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Viscosity Behavior of Mixtures of CO₂ and Lubricant Oil

Experimental data on the viscosity of mixtures of CO₂ and lubricant oil were acquired and correlated using an excess-property approach based on the classical Eyring liquid viscosity model. Three oils of different types and viscosity grades (alkylbenzene AB ISO 32, mineral MO ISO 50 and polyol ester POE ISO 68) were evaluated at temperatures ranging from 36.5 to 82°C. The excess activation energy for viscous flow was successfully correlated as a function of temperature and concentration using Redlich-Kister polynomial expansions with up to three terms. Large departures from the ideal solution viscosity behavior have been identified in all mixtures. The nature of the observed deviations has been explored in the light of their dependence on temperature, refrigerant concentration and oil type. The Katti and Chaudry (1964) model of the activation energy of viscous flow displayed the best correlation of the experimental data, with RMS deviations of 4.6% (AB ISO 32), 3.3% (MO ISO 50) and 2.8% (POE ISO 68).

Keywords: CO₂-lubricant oil mixture, viscosity, experimental data, modeling

Introduction

There are several refrigeration applications where CO₂ (R-744) is well suited as a primary refrigerant (Pettersen, 1999). Due to its unique characteristics, such as chemical stability, non-toxicity, non-flammability, low-cost, low GWP (Global Warming Potential) and zero ODP (Ozone Depletion Potential), the use of CO₂ in some specific applications appears especially promising in a global scenario where more stringent environmental legislation is applied.

As far as CO₂ refrigeration systems are concerned, the compressor design requires special attention due to the inherently lower system performance encountered in the CO₂ trans-critical cycle when compared with the standard sub-critical cycle usually associated with other refrigerants (Gosney, 1982). In this context, a crucial aspect of the CO₂ compressor design is related to the performance and reliability of the bearings system. It is of utmost importance that the variation of the physical properties of the lubricant mixture with respect to temperature and refrigerant concentration are taken into account during all stages of the system design. Despite the increasing number of publications dealing with the determination of physical properties of mixtures of CO₂ and lubricant oils and some of their precursors (Yokozeki, 2007; Bobbo et al., 2007; Pensado et al., 2008), more research is still needed before the most suitable lubricants, with specific lubrication characteristics for a variety of CO₂ refrigeration applications, are finally selected.

Recent appraisals of the experimental and theoretical work on the determination of the physical properties of mixtures of CO₂ and lubricant oil have been presented by Seeton and Hrnjak (2006), Bobbo et al. (2006), Marcelino Neto and Barbosa (2007) and Yokozeki (2007). Undoubtedly, in the open literature, the most extensively studied mixtures of CO₂ and oil are those involving polyol esters (POE) and polyalkylene glycols (PAG). Significantly less work has been devoted to characterizing mixtures of CO₂ and other oil types, such as mineral (MO), polyalphaolefin (PAO) and alkylbenzene (AB). Moreover, the number of works in which the viscosity behavior of CO₂-lubricant mixtures is investigated in detail is still rather limited.

The objective of this paper is to present experimental data on the liquid viscosity of mixtures of CO₂ and three lubricants, namely, a POE ISO 68, a MO ISO 50 and an AB ISO 32. The data points were obtained at pressures between 0.9 and 5.7 MPa and temperatures between 36.5 and 82°C. The liquid mixture viscosity was correlated

using an excess-property approach based on the classical Eyring liquid viscosity model. The Eyring theory correlations of Grunberg and Nissan (1949), Katti and Chaudry (1964) and McAllister (1960) have been utilized. The best agreement between model and experimental data was obtained with the Katti and Chaudry (1964) equation.

Nomenclature

G_i^+	= Activation energy for viscous flow, J/kmol
G^{E+}	= Excess activation energy for viscous flow, J/kmol
h	= Planck's constant [6.626×10^{-34}], Js
M	= Molar mass, kg/kmol
N_a	= Avogadro's number [6.023×10^{26}], kmol ⁻¹
P	= Pressure, MPa
R	= Universal gas constant [8.314], kJ/kmol.K
T	= Temperature, K or °C
V	= Molar volume, m ³ /kmol
V_i	= Molar volume of component i , m ³ /kmol
w_i	= Mass fraction of component i , kg/kg
x_i	= Mole fraction of component i , kmol/kmol

Greek Symbols

η	= Dynamic viscosity, cP or mPa.s
ν	= Kinematic viscosity, m ² /s
ρ	= Mass density, kg/m ³

Experimental Work

Experimental Apparatus and Procedure

The experimental facility is schematically illustrated in Fig. 1 (Marcelino Neto and Barbosa, 2007, 2009). A specified amount of lubricant oil is placed in the 4 L equilibrium cell (2). A vacuum of 0.04 mbar is generated in the apparatus to remove moisture and dissolved gases. An initial amount of refrigerant is fed into the cell. The system temperature is set by a thermostatic bath (1) that circulates service water through a tank (6) in which the equilibrium cell is fully immersed. In the present experiments, the pressure of the oil-refrigerant mixture is, therefore, the dependent variable. The equilibrium cell is instrumented for absolute pressure, P , and the temperature of the fluids in the cell is recorded by three type-T thermocouples (T_1 , T_2 , T_3) located at three distinct heights to measure the temperatures of the liquid and vapor phases. A gear

pump (5) moves the liquid oil-refrigerant mixture through the experimental facility. The speed of the electrical motor is set at its minimum value (12 Hz). The mixture first flows through a Coriolis-type mass flow transducer (4) that records flow rate, temperature and liquid density. Then, an oscillating piston viscometer (3) registers temperature and dynamic viscosity of the liquid mixture. The solubility of the mixture is measured gravimetrically using a liquid mixture sample collected in a 150 mL cylinder (7). The experimental apparatus is integrated with a signal conditioning module (8) and a computerized system for data acquisition and treatment (9). The tank (top, sides and bottom), connection tubing and instrumentation (Coriolis flow meter, pump and sampling cylinder) are thermally insulated to prevent heat loss. The temperature variation between the viscometer, the mass flow/density meter and test cell was within the uncertainty level set during the calibration of the thermocouples. The experimental procedure for obtaining the mixture solubility has been outlined in Marcelino Neto and Barbosa (2007, 2009).

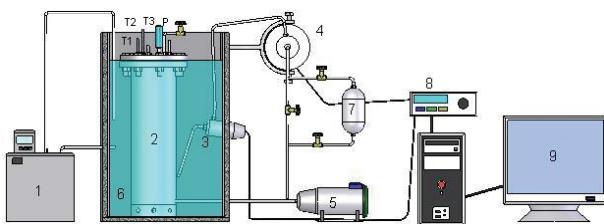


Figure 1. Schematic representation of the experimental rig.

The temperature measurement uncertainty was estimated at $\pm 0.2^\circ\text{C}$ (68% confidence level). The uncertainty of the density measurements (estimated from the manufacturer specifications) was $\pm 1\%$ of the absolute reading (95% confidence level). The uncertainty of the viscosity measurements (estimated from the manufacturer specifications) was $\pm 1\%$ of the full scale (68% confidence level). It should be observed that, because of interchangeable pistons, the uncertainty is $\pm 0.1 \text{ cP}$ for the 0.1-10 cP piston, and $\pm 1 \text{ cP}$ for the 1-100 cP piston. The uncertainty of the pressure transducer (estimated from the manufacturer specifications) was $\pm 0.15\%$ FS (0.3 bar) and that of the balance was $\pm 0.3 \text{ g}$. After an error propagation analysis (Marcelino Neto, 2006), the uncertainty in the solubility measurement was determined at $\pm 0.5 \text{ g/kg}$. The experimental procedure was validated with vapor pressure and density measurements of pure CO₂ and with density and viscosity of an ISO 10 lubricant oil, whose properties have been made available from its manufacturer (Marcelino Neto and Barbosa, 2007).

Experimental Conditions

The experimental conditions of the present experiments are as follows. Solubility, liquid density and viscosity of CO₂/MO ISO 50, CO₂/AB ISO 32 and CO₂/POE ISO 68 oil mixtures were measured at temperatures between 36.5 and 82°C. CO₂ was supplied by AGA (99.9% pure) and the lubricants were supplied by Embraco. The molar masses of the lubricants are 355 kg/kmol (MO), 375 kg/kmol (AB) and 763 kg/kmol (POE). The molecular structures of the lubricants have not been disclosed to the present authors as they represent proprietary information.

Viscosity Modeling

Liquid Viscosity Models

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):

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

or in a more suitable form:

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

where η is the mixture dynamic viscosity, η_i is the dynamic viscosity of component i , V is the mixture molar volume, V_i is the molar volume of component i , N_a is the Avogadro constant, h the Planck constant, R is the universal gas constant, T is the absolute temperature, x_i is the mole fraction of component i , G_i^+ is the activation energy for viscous flow of component i and G^{E+} is the excess activation energy for viscous flow. Subscripts 1 and 2 stand for the refrigerant and lubricant oil, respectively.

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

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

where D_i are constants with $i = 0, 1, 2, \dots, N$. In the present work, the Katti and Chaudry expansion was truncated in the third term.

The Grunberg and Nissan (1949) model assumes an ideal solution behavior ($V = x_1 V_1 + x_2 V_2$) in which the mixture viscosity is correlated from

$$\begin{aligned} \ln(\eta) = & x_1 \ln(\eta_1) + x_2 \ln(\eta_2) + G_0 x_1 x_2 + G_1 (2x_1 - 1)(x_1 - x_1^2) + \\ & + G_2 (2x_1 - 1)^2 (x_1 - x_1^2) + \dots \end{aligned} \quad (4)$$

In the present paper, the Grunberg and Nissan correlation has been employed in two different ways. In the first approach, henceforth referred to as the G-N approach, G_0 is correlated as a linear function of the CO₂ mole fraction for each temperature (Tomida et al., 2007):

$$G_0 = \Gamma_0(T) + \Gamma_1(T)x_1 \quad (5)$$

and the remaining coefficients G_i are assumed equal to zero. In the second approach, the G-N2 approach, the series are truncated in the third term and the coefficients G_0 , G_1 and G_2 are assumed independent of temperature and are determined from a best-fit to the experimental data.

The McAllister (1960) liquid mixture viscosity model is based on Eyring's theory of absolute reaction rates. For a binary mixture, the McAllister semi-theoretical equation is written as:

$$\begin{aligned} \ln \nu = & x_1^4 \ln \nu_1 + 4x_1^3 x_2 \ln \nu_{1112} + 6x_1^2 x_2^2 \ln \nu_{1122} + 4x_1 x_2^3 \ln \nu_{2221} x_2^4 \ln \nu_2 \\ & - \ln \left[x_1 + x_2 \frac{M_2}{M_1} \right] + 4x_1^3 x_2 \ln \left[\frac{3}{4} + \frac{M_2}{4M_1} \right] + 6x_1^2 x_2^2 \ln \left[\frac{1}{2} + \frac{M_2}{2M_1} \right] \\ & + 4x_1 x_2^3 \ln \left[\frac{1}{4} + \frac{3M_2}{4M_1} \right] + x_2^4 \ln \left[\frac{M_2}{M_1} \right] \end{aligned} \quad (6)$$

where ν is the kinematic viscosity of mixture, ν_i is the kinetic viscosity of pure component i at the same temperature and pressure as the mixture, and M_i is the molar mass. ν_{1112} , ν_{1122} , ν_{2221} are fitting coefficients calculated from a best-fit to the experimental data.

Implementation

The models were implemented in the Engineering Equation Solver – EES (Klein, 2007), and the genetic algorithm routines available in EES were employed in the determination of the models' coefficients. The physical properties of pure CO₂ were obtained from EES and the properties of the pure lubricants (density and viscosity) were derived from polynomial fittings to our own experimental data. To evaluate the prediction ability of the correlations, the following quantities can be defined

$$\text{RMS} = \frac{100}{n} \sqrt{\sum_{i=1}^n \frac{(\eta_{\text{cal},i} - \eta_{\text{exp},i})^2}{\eta_{\text{exp},i}^2}} \quad (7)$$

$$\text{AAD} = \frac{100}{n} \sum_{i=1}^n \left| \frac{\eta_{\text{cal},i} - \eta_{\text{exp},i}}{\eta_{\text{exp},i}} \right| \quad (8)$$

$$\text{Bias} = \frac{100}{n} \sum_{i=1}^n \frac{\eta_{\text{cal},i} - \eta_{\text{exp},i}}{\eta_{\text{exp},i}} \quad (9)$$

where, in the McAllister (1960) model, η is substituted by ν in the above equations.

Results and Discussion

The experimental data for the mixture viscosity, density and molar solubility are presented in Table 1 as a function of pressure and temperature. The model predictions are summarized in Table 2, where the search intervals employed in the calculation of the fitting parameters are also presented.

A direct comparison between the G-N2 model and the Katti and Chaudry (1964) correlation (both with a three-term expansion of the activation energy parameter) indicates that the latter has predicted

the data with a better agreement. Although there may be several reasons for this particular result, perhaps the most apparent explanation seems to be the fact that the severe non-ideality of the mixture molar volume is not considered in the Grunberg and Nissan (1949) model. The McAllister (1960) model does not perform well under the conditions evaluated in the present work.

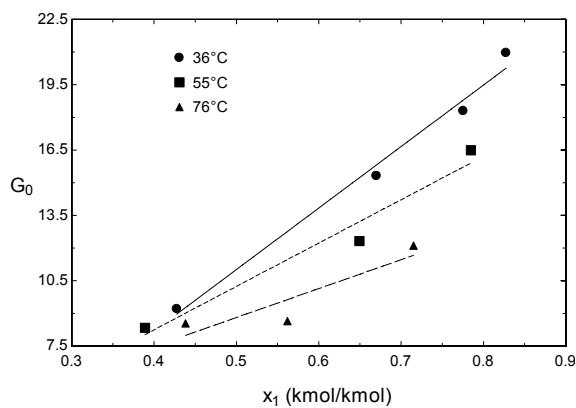
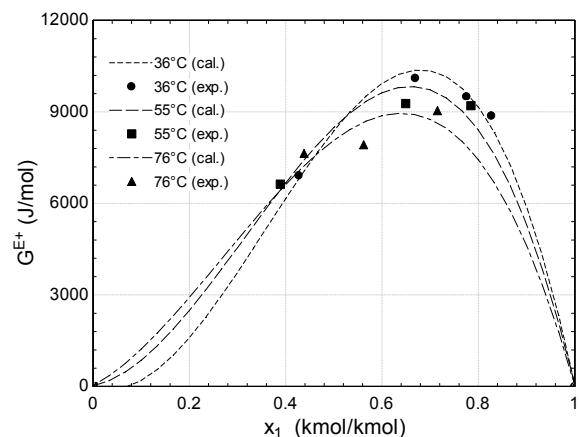
Contrary to what has been observed for the CO₂/MN ISO 50 and CO₂/AB ISO 32 mixtures, the performance of Grunberg and Nissan (1949) for the CO₂/POE ISO 68 mixture does not necessarily improve when the temperature dependence approach of Tomida et al. (2007) is adopted. In Table 2, the correlations for G_0 as a function of temperature are presented as a function of increasing temperature from top to bottom and Figures 2 to 4 present the dependence of G_0 on temperature and composition and the variations of G^{E^+} (the activation energy of Katti and Chaudry) and of G (the three term expansion on the right hand side of the Grunberg and Nissan model – Eq. 4) as a function of temperature and composition for the CO₂/POE ISO 68 mixture. Positive deviations are observed at all temperatures, which characterize stronger interactions between molecules of different species (Grunberg and Nissan, 1949).

Table 1. Experimental data.

Mixture	T(°C)	P(MPa)	x _i (kmol/kmol)	ρ (kg/m ³)	η (cP)
R-744 + POE ISO 68	36.5	0.9	0.4275	965.3	26.6
		2.2	0.6692	967.5	12.8
		4.1	0.7755	968.7	4.6
		5.2	0.8274	968.8	2.5
	55.5	1.1	0.4381	952.7	16.6
		3.4	0.5619	952.3	6.6
		5.8	0.7150	950.6	2.7
		7.6.5	1.1	0.3890	938.7
	76.5	3.4	0.6493	938.0	4.8
		5.7	0.7846	930.4	2.7
		40.5	1.0	0.2070	911.6
		3.4	0.3653	914.9	9.9
R-744 + MO ISO 50	59.7	5.5	0.5085	917.6	4.3
		1.2	0.1417	902.0	11.3
		3.4	0.3540	904.2	5.9
		5.6	0.4237	905.0	2.7
	81.5	1.2	0.1520	898.1	5.6
		3.8	0.2946	892.9	2.8
		5.5	0.4044	895.9	1.9
		40.5	1.4	0.2512	873.1
R-744 + AB ISO 32	57.7	3.6	0.5173	861.4	8.7
		5.1	0.6681	869.7	4.3
		1.3	0.1942	864.2	12.4
		3.5	0.4053	856.8	5.5
	69.2	5.4	0.5286	866.6	3.6
		1.4	0.2115	859.1	7.0
		3.5	0.3611	859.1	3.8
		5.5	0.4702	858.7	2.0

Table 2. Summary of the data correlation.

Mixture	Model	Parameter	Search interval	RMS (%)	AAD (%)	Bias (%)
R-744 + POE ISO 68	G-N2	$G_0 = 22.09$	-10000 to 10000	5.0	13.2	-5.9
		$G_1 = 5.12$				
		$G_2 = 8.47$				
	G-N	$G_0 = -3.1 + 28.23x_1$	-10000 to 10000	2.9	5.8	0.6
		$G_0 = 0.29 + 19.90x_1$		5.2	8.4	0.7
		$G_0 = 2.18 + 13.26x_1$		6.3	9.7	1.2
	Katti- Chaudry	$D_0 = 29438$	-100000 to 100000	2.8	6.1	-3.8
		$D_1 = 15907$				
		$D_2 = 51093$				
	McAllister	$\nu_{1112} = 1$	-10000 to 10000	10.9	27.7	-10.4
		$\nu_{1122} = 1.8$				
		$\nu_{2221} = 768.7$				
R-744 + MO ISO 20	G-N2	$G_0 = 8.29$	-10000 to 10000	3.6	10.2	1.4
		$G_1 = 2.64$				
		$G_2 = 4.22$				
	G-N	$G_0 = 5.35 - 1.04x_1$	-10000 to 10000	3.0	4.7	0.4
		$G_0 = 4.39 - 1.62x_1$		5.4	8.5	0.8
		$G_0 = 7.54 - 9.84x_1$		3.8	5.9	0.5
	Katti- Chaudry	$D_0 = 15646$	-100000 to 100000	3.3	6.8	1.4
		$D_1 = 4997$				
		$D_2 = 6543$				
	McAllister	$\nu_{1112} = 932$	-10000 to 10000	11.4	33.9	-17.8
		$\nu_{1122} = 1$				
		$\nu_{2221} = 106.5$				
R-744 + AB ISO 32	G-N2	$G_0 = 16.75$	-10000 to 10000	6.3	16.0	-6.6
		$G_1 = 12.16$				
		$G_2 = 5.03$				
	G-N	$G = 1.58 + 13.9x_1$	-10000 to 10000	2.6	4.0	0.5
		$G = 4.71 + 3.16x_1$		2.3	3.6	0.3
		$G = 2.14 + 1.68x_1$		1.2	1.9	-0.02
	Katti- Chaudry	$D_0 = 16364$	-100000 to 100000	4.6	10.6	1.1
		$D_1 = 30433$				
		$D_2 = 46503$				
	McAllister	$\nu_{1112} = 57.5$	-10000 to 10000	11.4	33.9	-17.8
		$\nu_{1122} = 1$				
		$\nu_{2221} = 6.6$				

Figure 2. Dependence of G_0 on mole fraction and temperature for the $\text{CO}_2/\text{POE ISO 68}$ mixture.Figure 3. Correlation of the activation energy of Katti and Chaudry (1964) for the $\text{CO}_2/\text{POE ISO 68}$ mixture.

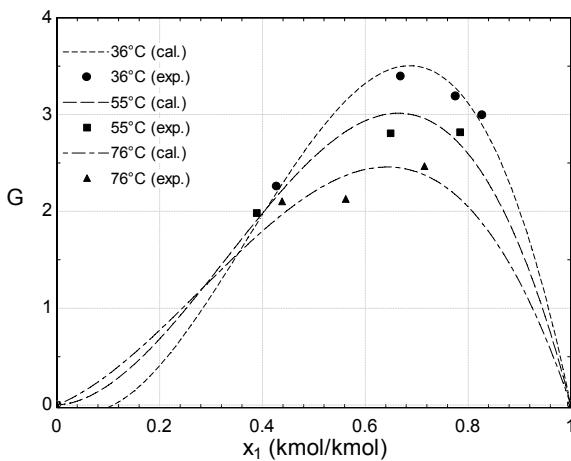


Figure 4. Correlation of the non-ideal three term expansion parameter of the Grunberg and Nissan (1949) correlation (CO₂/POE ISO 68 mixture).

The mass density of CO₂/MO ISO 50 and CO₂/AB ISO 32 as a function of temperature is presented in Figs. 5 and 6, where a significant departure from the ideal mixture behavior (calculated from Eq. 10) is clearly observed.

$$\frac{1}{\rho^{id}} = \frac{\sum x_i V_i}{\sum x_i M_i} \quad (10)$$

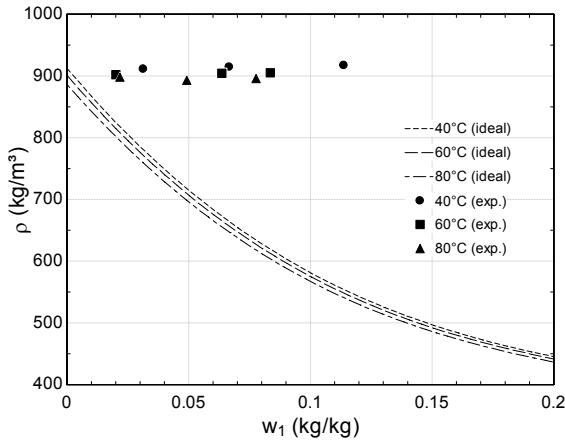


Figure 5. Experimental liquid mass density of the CO₂/MO ISO 50 mixture.

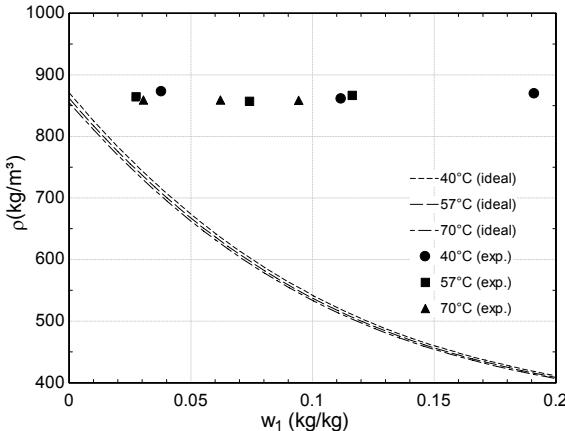


Figure 6. Experimental liquid mass density of the CO₂/AB ISO 32 mixture.

Figures 7 to 9 present comparisons between the Katti and Chaudry (1964) correlation and the G-N approach for the three mixtures. Despite the better agreement of the latter model for the CO₂/MO ISO 50 and CO₂/AB ISO 32 mixtures, the Katti and Chaudry (1964) model is recommended for all three mixtures, because it is easier to apply at intermediate temperatures that have not been correlated during the experiments.

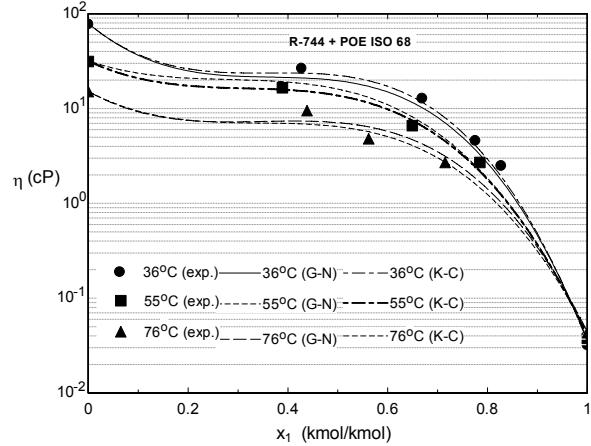


Figure 7. Viscosity of the CO₂/POE ISO 68 mixture and comparison with the Katti and Chaudry (1964) and G-N models.

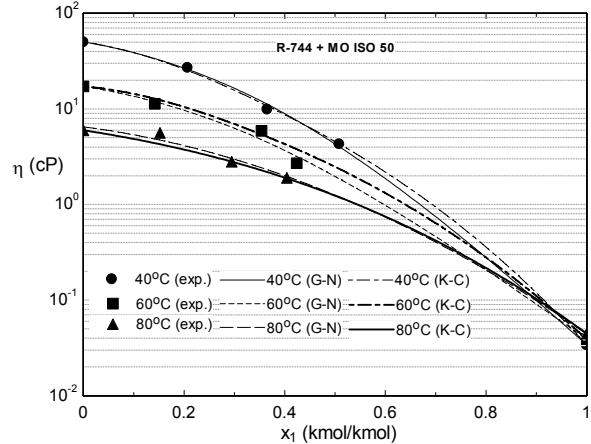


Figure 8. Viscosity of the CO₂/MO ISO 50 mixture and comparison with the Katti and Chaudry (1964) and G-N models (Grunberg and Nissan, 1949).

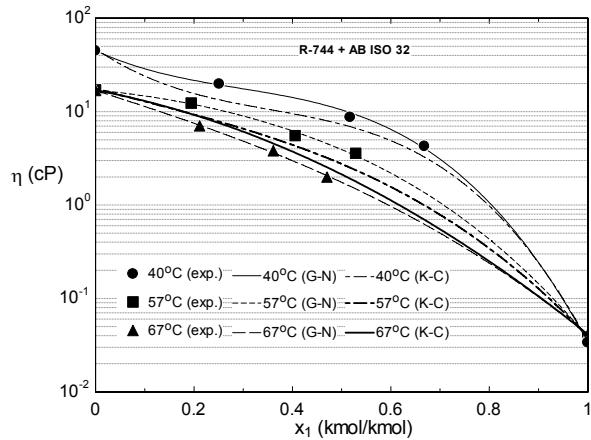


Figure 9. Viscosity of the CO₂/AB ISO 32 mixture and comparison with the Katti and Chaudry (1964) and G-N models (Grunberg and Nissan, 1949).

Conclusions

The present paper put forward new data on the viscosity of a mixture of CO₂ (R-744) and three lubricant oils (POE ISO 68, MO ISO 50 and AB ISO 32). An experimental facility that enables the simultaneous measurement of the viscosity, density and solubility has been utilized. The data were obtained at three different temperatures in the range of 36.5 and 82°C. The viscosity data have been correlated with the Grunberg and Nissan (1949), Katti and Chaudry (1964) and McAllister (1960) correlations. When a three-term polynomial expansion of the excess activation energy with temperature independent coefficients is employed in the Grunberg and Nissan (1949) and Katti and Chaudry (1964) models, the latter presents a better correlation of the experimental data. Contrary to what has been observed for the MN ISO 50 and AB ISO 32 mixtures, using the Grunberg and Nissan (1949) approach with temperature dependent coefficients does not improve the prediction for the POE ISO 68 mixture. The performance of the McAllister (1960) correlation has been less satisfactory than those of the other models.

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