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Prediction of Excess Molar Volumes for Quaternary Liquid Mixtures at 298.15 K

Hüseyin ARSLAN[*1]

[1]*Kahramanmaras Sütçüimam University, Science Faculty, Physics Department, 46100, Onikisubat / Kahramanmaras, Turkey*

**corresponding author e-mail: arslan@ksu.edu.tr*

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Abstract: The excess molar volumes for the ternary n-hexanol + ethanenitrile + dichloromethane and quaternary system n-hexanol + ethanenitrile + dichloromethane + tetrahydrofuran at 298.15K has been calculated by using symmetric and asymmetric models. Chou's General solution model has been employed to evaluate the excess molar volume of the ternary n-hexanol + ethanenitrile + dichloromethane and quaternary systems in the n-hexanol + ethanenitrile + dichloromethane + tetrahydrofuran. The good agreement between experimental data and calculated results of excess molar volumes for both mixture systems indicates that this approach can be successfully used to predict the excess molar volumes of multicomponent systems.

***Key words*:** Excess molar volumes, Redlich-Kister polynomial, General solution model, Quaternary systems.

298,15 K de Dörtlü Sıvı Karışımların Fazla Molar Hacimlerinin Tahmini

Özet: 298.15 K'de üçlü n-hekzanol + etanitril + diklorometan ve dörtlü sistem n-hekzanol + etanitril + diklorometan + tetrahidrofuran için fazla molar hacimler simetrik ve asimetrik modeller kullanılarak hesaplanmıştır. Chou'nun Genel çözüm modeli, n-heksanol + etanitril + diklorometan + tetrahidrofuranda üçlü n-hekzanol + etanitril + diklorometan ve dörtlü sistemlerin fazla molar hacmini değerlendirmek için kullanılmıştır. Her iki karışım sistemi için deneysel veriler ve fazla molar hacimlerin hesaplanan sonuçları arasındaki iyi uyum, bu yaklaşımın çok bileşenli sistemlerin fazla molar hacimlerini tahmin etmek için başarıyla kullanılabileceğini gösterir.

Anahtar kelimeler: Fazla molar hacim, Redlich-Kister poinomu, Genel çözüm modeli, Dörtlü sistem.

1. Introduction

From a scientific and technical point of view, multicomponent structures are of interest. It is well known that it is costly and time consuming to experimentally evaluate the excess functions of multicomponent structures. In addition, with the number of materials, the experimental problems increase dramatically. As a result, several papers on the prediction of physicochemical properties such as molar volume, molar enthalpy, density, viscosity and refractive indices have been published in recent decades [1-9]. In this paper, the excess molar volume of quaternary system n-hexanol + ethanenitrile +

dichloromethane + tetrahydrofuran at 298.15 K is determinate by using Toop model [10], Hillert model [11], Kohler model [12], Muggianu model [13] and Chou model [14, 15]. The geometric models have been used in quaternary system based on the binary information. Multicomponent system properties can be represented as a mixture of all binary system properties. For different selections of binary systems can be used symmetric (Kohler and Muggianu) and asymmetric (Toop and Hillert) models. The current models have unsuitable assumed that the selected binary system is independent of multicomponent system itself. That is causes inherent defects of both and asymmetric models. In order to overcome of this problem was proposed General Solution Model (GSM) by Chou. GSM can overcome the defects of a symmetric and asymmetric model through the introduction of a parameter "similarity coefficient". If two of the three components are similar, the new geometric model can be converted to a binary model. However, conventional geometric models do not do this. Thus, GSM has been successfully applied by many researchers for industrial topics including thermophysical and thermochemical property calculations [16-22]. In this study, we are reformulated Chou's model in terms of the Redlich-Kister (R-K) polynomial [23], which will give an accurate result and simplify the quaternary property calculations. The excess molar volume was calculated from GSM. The results are also compared with experimental data [24] and traditional models, such as Toop, Hillert, Kohler and Muggianu model.

2. Calculation Models

Redlich-Kister polynomials [23] can express the excess molar volume binary multicomponent system sub-systems as follows:

$$V^{ex} = x_i x_j \sum_{k=0}^{n} L_{ij}^{k} \left(x_i - x_j\right)^k \tag{1}$$

The parameters of Redlich-Kister for the binary system i-j, which is independent of the variable, are represented by L_{ij}^{k}. xi and xj, respectively, are the mole fractions of the components i and j in the binary scheme i-j. Calculation of excess molar volumes for the quaternary system have been done using different prediction methods, such as Toop [10], Hillert [11], Kohler [12], and Muggianu [13] at 298.15 K. The fundamental equations of these methods of prediction for the quaternary structure, the following are given:

Toop model:

$$\begin{aligned} V^{ex} = {} & \frac{x_2}{1-x_1} V_{12}^{ex}(x_1; 1-x_1) + \frac{x_3}{1-x_1} V_{13}^{ex}(x_1; 1-x_1) + \frac{x_4}{1-x_1} V_{14}^{ex}(x_1; 1-x_1) \\ & + (x_2 + x_3)^2 V_{23}^{ex}\left(\frac{x_2}{x_2+x_3}; \frac{x_3}{x_2+x_3}\right) + (x_2 + x_4)^2 V_{24}^{ex}\left(\frac{x_2}{x_2+x_4}; \frac{x_4}{x_2+x_4}\right) \\ & + (x_3 + x_4)^2 V_{34}^{ex}\left(\frac{x_3}{x_3+x_4}; \frac{x_4}{x_3+x_4}\right) \end{aligned} \tag{2}$$

Hillert model:

$$\begin{aligned} V^{ex} = {} & \frac{x_2}{1-x_1} V_{12}^{ex}(x_1; 1-x_1) + \frac{x_3}{1-x_1} V_{13}^{ex}(x_1; 1-x_1) + \frac{x_4}{1-x_1} V_{14}^{ex}(x_1; 1-x_1) \\ & + \frac{x_2 x_3}{\nu_{23}\nu_{32}} V_{23}^{ex}(\nu_{23}; \nu_{32}) + \frac{x_2 x_4}{\nu_{24}\nu_{42}} V_{24}^{ex}(\nu_{24}; \nu_{42}) + \frac{x_3 x_4}{\nu_{34}\nu_{43}} V_{34}^{ex}(\nu_{34}; \nu_{43}) \end{aligned} \tag{3}$$

where is: $v_{ij} = \frac{1+x_i-x_j}{2}$

Kohler model:

$$V^{ex} = (x_1 + x_2)^2 V_{12}^{ex}\left(\frac{x_1}{x_1+x_2};\frac{x_2}{x_1+x_2}\right) + (x_1 + x_3)^2 V_{13}^{ex}\left(\frac{x_1}{x_1+x_3};\frac{x_3}{x_1+x_3}\right)$$
$$+(x_1 + x_4)^2 V_{14}^{ex}\left(\frac{x_1}{x_1+x_4};\frac{x_4}{x_1+x_4}\right) + (x_2 + x_3)^2 V_{23}^{ex}\left(\frac{x_2}{x_2+x_3};\frac{x_3}{x_2+x_3}\right)$$
$$+(x_2 + x_4)^2 V_{24}^{ex}\left(\frac{x_2}{x_2+x_4};\frac{x_4}{x_2+x_4}\right) + (x_3 + x_4)^2 V_{34}^{ex}\left(\frac{x_3}{x_3+x_4};\frac{x_4}{x_3+x_4}\right) \quad (4)$$

Muggianu model:

$$V^{ex} = \frac{4x_1x_2}{(1+x_1-x_2)(1+x_2-x_1)} V_{12}^{ex}\left(\frac{1+x_1-x_2}{2};\frac{1+x_2-x_1}{2}\right) + \frac{4x_1x_3}{(1+x_1-x_3)(1+x_3-x_1)} V_{13}^{ex}\left(\frac{1+x_1-x_3}{2};\frac{1+x_3-x_1}{2}\right)$$

$$+ \frac{4x_1x_4}{(1+x_1-x_4)(1+x_4-x_1)} V_{14}^{ex}\left(\frac{1+x_1-x_4}{2};\frac{1+x_4-x_1}{2}\right) +$$
$$\frac{4x_2x_3}{(1+x_2-x_3)(1+x_3-x_2)} V_{13}^{ex}\left(\frac{1+x_2-x_3}{2};\frac{1+x_3-x_2}{2}\right)$$

$$+ \frac{4x_2x_4}{(1+x_2-x_4)(1+x_4-x_2)} V_{24}^{ex}\left(\frac{1+x_2-x_4}{2};\frac{1+x_4-x_2}{2}\right) +$$
$$\frac{4x_3x_4}{(1+x_3-x_4)(1+x_4-x_3)} V_{34}^{ex}\left(\frac{1+x_3-x_4}{2};\frac{1+x_4-x_3}{2}\right) \quad (5)$$

In all equations V^{ex} correspond to the excess molar volume for quaternary system, although x_1, x_2, x_3 and x_4 in the evaluated method correspond to the mole fraction of the components.

Basic theoretical explanations are given for the general solution model [14]:

$$V^{ex} = \sum_{i,j=1\ i\neq j}^{n} W_{ij} V_{ij}^{ex} \quad (6)$$

The molar volume property for an *ij* binary system is V_{ij}^{ex}, and the probability weight of the *ij* binary is W_{ij}. It is possible to express the probability weight of the IJ binary system as follows:

$$W_{ij} = \frac{x_i x_j}{X_{i(ij)} X_{j(ij)}}, \begin{bmatrix} i,j = 1 \text{ to } 4 \\ i \neq j \end{bmatrix} \quad (7)$$

in which the quaternary compositions are represented by x_i and x_j, and $X_{i(ij)}$ and $X_{j(ij)}$ represent the chosen binary compositions that are different for different types of geometric models. When extending the excess molar volume to a quaternary scheme, it can be expressed as:

$$V^{ex} = W_{12}V_{12}^{ex} + W_{13}V_{13}^{ex} + W_{14}V_{14}^{ex} + W_{23}V_{23}^{ex} + W_{24}V_{24}^{ex} + W_{34}V_{34}^{ex} \quad (8)$$

$X_{i(ij)}$ and $X_{j(ij)}$ in Eq.(7) the following can be determined in terms of:

$$X_{1(12)} = x_1 + x_3.\xi^{(3)}_{1(12)} + x_4.\xi^{(4)}_{1(12)}; \qquad X_{1(13)} = x_1 + x_2.\xi^{(2)}_{1(13)} + x_4.\xi^{(4)}_{1(13)}$$
$$X_{1(14)} = x_1 + x_2.\xi^{(2)}_{1(14)} + x_3.\xi^{(3)}_{1(14)}; \qquad X_{2(23)} = x_2 + x_1.\xi^{(1)}_{2(23)} + x_4.\xi^{(4)}_{2(23)}$$
$$X_{2(24)} = x_2 + x_1.\xi^{(1)}_{2(24)} + x_3.\xi^{(3)}_{2(24)}; \qquad X_{3(34)} = x_3 + x_1.\xi^{(1)}_{3(34)} + x_2.\xi^{(2)}_{3(34)} \quad (9)$$

In Eq. (9), ξ is the similarity coefficient which is defined as:

$$\xi^{(k)}_{i(ij)} = \frac{\eta(ij,ik)}{\eta(ij,ik)+\eta(ji,jk)} \quad (10)$$

Here $\eta(ij,ik)$ is called the "deviation sum of squares" and can be calculated as follows:

$$\eta(ij,ik) = \int_0^1 \left(V^{ex}_{ij} - V^{ex}_{jk}\right)^2 dX_i \quad (11)$$

Within Eq. (11), assessing the integration makes the measurement a little awkward. If the relations between the coefficient of similarity and the Redlich-Kister polynomial parameters are known, the algebraic equation can be carried out. A general relationship that enables us to go to any quaternary structure has recently been derived [16]:

$$\eta(ij,ik) = \sum_{l=0}^{n} \frac{1}{2(2l+1)(2l+3)(2l+5)} \left(L^l_{ij} - L^l_{ik}\right)^2 + \sum_{l=0}^{n}\sum_{m>l}^{n} \frac{1}{(l+m+1)(l+m+3)(l+m+5)} \times \left(L^l_{ij} - L^l_{ik}\right)\left(L^m_{ij} - L^m_{ik}\right) \quad (12)$$

Substituting Eq. (12) into Eq. (10), it is possible to define coefficients of similarity. If *n* Eqs. (11-12) is equal to 4, and then it is possible to express "deviation sum of squares" We should write for the η(12,13) as follows:

$$\eta(12,13) = \frac{1}{30}(L^0_{12} - L^0_{13})^2 + \frac{1}{210}(L^1_{12} - L^1_{13})^2 + \frac{1}{630} + \frac{1}{1386}(L^3_{12} - L^3_{13})^2 + \frac{1}{2574}(L^4_{12} - L^4_{13})^2 + \frac{1}{105}(L^0_{12} - L^0_{13})(L^2_{12} - L^2_{13}) + \frac{1}{315}(L^0_{12} - L^0_{13})(L^4_{12} - L^4_{13}) + \frac{1}{315}(L^1_{12} - L^1_{13})(L^3_{12} - L^3_{13}) + \frac{1}{693}(L^2_{12} - L^2_{13})(L^4_{12} - L^4_{13}) \quad (13)$$

Similar $\eta(ij,ik)$ values are also calculated from Eq. (12).

3. Results and Discussions

The excess molar volume on mixture systems is one of the most interesting properties of mixing functions since exploring the behavior of liquid systems is a good thermodynamic method. It is also known that a sensitive indicator to accuracy of liquid mixture theories. The positive excess molar volume indicates volume expansion on mixing as seen a maximum at equicomposition = 0.5 in the binary HE(1)+EN(2) and HE(1)+DCM(3) and thus repulsive interaction of mixing alloys or weaker interactions than the interactions of the pure components. The negative excess amount, on the other hand, indicates stronger mixed molecular interactions as seen negative deviations from the linear law and for example minimums at equicomposition = 0.5 in the binary EN(2)+DCM(3) and EN(2)+THF(4) than individual molecules before mixing. Therefore, these results signify that the interaction between EN(2) or THF(4) becomes relatively attractive at the mixture forming composition. Moreover, as shown in Figure

1 the excess molar volumes of the alloy HE(1)+THF(4) and DCM(3)+THF(4) are almost zero at 298.15 K over the entire spectrum of concentration. This situation suggests that the liquid these mixtures can be treated as an ideal mixture and are indicative of an ideal mixture at 298.15 K.

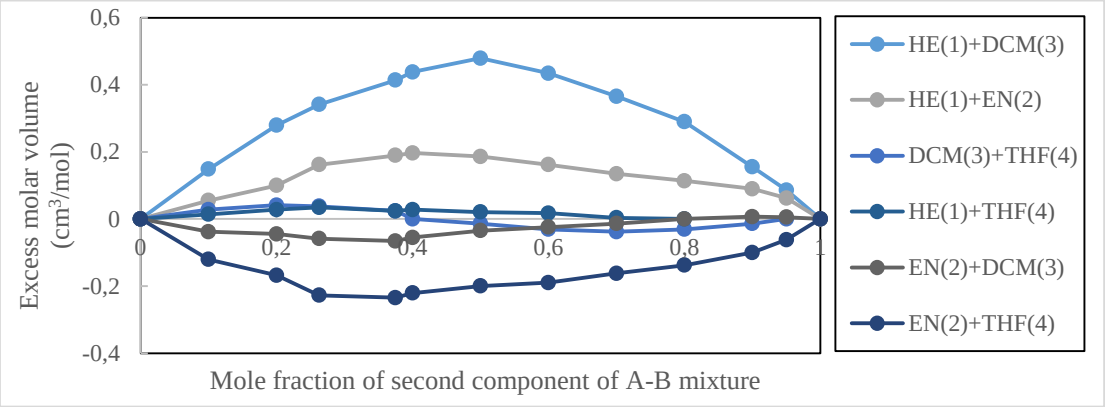

Figure 1. For the three binary systems, experimental values of excess molar volume as a function of n-hexanol molar fraction: symbols apply to the experimental data while the lines reflect the results of the ternary Hexanol + Ethanenitrile + Dichloromethane + tetrahydrofuran quaternary method determined from the Redlich-Kister-Muggianu formalism.

Table 1. Redlich-Kister parameters for n-hexanol (1) + ethanenitrile (2) + dichloromethane (3) + tetrahydrofuran (4) binary system at 298.15 K.

System	L_{ij}^0	L_{ij}^1	L_{ij}^2	L_{ij}^3	L_{ij}^4
HE(1)+EN(2)	0.77419	–0.39025	0.15884	–0.08105	0.22976
HE(1)+DCM(3)	1.93148	–0.10038	–0.58252	0.30740	0.53022
HE(1)+THF(4)	0.11280	–0.09130	0.00338	0.04194	–0.02595
EN(2)+DCM(3)	–0.13828	0.32532	–0.14651	0.00268	0.02211
EN(2)+THF(4)	–0.84025	0.63122	–0.74114	–0.07811	0.57113
DCM(3)+THF(4)	0.05575	–0.40898	–0.21286	0.04548	0.14401

A quaternary system contains six binary system, and the value before using the binary form, all of these binary systems should be understood symmetric and asymmetric models. Excess molar volumes of the quaternary liquid mixtures n-hexanol + ethanenitrile + dichloromethane + tetrahydrofuran have been measured at 298.15 K [24] and [25]. The R-K form polynomial was used to measure all data for six binary systems and the R-K parameter values for the constituent binary systems given in Table 1 [24] were used for the calculation.

The values of excess molar volume in terms of $cm^3\ mol^{-1}$ for different mixtures of four component of quaternary system have been calculated using Toop, Hillert, Kohler, Muggianu and Chou model and Eqs. (1-13). x_1, x_2, x_3 and x_4 in Table 1 represent mole fraction of n-hexanol, ethanenitrile, dichloromethane and tetrahydrofuran, respectively. For comparison, the results of asymmetric model (Toop, Hillert) and symmetric model (Kohler, Muggianu) as well as the experimental data are also included.

In the ternary n-hexanol + ethanenitrile + dichloromethane and quaternary system n-hexanol + ethanenitrile + dichloromethane + tetrahydrofuran at 298.15 K, the excess molar volumes of mixing are shown in Figures 2, 3 and Figures 4-7 for comparison, respectively. From the Figures 2 and 3, the molar volume values of the ternary n-hexanol + ethanenitrile + dichloromethane for the sections He/En=1.49049 and 4.089

show nonlinear dependence strongly at 298.15 K for all the compositions in the ranges of 0-0.6 and 0.8. In addition, it can be seen from Figures 4-7 that the molar volume values of the quaternary system n-hexanol + ethanenitrile + dichloromethane + tetrahydrofuran at 298.15 K show also nonlinear dependence strongly in the ranges of the related sections He/En and DCM+THF.

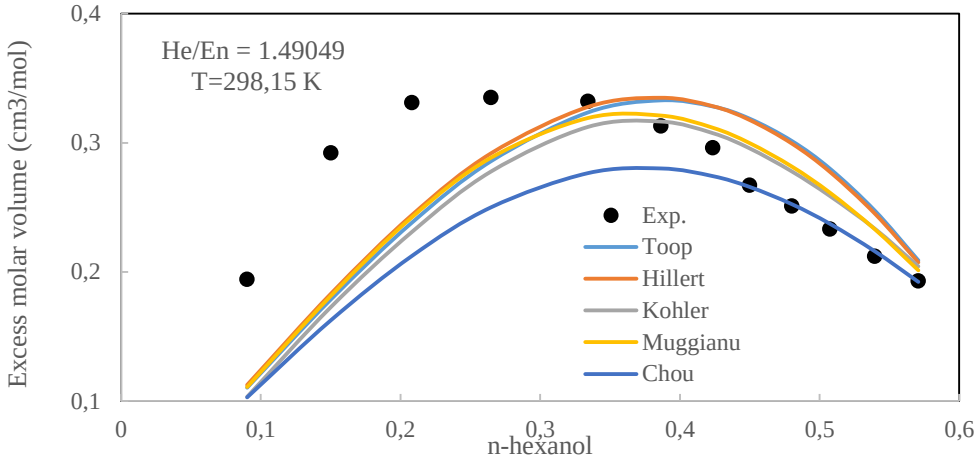

Figure 2. Comparison of the traditional models, Chou model and experimental data associated with the excess molar volume for the section x_1/x_2 = 1.49049 in the ternary Hexanol + Ethanenitrile + Dichloromethane.

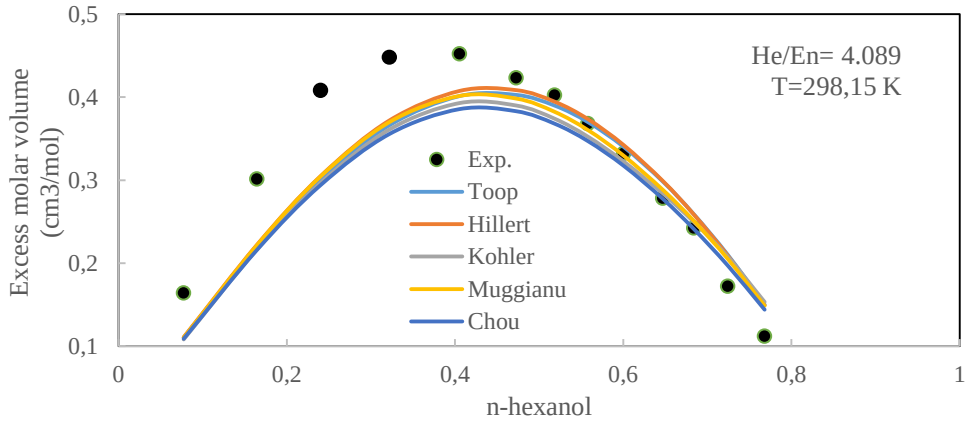

Figure 3. Comparison of the traditional models, Chou model and experimental data associated with the excess molar volume for the section x_1/x_2 = 4.089 in the ternary Hexanol + Ethanenitrile + Dichloromethane.

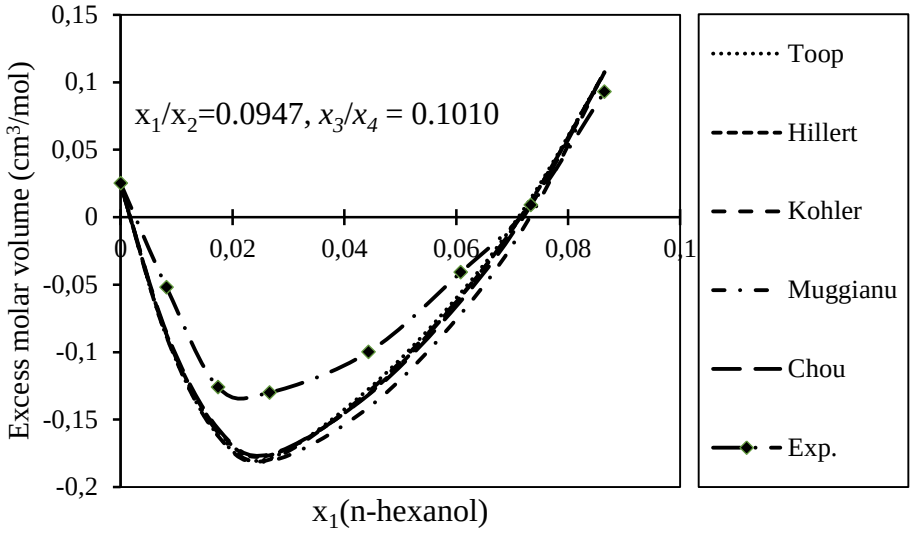

Figure 4. Comparison of the traditional models, Chou model and experimental data of excess molar volume for the section x_1/x_2 = 0.0947; x_3/x_4 = 0.1010 in the quaternary Hexanol + Ethanenitrile + Dichloromethane + tetrahydrofuran quaternary system.

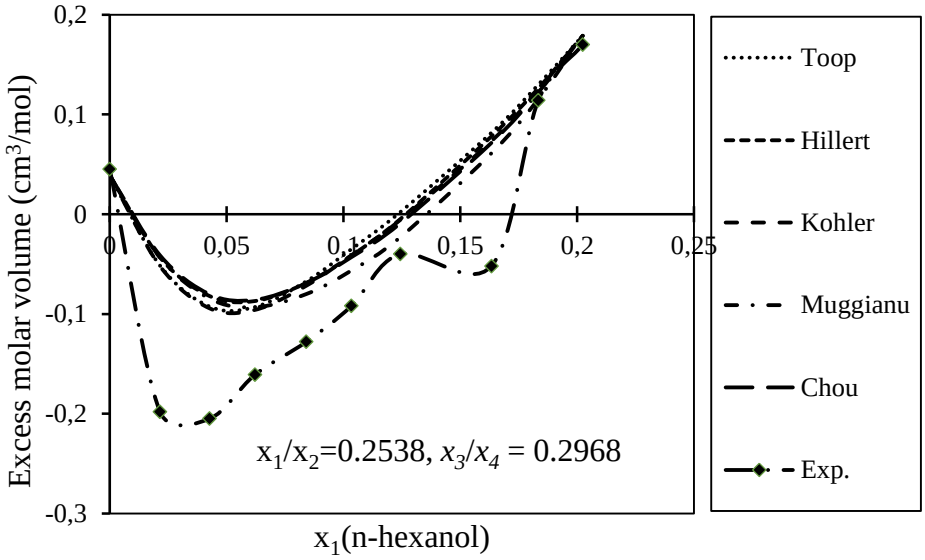

Figure 5. Comparison of the traditional models, Chou model and experimental data of excess molar volume for the section x_1/x_2 = 0.2538; x_3/x_4 = 0.2968 in the quaternary Hexanol + Ethanenitrile + Dichloromethane + tetrahydrofuran quaternary system.

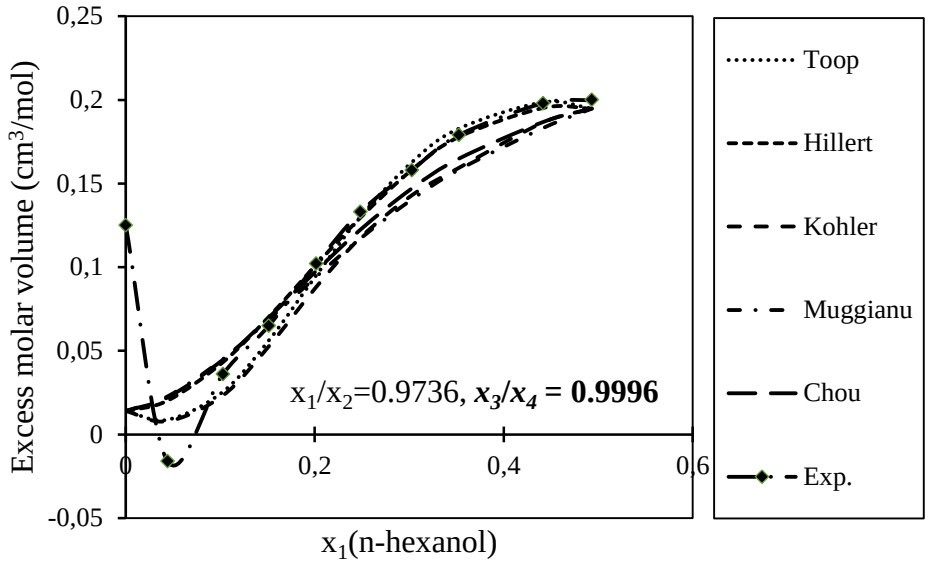

Figure 6. Comparison of the traditional models, Chou model and experimental data of excess molar volume for the section x_1/x_2 = 0.9736; x_3/x_4 = 0.9996 in the quaternary Hexanol + Ethanenitrile + Dichloromethane + tetrahydrofuran quaternary system.

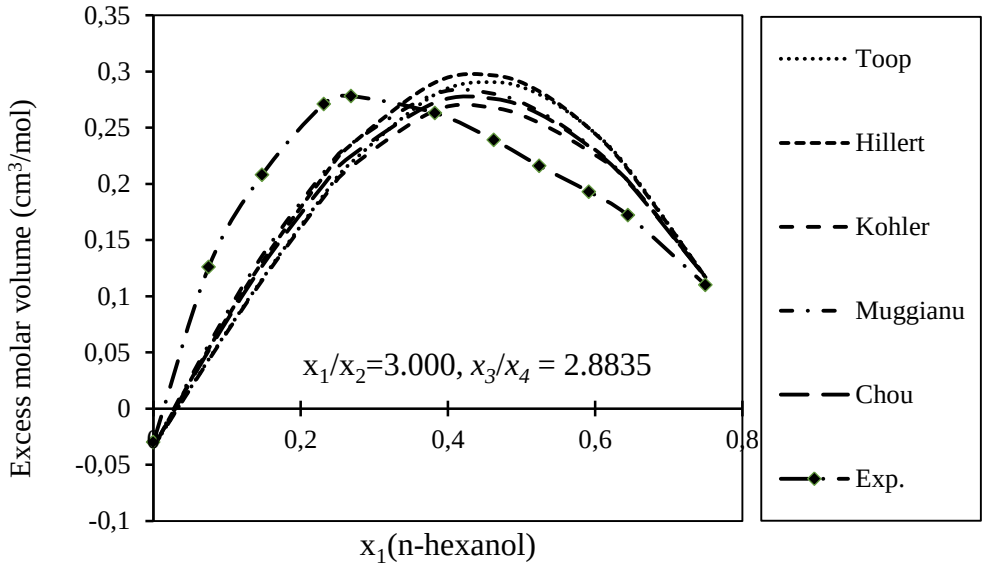

Figure 7. Comparison of the traditional models, Chou model and experimental data of excess molar volume for x_1/x_2 = 3.0000; x_3/x_4 = 2.8835 in the quaternary Hexanol + Ethanenitrile + Dichloromethane + tetrahydrofuran quaternary system.

All models treated in the present study lead to a little deviation from the experimental values due to boundary between symmetric and asymmetric models. In order to accurately the deviation between used models and experimental data [24], the mean deviation (*MD*) analysis was applied to excess molar volumes data:

$$MD = \frac{1}{n}\sqrt{\sum_{i=1}^{n}\left(V_{exp,i}^{ex} - V_{cal,i}^{ex}\right)^2} \qquad (14)$$

in which, n the number of counting points, V_{exp}^{ex} experimental data results, V_{cal}^{ex} calculated values for excess molar volume. The results of *MD* analysis are presented in Table 2.

Table 2. In the current study for Hexanol + Ethanenitrile + Dichloromethane + Tetrahydrofuran quaternary method, the measured mean deviation corresponds to the experimental results for each model.

Sections		**Mean Deviation Values (MD)**					
He/En with Exp.	**Dcm/Thf**	**Toop**	**Hillert**	**Kohler**	**Muggianu**	**Chou**	**Model Agreeing**
0.0947	0.1010	0.00983	0.00960	0.01008	0.01109	0.00954	Chou
3.0000	2.8835	0.0256	0.0263	0.0246	0.02450	0.02610	Muggianu
0.9736	0.9996	0.01151	0.01173	0.01200	0.01233	0.01203	Toop
0.2538	0.2968	0.01759	0.01592	0.0160	0.01365	0.01472	Muggianu
1.49049	-	0.01694	0.01618	0.01647	0.01513	0.01883	Muggianu
4.089	-	0.01429	0.01475	0.01519	0.01382	0.01563	Muggianu

It can be seen results of the *MD* analysis in Table 2 that a good agreement has been demonstrated between the predictions of the models and the experimental data. Moreover, Chou's model also agrees with the experimental results while a good agreement is obtained mutually one another the models treated in the ternary and quaternary mixtures. When a comparison was done with experimental results for the section, He/En = 0.0947, Dcm/Thf= 0.1010 in the quaternary system, it is seen that a reasonable agreement with Chou's model. Chou's model appears to be valid and could be successfully applied for the prediction of these types of ternary systems. It can be seen for other quaternary systems that a reasonable agreement with Toop and Muggianu models for the sections He/En= 0.9736, Dcm/Thf =0.9996, He/En 3.0000, Dcm/Thf =2.8835 and He/En=0.2538, Dcm/Thf =0.2968, respectively. It is observed from Table 2 that there are the agreements with the experimental results for both sections in the quaternary mixtures. Muggianu models also appear to be valid and could be successfully applied for the prediction of these types of ternary systems.

4. Conclusion

In this work, the ternary mixtures are estimated to calculate the excess molar volumes of the mixtures n-hexanol + ethanenitrile + dichloromethane and quaternary system n-hexanol + ethanenitrile + dichloromethane + tetrahydrofuran at 298.15 K associated with ternary and quaternary systems, the Chou's model was extended to ternary and quaternary systems using binary mixture properties. According to the ternary systems, the only difference was including the extra binary terms in both the excess properties. The benefit of the current model is that human intervention in defining the asymmetrical variable is not necessary. In addition, one of the important contributions to Chou's model that the high order framework can be easily expanded, such as quaternary system readily the inclusion of the comparable extra words in the excess molar volume formulae associated with ternary system. Moreover, the analysis of mean deviation has been carried out and in line with this analysis: the results obtained have displayed reliability of the Chou's model for the section He/En=0.0947, Dcm/Thf=0.1010 in the quaternary system, except for the other sections. It is concluded that Muggianu model

for ternary mixture system appears to be valid and could be successfully applied for the prediction of these types of ternary systems for the two sections, He/En=3.0000, Dcm/Thf =2.8835 and He/En =0.2538, Dcm/Thf =0.2968.

Author Statement

Hüseyin Arslan: Investigation, Original Draft Writing, Review and Editing., Investigation, Review and Editing, Supervision, Observation, Advice.

Conflict of Interest

As an only one author of this study, we declare that we do not have any conflict of interest statement.

Ethics Committee Approval and Informed Consent

As an authors of this study, I declare that I do not have any ethics committee approval and/or informed consent statement.

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