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Composition and Temperature Dependence of Excess Volume of Heavy Oil-Stocks Mixtures + (Gas oil or Toluene or Reformate)

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Abstract

Binary mixtures of three, heavy oil-stocks was subjected to density measurements at temperatures of 30, 35 and 40 °C. and precise data was acquired on the volumetric behavior of these systems. The results are reported in terms of equations for excess specific volumes of mixtures. The heavy oil-stocks used were of good varity, namely 40 stock, 60 stock, and 150 stock. The lightest one is 40 stock with °API gravity 33.69 while 60 stock is a middle type and 150 stock is a heavy one, with °API gravity 27.74 and 23.79 respectively. Temperatures in the range of 30-40 °C have a minor effect on excess volume of heavy oil-stock binary mixture thus, insignificant expansion or shrinkage is observed by increasing the temperature this effect becomes more significant although the heavy oil-stocks is spiked with hydrocarbons like (gas oil, toluene and reformate). Blending of Heavy oil-stocks with hydrocarbons spikes (gas oil, toluene and reformate) form non-ideal mixtures, for which excess volume can be positive or negative depending on nature species. Spiking of Heavy oil-stocks with either gas oil or reformate resulted in negative excess volume. This shrinkage is greater for the lowest boiling point spike as in the case of reformate, While, the presence of methyl groups in aromatic rings results in a positive excess volume, as shown in toluene when blended with 40 stock but a negative excess volume was found when blended with 60 stock and 150 stock. The API gravity of heavy oil-stocks has an effect on excess volume when the oil-stocks spiked with hydrocarbons like (gas oil, toluene and reformate). This 40 stocks as a typical light types resulted in minimum negative excess volume of -0.47 at 30 °C, when it was spiked with the gas oil; while the spiked heavy oil-stock with kerosene shows a maximum excess volume of -15.56 at 40 °C.

Keywords: *Excess volume, heavy oil-stocks, temperature dependence.*

1. Introduction

When two liquids are mixed together, the resulting changes in physical and thermodynamic properties can be considered as a sum of several contributions due to free volume change, change in energy, change in molecular orientations and steric hindrances. The mixing of different compounds gives rise to solutions that generally do not behave as ideal solutions. A solution which obeys Raoult's and Dalton's laws is said to be an ideal solution. In terms of interaction if unlike molecules interaction are equal with like molecules interaction, the solution is said to be an ideal solution. The deviation from ideality is expressed by many thermodynamic variables particularly by excess or residual extensive properties. Excess thermodynamic properties of mixtures are useful in the study of molecular interactions and arrangements [1].

Thermodynamic studies provide the additional advantage of using the analysis of microscale interactions for understanding the macroscale behavior of gases and liquids upon mixing. Through the last decade, a considerable effort has been developed in the field of thermodynamic properties, although a scarcity of published data is still encountered for the mixtures. Such properties are strongly dependent on the hydrogen-bond strength of hydroxyl groups, chain length, isomeric structures, and molecular interactions [2].

Excess thermodynamic properties of mixtures provide sight into the molecular interactions between the various components and can be used for the development of molecular models describing the thermodynamic behavior of mixtures. Recently, a considerable upsurge in the theoretical and experimental investigations of the excess thermodynamic properties of binary liquid mixtures has been done [3].

Blend of petroleum components having different physical properties, endure a change in excess volumes since their constitutes do not form ideal solutions. In an ideal solution, the total solution volume is equal to the sum of the volumes of the components. For a solution to approach ideality, the molecules of the materials blended together must be similar in size, shape, and properties. If the nature of the components differs appreciably, then deviation from ideal behavior may be expected. This deviation may be either positive or negative; that is, the total volume may be smaller or larger than the volumes of its components [4]. The blending of oil stocks results in volume changes, caused by the non-ideal behavior of oil systems as compared with the calculated ideal volume. Since the oil industry uses volume measurement in its balances, the apparent discrepancies in material may cause financial complications, which in some cases have led to litigation [4].

The excess volume behavior of heavy oil-stocks mixtures is important. Since only small amount of database was published, especially on mixtures of heavy oil-stocks with pure hydrocarbons; while no or little studies were published on mixtures of different types of heavy oil-stocks [4].

A full understanding of thermodynamic and transport properties of binary liquid systems is essential in many chemical engineering processes such as design calculation, heat transfer, mass transfer, fluid flow, and so forth [5]. We started a research program on the excess properties of mixtures containing (heavy oil-stocks + spikes). In this work we reported experimental data such as density over the whole range of composition for binary liquid mixtures.

Density is temperature-dependent; in most cases a fluid has less dense as the temperature rises [6]. The variation of density with temperature is a property of great technical importance, since most petroleum products are sold by volume and specific gravity which are usually determined at the prevailing temperatures rather than at the standard 15.6 °C [7].

The present work is aimed to evaluate the volumetric behavior of blends of the heavy oil-stocks at temperature range of 30-40 °C. The work is also extended to study the effect of hydrocarbons spikes such as (gas oil, toluene and reformate) on the excess volume of these mixtures, also in Temperature range of 30-40 °C.

2. Experimental Work

Three heavy oil-stocks were obtained from al-Durra Refinery, namely 40 stock, 60 stock, and 150 stock. The lightest one is 40 stock with °API gravity 33.69 while 60 stock is middle type and 150 stock the heaviest of °API gravity 27.74 and 23.79 respectively. The main properties of oil stocks (40 stock, 60 stock, and 150 stock) were measured in al-Durra Refinery laboratories according to API and ASTM specification, as listed in Table 1.

Table 1,
Properties of Oil Stocks.

Specification	40 stock	60 stock	150 stock
Kin. Viscosity at 40 °C, cSt	18.11	56.12	501.98
Kin. Viscosity at 100 °C, cSt	3.14	7.69	33.38
Viscosity index (VI)	95	95	93
Specific gravity at 15.6 °C	0.85	0.88	0.90
°API gravity	33.69	27.74	23.79
COC Flash,°C	n.d.	200-300	290-300
P.M. Flash,°C	160	n.d.	n.d.
Pour point,°C	-24	-6	-3
Sulfur content, wt.%	0.62	1.40	2.00

Gas oil was used as petroleum fraction to study the excess volume phenomena of heavy oil-stocks. This fraction was supplied by al-Duora Refinery, while the toluene was supplied from MERCK Company, Germany. Reformate, supplied from Al- Duora Refinery, was used to study the excess volume phenomena of heavy oil-stocks. Its general properties are about 58 API gravity, initial boiling point 40ºC, end point 200ºC, an octane number of about 91.

The method of mixing process occurred by electrical mixer at room temperature (20-25 °C). Density measurements was measured just, after preparing the mixtures to avoid deposit formation or vaporizing the light ends. All density measurements are carried out at atmospheric pressure.

The following mixtures were prepared:

- Three heavy oil-stocks binary mixtures, at temperature range of 30 - 40 °C.
- Binary mixtures of heavy oil-stocks with spikes (gas oil, toluene and reformate), at temperature range of 30 - 40 °C.

Fig.1. Water Bath.

Density determination of heavy oil-stocks, spikes and their blends was carried out using Pyknometer with a size of 50 cm^3 according to the standard methods (IP 190) [8].

The Pyknometer was placed in a water bath type (Julabo F25) as shown in Fig. (1), which was capable of maintaining the temperature within an accuracy of ± 0.1 °C. Pyknometer and the stopper end were thoroughly cleaned using gasoline and acetone. To eliminate all traces of moisture Pyknometer and stopper capillary are dried using hot air. Wiping the outside of Pyknometer and stopper with a clean, lint-free cloth normally.

3. Results and Discussion

The excess thermodynamic properties such as excess volumes of mixtures of heavy oil-stocks are considerably interested in the field of transportation. Normally light oil-stock blendes with medium and heavy oil-stock were used to satisfy such specification for lubricating oil and another uses. Although these blends would lend to considerable change into thermodynamic behavior of these mixtures, but it directly led to loss in volume of these mixtures [4].

Measurement densities at temperatures range of 30-40 °C for individual oil-stocks are listed in Table (2).

Table 2,
Densities for Individual Oil Stocks.

Oil-Stocks	Density in g/cm^3	Temperature 30 °C	35 °C	40 °C
Stock 40	ρ_{exp}	0.8566	0.8544	0.8524
Stock 60	ρ_{exp}	0.8886	0.8866	0.8826
Stock 150	ρ_{exp}	0.9112	0.9092	0.9072

Three binary mixtures of heavy oil-stocks have been prepared. The volumetric behavior of the binary mixtures of heavy oil-stocks with different densities was evaluated. The ideal volume was calculated by a linear expression in terms of mass fraction of blending component as follows [9].

$$V^{id} = \frac{\rho_2^o + x_2(\rho_1^o - \rho_2^o)}{\rho_1^o \rho_2^o} \left(\frac{cm^3}{kg}\right) \qquad ...(1)$$

Excess volume is defined by the equation:-

$$V^E = V_{mix} - V_{ideal} \qquad ...(2)$$

Where V_{mix} is the actual specific volume, which is equal to $1/\rho_{mix}$ in cm^3/kg and ρ_{mix} is the measured density in kg/m^3.

Excess volumes for binary heavy oil-stocks mixtures are summarized in Tables (3), (4) and (5).

Table 3,
Excess volume of Binary Systems; of Stock 40 with Stock 60 at 30, 35 and 40 °C.

Stock 40 X_2	V^E	Temperature 30 °C	35 °C	40 °C
0.2	V^E_{exp}	-0.7838	-0.9918	-1.2474
0.4	V^E_{exp}	-1.0909	-1.3503	-1.5725
0.5	V^E_{exp}	-1.5105	-1.7161	-2.0788
0.6	V^E_{exp}	-1.1479	-1.357	-1.6335
0.8	V^E_{exp}	-0.7661	-1.0096	-1.1998

Table 4,
Excess Volume of Binary Systems; of Stock 40 with Stock 150 at 30, 35 and 40 °C.

Stock 40 X_2	V^E	Temperature		
		30 °C	35 °C	40 °C
0.2	V^E_{exp}	-2.0589	-2.3717	-3.6435
0.4	V^E_{exp}	-3.3522	-3.8262	-4.4801
0.5	V^E_{exp}	-3.9800	-4.6623	-5.6093
0.6	V^E_{exp}	-3.3812	-3.8554	-4.4510
0.8	V^E_{exp}	-1.7825	-2.2953	-3.7838

Table 5,
Excess Volume of Binary Systems; of Stock 60 with Stock 150 at 30, 35 and 40 °C.

Stock 40 X_2	V^E	Temperature		
		30 °C	35 °C	40 °C
0.2	V^E_{exp}	-0.5140	-0.7470	-1.1970
0.4	V^E_{exp}	-1.0893	-1.9432	-2.2325
0.5	V^E_{exp}	-1.5456	-2.2004	-2.3360
0.6	V^E_{exp}	-0.9790	-1.8282	-2.1924
0.8	V^E_{exp}	-0.5393	-0.7937	-1.2701

If the data in the form of excess volumes are plotted against mass fraction of reference components at temperatures range of 30-40 °C, smooth curves are obtained as shown into Figs. (2), (3) and (4). The curves pass through zero at 0 wt% and 100 wt% reference component, while the maximum excess volume occurs at, or close to, mass fraction of 0.5, indicating that V^E at this point should be a good indicator of the molecular interactions in the mixtures.

Excess volumes are negative for (stock 40 + stock 60), (stock 40 + stock 150) and stock (60 + stock 150) mixtures over the mass fraction range, as shown into Figs. (2), (3) and (4). Both oil stocks mixtures (stock 40 + stock 60 and stock 60 + stock 150) gave approximately the same behavior of excess volume at different temperatures as shown into Fig. (5). Due to the non-ideal behavior of the system as compared with the calculated ideal volume, it can be also noticed from Figs. (2), (3) and (4) that the increase in temperature was accompanied with an increase in the value of V^E of binary crude oils. Negative V^E is obtained over the whole mass fraction range for binary heavy oil-stock mixtures at 30, 35 and 40 °C, under concern.

Tables (3), (4), and (5) are depicted into Figs. (2), (3), and (4) in terms of function of mass fraction. Fig.(5) shows maximum negative deviation values of V^E for the (stock 40 + stock 150) mixtures (-5.6093 cm3/kg at 40 °C). V^E curves are almost symmetric with the occurrence of V^E maximum at 0.5 mass fractions. This negative excess volume (shrinkage) cames out from the changes in self-association (inter or intramolecular interactions) and physical interaction (van der Waals interaction and dipole–dipole interaction) than generated between like molecules and undo the decreases in the volume [10]. On the other hand, free volume effects, interstitial accommodation or interactions between unlike molecules lead to volume contraction.

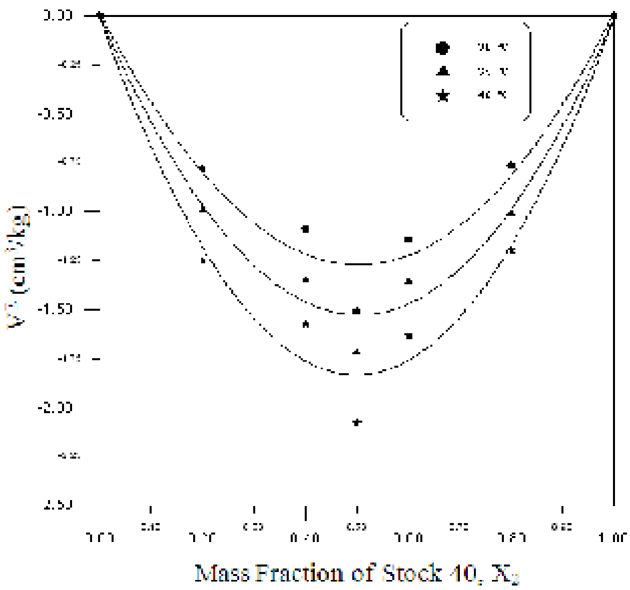

Fig.2. Excess Volume V^E for Stock 40 with Stock 60 at 30, 35 and 40 °C)

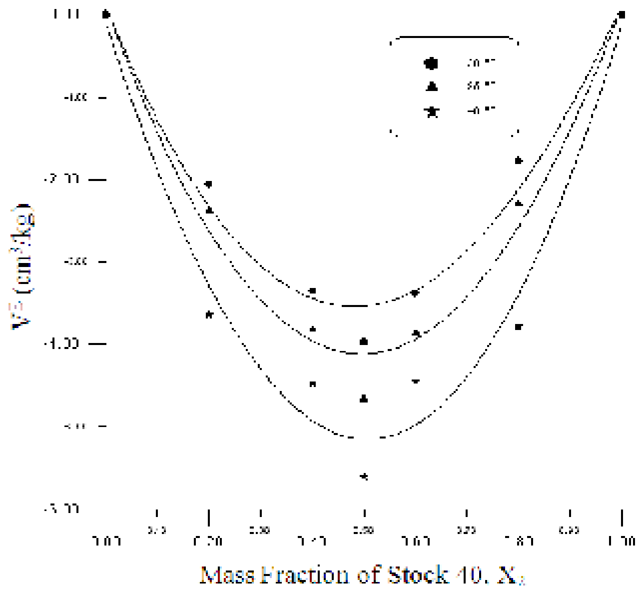

Fig.3. Excess Volume V^E for Stock 40 with Stock 150 at 30, 35 and 40 °C.

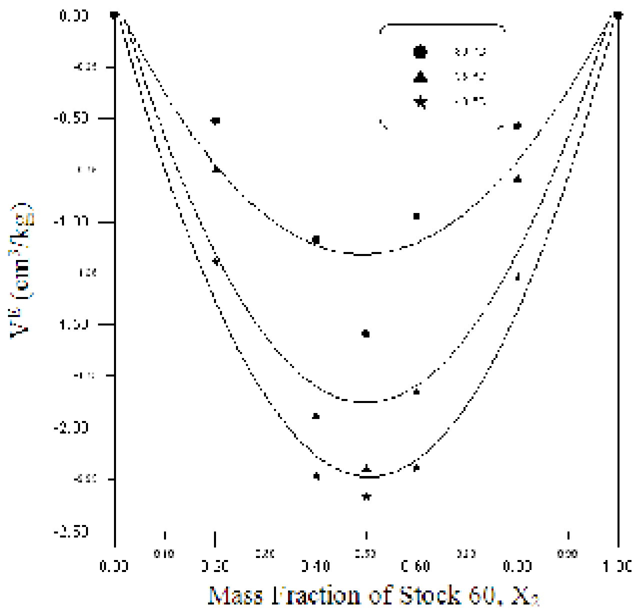

Fig.4. Excess Volume VE for Stock 60 with Stock 150 at 30, 35 and 40 °C.

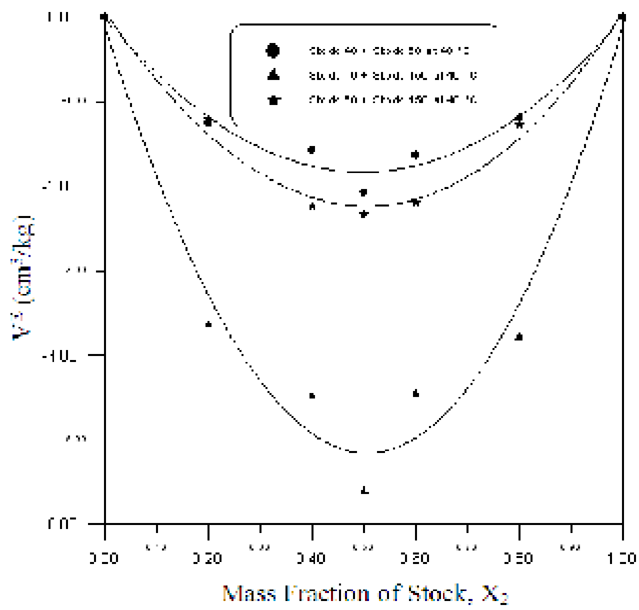

Fig.5. Excess Volume VE for Binary Mixtures of Stock Oil at 40 °C.

Table 6,
Excess Volumes V^E, for Spiked Heavy Oil-Stock at 30, 35 and 40 °C).

Stock 40 X_2	**Temperature**								
	30 °C			**35 °C**			**40 °C**		
	Gas oil	**Toluene**	**Reformate**	**Gas oil**	**Toluene**	**Reformate**	**Gas oil**	**Toluene**	**Reformate**
0.2	-0.5871	0.4511	-3.4789	-1.3803	0.7125	-3.8057	-1.5108	1.1529	-3.9150
0.4	-0.8048	0.7359	-4.6910	-1.6439	1.2779	-4.4355	-1.7034	3.1330	-4.8383
0.5	-1.3024	1.7242	-5.1969	-1.8833	2.4827	-6.3609	-2.2185	3.5718	-7.4587
0.6	-0.9284	0.7868	-4.6368	-1.5058	1.2361	-4.2851	-1.7896	2.9824	-5.1082
0.8	-0.4785	0.4281	-3.2529	-1.2144	0.6504	-3.7755	-1.4638	1.2009	-4.1677
Stock 60 X_2	**Temperature**								
	30 °C			**35 °C**			**40 °C**		
	Gas oil	**Toluene**	**Reformate**	**Gas oil**	**Toluene**	**Reformate**	**Gas oil**	**Toluene**	**Reformate**
0.2	-0.8387	-0.6438	-3.9328	-1.4588	-0.9120	-4.8113	-1.7580	-1.3359	-6.9053
0.4	-1.3824	-0.9887	-6.6942	-1.8582	-1.5394	-6.9254	-2.6400	-2.0288	-8.9683
0.5	-1.8798	-1.7571	-8.7438	-2.5500	-2.1099	-9.5726	-3.0878	-2.9151	-10.9243
0.6	-1.3277	-0.8576	-6.7819	-1.8639	-1.3398	-7.1612	-2.4687	-1.8970	-9.2383
0.8	-1.1430	-0.6310	-4.0481	-1.5024	-0.9636	-5.0424	-1.8773	-1.2794	-7.4807
Stock 150 X_2	**Temperature**								
	30 °C			**35 °C**			**40 °C**		
	Gas oil	**Toluene**	**Reformate**	**Gas oil**	**Toluene**	**Reformate**	**Gas oil**	**Toluene**	**Reformate**
0.2	-1.3590	-0.5970	-5.7449	-2.1173	-0.9323	-8.6605	-3.5738	-1.1149	-9.8580
0.4	-2.4588	-0.6587	-8.0783	-3.9455	-1.1873	-10.8835	-4.5397	-1.3392	-13.2253
0.5	-5.0172	-0.7806	-11.2640	-3.7366	-1.3139	-14.0229	-5.4360	-1.7155	-15.5622
0.6	-2.7736	-0.6445	-7.8434	-4.1536	1.2624	-10.8351	-4.7196	-1.2954	-12.8288
0.8	-1.4146	-0.6138	-5.6497	-2.5497	0.8964	-8.6506	-3.8789	-1.0963	-10.2505

The lower values of the V^E in comparison to other mixture is the result of the ideality of the stock 40 + stock 60 mixture, i.e. the difference between the mixture volume V_{mix} and the ideal mixture molar volume $V_{idel} = xV_1 + (1-x)V_2$ is very small (maximum value of about (-1.51) cm^3/kg at 30 ° and 0.5 mass fraction.

The three heavy oil-stocks which had been blended with hydrocarbons spikes (gas oil, toluene and reformate), have been subjected to density measurements to evaluate its volumetric behavior at temperature range 30-40 °C. The density data obtained for each heavy oil-stock/spike pairs are reported in the form of excess volume VE at a given mass fraction of spike X2 as shown in Table (6).

If the data in the form of specific excess volume are plotted against mass fraction of spike of blend, smooth curves are obtained as shown in Figs. (6), (7) and (8) at 40 °C. The curves pass through zero at 0 wt% spike and 100 wt% spikes, while the maximum excess volume occur at, or close to, mass fraction of 0.5, indicating that V^E at this point would be good indication of the molecular interactions in these systems.

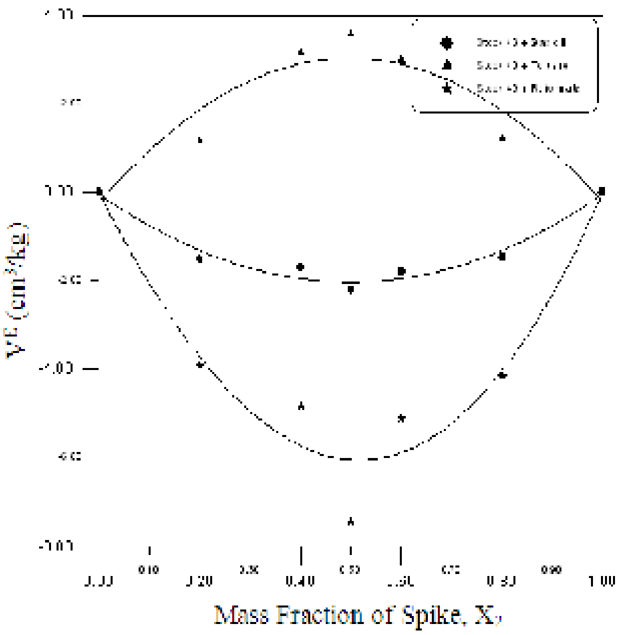

Fig.6. Excess Volume V^E for Spiked Stock 40 at 40°C.

It is generally observed that the addition of middle petroleum fractions, such as gas oil to the three heavy oil-stocks of different densities produces negative excess volumes. In other words "shrinkage" occurs in comparison with the calculated ideal volume; as shown in Fig. (9).

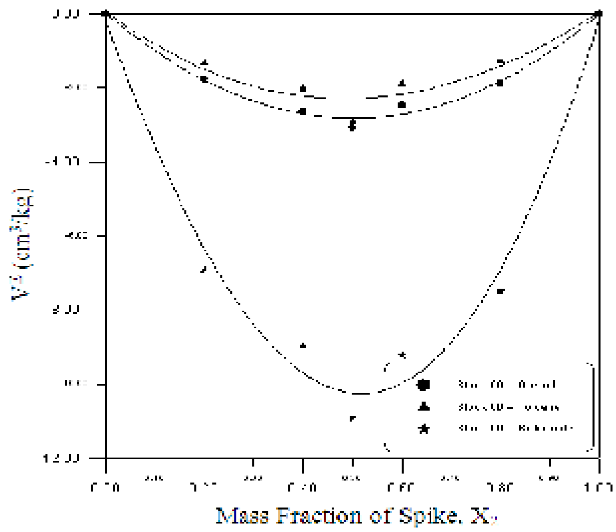

Fig.7. Excess Volume VE for Spiked Stock 60 at 40°C.

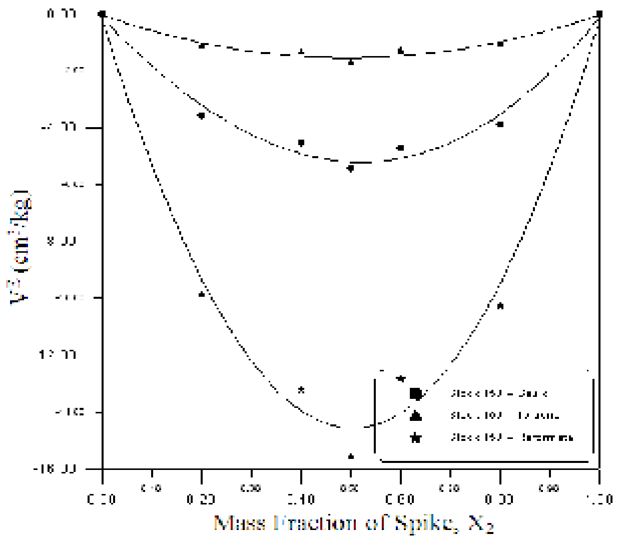

Fig.8. Excess Volume V^E for Spiked Stock 150 at 40°C.

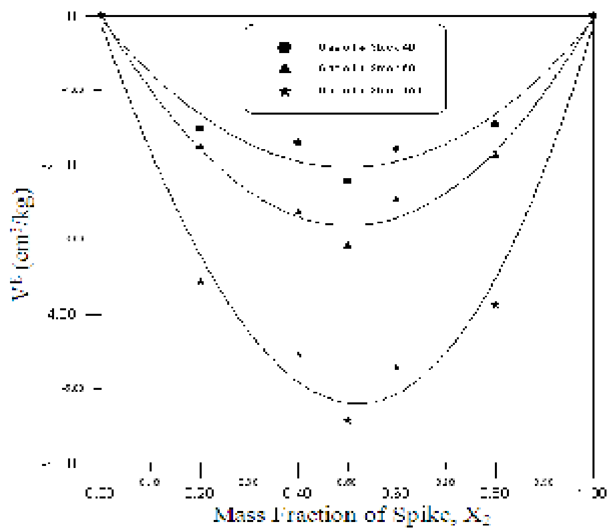

Fig.9. Excess Volume VE for Binary Mixtures of Stock Oil/ Gas Oil at 40 °C.

Also, the addition of the lowest boiling point spike as in the case of reformate to the three heavy oil-stocks of different densities produces negative excess volumes; in other words "shrinkage" occurs, as shown in Fig. (10).

This shrinkage effect is directly proportional to the specific gravities of the oil-stocks, thereof when the density of oil-stock increase the negative V^E will be increased. As noticed in Fig. (10), the derived values of V^E for the (reformate + 150 oil stock) mixtures records negative (maximal value at about (-15.56) cm^3/kg).

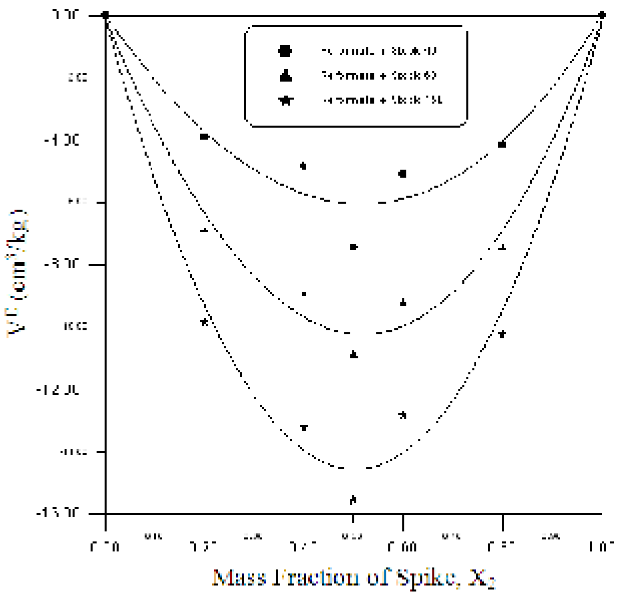

Fig.10. Excess Volume VE for Binary Mixtures of Stock Oil/ Reformate at 40 °C.

The negative V^E values arise due to the dominance of the following factors [11]:

(a) Chemical interaction between constituent molecules such as hetero-molecular association through the formation of H-bond, often termed as strong specific interaction.
(b) Association through weaker physical forces such a dipolar force or any other forces of this kind.
(c) Accommodation of molecules of one component into the interstitial positions of the structural network of molecules of the other component.
(d) Geometry of the molecular structure that favors fitting of the component molecules with each other.

The blending of heavy oil-stocks with aromatic spikes, such as toluene give a positive V^E and negative V^E depending upon the specific gravity of heavy oil-stock that blends with it. So that the blending of toluene with stock 40 gives positive excess volume and this phenomena refers to the interactions between molecules that are weak and it is better to say that the interactions between dissimilar molecules are much weaker between the similar ones. Give rise positive deviations. But the blending of toluene with stock 60 and stock 150 give negative excess volume and this belong to the effect of the two methyl groups of toluene. The binary mixtures of stock 40 with toluene show positive values of excess volume, and thus exhibit expansion, as shown in Fig. (11). This effect is appreciably in relation to the fact, that such compounds are "structure breaking" materials leading to positive excess volume [12].

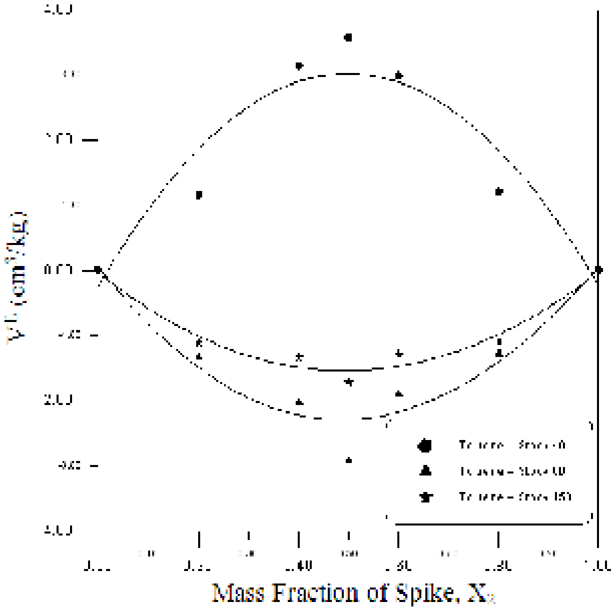

Fig.11. Excess Volume V^E for Binary Mixtures of Stock Oil/ Toluene at 40 °C.

The factors that are mainly responsible for the volume expansion i.e., positive values of V^E, are [11]:

(a) Disruption of one or both components in a solution system. A suitable example of this, is the rupture of H-bonding of one compound by the other, or breaking up of associates held together by weaker physical forces such as dipole or dipole-induced dipole interactions or by any other van der Waals forces.
(b) The geometry of molecular structure which does not favor fitting of the molecules with each other.

(c) (c) Steric hindrance which opposes the proximity of the constituent molecules.

The binary mixtures of stock 60, 150 with toluene shows negative values of excess volume over the whole composition range as shown in Fig.(11). V^E curves are almost symmetric with their maximal point at 0.5 mass fraction. This negative excess volume (shrinkage) come from the changes in self-association (inter or intramolecular) and physical interaction between similar molecules which increase the volume [10].

4. Conclusions

It is general, it was observed that the blending of light medium oil-stocks and heavy oil-stock results in volume "losses" caused by the non-ideal behavior of those system as compared with their calculated ideal volumes which is accompany increased by increasing in temperature. Also it was shown that the mixing of moderate petroleum fractions, such as gas oil and the addition of lowest boiling point spike as in the case of reformate with heavy oil-stocks produces negative excess volume.

While the blends of aromatics hydrocarbons with heavy oil-stocks produces positive excess volume except for stocks 60, 150 at temperature range of 30-40 °C.

Nomenclature

x_2 mass fraction of component 2

V^E excess volume, cm3/kg

ρ_{mix} density of the mixture for binary system, g/cm^3

ρ_i density of component i, g/cm^3

5. References

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تاثير النسب الوزنية ودرجة الحرارة على الحجم الزائد للاساسات زيت التزليق بوجود المضافات الهيدركاربونية (زيت الغاز+ التلوين + الريفورميت)

عزت نيازي سليمان *   حيدر موفق توفيق **   شهد زهير عطا **

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** قسم الهندسة الكيمياوية/ كلية الهندسة/ جامعة النهرين

الخلاصة

تم استخدام ثلاث انواع من اساسات زيت التزليق وهي الاستوك ٤٠ وهو الاخف من حيث الوزن الجزيئي والمتوسط وهو الاستوك ٦٠ والاثقل وزنا جزئيا الاستوك ١٥٠ لمعرفة السلوك الحجمي لمخاليط تلك الزويت وكذلك سلوك تلك الزيوت حجميا عند اضافة المضافات الهيدروكاربونية مثل (زيت الغاز و التلوين و الريفورميت) وكذلك بدرجات حرارة متغيرة (٣٠-٤٠ °م). حيث ان اضافة هذه المواد تؤدي الى زيادة في الحجم قد تكون موجبة او سالبة بمعنى اخر ان هذه الاضافات قد تودي الى انكماش او تمدد حجمي في تلك الخلائط. كما ان لدرجة الحرارة تاثير ملحوظ على الزيادة الحجمية لكل مزيج تمت دراسته. حيث تبين ان اضافة المادة الهيدروكاربونية ذات الطابع البرافييني (زيت الغاز) يؤدي الى حدوث انكماش ملحوظ في حجم الخلائط وكذلك المواد الهيدروكاربونية ذات درجة الغليان الواطى كما هو الحال في (الريفورميت) بينما اضافة المادة الهيدروكاربونية ذات الطابع الوروماتي (التلوين) يؤدي الى تمدد تلك الخلائط ماعدا خلائط الاستوك ١٥٠و ٦٠ حيث ان اضافة التلوين الى تلك الخلائط يؤدي الى انكماشها والسبب في سلوك الزايلين بصورة عامة يرجع الى وجود مجموعتي المثيل في الحلقة (التلوين). تم الحصول على اعلى نسبة من الوزن الزائد وهي ١٥,٥٦ عند اضافة الريفورميت الى الاستوك ١٥٠.