

# A New Model for Methane Viscosity Based on the Theory of Averaged Intermolecular Potential Field

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**ABSTRACT:** Methane is the simplest alkane and the main constituent of natural gas, shale gas, and gas hydrate. Its viscosity has both academic and industrial importance. A new model for the methane viscosity was developed based on the theory of the averaged intermolecular potential field. The derived equations of the model were fitted to 1669 experimental data points in good agreement over the parameter ranges: from 100 to 523 K with the pressure from 0.047 up to 1000 MPa. The new model also comprehensively performed better than the famous samples in comparison. The physical base, good numerical behavior, and better-comparing performance suggest that the model has considerable theoretical significance and practical application potentials.

**KEYWORDS:** Viscosity; Methane; Model; Molecular interaction; Molecular potential.

## INTRODUCTION

Methane is the simplest alkane and the main constituent of natural gas, shale gas, and gas hydrate. Its physical properties have both academic and industrial importance. One of the most important properties is the viscosity that is required in many designs and industrial applications. Consequently, the viscosity of methane has long been studied via various methods. Notable among these studies may trace back to the work by Swift *et al.* In 1959[1], Who measured the methane viscosity via a cylindrical falling body viscometer with a relative experimental error of up to 8%. In 1966, Huang *et al.* [2] reported the viscosities of methane at pressures to 5000 lb/sq. in. abs. over a temperature range from -170 °C to 0 °C in the declared uncertainty of 1.2%. In 1987, a set of thermophysical properties of methane were calculated, including the viscosity [3]. In the same year, Trengove and

Wakeham [4] reported the viscosity of the three polyatomic gases, carbon dioxide, methane, and sulfur hexafluoride, in the limit of zero density. Two years later, Dymond and Awan [5] correlated high-pressure diffusion and viscosity coefficients for members of the n-alkane series on the limited data at that time. Friend *et al.* [6] built correlations for thermophysical properties of methane containing viscosity on a critical evaluation of the available experimental data. In 1990, Assael *et al.* [7] used semiempirical schemes to the correlation of high-pressure thermal conductivity, viscosity, and diffusion coefficients for n-alkanes. In 2010, Heidaryan *et al.* [8] reported a mathematical model for predicting methane viscosity at high pressures and high temperatures. Three years later, Rabah [9] presented a generalized viscosity Arrhenius correlation for normal alkanes at atmospheric pressure.

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In recent years, some new models for viscosity representation were reported, such as the work by *Khosharay Shahin, et al.* [10] for the viscosity of refrigerants and there is also methane in their work, the neural network method by *Rocabruno-Valdés, C. I., et al.* [11] and *Hosseini, Sayed Mostafa, et al.* [12] as one of the most powerful methods in viscosity representation.

Except for the above general topics, experimental works were also carried out for their substantial importance. In 1980, *Diller* [13] measured the viscosity of compressed gaseous and liquid methane at temperatures between 100 and 300 K and at pressures up to 30 MPa using a torsionally oscillating quartz crystal viscometer. After that, *Gulik et al.* [14,15] measured the methane viscosity coefficients at 25 °C and 273 K at pressures from 0.1 to 1000 MPa employing the vibrating wire viscometer. In 2002, *Evers et al.* [16] presented an apparatus for the simultaneous measurement of viscosity and density of fluids and the viscosity of nitrogen, methane, helium, neon, argon, and krypton. In 2004, *Schley et al.* [17] measured the viscosity of methane and two natural gases using a vibrating-wire viscometer of very high precision. In 2007, *May et al.* [18] determined the zero-density viscosity of hydrogen, methane, and argon in the temperature range from 200 to 400 K via a two-capillary viscometer. In 2010, *Atilhan et al.* [19] reported the dynamic viscosity of pure methane utilizing an electromagnetic piston viscometer over the temperature range (250 to 450) K and pressure range (10 to 70) MPa. In the same year, *Ling* measured the methane viscosity at high pressure and high temperature via a falling body method for his Ph.D. thesis [20].

Among these interests, special attention has been paid to the models for methane viscosity. They may be classified into four categories: 3 terms correlation, hard-sphere model, friction theory, and others. The 3 terms correlation commonly separates the viscosity ( $\eta$ ) into three terms [6,3,21],

$$\eta = \eta_0(T) + \eta_1(T)\rho + \eta_2(T,\rho) \quad (1)$$

Here,  $T$  denotes the absolute temperature and  $\rho$  stands for the density. The first term in Eq. (1) is from the contribution of dilute gas, which can be described by the formulae from Chapman-Enskog theory in the zero density limit. The second term represents the contribution of the moderately dense fluid. By the Rainwater-Friend

theory, this term has a linear-in-density form. The third term comes from the contributions of dense fluid, which is a bivariate function of temperature and density. Although, the expression for the last term was empirically suggested by various authors, at least 2 equations and 15 coefficients are needed in order to express this term. Such separation enables an independent treatment of each contribution, so the most recent advances of the kinetic theory may be used as a guide in the improvement of the three terms.

For the hard-sphere model, approximate expressions for the coefficients of thermal conductivity, viscosity, and self-diffusion was given by the Enskog theory. The expressions made success to n-alkanes [7,22]. In recent years, the extended hard-sphere model was applied to predict the viscosity of gases, liquids, and supercritical fluids [23], even the long-chain n-alkanes [24], after the roughness factors and molar core volumes were related to reduced viscosity and a universal function of reduced volume.

Referring to the friction theory based on principles of mechanics and thermodynamics, the theory for viscosity modeling treated the residual viscosity as mechanical friction, which is related to the van der Waals attractive pressure ( $p_a$ ) and repulsive pressure ( $p_r$ ). For a simple hydrocarbon, it was shown that the friction theory models consisting of a linear relation to  $p_a$  and a quadratic relation to  $p_r$  can accurately represent the viscosity over wide ranges of temperature and pressure,

$$\eta = \eta_0 + (\kappa_a p_a + \kappa_r p_r + \kappa_{rr} p_r^2) \quad (2)$$

Here  $\kappa_r$ ,  $\kappa_a$ , and  $\kappa_{rr}$  are the temperature-dependent friction coefficients [25].

With respect to the other models, the first example was the three-parameter generalized corresponding states principle for the prediction of the viscosity of pure liquids [26], which belongs to the semi-theoretical method. The second is based on the significant structure theory and calculated the viscosity of a liquid from weighted two main components: a gas-like and a solid-like contribution [27].

The above consistent investigations into the methane viscosity reflect the theoretical and practical importance as well as the deep concerns and big interests of researchers in different areas. The determination of the methane viscosity belongs to a particularly important question in natural gas science and engineering. To obtain viscosity data, predicting models based on the theoretical

framework are always in high demand. The theoretical model of methane viscosity can not only give insights into properties of alkanes, but also have the benefits to getting the methane viscosity data much more quickly and cheaply in acceptable uncertainty than measurements and to predict the data even in the conditions at which a measurement cannot be carried out. The aim of this paper is to develop a new theoretical model for the methane viscosity.

The paper has four parts. First, it reviews the extant literature relevant to the viscosity of methane. Then the modeling method is introduced. Next, the findings are presented and discussed. Finally, the paper concludes with core findings and their practical implications.

## THEORETICAL SECTION

### Modeling methane viscosity

With the development of molecular science, emerging theories and methods are always promising in successfully modeling methane viscosity. One of such hopeful routes can be found in the theory of averaged intermolecular potential field [28] that was developed for methane viscosity based on the symmetric molecular interaction approximation [29-31]. This theory provides a solid base to develop a more reliable and confident model for methane viscosity.

According to the averaged intermolecular potential field theory, the methane viscosity can be derived as [28],

$$\eta = K \sqrt{T} \left( 1 + \xi \frac{\rho}{\rho_{ref}} \right)^{\frac{2}{3}} \exp \left( \frac{u}{kT} \right) \quad (3)$$

Where,  $K$  notes a coefficient,  $k$  the Boltzmann's constant,  $T$  the absolute temperature.  $\rho$  stands for the density and  $\rho_{ref}$  is a reference value of the density.  $\xi$  is a variable relying on temperature, and  $u$  is a bivariate function of temperature and density. To obtain a better fit to experimental data, the expressions for the last two variables are upgraded to the following new versions,

$$\xi = \lambda + \left( \frac{T_{\xi}}{T} \right)^{1.5} \quad (4)$$

$$u = \phi_{\sigma} \left( 1 + \frac{T_{\sigma}}{T} \right) \left[ \left( \frac{\rho}{\rho_{ref}} \right)^{\beta/3} - \left( \frac{\rho}{\rho_{ref}} \right)^{\alpha/3} \right] + u_{m0} \left( 1 + \frac{T_m}{T} \right) \quad (5)$$

Here,  $\lambda$ ,  $T_{\xi}$ ,  $\phi_{\sigma}$ ,  $T_{\sigma}$ ,  $\beta$ ,  $\alpha$ ,  $u_{m0}$ ,  $T_m$  and  $\rho_{ref}$  are adjustable coefficients to be fitted with experimental data.

Eqs. (3-5) mathematically present the new model for the methane viscosity. In Eq. (3), the methane viscosity is determined by two terms: the root square of temperature multiplying 2/3th power of density as well as the exponent of the ratio of the averaged intermolecular potential field to average kinetic energy. Physically, the first term is due to the momentum transfer resulting from molecular transportation through Brownian motion, the second is a bulk statistical effect arising from molecular interaction.

In Eq.(5), the averaged intermolecular potential field inside the methane fluid is composed of two terms: the averaged binary potential appearing as the extended Lennard-Jones potential, as well as the total contributions of triple and higher interaction mechanisms [28].

Except for the clear physical meanings of each term, the advantages of the new model are simple and available at different phases. The simplicity means that there are only three explicit functions of temperature and density with merely ten coefficients to be fitted, which is much simpler than the existing models in question. The availability implies the Eqs. (3-5) are applicable to methane samples regardless of their phase states, due to the presentation in terms of temperature and density, in contradict to the presentation using temperature and pressure as independent variables.

When the density approaches zero, we obtain the formula for the methane viscosity in the zero density limit,

$$\eta_0 = K \sqrt{T} \exp \left[ \frac{u_{m0}}{kT} \left( 1 + \frac{T_m}{T} \right) \right] \quad (6)$$

Defining a reduced viscosity as  $\eta_r = \eta/\eta_0$ , as well as a reduced density  $\rho_r = \rho/\rho_{ref}$ , it yields

$$\eta_r = (1 + \xi \rho_r)^{2/3} \exp \left[ \frac{1}{kT} \phi_{\sigma} \left( 1 + \frac{T_{\sigma}}{T} \right) \left( \rho_r^{\beta/3} - \rho_r^{\alpha/3} \right) \right] \quad (7)$$

These reduced variables provide a simpler and more concise form to represent the new model of methane viscosity.

## RESULTS AND DISCUSSION

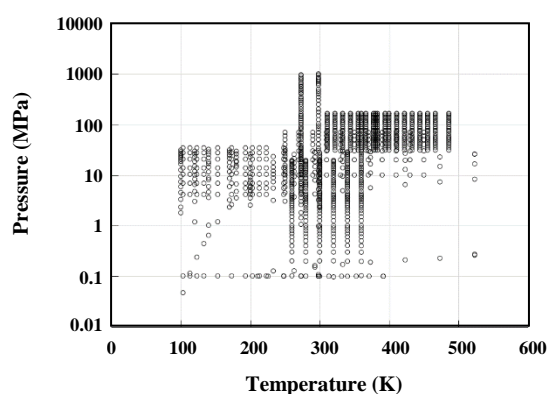
### Viscosity data source

With the development of the measuring technique, advanced viscosity data of methane have been measured. They offer a new base to build a more reliable and confidential model for methane viscosity. Table 1 summarizes 1699 data points measured via relevant

**Table 1: Viscosity data sources of methane.**

Ref.	Year Published	Technique employed <sup>a</sup>	Uncertainty (%)	No. of data	Temperature range (K)	Pressure range (MPa)
Huang[2]	1966	FB	1.2	114	103.2-273.2	0.047-34.47
Diller[13]	1980	TCr	0.5, or 2	116	100-300	0.6-30
Gulik[14]	1988	VBW	0.5	39	298.15	1-1000
Gulik[15]	1992	Abs	1	125	273.1	1-953
Evers[16]	2002	Abs	0.4	61	233-523	0.13-30
Schley[17]	2004	VBW	0.3	345	260-360	0.1-29
May[18]	2007	Cap	0.096	13	210-391	0.1
Atilhan[19]	2010	EPV	2	77	250-450	10-70
Ling[20]	2010	FB	1	779	310.9-485.9	31.1-169.1
Overall	9 refs.			1669	100-523	0.047-1000

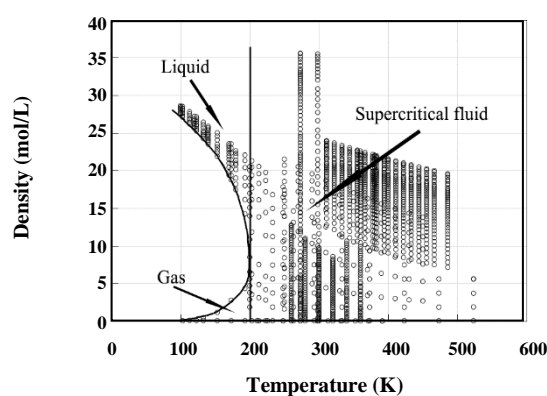
<sup>a</sup>Abs, Absolute; Cap, Capillary; FB, Falling Body; TCr, Torsional Crystal; VBW, Vibrating Wire; EPV, Electromagnetic Piston Viscometer



**Fig. 1: Temperature-pressure ranges of selected experimental viscosity data of methane.**

methods from 9 references that are critically evaluated by us. The uncertainties ascribed by the authors are below 2%. The temperatures at which the viscosity was measured cover a wide range from 100 to 523 K. The pressure ranges from 0.047 up to 1000 MPa. Fig. 1 visually shows the distribution of the 1699 data points on a temperature-pressure plot, which demonstrates that there are enough data locate in both the LTLP (low temperature and low pressure) and HTHP (high temperature high pressure) regions.

It is needed to point out that a small portion of the viscosity data listed in Table 1 were reported in terms of temperature and pressure. For these data, the Helmholtz energy type Equation of State (EOS) of pure methane provided by REFPROP Version 8.0 [32] was then used to calculate the density at the given temperature and pressure.

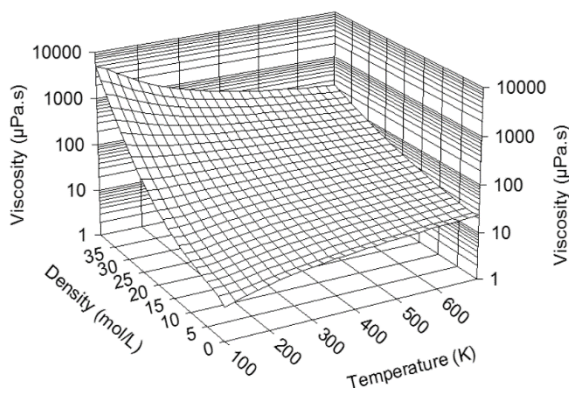


**Fig. 2: Temperature-density ranges of selected experimental viscosity data of methane.**

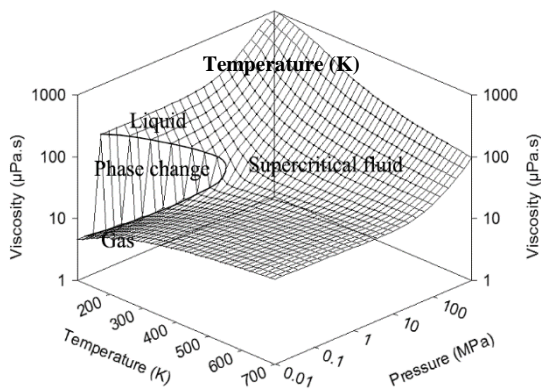
After the calculation, the data distribution in a temperature-density plot can thus be given in Fig.2. It is easy to find that the phases of the experimental samples cover gaseous, liquid, and supercritical methane, although the majority of the data is located in the supercritical region. The phase boundary may also be clearly distinguished as well. Putting together the 1669 viscosity data points that were precisely measured via advanced equipment, yields a good dataset to determine the coefficients of the new model.

#### Viscosity surface

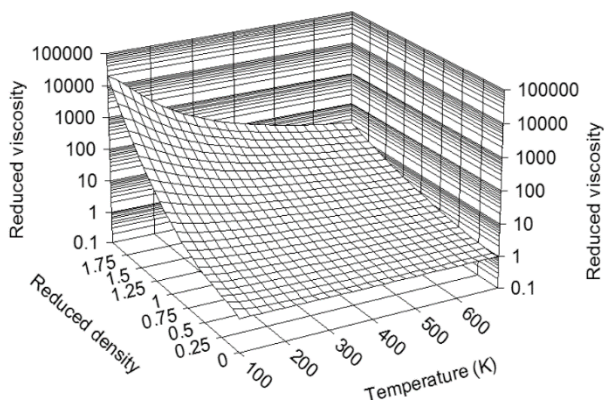
Based on the above-mentioned viscosity dataset and using the least square method carried out with the software named TableCurve 3D v4.0 [33], the optimized coefficients in the Eqs. (3-5) were determined as:  $K=1.093$ ,



**Fig. 3:** Viscosity surface of methane over temperature and density by the author's model.



**Fig. 4:** Viscosity surface of methane over temperature and pressure.



**Fig. 5:** Reduced viscosity surface of methane over temperature and reduced density.

$u_{m0}/k=-151.8$  (K),  $T_m=-48.39$  (K),  $\lambda=-0.2302$ ,  $T_\xi=1624$  (K),  $\rho_{ref}=24.21$  (mol/L),  $\phi_{\sigma}/k=548.3$  (K),  $T_\sigma=-50.27$  (K),  $\beta/3=1.920$ ,  $\alpha/3=0.4266$ , with a  $r^2$  of 0.998. Here the units of variables are: viscosity in ( $\mu\text{Pa}\cdot\text{s}$ ), temperature in (K), and density in (mol/L).

Using Eqs. (3-5) and these coefficients, a viscosity surface of methane was calculated as Fig. 3 in the wide ranges from 100 to 700 K with density up to 40 mol/L. The surface shows that the viscosity unanimously increases with density at a fixed temperature. In the zero density limit, the viscosity increases with temperature. However, the viscosity turns to decrease with temperature as the density takes the amounts larger than a critical value. In other words, the trend for the viscosity varying with temperature reverses at the critical density.

Based on the new model along with the aforementioned equation of state for pure methane, a viscosity surface of methane over temperature and pressure was determined in Fig.4, which shows that the viscosity monotonously increases with pressure going up from 0.01 to 1000 MPa for temperatures higher than the critical temperature (190.56 K). Consequently, the supercritical methane behaves as a pressure-induced thick fluid. When the temperature is less than the critical value, there is a viscosity jump due to the density difference before and after the phase change.

Using Eq. (7), the reduced viscosity over temperature and reduced density were calculated as Fig.5, which indicates that the reduced viscosity accordingly increases as the reduced density rises from 0 to 2, and/or the temperature decreases from 625 to 100 K. The variation trend with temperature does not change in the entire reduced density region (0-2) in contradict to the trend in Fig. 3.

The above 3D surfaces visually illustrate the major relations among viscosity and thermodynamic parameters by the new model. These results are highly expected by the authors of this paper to be used in correlating the methane viscosity, whose accuracy and advantages need to be evaluated in more detail.

### Evaluation of the model

In order to give an overall impression of the performance of the developed new model, we plot the 1669 predicted viscosity points against the measured viscosity points in Fig.6. It shows that almost all the points

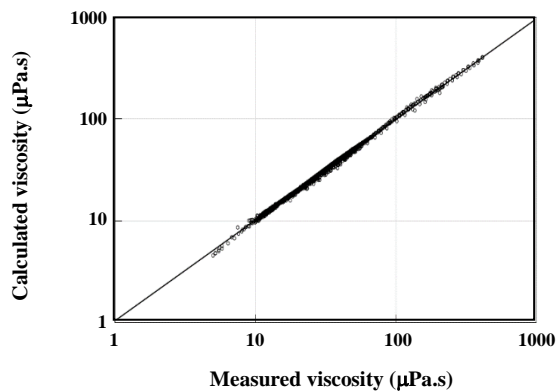


Fig. 6: Plot of the measured viscosity versus predicted viscosity.

narrowly locate along the diagonal line, suggesting a success for the model to describe the advanced viscosity data gathered from the references listed in Table 1.

With respect to deviation, an Absolute Percent Deviation (APD) between predicted and measured data is defined as usual,

$$APD = \left| 1 - \frac{\eta_{\text{predicted}}}{\eta_{\text{measured}}} \right| \times 100 \quad (8)$$

Here,  $\eta_{\text{predicted}}$  and  $\eta_{\text{measured}}$  are the predicted and measured viscosity, respectively.

Fig. 7 plots the APDs of the 1669 points of methane viscosity with respect to temperature, while the predicted data is calculated with the developed new model and the optimized coefficients. It shows that the APD randomly distribute with temperature and the majority of the points have the APDs below 5%. The few large APDs mainly position under the critical temperature (190.56 K), where the methane behaves itself as either gas or liquid including phase change. The average and maximum APD is 2% and 15%, respectively. As a reference for readers, the prediction by NIST database to the 1669 points gives similar APDs with 10 undefined points (unpredictable using the database).

Concerning the advantages of the developed new model, a comparison to the three famous models mentioned in the introduction may demonstrate more clearly. The main behaviors of the models are listed in Table 2. It shows that the author's new model is much simpler, and bases on the fundamental molecular theory with the new method.

In the review of the evaluating results, we see a very successful new model for methane viscosity, which

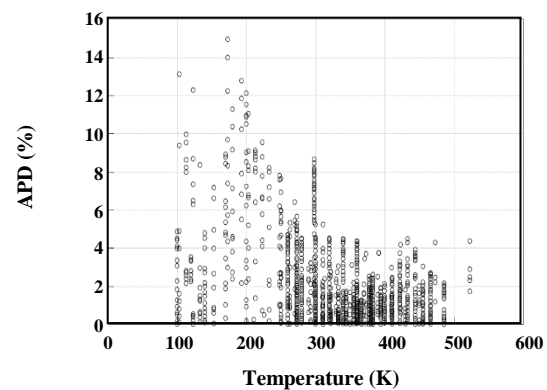


Fig. 7: Absolute percent deviations of the predicted data from the experimental data versus temperature.

comprehensively performs better than the famous samples in question. With respect to the reason for success, it is suggested that the new model remains the core laws that govern the variation of methane viscosity with temperature and density. And thus, the new model can extrapolate in a physically reasonable manner.

## CONCLUSIONS

This work developed a new model for the viscosity of methane based on the theory of averaged intermolecular potential field using the symmetric molecular interaction approach. The derived equations of the model were expressed as functions of temperature and density. Each term in the functions has a corresponding physical meaning. The functions are easily and conveniently calculable and applicable at gaseous, liquid, and supercritical phases over a wide parameter range: from 100 to 523 K with the pressure from 0.047 up to 1000 MPa. The model has been fitted to the 1669 experimental data points that were reasonably selected for the methane viscosity in good agreement. The physical and numerical good behaviors imply success, reliability, and confidence for the model to predict methane viscosity. The new model also comprehensively performs better than the famous samples in comparison. Therefore, the model has considerable theoretical significance and practical application potentials in natural gas, shale gas, and gas hydrate science and engineering.

## Nomenclatures

### English symbols

k	Boltzmann's constant (-)
K	Coefficient (-)
$p_a$	Attractive pressure (-)

Table 2: Main behaviors of author's new model compared to the three famous ones.

Theory	Required equations	Fitted coefficients	Required inputs	Bases
This work	3	10	T, $\rho$	Theory of averaged intermolecular potential field, symmetric molecular interaction approach, Maxwell-Boltzmann distribution function, extended Lennard-Jones' potential
3 terms correlation	8	$\geq 30$	T, $\rho$	Chapman-Enskog theory for the term in the zero density limit, Rainwater-Friend theory for the linear-in-density term, empirical higher density terms
Extended hard-sphere model	6	$\geq 20$	T, V	Chapman-Enskog theory for the term in the zero density limit, Hard-sphere theory, empirical correlations
Friction theory	$\geq 6$	$\geq 15$	T, p, $M_w$	Chapman-Enskog theory for the term in the zero density limit, mechanical friction, and EoS

$p_r$	Repulsive pressure (-)
T	Absolute temperature (K)
$T_\xi$	Coefficient (-)
$T_\sigma$	Coefficient (-)
$T_m$	Coefficient (-)
u	Variable (-)
$u_{m0}$	Coefficient (-)

#### Greek symbols

$\alpha$	Coefficient (-)
$\beta$	Coefficient (-)
$\eta$	Viscosity ( $\mu\text{Pa}\cdot\text{s}$ )
$\kappa_r$	Friction coefficient (-)
$\kappa_a$	Friction coefficient (-)
$\kappa_{rr}$	Friction coefficient (-)
$\lambda$	Coefficient (-)
$\xi$	Variable (-)
$\rho$	Density (mol/L)
$\rho_{ref}$	Reference density (mol/L)
$\phi_\sigma$	Coefficient (-)

#### Subscripts

a	Attractive
r	Repulsive, reduced variable
ref	Reference

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