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RESEARCH ARTICLE

EVALUATION OF REFRACTIVE INDICES AND THE ACCURACY OF SOME RULES FOR MIXING BINARY MIXTURES

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Abstract

The determination of light refractive index is an important step in the characterization of liquid mixtures with different industrial applications. Through it, the purity of their substances can be assessed and, together with density and viscosity, it has a significant role in characterizing and understanding the thermodynamic properties of the liquids. Theaim of the present studywas to evaluate the accuracyofsomeofthe common mixingrules asappliedto the followingbinarymixtures: Benzene – Toluene, n-Heptane–n-Hexane, Chloroform – Benzene and Chloroform – Tolueneatdifferentconcentrationsandtemperatures.Themixtures were chosen becausedozens of millions of tonsof them areusedworldwide andthus they haveagreattechnologicalsignificanceandareessentialinalmostallaspectsofthe chemicalsynthesis.Inaddition, on the basisoftheresultsobtained,amodifiedequationbasedontheArago-Biotmixingrule was developed.The equationismoreaccurateandeasiertousethanmostexistingexpressionsinthe literature.

Introduction:-

The refractive index of light is a useful parameter for characterizing liquids that are widely applied in the industry, such as oils, waxes, sugar syrups, etc. It is a fundamental physical property that measures the speed of light in a material and characterizes its optical properties [1]. It has been used for years for accurate identification and characterization of pure liquids and mixtures [2]. A number of authors [3, 4] have noted that the refractive index, density and viscosity are essential for characterizing and understanding the thermodynamic properties of liquids. The molecular interaction in a fluid mixture can also be estimated from the refractive index and the density of its pure components [5, 6]. The refractive index is useful in the indirect measurement of density and salinity and in the detection of structural properties of liquid-liquid mixtures. Its application has also led to the development of alternatives to fuel substitutes, additives and oil treatment with chemicals [7].

By using mixing rules [8], the composition of an unknown mixture as well as the presence of molecular interactions in binary mixtures can be determined. Quite a few researchers have noticed a discrepancy between the theoretical and experimental values of the refractive indices of mixtures, and such deviations can be reduced by taking the concept of excess volume into account.

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Thevaluesoftherefractiveindicesforverypureliquidsareknownorreadilyavailableintheliterature.Besides, therefractiveindicesofpure,two-andmulticomponentmixturescanbeeasilymeasureddirectlybymeansofrefractometers.However, whenexperimentaldata are notavailable, the refractiveindicesofbinaryandmulticomponentmixturesareoftenestimatedfromtheirpurecomponentsusingmixingrules. Themostcommonlyusedmixingrulesare the Lorentz-Lorenzequation, theWeinerrelation,and the Heller,Arago-BiotandGladstone-Daleequations.Thesemixingrulesrequirestrictproceduresthatsometimescause inaccuracies.

Someauthors[9]proposemodifiedversions of thegenerallyknownmixingruleswhere the presenceofa temperaturedependenceisalsotakenintoaccount.

The aim of the present study was to evaluate the accuracy of some of the common mixing rules as applied to the following binary mixtures: Benzene – Toluene, n-Heptane – n-Hexane, Chloroform – Benzene and Chloroform – Toluene at different concentrations and temperatures of 20^0C and 40^0C. In addition, on the basis of the results obtained, a modified equation based on the Arago-Biot mixing rule was developed.The equation has a wider range of applicability due to the temperature dependence included.

Computational

In the process of mixing two or more components, effects such as structural reorientation occur due to differences in the shape and size of the molecules of the components and the molecular interaction [10]. Various empirical and semiempirical relations were formulated and tested by a number of authors [11, 12]. The validity of these mixing rules was also tested by many other authors [13 ÷ 18]. The relative merits of these mixing rules were later studied and discussed by other investigators [19 ÷ 24]. The usefulness of the mixing rules in the determination of binary refractive indices and density data has also been pointed out [25].

Themostwidelyusedtheoreticalrulesweresproposedby Lorentz [27]andWeiner[28].By assumingthatthe volumesofpurecomponents can beaddedwhenanalyzingfluidmixturesandusing the meanpolarizabilitywhichapproximates the averagepossibleorientationsof amolecule,theLorentz-Lorentzrelationshipcanbeobtained[29].The Lorentz-Lorenz equation wasnamedafterDanishmathematician and scientistLudvigLorenz, who publishedit in 1869, andDutchphysicistHendrikLorentz,whodiscovereditindependentlyin1878.

TheLorentz-Lorenz equation fora binary mixtureisexpressedas:

$$\frac{n_m^2-1}{n_m^2+2} = y_1\frac{n_1^2-1}{n_1^2+2} + y_2\frac{n_2^2-1}{n_2^2+2} \quad (1)$$

Weiner’s relation [28]is given as:

$$\frac{n_m^2-n_1^2}{n_m^2+2.n_1^2} = y_2\frac{n_2^2-n_1^2}{n_2^2+2.n_1^2} \quad (2)$$

While Heller’s relation is [12]:

$$\frac{n_m-n_1}{n_1} = \frac{3}{2}.y_2.\frac{m^2-1}{m^2+2} \quad (3)$$

where: $m = n_2/n_1$

If we assumethatthereis volume additivityduringmixing,wecanusethesimpleArago-Biotrelationship[30]:

$$n_m = n_1.y_1 + n_2.y_2 \quad (4)$$

Inthe present work,anothermodifiedversionoftheArago-Biotequation[30] is proposed on the basis of the obtainedexperimentaldataon the refractiveindices of the studiedbinarymixtures:

$$n_m = \left[(n_1.y_1 - k_1) + (n_2.y_2 - k_2)\right] \quad (5)$$

where:

$$k_1 = y_1.10^{-3} \quad (6)$$

$$k_2 = y_2.10^{-4} \quad (7)$$

For pure components,$k_1$and$k_2$are equal to 0.

Materials and Methods:-

For the aims of the experiment, the light refractive indices of samples of the following pure compounds were measured (Table 1):

Compound	**Purity %**	**Producer**
Benzene	99.80	Merck, Darmstadt
Toluene	99.93	Marvin OOD, Dimitrovgrad
n-Heptane	99.93	Marvin OOD, Dimitrovgrad
n-Hexane	97.00	Riedel-de Haën
Chloroform	99.40	Riedel-de Haën

Literature data (Table 2) on some of the physical properties of the pure components in the experiment were used [31].The measured values of the refractive indices of the pure substances were also compared withthe respective refractive indices in the literature [32].

Table 2:-

Chemical formula	**Density, kg/m^3**		**T_{BP} ^0C**	**n_D^{20} [34]**	**n_D^{20} exp.**	**n_D^{40} exp.**
	20^0C	**40^0C**				
C_6H_6	879	858	80.2	1.50112	1.50104	1.48848
C_7H_8	866	847	110.8	1.49693	1.49649	1.48478
C_7H_{16}	670	650	98.0	1.38764	1.38734	1.37628
C_6H_{14}	660	641	69.0	1.37499	1.37427	1.36463

For the purpose of the experiment, an Abbe - KRÜSS AR4D refractometer with a double prism, 589 nm monochromatic light source and refractive index measurement accuracy of ± 0.0001 connected to a $50^0C \div 200^0C$ digital thermometer was used.

Since accurate measurements depend on the careful calibration of the refractometer, the latter was calibrated at 20°C using a thermostat connected to it. The measured and averaged value of the refractive index of the distilled water used as a calibration solution was 1.33289.

During the second stage of the study, the binary mixtures Benzene – Toluene, n-Heptane – n-Hexane, Chloroform – Toluene and Chloroform - Benzene were prepared by mixing the pure components in different volumes: 5 to 0 ml, 4.5 to 0.5 ml, 4 to 1 ml, 3.5 to 1.5 ml, 3 to 2 ml, 2.5 to 2.5 ml, 2 to 3 ml, 1.5 to 3.5 ml, 1 to 4 ml, 0.5 to 4.5 ml and 0 to 5 ml.

A KERNABS220-4analyticalbalancewasusedas a weighing system.

The refractiveindices of thepurecomponents and binarymixtures were measuredat 20^0C and 40^0C by means ofathermostatconnectedto the refractometer. Theresultswererepeatedtoensureaccuracyofthe measurements.

To find the mass concentration [g/g] of the highly volatile component the following formula was used [31]:

$$\bar{x} = \frac{m_{HiVC}}{m_{mix}}.100, \% \qquad (8)$$

where:

m_{HiVC}isthe mass of the highly volatile component in the mixture, g/g;

$m_{mix}$is the total mass of the binary mixture, g/g.

$$m_{MIX} = m_{HiVC} + m_{HaVC} \qquad (9)$$

To recalculate the composition (in liquid phase) of the three mixtures from mass [g/g] to mol [mol/mol], the following formula was used [31]:

$$x_i = \frac{\frac{\bar{x}_i}{M_A}}{\frac{\bar{x}_i}{M_A}+\frac{(1-\bar{x}_i)}{M_B}} \qquad (10)$$

where$M_A$isthe molecular mass of the highly volatile component (HiVC) in the mixture, [kg/kmol];

$M_B$isthe molecular mass of the hardly volatile component (HaVC) in the mixture, [kg/kmol].

Results and Discussion:-

Tables 3 to 10 and Figures 1 to 4 show the results obtained from the experiments that were carried out. It is evident that the refractive indices of pure liquids and mixtures decrease with the increase of temperature. This is consistent with the results of previous investigations [17, 33, 34] and can be attributed to changes in the liquid density due to temperature. At higher temperatures, the density of the liquid decreases, causing the light to travel faster in the medium, which results in a lower refractive index. The variation of refractive indices with temperature can also be attributed to the structural changes that occur during mixing.

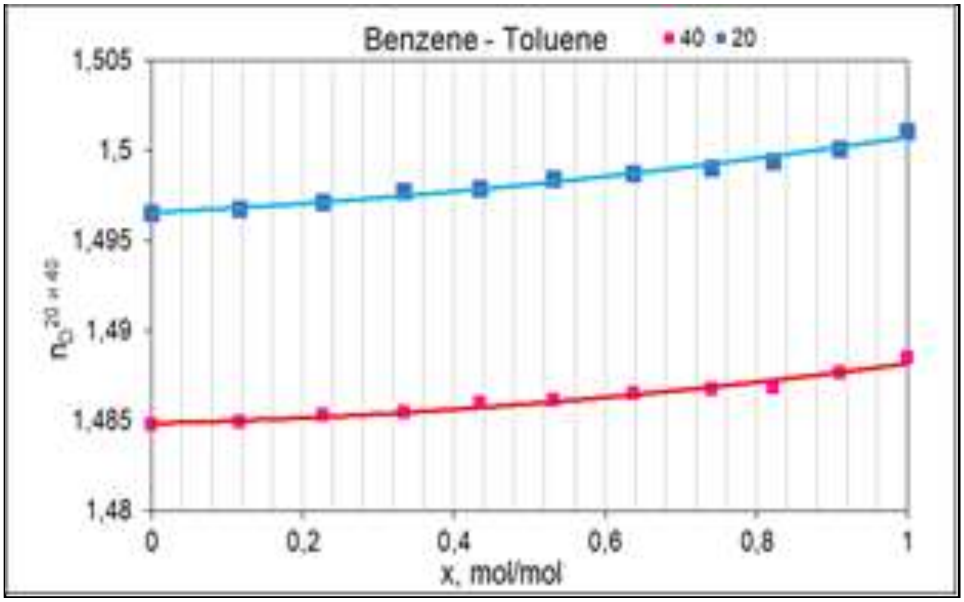

Fig. 1:- Dependence of the refractive index on the composition of the Benzene – Toluene mixture at 20^0C and 40^0C.

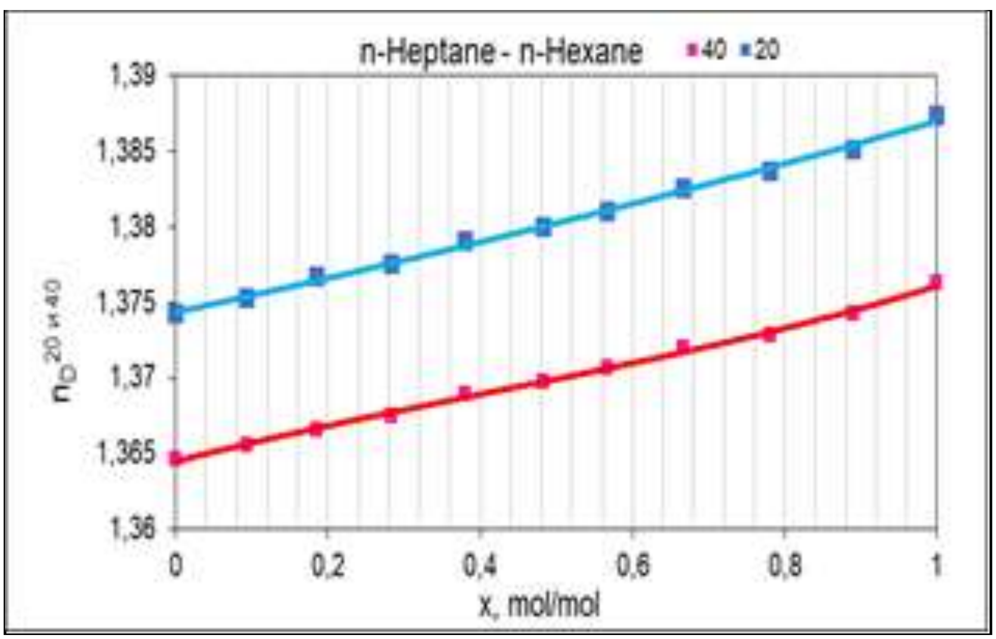

Fig. 2:- Dependence of the refractive index on the composition of the n-Heptane – n-Hexane mixture at 20^0C and 40^0C.

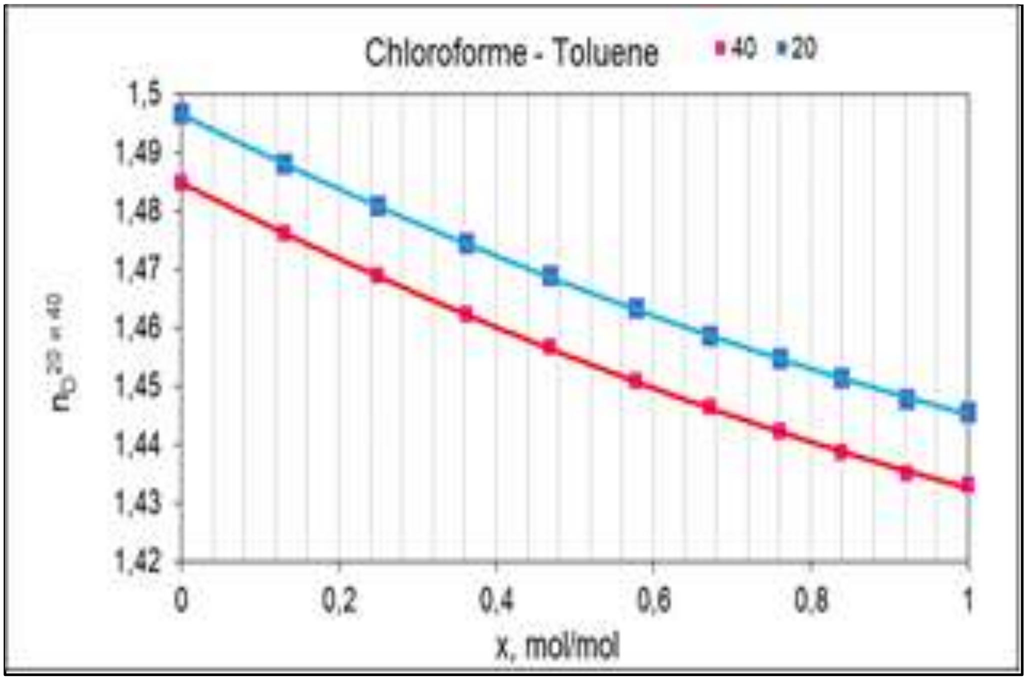

Fig. 3:- Dependence of the refractive index on the composition of the Chloroform – Toluene mixture at 20^0C and 40^0C.

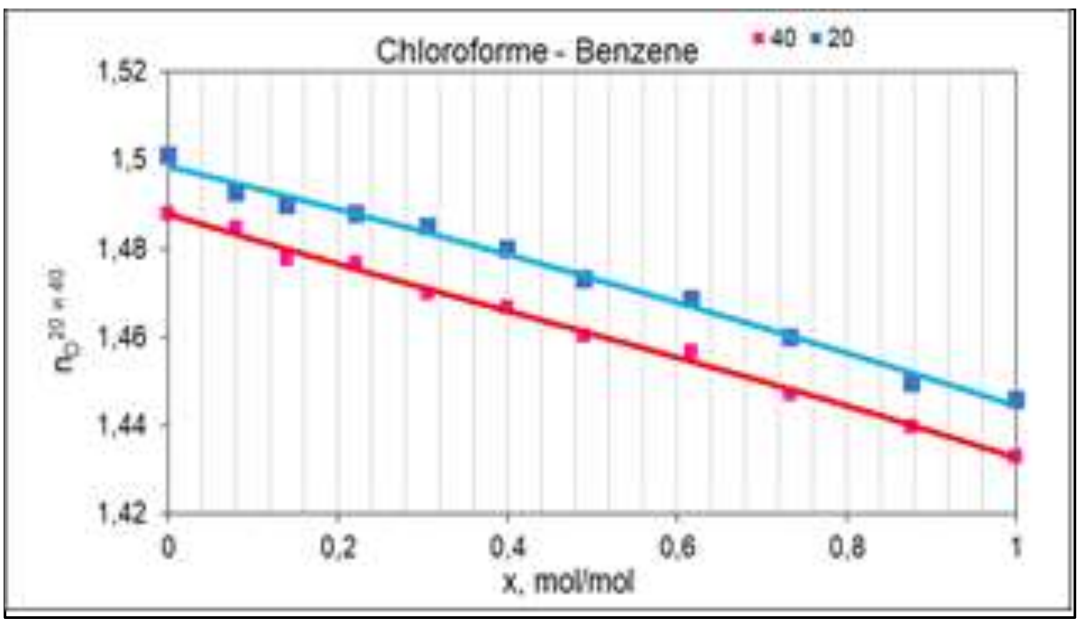

Fig. 4:- Dependence of the refractive index on the composition of the Chloroform – Benzene mixture at 20^0C and 40^0C

The experimental results were used to check the accuracy of some commonly used mixing rules. The results show that all refractive indices calculated using the mixing rules were reasonably close to the measured values for the studied binary mixtures. Moreover, all the mixing rules considered show remarkable changes in the refractive indices of the mixtures with the increase of temperature.

Table 3:- Refractive indices from experimental data for the Benzene – Toluene mixture at 20°C.

x_i	Experimental	Lorentz-Lorenz	Weiner	Heller	Arago-Biot	Arago-Biot modified
0.00	1.48478	1.48478	1.48478	1.48478	1.48478	1.48478
0.25	1.48530	1.48570	1.48570	1.48570	1.48571	1.48538
0.50	1.48604	1.48663	1.48663	1.48663	1.48663	1.48608
0.75	1.48671	1.48755	1.48755	1.48755	1.48756	1.48678
1.00	1.48848	1.48848	1.48848	1.48848	1.48848	1.48848

The results obtained from the experiment are also presented graphically using the calibration curve method. Fig. 5 shows the dependence of the refractive indices on the concentration of the Benzene – Toluene mixture for the different calculated mixing rules.

It is evident from the figure that there is an almost complete match of the refractive indices from the experimental data with those calculated by the Arago-Biot equation modified in this work at both 20^0C and 40^0C. It is also evident that the refractive indices decrease with the increase of temperature.

Table 4:- Refractive indices from experimental data for the Benzene – Toluene mixture at 40°C.

x_i	Experimental	Lorentz-Lorenz	Weiner	Heller	Arago-Biot	Arago-Biot modified
0.00	1.49649	1.49649	1.49649	1.49649	1.49649	1.49649
0.25	1.49722	1.49763	1.49763	1.49763	1.49763	1.49730
0.50	1.49822	1.49876	1.49876	1.49876	1.49877	1.49822
0.75	1.49906	1.49990	1.49990	1.49990	1.49990	1.49913
1.00	1.50104	1.50104	1.50104	1.50104	1.50104	1.50104

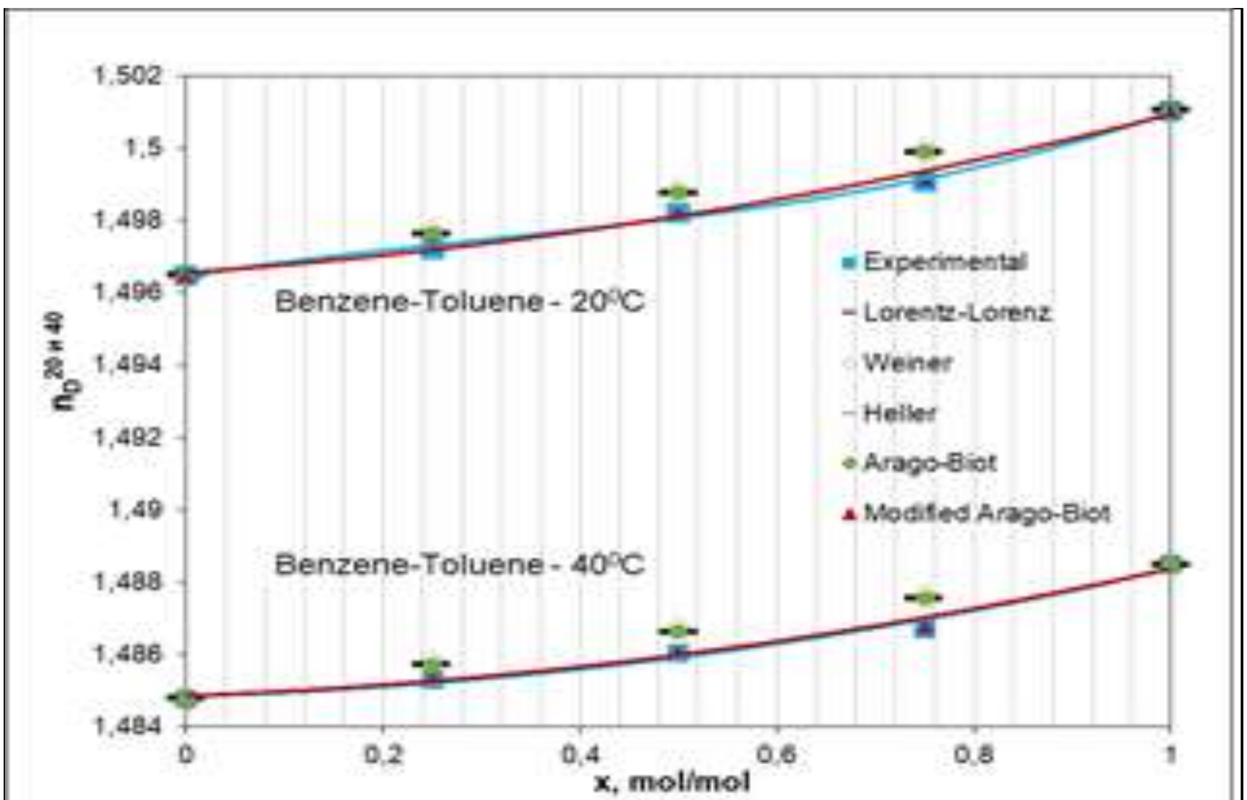

Fig.5:- Dependence of the refractiveindices on the composition of the Benzene – Toluene mixture at 20^0C and 40^0C.

The experimentally obtained results for the other three mixtures are presented in Tables 5 to 10 and Figures 6 and 7.

Table 5:- Refractive indices from experimental data for the n-Heptane – n-Hexane mixture at 20°C.

x_i	Experimental	Lorentz-Lorenz	Weiner	Heller	Arago-Biot	Arago-Biot modified
0.00	1.37427	1.37427	1.37427	1.37425	1.37427	1.37427
0.25	1.37721	1.37753	1.37753	1.37752	1.37754	1.37721
0.50	1.38022	1.38079	1.38080	1.38079	1.38081	1.38026
0.75	1.38330	1.38406	1.38407	1.38407	1.38407	1.38330
1.00	1.38734	1.38734	1.38734	1.38734	1.38734	1.38734

Table 6:- Refractive indices from experimental data for the n-Heptane – n-Hexane mixture at 40°C.

x_i	Experimental	Lorentz-Lorenz	Weiner	Heller	Arago-Biot	Arago-Biot modified
0.00	1.36463	1.36463	1.36463	1.36461	1.36463	1.36463
0.25	1.36721	1.36753	1.36754	1.36753	1.36754	1.36722
0.50	1.36996	1.37044	1.37045	1.37045	1.37046	1.36991
0.75	1.37258	1.37336	1.37336	1.37336	1.37337	1.37259
1.00	1.37628	1.37628	1.37628	1.37628	1.37628	1.37628

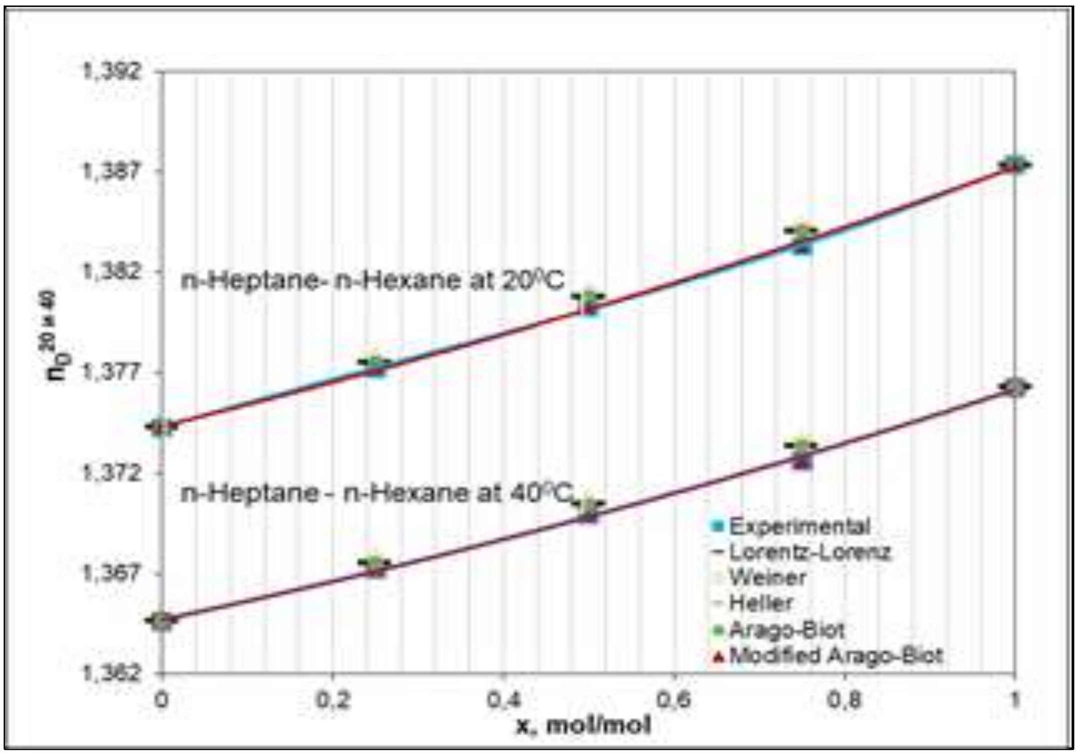

Fig.6:-Dependence of the refractiveindices on the composition of the n-Heptane – n-Hexane mixture at 20^0C and 40^0C.

Table 7:- Refractive indices from experimental data for the Chloroform – Toluene mixture at 20°C.

x_i	Experimental	Lorentz-Lorenz	Weiner	Heller	Arago-Biot	Arago-Biot modified
0.00	1.49649	1.49649	1.49649	1.49618	1.49649	1.49649
0.25	1.48339	1.48359	1.48371	1.48354	1.48377	1.48341
0.50	1.47045	1.47082	1.47097	1.47090	1.47106	1.47051
0.75	1.45750	1.45816	1.45828	1.45826	1.45834	1.45756
1.00	1.44562	1.44562	1.44562	1.44562	1.44562	1.44562

Table 8:- Refractive indices from experimental data for the Chloroform – Toluene mixture at 40°C.

x_i	Experimental	Lorentz-Lorenz	Weiner	Heller	Arago-Biot	Arago-Biot modified
0.00	1.48478	1.48478	1.48478	1.48445	1.48478	1.48478
0.25	1.47145	1.47164	1.47176	1.47158	1.47183	1.47150
0.50	1.45827	1.45863	1.45879	1.45871	1.45888	1.45833
0.75	1.44510	1.44574	1.44586	1.44584	1.44592	1.44515
1.00	1.43297	1.43297	1.43297	1.43297	1.43297	1.43297

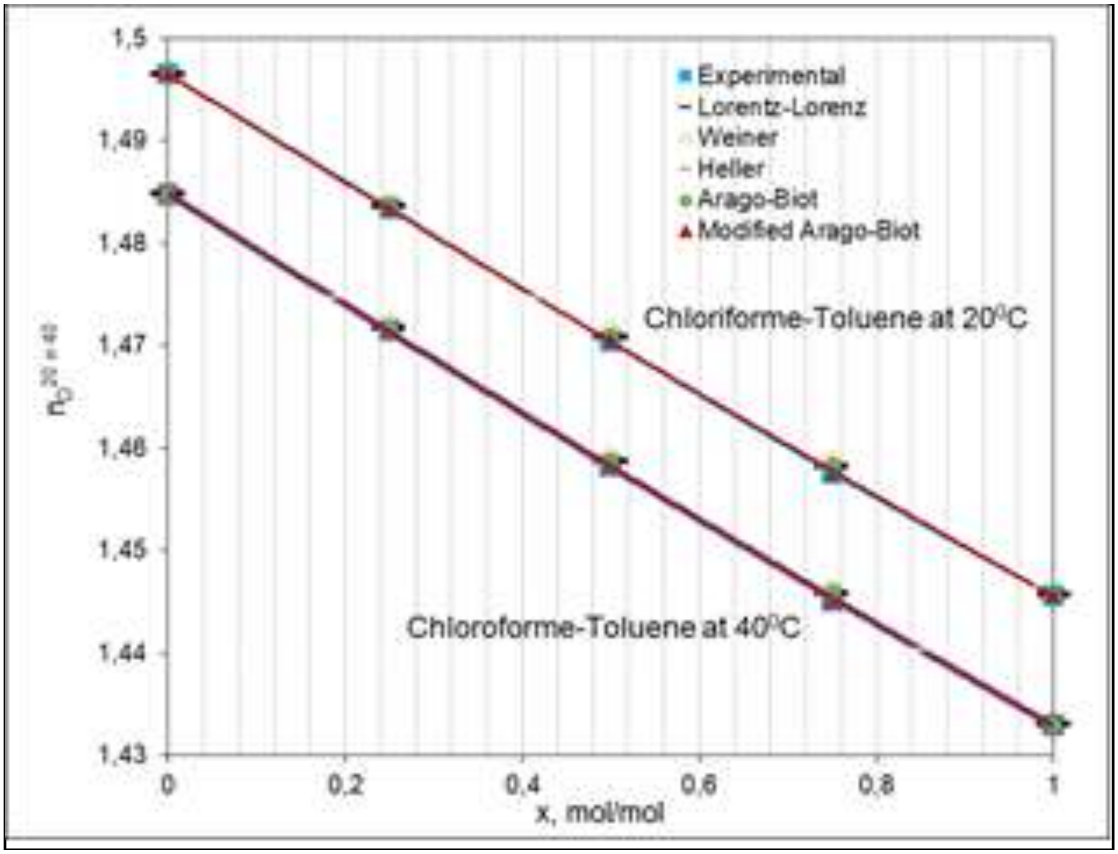

Fig. 7:- Dependence of the refractiveindices on the composition of the Chloroform – Toluene mixture at 20^0C and 40°C.

Table 9:- Refractive indices from experimental data for the Chloroform – Benzene mixture at 20°C.

x_i	Experimental	Lorentz-Lorenz	Weiner	Heller	Arago-Biot	Arago-Biot modified
0.00	1.50104	1.50104	1.50104	1.50067	1.50104	1.50104
0.25	1.48678	1.48697	1.48711	1.48691	1.48719	1.48686
0.50	1.47271	1.47305	1.47323	1.47314	1.47333	1.47278
0.75	1.45867	1.45927	1.45940	1.45938	1.45948	1.45870
1.00	1.44562	1.44562	1.44562	1.44562	1.44562	1.44562

Table 10:- Refractive indices from experimental data for the Chloroform – Benzene mixture at 40°C.

x_i	Experimental	Lorentz-Lorenz	Weiner	Heller	Arago-Biot	Arago-Biot modified
0.00	1.48848	1.48848	1.48848	1.48810	1.48848	1.48848
0.25	1.47426	1.47439	1.47453	1.47432	1.47460	1.47428
0.50	1.46015	1.46044	1.46063	1.46054	1.46073	1.46018

0.75	1.44606	1.44664	1.44678	1.44675	1.44685	1.44607
1.00	1.43297	1.43297	1.43297	1.43297	1.43297	1.43297

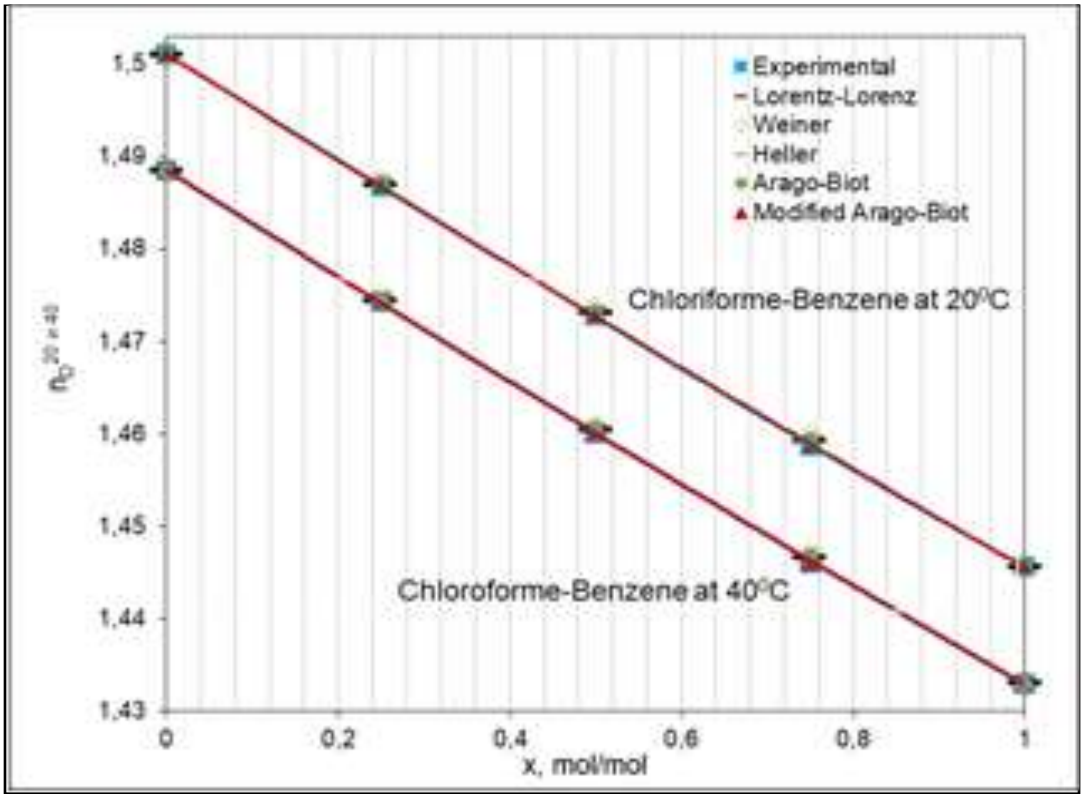

Fig. 8:- Dependence of the refractiveindices on the composition of the Chloroform – Benzene mixture at 20^0C and 40^0C.

Conclusions:-

On the basis of the experiments and the analysis of the obtained results, the following conclusions can be made:

1. The refractive indices of five pure compounds,namely Benzene, Toluene, n-Heptane, n-Hexane and Chloroform, have been determined and presented.
2. The refractive indices of four binary mixtures, namely Benzene – Toluene, n-Heptane – n-Hexane, Chloroform – Toluene and Chloroform – Benzene, have been determined at 20^0C and 40^0C and presented.
3. Due to lack of literature data, the dependence of the refractive index on the composition of the mixtures Benzene – Toluene, n-Heptane – n-Hexane, Chloroform – Toluene and Chloroform – Benzene has been determined by the weight method at 20^0C and 40^0C.
4. As a result of the study, it has been found out that the refractive indices of the pure components decrease with the increase of temperature.
5. The refractive indices of all four binary mixtures have also been found to decrease with the increase of temperature.
6. A modified Arago-Biot mixing rule has been proposed; it is easier to use and has a wider scope of application than some of the existing expressions in literature sources.

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