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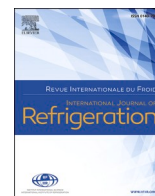
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Propylene and DME solubility in PAG oil: Experimental investigations and simplified modeling[☆]

Solubilité du propylène et du diméthyle éther dans l'huile polyalkylène glycol : études expérimentales et modélisation simplifiée

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ABSTRACT

This work experimentally investigated the solubility of Propylene and Dimethyl Ether (DME) in polyalkylene glycol (PAG) oil. The solubility experimental data were used to fit different local composition models from the literature, the Wilson and Heil equations, and the Non-Random Two-Liquid (NRTL) model. Two new empirical correlations were proposed to decrease the deviation between experiments and the models. The first correlation used six fitting coefficients and resulted in an average deviation of 1.2 % for Propylene and 2.8 % for DME. The second empirical correlation was proposed with a single solubility coefficient (SSC) and a compact form, which allows solubility to be determined explicitly. The correlation resulted in average deviations from experimental data of 1.6 % for Propylene and 2.5 % for DME. Moreover, the SSC correlation showed performance robustness against the decrease of available data points. The correlation was used to compare the solubility of Propylene and DME with the solubility of Propane. At the same conditions, Propane showed the lowest solubility while DME resulted in the most soluble refrigerant.

1. Introduction

The developments of the F-gas regulation and the regulation on the registration, evaluation, authorisation and restriction of chemicals (REACH) are accelerating the adoption of natural refrigerants such as Propane, Isobutane, and CO₂ in heat pump and refrigeration applications. Hydrocarbons are increasingly being adopted in refrigeration and heat pump applications, also thanks to their good thermophysical properties and low Global Warming Potential (GWP). Their main drawback is flammability, which introduces limits to the allowed refrigerant charge in indoor applications, according to [International](#)

[Standard IEC 60335-2-40 \(2022\)](#). Maintaining the charge amount below four times the lower flammability limit of the refrigerant allows to avoid installation restrictions and additional safety measures. These charge limits are 152 g for Propane, 184 g for Propylene, and 256 g for Dimethyl Ether (DME).

[Fernando et al. \(2004\)](#) investigated the refrigerant solubility of propane in different oils in the oil sump of a Propane compressor. Propane and hydrocarbons were found to be highly soluble in most lubricants, and careful oil selection and oil quantity minimisation are fundamental to reducing the amount of refrigerant in the oil. [Sánchez-Moreno-Giner et al. \(2023\)](#) presented the results of the performance and refrigerant distribution of a Propane low-charge

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Nomenclature		
<i>Symbols</i>		
p	Pressure (MPa)	(dimensionless or J/mol)
T	Temperature (K or °C)	α NRTL's non-randomness coefficient
ρ	Density (kg/m ³)	r Refrigerant
σ	Standard deviation	φ Fugacity coefficient
B	Second virial coefficient (m ³ /kg)	
M	Mass (kg)	<i>Acronyms and initialisms</i>
x	Molar concentration (mol/mol)	NRTL Non-Random Two-Liquid
X	Mass concentration (kg/kg)	COP Coefficient of Performance
l	Liquid	LFL Low Flammability Limit
v	Vapor	FS Full Scale
a	Coefficients for empirical correlations	GWP Global Warming Potential
a_1	Single solubility coefficient (1/K)	SCC Single Solubility coefficients
R	Molar gas constant (8.314472 J mol ⁻¹ K ⁻¹)	MAPE Mean Absolute Percentage Error
V	Volume (m ³)	RMSPE Root Mean Squared Percentage error
v	Molar volume (m ³ mol ⁻¹)	REACH Regulation on the registration, evaluation, authorisation, and restriction of chemicals
Λ	Interaction parameter	sat Saturation
τ	NRTL's binary adjustable interaction parameter	exp Experimental
		tot Total

brine-to-water heat pump. 41 % of the refrigerant was found in the compressor, mainly dissolved in the oil. One of the study's conclusions was that further charge reduction had to be focused on the charge stored in the oil, reducing solubility or oil quantity. Compressors designed with low oil amounts for use in low-charge Propane units were proposed by Gao et al. (2014) to reduce the refrigerant quantity absorbed by the lubricant. The adoption of polyalkylene glycol (PAG) oil allowed the reduction of solubility, as investigated by Ginies and Dewitte (2014). The solubility, viscosity and miscibility characteristics were compared with those of other lubricants. Polyolester (POE) oil has often been preferred to PAG thanks to its lower viscosity at low temperatures, lower film thickness and better performance, as investigated by Shi et al. (2022). Moreover, the same study found that under any condition, the oil sump solubility was uniform and was close to the oil sump inlet saturation solubility. PAG and POE lubricants are both synthetically produced. However, in general, the specific price of PAG oil is expected to be higher compared to POE oil. On the other hand, since PAG oil is usually adopted in low-charge systems, its quantity is often minimised. This may result in a reduction of the impact of the oil on the overall compressor cost. Nevertheless, PAG oil has been increasingly adopted by manufacturers of compressors for applications with low refrigerant charge.

The non-fluorinated refrigerants Dimethyl Ether (DME or R-E170) and Propylene (R-1270) were proposed by Granryd (2001) as long-term solutions among other hydrocarbons. DME is characterised by a high coefficient of performance (COP) and Propylene by a relatively high volumetric heating capacity. Another attractive characteristic of DME is the relatively high lower flammability level (LFL), when compared to Propane, allowing higher refrigerant charge limits. DME solubility in squalane was experimentally investigated by Sun et al. (2021). The author adopted the local composition Non-Random Two-Liquid model (NRTL) from Renon and Prausnitz (1968) and fitted it to experimental data. Wang et al. (Wang et al., 2021) adopted a similar experimental and modeling approach for Propane in mineral oils. Different thermodynamic local composition models were compared for multiple binary refrigerant-oil mixtures by Martz and Jacobi (1994) and Youbi-Idrissi (2003). Local composition models from Wilson and Wilson (1964), Heil and Prausnitz (1966) and Renon and Prausnitz (1968) were found to be able to reach a good level of accuracy.

As previously discussed, due to its lower solubility, PAG oil is particularly suitable for systems which aim to minimise refrigerant charge. This is an essential requirement of refrigeration equipment that

uses flammable refrigerants. PAG oil has also been adopted for CO₂ applications. Ikeda et al. (2004) evaluated the chemical and physical properties of different lubricants for CO₂. The study found that PAG was the most suitable oil for CO₂ applications. The reasons were suitable miscibility, higher chemical stability and improved lubricity under high pressure. While PAG was investigated and adopted for Propane and CO₂ applications, there is a lack of available experimental data on the interaction of PAG oils with other non-fluorinated refrigerants.

Despite the good accuracy, local composition models, commonly used for modeling refrigerant-oil solubility, also encounter some drawbacks. (i) They are semi-empirical models and rely on multiple coefficients. (ii) Their fitting requires refrigerant-lubricant solubility data that is time-consuming to obtain. (iii) The formulation of solubility is implicit. The coefficient fitting procedure, as well as the utilization of the fitted model, requires the adoption of iterative numerical methods. To incentivize the adoption of refrigerant-lubricant solubility models, the development of simplified and robust models with low implementation and execution time requirements may be needed.

This study is an extension of the work by Caramaschi et al. (2023). It investigated experimentally the solubility of the non-fluorinated refrigerants Propylene and DME in PAG oil. The newly obtained datasets were presented in tabular and graphical form. Local composition methods from Wilson and Wilson (1964), Renon and Prausnitz (1968) and Heil and Prausnitz (1966) were applied to fit the experimental data. Two new empirical correlations to characterize solubility as a function of pressure and temperature were proposed. A first correlation using six coefficients and a second simplified correlation using a single solubility coefficient (SSC) were developed. The SSC correlation was tested on other datasets available in the literature and its performance was assessed. At last, the solubility of Propylene and DME in PAG oil was evaluated and compared against the solubility of Propane in PAG oil, and the implications on the refrigerant charge in compressors of a heat pump were discussed.

2. Methods

The methods of this study are divided into six subsections. The investigated media are initially introduced in Section 2.1. Then, the experimental set-up is presented in Section 2.2 followed by the calculation of the solubility in Section 2.3, as part of the experimental procedure. The solubility data obtained were then fitted by different models, as described in the last three subsections. In Section 2.4, the

semi-empirical models from the literature are introduced. They are followed by the presentation of a new empirical correlation based on six fitting coefficients in Section 2.5. The method section concludes with the presentation of a new user-friendly correlation with a single fitting coefficient.

2.1. Media

The characteristics of the refrigerants investigated in this study are described in Table 1. Information about the PAG oil density follow in Table 2. The oil investigated was characterized by a kinematic viscosity at 40 °C of 64 mm²/s and 10 mm²/s at 100 °C.

2.2. Experimental setup

An experimental setup at the refrigeration laboratory (Lafset) of Conservatoire National des Arts et Métiers (CNAM) described also by Ginies and Dewitte (2014) and Notturmo et al. (2023) allowed to measure the solubility of refrigerant in PAG oil. The studied oil was a PAG66 oil and had similar characteristics to the PAG40 investigated by Shi et al. (2022). The test stand was composed of (i) a temperature-controlled oil bath; (ii) two sealed test cells equipped with a pressure transducer and a temperature sensor; and (iii) the sensors were connected to a computer for data collection. The volumes of the test cells were of 64.95 cm³ and 64.72 cm³. A simplified schematic of the test installation is in Fig. 1.

The experimental procedure, described in Fig. 2, started with the preparation of the cell, air removal (i), and weighing (ii). Oil density was measured using a densimeter. The results are presented in Table 2 (iii). A polynomial characterized the oil density as a function of temperature. After selecting the wanted refrigerant composition (iv), the refrigerant and oil quantities necessary for the tests were determined (v). Oil was then added to the cell (vi), and residual air was removed from the cell with a vacuum pump (vii). The cell containing oil was weighed to determine the exact quantity of oil introduced. Refrigerant was introduced in the cell and weighed (viii). At this point, the mixture in the cell was composed of three zones. One in vapour phase and two in the liquid phase. The cell was then manually shaken to reach only one zone in vapour phase and one zone in liquid phase. A first miscibility check was then made (ix). The desired bath temperature set-point was selected (x) and the solubility tests were carried out by immersing the test cell in the oil bath (xi).

The time required to reach steady state was at least two hours and it varied depending on the conditions and the refrigerant tested (xii). When a steady-state condition was achieved, the pressure and temperature of the cell were measured for 20 min or more (xiii). The measurements were taken every 5 s. At the end of the measurement, miscibility was visually checked (xiv). From the measurement of pressure and temperature, knowing the internal volume of the cell and the quantity of oil and refrigerant, it was possible to first calculate the quantity of vapour refrigerant, and then the solubility (xv) defined as the liquid refrigerant in the oil. It was assumed that given the low vapour pressure of oil, no oil is contained in the vapour phase. The refrigerant concentrations tested were between 8 % and 50 %, with temperatures varying from -10 °C to 90 °C for Propylene and to 100 °C for DME. The temperature and concentration ranges were selected with the goal to cover the typical operating conditions of compressors for heat pump applications. The test range was also determined by the safety

Table 2
Oil density measurements.

Density [g/cm ³]	Temperature [°C]
1.0018	20.0
0.9920	30.0
0.9824	40.0
0.9737	50.0
0.9659	60.0
0.9589	70.0
0.9528	80.0

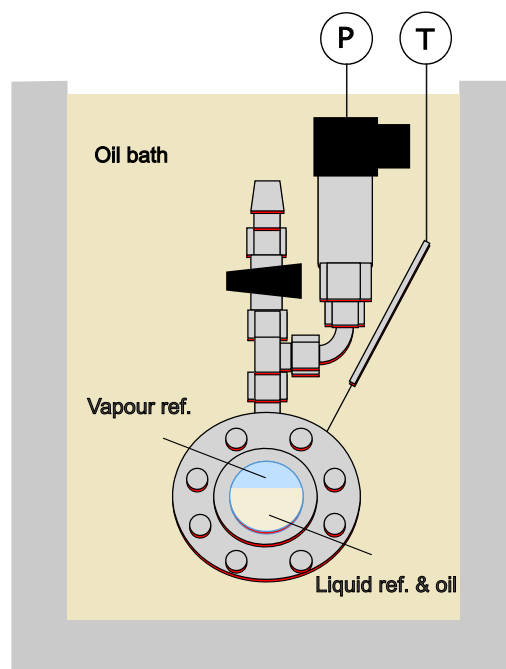


Fig. 1. Schematic of the experimental setup.

requirements and physical limitations of the test bench, which constrained the maximum cell pressure.

The methods of this work were implemented in the Python (Van Rossum and Drake, 2009) programming language using an object-oriented approach. The modelling work was divided into five parts: (i) calculation of solubility from the measured temperatures and pressures; (ii) uncertainty analysis using Monte Carlo simulations; (iii) local-composition models; (iv) empirical correlations; (v) the utilization of the solubility models for solubility prediction.

2.3. Calculation of the refrigerant solubility

The extended derivation of the method applied to determine the solubility was described by Youbi-Idrissi (2003). The main equation to calculate solubility $X_{r,l}$ was given:

$$X_{r,l} = \frac{M_{r,l}}{M_{oil} + M_{r,l}} = \frac{M_r - M_{r,v}}{M_{oil} + M_r - M_{r,v}} \quad (1)$$

Where $X_{r,l}$ is the mass concentration, r stands for refrigerant, l for

Table 1

Characteristics of the refrigerants considered in this study. The data are from ISO 817 (ISO 817, 2014), EN 378-1 (EN 378, 2016) and ASHRAE 34 (ASHRAE 34, 2022). GWP is the global warming potential, LFL is the lower flammability level, AIT is the autoignition temperature, NBP is the normal boiling point.

Name	R number	Chemical formula	Safety class	GWP	Molar mass [g/mol]	LFL [g/m ³]	AIT [°C]	NBP [°C]
Propylene	R-1270	C ₃ H ₆	A3	2	42.1	46	455	-48
DME	R-E170	C ₂ H ₆ O	A3	1	46.0	64	235	-25

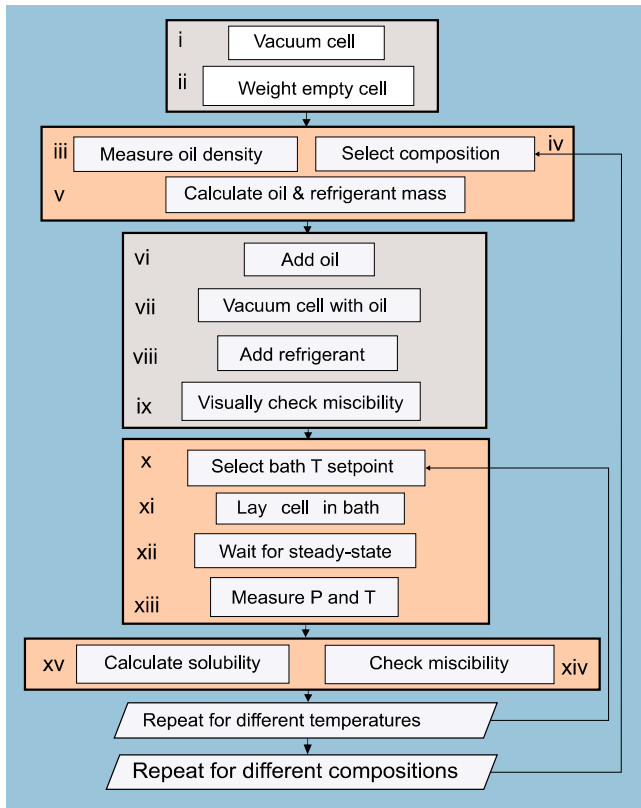


Fig. 2. Test procedure steps.

liquid, and v for vapour. Neglecting excess volume, the vapour volume $V_{r,v}$ was calculated in Eq. (2) as the difference between the total cell volume V_{tot} and the liquid volume, composed by refrigerant volume $V_{r,l}$ and the oil volume V_{oil} . The mass of refrigerant vapour $M_{r,v}$ is the only unknown and it can be calculated in Eq. (5) by combining Eqs. (2), (3), and (4):

$$V_{r,v} = V_{tot} - V_{r,l} - V_{oil} \quad (2)$$

$$M_{r,v} = V_{r,v} \rho_{r,v} = (V_{tot} - V_{r,l} - V_{oil}) \rho_{r,v} \quad (3)$$

$$V_{r,l} = \frac{M_{r,l}}{\rho_{r,l}(P)} = \frac{M_r - M_{r,v}}{\rho_{r,l}(P)} \quad (4)$$

$$M_{r,v} = \frac{\left(V_{tot} - \frac{M_{oil}}{\rho_{oil}(T)} - \frac{M_r}{\rho_{r,l}(P)} \right) \rho_{r,l}(P) \rho_{r,v}(P, T)}{\rho_{r,l}(P) - \rho_{r,v}(P, T)} \quad (5)$$

V_{tot} is the total volume of the cell and ρ is the density, calculated as a function of the cell temperature T and/or of the equilibrium pressure P . The main assumptions taken for the calculations were the following.

- The vapour was assumed to contain only refrigerant and no oil, due to the negligible oil vapor pressure.
- The refrigerant dissolved into the oil was assumed to be in liquid phase.
- The excess volume of the refrigerant-oil mixture was neglected, assuming ideal refrigerant-oil mixing.
- Full miscibility of the refrigerant-oil mixture.

An uncertainty analysis on the measurement of solubility was carried out using the standard uncertainties of Table 3 and propagating the uncertainties using Latin Hypercube sampling (Helton and Davis, 2003) and Monte Carlo simulations. The uncertainty distribution was assumed to be normal for all parameters.

Table 3

Uncertainties. FS stands for Full Scale and σ is the standard deviation.

Parameter measured	Uncertainty (2σ)
Mass of oil	0.03 g
Mass of refrigerant	0.03 g
Pressure	0.4 % FS
Temperature	0.25 K
Oil density	0.5 kg/m ³

2.4. Semi-empirical local composition models

Different solubility models in the literature were adopted to fit the experimental data. The activity coefficient of the refrigerant was calculated using the Wilson equation (Wilson and Wilson, 1964), the NRTL model from Renon and Prausnitz (1968) and Heil's equation (Heil and Prausnitz, 1966). Their parameters were fitted to the experimental data using an optimization procedure to minimize the difference between experimental data and measurements. For each component i of a binary mixture, the equilibrium between the two phases is characterized by the equality of fugacities of the vapour and liquid phases:

$$x_{i,v} \varphi_{i,v} p = x_{i,l} \varphi_{i,l} p \quad (6)$$

Where v and l are the vapour and liquid phases, x is the molar concentration, p is the equilibrium pressure and φ are the fugacity coefficients. Due to the assumption of negligible oil vapour pressure, $x_{oil,v} = 0$ and $x_{r,v} = 1$. It was possible to determine the activity coefficient of the refrigerant γ_r following Van Ness and C.H. (1982) as:

$$\gamma_r = \frac{p \varphi_r}{x_{r,l} p_{sat}} \quad (7)$$

The fugacity coefficient of the refrigerant φ_r , despite having a value very close to 1, was calculated by using Eq. (8) as adopted by Wang et al. (2021):

$$\varphi_r = \exp\left(\frac{(B_1 - V_l)(p_{sat} - p)}{RT}\right) \quad (8)$$

Where B_1 is the second virial coefficient in [m³/mol], V_l is the molar liquid volume in [m³/mol] from REFPROP 10 by Lemmon et al. (2018).

The activity coefficients were calculated using three different local composition models from the literature. The Wilson semi-theoretical equation (Wilson and Wilson, 1964) derived from Flory-Huggins's equation introduced an equal interaction energy parameter between two components. The Non-Random Two-Liquid model (NRTL) developed by Renon and Prausnitz (1968) model adopts an equation composed of three interaction parameters.

$$\text{Wilson} \ln \gamma_r = -\ln(x_r + \Lambda_{r,oil} x_{oil}) + x_{oil} \left(\frac{\Lambda_{r,oil}}{x_r + \Lambda_{r,oil} x_{oil}} - \frac{\Lambda_{oil,r}}{x_{oil} + \Lambda_{oil,r} x_r} \right) \quad (9)$$

$$\text{NRTL} \ln \gamma_r = x_{oil}^2 \left(\tau_{r,oil} \left(\frac{\Lambda_{oil,r}}{x_r + x_{oil} \Lambda_{oil,r}} \right)^2 + \frac{\tau_{r,oil} \Lambda_{r,oil}}{x_{oil} + x_r \Lambda_{oil,r}} \right) \quad (10)$$

Where $\Lambda_{r,oil}$ and $\Lambda_{oil,r}$ are interaction parameters related to the local composition of refrigerant around oil and vice versa. The NRTL model adopts two binary adjustable interaction parameters τ and a third fitting coefficient α , which can be set to a fixed value. This study also adopted the Heil equation (Heil and Prausnitz, 1966) which is a combination of the Wilson equation and the NRTL model.

For the Wilson and Heil models:

$$\Lambda_{r,oil} = \frac{V_r}{V_{oil}} \exp(-\tau_{r,oil}) \quad (11)$$

$$\Lambda_{oil,r} = \frac{V_{oil}}{V_r} \exp(-\tau_{oil,r}) \quad (12)$$

For the NRTL model:

$$\Lambda_{r,oil} = \exp(-\alpha\tau_{r,oil}) \quad (13)$$

$$\Lambda_{oil,r} = \exp(-\alpha\tau_{oil,r}) \quad (14)$$

For the NRTL and Heil models, the interaction parameters were modelled using two different approaches. In the first case, a conventional formulation was found in [Renon and Prausnitz \(1968\)](#).

$$\tau_{r,oil} = \frac{\tau_{r,oil}(1) - \tau_{r,oil}(2)}{RT} \quad (15)$$

$$\tau_{oil,r} = \frac{\tau_{oil,r}(1) - \tau_{oil,r}(2)}{RT} \quad (16)$$

Where $\tau_{r,oil}(1)$, $\tau_{r,oil}(2)$, $\tau_{oil,r}(1)$ and $\tau_{oil,r}(2)$, typically defined as energy parameters, were set as four fitting coefficients in J/mol. α is a fifth coefficient, the non-randomness parameter, available only for the NRTL.

In a second approach, aimed to reduce the deviation between the models and the experimental data, the binary interaction parameters were formulated as function of temperature, similarly to part of the Antoine equation ([Antoine, 1888](#)) as suggested by [Leon Poesy \(1996\)](#) and adopted by [Wang et al. \(2021\)](#).

$$\tau_{r,oil} = \tau_{r,oil}(1) + \frac{\tau_{r,oil}(2)}{T} \quad (17)$$

$$\tau_{oil,r} = \tau_{oil,r}(1) - \frac{\tau_{oil,r}(2)}{T} \quad (18)$$

In the result section, the NRTL and Heil models adopting this alternative formulation of the interaction parameters were called NRTL 2 and Heil 2, respectively.

2.5. Empirical correlation with 6 coefficients

This work proposed two empirical correlations. The first proposal came from the commonly used correlation from [Mermond et al. \(1999\)](#) adopted by [Wu et al. \(2018\)](#), written below as a reference:

$$\log_{10}\left(\frac{P}{10^5}\right) = a_1 + \frac{a_2}{T} + \frac{a_3}{T^2} + \log_{10}(X_{r,l})\left(a_4 + \frac{a_5}{T} + \frac{a_6}{T^2}\right) + \log_{10}^2(X_{r,l})\left(a_7 + \frac{a_8}{T} + \frac{a_9}{T^2}\right) \quad (19)$$

Some modifications were implemented to the existing correlation:

- (i) The first three coefficients were removed and substituted by the calculated saturation pressures at solubility $X_{r,l} = 1$.
- (ii) The first-degree term in [Eq. \(11\)](#) was substituted with a term of the third degree.

This allowed us to obtain an equation with a derivative equal to 0 at $X_{r,l} = 1$, hence assuring a monotone curve with a horizontal tangent. The equation had six, instead of nine, parameters a_i and it determined pressure as a function of solubility, temperature, and the calculated saturation pressure. The proposed correlation resulted in:

$$\beta = \ln^2(X_{r,l})\left(a_1 + \frac{a_2}{T + a_3}\right) + \ln^3(X_{r,l})\left(a_4 + \frac{a_5}{T + a_6}\right) \quad (20)$$

$$p = p_{sat}(T)e^{\beta} \quad (21)$$

In the initial study from [Caramaschi et al. \(2023\)](#) for the case of DME, the term $-1e^{-4/X_{r,l}^2}$ was added to the polynomial β to improve the behaviour at low refrigerant concentrations. In the current study instead, the additional term was removed. The formulation of the correlation changed from $a_i + \frac{a_{i+1}}{T} + \frac{a_{i+2}}{T^2}$ to a form that resembles Antoine's equation $a_i + \frac{a_{i+1}}{T + a_{i+2}}$. The final form of the first proposed correlation can

be rewritten as:

$$\ln\left(\frac{P}{p_{sat}(T)}\right) = \ln^2(X_{r,l})\left(a_1 + \frac{a_2}{T + a_3}\right) + \ln^3(X_{r,l})\left(a_4 + \frac{a_5}{T + a_6}\right) \quad (22)$$

2.6. Empirical correlation with a single solubility coefficient (SSC)

Empirical correlations with multiple coefficients, as the one developed in the previous section, require a high amount of experimental data in order to be accurate. To simplify the understanding and the application of refrigerant-oil solubility models, and reduce the need for experimental data, a new user-friendly correlation was proposed.

The correlation characterized the equilibrium pressure as function of temperature and solubility and a single coefficient.

$$p = p_{sat}(T) - \frac{p_{sat}(T)}{\exp\left(\frac{X_{r,l}}{a_1 T}\right)} \quad (23)$$

Where p is the pressure of the refrigerant-oil mixture in MPa, p_{sat} is the saturation pressure of the pure refrigerant, T is the equilibrium temperature in Kelvin, $X_{r,l}$ is the solubility and a_1 is a single fitting parameter. The simple form of the equation allows to determine solubility $X_{r,l}$ explicitly as:

$$X_{r,l} = a_1 T \ln\left(\frac{p_{sat}(T)}{p_{sat}(T) - p}\right) \quad (24)$$

At raising pressure p , solubility increases. At increasing difference between saturation pressure and equilibrium pressure $p_{sat}(T) - p$ solubility instead drops. The effect of temperature on solubility depends also on how pressure is affected, and it is not trivial. On the other hand, the coefficient a_1 was found proportional to solubility. In this work a_1 was named single solubility coefficient (SSC).

All models were fitted to the experimental data through an optimization procedure which applied the global optimization algorithm Basin-Hopping first described by [Wales and Doye \(1997\)](#) and implemented in the Scipy library by [Millman et al. \(Virtanen et al., 2020\)](#). The objective function to minimize was the mean absolute percentage error (MAPE) between modelled and measured pressures:

$$\text{MAPE} = \frac{1}{n} \sum_{i=1}^n \left| \frac{P_{exp,i} - P_{model,i}}{P_{exp,i}} \right| \cdot 100 \quad (25)$$

Where P_{exp} are the measured pressures, P_{model} the predicted pressures and n is the number of data points.

The deviations between experimental data and modelled data were compared for different models. Moreover, for the single-coefficient correlation, it was evaluated how the number of available data points can affect the accuracy of the correlation. Finally, the correlation's accuracy was tested on datasets in the literature.

3. Results

3.1. Miscibility

After the refrigerant filling process in the cell containing oil, it was possible to observe three phases: a vapour phase containing refrigerant and a negligible amount of oil, a liquid phase rich in liquid refrigerant, and another liquid phase rich in oil. An example can be seen in [Fig. 3](#). After shaking the cell and mixing the two components, full miscibility could be obtained. The miscibility of the refrigerant-oil mixture was visually confirmed for all the test points, for both Propylene and DME. For the tested compositions, no color change, and no phase separation were observed. An example can be observed in [Fig. 4](#).



Fig. 3. Refrigerant not fully miscible in oil, immediately after refrigerant filling.



Fig. 4. Example of Propylene miscible in PAG oil.

3.2. Solubility

The propagated uncertainty on the measurement of solubility resulted in between 0.2 % and 1.6 % of the calculated value. Pressure had the highest sensitivity on refrigerant solubility in oil. The experimental data for Propylene and DME are presented in Tables 4. and 5. respectively.

All the studied models were fitted to the experimental data through an optimization procedure. Table 6 summarizes the resulting coefficients.

The fitted correlations were used to predict the Pressure-Temperature-Solubility curves, also outside of the experimental range. Results were presented in Figs. 5–8. The curves are monotone and as it can be expected, the lower the refrigerant concentration, the lower the measured equilibrium pressure.

As depicted in Figs. 5 and 6, for Propylene the empirical correlations show a generally satisfactory match with the experimental results. The mean absolute percentage error (MAPE) for the correlation with 6 coefficients and the one with a single coefficient is found to be 1.2 % and 1.6 % respectively. At conditions of pure refrigerant, with a mass fraction of 1, the pressures were calculated using REFPROP through

Table 4
Experimental data Propylene.

Temperature [K]	Pressure [MPa]	Refrigerant solubility [-]
263.78	0.238	0.095 ± 0.001
263.88	0.397	0.295 ± 0.001
264.01	0.342	0.196 ± 0.001
264.10	0.409	0.396 ± 0.001
283.19	0.401	0.093 ± 0.001
283.26	0.596	0.193 ± 0.001
283.57	0.693	0.291 ± 0.001
283.59	0.743	0.392 ± 0.001
303.33	0.978	0.189 ± 0.001
303.36	0.638	0.092 ± 0.001
303.43	1.244	0.386 ± 0.001
303.48	1.157	0.286 ± 0.001
313.02	1.200	0.187 ± 0.001
313.12	1.550	0.383 ± 0.001
313.13	0.767	0.091 ± 0.001
313.21	1.424	0.283 ± 0.001
323.07	1.926	0.378 ± 0.002
323.07	1.475	0.184 ± 0.001
323.19	1.752	0.280 ± 0.001
323.2	0.930	0.090 ± 0.001
333.03	1.775	0.181 ± 0.001
333.03	2.362	0.373 ± 0.002
333.18	2.120	0.276 ± 0.002
333.22	1.098	0.089 ± 0.001
342.98	2.871	0.366 ± 0.003
343.00	2.097	0.178 ± 0.002
343.15	2.540	0.271 ± 0.002
343.21	1.276	0.088 ± 0.001
352.92	2.444	0.175 ± 0.002
353.20	1.464	0.087 ± 0.001
362.88	2.808	0.172 ± 0.002
363.19	1.661	0.086 ± 0.001

Table 5
Experimental data DME.

Temperature [K]	Pressure [MPa]	Refrigerant solubility [-]
263.57	0.072	0.097 ± 0.001
263.60	0.169	0.498 ± 0.003
263.73	0.143	0.299 ± 0.001
283.68	0.345	0.496 ± 0.001
283.69	0.143	0.096 ± 0.001
283.72	0.292	0.298 ± 0.001
293.33	0.394	0.297 ± 0.001
293.35	0.458	0.495 ± 0.001
293.55	0.217	0.095 ± 0.001
303.28	0.517	0.296 ± 0.001
303.36	0.613	0.494 ± 0.001
303.55	0.255	0.095 ± 0.001
313.33	0.673	0.295 ± 0.001
313.40	0.799	0.493 ± 0.001
313.47	0.331	0.094 ± 0.001
323.11	0.853	0.294 ± 0.001
323.11	0.411	0.094 ± 0.001
323.17	1.017	0.492 ± 0.001
333.07	1.069	0.293 ± 0.001
333.08	0.510	0.093 ± 0.001
333.14	1.278	0.490 ± 0.001
343.02	1.320	0.291 ± 0.001
343.03	0.617	0.092 ± 0.001
343.12	1.583	0.489 ± 0.001
352.98	1.609	0.289 ± 0.001
352.99	0.742	0.091 ± 0.001
353.02	1.945	0.486 ± 0.001
362.92	0.877	0.091 ± 0.001
362.92	1.938	0.288 ± 0.001
362.99	2.362	0.484 ± 0.001
372.82	1.022	0.090 ± 0.001
372.88	2.302	0.286 ± 0.001
372.94	2.838	0.482 ± 0.002

Table 6
Fitting coefficients for the local composition models and empirical correlation.

Local composition semi-empirical models – 4 and 5 coefficients						
Refrigerant	Model	α	$\tau_{r,oil}(1)$	$\tau_{r,oil}(2)$	$\tau_{oil,r}(1)$	$\tau_{oil,r}(2)$
Propylene	Wilson	–	-36,581	24,607	7979	5099
	NRTL	0.010	-1,000,000	1,000,001	-5532	6442
	NRTL 2	0.807	13.2	-3045	-2.55	304
	Heil	–	-9721.9	1881	1518	196
	Heil 2	–	-10.7	4278	4.81	-4471
DME	Wilson	–	-3710	-1055	-10,930	-11,730
	NRTL	0.750	-1706	-3116	-779	4028
	NRTL 2	1.276	1705	-10,300	-2.2	287
	Heil	–	-2715	-1110	181.6	-229
	Heil 2	–	0.988	151	-16.5	-241
Empirical correlation six coefficients						
	a_1	a_2	a_3	a_4	a_5	a_6
Propylene	-0.0462	216	-16,770	-0.219	-286	-1461
DME	-0.204	460	-19,355	-0.022	24	9559
Empirical correlation single solubility coefficient						
			SSC a_1			
Propylene			$4.588 \cdot 10^{-4}$			
DME			$6.530 \cdot 10^{-4}$			

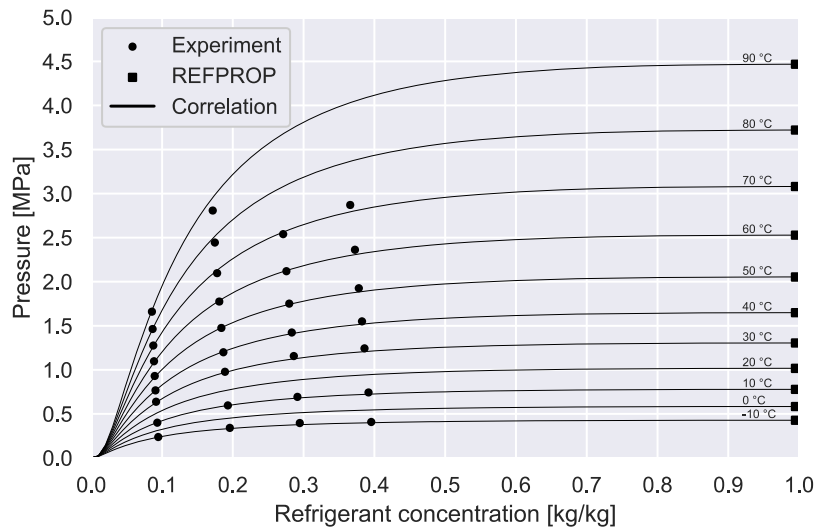


Fig. 5. Propylene data and correlation 6 coefficients. Average deviation 1.2 %.

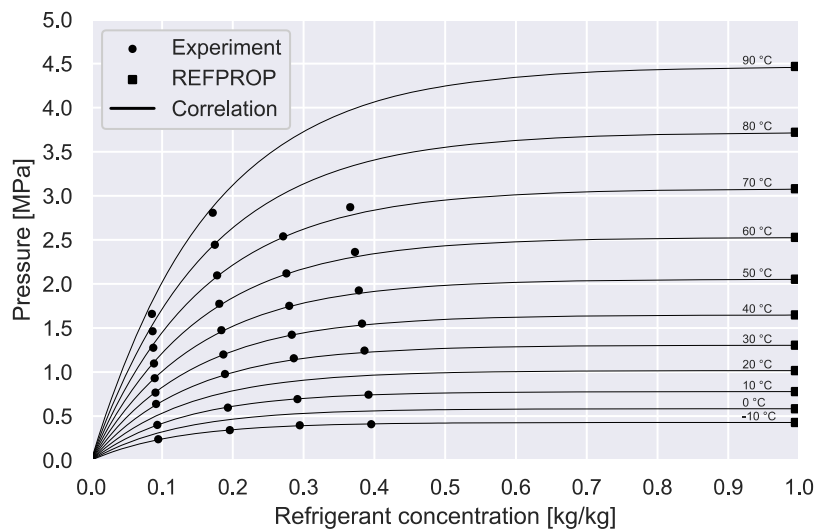


Fig. 6. Propylene data and correlation single coefficient. Average deviation 1.6 %.

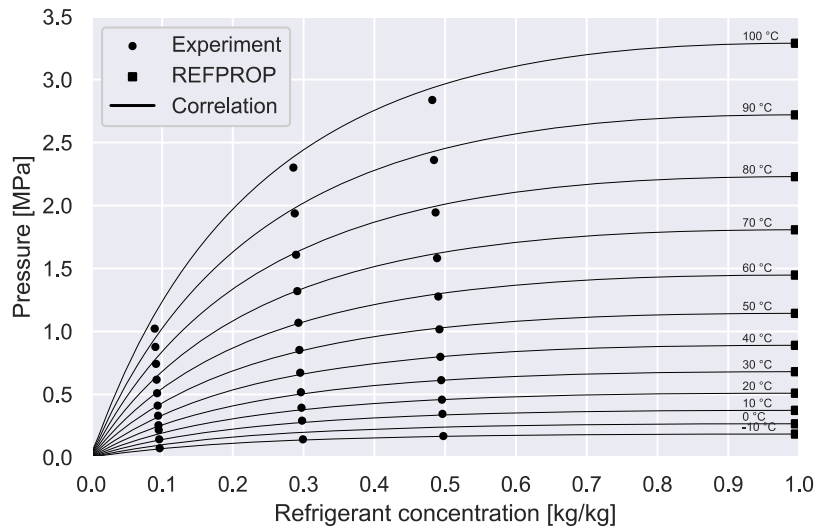


Fig. 7. DME data and correlation 6 coefficients. Average deviation 2.8 %.

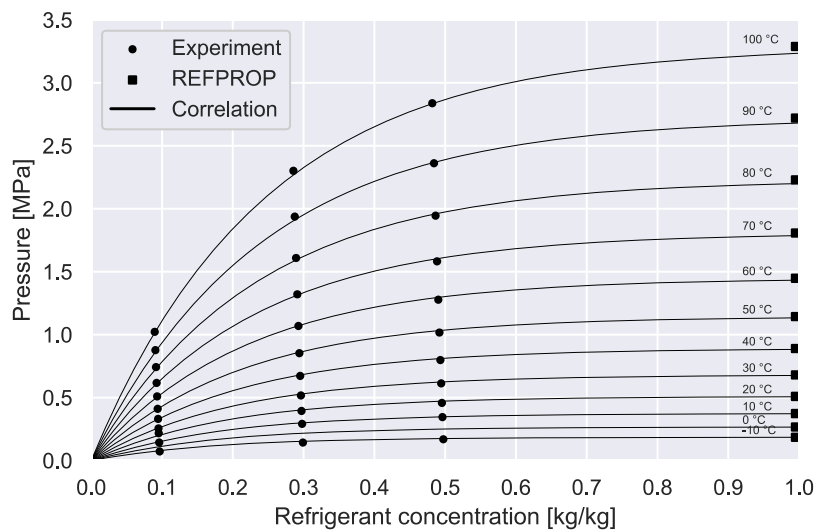


Fig. 8. DME data and correlation 1 coefficient. Average deviation 2.5 %.

CoolProp (Bell et al., 2014). These data points were excluded from the calculation of the average deviation. However, it can be stated that for Propylene, both correlations successfully predict the pressure behavior

of the pure refrigerant conditions, at all temperatures. Moreover, pressure tends to zero when no refrigerant is in the system.

The two empirical correlations matched DME data with an average

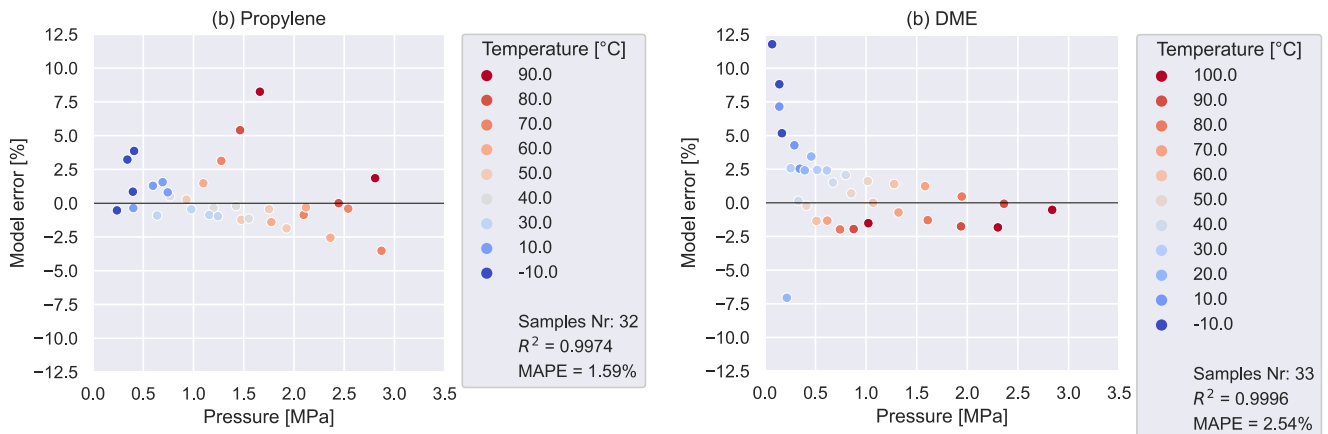


Fig. 9. Deviation for each datapoint using the single-coefficient correlation vs measured pressures, at different temperatures, for Propylene (a) and DME (b).

deviation of 2.8 % when 6 coefficients were adopted, and 2.5 % with the SSC correlation. For DME, in Fig. 7, the 6-coefficient correlation predicted well the points with pure refrigerant and tends to overestimate pressures at high temperatures. The SSC correlation, in Fig. 8 introduces a slight deviation at a refrigerant concentration of unity and high temperatures while reducing the deviation at lower concentrations.

In Fig. 9, the errors of the correlation with a single coefficient were plotted versus pressures and temperatures. The two refrigerants present different trends. For Propylene, in Fig. 9a, the maximum deviations occur between 1 MPa and 2 MPa, and the highest error of 8.2 % was found at the temperature of 90 °C. For DME, in Fig. 9b, the maximum deviations occur at low pressures and low temperatures, with the largest value being 11.7 %.

Furthermore, additional error analysis was performed to evaluate the effect of the number of data points used for fitting the model on the error. The data points selected for training were kept in consecutive order and tested $n_{data,tot} - n_{data,training}$ times. As shown in Fig. 10a, it was found that for Propylene, reducing the number of data points used during model fitting to only two data points increased the percentage error, on average, from 1.6 % to 2 %. Moreover, in light blue, it is possible to observe that depending on the chosen data points, the average error can vary between 1.6 and 4.5 %. The maximum error, on average, remained stable and slightly decreased from 8.2 % to 8 %. Using three data points, the possible error variation increased, ranging from about 4.8 % to 19 %. For DME, the average error increase was more pronounced when the number of data points decreased to two (Fig. 10b). With two data points, the average error increased from 2.5 % to 3.8 %, and the maximum error moved, on average, from 12 % to 13.9 %. On the other hand, using two and three data points increased the uncertainty significantly, with the average error ranging between 2.5 % and 9 % and the maximum error varying between 9 and 21 %. Overall, while the mean values of the average and maximum errors only slightly increased with the reduction of data points, their uncertainty was raised significantly for all the cases, especially when the number of data points used for training was reduced below four.

3.3. Model comparison

The accuracy of the seven models considered in this study were compared for both Propylene and DME. The results are reported in Fig. 11. For both refrigerants the local composition models (Wilson, NRTL and Heil) adopting conventional formulation of the binary interaction parameters resulted in the highest deviations, in the range between 4.2 % and 9 %. The alternative form of the binary interaction parameters, $\tau_{ij} = \tau_{ij}(1) + \frac{\tau_{ij}(2)}{T}$ resulted in a significant improvement of the performance. NRTL 2 lowered the error to 1.1 % for Propylene and Heil 2 reached an error of 1.3 % for DME. The empirical correlations

outperformed the local composition models, while tend to underperform compared to one model between NRTL 2 and Heil. The empirical correlations ranked among the top three models for Propylene and among the top four options for DME.

3.4. Test of the SSC correlation with other datasets in the literature

The correlation with a single fitting coefficient was tested on other datasets, and its performance was evaluated. As can be observed from Table 7, the mean average percentage error (MAPE) was between 1.6 and 6.2 %, the model performed better than the existing models for 3 datasets of the 7 tested. While the correlation introduces a lower performance for the majority of the models it was compared with, a performance improvement of 1.7 % points was obtained on the dataset from Stöckel et al. (2023), which however is only composed of 8 data points. The worst case was instead for DME-Squalane where MAPE increased by 3.9 % compared to the NRTL model applied by Sun et al. (2021), similar to the NRTL 2 model applied in this work. Here the minimum value for R^2 was reached at 0.9928.

At last, the solubilities of different refrigerants in PAG oil were calculated with the proposed SSC correlation. In Fig. 12, data on the solubility of different refrigerants were compared, at the saturation temperature of 0 °C and 60 °C. Compared to the data presented by Caramaschi et al. (2023) which applied an empirical correlation with 6 coefficients, in this study the solubility curves were obtained by applying the correlation with a single coefficient. As expected, solubility increases at higher saturation temperatures (pressures) and at decreasing superheat temperature differences. DME resulted in having the highest solubility values, especially at higher saturation temperatures, with a solubility of 33 % at a superheat value of 10 K and a saturation temperature of 0 °C. At the same conditions, Propylene showed a lower solubility, with a value of about 26 %. Propane, used as a reference, showed the lowest solubility of 19 %. At the saturation temperature of 60 °C and 20 K of superheating, DME had a solubility of 35 %, followed by Propylene at 27 % and Propane at 19 %.

4. Discussions

Despite miscibility being visually checked in all the test points, it could not be guaranteed in the entire composition range. The study had a limited solubility range, from about 9 % to a maximum of 50 %, in the case of DME, and to a maximum of about 40 % for Propylene. Immiscibility at higher refrigerant concentrations could not be excluded, especially when considering that the PAG oil studied by Shi et al. (2022), with similarities to the one in the current study, was found to be partially miscible with Propane. The choice of the testing range was made to cover solubility levels typically found in compressors of residential heat

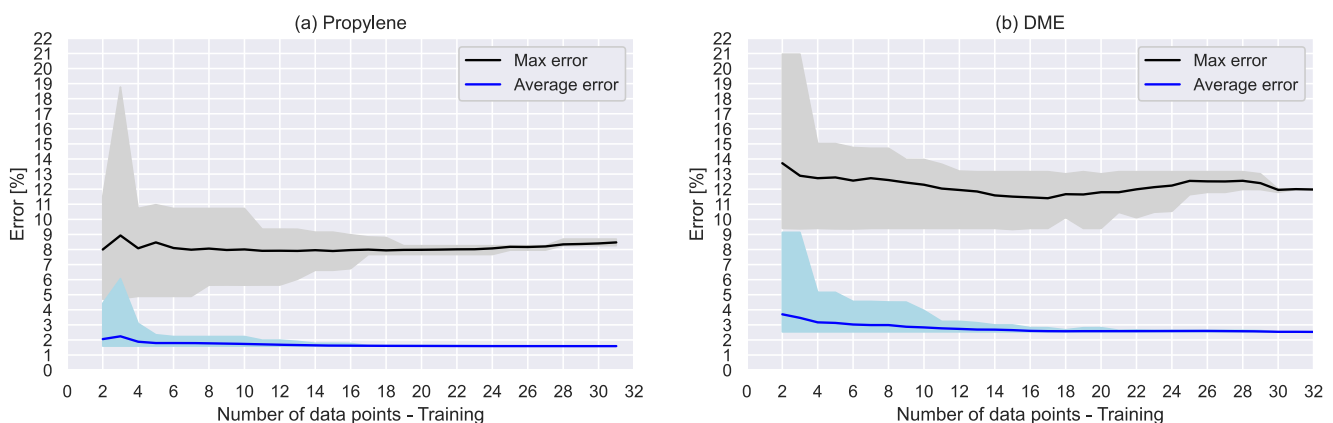


Fig. 10. Average and maximum model deviations as a function of the number of data points used for fitting the single solubility coefficient (SSC) a_1 . The two lines represent the mean values. The areas in grey and light blue show the ranges of the maximum and average errors, respectively.

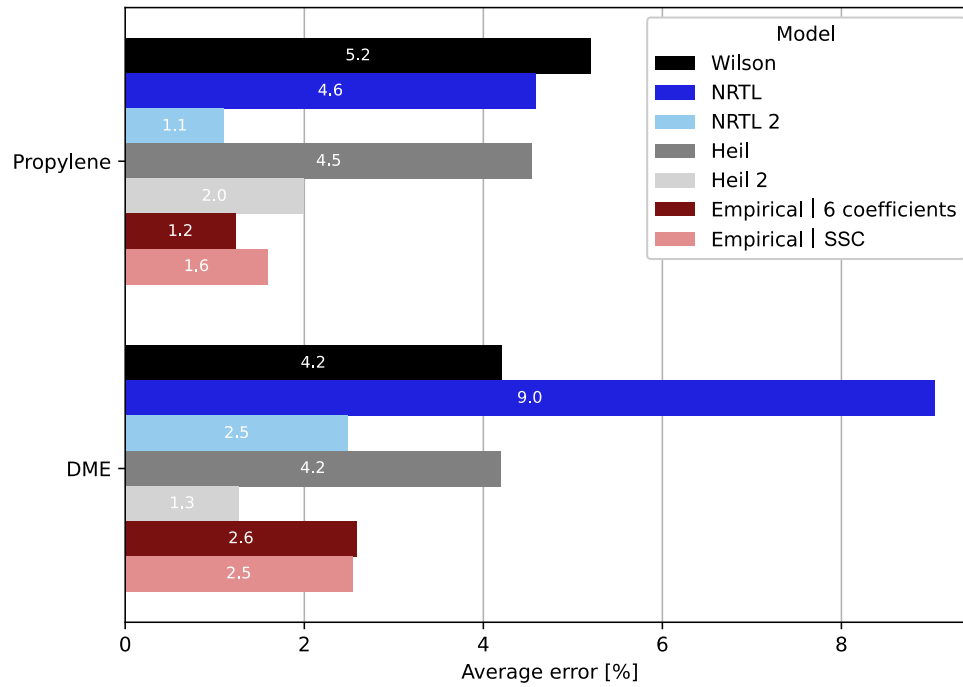


Fig. 11. Average deviation comparison between different models and correlations for refrigerant-oil solubility.

Table 7

Evaluating the performance model with one fitting parameter using other datasets. Solubility is considered Low for SSC $a_1 < 4 \cdot 10^{-4}$, high for SSC $a_1 > 6 \cdot 10^{-4}$, and Medium in between.

Oil	Ref	Source	Data points	SSC correlation						Model in literature		Δ MAPE [%]
				Solubility	SSC a_1 [1/K]	R^2	Max E [%]	RMSPE [%]	MAPE [%]	Model name	MAPE [%]	
PAG	Propylene	This work	32	Medium	$4.59 \cdot 10^{-4}$	0.9974	8.2	2.8	1.6	NRTL	1.1	+0.5 %
PAG	DME	Stöckel et al. (2023)	33	High	$6.53 \cdot 10^{-4}$	0.9996	11.7	1.7	2.5	Heil	1.3	+1.2 %
POE	DME		8	High	$6.378 \cdot 10^{-4}$	0.9980	6.7	3.1	2.4	Empirical	4.1	-1.7 %
Squalane	DME	Sun et al. (2021)	56	Medium	$5.613 \cdot 10^{-4}$	0.9928	20.8	6.6	6.2	NRTL	2.3	+3.9 %
Squalane	Isobutane	Sun et al. (2021)	49	High	$7.697 \cdot 10^{-4}$	0.9994	5.4	1.8	1.7	NRTL	0.8	+0.9 %
MO 3GS	Propane	Wang et al. (2021)	49	Medium	$5.461 \cdot 10^{-4}$	0.9980	9.8	4.5	3.0	NRTL	1.9	+1.1 %

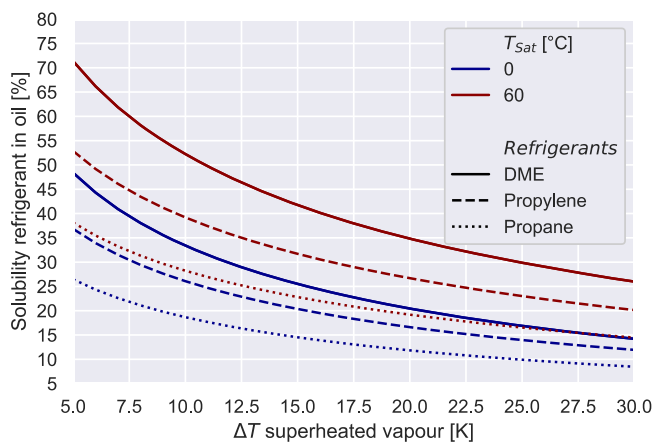


Fig. 12. Solubility as a function of the level of superheated vapour for different refrigerants.

pumps.

For the NRTL methods, the formulation of the interaction parameters was found to be critical for the minimisation of errors. The temperature-dependent form similar to Antoine’s equation (Antoine, 1888) as suggested by Leon Poesy (1996) was found to be best. The form of the interaction parameters was adopted also by other studies, such as the one by Wang et al. (2021).

The simple and explicit form of the developed SSC correlation may incentivize refrigerant-oil solubility characterization also in those models where computational power or allowed running time is limited. Existing semi-empirical models in the literature were found to reach a high level of accuracy, however, their implementation may be considered complicated. Moreover, existing models, both semi-empirical and empirical, are formulated in implicit form, which requires solubility to be determined iteratively. The proposed SSC correlation, on the other hand, thanks to its compact and explicit formulation allows for a simple implementation and a fast execution. The latter may be a requirement in screening and parametric analysis or dynamic simulations. Overall, refrigerant-oil solubility is more likely to be included in the modelling of heat pumps and refrigeration equipment.

Moreover, for the tested datasets, the SSC correlation was found to be

robust and maintained an acceptable mean accuracy also at a very low number of data points. On the other hand, the rising uncertainty of the errors at the low amount of data points suggests that the choice of the data points may be critical to error minimization. Further analysis of the most effective selection criteria is recommended. Furthermore, particular care shall be taken when using the SCC to predict solubility at high refrigerant concentrations and low operating pressures and temperatures. Further testing and efforts to improve the SSC correlation are suggested.

The SSC correlation may also find use in the early design of new refrigerants and oils. Knowing the single coefficient of a specific refrigerant-oil combination with satisfactory solubility characteristics, the coefficient can be used as a reference or target for the design of new oils and refrigerants. For instance, thanks to this study it was found that Propane with the same PAG oil was able to reach an SSC a_1 equal to $3.34 \cdot 10^{-4}$ with the lowest resulting solubility. For the development of oils which offer low DME solubility, a similar a_1 target could be set. The existing DME-PAG single solubility coefficient a_1 is $6.53 \cdot 10^{-4}$, 95 % higher than Propane suggesting significant efforts may be required.

The difference between solubility levels of DME and Propylene compared to Propane was found to be significant. At the saturation temperature of 60 °C, DME would require about an additional 26.5 K in superheated vapour to match the same solubility of Propane. Propylene instead would require 7 K to 11 K additional superheating. From the results, it might be obvious to deduct that higher refrigerant solubility is expected in heat pump compressors using DME and Propylene compared to Propane. However, heat pumps using DME, and Propylene are expected to also operate with higher discharge temperatures at the same condensing temperatures. Since the superheated vapour level on the compressor high-pressure side is expected to be larger, a negative effect on solubility is expected. Solubility data are not enough to determine the refrigerant quantity in a compressor. Operating temperatures, pressures, densities, volumes, and kinetics may also play an important role. For having a realistic picture of the quantity of refrigerant dissolved in compressor oil for heat pumps, the modelling of a compressor and a full heat pump analysis was recommended.

The obtained results do not give information on the charge levels to be expected in the compressor. However, they may give insights into the solubility levels at the low- and high-pressure side. While the temperature on the suction side of a rotary compressor is close to the evaporator outlet condition, higher temperature uncertainties can be expected for the oil sump on the high-pressure side. The developed solubility models could be further integrated into a compressor or evaporator model that determines the amount of refrigerant contained by the single component.

The study did not investigate the effect that solubility has on viscosity and lubricity. Great care shall be dedicated to this aspect for the development of reliable and efficient compressors and vapour-compression cycles. Particularly for conditions at high discharge temperatures and high pressures, where both high solubility and low viscosity can be expected to be the most critical. These challenges may be expected for refrigerants with high solubility in oil, such as DME. Moreover, despite the foamy mixtures were not experienced in the experiments, it cannot be excluded in the case experiments are carried out on a compressor.

As discussed in the study from [Fernando et al. \(2004\)](#), the minimization of solubility should be a priority, especially for compressors in low-charge applications. The PAG oil tested was developed for Propane and small charge systems. For the future investigation and development of DME as a refrigerant, the custom design of an oil optimized for use with DME can be considered.

5. Conclusions

This study investigated the solubility and miscibility of Propylene and Dimethyl Ether (DME) in polyalkylene glycol oil for Propane

compressors. The solubility experimental data were used to fit different local composition models from the literature, the Wilson and Heil equations, and the Non-Random Two-Liquid model. It was found that the formulation of the binary interaction parameters of the NRTL and Heil models had a high influence on the accuracy of the models. NRTL with the second formulation of interaction parameters reduced deviations to an average of 1.1 % for Propylene. On the other hand, The Heil model achieved the best accuracy for DME, with an average error of 1.3 %.

Two new empirical correlations were introduced, and they were fitted to the experimental data. The first empirical correlation used six fitting coefficients and resulted in an average deviation of 1.2 for Propylene and 2.8 % for DME.

The second empirical correlation, formulated in a compact form and with a single solubility coefficient, was performed with an average deviation between experiments and the models of 1.6 % for Propylene and 2.5 % for DME. The fitted coefficient was found to be proportional to the solubility of the refrigerant.

Moreover, the SSC correlation showed performance robustness against the decrease of available data points. For Propylene, reducing the number of data points used during model fitting to only two data points, increased the percentage error, on average, from 1.6 % to 2 %. The maximum error, on average, remained stable, and slightly decreased from 8.2 to 8 %. For DME using only two data points, the average error increased from 2.5 % to 3.8 %, and the maximum error changed from 12 to 13.9 %. On the other hand, it was found that, when the amount of training data points were reduced below four, error uncertainties increased significantly. The results suggest that data points selection may have an important role on error minimization and further analysis on effective selection methods is recommended. The proposed SSC correlation, thanks to its compact and explicit formulation, may allow a simple implementation and a fast execution. It may incentivize the characterization of refrigerant-oil solubility in those applications where computational power or allowed running time are limited.

The same correlation allowed for comparing the solubility of Propylene and DME with the reference refrigerant Propane, at different temperatures, pressures, and compositions. It was found that at the same conditions, Propane had the lowest solubility while DME had the highest, and that the difference between solubility levels increased at growing pressures. For the potential future adoption of DME in low-charge heat pumps, the design of new low-solubility lubricants might be needed.

CRedit authorship contribution statement

Matteo Caramaschi: Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Software, Validation, Visualization, Writing – original draft, Writing – review & editing. **Jonathan Notturmo:** Conceptualization, Formal analysis, Methodology, Supervision, Writing – review & editing, Data curation, Investigation, Software. **Guglielmo Vaccaro:** Conceptualization, Data curation, Methodology, Software, Writing – original draft, Writing – review & editing. **Wiebke Meesenburg:** Conceptualization, Methodology, Supervision, Writing – review & editing. **Jonas Jensen:** Conceptualization, Methodology, Project administration, Writing – review & editing, Supervision. **Brian Elmgaard:** Conceptualization, Funding acquisition, Methodology, Project administration, Resources, Supervision, Writing – review & editing. **Brice Tremeac:** Project administration, Resources, Supervision, Writing – review & editing. **Pascal Tobaly:** Conceptualization, Project administration, Resources, Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence

the work reported in this paper.

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