Viscosity Model for Pure Liquids Based on Eyring Theory and Cubic EOS

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Viscosity is an important transport property in process design and development. Knowledge of viscosity of pure liquids and of the liquid mixtures plays a key role in solving chemical engineering problems dealing with heat-transfer and mass-transfer operations, and fluid-flow processes. Much effort has been directed towards the development of reliable models for correlating and predicting viscosities of pure nonpolar and polar liquids. However, there is still a lack of a sound theory for the accurate prediction of liquid viscosities within a wide range of temperatures and pressures (Reid et al., 1987). Furthermore, viscosity models based on similarities either with gas-like or solid-like behavior have not successfully been proven to yield values of liquid viscosity for pure liquids and mixtures as functions of temperature (Chhabra, 1992). Consequently, most of the viscosity models reported in the literature have been developed under semi-empirical and empirical considerations. Pertinent literature on viscosity models for pure liquids has been reviewed and evaluated by Przezdziecki and Sridhar (1985), Reid et al. (1987), Mehrotra (1991), Monnery et al. (1995), Mehrotra et al. (1996), and Poling et al. (2001).

A thorough review of the literature reveals that numerous empirical and semiempirical models have been published for viscosities of pure liquids with exponential viscosity-temperature functions receiving most of the attention. The corresponding states principle along with the Eyring's absolute rate theory are the most widely used approaches to represent the viscosity of pure liquids. Recently, Lei et al. (1997) presented a two-parameter model based on the Eyring's absolute rate theory for the correlation of the viscosity of pure liquids under saturated conditions. Their correlating results yielded an overall *ADD* of 1.51% in the representation of experimental liquid viscosities of 106 pure nonpolar and polar fluids. An important feature of the model proposed by Lei et al. is that, at a given temperature, the liquid viscosity can directly be computed from equilibrium properties such as vapor pressure, heat of vaporization, and compressibility factors of the phases at equilibrium. Based on the two-parameter model of Lei et al. (1997), the purpose of the present study is three fold. The first purpose is to make use of a well-known cubic equation of state to calculate all of the thermodynamic properties required in the model, thus enabling the simultaneous calculation of vapor-liquid equilibria and saturated liquid viscosity. The second purpose is to improve the correlation of liquid viscosity over a wider temperature range by modifying the functionality between the activation energy and the internal energy of vaporization, and, the third purpose is to extend the use of the model to the calculation of compressed liquid viscosities (at pressures larger than the saturation point).

Model Development

On the basis of the Eyring's Absolute Rate Theory (Glasstone et al., 1941), Lei et al. (1997) derived the following two-parameter model for the viscosity calculation of a pure saturated liquid η_l^s

$$\eta_l^s = \frac{RT}{v_l^s} \cdot \frac{1}{\gamma} \exp\left[\alpha \frac{\Delta U_{\mathrm{vap}}}{RT}\right] \exp(Z_l^s) \tag{1}$$

where γ represents the frequency in Hz or s^{-1} of a molecule that jumps from an activated position to a vacancy site, and α is the proportionality factor between the activation energy $\Delta G^{\neq}$ and the internal energy of vaporization ΔU_{vap}. The other variables v_l^s and Z_l are the molar volume and the compressibility factor of the liquid, respectively, at the temperature and pressure of saturation. Lei et al. reported individual values of γ and α for 106 nonpolar and polar compounds. One of the main shortcomings of Eq. 1 is attributed to the assumption of the linear relationship between $\Delta G^{\neq}$ and ΔU_{vap}, since it may be not valid for some fluids, particularly

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near the critical point. In an attempt to extend the applicability of Eq. 1 to a wider temperature range, a power-law dependence between $\Delta G^{\neq}$ and ΔU_{vap} was adopted as follows

$$\frac{\Delta G^{\neq}}{RT} = \alpha \cdot \left(\frac{\Delta U_{\text{vap}}}{RT} \right)^{\beta} \tag{2}$$

Knowing that $\Delta U_{\text{vap}} = \Delta H_{\text{vap}} - P\Delta v_{\text{vap}} = \Delta H_{\text{vap}} - RT\Delta Z_{\text{vap}}$, the first exponential term in Eq. 1 is modified accordingly

$$\eta_l^s = \frac{RT}{v_l^s} \cdot \frac{1}{\gamma} \exp\left[\alpha \left(\frac{\Delta H_{\text{vap}}}{RT} - \Delta Z_{\text{vap}} \right)^{p} \right] \exp(Z_l^s) \tag{3}$$

here ΔH_{vap} is the latent heat of vaporization and $\Delta Z_{\text{vap}} = Z_\nu - Z_l$. In this work, the above equation served to reproduce the experimental liquid viscosities for some commonly encountered compounds at saturated conditions. For each compound, the best values of the characteristic parameters γ, α, and β were obtained via a nonlinear regression of the experimental viscosity-temperature data from the normal boiling point to near the critical point. At a given temperature and pressure, the equilibrium properties v_l^s, ΔH_{vap}, Z_ν^s and Z_l^s in Eq. 3 were conveniently calculated by means of a cubic equation of state. For this purpose, the equations of state proposed by Soave (1972) (SRK) and Peng and Robinson (1976) (PR) were used. At pressures higher than the saturation point, the effect of pressure on liquid viscosity is important, particularly at high-reduced temperatures. An expression similar to the Eyring's viscosity model for liquid mixtures was also devised to correct the value of liquid viscosity obtained from Eq. 3 due to an increase in pressure

$$\ln\left[\frac{\eta_l v_l}{(\eta_l v_l)^s} \right] = \frac{\Delta G^{\neq,hp}}{RT} \tag{4}$$

where superscript s stands for saturated conditions and $\Delta G^{\neq,hp}$ is a high-pressure activation energy and represents the energy required to compress the liquid from its vapor pressure P^s to a pressure of interest P $(P > P^s)$ at constant temperature. The following functionality can be used to relate $\Delta G^{\neq,hp}$ to well-known thermodynamic variables

$$\begin{aligned} \Delta G^{\neq,hp} &= -\sigma \cdot \Delta U_l^{hp} = -\sigma \cdot \{ \Delta H_l^{hp} + \Delta(P v_l) \} \\ &= \sigma \cdot \{ (H_l^s - H_l) - (P^s v_l^s - P v_l) \} \end{aligned} \tag{5}$$

where σ is an adjustable proportionality factor. Substituting the above equation into Eq. 4, we obtain an expression to correct η_l^s at elevated pressures

$$\eta_l = \eta_l^s \cdot \frac{v_l^s}{v_l} \exp\left\{ \sigma \left[\frac{(H_l^s - H_l)}{RT} - (Z_l^s - Z_l) \right] \right\} \tag{6}$$

Table 1. Regression Results for Saturated Liquid Viscosities

		Parameter α		Parameter β		Parameter $\gamma \times 10^{-11}$ [s^{-1}]			AAD [%]		AADmax [%]	
Compound	N	SRK	PR	SRK	PR	SRK	PR	T [K]	SRK	PR	SRK	PR
Hydrogen	9	0.149	0.125	1.256	1.375	16.45	17.39	20.4−32	0.45	0.48	1.04	1.11
Nitrogen	9	0.066	0.053	1.467	1.578	4.525	4.880	77.4−120	1.44	1.34	4.82	4.52
CO_2	9	0.030	0.023	1.827	1.965	6.610	7.140	216.6−300	2.01	1.98	4.24	4.22
CCl_4	9	0.466	0.388	0.753	0.821	6.598	6.606	349.9−525	3.02	2.98	8.84	8.70
Methane	10	0.076	0.060	1.427	1.544	10.51	11.25	111.4−190	0.82	0.73	3.14	2.43
Acetylene	9	1.106	0.860	0.419	0.486	36.00	30.27	192.2−290	1.75	1.76	3.18	3.23
Ethylene	10	0.198	0.194	0.963	0.983	8.597	9.706	169.4−281	1.69	1.97	4.73	7.27
Propane*	28	0.155	0.126	1.118	1.207	7.346	7.763	230−365	0.50	0.62	2.35	2.93
1,3-Butadiene	9	0.024	0.019	1.832	1.942	5.229	5.726	268.7−410	1.81	1.99	5.38	6.09
n-Pentane	9	0.173	0.141	1.039	1.121	5.814	6.136	309.2−440	0.64	0.70	0.91	1.11
Benzene	9	0.051	0.041	1.557	1.657	4.857	5.261	353.3−550	0.75	0.71	1.64	1.62
Cyclohexane	9	0.019	0.015	1.943	2.063	2.578	2.822	353.9−535	0.58	0.62	1.35	1.18
Toluene	9	0.010	0.007	2.091	2.235	2.970	3.260	383.8−575	1.14	1.34	3.35	4.10
n-Heptane	9	0.097	0.077	1.236	1.334	4.037	4.296	371.6−520	0.67	0.75	2.09	2.37
p-Xylene	9	0.155	0.122	1.043	1.136	4.798	5.025	412−605	2.05	2.09	6.00	6.36
n-Decane†	50	0.239	0.233	0.918	0.938	4.751	5.404	283−600	1.51	1.59	4.30	6.42
HCl	9	0.349	0.312	0.925	0.976	19.93	21.16	188.1−305	1.70	1.81	7.07	7.61
Water	17	0.012	0.009	1.899	2.024	20.49	22.40	373.2−643.2	0.39	0.39	1.73	0.72
Ammonia	9	0.068	0.057	1.383	1.464	14.88	16.17	239.8−390	1.28	1.13	5.01	4.30
Methanol	9	0.067	0.056	1.352	1.426	11.84	12.87	337.9−493.2	1.95	2.10	7.45	8.08
Acetic Acid	9	0.100	0.084	1.243	1.315	7.444	8.044	391.2−560	0.43	0.41	1.33	1.25
Acetone	9	0.412	0.337	0.733	0.803	11.40	11.46	329.3−480	1.29	1.34	3.15	3.35
Methyl Acetate	9	0.216	0.174	0.933	1.014	7.092	7.367	331−490	1.27	1.33	3.57	3.89
iso-Propanol	9	0.027	0.022	1.697	1.791	4.885	5.321	355.7−498	1.15	1.31	4.66	5.53
MTBE	9	0.157	0.128	1.090	1.173	5.357	5.664	331.2−480	0.95	1.01	2.44	2.72
Phenol	9	0.028	0.023	1.739	1.826	7.077	7.776	455−665	3.07	3.20	8.33	9.01
Aniline	9	0.006	0.005	2.237	2.369	3.844	4.242	457.8−675	1.49	1.70	4.91	5.71
TOTAL	**313**								**1.28**	**1.34**	**3.96**	**4.29**

*Vogel et al. (1998).

†Yaws (1999).

$$AAD = \frac{100}{N} \cdot \sum_{i=1}^{N} \left| 1 - \frac{\eta_i^{\text{cal}}}{\eta_i^{\text{exp}}} \right|$$

A better performance of the above expression was obtained by excluding the evaluation of the enthalpy difference $(H_l^s - H_l)$. Furthermore, for most fluids, the contribution of the term $(H_l^s - H_l)$ is not significant at isothermal conditions and at pressures higher than the saturation point. Consequently, at constant temperature Eq. 6 reduces to

$$\eta_l = \eta_l^s \cdot \frac{PZ_l^s}{P^s Z_l} \exp\left[\sigma\left(Z_l - Z_l^s\right)\right] \qquad (7)$$

where σ was found to be a function of the reduced temperature as follows $\sigma = \sigma_0 + \sigma_1 T_r^2$. The parameters σ_0 and σ_1 were adjusted to individually fit the experimental data of compressed liquid viscosities. Evidently, Eq. 7 provides a simple way to correct the value of liquid viscosity at pressures beyond the saturation point of the fluid. It only requires the calculation of the high-pressure property Z_l using either the SRK or PR cubic equation of state at the temperature and pressure of interest.

Results and Discussion

Initially, Eq. 3 was applied to the reproducibility of experimental saturated viscosities of 27 pure liquids. The experimental viscosity data of the pure liquids considered in this study were obtained from Beaton and Hewitt (1989) with some exceptions: for the case of propane, the reference data of Vogel et al. (1998) was used, whereas the regressed experimental data of Yaws (1999) was considered for *n*-decane. The correlating results obtained in this work are summarized in Table 1 for 16 nonpolar and 11 polar fluids. These results include the regressed constants γ, α and β for the two modeling approaches (Eyring-SRK and Eyring-PR), the correlating temperature range, and the resulting *AAD* values between calculated and observed viscosity data. Unlike Lei et al. (1997), we used a wider temperature range to correlate liquid viscosity data: from the normal boiling temperature of the fluid of interest to near its critical point (where $\ln\eta$ is no longer a linear function of T_r^{-1}), except for *n*-decane for which a much wider temperature range was adopted. In fact, Lei et al. mostly correlated their viscosity data using temperatures from the triple point to near the normal boiling point of the compound (where $\ln\eta$ varies linearly with T_r^{-1}). Unfortunately, they gave no information about the calculation methods used to compute the equilibrium properties incorporated into their viscosity model (Eq. 1). It was, therefore, not possible to reproduce their viscosity results within the same temperature ranges considered in this study. As shown in Table 1, the performance of Eq. 3 in correlating saturated liquid viscosities of highly polar fluids was remarkably good with overall *AAD* values of 1.29% using the Eyring-SRK model and 1.35% using the Eyring-PR model, whose values are within experimental uncertainty. For some of the compounds considered in the present work, Figures 1 and 2 show the graphical comparison between observed saturated liquid viscosities and those calculated using the proposed approach. Evidently, the reproducibility results are excellent in all cases, including highly polar fluids such as water, acetone, and aniline. The success of the present approach in reproducing the experimental viscosity data of nonpolar and polar compounds is rather attributed to the mathematical form of the Eyring theory itself (Eq. 2) and not to the nature of the two cubic EoS used to compute the equilibrium properties: v_l^s, ΔH_{vap}, Z_v^s and Z_l^s. Both EoS poorly represent the liquid-phase behavior of most substances, particularly highly polar fluids. However, they at least give the correct temperature dependency within the framework of the Eyring theory. Despite the improvements that the PR EoS offers in the representation

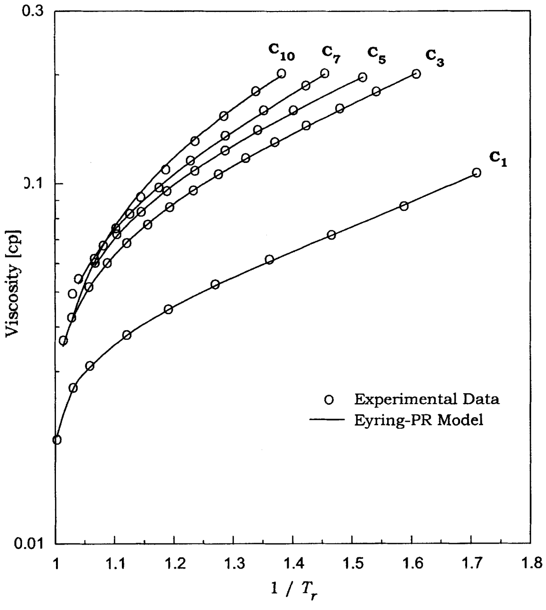

Figure 1. Reproducibility of observed saturated liquid viscosities using the Eyring-PR model.

Nonpolar fluids: normal paraffins.

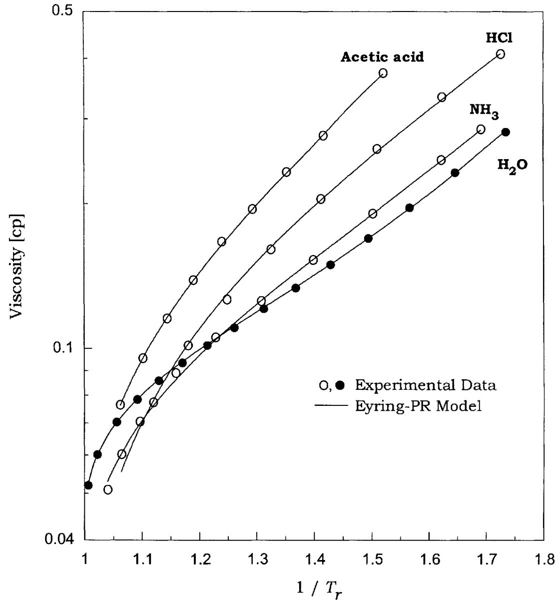

Figure 2. Reproducibility of observed saturated liquid viscosities using the Eyring-PR model.

Polar fluids: inorganic substances.

of thermodynamic properties in the liquid phase, the Eyring-SRK model surprisingly yielded better reproducibility results than the Eyring-PR model with lower *AAD* values for the majority of the compounds listed in Table 1. One possible cause is that the capability differences between the two equations of state vanish during the regression process of the viscosity data, while being finally absorbed by the regressed parameters γ, α and β. Under these circumstances, one may conclude that the temperature dependency of the equilibrium properties embedded in the viscosity model and calculated by the SRK equation of state is more *ad hoc* to the Eyring theory for some fluids. The validity of Eq. 3 was also verified for temperatures below the normal boiling point down to the freezing point. The performance of the model within this temperature range was acceptable for some selected compounds. The correlating capabilities of Eq. 7 were tested in the representation of experimental liquid viscosities above the saturation pressure for some of the compounds considered in this study (7 nonpolar and 3 polar fluids). Table 2 shows the parameters σ_0 and σ_1 obtained for the two Eyring-EoS-based models and their corresponding *AAD* values per compound. As shown in Table 2, the low *AAD* values obtained for the two Eyring models clearly demonstrate the suitability of Eq. 7 in reproducing compressed liquid viscosities at different temperatures and within a wide pressure range. Figures 3, 4, and 5 graphically show the experimental compressed liquid viscosities along with those calculated using Eq. 7 at various temperatures for the case of propane, *n*-decane, and water, respectively. For comparison, liquid viscosities obtained from the recently published *f-theory* model (Quiñones-Cisneros et al., 2000) have also been plotted in Figures 3 and 4 at the same temperature and pressure conditions. As shown in Figures 3 and 4, correlated liquid viscosities using Eq. 7 are in good agreement with the experimental data reported by Vogel et al. (1998) for propane and Ducoulombier et al. (1986) for *n*-decane over a wide pressure range of 1–1,000 bar. Also, as depicted by Figures 3 and 4, the *f-theory* SRK model in its five-constant parametric version performs quite well in reproducing compressed liquid viscosities for all the isotherms. For the case of propane, the *f-theory* model performs much better than the present approach within the pressure range of 100–500 bar, particularly at the highest temperature (360 K). At pressures above 600 bar, however, Eq. 7 does a better job for the three isotherms in comparison to the *f-theory* formulation. For *n*-decane, although both approaches yield comparable results, at 293.15 K, the Eyring-SRK model gives an improved representation of liquid viscosities as the maximum pressure is approached (Figure 4). For the case of water (Figure 5), there is an excellent agreement between calculated and observed viscosity data over the entire pressure range of 10–1,000 bar, particularly at the highest temperature. In fact, the resulting *AAD* value is of only 0.78%, which is within experimental uncer-

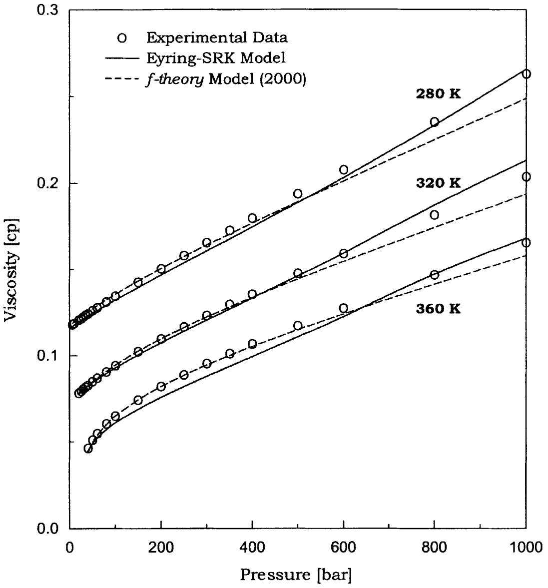

Figure 3. Compressed liquid viscosities of propane using the Eyring-SRK model.

Table 2. Performance of the Two Eyring-CEoS-based Models at High Pressures

		Parameter σ_0		Parameter σ_1				*AAD* [%]	
Compound	*N*	SRK	PR	SRK	PR	*T* range [K]	*P* range [bar]	SRK	PR
Hydrogen*	21	0.305	0.347	−0.022	−0.03	20–30	1–300	1.27	1.41
Methane*	45	0.512	0.587	−0.338	−0.396	110–190	1–500	2.07	2.02
Propane†	95	0.029	0.048	0.289	0.312	280–360	6–1,000	1.93	2.19
n-Pentane‡	31	−0.08	−0.098	0.531	0.633	298–373.4	1–246.3	2.02	2.13
Benzene*	53	0.155	0.184	0.106	0.12	375–550	2–500	1.02	1.01
n-Heptane*	54	−0.002	0.008	0.307	0.339	373.2–533.2	1–506.6	1.71	1.66
n-Decane§,‖	66	0.074	0.082	0.105	0.127	293.2–373.4	1–1,000	2.11	2.28
Water#	66	0.233	0.278	−0.224	−0.254	423.2–623.2	10–1,000	0.91	0.78
Ammonia*	41	0.139	0.161	0.223	0.263	313.2–373.2	50–800	1.64	1.71
iso-Propanol*	37	0.343	0.392	−0.009	−0.009	428.2–503.2	25.3–810.6	1.72	1.8
TOTAL	**509**							**1.65**	**1.72**

*Vargaftik (1975).
†Vogel et al. (1998).
‡Estrada-Baltazar et al. (1998a).
§Ducoulombier et al. (1986).
‖Estrada-Baltazar et al. (1998b).
#Beaton and Hewitt (1989).

$$AAD = \frac{100}{N} \cdot \sum_{i=1}^{N} \left| 1 - \frac{\eta_i^{\text{cal}}}{\eta_i^{\text{exp}}} \right|$$

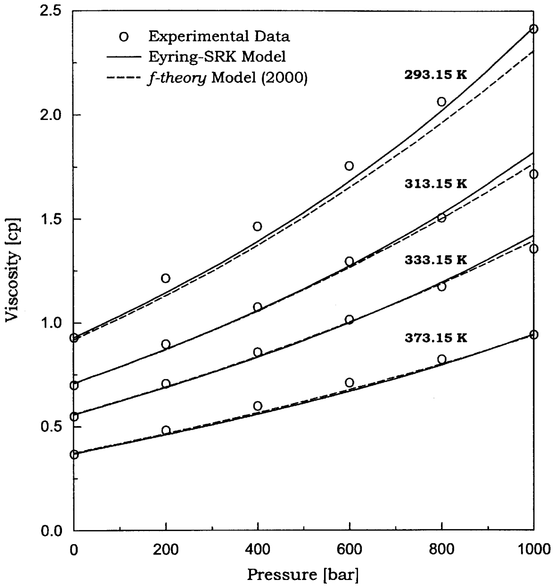

Figure 4. Compressed liquid viscosities of *n*-decane using the Eyring-SRK model.

tainty. These results again demonstrate the potential of the present approach in accurately modeling compressed liquid viscosities of nonpolar and polar fluids above their vapor pressures. The accuracy of Eq. 7 appears to depend on how well saturated liquid viscosities of a fluid of interest were originally correlated with temperature via the use of Eq. 3. This explains the excellent reproducibility results obtained for water.

Conclusions

On the basis of the Eyring theory for pure fluids, a new three-parameter viscosity model was developed for the calculation of saturated viscosities of pure liquids. The model adequately describes the viscosity-temperature relationship in terms of equilibrium properties easily determined via the use of a well-known cubic equation of state. A two-parameter model was also devised to correct saturated liquid viscosity data at pressures higher than the saturation point. The following conclusions can be drawn from this work:

- The accuracy of the proposed model appears to depend on the mathematical form of the Eyring theory regardless of the nature of the equation of state used to calculate the equilibrium properties. On the other hand, the use of a simple cubic EoS will allow the simultaneous determination of phase equilibria and saturated liquid viscosity with reliable results.
- For all fluids considered in this study (16 nonpolar and 11 polar liquids), the reproducibility of experimental saturated viscosities was excellent with an overall average deviation *AAD* of 1.28% using the Eyring-SRK model and 1.34% using the Eyring-PR model for a total of 313 data points.
- The proposed power-law dependence between the activation energy and the internal energy of vaporization gave improved viscosity results within a wider temperature range (from the normal boiling point to near the critical point). The resulting model is, therefore, capable of accurately describing the viscosity-temperature behavior over two important temperature regions: at $T_r < 0.7$ down to the triple point and at $T_r > 0.7$ up to the critical point of the fluid.
- The results confirmed the suitability of the proposed approach to calculate compressed liquid viscosities (above the saturation pressure) for 7 nonpolar and 3 polar fluids with *AAD* values of 1.65% using the SRK EoS and 1.72% using the PR EoS for a total of 509 points.

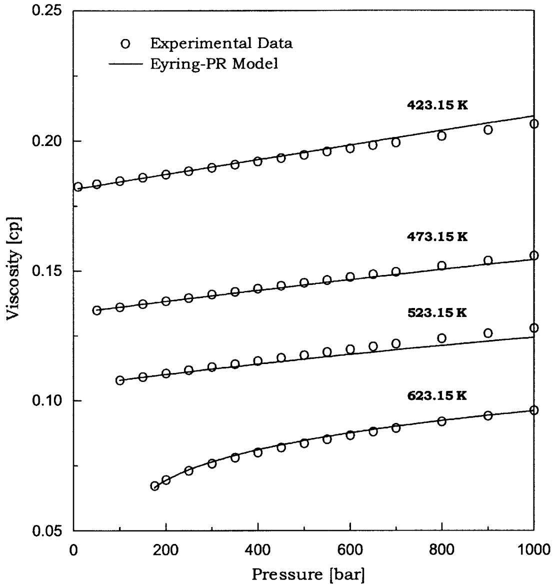

Figure 5. Compressed liquid viscosities of water using the Eyring-PR model.

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Notation

ΔG = Gibbs energy
H = enthalpy
ΔH_{vap} = latent heat of vaporization
P = pressure
R = ideal gas constant
T = temperature
ΔU_{vap} = internal energy of vaporization
v = molar volume
Z = compressibility factor
α = regression constant in Eq. 3
β = regression constant in Eq. 3
γ = regression constant in Eq. 3
η = dynamic viscosity
σ_0 = regression constant
σ_1 = regression constant

Subscripts

l = liquid phase
v = vapor phase

Superscripts

hp = high-pressure property
s = saturated conditions
$\neq$ = activation state

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