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Research Article

Thermodynamic Properties of R1234yf and DMAC Binary Solution

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Abstract

To assess the performance of potential refrigerant-absorbent pairs, it is essential to have thermodynamic properties of both the pure components and their mixtures. Since these mixtures do not behave ideally, the properties of the solutions can only be obtained through experimental means. This paper's proposed candidate pair is the environmentally friendly refrigerant 2,3,3,3-Tetrafluoropropene (R1234yf) and the organic solvent Dimethylacetamide (DMAC). For this purpose, an experimental setup was designed to obtain data at the equilibrium point between the gas and liquid phases. The collected data was analyzed using models based on the vapor-liquid equilibrium of mixtures. Correlations were established for pressure-temperature and refrigerant concentration in the liquid phase and the solution's enthalpy. These results can facilitate further investigations into the solution's compatibility as an alternative working pair.

Keywords: *Vapor liquid equilibrium; binary solutions; refrigerant 2,3,3,3-tetrafluoropropene (R1234yf) and dymethylacetamide (DMAC); thermodynamic properties.*

1. Introduction

The use of environmentally friendly refrigerants, such as hydrocarbons (HCs), hydrofluoroolefin (HFO), R744 (carbon dioxide), and eco-safe nano refrigerants, can help to reduce ozone depletion potential (ODP) and global warming potential (GWP). When selecting refrigerants, it is important to consider other environmental factors such as toxicity, flammability, vapor pressure, solubility, stability, and lubricity [1]. HCs have gained popularity since the 1990s due to their relatively low environmental impact. However, the introduction of perfluorocarbons and HFCs (Hydrofluorocarbon) faced criticism in the Kyoto Protocol of 1997 due to their high GWP. Recently, HFO refrigerants, including R1234yf, R1234ze, R1336mzz(Z), and R1336mzz(E), have been implemented, showing significant progress in the development of environmentally friendly options since 2008.

In commercial diffusion absorption cooling systems, the options for binary solutions used with an auxiliary gas are limited to ammonia-water or water-lithium bromide combinations. The ammonia-water mixture is widely used in diffusion-absorption refrigeration systems due to its chemical stability across a wide range of pressures and temperatures. Additionally, ammonia has a high latent evaporation heat and a low freezing point (-77°C), making it suitable for applications requiring low evaporation temperatures. However, since ammonia water is volatile, a rectifier is usually required to separate the evaporated water from ammonia, leading to heat loss and reduced energy efficiency. Despite these drawbacks, the advantages of ammonia water include its environmental friendliness and production cost (10-20%). Its thermophysical properties can be obtained from various sources [2-5]. One disadvantage of using ammonia water as a working fluid is its high operating pressure, toxicity, and corrosive nature towards copper and copper alloys, limiting its use to materials like carbon steel.

Although ammonia-water-hydrogen has been used as a working fluid in diffusion-absorption refrigeration systems for many years, researchers have explored alternative working mixtures to minimize activation energy. Pfaff et al. [6] suggest the use of the lithium bromide-water mixture as a working fluid in diffusion-absorption systems. The input temperatures in the generator range from 66°C to 78°C, which activates the bubble pump and makes this mixture appealing for solar refrigeration applications. Water, when used as a refrigerant fluid, has an evaporation temperature limited to values above 0°C, making it a viable alternative for air-conditioning applications. The advantage of the lithium bromide-water mixture is that the absorbent is non-volatile, eliminating the need for rectifiers used in ammonia-water systems. However, vacuum pressures pose a disadvantage, and high concentrations of lithium bromide can lead to crystallization problems. Due to the corrosive nature of salts, installations using this mixture require construction materials like copper. Thermo-physical properties of this mixture can be obtained from various sources [7-11].

Fluoride refrigerants are recommended for their good solubility with organic solvents such as N, N′-dimethylformamide (DMF) and N, N′-dimethylacetamide (DMAC). Compared to ammonia, these refrigerants have the advantage of being less toxic, and compared to water, they can reach temperatures below 0°C suitable for refrigeration. They exhibit chemical stability, non-corrosiveness, and complete miscibility over a wide range of temperatures [12,13]. DMF is commonly used as an absorbent in absorption refrigeration systems and has been utilized in diffusion-absorption systems as well. However, precautions

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must be taken to prevent leaks when DMF reacts with certain metals in the presence of oxygen. Various researchers [14-17] have studied the properties of halogenated refrigerants combined with DMF. Koyfman et al. [18] conducted an experimental study using R22 and DMF as the working fluid, achieving evaporation temperatures below 0°C with activation temperatures in the generator between 50°C and 90°C, resulting in an energy efficiency of 0.35. Zohar et al. [19] incorporated refrigerants R32, R124, R125, and R134a into their analysis using DMF as an absorbent in all cases, observing valid results under different operating conditions. They found that these mixtures could be activated at a lower temperature (150°C), but with a lower coefficient of performance and higher condensation and evaporation temperatures compared to the ammonia-water mixture. DMAC, another commercially available organic compound with good solubility, is considered an absorbent for refrigeration systems [16]. It has been combined with halogenated refrigerants in diffusion-absorption applications. Since the boiling temperatures of both compounds are very close, a certain amount of DMAC evaporates in the generator, necessitating the use of a rectifier to avoid condensing DMAC, which reduces cooling capacity and increases manufacturing costs. Ezzine et al. [20] showed that R124-DMAC is a good working pair, achieving lower vapor pressures than the mixture with DMF and activation temperatures between 80°C and 180°C, making them ideal for activation using solar energy, geothermal sources, residual heat, or other sources. Light hydrocarbons with organic solvents have also been explored in the literature [21]. Hydrocarbons and mixtures of alkanes have been extensively studied as refrigerants in vapor-compression refrigerating machines and heat pumps, as evidenced in existing literature [22, 23]. Simulation results using Fortran for an absorption refrigeration model, considering ten alkane mixtures with both air and water cooling were presented by [24]. Dardour et al. [25] performed simulations to analyze the overall behavior and performance of an absorption-diffusion system utilizing propane-nonane-hydrogen as the working fluid. Propane served as the refrigerant, nonane as the absorbent, and hydrogen as the inert auxiliary gas. The Peng-Robinson equation of state was employed to extract the thermodynamic characteristics of individual substances, as well as their binary solution properties, from the Aspen software databank. Hydrocarbon mixtures can be activated at temperatures between 120°C and 150°C, but their coefficient of performance is sometimes lower than the previously described mixtures.

The study of [26] investigated the use of a nano ferrofluid (Fe_3O_4-ammonia/water) as a binary working solution in DAR system and the research presented an experimental study demonstrating the enhancement in DARS (Diffusion Absorption Refrigeration Systems) performance using the nano ferrofluid with helium as an inert gas. The experiments were conducted with nano ferrofluids containing 0.05 wt.% and 0.1 wt.% Fe3O4 nanoparticles concentrations in 300 ml NH3/H2O base-fluid, with the addition of 1 wt.% concentration of polyvinyl pyrrolidone surfactant, both with and without an external magnetic field. The results showed that the DARS with 0.1 wt.% nano ferrofluid under the external magnetic field exhibited the best performance.

While the thermodynamic properties of pure absorbents and refrigerants are well known, the thermodynamic properties of binary organic mixtures suitable for absorption refrigeration systems have not been fully investigated. Additionally, the behavior of Hydrofluoroolefin (HFO) refrigerants with various absorbents remains largely unexplored. Since the thermophysical properties of the working fluids depend on the refrigerant's concentration in the solution, the objective of this study is to experimentally determine the pressure-temperature-concentration of binary mixtures at equilibrium, as well as the enthalpy-temperature-concentration data. The investigated refrigerants include R1234yf and DMAC (N,N'-dimethylacetamide, C_4H_9NO)

2. **Research Design and Methods**

For the absorbent refrigerant pair to be acceptable, the refrigerant and absorbent must have a strong molecular attraction. Since these forces cause the fluids to deviate from ideal fluid behavior, it is required that the transport and thermodynamic properties of the refrigerant and solution will be determined experimentally.

The research design is based on the work of [27] and [28]. The experimental set up consists of 300 ml Parr 4383 reactor equipped with pressure and temperature measuring devices to determine the system’s equilibrium data (see Figure .1). To keep the reactor at constant temperature, it was heated from the sides by a controlled electrical heating jacket, and was insulated around and above. The contents of the vessel were stirred magnetically with a Teflon-clad stirring capsule. The two thermocouples inserted into the reactor measured the temperature of the liquid phase and temperature of the gas phase above the solution. The system equilibrium pressure in the vessel was measured with the pressure gauge. The equilibrium vapor pressure-temperature-concentration relationships was determined according to the following procedure.

1. First, the reactor was weighed.
2. A predetermined amount of the absorbent, was introduced to the reactor, and the reactor was reweighed.
3. A vacuum pump degassed the absorbent, and the vessel was weighed again. Then, the refrigerant was inserted and the vessel was weighed again.
4. The mixture was heated up in steps of 5ºC at high refrigerant concentrations and 10ºC at low concentrations.
5. When thermal equilibrium (showed by the equality in the temperature readings) between the gas phase and the mixture was achieved, the readings were logged.
6. After the completion of a series of equilibrium measurements, the vessel was cooled down to the ambient temperature, a certain amount of gas was inserted, and the reactor was weighed again.

Figure 1. Experimental setup.

3. Data Analysis Based on Vapor Liquid Mixture Equilibrium

At the pressure vessel, the liquid phase is at the bottom and the gaseous phase is above it. The mass concentration of the refrigerant in the liquid phase is determined by the following procedure. When thermal equilibrium is reached, i.e., the temperature of the liquid phase equals the gas phase, the total volume of the vessel (known parameter) is:

$$V_T = V_S + V_G \tag{1}$$

The total volume V_T consists of the sum of the solution volume, V_S , and the gas volume, V_G .

The mass of the components m_R and m_A (refrigerant and absorbent) in the vessel is the sum of each component in the liquid phase and in the gas phase (also known parameters):

$$m_R = m_{RS} + m_{RG} \tag{2}$$

$$m_A = m_{AS} + m_{AG} \tag{3}$$

where m_{RS} is the mass of the refrigerant in the liquid phase, m_{RG} is the mass of the refrigerant in gas phase, m_{AS} is the mass of the absorbent in the liquid phase and m_{AG} is the mass of the absorbent in the gas phase.

The volumes of the liquid and the gas phases, V_S and V_G are:

$$V_S = m_{RS} v_{RS} + m_{AS} v_{AS} \tag{4}$$

$$V_G = m_{RG} v_{RG} + m_{AG} v_{AG} \tag{5}$$

where v_{RS} , v_{AS} , v_{RG} , v_{AG} are the specific volumes of the refrigerant and the absorbent in the liquid and vapor phases as function of the temperature and/or the pressure of the system. The specific volumes of v_{RS} , v_{AS} can be calculated from the densities of the refrigerant and absorbent in the liquid phase:

$$v_{RS} = \frac{1}{\rho_{RS}} \tag{6}$$

$$v_{AS} = \frac{1}{\rho_{AS}} \tag{7}$$

In general, the density of the refrigerants can be obtained from the EES (Engineering Equation Solver) and density of the absorbent component in the liquid phase can be calculated by following polynomial equation:

$$\rho_{iS} = \rho_0 + \rho_1 T + \rho_2 T^2 \tag{8}$$

Note that the density is in the units of ${gr}/{cm^3}$ and the temperature in Celsius. The density constants ρ_0, ρ_1, ρ_2 are presented in Table 1.

Table 1. Density constants [26].

	ρ_0	ρ_1	ρ_2
DMAC	0.9528000	0.0005856	-0.000003363

The specific volume of the components in the gas phase v_{RG} and v_{AG} are the ratio of molar volume to molecular weight and will be calculated from Peng-Robinson equation of state (EOS).

$$v_{iG} = \frac{V_{iG}}{M_i} \tag{9}$$

$$P = \frac{RT}{V_{iG} - b_i} - \frac{a_i}{{V_{iG}}^2 + 2b_i V_{iG} - b_1^2} \tag{10}$$

$$a_i = 0.45724 \frac{R^2 {T_{ci}}^2}{p_{ci}} \left[1 + f_w \left(1 - {T_{ri}}^{0.5}\right)\right]^2 \tag{11}$$

$$b_i = 0.0778 \frac{R T_{ci}}{p_{ci}} \tag{12}$$

$$f_w = 0.37464 + 1.54226\omega + 0.26992\omega^2 \tag{13}$$

where, a_i and b_i are cubic EOS parameters, R is the ideal gas constant, T_{ci} and p_{ci} are critical temperature and pressure of the substance , T_{ri} is the reduced temperature and ω is the acentric factor. The data for the components is presented in Table 2.

Table 2. Data for the components.

	$T_c\left[{}^{O}C\right]$	$p_c\left[bar\right]$	$M\left[{gr}/{mol}\right]$	ω
R1234yf	94.7	33.82	114	0.276
DMAC	382.4	42.11	87.12	0.363

After the specific volumes are calculated, the weight fraction of the refrigerant in liquid phase can be defined by:

$$\xi_R = \frac{m_{RS}}{m_{RS} + m_{AS}} \tag{14}$$

and the weight fraction of the absorbent was calculated by:

$$\xi_A = 1 - \xi_R \tag{15}$$

Similarly, the weight fractions of the refrigerant and the absorbent in gas phase can calculated by:

$$\psi_R = \frac{m_{RG}}{m_{RG} + m_{AG}} \tag{16}$$

$$\psi_A = 1 - \psi_R \tag{17}$$

The mole fractions of the refrigerant and the absorbent in liquid phase were calculated by:

$$x_R = \frac{\xi_R M_A}{\xi_R M_A + \xi_A M_R} \tag{18}$$

$$x_A = 1 - x_R \tag{19}$$

The mole fractions of the refrigerant and the absorbent in gas phase can be calculated by:

$$y_R = \frac{\psi_R M_A}{\psi_R M_A + \psi_A M_R} \tag{20}$$

$$y_A = 1 - y_R \tag{21}$$

There is one unknown parameter left. When there is an equilibrium between two phases, the fugasities are equal $f_i^V = f_i^L$ and for each component obtained:

$$y_i p = \gamma_i x_i p_i^{sat} \Phi_i \tag{22}$$

where y_i is the mole fraction of component i, p is the measured pressure of the system, γ_i is the activity coefficient, x_i is the molar concentration of component *i* in the liquid solution, p_i^{sat} is the saturation pressure of component i, Φ_i is the correction factor for pressure changes in the system and is calculated the following way:

$$\Phi_i = \exp\left[\frac{\left(V_i^L - B_i\right)\left(p - p^{sat}\right)}{RT}\right] \tag{23}$$

where V_i^L and B_i are the molar volume and second virial coefficient of component *i* in the liquid phase.

For a binary system consisting of an absorber and a refrigerant:

$$y_A p = \gamma_A x_A p_A^{sat} \Phi_A \tag{24}$$

$$y_R p = \gamma_R x_R p_R^{sat} \Phi_R \tag{25}$$

The activity coefficient for each of the components is going to be calculated from the Van Laar relation for the molar excess Gibbs free energy g^E and activity coefficient γ_i :

$$\frac{g^E}{RT} = \frac{A x_A x_R}{x_R\left(\frac{B}{A}\right) + x_A} \tag{26}$$

$$\ln \gamma_R = A\left(1 + \frac{A}{B}\left(\frac{x_R}{x_A}\right)\right)^{-2} \tag{27}$$

$$\ln \gamma_R = A\left(1 + \frac{A}{B}\left(\frac{x_R}{x_A}\right)\right)^{-2} \tag{28}$$

where A, B are constants depending on temperature. Division of Eqs. (27) and (28) gives a new relationship between the activity coefficients of the two components:

$$\frac{\ln \gamma_A}{\ln \gamma_R} = \frac{A}{B}\left(\frac{x_R}{x_A}\right)^2 \tag{29}$$

When the activity coefficients are substituted into the equilibrium equations, it yields:

$$\ln \gamma_A = \ln\left(\frac{y_A p}{x_A p_A^{sat} \Phi_A}\right) \tag{30}$$

$$\ln \gamma_R = \ln\left(\frac{y_R p}{x_R p_R^{sat} \Phi_R}\right) \tag{31}$$

Due to the lack of an equation, the following assumption can be used:

$$\frac{B}{A} = \frac{V_A^L}{V_R^L}$$

(32)

After obtaining pressure-temperature-concentration data, it will be expressed in as a polynomial function:

$$p = \sum_{j=0}^{j=5} \sum_{i=0}^{i=6} p_{ij} \xi_R{}^i T^j \tag{33}$$

p_{ij} values were obtained by regression of the equilibrium results. The temperature T is in Kelvin. The results of the pressure-temperature-concentration are presented in Figure 2. Values of p_{ij} are presented in Table 3.

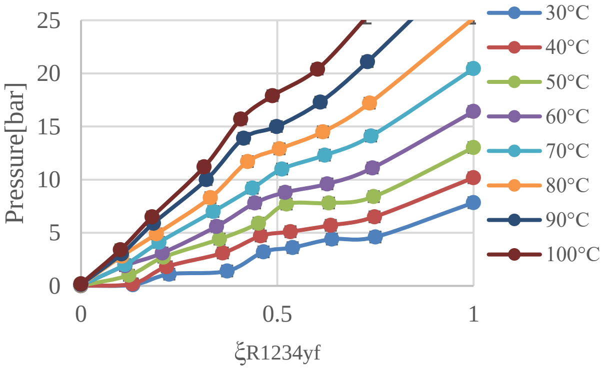

Figure 2. Pressure-temperature-concentration results in equilibrium.

Table 3. p_{ij} *coefficients of R1234yf and DMAC.*

i.j	0	1	2	3	4	5
0	277	-28.3	1.15	-0.0241	0.00028	0
1	-49	1.65	0.0986	-0.00437	0.00006	0
2	-8780	819	-29.9	0.527	-0.0045	0.00002
3	36700	-3390	123	-2.15	0.0182	-0.00006
4	-51700	4760	-172	3.01	-0.0255	0.00008
5	24100	-2170	78.7	-1.38	0.0117	-0.00004
6	-395	0	0	0	0	0

Table 4. h_{ij} *coefficients of R1234yf and DMAC.*

i,j	0	1	2	3	4	5	6
0	7670	192000	1440000	-4710000	6770000	-3270000	344000
1	638	16800	-127000	424000	632000	343000	0
2	18.2	-517	3960	13200	19800	-10700	0
3	-0.229	7.43	57.3	192	-288	157	0
4	0.00113	-0.00509	0.389	-1.31	1.97	1.07	0
5	0	0.00013	-0.001	0.00339	-0.00509	424000	0

In order to obtain the enthalpy of the solution, the following formula is used:

$$h = h_R \xi_R + h_A \left(1 - \xi_R\right) + h^E \qquad (34)$$

h_R and h_A are the specific enthalpies of the pure components, h^E is the excess specific enthalpy of mixing and h is the enthalpy of the solution in the liquid phase. The data for the calculations of the specific enthalpies of the absorbent and the refrigerant can be found in the literature. However, it is important to state that the reference point is $100\frac{kJ}{kg}$ at $0^O C$. The molar excess enthalpy H^E can be calculated the following manner:

$$H^E = -RT^2 \left(\frac{\partial \left(g/RT \right)}{\partial T} \right)_{p,x} \qquad (35)$$

Gibbs free energy g equals to:

$$g = RT\left(x_A \ln \gamma_A + x_R \ln \gamma_R\right) \qquad (36)$$

The results of the excess enthalpy with respect to molar refrigerant concentration are presented in Figure 3.

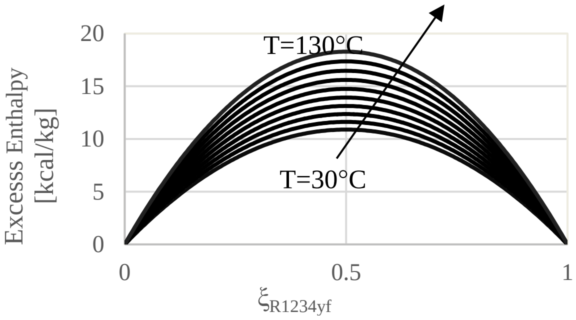

Figure 3. Excess enthalpy as a function of the refrigerant concentration for various equilibrium temperatures.

The specific excess enthalpy can be expressed in a polynomial form:

$$h^E = 4.1868 \sum_{i=0}^{i=5} \sum_{j=0}^{j=6} h_{ij} \xi_R{}^j T^i \qquad (37)$$

The excess enthalpy of the liquid solution is in kJ/kg and the temperature T is in Celsius. The coefficients h_{ij} were determined by regression of the equilibrium data. Their values are presented in Table 4.

Enthalpy of the liquid solution for various temperatures as a function of the refrigerant mass concentration is presented in Figure 4.

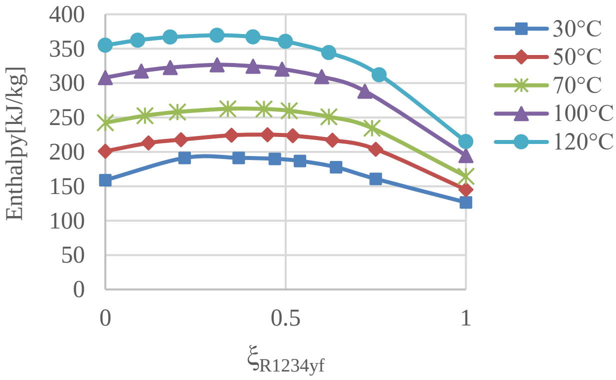

Figure 4. Liquid phase enthalpy as a function of the refrigerant concentration for various equilibrium temperatures.

4. Conclusions

This paper presents a thorough analysis of the thermodynamic characteristics of a new binary solution. The data utilized in this study was gathered through experimental measurements of mass, temperature, and pressure of the solution's phases at equilibrium. By employing a relatively simple setup and conducting complex data analysis, the thermodynamic properties of the binary solution consisting of R1234yf and DMAC were determined. The findings, published in this paper, provide researchers with the essential thermodynamic information needed for further investigations.

The outcomes, specifically the pressure-temperature-refrigerant concentration in the liquid phase, can be utilized to ascertain the solution concentration based on temperature and pressure at the absorber of the DAR system. Additionally, the enthalpy data is crucial for calculating the performance of the DAR system when utilizing R1234yf and DMAC as working fluids.

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Nomenclature

A	First virial coefficient	
a	EOS parameter	
B	Second virial coefficient	
b	EOS parameter	
f	Fugacity	$\left[bar\right]$
g	Gibbs free energy	$\left[{}^{kJ}/_{kg}\right]$
h	Enthalpy	$\left[{}^{kJ}/_{kg}\right]$
M	Molar mass	$\left[{}^{kg}/_{kmol}\right]$
m	Mass	$\left[kg\right]$
p	Pressure	$\left[bar\right]$
T	Temperature	$\left[{}^{o}C\right],\left[K\right]$
V	Volume	$\left[m^3\right]$
v	Specific volume	$\left[{}^{m^3}/_{kg}\right]$
x	Mole fraction in liquid phase	
y	Mole fraction in gas phase	

Greek symbols

γ	Activity coefficient	
ξ	Weight fraction in liquid phase	
ρ	Density	$\left[{}^{kg}/_{m^3}\right]$
Φ	Correction factor	
ψ	Weight fraction in gas phase	
ω	Acentric factor	

Subscripts

AG	Absorbent in gas phase
AS	Absorbent in liquid phase
c	Critical
G	Gas
i	Component index
RG	Refrigerant in gas phase
RS	Refrigerant in liquid phase
S	Solution
T	Total

Superscript

E	Excess
L	Liquid
V	Vapor

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