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Solubility, density and viscosity of mixtures of isobutane (R-600a) and a linear alkylbenzene lubricant oil

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ABSTRACT

Isobutane (R-600a) is a well established environmentally friendly substitute for hydrofluorocarbon and chlorofluorocarbon refrigerants in vapor compression refrigeration applications. In order to improve the performance and reliability of compressors and refrigerators, the behavior of the thermophysical properties of refrigerant–lubricant mixtures must be well understood, as the choice of lubricant oil plays an important role in determining the system equalizing pressure and in minimizing the friction losses in the compressor. Solubility, density and viscosity data for a system (i.e., several mixtures of the same components) of R-600a and a linear alkylbenzene lubricant oil (LAB ISO 5) were obtained experimentally at temperatures between 23 and 80 °C. The solubility and density data were correlated with the Peng–Robinson equation of state with a single interaction parameter. The dynamic viscosity data were predicted with an Eyring-type model in which the excess activation energy for viscous flow was modeled in terms of the excess Gibbs free energy derived from the Peng–Robinson equation of state.

1. Introduction

Isobutane (R-600a) is widely recognized as a substitute for hydrofluorocarbons (HFCs) and chlorofluorocarbons (CFCs) in household and small capacity vapor compression refrigeration. As with any other refrigerant, its absorption in the lubricant oil inside the compressor causes a significant departure from the pure lubricant thermodynamic and transport properties, such as solubility and dynamic viscosity. Since the compressor thermodynamic efficiency and reliability are strongly dependent on the lubrication efficacy under steady and transient conditions, a precise prediction of the properties of refrigerant–oil mixtures is a critical aspect of compressor design.

The literature on thermophysical properties of mixtures of natural refrigerants and lubricant oils has increased considerably in the recent years [1–8]. There remains, however, a lack of studies dealing specifically with mixtures involving R-600a. Marcelino Neto and Barbosa [9] measured the solubility, the liquid phase density and the viscosity of a system (i.e., several mixtures of the same components) of R-600a and a polyol ester lubricant oil (POE ISO 7) at temperatures ranging from 10 to 60 °C. The vapor-liquid equilibrium (VLE) data were correlated with the Heil–Prausnitz and Flory–Huggins activity models and with the Peng–Robinson [10] equation of state (EoS). The liquid density was correlated with the Peng–Robinson EoS and with a first-order Redlich–Kister expansion for the excess molar volume. The liquid dynamic viscosity was correlated with an excess-property approach based on the classical Eyring model. Satisfactory agreement was obtained between models and experimental data, with maximum root mean square (RMS) deviations of 1.1%, 0.2% and 3.0% in the predictions of VLE (bubble-point pressure), density and viscosity, respectively. Kumagai et al. [11] measured the viscosity of mixtures of R-600a and squalane between 0 and 60 °C at pressures up to 30 MPa and correlated the experimental data with a Tait-like equation and with a modified Kanti et al. [12] correlation with average absolute deviations (AAD) of 2.8% and 3.9%, respectively. Zhelezny et al. [13] presented experimental data for solubility, density and capillary constants for solutions of R-600a and a commercial mineral compressor oil over wide ranges of temperature (30–90 °C) and concentrations. The enthalpy of the liquid phase solution was calculated together with the excess thermodynamic functions. The paper also examined the nature of the experimental uncertainties in the investigation of thermodynamic properties of the refrigerant–oil mixtures. Zhelezny et al. [14] presented experimental data for the viscosity of mixtures of R-600a with two commercial mineral compressor oils at temperatures ranging from 13 to 75 °C (nominal)

Abbreviations: AAD, average absolute deviation; CFC, chlorofluorocarbon; EoS, equation of state; HFC, hydrofluorocarbon; LAB, linear alkylbezene; POE, polyol ester; PR, Peng–Robinson; RMS, root mean square; TV, volume translation; VLE, vapor–liquid equilibrium.

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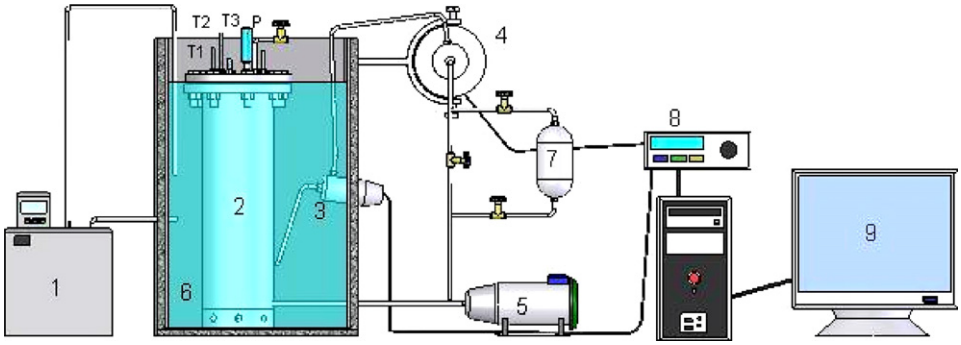

Fig. 1. Schematic diagram of the experimental apparatus.

and mass solubilities below 37%. The liquid mixture viscosity was measured using a rolling ball method. The viscosity data were successfully correlated via empirical relationships involving the molar solubility and the reduced temperature.

The present paper puts forward new data on mass solubility, liquid density and liquid dynamic viscosity of mixtures of R-600a and a linear alkylbenzene lubricant oil (LAB ISO 5), which is in fact a mixture of alkylbenzene compounds, at temperatures between 23 and 80 °C. The phase equilibrium and the density data were correlated using the Peng–Robinson [10] EoS. The liquid viscosity data were predicted with an Eyring-type model [15] in which the excess activation energy of viscous flow was modeled in terms of the component fugacity coefficients obtained from the Peng–Robinson EoS [16].

2. Materials and methods

2.1. Experimental apparatus and procedure

The experimental apparatus is shown in Fig. 1 [9,17]. A specified amount of the LAB 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 R-600a 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. Therefore, in the present experiments, the pressure of the R-600a/LAB mixtures is the dependent variable. The equilibrium cell is instrumented for absolute pressure, P (HBM P3MB transducer), 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 GC-M25 Micropump gear pump (5) moves the liquid 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 Danfoss Dl 1.5 Coriolis effect mass flow transducer (4) that records the mass flow rate, temperature and liquid density. In this gauge, the liquid mixture runs through U-shaped tubes that vibrate in a direction perpendicular to the flow. In the presence of fluid flow, inertia effects cause the vibrations of the two legs to be out of sync. The associated degree of phase-shift can be correlated to the mass flow through the tubes. The fluid density can be deduced from measurements of the natural frequency of the tube, which is proportional to the mass of fluid inside the tube. A Cambridge Applied Systems SPL 571 oscillating piston viscometer (3) registers temperature and dynamic viscosity of the liquid mixture. This type of gauge evaluates the viscosity of a given liquid based on the time taken for a calibrated piston to move, under the action of a magnetic force, between the two ends of a cylindrical cavity filled with the liquid. The mass solubility of R-600a in the mixture is measured gravimetrically with the aid of a KNWaagen (KN 4000) balance 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 National Instruments 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 losses to the environment. The temperature variation between the viscometer, the mass flow/density meter and test cell were within the uncertainty level associated with the thermocouples.

The experimental procedure for obtaining the mixture solubility has been described in detail in [17]. The temperature measurement uncertainty was estimated at ±0.2 °C (68% confidence level). The uncertainty of the density measurement (estimated from the manufacturer specifications) was ±1% of the absolute reading (95% confidence level). The uncertainty of the viscosity measurement (estimated from the manufacturer specifications) was 0.1 mPa s (±1% of the full scale, 68% confidence level). The uncertainty of the pressure transducer (estimated from the manufacturer specifications) was 3 kPa (68% confidence level) and that of the balance was ±0.03 g (determined by the present authors with a 95% confidence level). After an error propagation analysis, it was concluded that the uncertainty in the determination of the mass solubility was ±0.5 g kg^{-1} (68% confidence level). This value was subsequently confirmed through repeatability tests.

The mass solubility, liquid density and liquid dynamic viscosity measurements of the R-600a/LAB ISO 5 system were performed at temperatures between 23 and 80 °C (nominal). In total, 54 data points have been collected.

2.2. Materials

LAB ISO 5 (CAS 67774-74-7) is a mixture of alkylbenzenes with the general molecular structure given in Fig. 2. According to its manufacturer, the average molecular mass ranges from 238 to 245 g mol^{-1}. In our calculations, an effective average molecular mass of 239 g mol^{-1} has been used ($n+m=8.5$). The LAB mass density at 20 °C and the kinematic viscosity at 40 °C, derived from our own experimental database (see Section 3.4), are 868.4 kg m^{-3} and 4.2 mm^2 s^{-1}, respectively. The LAB was used as supplied by its

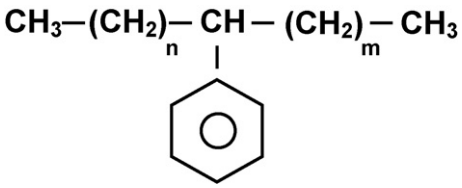

Fig. 2. Molecular structure of the alkylbenzenes that constitute the LAB ISO 5 lubricant oil ($n+m=7$–10) (n, $m=0$–10).