CORRELATIONS OF THE LIQUID VISCOSITY OF MIXTURES OF R-404A AND LUBRICANT OIL

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Abstract. *Understanding the viscosity behavior of refrigerant-oil mixtures is of crucial importance in the design and operation of refrigeration compressors and systems. Although a number of different methods and correlations have been proposed and evaluated for mixtures of a single component refrigerant (e.g., R-134a, R-600a and R-744) and lubricant oil, there is a shortage of correlations for the viscosity of mixtures involving refrigerant blends such as R-404A in the open literature.The purpose of the present paper is to conduct an evaluation of two widely used liquid viscosity models, i.e., Grunberg and Nissan (1949) and Katti and Chaudry (1964), in terms of their prediction of the viscosity of a mixture of R-404A (44% of R-125, 52% of R-143a and 4% of R-134a by weight) and a polyolester (POE) ISO 32 lubricant oil for temperatures of 20, 40 and 60^{o}C. The experimental data on the liquid mixture viscosity, together with liquid solubility and mixture density, were obtained from the reference database provided by Cavestri (1995).*

Keywords*: Refrigerant-oil mixture, Refrigerant blends, R-404A, Viscosity, Correlation*

1. INTRODUCTION

It is well known that the performance of refrigeration systems can be very much affected by a variation of the composition (and hence the thermophysical properties) of the working fluid due to the oil migration from the compressor to the system components (Lottin *et al.*, 2003a, 2003b; Cremaschi *et al.*, 2005; Marcinichen, 2006). By the same token, the refrigerant dissolved in the lubricant oil inside the compressor can also influence the mechanical (friction) losses because of the significant impact of the refrigerant solubility on the refrigerant-oil mixture viscosity (Prata and Barbosa, 2009). Although there have been a large number of studies in the open literature dealing with the prediction of the viscosity of refrigerant-oil mixtures (Marcelino Neto and Barbosa, 2010a, 2010b), the vast majority of such models are for single component refrigerants (e.g., R-134a, R-600a and R-744). Therefore, there is a shortage of correlations for the viscosity of mixtures involving refrigerant blends such as R-404A, R-407C and R-410A.

Cavestri (1995) reported on an extensive database of viscosity, solubility and density of mixtures of a lubricant (either a 32 ISO branched POE or a 32 ISO mixed acid POE) and HFC refrigerants (R-32, R-125, R-134a, R-143a) and HFC blends (R-404A, R-507, R-407C). The mixture viscosity was measured with an oscillating body-type viscometer, which was also capable of providing the liquid mixture density. For the HFC blend/oil mixtures, Cavestri (1995) also provided the equilibrium liquid mass fractions of each individual component, which were obtained via gas chromatography.

In a paper related to the subject of the present investigation, Yana Motta *et al.* (2002) analyzed the flow of a R-404A/POE oil mixture through a glass capillary tube, with special emphasis on the behavior of the vaporization point. The oil mass fraction ranged from 5.6 to 6.9% for a condensing pressure of 1825 kPa and subcooling ranging from 6.2 to 21.5^{o}C. They reported a shortening of the liquid region and a decrease of the critical flow rate for the R-404A/oil cases in comparison with those of pure R-404A. These results were attributed to the increase of the viscosity of the R-404A/oil mixture, which was the physical property that exhibited the largest variation with respect to the pure refrigerant.

Lebretton *et al.* (2001) developed an ultrasonic device to measure *in situ* and in real time the POE oil concentration in liquid R-410A in a calorimetric installation. The authors reported that the method required a time-consuming calibration due to the sensitive nature of the speed-of-sound measurement on the nature and concentration of both refrigerant and lubricant.

More recently, Zhang and He (2010) measured the kinematic viscosities of liquid mixtures of lubricant oil and R410A and R407C at the temperatures ranging from 256.15 to 336.15 K and mass fractions of POE oil of 2, 5, 8, and 10×10^{-5}. Despite the very small lubricant mass fractions, the experimental results show that these have a significant impact on the kinematic viscosity of R410A and R407C refrigerant-oil mixtures, with the mixture viscosity increasing with the refrigerant concentration. The authors have proposed a simple empirical correlation to take into account the variation of the mixture viscosity with both temperature and oil concentration.

The purpose of this paper is to investigate two widely used liquid viscosity models, i.e., the Grunberg and Nissan (1949) and the Katti and Chaudry (1964) models, in terms of their prediction of the viscosity of a mixture of R-404A (44% of R-125, 52% of R-143a and 4% of R-134a by weight) and the 32 ISO branched POE oil (Cavestri, 1995) for temperatures of 20, 40 and 60^{o}C. R-404A is a non-azeotropic blend with a temperature glide that spans from approximately 0.5^{o}C at 20^{o}C to slightly above 0.2^{o}C at 60^{o}C (Lemmon *et al.*, 2008). The experimental data on the liquid mixture viscosity, together with liquid solubility and mixture density, were obtained from the reference database provided by Cavestri (1995). Six correlation methods have been tested, and the best results were given by the correlations which treated R-404A as a pseudo-component and had only two fitting parameters.

2. CORRELATION METHODS

The Eyring theory (Glasstone *et al.*, 1941) for the viscosity of pure liquids can be extended for *binary* mixtures as follows (Oswal and Desai, 2001),

$$\eta V = N_a h \exp\left(\frac{x_1 G_1^+ + x_2 G_2^+ + G^{E+}}{RT}\right) \tag{1}$$

or, in a more suitable form,

$$\ln(\eta V) = x_1 \ln(\eta_1 V_1) + x_2 \ln(\eta_2 V_2) + \frac{G^{E+}}{RT} \tag{2}$$

Katti and Chaudry (1964) modeled the activation energy for viscous flow in Eq. (2) using a polynomial Redlich-Kister type expansion given by,

$$\frac{G^{E+}}{RT} = \frac{D_0 x_1 x_2}{RT} + \frac{D_1(2x_1-1)(x_1-x_1^2)}{RT} + \frac{D_2(2x_1-1)^2(x_1-x_1^2)}{RT} + \cdots \tag{3}$$

where D_i are fitting parameters with i = 0,1,2,...N. The Grunberg and Nissan (1949) model assumes an ideal solution behavior ($V = x_1V_1 + x_2V_2$) in which the mixture viscosity is correlated from,

$$\ln(\eta) = x_1 \ln(\eta_1) + x_2 \ln(\eta_2) + G_0 x_1 x_2 + G_1(2x_1-1)(x_1-x_1^2) + G_2(2x_1-1)^2(x_1-x_1^2) + \cdots \tag{4}$$

For multicomponent mixtures, the Grunberg and Nissan (1949) correlation can be cast in the following form,

$$\ln(\eta) = \sum_{i=1}^{N} x_i \ln(\eta_i) + \sum_{i=1}^{N}\sum_{j>1}^{N} x_i x_j G_{i,j} \tag{5}$$

In the present paper, six correlation methods have been tested, which consisted of variations of the Grunberg and Nissan (1949) and Katti and Chaudry (1964) correlations for the mixture R-404A/ISO 32 POE. In the *first method*, R-404A is treated as a pseudo-component and the polynomial Redlich-Kister expansion is truncated in the first term as follows,

$$\ln(\eta) = x_O \ln(\eta_O) + x_R \ln(\eta_R) + G_{O,R} x_O x_R \tag{6}$$

The *second method* consists of truncating the polynomial Redlich-Kister expansion in the second term. Thus,

$$\ln(\eta) = x_O \ln(\eta_O) + x_R \ln(\eta_R) + G_{O,R} x_O x_R + (2x_R-1)(x_R-x_R^2)C_{O,R} \tag{7}$$

where $G_{O,R}$ and $C_{O,R}$ are empirically determined coefficients. The *third method* takes into account the individual mole fractions of each component in the liquid phase, which have been made available by the gas chromatography data of Cavestri (1995). Thus, with the Redlich-Kister expansion truncated in the first terms, equation (5) can be written as,

$$\begin{aligned}\ln(\eta) = {} & x_O \ln(\eta_O) + x_1 \ln(\eta_1) + x_2 \ln(\eta_2) + x_3 \ln(\eta_3) + G_{O,1} x_O x_1 + G_{O,2} x_O x_2 + G_{O,3} x_O x_3 + G_{1,2} x_1 x_2 + \\ & G_{1,3} x_1 x_3 + G_{2,3} x_2 x_3\end{aligned} \tag{8}$$

where $G_{i,j}$ are empirically determined coefficients and the subscripts 1, 2 and 3 refer to R-125, R-143a and R-134a, respectively. The *fourth method* is an extension of the third in the sense that the polynomial expansion has been truncated in the second terms as follows,

$$\begin{aligned}\ln(\eta) = x_O \ln(\eta_O) + x_1 \ln(\eta_1) + x_2 \ln(\eta_2) + x_3 \ln(\eta_3) + G_{O,1} x_O x_1 + G_{O,2} x_O x_2 + G_{O,3} x_O x_3 + \\ G_{1,2} x_1 x_2 + G_{1,3} x_1 x_3 + G_{2,3} x_2 x_3 + G_{O,1,2} x_O x_1 x_2 + G_{O,1,3} x_O x_1 x_3 + \\ G_{O,2,3} x_O x_2 x_3 + G_{1,2,3} x_1 x_2 x_3\end{aligned} \tag{9}$$

where $G_{i,j}$ and $G_{i,j,k}$ are empirically determined coefficients. The *fifth method* consists of the Katti and Chaudry (1964) model with a truncation of the first term of the polynomial expansion. Thus,

$$\ln(\eta V) = x_O \ln(\eta_O V_O) + x_R \ln(\eta_R V_R) + x_O x_R \frac{W_{vis}}{RT} \tag{10}$$

where R is the universal gas constant and W_{vis} is a empirically determined coefficient. Finally, the *sixth method* takes into account the second term of the polynomial Redlich-Kister expansion of the Katti and Chaudry (1964) model. Thus,

$$\ln(\eta V) = x_O \ln(\eta_O V_O) + x_R \ln(\eta_R V_R) + x_O x_R \frac{W_{vis,1}}{RT} + (2x_R - 1)(x_R - x_R^2)\frac{W_{vis,2}}{RT} \tag{11}$$

where $W_{vis,1}$ and $W_{vis,2}$ are empirically determined coefficients.

The correlations for the density and kinematic viscosity of the pure oil has been obtained from the work of Seeton and Hrnjak (2006) as follows,

$$\rho_O = 985 - (T - 273.15) \tag{12}$$

and

$$\nu_O = 55605.5 - 621.253T + 2.60379T^2 - 4.8504 \times 10^{-3} T^3 + 3.38835 \times 10^{-6} T^4 \tag{13}$$

where T is the absolute temperature and the units of density and kinematic viscosity are kg/m^3 and cSt, respectively. The properties of pure R-404A were obtained from the refrigerant properties database embedded into the EES platform (Klein, 2007), which was used for the computational implementation of the six correlation methods. The prediction ability of the correlations was evaluated via the Absolute Average Deviation, AAD, defined as,

$$\text{AAD} = \frac{100}{n} \sum_{i=1}^{n} \left| \frac{\eta_{cal,i} - \eta_{\exp,i}}{\eta_{\exp,i}} \right| \tag{14}$$

where the subscripts *cal* and *exp* refer to the calculated and experimental values, respectively.

3. RESULTS

The results of the first method are shown in Figure 1. The predictions are reasonably good for the lower refrigerants mole fractions, but generally underpredict the experimental data as the refrigerant mole fraction increases. The fitting parameter $G_{O,R}$ has been found equal to 6.291 and the resulting absolute average deviation was calculated as 27.4%. As an attempt to improve the prediciton ability of the correlation methods, the second method predicts the experimental data with an average deviation of 9.8%, which is a significant improvement considering that only one additional fitting parameter has been added to the Grunberg and Nissan (1949) correlation. In the second method, the fitting parameters $G_{O,R}$ and $C_{O,R}$ are equal to 7.323 and 5.827, respectively.

In the third method, the components of R-404A are treated individually, so six binary interaction parameters for the refrigerants and the oil have been incorporated in the Redlich-Kister expansion. The fitting parameters of the third method are shown in Table 1.

Table 1. Interaction parameters of the third correlation method.

$G_{O,1}$	$G_{O,2}$	$G_{O,3}$	$G_{1,2}$	$G_{1,3}$	$G_{2,3}$
1	2.703	30	2.4	0.9716	42.78

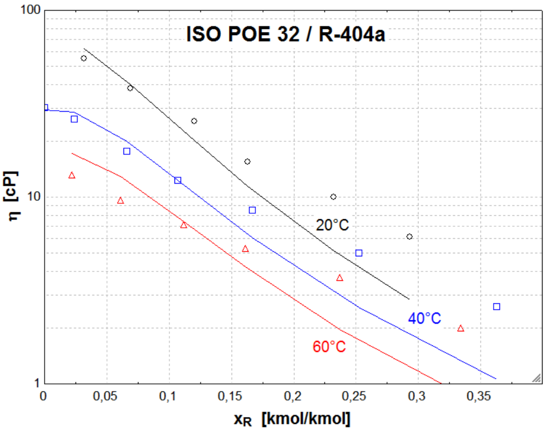

Figure 1. Predictions of the first method.

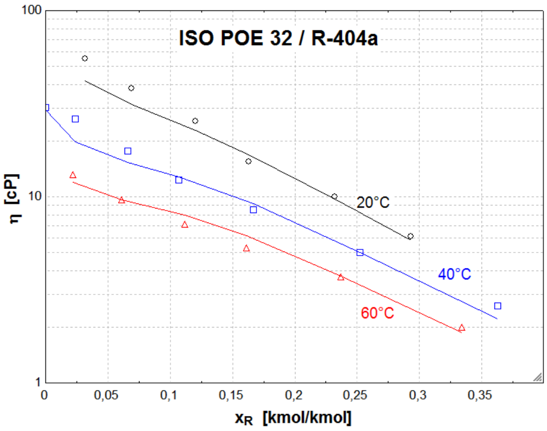

Figure 2. Predictions of the second method.

The third method, whose results are shown in Fig. 3, predicts the experimental data with an average error of 10.5%. Therefore, the increased complexity of the model is not justified. The fourth method predictions are shown in Fig. 4. As mentioned above, this correlation method is an extension of the third method in the sense that the polynomial expansion has been truncated in the second terms. The eleven fitting parameters of this correlation are shown in Table 2.

Table 2. Interaction parameters of the fourth correlation method.

$G_{O,1}$	$G_{O,2}$	$G_{O,3}$	$G_{1,2}$	$G_{1,3}$	$G_{2,3}$	$G_{O,1,2}$	$G_{O,1,3}$	$G_{O,2,3}$	$G_{1,2,3}$	$G_{O,1,2,3}$
0.88	3.045	17.44	9.905	1.192	41.67	10.52	-18.48	41.96	-137.2	-164.8

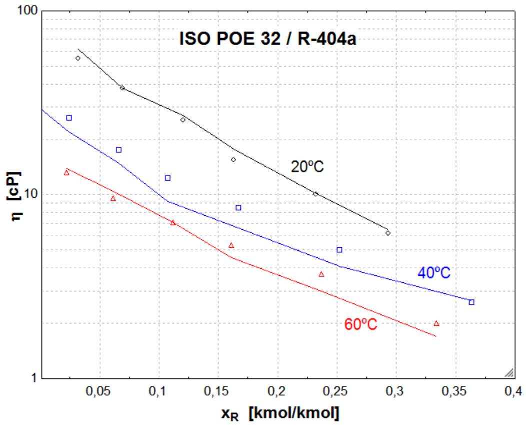

Figure 3. Predictions of the third method.

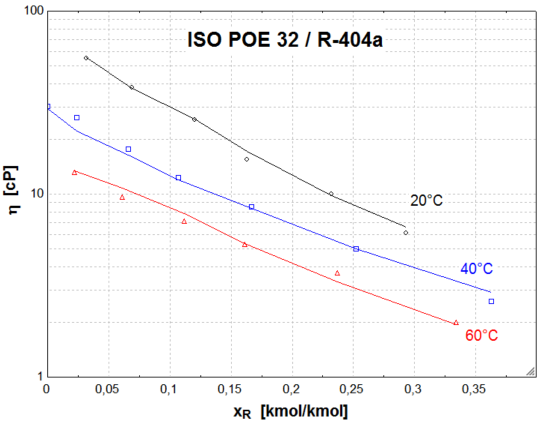

Figure 4. Predictions of the fourth method.

The absolute average deviation of the fourth method is 5.36%, which is a considerable improvement, but in a way also unsatisfactory due to the substantial increase in the number of fitting parameters. Once again, the small reduction of the prediction error is not justified.

The fifth method incorporates the molar volumes of the individual components and of the mixture in the correlation, since it is based on the correlation due to Katti and Chaudry (1964). The prediction ability of this method, which contains only one fitting parameter can be appreciated in Fig. 5. The absolute average deviation of the fifth method has been calculated at 30.2%, and the fitting parameter is equal to 20457. It seems that the incorporation of the effect of the molar volume has not brought any advantage (at least for the methods with only one fitting parameter), since the error has increased with respect to the first method by approximately 3%. Like the first method, the prediction at higher mole fractions is also poor.

The results of the sixth correlation method are shown in Fig. 6. This method is also based on the Katti and Chaudry (1964), but with the polynomial Redlich-Kister expansion truncated in the second term so that the model constants are W_{vis1} = 22616 and W_{vis2} = 16952. The absolute average deviation associated with the sixth method is 10.38% which, like the second method, makes it quite attractive for correlating the viscosity data of R-404A/ISO POE 32 mixtures due to the lack of complexity.

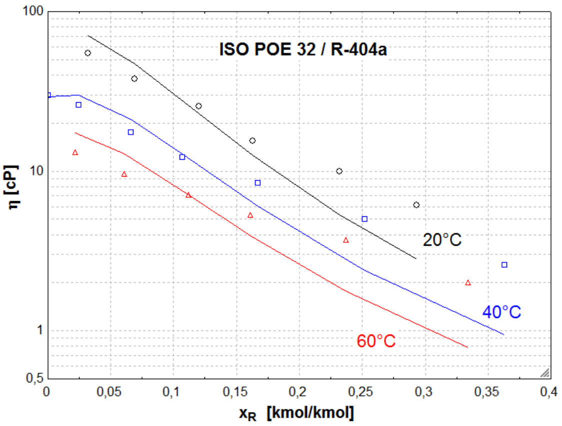

Figure 5. Predictions of the fifth method.

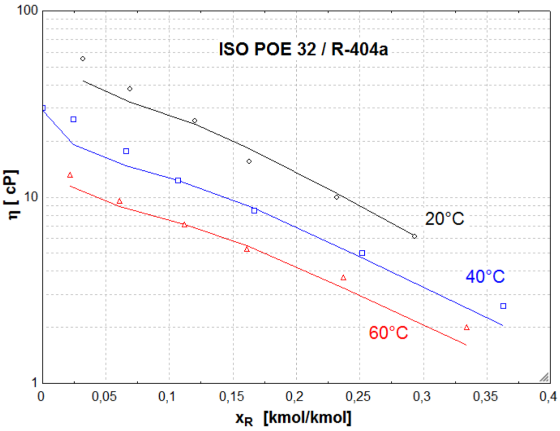

Figure 6. Predictions of the sixth method.

4. CONCLUSIONS

In the present paper, we have performed a comparison of six correlation methods for the liquid viscosity of a mixture of the near azeotropic blend R-404A (44% R-125, 52% R-143a and 4% R-134a by weight) and the 32 ISO POE lubricant oil. The experimental data were obtained from the extensive database of Cavestri (1995), which also contained information on the composition of the liquid mixture as a function of temperature and pressure. The results show that the best correlation methods (i.e., those that provide the smallest absolute average deviation) are those that treat R-404A as a pseudo-component, thus not taking into account the concentration of each individual component in the liquid phase. In this sense, the recommended methods are the second and sixth correlations, i.e., the Grunberg and Nissan (1949) and Katti and Chaudry (1964), both with polynomial Redlich-Kister expansions of the activation energy for viscous flow truncated in the second term.

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