

OPTIMIZATION OF UNIQUAC INTERACTION PARAMETERS FOR ABSORPTION REFRIGERATION MACHINE WORKING FLUIDS.

Ilhem LOUAER¹, Nardjess BOUNAB² et Abdeslam Hassen MENIAI²

1 Laboratoire de l'Ingénierie des Procédés de l'Environnement, Mentouri Brothers University Constantine, Alegria.

2 Laboratoire de l'Ingénierie des Procédés de l'Environnement, Constantine 3 University, Alegria.

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Abstract

To be able to pre-select new working fluids for absorption refrigeration systems, it is necessary to have a good understanding in the calculation or correlation of thermophysical properties of working fluids. The choice of the most appropriate thermodynamic model for the prediction or calculation of thermophysical properties is a crucial task. Consequently the present study deals with the determination of excess properties for refrigerant mixtures which are required for the calculation of the various involved enthalpies and hence the used refrigeration machine.

The present study considered the pairs (2-chloro-1,1,1,2-tetrafluoroethane) (R124) as a refrigerant in combination with an organic absorbent (N, N'- Dimethylacetamide) DMAC and (1,1,1,2-tetrafluoroethane) (R134a) as a refrigerant in combination with an organic absorbent dimethylether tetraethyleneglycol (DMETEG) by calculating the molecular interaction parameters for the UNIQUAC model using the excess enthalpy of mixing at eight different temperatures varying from 0°C to 140°C by a step of 20°C for the first pair and varying from 20°C to 140°C for the second pair.

The obtained interaction parameters values led to mixing excess enthalpy results very close to experimental values, contrary to the UNIFAC model which showed huge deviations. Therefore the optimization technique based on the Simplex method proved to be very reliable in the determination of interaction parameters for the UNIQUAC model.

Keywords: UNIQUAC; UNIFAC; Excess enthalpy; Refrigerant; Absorbent.

Résumé

Pour pouvoir présélectionner de nouveaux fluides de travail pour les systèmes de réfrigération à absorption, il est nécessaire de bien comprendre le calcul ou la corrélation des propriétés thermophysiques des fluides de travail. Le choix du modèle thermodynamique le plus approprié pour la prédiction ou le calcul des propriétés thermodynamiques est une tâche cruciale. En conséquence, la présente étude traite de la détermination des propriétés excédentaires des mélanges de réfrigérants nécessaires au calcul des différentes enthalpies impliquées et donc de la machine frigorifique utilisée.

La présente étude a examiné les paires (2-chloro-1,1,1,2-tetrafluoroéthane) (R124) en tant que frigorigène en combinaison avec un absorbant organique (N, N'-diméthylacétamide) DMAC et (1,1,1, 2-tetrafluoroéthane) (R134a) en tant que réfrigérant en association avec un diméthyléther éther tétraéthylène glycol (DMETEG) absorbant organique en calculant les paramètres d'interaction moléculaire pour le modèle UNIQUAC en utilisant l'enthalpie de mélange en excès à huit températures différentes variant de 0 ° C à 140 ° C par un pas de 20 ° C pour la première paire et variant de 20 ° C à 140 ° C pour la deuxième paire.

Les valeurs des paramètres d'interaction obtenues ont conduit à mélanger les résultats d'excès d'enthalpie très proches des valeurs expérimentales, contrairement au modèle UNIFAC qui présentait des écarts importants. Par conséquent, la technique d'optimisation basée sur la méthode Simplex s'est révélée très fiable pour déterminer les paramètres d'interaction du modèle UNIQUAC.

Mots clés: UNIQUAC; UNIFAC; Excès d'enthalpie; Réfrigérant; Absorbant.

ملخص

لكي تكون قادراً على تحديد سوائل عمل جديدة مسبقاً لأنظمة التبريد بالامتصاص ، من الضروري أن يكون لديك فهم جيد في حساب أو ترابط الخصائص الحرارية الفيزيائية للسوائل العاملة. إن اختيار أنسب نموذج ديناميكا حرارية للتنبؤ بالخصائص الحرارية أو حسابها هو مهمة حاسمة. وبناءً على ذلك ، تتناول الدراسة الحالية تحديد الخصائص الزائدة لمخاليط التبريد اللازمة لحسابات المستهلك المختلفة ومن ثم آلة التبريد المستخدمة.

نظرت الدراسة الحالية في أزواج) 2 (R124) (chloro-1,1,1,2-tetrafluoroethane) كمبرد مع الماصة العضوية (N) ، N'-

Dimethylacetamide) (DMAC) و) 1,1,1,2 (R134a) tetrafluoroethane) كمبرد مع توليف عضوي ثنائي ميثيلبر رباعي إيثيلينجلكول

(DMETEG) عن طريق حساب معاملات التفاعل الجزيئي لنموذج UNIQUAC باستخدام المحتوى الحراري الزائد للاختلاط عند ثمانية درجات حرارة مختلفة تتراوح

من 0 درجة مئوية إلى 140 درجة مئوية بخطوة مقدارها 20 درجة مئوية للزوج الأول وتتراوح من 20 درجة مئوية إلى 140 درجة مئوية للزوج الثاني.

أدت قيم معاملات التفاعل التي تم الحصول عليها إلى خلط نتائج المحتوى الحراري الفائض قريبة جداً من القيم التجريبية ، على عكس نموذج UNIFAC الذي أظهر

انحرافات ضخمة. ولذلك ثبت أن تقنية التحسين القائمة على طريقة Simplex موثوقة للغاية في تحديد معاملات التفاعل لنموذج UNIQUAC.

الكلمات المفتاحية: UNIQUAC. UNIFAC. المحتوى الحراري الزائد التبريد. ماص.

1. INTRODUCTION

The vapor absorption refrigeration system is a kind of important energy saving technology and emission reduction.

It can utilize renewable energy such as solar and geothermal energy or waste energy from industrial processes. In addition, the absorption system has no vibration and noise and therefore, environmentally friendly and becomes a competitive alternative to the conventional mechanical-driven vapour compression refrigeration systems [10]. In spite of above advantages of the absorption refrigeration system, it is not used for commercial purposes because of its low performance; so it needs improvement.

The performance of the absorption refrigeration cycle depends on its configuration and is critically dependent on the chemical and thermodynamic properties of the working pairs composed of a refrigerant and an absorbent [12, 13, 14, 18]. Accordingly, many researchers have attempted to develop new working pairs for the absorption refrigeration cycles to improve their performance [15, 16, 17]

A wide variety of refrigerant-absorbent combinations (both organic and inorganic) have been suggested for vapor absorption cooling systems in literature. A survey of absorption fluids provided by Marcriss [19] suggests that, there are some 40 refrigerant compounds and 200 absorbent compounds available. Most of these are two component systems. The two common working fluids in absorption refrigeration cycle are water-LiBr and ammonia-water. These two working fluids present drawbacks which directly affect the performance of these refrigeration systems. Hence to overcome the drawbacks associated with the conventional working pairs, finding a suitable mixture is inevitable.

Furthermore, to be able to evaluate the performance of the cycle, one must have at least the vapor liquid equilibrium data of the working fluid mixture. Obviously, the vapor liquid equilibrium data of the working fluid must be solid and robust to improve the quality of the results [10].

The calculation of the absorption systems requires the knowledge of the enthalpy of the liquid and vapor phase mixtures as a function of temperature, pressure and concentration. However most of the liquid systems deviate from ideality and therefore the calculations of specific excess enthalpies are necessary.

These are important thermodynamic properties are functions of the temperature and the composition of the considered solutions and do have an influence on the usual and necessary enthalpy-concentration diagrams. Hence the thermodynamic calculation of the key operating parameters of an absorption cycle, i.e. the coefficient of performance (COP) and the circulation ratio with a possible variation of the order of 15% [1].

In addition, to have solid enthalpy-concentration diagrams of the working fluid, it is necessary to have a good correlation of the specific excess enthalpy of the working fluid mixtures using and to choose appropriate thermodynamic model to describe the activity coefficient,

which is a measure of the deviation from ideality of the liquid system. Various models for the activity coefficient are available in the literature and most of them are based on the variation of the excess Gibb free energy g^E with composition. They predict more or less depending on the type of the considered systems. Hence, the problem of the choice of the most appropriate thermodynamic model is not an easy task.

Thence, advance study on the thermodynamic model to describe the vapor liquid equilibrium properties of the new working fluids mixtures for the absorption cooling systems is undoubtedly necessary before investigating the performance of the absorption cycle using new working fluids.

In the literature, a number of studies were carried out on techniques to improve the performance of absorption refrigeration systems directly or indirectly. In many previous works, the Non-Random two-liquids (NRTL) activity coefficient model was used to correlate the vapor liquid equilibrium data of non ideality binary systems such as HFCs and organic solvents mixtures for absorption refrigeration cycles [10,11,20,21,22]. The (NRTL), Redlich-Kwong-Soave cubic equation of state (RKS), Perturbed-chain Statistical Associating fluid Theory (PC-SAFT) and UNIFAC models were studied by Hifni M Ariyadi [10] in order to select the appropriate model to be used to correlate the thermophysical properties needed to investigate the thermal performance of the absorption systems based on ammonia/ionic liquid working fluids.

The present work can be regarded as a continuation of a previous one [2] where the binary system R124-DMAC was considered in an absorption refrigeration machine and the modeling of the performance and the recirculation ratio was carried out by means of the UNIFAC, which is based on the contributions of groups of atoms on a structural basis ,and considers the liquide solution to be made up of groups rather than pure components as introduced by Fredenslund et al. [3]. Interaction parameters between the functional groups may be evaluated using table I [7,8]

The results obtained in [2] were not always in a reasonable agreement with experimental data and this may certainly be due to errors induced by the approximations upon which is based the group contribution concept, particularly the property additivity.

Therefore the aim of the present work is to show the influence of the chosen thermodynamic model on the performance of an absorption refrigeration machine, rather using the UNIQUAC model which is the precursor of UNIFAC and is based on molecular rather than group interaction parameters [4]. Two working fluids were considered in this study, the first one is (2-chloro-1,1,1,2-tetrafluoroethane) (R124) as a refrigerant in combination with an organic absorbent (N, N'- Dimethylacetamide) DMAC and the second one is (1,1,1,2-tetrafluoroethane) (R134a) as a refrigerant in combination with an organic absorbent dimethylether tetraethyleneglycol (DMETEG).

II. CALCULATION PROCEDURE

The enthalpy of the solution in the liquide phase can be regarded as the sum of the weighted contributions of the two components ,as a function of the temperature and

concentration and can be calculated from the specific enthalpy of the pure components and the excess specific enthalpy of mixing as follows:

$$h = x h_r + (1-x) h_a + h^E \quad (1)$$

with h_r is the specific enthalpy of the refrigerant, h_a the specific enthalpy of the absorbent, x is the weight fraction of refrigerant in the solution and h^E the excess specific enthalpy of mixing in [kJ/kg].

A. Excess thermodynamic properties :

The excess molar Gibbs energy of mixing g^E of the solution is the point of departure for calculating other excess properties. The excess molar Gibbs energy of mixing is expressed in terms of the molar concentration and the activity coefficient as follows:

$$g^E = RT \sum (x_i \cdot \ln(\gamma_i)) \quad (2)$$

$$S^E = -\frac{\partial g^E}{\partial T} \quad (3)$$

From equation (2) and (3) the specific excess enthalpy h^E may be expressed as:

$$h^E = g^E - T \frac{\partial g^E}{\partial T} \quad (4)$$

$$h^E = R \cdot T \cdot \sum_i x_i \cdot \ln \gamma_i - R \cdot T \cdot \left(\sum_i x_i \cdot \ln \gamma_i + T \cdot \sum_i x_i \cdot \frac{\partial \ln \gamma_i}{\partial T} \right) \quad (5)$$

$$h^E = -RT^2 \sum x_i \frac{\partial \ln \gamma_i}{\partial T} \quad (6)$$

B. Activity coefficient models :

Various models for the calculations of the activity coefficients γ_i are proposed in the literature.

Each one depends on the manner, the expression for the Gibbs energy g^E is chosen.

The UNIQUAC and UNIFAC models were used to evaluate the activity coefficient γ_i in the liquid phase.

C. UNIQUAC (Universal Quasi-chemical Activity coefficient) model :

The coefficient of activity is made of two terms as [5]:

$$\ln \gamma_i = \ln \gamma_i^C + \ln \gamma_i^R \quad (5)$$

The first term is the combinatory and the second is the residual one and are given as follows:

$$\ln \gamma_i^C = \ln \frac{\Phi_i}{x_i} + \frac{Z}{2} q_i \ln \frac{\Theta_i}{\Phi_i} + l_i \frac{\Phi_i}{x_i} \sum_j x_j l_j \quad (6a)$$

$$\ln \gamma_i^R = q_i \left[1 - \ln \left[\sum_j \theta_j \tau_{ji} \right] - \sum_j \frac{\theta_j \tau_{ji}}{\sum_k \theta_k \tau_{kj}} \right] \quad (6b)$$

$$l_i = \frac{z}{2} (r_i - q_i) - (r_i - 1) \quad (6c)$$

Where r_i and q_i are the van der Waals molecular volume and surface parameters, respectively, z is the coordination number set equal to 10.

τ_{ji} is the energy parameter.

D. UNIFAC (UNIQUAC Functional Activity Coefficient) model :

Similarly to the UNIQUAC, the UNIFAC is also made of two terms, combinatorial and residual as shown by Equation 5.

The first term is the combinatorial which is expressed by Equation 6a and the second one is the residual and is expressed as:

$$\ln \gamma_i^R = \sum_k v_k^{(i)} (\ln \Gamma_k - \ln \Gamma_k^{(i)}) \quad (7a)$$

Where Γ_k and $\Gamma_k^{(i)}$ are the residual activity coefficients of group k in the mixture and in pure liquid i , respectively.

$$\ln \Gamma_k = Q_k \left[1 - \ln \left(\sum_m \Theta_m \cdot \Psi_{mk} \right) - \sum_n \frac{\Theta_m \cdot \Psi_{km}}{\sum_n \Theta_n \cdot \Psi_{nm}} \right] \quad (7b)$$

$$\Theta_m = \frac{Q_m \cdot X_m}{\sum_n q_n \cdot x_n} \quad (7c)$$

$$X_m = \frac{\sum_j v_m^{(j)} \cdot x_j}{\sum_j \sum_n v_n^{(j)} \cdot x_j} \quad (7d)$$

The activity coefficient is a function of temperature and is differentiated as follows:

$$\frac{\partial \ln \gamma_i}{\partial T} = \frac{\partial \ln \gamma_i^C}{\partial T} + \frac{\partial \ln \gamma_i^R}{\partial T} \quad (8)$$

$\frac{\partial \ln \gamma_i^C}{\partial T} = 0$ This term doesn't depend on temperature

Thus, we have to differentiate this term $\frac{\partial \ln \gamma_i^R}{\partial T}$.

$$\frac{\partial \ln \gamma_i}{\partial T} = \frac{\partial \ln \gamma_i^R}{\partial T} = \sum_K v_K^{(i)} \left(\frac{\partial \ln \Gamma_k}{\partial T} - \frac{\partial \ln \Gamma_k^{(i)}}{\partial T} \right) \quad (9)$$

$$\frac{\partial \ln \Gamma_k}{\partial T} = Q_k \left[\frac{\sum_m \theta_m \cdot \Psi_k \cdot \frac{a_{mk}}{T^2}}{\sum_m \theta_m \cdot \Psi_{mk}} - \frac{\left(\theta_m \cdot \Psi_{km} \cdot \frac{a_{km}}{T^2} \right) \cdot \left(\sum_n \theta_n \cdot \Psi_{nm} \right) - \left(\theta_m \cdot \Psi_{km} \right) \cdot \left(\sum_n \theta_n \cdot \Psi_{nm} \cdot \frac{a_{nm}}{T^2} \right)}{\sum_m \theta_m \cdot \Psi_{mk}} \right] \quad (10)$$

The derivative of $\left(\frac{\partial \ln \Gamma_k^{(i)}}{\partial T} \right)$ can be obtained similarly to

$$\left(\frac{\partial \ln \Gamma_k}{\partial T} \right)$$

III. EXPERIMENTAL EXCESS ENTHALPY :

Experimental excess enthalpy is expressed as a function of temperature and concentration for the R124-DMAC and R134a-DMETEG systems as a polynomial function expressed as [1,2]:

$$h^E = 4.1868 \sum_{j=0}^7 \sum_{i=0}^7 h_{ij} x^i T^j \quad (11)$$

h^E is expressed in [kJ/ kg] and x is the mole fraction of the refrigerant in liquid phase. The coefficients h_{ij} for R124-DMAC and for R134a-DMETEG are given in [1,2].

To be able to analyse the results and to compare with the experimental data, the absolute average deviation of the results is calculated. The absolute average deviation is calculated using equation 12

$$AAD(\%) = \frac{100}{npts} \sum_{i=1}^{npts} \left| \frac{h_i^{exp} - h_i^{calc}}{h_i^{exp}} \right| \quad (12)$$

IV. Optimization of interaction parameters :

The UNIFAC model was used for the determination of the activity coefficients of the constituents R124 (2-chlor-1.1.1.2-tetra-fluoroethane) and DMAC (N'-N'-dimethylacetamide), and the UNIQUAC model for the two considered working pairs (R124-DMAC, R134a-DMETEG). For the UNIQUAC model the required interaction parameters are obtained using the experimental data available in the literature [1,2], by minimizing the objective function F defined as follows:

$$F = \sum_1^N (h_{i,exp}^E - h_{i,calc}^E)^2 \quad (13)$$

With h^E the excess enthalpy, the subscripts exp and calc. denoting experimental and calculated, respectively and N the number of experimental data points.

The adopted minimization technique was based on the Nelder-Mead version Simplex method which is extensively described in details in the literature [6].

For the UNIFAC model the group interaction parameters between the involved groups as well as the van der Waals volume and surface area parameters are available in the literature [7, 8] and are as shown in the following Table I. The decomposition into groups of the two considered constituents is as follows:

R124 (2-chloro-1, 1, 1, 2-tetrafluoroethane): 1C, 1CH, 1Cl, 4F

DMAC (N, N'- Dimethylacetamide): 1CH₃, 1 CON(Me)₂

TABLE I GROUP VOLUME SURFACE AND INTERACTION PARAMETERS FOR THE UNIFAC MODEL

m/n	C	CH	CH ₃	Cl	F	CON(Me) ₂
R _k	0.2195	0.45	0.90	0.77	0.377	2.86
Q _k	0	0.29	0.848	0.75	0.44	2.43
C	0	0	0	523	266	729
CH	0	0	0	523	266	729
CH ₃	0	0	0	523	266	729
Cl	-120	-120	-120	0	28	-678.52
F	34	34	34	870	0	156.812
CON(Me) ₂	-11.1	-11.1	-11.1	852.49	631.25	0

V. RESULTS :

The detail results of UNIQUAC molecular interaction parameters of the selected working fluids obtained after minimization of the objective function described by Equation 13 for the considered pairs are shown in the following tables:

TABLE 2 : MOLECULAR INTERACTION PARAMETERS FOR UNIQUAC MODEL FOR R124(1) -DMAC (2) SYSTEM

Temperature (°C)	a12	a21
0	-194.82	-256.46
20	-129.00	-273.00
40	-68.60	-277.00
60	52.77	-311.85
80	78.00	-299.57
100	228.87	-330.59
120	202.44	-307.80
140	228.531	-300.95

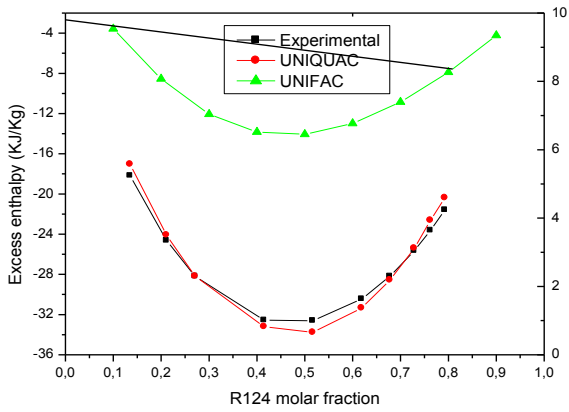
TABLE 3: MOLECULAR INTERACTION PARAMETERS FOR UNIQUAC MODEL FOR R134A (1) -DMETEG (2) SYSTEM

Temperature (°C)	a12	a21
20	-251.125	56.218
40	-205.8	56.218
60	-163.99	57.75
80	-122.488	58.628
100	-96.557	73.117
120	-123.875	181.435
140	-57.328	98.974

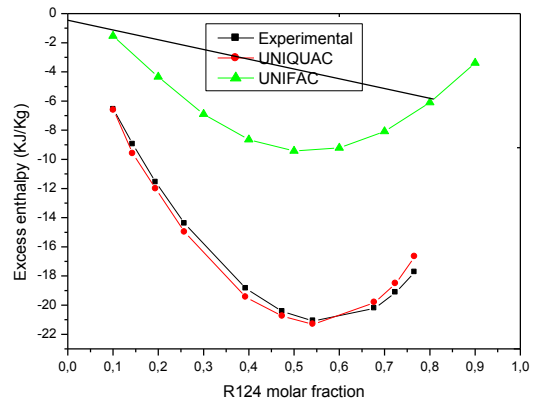
The results of specific excess enthalpy with UNIFAC and UNIQUAC models at several temperatures are presented in figures 1a-h and 2a'-g'. Among the models studied in this research, both UNIFAC and UNIQUAC models show their ability to calculate the activity coefficient and thus specific excess enthalpy of the R124-DMAC with relative accuracy. Although, the calculation results using UNIFAC model were less accurate than those of UNIQUAC model for the R124-DMAC pair. Subsequently, UNIQUAC model is chosen to evaluate the excess enthalpy of the R134a-DMETEG mixture.

Fig1a-h show the excess enthalpy variations with the refrigerant (R124) molar fraction at different temperatures in terms of molar fraction for the R124-DMAC pair considered in [1], the values of which are compared with the results obtained in the present work.

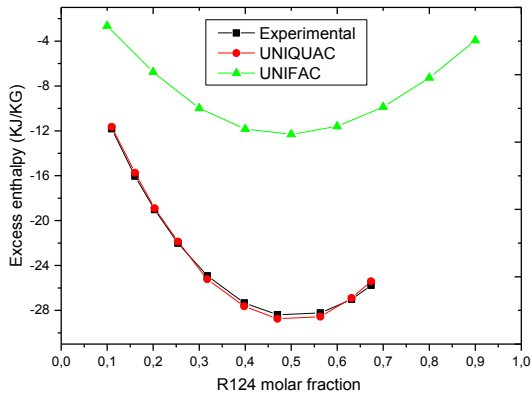
OPTIMIZATION OF UNIQUAC INTERACTION PARAMETERS FOR ABSORPTION REFRIGERATION MACHINE WORKING FLUIDS.



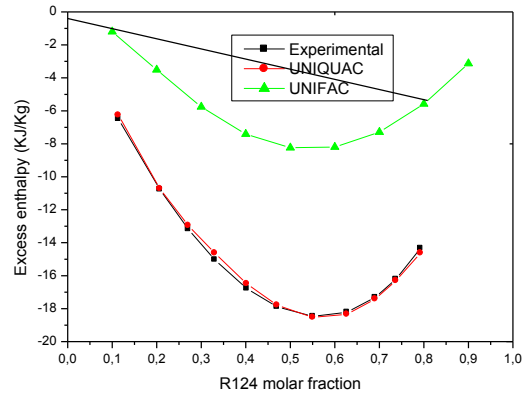
(a)



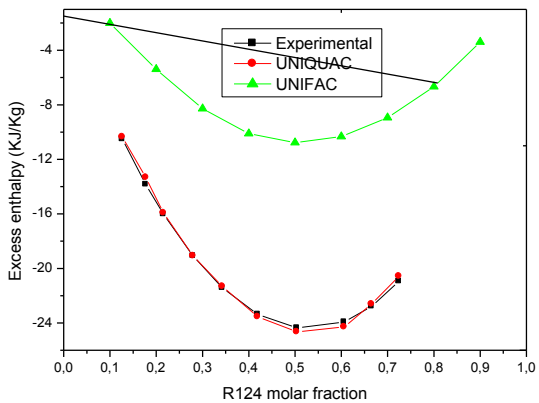
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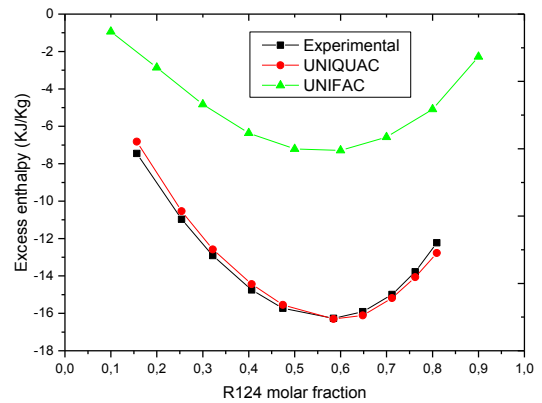
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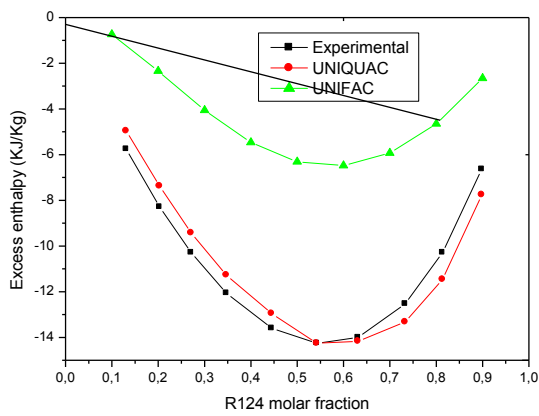
(e)



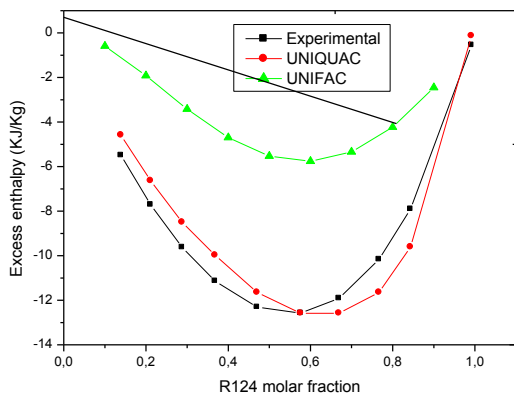
(c)



(f)



(g)



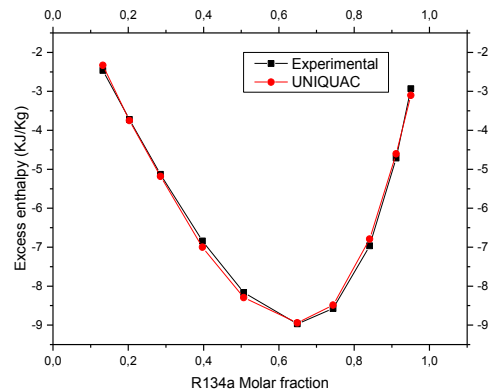
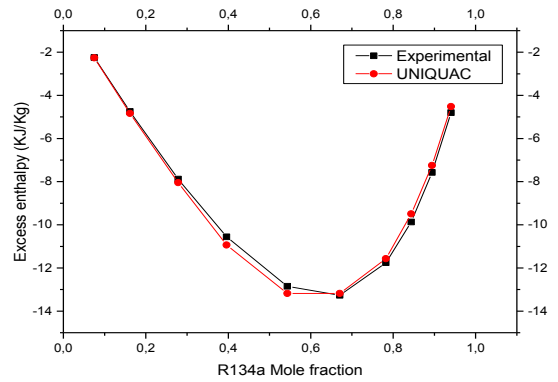
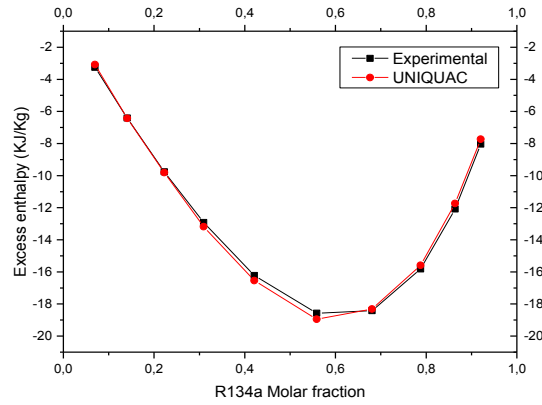
(h)

Fig1. Comparison of experimental, UNIFAC and UNIFAC specific excess enthalpies at: a) 0°C; b) 20°C; c) 40°C; d) 60°C; e) 80°C; f) 100°C; g) 1) 20°C; h)140°C.

Similar with those of R124 and DMAC mixture Fig2a'-h' show the excess enthalpy variations with the refrigerant (R134a) molar fraction at different temperatures in terms of molar fraction for the R134a-DMETEG pair considered in [2], the values of which are compared with the results obtained in the present work.

From the calculated excess enthalpy for the investigated working mixtures at temperature varying from 20°C to 140°C for R134a-DMETEG, It is interesting to see at the figure 2 a',g' that the results of the specific excess enthalpy have good agreement with the experimental data.

It is also interesting to observe that at higher temperature beyond 100°C the excess enthalpy was positive which means that the mixing process is endothermic. On contrary, at lower temperature below 100°C, the excess enthalpy was negative which means that the mixing process is exothermic.



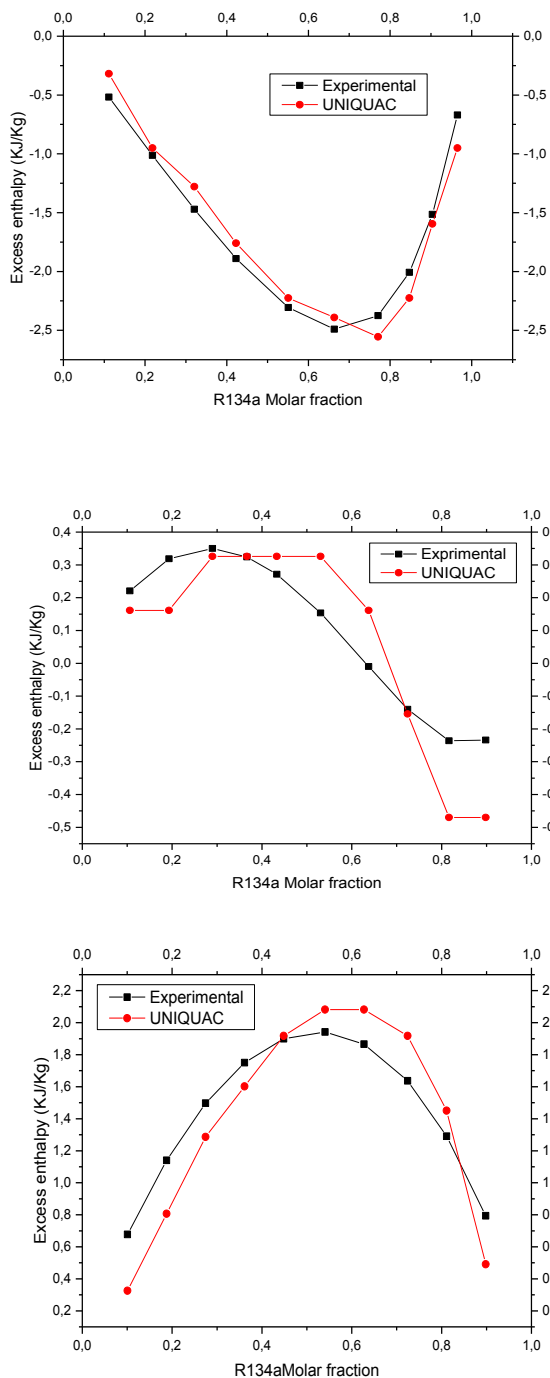


Fig2. Comparison of experimental, and UNIQUAC specific excess enthalpies at: a') 20°C; b') 40°C; c') 60°C; d') 80°C; e') 100°C; f') 120°C; g') 140°C.

From the previous figures, it can be seen that qualitatively both the UNIQUAC and the UNIFAC models led to similar shapes of the corresponding curves of h^E versus the refrigerant molar fraction for R124-DMAC binary system.

For R124-DMAC and for R134a-DMETEG systems, UNIQUAC calculations reproduce correctly the experimental data although UNIQUAC values give always a better correlation Fig1a-h and Fig2a'-h'. Both models give acceptable results nevertheless UNIQUAC

reproduces better the experimental data and can be used with more confidence to predict the activity coefficients, thus specific excess enthalpy.

VI. CONCLUSION

Through this optimization study it has been clearly shown that the group contribution concept based models like the UNIFAC for the activity coefficients have serious limitations as illustrated by the obtained results where the corresponding curves are far away from those of the experimental and the UNIQUAC which in turn were in an excellent agreement.

To have solid enthalpy-concentration diagrams of the working fluid used in absorption refrigeration systems, it is necessary to have a good correlation of the specific excess enthalpy of the working fluid mixtures using and to choose appropriate thermodynamic model to describe the activity coefficient. They predict more or less depending on the type of the considered systems. Hence, the problem of the choice of the most appropriate thermodynamic model is not an easy task.

Also the main results is that molecular interaction parameters become available for the R124-DMAC and for the R134a-DMETEG systems which are not available. With powerful minimization techniques like Nelder-mead version Simplex method, the obtained results are very reliable and can be used to perform computer experiments at will.

From this analysis, it can be concluded that the use of default binary interaction parameter is not able to calculate the specific excess enthalpy of R124 and DMAC also of R134a and DMETEG mixtures with high accuracy.

Finally through this study one can see that the choice of the most adequate method of optimization is a crucial task upon which will depend the reliability of the results.

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