCup	1.15	6	1.0
Knapstad ⁵⁰	1989	OCup	0.33-0.56	7	0.3
Grigor'ev ⁵¹	1988	Cap	0.9^{c}	4	0.7
Bauer ⁵²	1984	Cap	0.3	4	0.6
Dymond ⁵³	1980	Cap	0.5	12	0.7
Dymond ⁵⁴	1980	FB	2	4	1.5
Agaev ⁵⁵	1963	Cap	1-3 ^c	21	1.6
		n-(C8H18		
- 12		Reference	correlation		
Huber ¹³	2004				0.8
.52		Prima	ry data		
Dymond ⁵³	1980	Cap	0.5	8	0.8
Badalyan ⁵⁶	1986	Cap	1	10	2.0
Knapstad ⁵⁰	1989	OCup	0.33-0.56	5	0.5
Keramidi ⁵⁷	1991	Cap	$1-3^{d}$	6	3.2
Oliveira ⁴⁷	1992	VW	0.5 - 2.4	3	0.4
Harris ⁵⁸	1997	FB	2	5	0.4
Caudwell ⁵⁹	2009	VW	2	4	0.7
		<i>n-</i> C	$C_{10}H_{22}$		
. 12		Reference	correlation		
Huber ¹⁵	2004				1.6
.(0		Prima	iry data		
Dymond ⁶⁰	1981	Cap	0.5	8	0.5
Knapstad ³⁰	1989	OCup	0.33-0.56	11	0.5
Assael ⁶¹	1992	VW	0.5	6	0.5
Oliveira ⁴⁷	1992	VW	0.5–2.4	3	0.7
Caudwell ⁵⁹	2009	VW	2	4	0.2
		<u>n-C</u>	C ₁₆ H ₃₄		
14	2010	Reference	correlation		0.0
Meng	2018	D			0.9
$\frac{\text{Primary data}}{1047} = \frac{1047}{1047} = \frac{10}{10}$					
Nederbragt	1947	Cap	N/A^{2}	3	1.0
Dymond ¹⁵⁴	1980	Cap	0.5	4	0.4
Dymond ¹	1980	гв	2	10	0.5
Wakefield ⁶⁵	1987	Cap	0.5	3	0.5
Wakefield ^{or}	1988	Cap	0.5	2	0.2
Tanaka ⁰³	1991	TC	2	3	1.1
Wu ⁰⁰	1998	Сар	0.1	4	1.0

^aCap, Capillary viscometer; FB, Falling body viscometer; OCup, Oscillating cup viscometer; TC, Torsional crystal viscometer; VW, Vibrating wire viscometer.
 ^bAbsolute average deviation between the literature data set and eq 2

^cUncertainties taken from Reference 46

^dUncertainties taken from Reference 13

^eUncertainties are not specified

For the liquid viscosity of n-C₁₆H₃₄, the reference correlation developed by Meng *et al.*¹⁴ with a specified uncertainty of 1 % as well as the seven primary data sets along the saturation line are available for data comparison in Figure 3d. Over the entire temperature range, all literature data agree with our correlation, eq 2, within combined uncertainties, resulting in AAD values below 1.1 %. As it can be seen in Figure 3d, the reference correlation follows the experimental data from Nederbragt and Boelhouwer⁶² who performed measurements at temperatures between (298.15 and 518.15) K.

Surface tension. For the surface tension of *n*-C₆H₁₄ shown in Figure 4a, agreement between our data and the reference correlation from Mulero *et al.*¹¹ within relative absolute deviations of 2 % is found over the entire temperature range between (283.15 and 473.15) K. The same statement holds also for most of the totally 41 different experimental data sets, including the measurements of Grigoryev *et al.*⁶⁹ which follow our data and the reference correlation up to 473.15 K. At temperatures below 340 K, larger positive and negative deviations from eq 3, which are up to 7.5 % and outside combined uncertainties, are observed for the data measured by Dutoit and Friedrich,⁷⁰ Harkins *et al.*,⁷¹ Murphy *et al.*,⁷² and Ridgway and Butler.⁷³ At temperatures above 400 K, the three further data sets from Skripov and Sinitsyn,⁷⁴ Skripov and Firsov,⁷⁵ and Bagdasaryan⁷⁶ show increasing negative relative deviations from the present work with values up to –9 %.



Figure 4. Relative deviations for the surface tension of investigated *n*-alkanes at saturation conditions from the fits of our experimental data (eq 3, —) as a function of temperature. (a) n-C₆H₁₄: \blacklozenge , this work; -----, reference correlation from Mulero *et al.*;¹¹ ; \diamondsuit , experimental literature data as given in Table 6. (b) n-C₈H₁₈: \blacksquare , this work; -----, reference correlation from Mulero *et al.*;¹¹ \square , experimental literature data as given in Table 6. (b) n-C₈H₁₈: \blacksquare , this work; -----, reference correlation from Mulero *et al.*;¹¹ \square , experimental literature data as given in Table 6. (c) n-C₁₀H₂₂: \blacktriangle , this work; ------, reference correlation from Mulero *et al.*;¹¹ \triangle , experimental literature data as given in Table 6. (d) n-C₁₆H₃₄: \blacklozenge , this work; ------, reference correlation from Mulero *et al.*;¹² \triangle , experimental literature data as given in Table 6. (d) n-C₁₆H₃₄: \blacklozenge , this work; ------, reference correlation from Huber;¹² \bigcirc , experimental literature data as given in Table 6. (d) n-C₁₆H₃₄: \blacklozenge , this work; ------, reference correlation from Huber;¹² \bigcirc , experimental literature data as given in Table 6. Dotted lines (-----) represent the uncertainties of the fits based on the average uncertainty of our experimental data.

Table 6. Reference Correlations and Available Experimental Data Sets for Surface Tension of $n-C_6H_{14}$, $n-C_8H_{18}$, $n-C_{10}H_{22}$, and $n-C_{16}H_{34}$ Used for Comparison with the SLS Measurements

1 st author	year of publication	technique employedª	$100U_{\rm r}$	number of data points	AAD^b / %	
		n-C ₆ H	I ₁₄			
Reference correlation						
Mulero ¹¹	2012				1.2	
	Primary data					
Schiff ⁷⁷	1884	CR	1.75–2.4	2	1.2	
Dutoit ⁷⁰	1900	CR	N/A^{c}	2	5.0	
Morgan ⁷⁸	1913	DW	N/A^{c}	2	2.4	
Harkins ⁷¹	1917	DW	N/A^{c}	1	5.3	
Harkins ⁷⁹	1921	DW	N/A^{c}	4	0.8	

II (D. 1. 180	1021	CD		2	0.0
Hennaut-Roland ⁸⁰	1931	CR	N/A ^c	3	0.9
Irieschmann ⁶¹	1935	MBP	0.05	1	2.4
Wibaut ⁶²	1939	MBP	0.15	l	0.8
Quayle	1944	MBP	N/A^c	2	0.3
Vogel	1946	CR	N/A ^c	7	1.3
Jasper ⁶⁵	1953	CR	0.05	7	0.8
Jasper ⁸⁰	1955	CR	0.05	7	0.6
Murphy ⁷²	1957	RD	N/A^{c}	1	7.5
Ben'kovskii ⁸⁷	1964	CR + MBP	0.5	3	0.9
Clever ⁸⁸	1963	MBP	N/A^{c}	3	0.7
Schmidt ⁸⁹	1966	MBP	N/A^{c}	4	0.7
Ridgway ⁷³	1967	RD	1	1	3.8
Skripov ⁷⁵	1968	CR	0.6 - 1.5	6	5.8
Skripov ⁷⁴	1968	DV	N/A^{c}	3	7.5
Bagdasaryan ⁷⁶	1979	N/A ^c	N/A^{c}	20	3.5
Ross ⁹⁰	1979	PD	0.35	1	0.9
Grigoryev ⁶⁹	1985	CR	N/A^{c}	20	0.6
Grigorvev ⁹¹	1992	CR	N/A^{c}	6	1.5
Papaioannou ⁹²	1994	CR	0.25	1	0.2
Pineiro ⁹³	1999	DV	0.25	1	0.9
Limenez ⁹⁴	2000	DV	0.06	1	0.7
Penas ⁹⁵	2000	DV	0.00	1	0.9
Pandev ⁹⁶	2000		0.15	1	0.9
A zizion ⁹⁷	2001		4	1	1.2
AZIZIAII Dominguoz Doroz ⁹⁸	2000		0.00	5	1.0
Dominguez-Perez	2006		0.06	1	0.8
Giner	2007	DV	0.06	7	1.4
Azizian	2008	RD	0.1	5	1.0
Giner ¹⁰¹	2008	DV	0.01	7	1.4
Perez-Gregorio ¹⁰²	2009	DV	0.06	7	1.4
Dominguez-Perez ¹⁰³	2010	DV	0.06	1	0.8
Perez-Navarro ¹⁰⁴	2010	DV	0.06	1	1.6
Wang	2011	RD	0.5	5	2.0
Mejia ¹⁰⁶	2012	MBP	1.1	1	0.7
Garrido ¹⁰⁷	2014	PD	0.06	4	0.9
Mejia ¹⁰⁸	2014	MBP	0.06	1	0.7
Tahery ¹⁰⁹	2017	PD	0.5	1	0.7
		<i>п</i> -С ₈ Н	18		
		Reference con	rrelation		
Mulero ¹¹	2012				0.9
		Primary	data		
Harkins ⁷¹	1917	DW	N/A ^c	1	1.8
Harkins ⁷⁹	1921	DW	N/A ^c	4	1.7
Richards ¹¹⁰	1924	CR	N/A ^c	2	1.6
Hennau-Roland ⁸⁰	1931	CR	N/A ^c	3	1.8
Wibaut ⁸²	1939	MRP	0.15	1	1.0
Manzoni-Ansidei ¹¹¹	1940	N/A ^c	N/A ^c	5	1.5
	10//	MPD	N/Λ^{c}	4	1.1
Smith ¹¹²	1944		1 V / 2 A	т 1	1.5
$V_{2} \sim 1^{84}$	1744		1N/2A NT/AC	1	1.J 1 1
vogei	1940		IN/A	5	1.1
Jasper ³⁶	1953	CK	0.05	0 10	1.5
Jasper ⁶⁶	1955		0.05	10	0.7
Ben'kovsk11°'	1964	CR + MBP	0.5	3	1.9
Baglay	1988	CR	0.5	4	1.4
Ramkumar ¹¹⁴	1989	CR	7.7	3	0.2

Grigorvev ⁹¹	1992	CR	N/A^{c}	8	2.9
Pineiro ⁹³	1999	DV	0.2	1	1.3
Penas ⁹⁵	2000	DV	0.15	1	1.3
Segade ¹¹⁵	2003	DV	N/A^c	1	1.2
Mosteiro ¹¹⁶	2009	DV	0.4	1	0.4
Meija ¹¹⁷	2011	MBP	0.5	3	0.9
Gavol ¹¹⁸	2013	DV	0.5	1	0.5
Lopez-Lazaro ¹¹⁹	2015	DV	0.15	5	1.9
Tahery ¹⁰⁹	2013	PD	0.15	1	1.9
	2017	<i>n</i> -C ₁₀ H	22	1	1.1
		Reference cor	relation		
Mulero ¹¹	2012				1.3
		Primary o	lata		
Hennaut-Roland ⁸⁰	1931	CR	N/A^{c}	3	3.3
Quayle ⁸³	1944	MBP	N/A^{c}	3	2.9
Vogel ⁸⁴	1946	CR	N/A^{c}	6	2.2
Mumford ¹²⁰	1950	MBP	N/A^{c}	1	3.7
Jasper ⁸⁵	1953	CR	0.05	6	2.8
Jasper ⁸⁶	1955	CR	0.05	10	1.8
Ben'kovskii ⁸⁷	1964	CR + MBP	0.5	3	3.2
Korosi ¹²¹	1981	CR	0.15	2	1.6
Nagarajan ¹²²	1986	PD	2-3	2	1.0
Pineiro ⁹³	1999	PD	0.2	1	2.7
Penas ⁹⁵	2000	PD	0.2	1	2.4
Domanska ¹²³	2002	RD	0.1	2	2.7
Gomez-Diaz ¹²⁴	2002	WP	N/A^c	1	5.4
Rolo ¹²⁵	2002	WP	1	5	4.7
Oueimada ¹²⁶	2005	WP	0.1	6	4.0
Mosteiro ¹¹⁶	2009	DV	0.4	1	1.7
Mohsen-Nia ¹²⁷	2010	MBP	0.2	6	2.2
Meiia ¹¹⁷	2011	MBP	0.5	3	2.8
Gavol ¹¹⁸	2013	DV	0.5	1	1.0
Luning Prak ¹²⁸	2017	PD	0.8	1	3.4
0		<i>n</i> -C ₁₆ H	34		
		Reference cor	relation		
Huber ¹²	2017				2.1
		Primary o	lata		
Vogel ⁸⁴	1946	CR	N/A^{c}	5	1.6
Jasper ⁸⁵	1953	CR	0.05	9	1.7
Fox ¹²⁹	1955	RD	0.1	1	1.2
Jasper ⁸⁶	1955	CR	0.05	9	1.1
Koefoed ¹³⁰	1958	CR	0.10	2	0.7
Myers ¹³¹	1969	MBP	0.15	1	1.2
Korosi ¹²¹	1981	CR	0.15	2	1.0
Rolo ¹²⁵	2002	WP	1.00	6	2.8
Mohsen-Nia ¹³²	2011	MBP	0.16	10	1.4
Luning Prak ¹³³	2014	PD	1	1	0.5
Luning Prak ¹³⁴	2016	PD	1	1	0.4
Luning Prak ¹²⁸	2017	PD	1	1	1.0
Luning Prak ¹³⁵	2017	PD	1	1	0.0
Luning Prak ¹³⁶	2017	PD	1	1	0.0

^cUncertainties are not specified

Figure 4b shows the comparison of surface tension data of n-C₈H₁₈. With the exception of the data reported by Grigoryev *et al.*⁹¹ all further 22 experimental data sets and the reference correlation from Mulero *et al.*¹¹ agree well with our measurement results with an AAD of 1.4 %. It should be mentioned that the measurements of Grigoryev *et al.*⁹¹ match with our correlation, eq 3, inside combined uncertainties for temperatures below 400 K, before larger relative deviations occur at larger temperatures. While the surface tension data of Grigoryev *et al.*⁹¹ deviates by +6.3 % at 428.15 K, a further temperature increase results from positive deviations crossing eq 3 to negative deviations of -10.4 % at 522.48 K. The same trend seems also to be reflected in the behavior of the reference correlation from Mulero *et al.*¹¹ relative to our data at temperatures above 450 K.

In connection with the comparison of the surface tension of n-C₁₀H₂₂ illustrated in Figure 3c, the experimental literature data available up to 373 K as well as the reference correlation of Mulero *et al.*¹¹ are slightly larger than our experimental data. For temperatures between (283.15 and 373.15) K, the AAD of all literature sources from eq 3 is 2.7 %. Due to the lack of literature data above 373 K, the reference correlation is purely predictive where its extrapolation can represent the temperature-dependent trend of our SLS data.

The situation for the surface tension of n-C₁₆H₃₄ shown in Figure 4d is similar to that for n-C₁₀H₂₂. For temperatures up to 373 K where experimental data are present, the literature data including the reference correlation from Huber¹² are generally larger than our surface tension values represented by eq 3 with an AAD of 1.4 %. The increasing deviations of the reference correlation from our fit at temperatures above 500 K can again be related to the extrapolation at states where no experimental data could be considered in the development of the correlation.

^{*a*}CR, Capillary rise method; DW, Drop weight method; MBP, Maximum bubble pressure method; RD, Ring detachment method; PD, Pendent drop method; DV, Drop volume method; WP, Wilhelmy plate method

^bAbsolute average deviation (AAD) between the literature data set and eq 3

Development of empirical models for liquid viscosity and surface tension of *n*-alkanes. In this section, simple empirical models for the saturated liquid viscosity and surface tension of *n*-alkanes as a function of temperature and number of carbon atoms are presented. For this purpose, our experimental results for *n*-C₆H₁₄, *n*-C₈H₁₈, *n*-C₁₀H₂₂, and *n*-C₁₆H₃₄ presented in this work in Table 2 as well as for *n*-C₁₂H₂₆ and *n*-C₂₈H₅₈ reported in our previous study³¹ were used as target set. In a first step, the data for the viscosity η' and surface tension σ were fitted as a function of temperature for the six *n*-alkanes. In a second step, the fitting parameters obtained from the first step were then fitted as a function of the number of carbon atoms in the respective *n*-alkanes.

For the correlation of the liquid dynamic viscosity of *n*-alkanes, we used an empirical correlation in terms of the reduced temperature $T_{\rm R}$ (= $T/T_{\rm C}$) according to

$$\eta'_{\rm corr} = \exp\left(\sum_{i=0}^{3} \eta'_{i,\rm C} T_{\rm R}^{\ i}\right),\tag{4}$$

where $\eta'_{i,C}$ are fitting parameters which are a function of the number of carbon atoms ranging between 6 and 28. In comparison with the data correlation in eq 2, we found that it is appropriate to terminate the polynomial extension after the third term to obtain fitting parameters which show a clear dependency of the number of carbon atoms. For the critical temperature T_C of the *n*-alkanes, the REFPROP database⁹ was employed. The coefficients $\eta'_{i,C}$ are fitted by a second order polynomial as a function of the number of carbon atoms n_C according to

$$\eta'_{i,C} = A_i + B_i n_C + C_i n_C^2,$$
(5)

where A_i , B_i , and C_i are fitting parameters given in Table 7. In the fitting procedures, the same statistical weight of each measured data point for η' is assumed. Based on the empirical fit equations according to eqs 4 and 5, the totally 72 experimental data points of the six *n*-alkanes used as training set can be represented with an AAD of 1.9 %.

Table 7. Parameters in the Empirical Models for the Liquid Dynamic Viscosity $\eta'_{i,C}$ and Surface Tension $\sigma_{i,C}$ of *n*-Alkanes at Saturation Conditions as a Function of Temperature and Number of Carbon Atoms Ranging Between 6 and 28

Parameters for $\eta'_{i,C}$ in eq 5 used in eq 4					
$A_0 = -1.1147 \cdot 10^{-1}$	$B_0 = 1.1829$	$C_0 = -1.7983 \cdot 10^{-2}$			
$A_1 = -1.4660$	$B_1 = -4.1141$	$C_1 = 4.8808 \cdot 10^{-2}$			
$A_2 = -2.7482$	$B_2 = 5.0579$	$C_2 = -3.5541 \cdot 10^{-2}$			
$A_3 = 1.3182$	$B_3 = -2.1079$	$C_3 = 1.4650 \cdot 10^{-3}$			
Parameters for $\sigma_{i,C}$ in eq 7 used in eq 6					
$D = 5.6792 \cdot 10^1$	$E = -4.5458 \cdot 10^{-1}$	$F = 6.0013 \cdot 10^{-3}$			

For the correlation of the surface tension of *n*-alkanes, the van der Waals equation¹³⁷

$$\sigma_{\rm corr} = \sigma_{\rm C} \left(1 - T_{\rm R} \right)^{1.26} \tag{6}$$

was found to be appropriate for the representation of the measured σ data over a wide temperature range. Though eq 6 with the universal critical exponent 1.26 is derived from the scaling theory for the description of the surface tension in vicinity of the critical point, we found that it can also be used for a broader range of surface tensions as studied in this work. In eq 6, the parameters $\sigma_{\rm C}$ is fitted as a function of the number of carbon atoms $n_{\rm C}$ via

$$\sigma_{\rm C} = D + E n_{\rm C} + F n_{\rm C}^2. \tag{7}$$

The coefficients in eq 7 are summarized in Table 7. The same statistical weight for each data point entering into the fitting procedures was assumed. With the developed correlation given by eqs 6 to 7, the 72 surface tension data of the considered six *n*-alkanes are modeled with an AAD of 1.6 %.

For probing the transferability of the developed correlations to *n*-alkanes that were not considered in the model development, the four *n*-alkanes *n*-heptane (n-C₇H₁₆), *n*-nonane (n-C₉H₂₀), *n*-undecane (n-C₁₁H₂₄) and *n*-eicosane (n-C₂₀H₄₂) with carbon numbers between 7 and 20 were used as a validation set. For these systems, the critical temperatures required in the empirical correlations were taken from the REFPROP database.^{9,12,40,138,139} The results for liquid viscosity and surface tension predicted by eqs 4 and 6 including their corresponding fit

parameters are compared with available reference correlations. For n-C₇H₁₆, n-C₉H₂₀, and n-C₁₁H₂₄, the corresponding reference correlations^{11-13,140-142} for η' and σ implemented in REFPROP⁹ were adopted. In detail, the reference correlations developed by Michailidou *et al.*¹⁴⁰ for the viscosity and by Mulero *et al.*¹¹ for the surface tension were used for n-C₇H₁₆. For n-C₉H₂₀, the correlations reported by Huber *et al.*¹³ and Mulero *et al.*¹¹ for viscosity and surface tension were employed. While the reference correlations from Assael *et al.*¹⁴¹ and Mulero and Cachadiña¹⁴² were used for the viscosity and surface tension of n-C₁₁H₂₄, those reported by Huber¹² were considered for n-C₂₀H₄₂.

For the comparison between the empirical correlations developed in this work and the reference correlations, temperature ranges between (283.15 and 520.15) K for *n*-C₇H₁₆, between (283.15 and 573.15) K for *n*-C₉H₂₀ and *n*-C₁₁H₂₄ as well as between (313.15 and 573.15) K for *n*-C₂₀H₄₂ were taken into account. The smaller temperature ranges for *n*-C₇H₁₆ and *n*-C₂₀H₄₂ are required to exclude the regions in vicinity of the critical point of *n*-C₇H₁₆ ($T_C = 540.2 \text{ K}$)⁹ and the melting point of *n*-C₂₀H₄₂ at *T* = 309.85 K.¹⁴³ For the four *n*-alkanes used as a validation set, agreement between the correlations according to eqs 4 and 6 as well as the reference correlations was found, resulting in AAD values of (4.8 and 2.3) % for the liquid dynamic viscosity and the surface tension, respectively. This indicates the transferability of the developed empirical correlations to *n*-alkanes with varying carbon number.

CONCLUSIONS

The present study has contributed to a reliable experimental database for the liquid viscosity and surface tension of *n*-hexane, *n*-octane, *n*-decane, and *n*-hexadecane at saturation conditions over a broad temperature range from (283 up to 573) K. With surface light scattering in thermodynamic equilibrium, total average measurement uncertainties (k = 2) of (2.0 and 1.7) % could be achieved for the liquid dynamic viscosity and surface tension. For the studied *n*- alkanes, agreement between the experimental data, which were represented by a Vogel-type equation for the dynamic viscosity as well as a modified van der Waals equation for the surface tension as a function of temperature, and the available literature data was found. Based on the measured data, empirical models for the liquid viscosity and surface tension of *n*-alkanes were developed as a function of temperature and carbon number ranging between 6 and 28. Their transfer to further selected *n*-alkanes which were not considered in the development procedure showed reasonable agreement.

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ACKNOWLEDGMENTS

This work was financially supported by the German Research Foundation (Deutsche Forschungsgemeinschaft, DFG) through funding the Erlangen Graduate School in Advanced Optical Technologies (SAOT) within the German Excellence Initiative and via the project Grant FR 1709/15-1.

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TOC Graphics

