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## Absorption of Isobutane (R-600a) in Lubricant Oil

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### ABSTRACT

A detailed knowledge of refrigerant absorption in refrigerant-oil mixtures is important for understanding the behavior of hermetic compressors under transient conditions. In this work, we study the absorption of R-600a through the free surface of a stagnant layer of alkylbenzene lubricant oil in a closed system. Experiments were conducted in a transparent test section that enables visualization of the absorption process. The system pressure and the temperatures of the vapor, liquid and interface regions were recorded as a function time for ambient temperatures of 25, 40, 55 and 70°C. The data were used to validate a transient mass diffusion model that takes into account the departure from ideal solution behavior.

### 1. INTRODUCTION

Refrigerant absorption (or dissolution) in lubricant oil is important in many aspects of the design and operation of hermetic refrigeration compressors. To predict the rates of refrigerant absorption in lubricant oil, specific models are needed for phase equilibrium and physical properties such as liquid viscosity and mass diffusivity.

So far, the general approach to the investigation of gas dissolution in non-volatile liquids such as lubricant oil has been to determine the mass diffusivity and/or the interfacial mass transfer coefficient by means of either the history matching of calculated pressures or mass of solute with experimental data (Yokozeki, 2002; Tharanivasan *et al.*, 2004; Gessner and Barbosa, 2006), or by analytical approaches involving regression of pressure decay data for closed systems (Civan and Rasmussen, 2006; Sheikha *et al.*, 2006; Rasmussen and Civan, 2009). In these studies, non-ideal mixture effects are usually ignored, i.e., phase equilibrium is calculated according to Raoult's law, and the mass diffusivity is assumed independent of temperature and concentration.

In the specific case of R-600a, a number of papers have been published on the subject of phase equilibrium in recent years (Kumagai *et al.*, 2007; Zhelezny *et al.*, 2007; Marcelino Neto and Barbosa, 2008; Zhelezny *et al.*, 2009; Marcelino Neto and Barbosa, 2010). However, only a few studies deal with the actual time-dependent dissolution of R-600a in lubricant oil. Fukuta *et al.* (2005) discussed the transient mixing of R-600a and a mineral oil at constant pressure. In their one-dimensional diffusion model, the mass diffusivity was assumed independent of concentration and the solution density was assumed constant. Barbosa and Ortolan (2008) investigated experimentally the absorption of R-134a and R-600a through the top interface of an oil layer. The tests were conducted with three polyol ester (POE) oils of different viscosity grades, and the effect of the initial height of the oil layer on the absorption rate was assessed. For the absorption of R-134a, a macroscopic control volume model based on the solution of mass balance equations for the vapor and liquid phases was proposed. For R-600a, however, the data were correlated with an empirical correlation based on the Buckingham-Pi theorem.

In this paper, the absorption of R-600a through the free surface of a stagnant layer of lubricant oil in a closed system is investigated experimentally. The test section was specially constructed to allow visual observation of the dissolution process. In the mathematical model proposed for calculating the pressure decay as a function of time, the

departure from the ideal solution behavior has been incorporated into the calculation of the solution density, mass solubility and mass diffusivity by means of thermodynamic models based on equations of state (EoS) and activity coefficients for the R-600a/AB ISO 5 mixture (Marcelino Neto and Barbosa, 2010). Therefore, no data regression is performed and an actual comparison of the calculated instantaneous pressure with experimental data is presented.

## 2. EXPERIMENTAL WORK

The experimental apparatus is shown in Fig. 1(a). The test cell, shown in detail in Fig. 1(b), is a 153-mm high 74-mm ID glass beaker mounted between two stainless steel plates fastened with four sets of screws that press them against both ends of the beaker. A silicon gasket seals the contact between the plate and the top surface of the beaker. The cell is instrumented for absolute pressure,  $P$ , and the temperatures of the fluids are recorded by three Pt-100 RTDs located at three distinct heights to measure the temperature of the vapor,  $T_V$ , the interface,  $T_I$ , and the liquid,  $T_L$ . The measurement uncertainties of pressure and temperature are  $\pm 3\text{kPa}$  and  $\pm 0.15^\circ\text{C}$ , respectively.

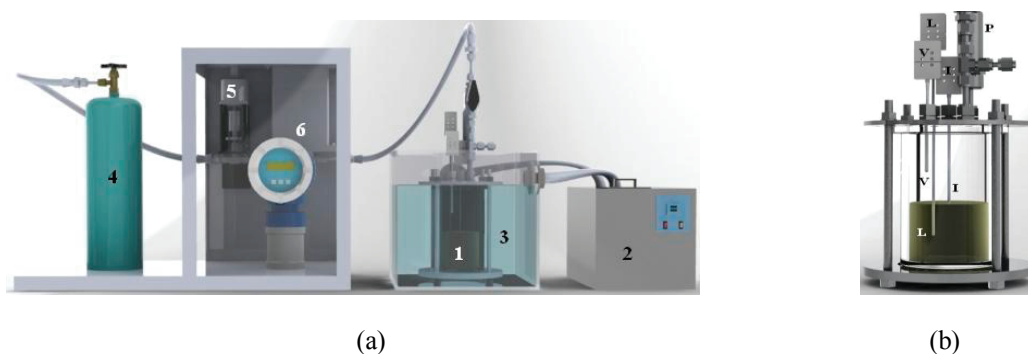


Figure 1: The experimental apparatus for absorption of refrigerant in lubricant oil.

The experimental procedure is as follows. Firstly, a specified amount of oil is placed in the cell (1). A vacuum of 0.04 mbar is generated in the apparatus to remove moisture and dissolved gases. The system temperature is set by a thermostatic bath (2) that circulates service water through a Perspex tank (3) in which the test cell is fully immersed. Time is allowed for the cell to reach thermal equilibrium with the water. Refrigerant vapor is kept at thermodynamic equilibrium with its liquid at room temperature inside a 2 L reservoir (4). An automatic needle valve (5) that connects the reservoir to the test cell is opened slowly for a specified amount of time (typically 8 s). This time allows the equalization of pressures in the reservoir and in the test cell. The mass of vapor that enters the test cell is measured by a Coriolis mass flow meter (6) with an error of  $\pm 0.15\%$  of the absolute reading. The valve is closed automatically and the pressure in the cell decreases with time as result of refrigerant absorption in the lubricant oil layer. The height of the liquid layer is measured with a milimetric scale on the outside of the glass beaker. The experimental apparatus is integrated with a signal conditioning and data acquisition module.

The absorption experiments were performed at four different temperatures (25, 40, 55 and  $70^\circ\text{C}$ ), for two different initial heights of the oil layer: 74 mm ( $1.0 D$ ) and 37 mm ( $0.5 D$ ), where  $D$  is the internal diameter of the test cell. R-600a was 99.5% pure according to its manufacturer/supplier. The chemical structure and properties of the lubricant oil have been reported elsewhere (Marcelino Neto and Barbosa, 2010).

## 3. MODELING

The difference between the refrigerant chemical potential in the vapor and liquid phases is the driving force for the vapor absorption in the liquid mixture. As a result, the refrigerant is gradually dissolved into the oil until the latter becomes completely saturated. A schematic diagram of the refrigerant-oil system is shown in Fig. 2, where the vapor-liquid interface is located at  $z = 0$ , and the bottom of the cell is at  $z = L$ .

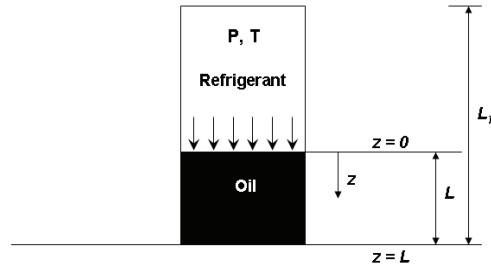


Figure 2: A schematic of refrigerant-oil system.

In the liquid layer, the time-dependent one-dimensional molecular diffusion is modeled according to Fick's law. Thus, the solute mass conservation equation is given by (Mills, 2001),

$$\rho \frac{\partial m_1}{\partial t} = \frac{\partial}{\partial z} \left( \rho D_{12} \frac{\partial m_1}{\partial z} \right) \quad (1)$$

where  $m_1$  is the refrigerant mass fraction,  $\rho$  is the solution mass density and  $D_{12}$  is the mass diffusivity. The mass diffusion process has been assumed isothermal and, based on visual observations of the absorption process, liquid swelling was neglected. As the oil is initially under vacuum, the initial condition is given by,

$$m_1(z, t)|_{t=0} = 0 \quad 0 \leq z \leq L \quad (2)$$

For the impermeable boundary at the bottom of the test cell, the mass flux at any time is equal to zero. Therefore,

$$\left. \frac{\partial m_1(z, t)}{\partial z} \right|_{z=L} = 0 \quad t > 0 \quad (3)$$

The interfacial mass transfer resistance is assumed negligible. Therefore, thermodynamic equilibrium holds at the vapor-liquid interface and the resulting boundary condition is given by,

$$m_1(z, t)|_{z=0} = m_1^{sat}(P, T) \quad t > 0 \quad (4)$$

where  $P$  is the instantaneous pressure. The refrigerant mass solubility and the solution mass density were calculated via the Peng and Robinson (1976) EoS with a single interaction parameter ( $k_{12} = -0.02668$ ) obtained from experimental data (Marcelino Neto and Barbosa, 2010).

The method of Vignes (1966) was used to take into account the composition effect on the mass diffusivity. Thus,

$$D_{12} = [(D_{12}^\circ)^{x_2} (D_{21}^\circ)^{x_1}] \alpha \quad (5)$$

where the binary diffusivities at infinite dilution were calculated via the Hayduk and Minhas (1982) correlation as follows,

$$D_{ij}^\circ = 1.55 \times 10^{-8} \frac{V_j^{0.27} T^{1.29} \sigma_j^{0.125}}{V_i^{0.42} \eta_i^{0.92} \sigma_i^{0.105}} \quad (6)$$

where  $T$  is the absolute temperature (K),  $V_i$  is the component molar volume at its normal boiling temperature ( $\text{cm}^3 \text{mol}^{-1}$ ),  $\eta_i$  is the component viscosity (cP) and  $\sigma_i$  is the surface tension ( $\text{dyn cm}^{-1}$ ). The oil viscosity and molar volume were obtained from experimental data (Marcelino Neto and Barbosa, 2010), and the normal boiling temperature of the oil was estimated with the group contribution method of Constantinou and Gani (1994). The surface tension of the oil was calculated with the Brock and Bird corresponding states method (Poling *et al.*, 2000). The physical properties of R-600a were obtained from REFPROP 8.0 (Lemmon *et al.*, 2007).

The thermodynamic correction factor,  $\alpha$ , was calculated with the NRTL activity coefficient model (Renon and Prausnitz, 1968),

$$\alpha = 1 + \left. \frac{\partial \ln \gamma_1}{\partial \ln x_1} \right|_{P,T} = 1 - 2x_1x_2 \left[ \frac{\tau_1 G_{21}^2}{(x_1 + x_2 G_{21})^3} + \frac{\tau_2 G_{12}^2}{(x_2 + x_1 G_{12})^3} \right] \quad (7)$$

where  $x_i$  is the component mole fraction and,

$$G_{ij} = \exp(-\psi\tau_j) \quad \tau_i = \frac{b_i}{RT} \quad (8)$$

where  $\psi = 0.3$ ,  $b_1 = 1061$  and  $b_2 = -2060$  were obtained from a regression of experimental data.

The instantaneous pressure is determined by a mass balance in the vapor phase. Assuming that the vapor pressure of the oil is negligible, the vapor behaves as an ideal gas and the system is isothermal, one has,

$$\frac{dP}{dt} = \frac{RT}{(L_T - L)} \left( \rho D_{12} \frac{\partial m_1}{\partial z} \right)_{z=0} \quad (9)$$

where  $R$  is the ideal gas constant of R-600a and  $L_T$  is the height of the cell.

The model equations were implemented in the Engineering Equation Solver – EES (Klein, 2007). Equations (1) to (4) were solved by the method of finite differences with first-order centered difference schemes for the spatial term and a first-order explicit scheme for the temporal term. The height of the liquid layer was divided into 20 equally spaced nodes and a constant time step of 200 s was used with a total integration time of  $8.0 \times 10^4$  s for each run. The initial condition of Eq. (9) is  $P(t=0) = P_0$ , where  $P_0$  was obtained from the experiments and corresponds to the maximum pressure in the test cell.

#### 4. RESULTS AND DISCUSSIONS

Figure 3 presents photographs taken at 0, 0.5, 2 and 24 hours into the absorption experiment for a temperature of 25°C and a height of the liquid layer of  $0.5D$ . The tank was filled with glycerin to minimize image distortion. A piece of striped paper was placed behind the test cell to allow the visual identification of the mass transfer penetration thickness, which can be clearly distinguished due to a change in the solution refractive index with the local refrigerant concentration. As can be seen in the pictures, the absorption process is diffusion-controlled due to the lower density of liquid R-600a in comparison to that of the oil (Fukuta *et al.*, 2005; Barbosa and Ortolan, 2008; Barbosa *et al.*, 2008). Furthermore, liquid swelling due to refrigerant absorption is negligible, as can be verified against the milimetric scale on the right-hand side of the pictures.

Figures 4(a) and (b) show the behavior of the system pressure as a function of time for the two heights of the liquid layer. Pressure and time were non-dimensionalized as follows:  $P^* = P/P_0$  and  $Fo = tD_{12}^0/L^2$ , where  $Fo$  is the mass transfer Fourier number. As can be seen, the pressure decreases as a result of vapor absorption in the oil, and the rate of decrease is inversely proportional to the temperature. This behavior can be explained by the fact that the mass solubility decreases with temperature, which reduces the mass transfer driving force. The maximum pressure and the mass of refrigerant that entered the test cell for the two sets of experiments are shown in Table 1. Less mass enters the test cell at higher temperatures because the vapor expands as exchanges heat with the liquid layer and the beaker wall. In both sets of tests, thermodynamic equilibrium was not attained before the end of each test (after approximately  $8 \times 10^4$  s), and the low absorption rates can be explained by the fact that the absorption process is diffusion-controlled. A good agreement between the model and the experimental data was observed, as can be seen from Figs. 5(a) and (b) which show the instantaneous error of the pressure prediction as a function of time. The average absolute errors for each run are also summarized in Table 1.

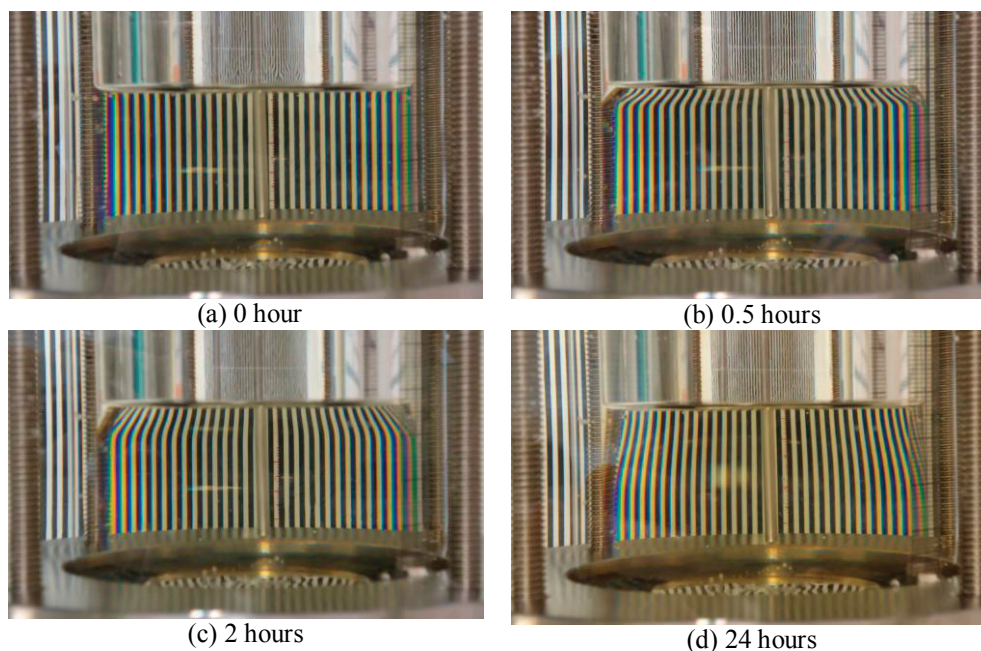


Figure 3: Photographs of the refrigerant absorption at 25°C for a 0.5D-high oil layer.

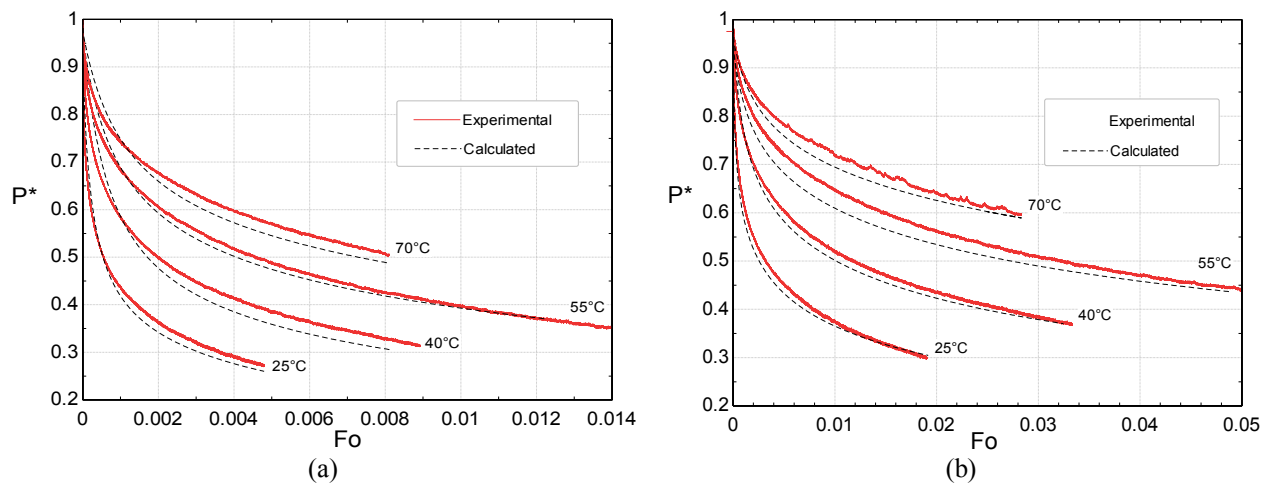


Figure 4: Pressure behavior in the R-600a/LAB ISO 5 system for a liquid height of (a) 1.0D and (b) 0.5D.

Table 1: Summary of experimental data.

Height of the oil layer	$T$ (°C)	Refrigerant Mass (g)	$P_0$ (kPa)	Average absolute error (%)
1.0D	25	2.73	271.5	4.86
	40	2.55	273.9	5.94
	55	2.40	261.6	3.65
	70	2.36	270.8	3.53
0.5D	25	3.57	245.1	2.47
	40	3.39	245.8	2.69
	55	3.25	252.0	4.03
	70	3.18	261.8	2.48

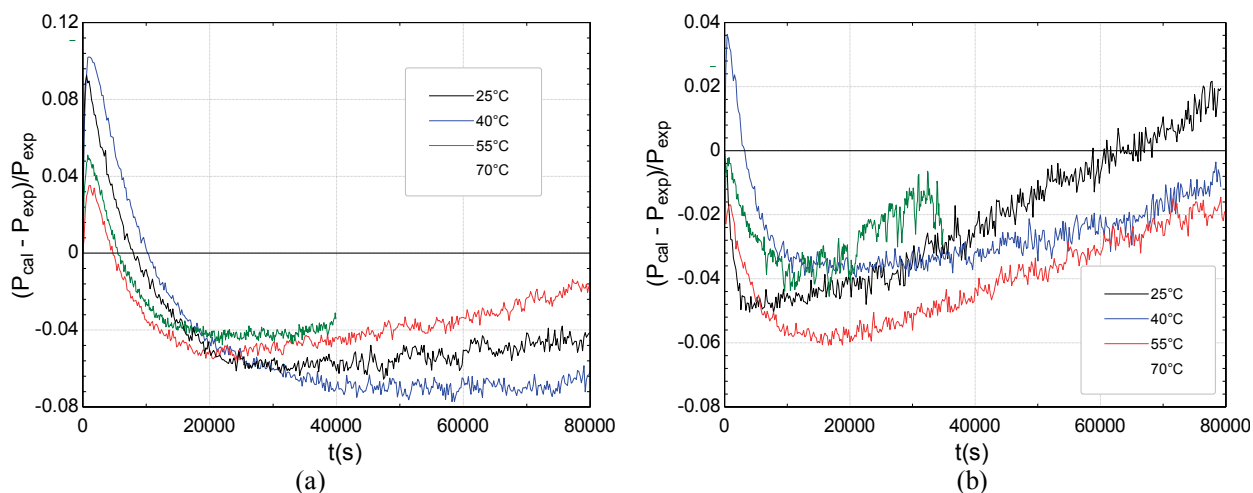


Figure 5: Instantaneous error of the pressure prediction for a liquid height of (a)  $1.0D$  and (b)  $0.5D$ .

Figures 6(a) and (b) show the temperature behavior during the first hour of the absorption tests at  $25^{\circ}\text{C}$  for the liquid heights of  $1.0D$  and  $0.5D$ , respectively. The temperatures of the interface and of the vapor exhibit the greatest increase due to the latent heat of absorption that is released at the interface. The liquid temperature does not change significantly because the sensor was positioned near the bottom of the test cell. After a short period of time, approximately 0.5 hour, the three temperatures return to the initial values.

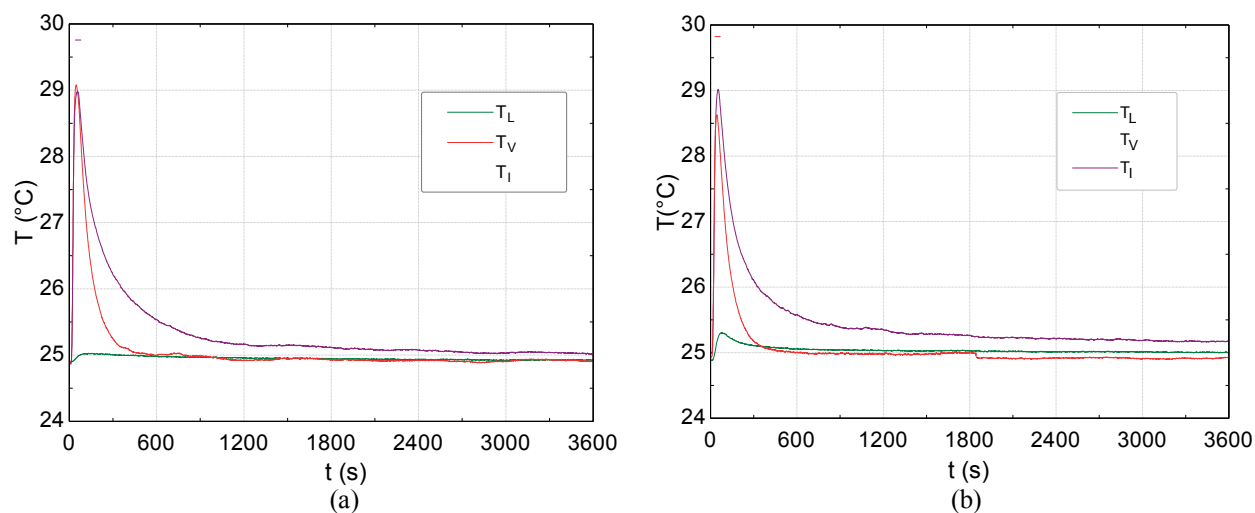


Figure 6: Temperature behavior in the R-600a/LAB ISO 5 system for a liquid height of (a)  $1.0D$  and (b)  $0.5D$ .

Figure 7 illustrates the dependence of the mass diffusivity on concentration for  $25^{\circ}\text{C}$ . A significant departure from the ideal behavior (i.e., a linear interpolation between the binary diffusivities at infinite dilution for both components) is observed. In the case presented, the binary diffusivities at infinite dilution vary considerably ( $D_{12}^{\circ} = 3.294 \times 10^{-10}$  and  $D_{21}^{\circ} = 4.270 \times 10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$ ). Taking into account this dependence of the diffusion coefficient on concentration contributed greatly to the prediction ability of the model. Figure 8 shows the dependence of the calculated interfacial solution density and the interfacial mass fraction (solubility) on the instantaneous pressure. Despite the fact that the deviation from Raoult's law is not significant for mixtures involving R-600a (Marcelino Neto and Barbosa, 2008), the significant variation of the interfacial solution density justifies the assumption that the density is not constant.

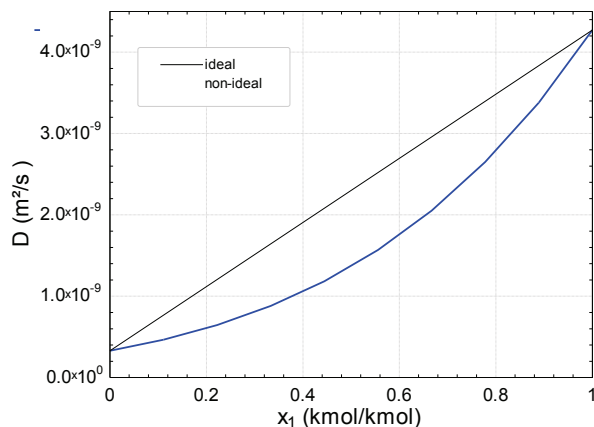


Figure 7: Mass diffusivity as a function of refrigerant mole fraction at 25°C.

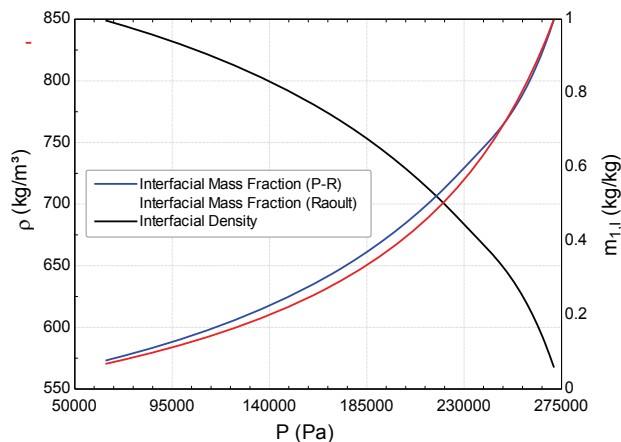


Figure 8: Density and interfacial mass fraction dependence on pressure at 25°C.

## 6. CONCLUSIONS

The absorption of R-600a through the free surface of a stagnant layer of lubricant oil was investigated in this paper. An experimental facility was constructed to enable visual observation of the process and measurements of system pressure, refrigerant mass and temperatures of the liquid, vapor and interface regions. A time-dependent mass diffusion model which took into account non-ideal effects was implemented and the results were compared with the experimental data. A good agreement was observed between the model and the experiments.

## NOMENCLATURE

$D_{12}$	diffusion coefficient	( $\text{m}^2 \text{s}^{-1}$ )
$L$	liquid height	(m)
$m$	mass fraction	( $\text{kg kg}^{-1}$ )
$P$	pressure	(Pa)
$t$	time	(s)
$T$	temperature	(K)
$R$	specific gas constant	( $\text{J kg}^{-1} \cdot \text{K}^{-1}$ )
$x$	molar fraction	( $\text{kmol kmol}^{-1}$ )
$z$	position	(m)
$\alpha$	thermodynamic factor	(-)
$\gamma$	activity coefficient	(-)
$\eta$	viscosity	(cP)
$\sigma$	surface tension	( $\text{dyn cm}^{-1}$ )

### Subscripts

I	interface
L	Liquid
V	vapor
1	refrigerant
2	oil
$i$	component $i$
$j$	component $j$

### Superscripts

$^{\circ}$	infinity dilution
$^{sat}$	saturation

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