Predicting the viscosity of liquid mixtures consisting of *n*-alkane, alkylbenzene and cycloalkane species based on molecular description

Thanh-Binh Nguyen,[1,2,3] and Velisa Vesovic[2,*]

1. Faculty of Chemical Engineering, The University of Danang - University of Science and Technology, 54 Nguyen Luong Bang St., Danang city, Vietnam.

2. Department of Earth Science and Engineering, Imperial College London, South Kensington Campus, London SW7 2AZ, United Kingdom.

3. Qatar Carbonates and Carbon Storage Research Centre (QCCSRC), Imperial College London, London SW7 2AZ, UK

* To whom correspondence should be addressed. E-mail: v.vesovic@imperial.ac.uk

Abstract

1-component Extended Hard-Sphere (1-cEHS) model has been developed recently to predict the viscosity of liquid, *n*-alkane mixtures. It represents a mixture by a single pseudo-component characterized by an appropriate molecular weight and calculates the viscosity by means of the modified, extended hard-sphere model (EHS) that makes use of a universal function relating reduced viscosity to reduced volume. In this work we have extended the model to also predict the viscosity of mixtures containing alkylbenzene and cycloalkane species. Furthermore, we have developed a new 3-component Extended Hard-Sphere (3-cEHS) model which requires only a knowledge of the overall composition of *n*-alkane, alkylbenzene and cycloalkane species. Extensive comparison with the available experimental data indicates that both models (1-cEHS and 3-cEHS) predict the viscosity of binary and multicomponent mixtures containing *n*-alkane, alkylbenzene and cycloalkane species with uncertainty of 5-10%. The proposed models are a precursor of a new family of models that do not require a knowledge of the detailed composition of the mixture, but still take advantage of the underlying molecular description.

Keywords

1. Introduction

There is a growing need for accurate and reliable knowledge of thermophysical properties driven not only by the need to better characterize fluids used in current industrial applications, but also to develop and design more optimal and purpose-specific fluids by means of multiscale molecular approaches. The current trend is to achieve this by developing predictive, versatile models that can provide accurate and reliable values and that have been validated by comparing with a wide range of experimental data of low uncertainty. For thermodynamic properties we have a range of tools for such an endeavour that are based on strong underlying fundamentals [1]. For transport properties the situation is mixed [1,2]. At low pressure, recent developments and well understood kinetic theory allow us to calculate the transport properties of gases and their mixtures with an uncertainty commensurate with the best experimental measurements over a wide temperature range [3–9] at least for simple molecules. The situation for liquids is markedly different as the underlying theory is incomplete. This state of affairs is particularly acute for liquid viscosity which exhibits a rapid increase as one approaches the solidification line, thus putting severe constraints on the accuracy with which the viscosity can be predicted, as any proposed prediction model will be very sensitive to the value of the input parameters, primarily temperature and density. The lack of underlying theory has led to a number of different approximate approaches that are based on kinetic theory [10–14], corresponding states theory [15–22], free-volume concept [23–26], friction theory [27–31], relationship with residual entropy [32–35], density scaling [36–38], effective carbon number approach [39,40] and the expanded fluid based approach [41–43] among others. The plethora of predictive models is useful for practitioners, but we are still lacking a comprehensive comparison against reliable experimental data in order to ascertain the accuracy and the range of validity of different models. Furthermore, most of the models are designed for compositionally well-characterized mixtures where accurate compositional data is used as one of the inputs.

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However, as we discussed previously [44] for quite a few fluids of industrial interest (*e.g.* oil, heavier refinery fractions, coal liquids, pharmaceutical) that may contain a large number of components, isomeric species or ill-defined large species, this is not the case. In this instance a different formulation is required that forfeits the full compositional formulation and usually relies on describing the mixture in terms of pseudo-components. The objective of this work is to provide the basis for such a model.

We have recently developed a one-component, extended hard-sphere (1-cEHS) model that possesses the right characteristics and have validated it by comparison with the experimental viscosity data for numerous liquid mixtures composed of *n*-alkanes [44]. The 1-cEHS model is based on the extended hard-sphere (EHS) model [14,45], that has its origins in the Assael and Dymond (AD) approach [46,47], where the viscosity of a fluid is represented by a universal function, by means of two scaling parameters. In the 1-cEHS model [44] the viscosity of the multicomponent fluid is represented by a viscosity of a single pseudo-component which is characterized by an appropriate molecular weight. Thus, eliminating the need for a full compositional characterization of the fluid and consequently the need for mixing rules, an ubiquitous feature of thermophysical property models. As pointed out previously [44], we use the term pseudo-component not to indicate a real component, but one that is used to simplify the compositional description of the mixture and hence the concept may differ from how it is used in other fields.

In this work we have extended the 1-cEHS model to include species from other chemical families, namely alkylbenzenes and cycloalkanes, in addition to *n*-alkanes. The choice of alkylbenzenes and cycloalkanes was primarily driven by the need for developing models to predict the viscosity of fluids encountered in the petrochemical and chemical industries. We have tested the proposed model by validating it against a large data set consisting of viscosity of relevant liquid mixtures and have compared its performance to that of the EHS model that requires mixing rules and a knowledge of the composition of the mixture. We do not expect the 1-cEHS model to be more accurate than the EHS model or

other similar models. By replacing the full compositional characterization by what will turn out to be essentially an effective molecular weight of the mixture, we have sacrificed accuracy for versatility. Hence the validation against the binary and subsequently multicomponent mixtures is to validate that the proposed model is reliable and of reasonable accuracy. Its ability to predict the viscosity of liquid mixtures that are not well compositionally characterized, is where we expect the model to come into its own.

In section 2 we summarize the modifications required to use the 1-cEHS model for alkylbenzene and cycloalkane species. We complete the section by providing the new EHS scaling parameters for a number of fluids belonging to the two chemical families. In section 3 we compare the two models (1-cEHS and EHS) in their ability to predict the viscosity of binary and multicomponent liquid mixtures consisting of n-alkane, alkylbenzene and cycloalkane species.

2. Methodology

In this work we take the recently proposed 1-cEHS model [44] as our starting point. The essence of the 1-cEHS model is to make use of a universal function, developed originally by Assael and Dymond [46,47] and subsequently modified [14,45], that correlates the reduced excess viscosity $\Delta\eta^*$ as a function of the reduced molar volume, V^*,

$$\log_{10}(1 + \Delta\eta^*) = \sum_{i=1}^{7} a_i/(V^*)^i \quad , \tag{1}$$

where the reduced molar volume ($V^* = V_m/V_0$) is the ratio of molar volume, V_m and molar core volume, V_0. The reduced excess viscosity, $\Delta\eta^*$, is defined as [46],

$$\Delta\eta^* \equiv \frac{16}{5}(2N_A)^{1/3}\left(\frac{\pi}{MRT}\right)^{1/2} V_m^{2/3}\left(\frac{\eta - \eta^{(0)}}{R_\eta}\right), \tag{2}$$

where M is the molar mass, N_A is Avogadro's constant, R is the gas constant, R_η is the roughness factor and $\eta^{(0)}$ is the zero-density viscosity. In order to apply the scheme to real fluids R_η and temperature-dependent V_0, are used as scaling parameters. It has been shown [2,14,45,46] that with appropriate choice of the scaling parameters the viscosity of the plethora of pure fluids can be represented to within 5%. For *n*-alkanes the scaling parameters are smooth functions of temperature, T, and carbon number, n, and it is possible to express them by means of empirical correlations. For instance, the molar core volume, V_0, for *n*-hexane to *n*-tetratetracontane is given by [46],

$$\begin{aligned} V_o\,(cm^3.mol^{-1}) =\ & 117.874 + 0.15(-1)^n - 0.25275\,T + 0.000548\,T^2 \\ & -4.246 \times 10^{-7}T^3 + (n-6)(1.27 - 0.0009\,T)(13.27 + 0.025\,n) \end{aligned} \tag{3}$$

where temperature is in degrees Kelvin. The expressions for V_0 for lighter *n*-alkanes and for roughness factor are available in literature [46]. In order to use the universal correlation to calculate the viscosity of a mixture, the original models made use of mixing rules for the scaling parameters. Although the mixing rules worked reasonably well when the species belonged to the same homologous family, there are indications [2,48] that when this is not the case or when the mixture contains species very different in size, the standard EHS model has difficulty in accurately predicting the viscosity. The recently developed 1-cEHS

model [44] drops the mixing rule requirement by postulating that what distinguishes one *n*-alkane molecule from another, as far as the prediction of the liquid viscosity is concerned, is only the molecular weight. Hence, the 1-cEHS model evaluates the mixture viscosity of *n*-alkanes using V_0 and R_η expressions developed for pure *n*-alkanes and replacing the molecular weight of a pure species by the molecular weight of the mixture. In order to extend the 1-cEHS model to other fluid families, we first examine the viscosity of pure fluids.

2.1 Alkylbenzenes

Assael and co-workers [49] were first to propose a correlation for V_o and a set of R_η for alkylbenzenes. We extend their work by including more recent data and alkylbenzenes larger than 1,3,5-trimethylbenzene. Table 1 summarizes the pseudo-experimental and experimental data that were used in developing the scaling parameters. For benzene, toluene, ethylbenzene and xylene isomers we have used the recommended viscosity correlations [50–55], to generate the pseudo-experimental input data, while for other alkylbenzene fluids we made use of the available literature experimental data. We have limited ourselves to the liquid region for each fluid, with temperature range limited to below 400 K, in order to stay within the range of the validity of the 1-cEHS model. As the methodology of finding the optimal scaling parameters is very sensitive to the values of density used, we have either used the experimental density provided by the authors or we have calculated it from the recommended EoS for each fluid by using Refprop [56]. Our analysis indicates that the molar core volume, V_0, for alkylbenzenes can be represented by

$$
\begin{aligned}
V_o(cm^3 \cdot mol^{-1}) = \ & 983.47565n^{-2} - 259.33764n^{-1} + 14.65385n \\
& + T(0.21932n^{-1} - 0.0189n + 8.118 \times 10^{-4}n^2) \\
& + T^2(-1.903 \times 10^{-4} + 5.378 \times 10^{-5}n - 2.917 \times 10^{-6}n^2)
\end{aligned}
\tag{4}
$$

while the roughness factor for each fluid is given in Table 1. We concur with the observation made by Assael and co-workers [49] that although the roughness factor of alkylbenzenes in general increases with the carbon number, no simple correlation is possible without

compromising the accuracy of the model. Figure 1 and Table 1 indicate that with this choice of the scaling parameters it is possible to correlate most of the data to within 5%, with a maximum deviation of -6.5%.

Table 1: Database used for determining the close-packing volume V_o and roughness factor R_η of alkylbenzenes.

Name	$T_{min} - T_{max}$, K	$P_{min} - P_{max}$, MPa	R_η	AAD [a], %	MD [b], %
Benzene [50]	280 – 400	$P_{sat.}$ - 70	0.984	0.3	0.9
Toluene [51]	270 – 400	$P_{sat.}$ - 60	1.027	0.6	-2.0
Ethylbenzene [52]	270 – 400	$P_{sat.}$ - 60	1.028	1.0	2.7
p-Xylene [52,53]	290 – 400	$P_{sat.}$ - 70	1.041	0.7	1.8
o-Xylene [54]	280 – 400	$P_{sat.}$ - 70	1.054	1.2	-3.8
m-Xylene [55]	280 – 400	$P_{sat.}$ - 70	0.989	1.1	-2.0
n-Propylbenzene [57–63]	273 – 373	0.1	1.150	1.6	6.4
1,3,5-Trimethylbenzene [64,65]	298 – 393	0.1 - 52	0.932	0.9	-4.1
n-Butylbenzene [58–60,66]	273 – 373	0.1	1.158	3.3	-6.5
n-Pentylbenzene [59,66]	288 – 303	0.1	1.321	2.1	-4.3
n-Hexylbenzene [67]	293 – 373	0.1	1.413	1.1	-3.4
n-Heptylbenzene [59,68]	293 – 353	0.1 - 100	1.555	1.6	4.7
n-Octylbenzene [67]	293 – 373	0.1	1.678	1.1	-3.7
n-Nonylbenzene [69]	313 – 353	0.1 - 40	1.913	2.2	5.3
n-Dodecylbenzene [67]	293 – 373	0.1	2.542	1.3	-2.2

$P_{sat.}$ – Saturation pressure. [a] Average Absolute Deviation = $(100/N) \sum |(\eta_{cal} - \eta_{exp})/\eta_{exp}|$. [b] Max Deviation
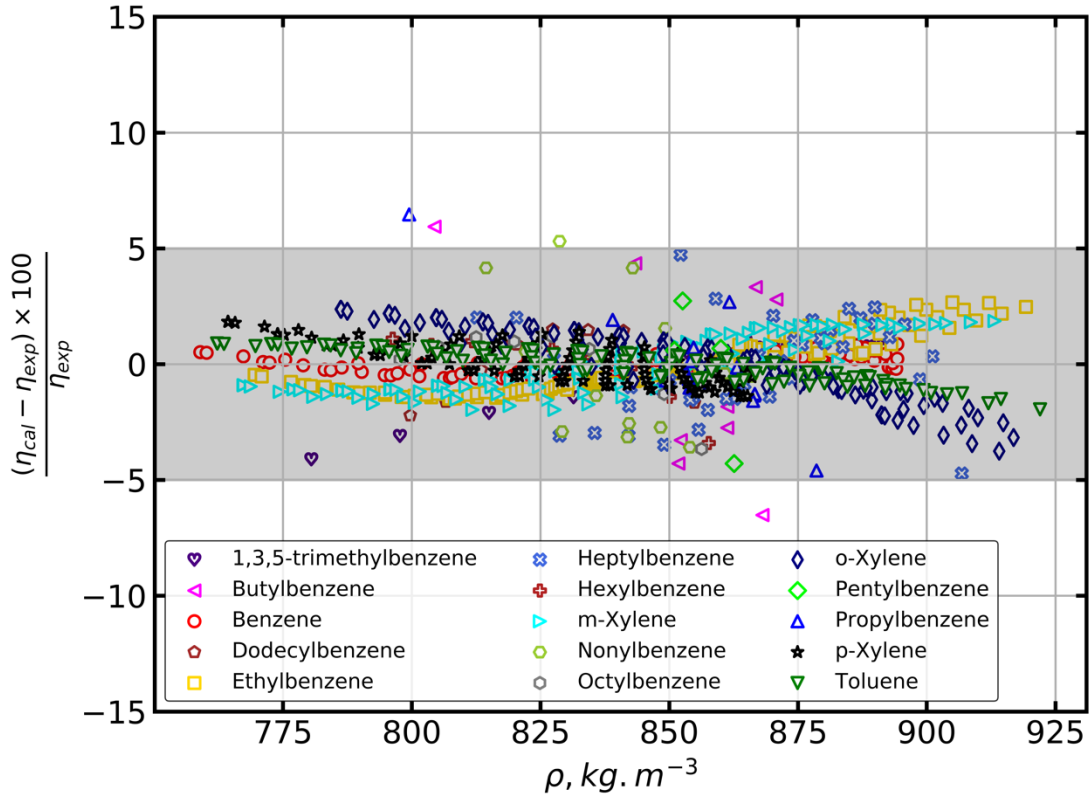
Figure 1: Percentage deviations $[100(\eta_{cal} - \eta_{exp})/\eta_{exp}]$ of the experimental data for alkylbenzenes from the calculated values, by means of EHS model, as a function of density.

2.2 Cycloalkanes

To the best of our knowledge, there is no correlation for the scaling parameters V_o and R_η for cycloalkanes, apart from cyclohexane [70]. In this work, primarily driven by the availability of viscosity data, we have examined cycloalkanes belonging to cyclohexane series and cyclooctane. Table 2 summarizes the experimental and pseudo-experimental data that were used in the analysis.

Table 2: Database used for determining the close-packing volume V_o and roughness factor R_h of cycloalkanes

Name	$T_{min} - T_{max}$, K	$P_{min} - P_{max}$, MPa	R_η	AAD, %	MD, %
Cyclohexane [71]	298 − 398	P_{sat} - 75	0.920	0.9	-2.3
Methylcyclohexane [72,73]	293 − 353	0.1 - 60	1.000	1.6	-6.4
Ethylcyclohexane [74]	293 − 343	0.1	0.981	3.0	-5.6
Butylcyclohexane [75,76]	293 − 353	0.1	1.243	0.9	-3.1
Hetylcyclohexane [68]	293 − 353	0.1 - 60	1.782	1.9	-3.8
Cyclooctane [76–78]	283 − 353	0.1	1.167	2.0	-5.9

Based on the same procedure used for alkylbenzenes, we propose the following expression the molar core volume, V_o

$$V_o(cm^3 \cdot mol^{-1}) = 15.141049 + 9.205496\, n + \left(0.568402 - 0.02191 \times T^{0.5}\right) n^2 \tag{5}$$

where n is the real carbon number of cycloalkanes considered in this work. The roughness factor for each fluid is given in Table 2. Figure 2 shows the percentage deviation of the experimental data from the EHS correlation for cycloalkanes, while Table 2 summarizes the deviations in terms of AAD and MD. Most of the experimental data are predicted within 5%, with a maximum deviation of -6.4%.
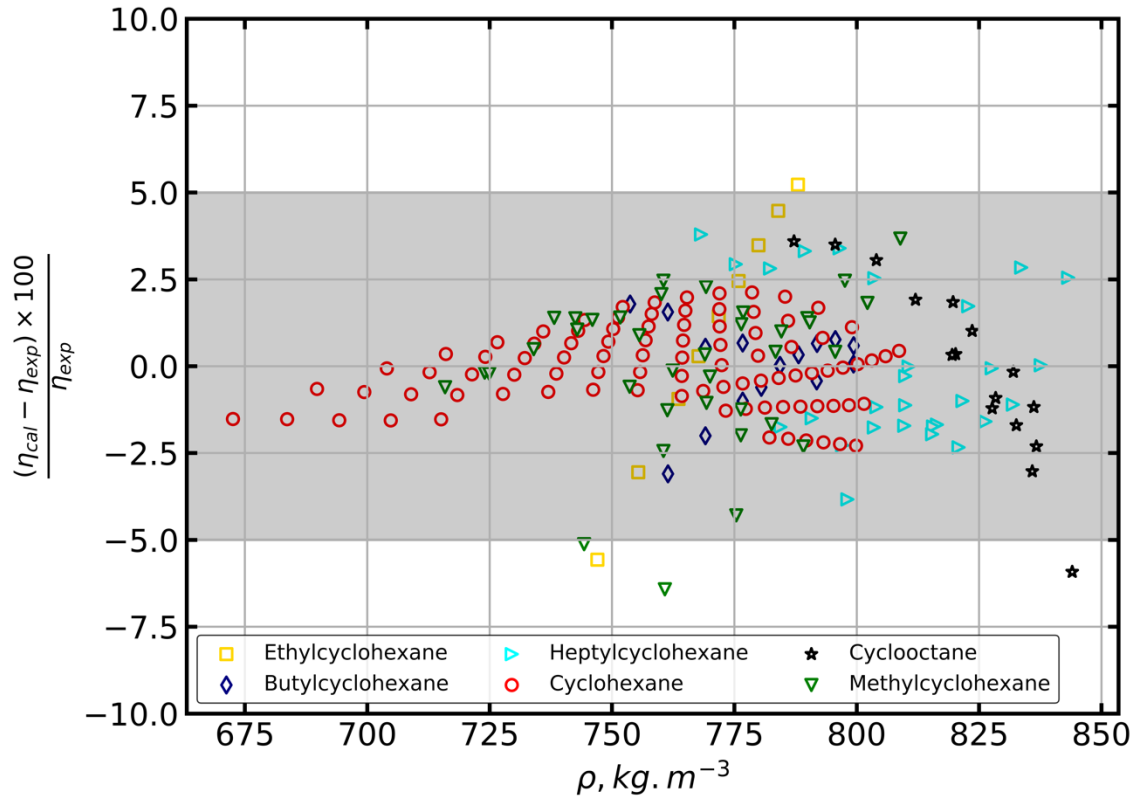
Figure 2: Percentage deviations $[100(\eta_{cal} - \eta_{exp})/\eta_{exp}]$ of the experimental data for cycloalkanes from the calculated values, by means of the EHS model, as a function of density.

2.3 Mixtures

In order to apply the developed models to calculate the viscosity of a mixture of a specified composition, one needs to know the value of the mixture scaling parameters V_o and R_η. In a traditional approach [2, 47] one would estimate the mixture parameters, from knowledge of pure species parameters, by the mole average mixing rules,

$$V_{o,mix} = \sum_{i=1}^{N} x_i V_{o,i} \qquad (6)$$

$$R_{\eta,mix} = \sum_{i=1}^{N} x_i R_{\eta,i} \; . \qquad (7)$$

In the rest of this work we refer to using these simple mixing rules, within the EHS scheme, to calculate the viscosity of a mixture, as the EHS model.

However, the main aim of this work was to extend the 1-cEHS model to evaluate the viscosity of mixtures involving n-alkanes, alkylbenzenes, and cycloalkanes. For this purpose, we need to develop a way of estimating the molar core volume of a mixture, V_o, by only knowing its molecular weight. One way of doing this is to treat non-alkane species in the mixture as pseudo-alkanes characterized by an effective molecular weight. We obtain the effective molecular weight by mapping V_o curves of alkylbenzenes and cycloalkanes onto the V_o curve of n-alkanes. In the process we intentionally sacrifice accuracy for versatility, the effect of which can be judged only at the validation stage. As a result of mapping, the effective molecular weight of alkylbenzenes $\widetilde{M}_\mathrm{aro}$ is given as,

$$\begin{aligned} \widetilde{M}_\mathrm{aro} = \ & -83.0383 + 0.6130T + (8.566 \times 10^{-3}T + 1.278 \times 10^{-3}M_\mathrm{aro})M_\mathrm{aro} \\ & +(15.1747 - 0.1363T)M_\mathrm{aro}^{0.5} - 9.9417 \times 10^{-6}TM_\mathrm{aro}^2 \end{aligned} \tag{8}$$

whereas the effective molecular weight of cycloalkanes series is given as,

$$\begin{aligned} \widetilde{M}_\mathrm{cyc} = \ & 15.6161259 + (0.62793565 + 0.00628002 \times T^{0.5})\,M_\mathrm{cyc} + \\ & (0.00010853 \times T^{0.5} - 3.6767 \times 10^{-6} \times T)\,M_\mathrm{cyc}^2 \end{aligned} \tag{9}$$

where M_aro and M_cyc are the real molecular weight of alkylbenzenes and cycloalkanes, respectively. For a general multicomponent mixture consisting of n-alkanes, alkylbenzenes, and cycloalkanes we calculate the effective molecular weight of a mixture, $\widetilde{M}_\mathrm{mix}$, as a simple mole average of effective molecular weight of each species, $\widetilde{M}_\mathrm{mix} = \sum_i^N x_i\widetilde{M}_i$, bearing in mind that in this scheme the effective molecular weight of n-alkane species is the same as its real molecular weight. The molar core volume of a mixture, $V_\mathrm{o,mix}$ can then be estimated from Eq. (3) by converting the effective molecular weight of the mixture $\widetilde{M}_\mathrm{mix}$ to the carbon number, $n = (\widetilde{M}_\mathrm{mix} - 2.01588)/14.02658$.

For the evaluation of the roughness factor R_η, we propose a slight modification of the expression already developed for alkane mixtures [44], namely the introduction of the term

$\left(\frac{\widetilde{M}_{\text{mix}}-\beta}{M}\right)$ that takes into account the presence of non *n*-alkane species. The new expressions read as follows,

$$
\begin{aligned}
R_\eta &= (0.995 - 0.0008944n_{\text{av}} + 0.005427n_{\text{av}}^2)\left(\frac{M_{\text{av}}}{M}\right)^{0.15}\left(\frac{\widetilde{M}_{\text{mix}}-17\,x_{\text{cyc}}^{0.6}}{M}\right) \quad n_{\text{av}} < 17 \\
R_\eta &= (2.6380 + 0.157(n_{\text{av}}-18))\left(\frac{M_{\text{av}}}{M}\right)^{0.15}\left(\frac{\widetilde{M}_{\text{mix}}-17\,x_{\text{cyc}}^{0.6}}{M}\right) \qquad\qquad n_{\text{av}} \geq 17
\end{aligned}
\tag{10}
$$

where M_{av} is defined as in our previous paper [44], $M_{\text{av}} = \left(\sum_i^N x_i M_i^\alpha\right)^{1/\alpha}$ and M is the real molecular weight of the mixture. The interested reader can find the definition of parameter α in the same paper. The parameter n_{av} is calculated from the weighted average molecular weight, M_{av}, by using a standard relationship between the number of carbon atoms and the molecular weight of *n*-alkane, $n_{\text{av}} = (M_{\text{av}} - 2.01588)/14.02658$.

As it stands the 1-cEHS model still requires knowledge of the composition in order to evaluate the value of M_{av}, as defined above. For compositionally complex fluid mixtures, encountered in industrial applications, full specification will not be available. To make the model applicable in such cases we characterize the mixture in terms of the chemical families present and for the purposes of this work in terms of total amount of *n*-alkane, alkylbenzene and cycloalkane species. In the terminology of the petroleum industry, we create a three component pseudo-mixture consisting of alkane, aromatic and cyclic pseudo components; each defined by its mole fraction ($x_{\text{alk.}}$, x_{aro} or $x_{\text{cyc.}}$) and its molecular weight (M_{alk}, M_{aro} or M_{cyc}). The M_{av} is then estimated from,

$$
M_{\text{av}} = \left(x_{\text{alk}}M_{\text{alk}}^\alpha + x_{\text{aro}}M_{\text{aro}}^\alpha + x_{\text{cyc}}M_{cyc}^\alpha\right)^{1/\alpha}
\tag{11}
$$

In the rest of the work we will refer to this model as 3-cEHS. We only propose to use this model for the prediction of the viscosity of multicomponent liquid mixtures which consist of species belonging to different families.

3. Results and Discussion

In this section, we evaluate the two new models (1-cEHS and 3-cEHS) by validating them against extensive data on the viscosity of mixtures consisting of *n*-alkane, alkylbenzene and cycloalkane species. The evaluation is initially performed using the data sets that pertain to binary mixtures, as such data are most plentiful. This is followed up with a validation using viscosity data of compositionally well-defined multicomponent mixtures. Such an analysis is carried out in order to test the versatility and reliability of the proposed model and to estimate the uncertainty limits of the predicted viscosity values. However, it should be emphasized here that our primary future aim is to use the developed models to predict the viscosity of compositionally ill-defined multicomponent mixtures.

3.1 Binary alkylbenzene mixtures

We have collected the data on 28 binary alkylbenzene mixtures measured in a variety of viscometers with the claimed uncertainty ranging from 0.1% to 0.4%. The available data set consists of approximately 440 data points covering a range of temperature (273 - 318 K) at atmospheric pressure. The following alkylbenzene species are present: benzene, toluene, m-, o- , p-xylene, ethylbenzene, *iso*-propylbenzene (cumene) and *tert*-butylbenzene. Figure 3 shows the deviations in predicting the viscosity of available binary alkylbenzene mixtures by means of the 1-cEHS model, while Table 3 summarizes the AAD and MD by both the 1-cEHS and EHS model.
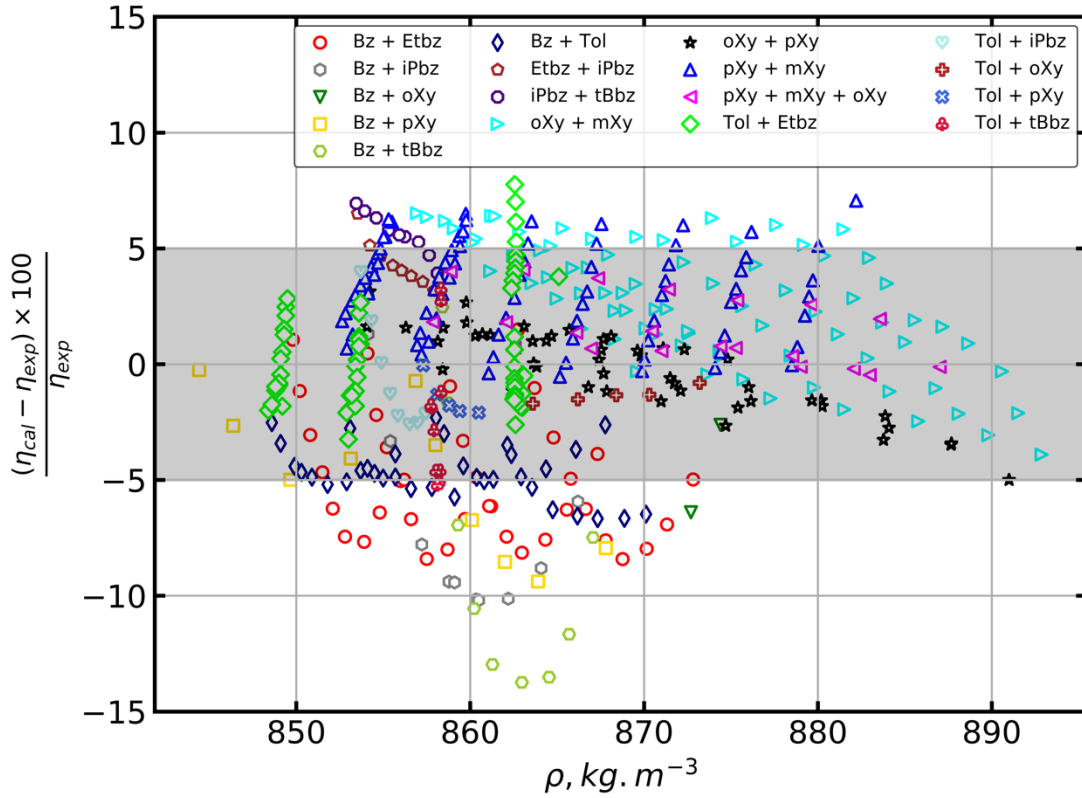
Figure 3: Percentage deviations $[100(\eta_{cal} - \eta_{exp})/\eta_{exp}]$ of the experimental viscosity data for binary, alkylbenzene mixtures from the calculated values, by means of the 1-cEHS model, as a function of density. See Table 3 for the legend description.

Table 3: Comparison of the prediction of the viscosity of binary, alkylbenzene mixtures by using the 1-cEHS and EHS models. All the experimental data are measured at 0.1 MPa

Binary mixtures containing	No of data	$T_{min} - T_{max}$, K	1-cEHS		EHS	
			AAD, %	MD, %	AAD, %	MD, %
Benzene (Bz)	84	298 – 318	4.9	-9.4	0.8	5.4
Toluene (Tol) and/or xylenes (Xy)	301	273 – 313	2.5	7.8	1.1	5.9
i-Propylbenzene (iPbz) and/or						
t-Butylbenzne (tBbz)	55	303 – 303	5.4	-13.7	-	-

We observe that the 1-cEHS model has particular difficulties in predicting the viscosity of mixtures containing benzene [79–83]. The overall AAD is 4.9% with maximum

deviation of -9.4%. As illustrated in Figure 3 the deviations increase with an asymmetry of the mixture. The effective molecular weight of benzene is 78.11 g.mol^{-1} which corresponds to roughly pentane among n-alkanes. The uncertainty in n-alkanes V_0 function is highest between n-butane and n-hexane and this translates into poor prediction for mixtures containing benzene, whose effective molecular weight is below that of n-hexane. We observed a similar deterioration in n-alkane binary mixtures involving n-pentane [44]. The EHS model predicts the mixtures containing benzene with a good accuracy with the resulting AAD and MD of 0.8% and 5.4%, respectively.

The 1-cEHS model predicts the viscosity of the mixtures containing toluene and/or xylene isomers [80,81,84–87] with acceptable AAD of 2.5% and MD of 7.8%. Nevertheless the predictions of the EHS model are marginally better, on a par with predictions for benzene containing mixtures, see Table 3. For *iso*-propylbenzene and *tert*-butylbenzene the literature viscosity data are scarce and it is not possible to get the values of V_0 and R_η; consequently, it is not possible to predict the viscosity of mixtures containing either of these two species by the EHS model. For the 1-cEHS model, that makes use of the effective molecular weight and relies on the n-alkane expressions for V_0 and R_η, such a calculation is feasible. The 1-cEHS model predicts the viscosity [80] with AAD of 5.4% and MD of -13.7%, see Table 3, thus illustrating the usefulness and versatility of the model when the viscosity of pure species is not known. The worst prediction is observed for benzene - *tert*-butylbenzene mixture where the 1-cEHS model underestimates the measured values by 7.0 to 13.7%. Some of the difficulties with predicting this system can be ascribed to the already noted difficulties in predicting the viscosity of mixtures containing benzene. Without considering this system the AAD for remaining mixtures reduces to 3.9 % and MD to -5.2 %.

We note that the 1-cEHS model predicts the viscosity of ternary aromatic mixtures with good accuracy, as illustrated in Figure 3 for ternary mixtures containing xylene isomers (m-, o-, p-xylene [86]) the only ternary aromatic mixture for which experimental results are available in open literature.

3.2 Binary n-alkane-alkylbenzene mixtures

The ability of the 1-cEHS and EHS models to accurately predict the viscosity was also tested using the available experimental data for binary mixtures consisting of alkylbenzenes and *n*-alkane species. The data set consisted of approximately 1687 data points measured by 14 investigators and covering a temperature range of (283.15 – 393.20 K). Most of the data were measured at atmospheric pressure, with the exception of the data reported by; (i) Dymond and Young [87] for mixtures of benzene with *n*-hexane, *n*-octane, *n*-decane, *n*-dodecane and *n*-hexadecane at saturation pressure; (ii) Dymond *et al.* [88] for mixtures of toluene with *n*-hexane at pressure up to 510 MPa; (iii) Kanti *et al.* [69] for mixtures of nonylbenzene with *n*-heptane at pressure up to 40 MPa. The choice of the data was governed by our analysis of the experimental data for *n*-alkane mixtures [81,84,89–93] from the same labs and by comparison of the data between different workers.

Figure 4 illustrates the deviations in predicting the viscosity by means of the 1-cEHS model, while Table 4 summarizes the AAD and MD for both the 1-cEHS and EHS models.
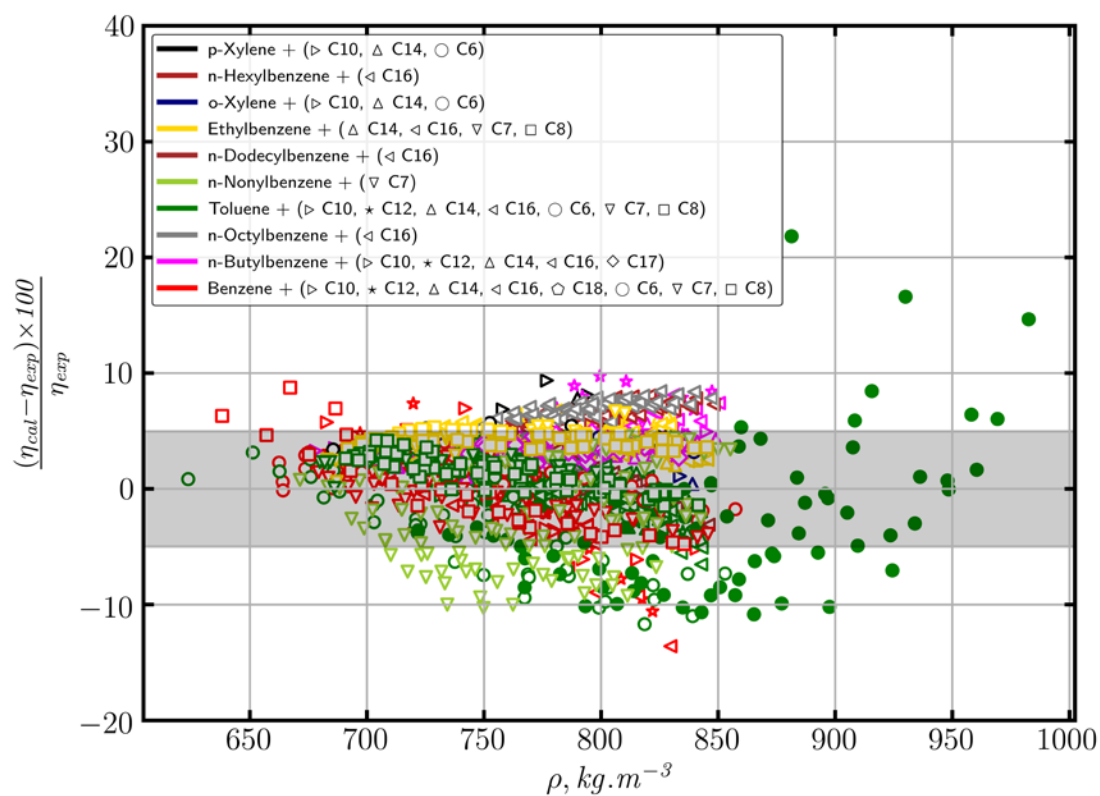
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Figure 4: Percentage deviations $[100(\eta_{cal} - \eta_{exp})/\eta_{exp}]$ of the experimental viscosity data for binary, *n*-alkane + alkylbenzene mixtures from the calculated values by means of the 1-cEHS model as a function of density. The solid symbols represent the experimental data at pressures over 75 MPa.

Table 4: Comparison of the prediction of the viscosity of binary, *n*-alkane + alkylbenzene mixtures by using the 1-cEHS and EHS models. For *n*-alkane species present see **Figure 4** legend.

Alkylbenzene	No of data	$T_{min} - T_{max}$, K	1-cEHS		EHS	
			AAD, %	MD, %	AAD, %	MD, %
at pressure of 0.1 MPa						
Benzene [81,89,90,94–98]	115	288 – 342	1.8	7.0	4.9	13.7
Toluene [79,81,88,99–102]	374	293 – 373	1.7	-6.5	4.6	10.9
Ethylbenzene [79,84,102,103]	245	293 – 373	3.9	6.9	4.0	9.4
p, m, o-Xylenes [81]	30	298 – 298	4.2	9.4	5.3	9.0
Butylbenzene [104]	372	293 – 373	3.8	9.7	1.9	6.0
Hexylbenzene [105]	81	293 – 373	5.5	8.1	3.1	4.5
Octylbenzene [105]	81	293 – 373	5.9	8.4	3.0	4.5
Nonylbenzene [69]	21	313 – 353	2.1	4.6	1.3	2.5
Dodecylbenzene [105]	81	293 – 373	1.9	4.3	2.9	4.5
at saturation pressure						
Benzene [87]	95	283 – 393	3.0	-13.6	4.3	10.0
at pressure up to 75 MPa						
Toluene [88]	35	298 – 373	5.4	-11.7	4.6	-10.5
Nonylbenzene [69]	84	313 – 353	4.5	-10.3	5.3	-10.3
at pressure from 77 up to 510 MPa						
Toluene [88]	73	293 – 373	5.5	21.8	5.2	22.7

As illustrated in Table 4, for all the analysed mixtures, the prediction of the 1-cEHS model of the viscosity measured at atmospheric pressure is within 10% of the experimental values. It is reassuring to see that the 1-cEHS model retains its accuracy as the pressure increases; the high pressure viscosity data of Dymond *et al.* (up to 75 MPa) [88] and of Kanti *et al.* (up to 40 MPa) [69] are predicted within AAD of 4.8% and MD of -11.7%. At pressure higher than 75 MPa, see Table 4 and Figure 4, we observe systematic increasing deviations with respect to measured Dymond *et al.* viscosity data (solid symbols on Figure 4). This is in line with what was reported for *n*-alkane mixtures previously [44], as has been observed that as the pressure increases above 30–75 MPa, the viscosity prediction based on the general expression for V_0, deteriorates. In summary, at pressure up to 75 MPa the 1-cEHS model predicts the experimental data with an overall AAD of 3.4%, and an MD of -13.6%. The maximum deviation was observed for the most asymmetric mixture studied (*n*-hexadecane + benzene [87]) at the saturation pressure, at the lowest temperature measured, 298.2 K.

The predictions obtained by the EHS model are overall comparable to the results obtained by the 1-cEHS model. It is interesting to note that based on the available experimental data, the 1-cEHS model gave more accurate predictions for mixtures involving small alkylbenzenes (*i.e.* benzene, toluene, ethylbenzene) and high asymmetric mixtures (*i.e.* toluene + *n*-tetradecane, toluene + *n*-hexadecane) than the EHS model. On the latter point, we observe that as the asymmetry of the binary mixture increases the EHS model tends to overpredict the viscosity data with increasing deviations. No such trend is found with the predictions of the 1-cEHS model. As an illustration, Figure 5 shows the comparison of the predictive performance of two models for the *n*-tetradecane + toluene mixture where asymmetry, measured by ΔC, is 7.
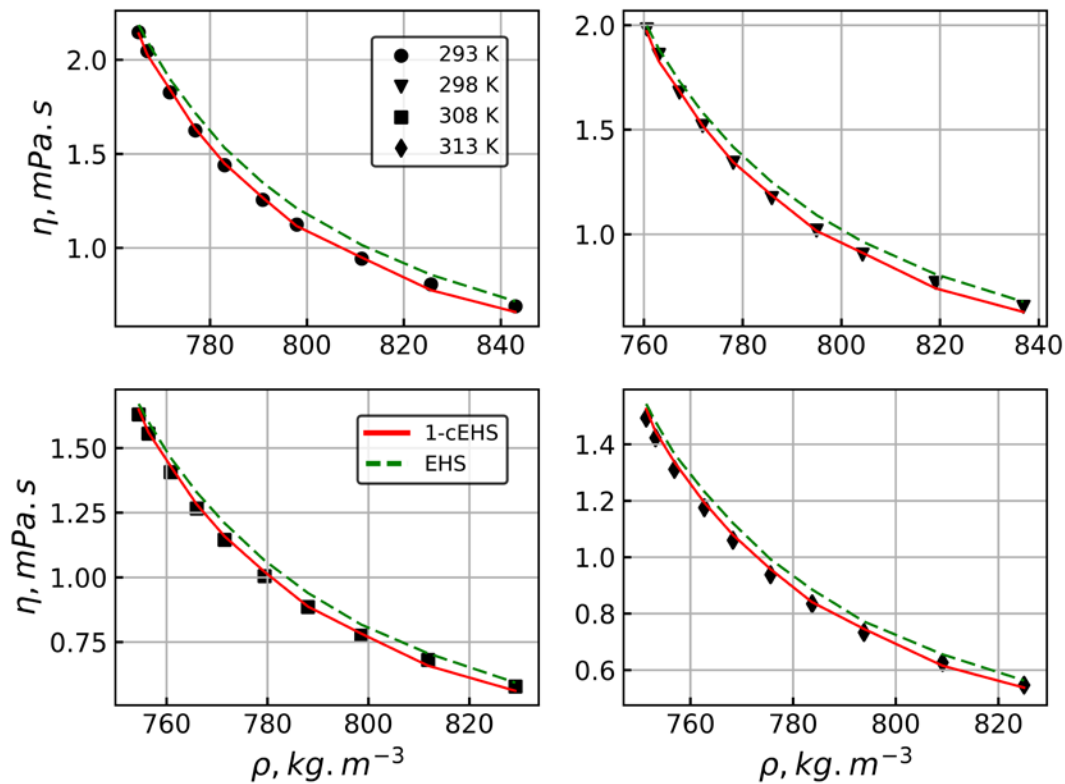

Figure 5: Comparison of the viscosity *n*-tetradecane + toluene mixture as a function of density predicted by the 1-cEHS and EHS models and the measured data [106].

3.3 Binary n-alkane + cycloalkane mixtures

Evaluation of the two models was also performed using collected experimental viscosity data on binary mixtures consisting of cycloalkane and *n*-alkane species. The data set consisted of approximately 970 data points measured by 14 investigators and covering a temperature range of (288.27 – 353.15 K). Most of the data were measured at atmospheric pressure, with the exception of the data reported by; (i) Isdale *et al.* [107] for (cyclohexane + *n*-hexane) mixture at pressures up to 50 MPa; (ii) Baylaucq *et al.* [72] for (methylcyclohexane + *n*-heptane) mixture at pressures up to 40 MPa; (iii) Tanaka *et al.* [108] for (cyclohexane + *n*-octane) mixture at pressures up to 71.3 MPa. Figure 6 illustrates the deviations in predicting the viscosity by means of the 1-cEHS model, while Table 5 summarizes the AAD and MD for both the 1-cEHS and EHS models. No results are given for the EHS model for mixtures containing 1,2,4-trimethylcyclohexane and 1,2-dimethylcyclohexane as there are insufficient viscosity data in literature to determine the pure species V_0 and R_η for these two fluids.
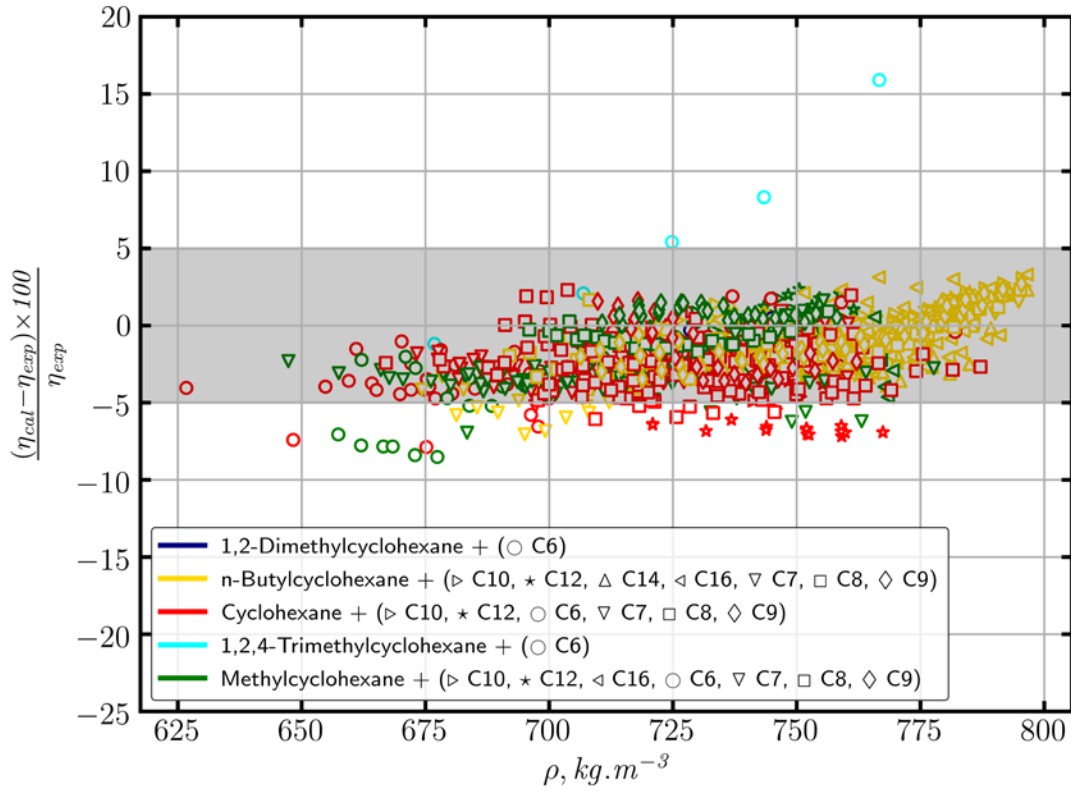
Figure 6: Percentage deviations $[100(\eta_{cal} - \eta_{exp})/\eta_{exp}]$ of the experimental viscosity data for binary, *n*-alkane + cycloalkane mixtures from the calculated values by means of the 1-cEHS model as a function of density.

Table 5: Comparison of the prediction of the viscosity of binary, *n*-alkane + cycloalkane mixtures by using the 1-cEHS and EHS models. For *n*-alkane species present see Figure 6 legend.

Cycloalkane	No of data	$T_{min} - T_{max}$, K	1-cEHS		EHS	
			AAD, %	MD, %	AAD, %	MD, %
At pressure of 0.1 MPa						
Cyclohexane [84,90,107–113]	259	288 – 348	2.7	-7.9	1.3	-5.3
Methyl cyclohexane [72,81,112,114]	207	298 – 343	1.9	-8.5	2.2	-4.7
Butyl cyclohexane [75,115]	393	293 – 353	1.6	-7.1	0.8	-4.9
1,2-Dimethyl cyclohexane [81]	3	298	0.6	-1.1	-	-
1,2,4-Trimethyl cyclohexane [81]	5	298	6.7	15.9	-	-
At pressure upto 73 MPa						
Cyclohexane [108]	61	298 – 348	3.4	-6.1	1.0	5.2
Methyl cyclohexane [72]	42	303 – 343	3.5	-6.2	1.0	-4.3

In general, the overall AAD of the predictions given by the 1-cEHS model, excluding

the (1,2,4-trimethylcyclohexane and *n*-hexane) and (1,2-dimethylcyclohexane and *n*-hexane)

mixtures, is 2.2% and the maximum deviation is -8.5%. The EHS model gives marginally better predictions. Most of the data are predicted to within 10%, in agreement with what we observed for n-alkane-alkylbenzene mixtures. For a mixture of 1,2,4-trimethylcyclohexane and n-hexane, measured by Chevalier *et al.* [81], we observe a systematic increase in deviations, as a function of density or as a function of mole fraction of 1,2,4-trimethylcyclohexane, leading to a maximum deviation of 15.9%, as illustrated in Figure 6. The cause of this behaviour is not currently clear. The deviations may be caused by the inclusion of the cyclic species which is a multi-branched cyclohexane, or by the fact that this molecule is not included in the procedure for determining the close-packing volume of the cyclohexane series. Nevertheless, we observe very good agreement for 1,2-dimethylcyclohexane and n-hexane mixture measured by the same authors in the same viscometer; 1,2-dimethylcyclohexane is only a slightly simpler molecule, and also it was not used in determining the close-packing volume.

We also observe, see Figure 6, that at low density the 1-cEHS model under predicts some of the measurements for the methylcyclohexane + n-hexane mixture, with a maximum deviation of 8.5%. These measurements were carried out at 298 and 303 K by Baragi *et al.* [114] for the mixtures lean in methylcyclohexane. Figure 7 illustrates the comparison of predicting the viscosity of the methylcyclohexane + n-hexane mixture measured by different authors at those two temperatures. Although all the authors indicate that their measurements are of low uncertainty, at low mole fractions of methylcyclohexane differences larger than 5% are observed. This further highlights the need to carry out a comprehensive analysis of the experimental measurements with a view to coming up with a primary data set that can be used for the purposes of model validation.
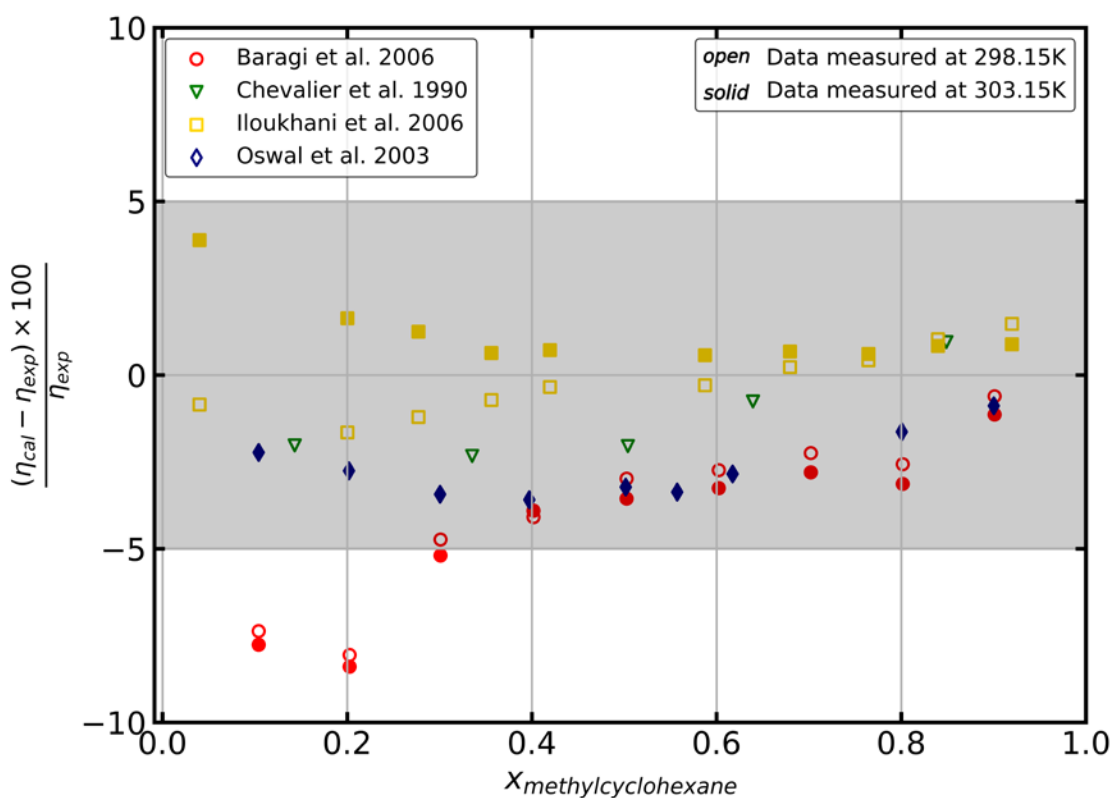
Figure 7: Percentage deviations $[100(\eta_{cal} - \eta_{exp})/\eta_{exp}]$ of the experimental viscosity data for binary, methylcyclohexane + *n*-hexane mixtures from the calculated values by means of the 1-cEHS model as a function of density. Measurements were performed by: ○ Baragi *et al.* [114] ; ▽ Chevalier *et al.* [81]; □ Iloukhani *et al.* [116] ; and ◇ Oswal *et al.* [112].

In the previous work [44] we have stressed the importance of getting accurate values of density in predicting the mixture viscosity, by any method that purports to predict the viscosity from a knowledge of temperature, density and the composition of the mixture. Here, we illustrate another example of this interdependency to further emphasize the point. Tanaka and co-workers have measured the viscosity of cyclohexane + *n*-dodecane mixture along three isotherms (298 K, 323 K and 348 K) to pressures up to 150 MPa, in their torsionally vibrating crystal viscometer which has been classified as a primary instrument by a number of workers who have been developing viscosity correlations. Figure 8 illustrates the deviation obtained by the 1-cEHS model from the experimental values by making use of the experimental density provided by Tanaka *et al.* [108] and the densities calculated form

Refprop [56]. We observe that for this system a change of 1% in density results in a change of up to 10% in predicted viscosity.
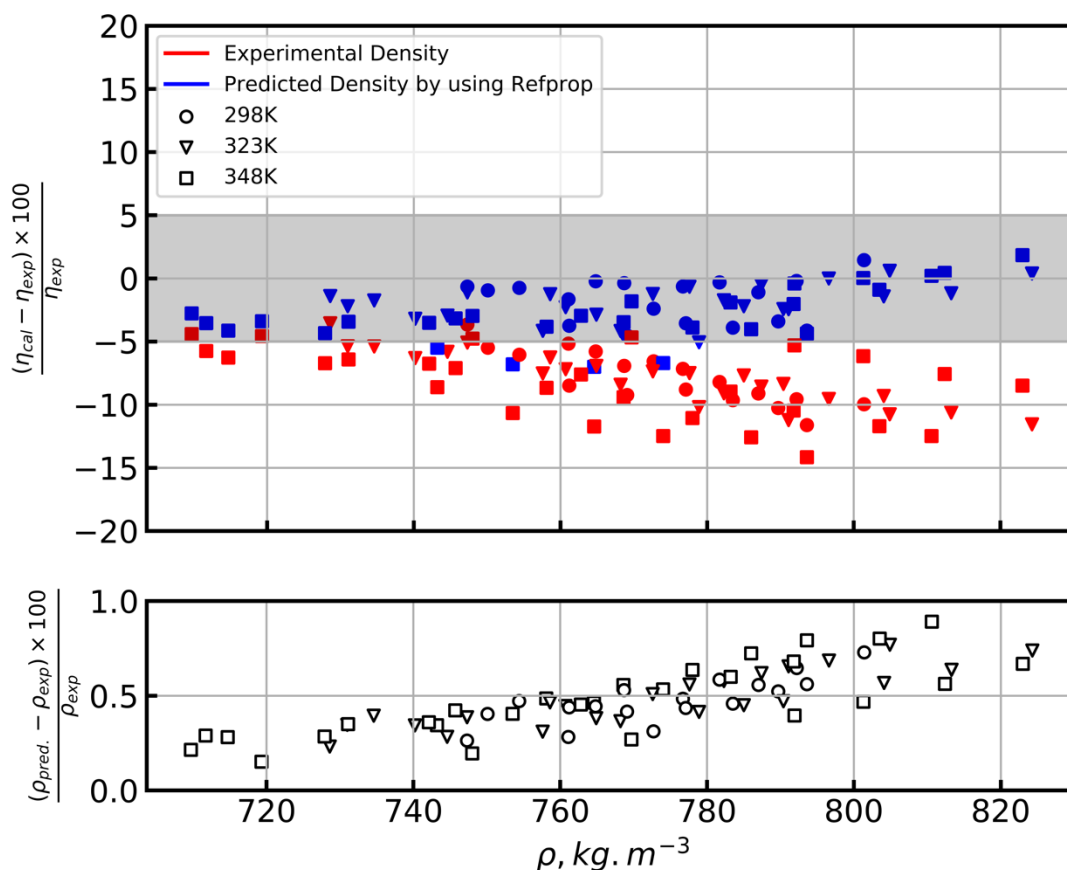

Figure 8: Sensitivity of the viscosity to the calculation of density for n-dodecane + cyclohexane mixture [108]. The density was predicted by using Refprop [56].

3.4 Binary alkylbenzene + cycloalkane mixtures

The final evaluation of the two models using binary mixture data was performed by examining the ability of the models to predict the experimental viscosity of alkylbenzene + cycloalkane systems. The collected data set is smaller than for previous systems and consists of approximately 367 data points measured by 10 investigators, covering a temperature range of (283.15 – 393.2 K) and pertaining to cyclohexane, methylcyclohexane and butylcyclohexane mixtures with benzene, toluene, ethylbenzene, isomers of xylene and propylbenzene. Most data were measured at atmospheric pressure, except the saturation pressure measurements for the cyclohexane + benzene mixture reported by Dymond and

Young [87]. Figure 9 illustrates the deviations in predicting the viscosity by means of the 1-cEHS model, while Table 6 summarizes the AAD and MD for both the 1-cEHS and EHS models.
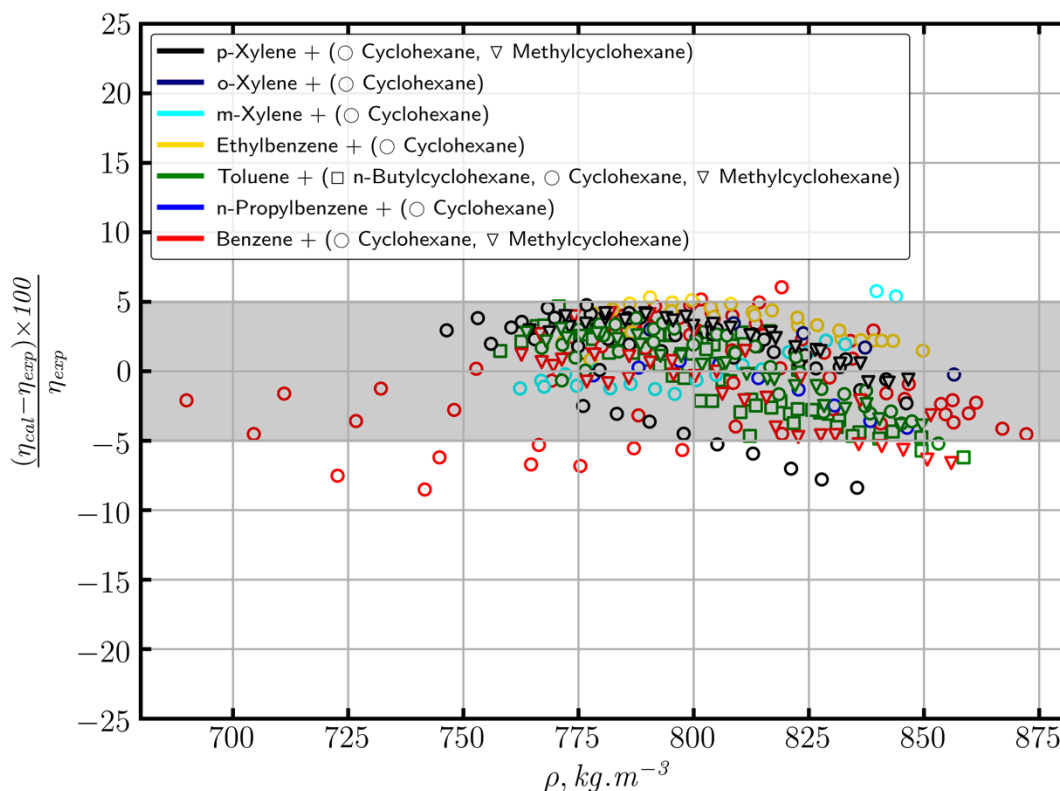

Figure 9: Percentage deviations $[100(\eta_{cal} - \eta_{exp})/\eta_{exp}]$ of the experimental viscosity data for binary, *n*-alkylbenzene + cycloalkane mixtures from the calculated values by means of the 1-cEHS model as a function of density.

Table 6: Comparison of the prediction of the viscosity of binary, alkylbenzene + cycloalkane mixtures by using the 1-cEHS and EHS models. For alkylbenzene species present see Figure 9 legend.

Cycloalkane	No of data	$T_{min} - T_{max}$, K	1-cEHS		EHS	
			AAD, %	MD, %	AAD, %	MD, %
Cyclohexane [81,84,85,87,112,113,117,118]	215	283 – 393	2.8	-8.5	4.5	12.4
Methylcyclohexane [112,114]	89	298 – 308	2.4	-6.6	7.0	14.5
Butylcyclohexane [75]	63	293 – 353	2.6	-6.2	4.1	8.2

In general, the 1-cEHS model gave an excellent prediction for these mixtures, with an overall AAD of 2.7% and a maximum deviation of -8.5%. Figure 9 illustrates that for the mixture of (benzene + cyclohexane) deviations higher than 5% are observed. This is in line with a discussion on alkylbenzene mixtures where the deviations of mixtures containing benzene species were attributed to the small effective molecular weight of benzene and higher uncertainty in evaluating parameters V_0 and R_η from the general n-alkane based correlations.

We also observe in Figure 9, that the prediction of the viscosity for the (p-xylene + cyclohexane) mixture, measured by Oswal et $al.$ (black circles) [112] gets progressively worse at high densities, that correspond to the higher mole fraction of p-xylene. The behaviour can be attributed to the uncertainty in the experimental density measured by Oswal et $al.$ for the mixtures containing high molar fraction of p-xylene. In Figure 10 we compare the viscosity deviations for the mixture measured by Oswal et $al.$ [112] with those measured by Yang et $al.$ [118], who measured the viscosity of the same mixture at the same conditions. It can be seen that the density of pure p-xylene measured by Oswal et $al.$ is smaller by about 1% than the one measured by Yang et $al.$, resulting in a difference in viscosity of approximately 9% between the two sets of workers. This further highlights the need, when validating different models, to not only use reliable and accurate values of density, but also to choose the validation dataset that is consistent, accurate and comes with reliable uncertainty estimates.

It is interesting to note, see Table 6, that for this family of mixtures the prediction of the EHS model is consistently worse than the prediction of the 1-cEHS model emphasizing the deficiency of the simple mixing rule.
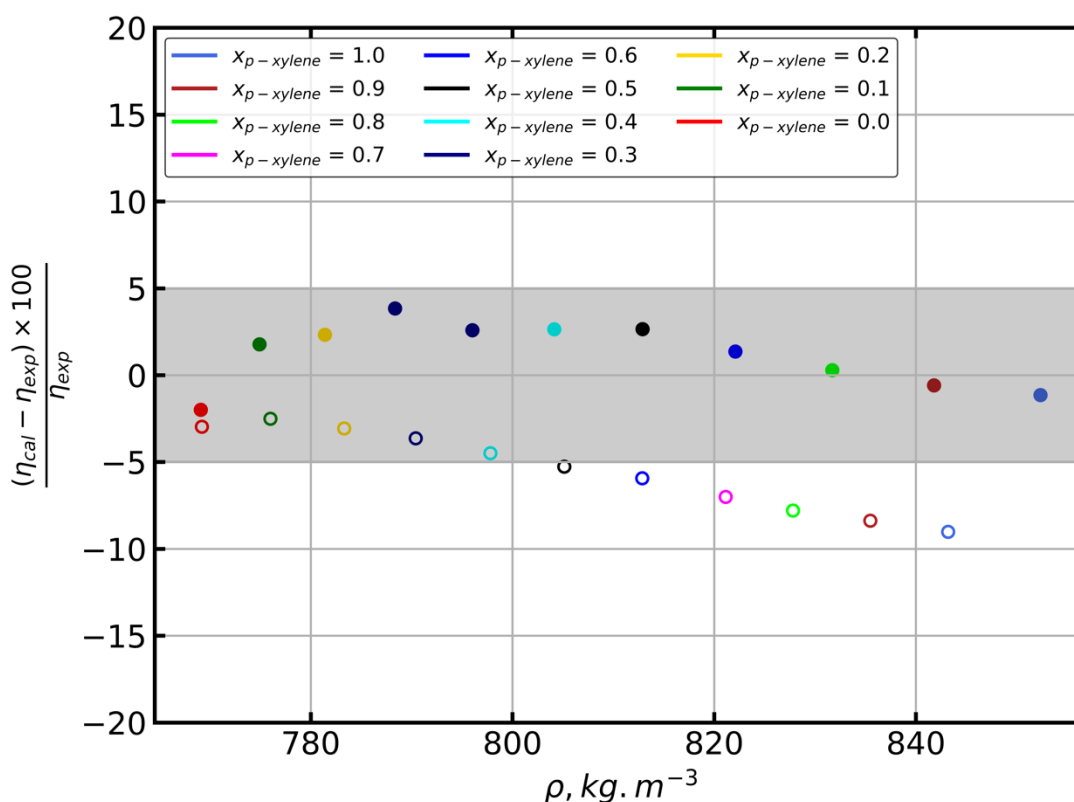
Figure 10: Percentage deviations $[100(\eta_{cal} - \eta_{exp})/\eta_{exp}]$ of the experimental viscosity data for cyclohexane + *p*-xylene mixture at 303 K and 0.1 MPa from the calculated values by means of the 1-cEHS model as a function of density. Solid symbols represent the data measured by Yang *et al.* [118] and open symbols by Oswal *et al.* [112]

3.5 Liquid multicomponent mixtures

The collected viscosity data pertaining to multicomponent mixtures consists of approximately 592 data points measured by 5 investigators. The data for ternary mixtures covers the temperature range of (293.15 − 353.15K), while the data on quaternary and quinary mixtures covers a narrow temperature range of (293.15 − 298.15K). All the data were measured at atmospheric pressure. Table 7 summarizes the experimental data and the prediction of the viscosity of multicomponent mixtures containing *n*-alkanes, alkylbenzenes and cyclohexane series by using the 1-cEHS, 3-cEHS, and EHS models. Figure 11, Figure 12 and Figure 13 illustrate the deviations in predicting the viscosity by means of 1-cEHS, 3-cEHS, EHS models, respectively. We observe that the 1-cEHS model provides good

estimates of viscosity with AAD of 2.8% and MD of 8.8%. In general, the deviations increase with density and based on a relatively limited experimental data set, decrease with the increase in the number of species present in the mixture. The latter behaviour has been observed for *n*-alkane mixtures [44] and bodes well for the application of the 1-cEHS model to predicting the viscosity of mixtures consisting of large number of species.

The data on the viscosity of multicomponent mixtures provides us with the opportunity of also testing for the first time the 3-cEHS model. The model groups alkanes, alkylbenzenes and cyclic alkanes into three pseudo component groups, respectively and as described in Section 2.3 makes use of Eq (11) to evaluate M_{av}. For ternary mixtures considered in this work the 3-cEHS model is equivalent to the 1-cEHS model and as expected the deviations, presented in Table 7 are the same for both models. For quaternary and quinary mixtures the 3-cEHS model predicts the experimental viscosity with AAD of 1.8 % and MD of -5.2% which is only marginally worse than the 1-cEHS model (AAD of 1.6% and MD of 5.1%). Here we also observe that the predictions improve as the number of components increases.

The traditional EHS model which makes use of mixing rules, predicts the viscosity of the mixtures listed in Table 7 with AAD of 3.8% and MD of 16.1%. Although the predictions of the viscosity of ternary mixtures is on a par with the 1-cEHS and 3-cEHS models, the deviations increase for quaternary and quinary mixtures.

Table 7: Comparison of the prediction of the viscosity of multicomponent mixtures containing *n*-alkanes, alkylbenzenes and cycloalkane species by using the 1-cEHS, 3-cEHS and EHS models.

Multicomponent mixtures	No of data	$T_{min} - T_{max}$, K	1-cEHS		3-cEHS		EHS	
			AAD, %	MD, %	AAD, %	MD, %	AAD, %	MD, %
Ternary mixtures								
C12 + ButylcC6 + ButylBz [119]	72	293 – 343	3.6	5.4	3.6	5.4	2.1	4.9
C12 + ButylcC6 + HexylBz [119]	72	293 – 343	5.1	8.8	5.1	8.8	2.6	4.3
C13 + HeptylcC6 + HeptylBz [119]	20	293 – 353	3.2	7.9	3.2	7.8	4.3	-6.9
C16 + ButylcC6 + Tol [119]	96	293 – 343	2.4	-7.8	2.4	-7.8	3.4	7.4
C16 + ButylcC6 + ButylBz [119]	72	293 – 343	2.5	5.8	2.5	5.8	1.7	3.7
C16 + ButylcC6 + HexylBz [119]	72	293 – 343	3.9	7.6	3.9	7.6	2.4	4.4
Quaternary mixtures								
C6 + C8 + cC6 + Tol [120]	16	293 – 298	1.7	-3.6	1.8	-3.8	3.8	6.7
C6 + C8 + cC6+ EtBz [120]	16	293 – 298	1.8	3.1	1.8	3.0	3.5	5.5
C6 + cC6 + Tol + EtBz [121]	16	293 – 298	1.5	4.0	1.4	3.5	4.5	7.2
C7 + C8 + cC6 + Tol [122]	16	293 – 298	1.2	-2.6	1.2	-2.6	4.2	6.9
C7 + C8 + cC6 + EtBz [122]	16	293 – 298	1.8	4.1	1.8	4.1	4.2	6.4
C7 + cC6 + Tol + EtBz [122]	16	293 – 298	1.9	5.1	1.7	4.6	5.0	8.5
C7 + cC8 + Bz + Tol [123]	12	293 – 298	0.9	-1.8	1.4	-2.7	12.0	15.5
C7 + cC8 + Bz + EtBz [123]	12	293 – 298	1.6	3.7	1.8	-4.1	11.3	15.0
C7 + cC8 + Tol + EtBz [123]	12	293 – 298	2.8	4.5	2.4	4.2	10.3	13.3
C8 + cC6 + Tol + EtBz [120]	12	293 – 298	2.0	4.4	1.8	4.1	5.8	7.2
cC8 + Bz + Tol + EtBz [123]	12	293 – 298	0.9	-2.3	2.7	-5.2	11.3	16.1
Quinary mixtures								
C6 + C8 + cC6 + Tol + EtBz [120]	12	293 – 298	1.1	-2.0	1.3	-2.3	4.2	5.9
C7 + C8 + cC6 + Tol + EtBz [120]	10	293 – 298	1.7	3.2	1.9	-3.2	4.3	6.7
C7 + cC8 + Bz + Tol + EtBz [123]	10	293 – 298	1.0	-2.2	3.0	-5.2	7.4	8.9
Overall	*592*	*293 - 353*	*2.8*	*8.8*	*2.9*	*8.8*	*3.8*	*16.1*

Cn stands for n-alkanes of n carbon number. Bz, Tol, EtBz, ButylBz, HexylBz and HeptylBz stand for Benzene, Toluene, Ethylbenzene, Butylbenzene, Hexylbenzene and Heptylbenzene, respectively. cC6, ButylcC6, HeptylcC6, cC8 stand for Cyclohexane, Butylcyclohexane, Heptylcyclohexane and Cyclooctane, respectively.
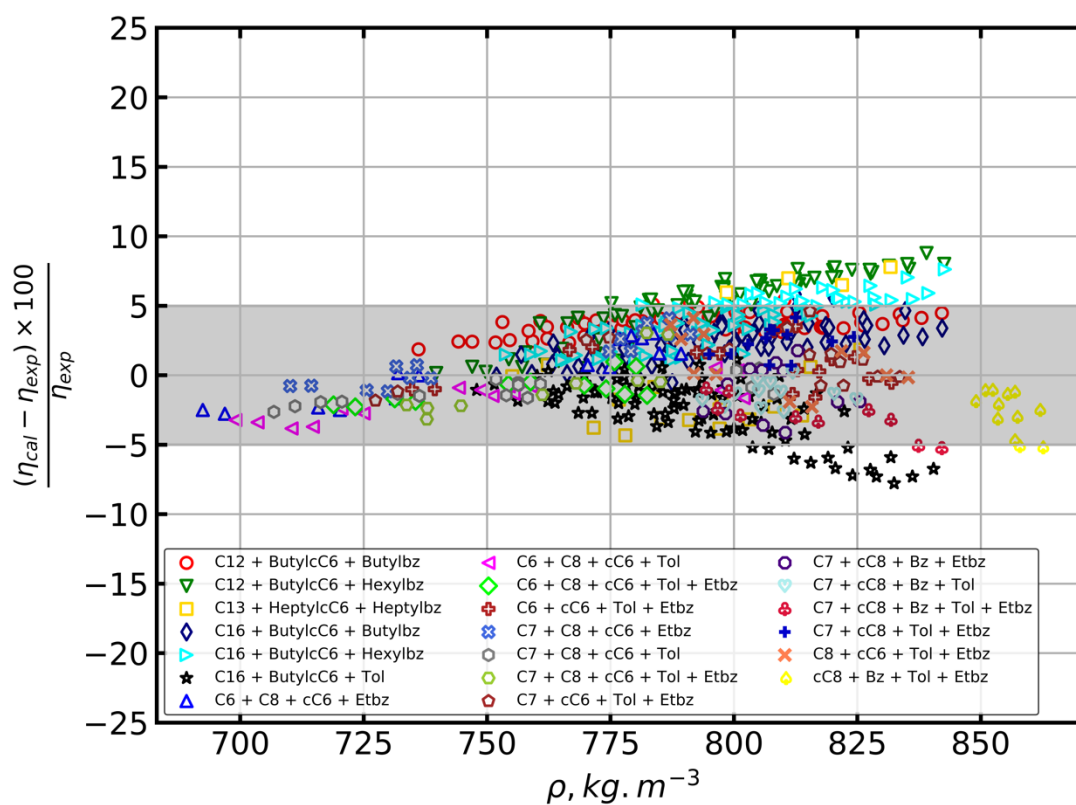
Figure 11: Percentage deviations $[100(\eta_{cal} - \eta_{exp})/\eta_{exp}]$ of the experimental viscosity data for multicomponent mixtures containing n-alkanes, alkylbenzenes, and cyclic species from the calculated values by means of the 1-cEHS model as a function of density.
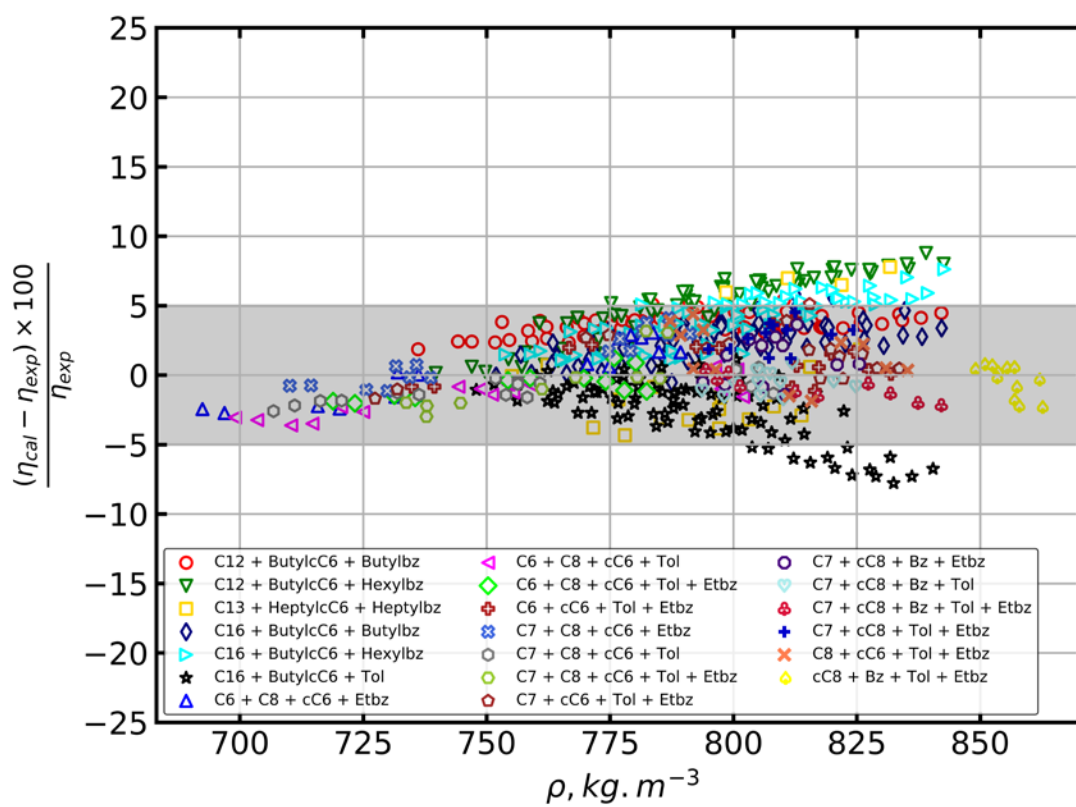
Figure 12: Percentage deviations $[100(\eta_{cal} - \eta_{exp})/\eta_{exp}]$ of the experimental viscosity data for multicomponent mixtures containing *n*-alkanes, alkylbenzenes, and cyclic species from the calculated values by means of the 3-cEHS model as a function of density.
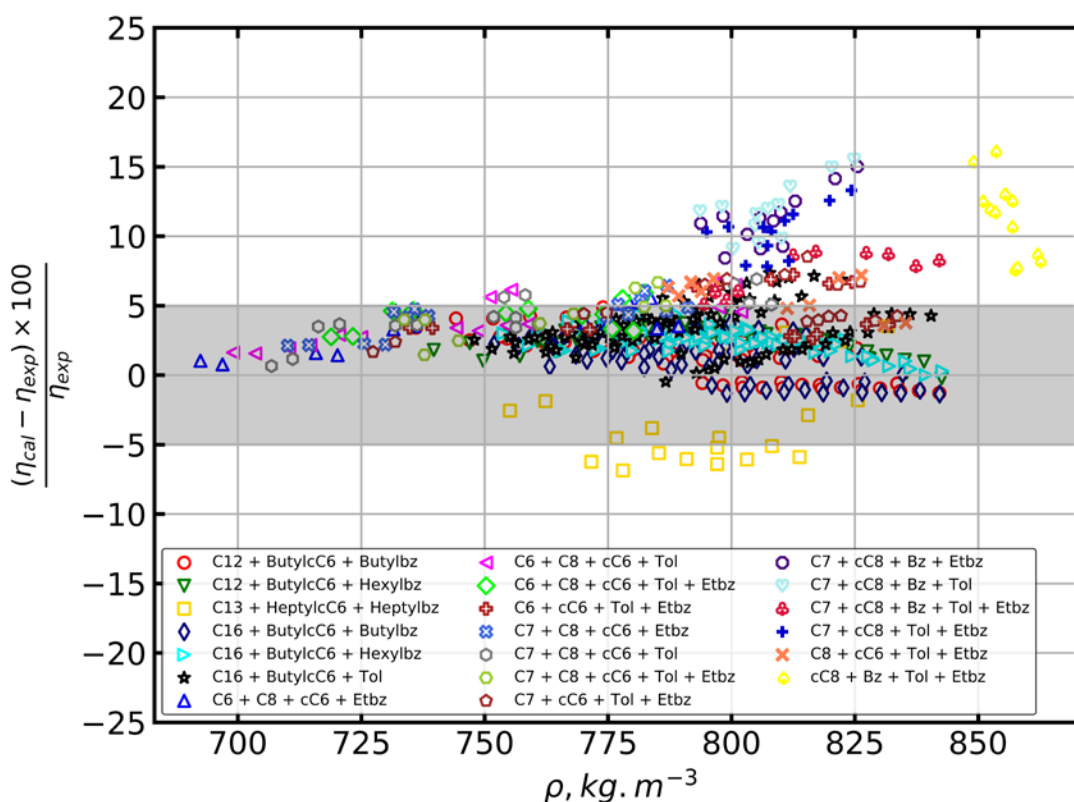
Figure 13: Percentage deviations $[100(\eta_{cal} - \eta_{exp})/\eta_{exp}]$ of the experimental viscosity data for multicomponent mixtures containing *n*-alkanes, alkylbenzenes, and cyclic species from the calculated values by means of the EHS model as a function of density.

4. Conclusions

One component extended hard-sphere (1-cEHS) model has been expanded to successfully predict the viscosity of liquid mixtures consisting of *n*-alkanes, alkylbenzene and cycloalkane species. Based on the comprehensive validation against the available experimental data, the uncertainty of the predicted viscosity values is adjudged to be in the region of 5-10%. The 1-cEHS model represents the mixture as a single pseudo-component characterized by the effective molecular weight. The evaluation of the effective molecular weight, in principle, requires knowledge of the overall molecular weights of *n*-alkane,

alkylbenzene and cycloalkane components, but does not completely eliminate the need for a detailed compositional specification of the mixture, especially if the mixture contains long-chain n-alkanes. In order to remove this constraint we have also developed the three component, extended, hard-sphere (3-cEHS) model. Here, we postulate that the multicomponent mixture can be represented as three pseudo-component mixtures consisting of n-alkane, alkylbenzene and cycloalkane pseudo-components. Each pseudo-component is characterized by the mole fraction and molecular weight only. Based on validation against the available experimental data, the uncertainty of the 3-cEHS model in predicting the viscosity is the same as the uncertainty of the 1-cEHS model, namely 5-10%.

The overall objective of this work was to provide the basis for new types of viscosity models that do not require, as one of the inputs, a full compositional characterization of a mixture, as for quite a few fluids of industrial interest a detailed characterization is neither available nor desirable. In developing such models we have purposely sacrificed accuracy for versatility, as we have replaced the full compositional characterization by a knowledge of molecular weight and mole fraction of each pseudo-component. The 1-cEHS and 3-cEHS models, that were validated against plentiful experimental data, demonstrate that these new types of models can predict reliably the viscosity of industrially relevant mixtures with reasonable accuracy and form a solid basis for further inclusion of species belonging to other chemical families. Further tests will be undertaken, in the near future, to validate the 1-cEHS and 3-cEHS models for mixtures that contain polar species, as such mixtures posed problems for the EHS model with traditional mixing rules.

It is prudent to currently limit the applications of both models to temperatures in the range 273 – 400 K and pressures below 75 MPa, as this is the current range of the general expression for the molar core volume for n-alkanes.

The prediction of viscosity, more often than not, requires accurate mixture density values. In the present work, a couple of examples were given where uncertainty in density

produces nearly an order of magnitude increase in uncertainty in the viscosity prediction. Thus, the need to supplement the viscosity models with thermodynamic packages that can fulfil density requirements. The work also highlighted the need for a critical assessment of the available experimental viscosity data to produce compilations of accurate and reliable datasets that can be used for validation purposes.

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