

Modeling of Refractive Indices for Binary Aqueous Solutions of Some Alkane Polyols at Constant Temperature and Pressure

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ABSTRACT: In this research, a theoretical study has been undertaken on the density and also on the relationship between refractive index and density for aqueous solutions. A simple linear equation is suggested to show this relationship. Also, a semi-empirical equation has been developed for estimating the constant of this linear equation. By using our suggested equations, the refractive indices of aqueous solutions of alkane polyols [$R(OH)_n$, $n = 2$ to 6 -OH,s) and also polyols with one ring (monosaccharide) can be calculated. Moreover, the refractive indices of some alcohols and other components have been collected from the various literature. Our suggested equations have been tested for aqueous solutions of these molecules. For aqueous solutions of alkane polyols [$R(OH)_n$, $n = 2$ to 6 -OH,s) and monosaccharide, a good agreement is observed between experimental refractive indices and theoretical ones. Other molecules show positive and/or negative deviations from the results of our suggested equations. For various liquid solutions, the refractive indices can be correlated and fitted with our linear equation. Also, the value of the constant of this linear equation, K_{ref} , shows the power of interaction between solute and solvent molecules. The interaction between solute and solvent molecules increases when the value of constant, K_{ref} increases.

KEYWORDS: Refractive index; Alkane polyols; Hydrogen bonding; Aqueous solution; Temperature, Linear equation.

INTRODUCTION

Refractive index (n_D or n) is one of the most important thermodynamic and optical properties of solutions [1-5]. It has many applications in various

industries like pharmaceutical [6,7], food [8], petroleum [9] and cosmetic [10]. In pharmaceutical and food industries, refractive index (RI) is often used to qualify control

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1021-9986/2019/5/221-237

17/\$/6.07

of crude intermediate and final products. Also, the refractive index gives us important information about various engineering calculations, for instance, RI is a key parameter for characterizing raw oils. The determination of this index through the experiment is capital-intensive and time-consuming [11]. On the other hand, the refractive index can be modified in the cosmetic materials as it is important in making clear emulsions used in cosmetic formulations [12].

Besides, it can be applied in academic and research works. The refractive index information can lead us to a better understanding of interactions between solute and solvent molecules [13-15].

For a medium like an aqueous solution, the refractive index is the ratio of the velocity of light in vacuum and phase velocity of light in the medium [16]. Also, the refractive index can be obtained from Maxwell's equation as follow [17]:

$$n = \frac{c}{v} = \sqrt{\frac{\epsilon\mu}{\epsilon_0\mu_0}} \quad (1)$$

In Equation (1), c is the velocity of light in vacuum, v is the velocity of light in the medium, ϵ and μ are the permittivity and permeability for medium and ϵ_0 and μ_0 are the permittivity and permeability for the vacuum, respectively.

The Refractive index has no dimension. Most of the materials have positive refractive index values and some others have negative ones [18-20].

Negative Index Metamaterial (NIM) has already been introduced in 1968 [21] by the general consideration of the electrodynamics properties of the materials with simultaneously negative values of the dielectric permittivity and magnetic permeability. In 1996, Pendry proposed that a negative epsilon can be made by arranging the metal wires in a simple cubic lattice [22]. But the main question was how we could get the negative μ . This question was answered by *Smith* in 2000 [23]. *Smith* proposed a structure consisting of a periodic array of split-ring resonators and continuous wires that produced the negative permeability and negative permittivity.

The refractive index of the medium varies as the wavelength of light changes [24]. In an apparatus which is used for measuring refractive index, a fixed source of light (with a certain wavelength) is applied for light generation. For example, in the Abbe refractometer, a sodium lamp generates the yellow light ($\lambda = 598.3$ nm) [25].

In recent years, some researchers have worked on refractive indices of pure compounds and solutions. *Shekaari* and *Jebali* reported refractive indices of aqueous solutions of (amino acid + Ionic Liquid ([BMIm]Br) at 298.15 K [26]. Tan and Huang studied the dependence of refractive index on concentration and temperature in an electrolyte solution, polar solution, nonpolar solution, and protein solution [27]. *Lorimer* reported refractive index increments of polymers in solution [28]. *Bai* and *Katz* researched the refractive index of sodium iodide solutions for index matching in PIV [29]. *Li et al.* reported values of sodium and potassium aluminate aqueous solutions [30]. *Chen et al.* determined the refractive index of an aqueous solution of CdTe quantum dots [31]. *Koralewski et al.* obtained optical properties of chitosan in aqueous solution [32]. *Proutiere et al.* reported refractive index variations in pure liquids [33]. *Gharagheizi et al.* reported a group contribution model for the estimation of refractive indices of organic compounds [34].

In this study, for aqueous solutions, in the first step we will show that there is a linear relationship between refractive index and molar concentration, especially in low molar concentration. In the second step, we will introduce a semi-empirical equation for the calculation of K_{ref} for binary aqueous solutions (water + glycerol, 1-propanol, 1,3-propanediol, butane-1, 3-diol, butane-1, 4-diol, butane-2, 3-diol, pentaerythritol, D-fructose, D-glucose, and D-mannitol) at constant temperature and atmospheric pressure (Table 1). However, other solute molecules (ethylene glycol, ethanol, acetone, lactic acid, acetic acid, citric acid, urea, and 1-amino-2-propanol) show a deviation (negative or positive) from the proposed equations.

CALCULATIONS

The Gladstone–Dale relation shows the relationship between density (ρ) and refractive index (n) for binary liquid solutions which is given as follows [35]:

$$n - 1 = B\rho \quad (2)$$

In Equation (2), B is the Gladstone–Dale constant. Equation 3 shows the relationship between variations of ρ and n when molar concentration of solute, c , varies in binary liquid solution:

$$\frac{dn}{dc} = B \frac{d\rho}{dc} \quad (3)$$

We assume that some non-ionic solute molecules are dissolved in a solvent (water) and a binary aqueous solution, with molar concentration c , is provided.

Table1: The experimental (from Equation (11)) and theoretical (from Equation (13)) values of K_{ref} for aqueous solutions of some alcohols and polyols at constant temperature and atmospheric pressure.

$c(\text{mol.L}^{-1})$	$K_{ref(\text{exp})}(\text{L/mol})$	$K_{ref(\text{cal})}(\text{L/mol})$	%RD (K_{ref})
Glycerol ^a (at 293.15 K)			
0.109	0.0110		3.6000
0.327	0.0107		0.9346
0.548	0.0106		0.0000
1.109	0.0106		0.0000
1.800	0.0106	0.0106	0.0000
2.752	0.0107		0.9346
4.771	0.0107		0.9346
6.385	0.0107		0.9346
8.680	0.0106		0.0000
10.498	0.0105		1.9048
D-Fructose ^a (at 293.15 K)			
0.112	0.0250		3.2000
0.283	0.0254		1.5748
0.517	0.0253		1.9763
1.072	0.0253		1.9763
1.600	0.0253	0.0258	1.9763
2.164	0.0251		2.7888
2.613	0.0250		3.2000
2.925	0.0250		3.2000
3.247	0.0250		3.2000
D-Glucose ^a (at 293.15 K)			
0.112	0.0250		3.2000
0.516	0.0256		0.7813
1.070	0.0255	0.0258	1.1765
2.014	0.0253		1.9763
3.071	0.0251		2.7888
4.081	0.0250		3.2000
D-Mannitol ^a (at 293.15 K)			
0.110	0.0264		1.1364
0.279	0.0262		0.3817
0.509	0.0260	0.0261	0.0000
0.626	0.0257		1.5564
0.746	0.0256		1.9531
0.867	0.0256		1.9531
Pentaerythritol ^b (at 298.15 K)			
0.3708	0.0175	0.0176	0.5714

Table1: The experimental (from Equation (11)) and theoretical (from Equation (13)) values of K_{ref} for aqueous solutions of some alcohols and polyols at constant temperature and atmospheric pressure.

$c(\text{mol.L}^{-1})$	$K_{ref(\text{exp})}(\text{L/mol})$	$K_{ref(\text{cal})}(\text{L/mol})$	%RD (K_{ref})
Pentaerythritol ^b (at 313.15 K)			
1.2838	0.0176	0.0176	0.0000
Ethanol ^a (at 293.15 K)			
0.6460	0.0028		32.1429
1.4980	0.0029		27.5862
2.9670	0.0032	0.0037	15.6250
5.0180	0.0033		12.1212
9.9190	0.0029		27.5862
Urea ^a (at 293.15 K)			
0.5020	0.0084		28.5714
1.5310	0.0086		30.2326
3.1390	0.0086	0.0060	30.2326
5.0140	0.0085		29.4118
6.1850	0.0084		28.5714
6.5840	0.0084		28.5714
Lactic acid ^a (at 293.15 K)			
0.111	0.0091		13.1868
0.560	0.0089		15.7303
1.017	0.0088	0.0103	17.0455
2.076	0.0087		18.3908
3.305	0.0086		19.7674
5.377	0.0085		21.176
Ethylene glycol ^a (at 293.15 K)			
0.161	0.0056		8.9286
0.484	0.0058		5.1724
0.972	0.0058	0.0061	5.1724
1.464	0.0058		5.1724
1.959	0.0058		5.1724
Citric acid ^a (at 293.15 K)			
0.105	0.0248		15.3226
0.320	0.0241		18.6722
0.541	0.0238	0.0286	20.1681
0.771	0.0239		19.6653
1.130	0.0237		20.6751
1.508	0.0235		21.7021

Table1: The experimental (from Equation (11)) and theoretical (from Equation (13)) values of K_{ref} for aqueous solutions of some alcohols and polyols at constant temperature and atmospheric pressure.

$c(\text{mol.L}^{-1})$	$K_{ref(\text{exp})}(\text{L/mol})$	$K_{ref(\text{cal})}(\text{L/mol})$	%RD (K_{ref})
1,3-Propanediol ^c (at 298.15 K)			
1.3188	0.0078		2.5641
2.6569	0.0080	0.0080	0.0000
5.3950	0.0081		1.2346
8.1987	0.0080		0.0000
1-Amino-2-propanol ^d (at 298.15 K)			
2.3961	0.0099		20.2020
3.1819	0.0100	0.0079	21.0000
4.1810	0.0102		22.5490
6.8122	0.0102		22.5490
Acetic acid ^a (at 293.15 K)			
0.1660	0.0042		38.0952
0.5010	0.0044		31.8182
1.0060	0.0043	0.0058	34.8837
2.0280	0.0042		38.0952
3.4140	0.0042		38.0952
5.1800	0.0040		45.0000
butane-1, 3-diol ^e (at 298.15 K)			
1.0282	0.0098	0.0100	2.0408
2.3244	0.0099		1.0101
butane-1, 4-diol ^f (at 298.15 K)			
2.3239	0.0100	0.0100	0.0000
6.2848	0.0099		1.0101
butane-2, 3-diol ^e (at 298.15 K)			
2.3348	0.0104	0.0100	3.8462
6.3085	0.0108		7.4074
1-Propanol ^a (at 293.15 K)			
0.4960	0.0054		3.7037
0.9870	0.0056		0.0000
1.9580	0.0057	0.0056	1.7544
2.9110	0.0056		0.0000
3.2230	0.0056		0.0000

Table1: The experimental (from Equation (11)) and theoretical (from Equation (13)) values of K_{ref} for aqueous solutions of some alcohols and polyols at constant temperature and atmospheric pressure.

$c(\text{mol.L}^{-1})$	$K_{ref(\text{exp})}(\text{L/mol})$	$K_{ref(\text{cal})}(\text{L/mol})$	%RD (K_{ref})
Acetone ^a (at 293.15 K)			
0.172	0.0041		29.2683
0.513	0.0043		23.2558
0.853	0.0042	0.0053	26.1905
1.191	0.0043		23.2558
1.360	0.0043		23.2558
1.696	0.0042		26.1905
Maltose ^a (at 293.15 K)			
0.119	0.0496		21.1694
0.303	0.0502		19.7211
0.497	0.0499	0.0601	20.4409
0.700	0.0497		20.9256
0.914	0.0498		20.6827
2.068	0.0493		21.9067

a) Ref [38]; b) Ref [58]; c) Ref [59]; d) Ref [60]; e) Ref [61]

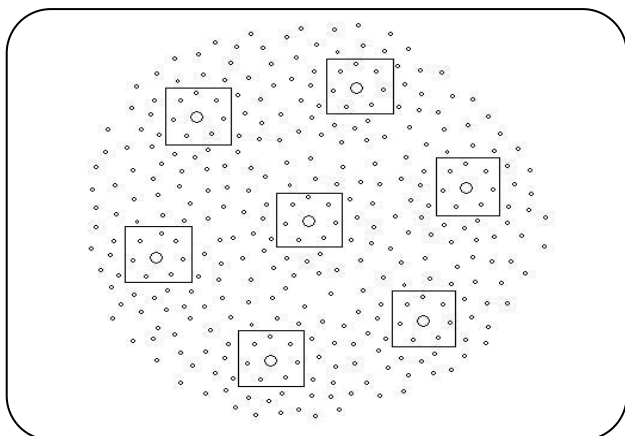


Fig. 1: The microscopic structure of the liquid solution. In this figure, the bigger molecules are solute and smaller molecules are solvent.

The microscopic structure of this binary aqueous solution can be seen in Fig. 1.

It is clear that in aqueous solutions, $c_o = 0$. Therefore:

$$dc = c - c_o = c \quad (4)$$

On the other hand, we can write:

$$\frac{d\rho}{dc} = \frac{\rho - \rho_o}{c} = \frac{\Delta\rho}{c} \quad (5)$$

$$\frac{dn}{dc} = \frac{n - n_o}{c} = \frac{\Delta n}{c} \quad (6)$$

In Equation (5), ρ and ρ_o are the density of binary aqueous solution and solvent (water), respectively. Also, in Equation (6), n and n_o are the refractive index of binary aqueous solution and solvent (water), respectively.

Using Equations (5) and (6), Equation (3) can be rewritten as Equation (7):

$$\frac{n - n_o}{c} = B \frac{\rho - \rho_o}{c} \quad (7)$$

It can be seen in some research works and literature [26,30,36-38] that for binary aqueous solutions, there is a linear relationship between density and concentration of solutes, especially in low concentrations, as below:

$$\rho = kc + \rho_o \quad (8)$$

In Equation (7), k is a constant. Equation (7) can be rewritten as below:

$$\frac{\rho - \rho_o}{c} = k \quad (9)$$

Equation (10) can be obtained by combining Equations (7) and (9):

$$\frac{n - n_0}{c} = Bk \quad (10)$$

In Equation (10), Bk is a constant that can be named K_{ref} . Therefore, Equation (10) can be presented as Equation (11):

$$n_{(T,P)} = K_{\text{ref}}c + n_0 \quad (11)$$

In Equation (11), $n_{(T,P)}$ is the refractive index of binary aqueous solution at given constant temperature and pressure, c is the molar concentration of this aqueous solution, and n_0 is the refractive index of solvent (water) at given constant temperature and pressure.

Equation (11) can be successfully applied in an aqueous solution of polyols and disaccharides, especially in low concentrations. However, as can be seen in Table 2 and Fig. 2, Equation (11) can also be applied in an aqueous solution of other solutes in higher molar concentration [13, 14, 38-44].

In Equation (11), for binary aqueous solutions, the molal concentration (instead of molar concentration) can be applied in low concentrations because in a specified Molar Concentration Range (MCR), the molality very closes to molarity. This molar concentration range varies in different solutes. In Table 3, these Molar Concentration Ranges (MCR) are listed for 48 solutes (acids, bases, alcohols, polyols, monosaccharide, disaccharide, and salts). As it can be seen in this table, among these solutes, the shortest molar concentration range belongs to sucrose ($0 \leq c \leq 0.10$).

Equation (11) can be expanded for ternary and quaternary liquid systems as below:

$$n_{(T,P)} = \sum_{i=1}^3 c_i K_{\text{ref}(i)} + n_0 \quad (12)$$

In Equation (12), the value of i for binary, ternary, and the quaternary aqueous solution is 1, 2, and 3, respectively. n_0 is the refractive index of the solvent at a given constant temperature.

We can use Equation (11) to obtain K_{ref} for binary aqueous solutions of various components such as alkane polyols, monosaccharides, organic molecules and acids, and ionic molecules. The values of K_{ref} which are obtained from Equation 11 can be named experimental K_{ref} [$K_{\text{ref}(\text{exp})}$].

On the other hand, we have chosen glycerol as a basic molecule and investigated on the K_{ref} of this molecule. Glycerol has 3 hydroxyl groups and generates hydrogen bonds with water molecules in aqueous solutions. We suggested the below semi-empirical equation to calculate the value of K_{ref} for glycerol, in aqueous solution, at atmospheric pressure and constant temperature (293.15 K):

$$K_{\text{ref}} = \left[M_{w(\text{solute})} - 18.01 \right] \times \left[0.0000052N_{(-O-)} + 0.000128 \right] \quad (13)$$

In Equation (13), M_w is the molar mass of solute and $N_{(-O-)}$ is the number of hydroxyl groups and/or oxygen atoms that are existed in the solute molecule. It is well known that in alkane polyols and monosaccharide, the oxygen atom of the solute molecule generates hydrogen bond with the water molecule.

We examined Equation (13) for some linear alkane polyols ($n = 2$ to 6 $-OH_s$) and polyols with one ring (monosaccharide) and we have found that this equation can be successfully applied for calculating K_{ref} of these molecules (please see Table 1). The K_{ref} which is obtained from Equation 13 can be named theoretical K_{ref} [$K_{\text{ref}(\text{calc})}$].

RESULTS AND DISCUSSION

As it has been mentioned in the introduction section [26-34], some researchers introduced equations that typically show the relationship between refractive indices of solutions and their concentrations. There are three important differences between our model and the models which have been reported in other literature. First, in most, this literature, the square, cube, and/or more complex equations have been suggested to predict/estimate values of refractive indices of liquids and solutions while in this article, for linear alkane polyols ($n = 2$ to 6 $-OH_s$) and polyols with one ring (monosaccharide), we suggested a simple linear equation with high accuracy (please see Table 1: %RD for calculation of K_{ref} are around 2% or less than %2) to predict values of the refractive index of solutions. Second, there is a constant, K_{ref} , in our linear equation that we suggested another semi-empirical equation (Equation (13)) for calculation of this constant but in other literature, we couldn't observe any equation for calculation of constants. Third, in other literature, the constants of

Table 2: The relationship between refractive index, n , and molar concentration, c , for aqueous solutions of various solutes (species) at $T = 293.15\text{ K}^a$.

Specie	M_w (g/mol)	Equation (n vs. c) ^b	R^2	Molar concentration range
Alkanols and Alkane polyols				
Methanol (CH ₃ OH)	32.04	$y = 0.0009x + 1.3328$	0.9986	0.156 – 10.580
Ethanol (C ₂ H ₅ OH)	46.07	$y = 0.0033x + 1.3327$	0.9986	0.216 – 7.370
1-Propanol (C ₃ H ₈ O)	60.10	$y = 0.0055x + 1.3331$	0.9992	0.166 – 3.838
Ethylene glycol (C ₂ H ₆ O ₂)	62.07	$y = 0.0059x + 1.3329$	0.9999	0.161 – 8.212
Glycerol (C ₃ H ₈ O ₃)	92.09	$y = 0.0107x + 1.3330$	1.0000	0.109 – 6.385
D-Glucose (C ₆ H ₁₂ O ₆)	180.16	$y = 0.0250x + 1.3333$	0.9999	0.056 – 4.081
D-Fructose (C ₆ H ₁₂ O ₆)	180.16	$y = 0.0251x + 1.3331$	1.0000	0.056 – 2.767
D-Mannitol (C ₆ H ₁₄ O ₆)	182.17	$y = 0.0254x + 1.3332$	0.9999	0.055 – 0.867
Lactose (C ₁₂ H ₂₂ O ₁₁)	342.30	$y = 0.0508x + 1.3330$	0.9999	0.029 – 0.565
Sucrose (C ₁₂ H ₂₂ O ₁₁)	342.30	$y = 0.0483x + 1.3332$	1.0000	0.059 – 2.255
Maltose (C ₁₂ H ₂₂ O ₁₁)	342.30	$y = 0.0493x + 1.3332$	1.0000	0.059 – 2.253
Organic acids				
Formic acid (CH ₂ O ₂)	46.03	$y = 0.0024x + 1.3332$	0.9983	0.109 – 4.074
Acetic acid (CH ₃ COOH)	60.05	$y = 0.0041x + 1.3332$	0.9996	0.166 – 4.470
Lactic acid (C ₃ H ₆ O ₃)	90.08	$y = 0.0085x + 1.3333$	0.9999	0.111 – 5.377
Oxalic acid (C ₂ H ₂ O ₄)	90.03	$y = 0.0095x + 1.3331$	0.9995	0.056 – 0.920
Citric acid (C ₆ H ₈ O ₇)	192.12	$y = 0.0233x + 1.3333$	1.0000	0.211 – 1.639
Organic solutes				
Ammonia (NH ₃)	17.03	$y = 0.0009x + 1.3329$	0.9994	0.292 – 5.622
Acetone (C ₃ H ₆ O)	58.08	$y = 0.0043x + 1.3330$	0.9999	0.172 – 1.696
Urea (CH ₄ N ₂ O)	60.06	$y = 0.0084x + 1.3333$	0.9999	0.167 – 8.234
Ionic solutes				
Sodium hydroxide (NaOH)	39.99	$y = 0.0099x + 1.3334$	0.9989	0.125 – 2.174
Sodium chloride (NaCl)	58.44	$y = 0.0092x + 1.3335$	0.9992	0.172 – 3.056
Sodium acetate (CH ₃ COONa)	82.03	$y = 0.0103x + 1.3335$	0.9992	0.122 – 2.993
Sodium nitrate (NaNO ₃)	84.99	$y = 0.0085x + 1.3335$	0.9989	0.118 – 2.689
Sodium bromide (NaBr)	102.89	$y = 0.0133x + 1.3334$	0.9998	0.197 – 2.837
Potassium hydroxide (KOH)	56.11	$y = 0.0097x + 1.3335$	0.9990	0.179 – 1.938
Potassium chloride (KCl)	74.55	$y = 0.0091x + 1.3334$	0.9993	0.135 – 2.701
Potassium nitrate (KNO ₃)	101.1032	$y = 0.0085x + 1.3333$	0.9991	0.099 – 2.240
Potassium bromide (KBr)	119.00	$y = 0.0130x + 1.3334$	0.9997	0.084 – 2.914
Potassium iodide (KI)	166.00	$y = 0.0207x + 1.3334$	1.0000	0.122 – 3.134
Hydrochloric acid (HCl)	36.46	$y = 0.0079x + 1.3333$	0.9996	0.137 – 4.099
Lithium chloride (LiCl)	42.39	$y = 0.0082x + 1.3335$	0.9995	0.118 – 4.118
Ammonium chloride (NH ₄ Cl)	53.491	$y = 0.0097x + 1.3333$	0.9997	0.187 – 3.128
Cesium Chloride (CsCl)	168.36	$y = 0.0121x + 1.3334$	0.9996	0.120 – 3.877
Nitric acid (HNO ₃)	63.01	$y = 0.0074x + 1.3334$	0.9996	0.159 – 3.937
Silver nitrate (AgNO ₃)	169.87	$y = 0.0168x + 1.3334$	0.9998	0.120 – 2.758
Manganese(II) sulfate (MnSO ₄)	151.00	$y = 0.0240x + 1.3336$	0.9996	0.135 – 1.063
Copper sulfate (CuSO ₄)	159.61	$y = 0.0265x + 1.3335$	0.9993	0.063 – 1.013

a) Ref [38] ; b) Eq. [24]

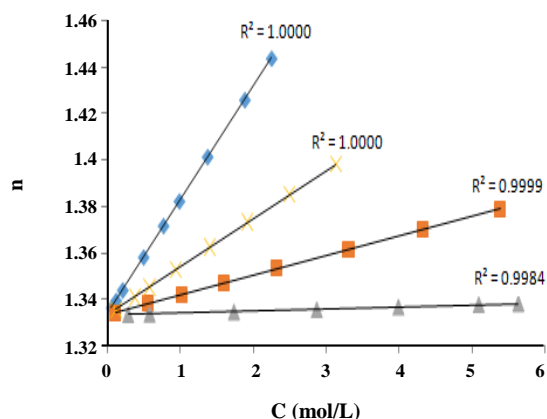


Fig. 2: The refractive index, n , versus molar concentration of solute, c , for binary mixtures of (♦): [water (1) + maltose (2)], (×): [water (1) + potassium iodide (2)], (■): [water (1) + lactic acid (2)], and (▲): [water (1) + ammonia (2)] at $T = 293.15$ K.

equations don't give us any information about the power of interactions between species in solutions while the values of K_{ref} can help us to find out the power of interactions between solute and solvent molecules. The higher value of K_{ref} shows the stronger interaction between the solute and solvent molecule. Therefore, for different liquid and especially aqueous solutions, we can study on power of interactions between solvent and solute molecules using the values of their K_{ref} . Of course, it should be noted that for better comparison between the power of solute-solvent interactions, the values of K_{ref} must be measured and considered in the same concentrations (preferably in low concentrations). Depend on the nature of molecules, in higher concentrations, the value of experimental K_{ref} decreases. In aqueous solutions, by increasing concentration, the solute-solute interactions increase and consequently, the interactions between solute and solvent molecules decrease.

The values of experimental and theoretical K_{ref} for aqueous solutions of some alkane polyols and monosaccharides are listed in Table 1. It can be seen in this table that there is a good agreement between experimental and theoretical K_{ref} for these alkane polyols (the Relative Deviation, RD , is less than %2 in most cases). It indicates that Equations 11 and 13 can be used for calculation of the refractive index of alkane polyols ($n = 2$ to 6 -OHs) and monosaccharides, especially in low concentrations (please see Table 2).

When light passes through a medium (like aqueous solution), the interactions occur between photons

(from light) and electrical and magnetic fields which are generated by solute and solvent molecules. The strength of these interactions depends on the strength of electrical and magnetic fields that are generated by components in aqueous solution. Of course, the sum of electrical, and or magnetic, fields generated by solute and solvent molecules interact with photons.

In aqueous solutions of polyols and monosaccharides, there are interactions between dipoles-dipoles, induced dipoles-induced dipoles (London dispersion force), and dipoles-induced dipoles [45,46]. London interactions generate the fluctuating electric fields and these fields affect the beam of light passing through the solution. These effects reduce the velocity of light and increase the value of the refractive index of the solution.

The energy of interaction between two uncharged molecules (A and B) can be approximated using the London formula as the below [47]:

$$E_{AB} \approx \left(-\frac{3}{2}\right) \left(\frac{I_A I_B}{I_A + I_B}\right) \left(\frac{\alpha_A \alpha_B}{r^6}\right) \quad (14)$$

Where I am the first ionization potentials of the molecule, α is the polarizability, and r is the distance between molecule A and molecule B. In alkane polyols and monosaccharide, the value of polarizability can be increased by increasing number of electrons and protons (molar mass) of solute. Also, the formula of alkanols is $R(OH)$. In these molecules, the -R group has an electron donor effect on the oxygen atom of the hydroxyl agent. It is well known that the electron donor effect increases by increasing the mass of -R group. Therefore, the oxygen atom in the heavier molecule is more negative and can generate a stronger hydrogen bond with water molecules (please see K_{ref} for ethanol and 1-propanol in Table 1).

Equation 13 shows that for alkane polyols and monosaccharides, in aqueous solution, the molar mass of solute, M_w , (please see Table 2) and several hydroxyl agents (-OH) and/or oxygen atoms are important factors that can be effective on the refractive index of these solutions.

It must be noted that for calculating values of K_{ref} for solutes that have nitrogen and fluorine, $N_{(-O)}$ is the number of oxygen atoms plus a number of these elements (N and F). Various solute molecules can be classified into three groups regarding Equation 13.

Table 3: Molar concentration ranges of solutes, Δc , for aqueous solutions that in this ranges, the molality is equal to molarity^a.

Species	Δc (mol.L ⁻¹)	Species	Δc (mol.L ⁻¹)	Species	Δc (mol.L ⁻¹)
acetic acid	$0 \leq c \leq 0.501$	copper(II) sulfate	$0 \leq c \leq 1.360$	lactic acid	$0 \leq c \leq 0.325$
acetone	$0 \leq c \leq 0.172$	ethanol	$0 \leq c \leq 0.432$	lactose	$0 \leq c \leq 0.11$
ammonia	$0 \leq c \leq 1.162$	ethylene glycol	$0 \leq c \leq 0.322$	Lithium chloride	$0 \leq c \leq 0.964$
ammonium chloride	$0 \leq c \leq 0.187$	iron(III) chloride	$0 \leq c \leq 0.388$	magnesium chloride	$0 \leq c \leq 0.895$
ammonium sulfate	$0 \leq c \leq 0.309$	formic acid	$0 \leq c \leq 0.436$	magnesium sulfate	$0 \leq c \leq 1.788$
barium chloride	$0 \leq c \leq 0.641$	D-fructose	$0 \leq c \leq 0.168$	maltose	$0 \leq c \leq 0.110$
calcium chloride	$0 \leq c \leq 0.768$	D-glucose	$0 \leq c \leq 0.168$	manganese(II) sulfate	$0 \leq c \leq 1.424$
cesium chloride	$0 \leq c \leq 0.573$	glycerol	$0 \leq c \leq 0.30$	potassium iodide	$0 \leq c \leq 0.350$
potassium bicarbonate	$0 \leq c \leq 0.510$	oxalic acid	$0 \leq c \leq 0.281$	potassium nitrate	$0 \leq c \leq 0.405$
potassium bromide	$0 \leq c \leq 0.526$	citric acid	$0 \leq c \leq 0.158$	phosphoric acid	$0 \leq c \leq 0.416$
potassium carbonate	$0 \leq c \leq 0.789$	nitric acid	$0 \leq c \leq 0.648$	hydrogen chloride	$0 \leq c \leq 1.11$
potassium chloride	$0 \leq c \leq 0.691$	D-manitol	$0 \leq c \leq 0.150$	potassium hydroxide	$0 \leq c \leq 1.729$
potassium permanganate	$0 \leq c \leq 0.327$	methanol	$0 \leq c \leq 0.621$	1-propanol	$0 \leq c \leq 0.166$
silver nitrate	$0 \leq c \leq 0.641$	sodium acetate	$0 \leq c \leq 0.371$	sodium bromide	$0 \leq c \leq 0.610$
sodium chloride	$0 \leq c \leq 0.885$	sodium nitrate	$0 \leq c \leq 0.607$	sodium phosphate	$0 \leq c \leq 0.535$
sucrose	$0 \leq c \leq 0.10$	urea	$0 \leq c \leq 0.450$	sulfuric acid	$0 \leq c \leq 0.526$

a) Ref [38]

Group 1: the solute molecules which follow Equation (13) [$K_{\text{ref(cal)}} = K_{\text{exp(exp)}}$]. These molecules (such as glycerol, propanediol, butane-1, 3-diol, butane-1, 4-diol, pentaerythritol, D-fructose, D-glucose, D-mannitol) have two or more hydroxyl agents and/or oxygen atoms and also, have R- group and/or groups with two or more carbon atoms (C_3, C_4, C_5, \dots). These R- groups have a good electron donor effect.

Group 2: the solute molecules which have a negative deviation from Equation (13). For these molecules, the value of experimental K_{ref} is less than calculated K_{ref} . These molecules have R- the group with small size, electron acceptor agents, and also, low number of hydroxyl agents (such as methanol and ethanol). Of course, ethylene glycol has a negative deviation from Eq.(13), but in a low amount. We can put the molecules with carbonyl agents (such as acetone, acetic acid, and citric acid) in group 2. Also, the molecules which have -R group with seven or more carbons and also, disaccharides (like maltose) can be classified as group 2 (please see Table 1).

Group 3: the solute molecules which have a positive deviation from Equation (13) (such as 1-amino-2-propanol and urea). For these molecules, the value of experimental K_{ref} is more than calculated K_{ref} . In this group, molecules have agents such as $-\text{NH}_2$. The nitrogen atom, an amino

group, has free electron pair and free electrons, compared to bonding electrons, generate a stronger magnetic field.

It must be taken into account in Eq.(13), N(-O-) can be several groups and/or atoms of solute molecules that can generate hydrogen bond with solvent molecules.

Application of Equations (11), (12), and (13)

There are some equations to fit refractive indices data of aqueous solutions but Equations (11) and (12) are important for fitting refractive indices data of aqueous solutions because the parameters of these equations (K_{ref}) help us to a better understanding of interactions between solvent and solute molecules [14]. The interaction between solute and solvent molecules increases as the value of K_{ref} increases.

Determination of refractive index of aqueous solutions

For aqueous solutions of alkane polyols ($n = 2$ to 6 -OH,s) and monosaccharides, we can calculate the value of K_{ref} for solutes by Equation (13) and then calculate refractive indices at various molar concentrations by Equations (11) or (12) with high accuracy (%RD < 0.15). Please see Table 4. Using Equations (13), (11) or (12), there is no need to refractometer for determination of refractive indices of the mentioned aqueous solutions.

Determination of molar concentration of aqueous solutions

For aqueous solutions of alkane polyols [R-(OH)_n, n = 2 to 6 -OHs] and monosaccharides, with having theoretical K_{ref} (obtained by Equation 13) and experimental refractive index, we can determine molar concentration using the following equation:

$$c = \frac{n_{exp} - n_0}{K_{ref(calcul)}} \quad (15)$$

Where, $K_{ref(calcul)}$ is calculated using Equation (13) and n_{exp} and n_0 are the experimental refractive index of aqueous solution and solvent, respectively. As can be seen in Table 4, Equation 15 can be used for the determination of the molar concentration of alkane polyols (n = 2 to 6 -OH_s) and monosaccharides aqueous solutions. The values of %RD for these determinations are less than 2. Equation 15 gives good results (uncertainty of ±0.001 to ±0.005) especially in low and moderate concentration (between 0.1 and 1 mol/L). For these solutions, the refractive index can be a good tool for the determination of concentration.

Study on interaction between solute and solvent molecules

The parameter K_{ref} (from Equation (13)) can show the strength of the interaction between solvent and solute molecules.

The -R group in alkane polyols, [R(OH)_n], have an electron donor effect on the oxygen atom of hydroxyl agents, and this effect increases by increasing the mass of -R group (or mass of solute molecules). In these molecules, the negative charge (-δ) of the oxygen atom (from hydroxyl agent) increases as the molar mass of solute increases. It means that in alkane polyols and monosaccharides, the strength of hydrogen bond which is formed between solute and solvent molecules, $HB_{(solu-solv)}$, increases by increasing the molar mass of solutes. Therefore, in an aqueous solution of alkane polyols and monosaccharides:

$$(\text{Strength of } HB_{(solu-solv)}) \propto (\text{molecular weight of solute}) \quad (16)$$

Two subjects should be considered when the number of hydroxyl agents, $N_{(-O-)}$, increases in alkane polyols and monosaccharides. The first subject, the number of $HB_{(solu-solv)}$ increases when the number of hydroxyl agents, in various solute molecules, increases. It should be noted that the electron donor effect of -R groups (C₂H_n, C₃H_n, C₄H_n, etc) is more than the electron donor effect of the hydrogen atom.

Therefore, in an aqueous solution of alkane polyols and monosaccharides, the hydrogen bond which is formed between solute and solvent molecules, $HB_{(solu-solv)}$, is stronger than hydrogen bond between solvent molecules, $HB_{(solv-solv)}$.

The second subject, in alkane polyols, the mass of -R (solute molar mass) mostly increases with increasing of the number of hydroxyl agents.

We can reach to the below proportionality with considering two above subjects:

$$(\text{Strength of } HB_{(solu-solv)}) \propto (N_{(-O-)}) \quad (17)$$

The proportionality 18 can be obtained by combination proportionalities 16 and 17:

$$(\text{Strength of } HB_{(solu-solv)}) \propto (M_{W(Solute)} \text{ and } N_{(-O-)}) \quad (18)$$

Equation (13) shows that K_{ref} depends on $M_{W(solute)}$ and $N_{(-O-)}$. The $K_{ref,s}$ for 1-propanol, 1,3 propandiol, glycerol, and also, ethanol and ethylene glycol show that the value of K_{ref} increases (about 0.0026) when the one hydroxyl group is inserted to -R group.

Comparison of Equation (13) and proportionality (18) results to the below:

$$(K_{ref}) \propto (\text{strength of } HB_{(solu-solv)}) \quad (19)$$

Proportionality (19) shows that in an aqueous solution of alkane polyols, the value of K_{ref} can indicate the strength of hydrogen bond which is formed between solute and solvent molecules, $HB_{(solu-solv)}$. The strength of $HB_{(solu-solv)}$ increases as the value of K_{ref} increases.

The K_{ref} can be used in industrial and research fields. It is well known that the H-bondings play an important role in many organic components and their solutions which are used in various industries such as pharmacy [48-50], food [51,52], detergents and cleaners [53,54].

In research works, $HB_{(solu-solv)}$ is a good tool for investigating interactions between molecules in aqueous solutions which have hydroxyl and/or carboxyl agents [14,55,56]. Of course, in research work, a nonclassical HB (hydrogen bonding involving a B-H...π interaction) was described [57].

As it was mentioned, in aqueous solutions, the K_{ref} shows the strength of $HB_{(solu-solv)}$ and it can be used in research works. For example, we can investigate positive and negative deviations from Equation (13).

Table 4: The values of calculated molar concentration (by Equation (15)) also, experimental and calculated refractive indices (by Equation (11)), n , for aqueous solutions of some polyol(s) in various molar concentration.

$c_{\text{calcul}}(\text{mol/L})^c$	$c_{\text{exp}}(\text{mol/L})$	n_{exp}	n_{calc}^b	RD (%) for n	RD (%) for c
Water + Glycerol (at 293.15 K)					
0.330	0.327 ^a	1.3365 ^a	1.3365	0.0000	0.9174
0.774	0.771 ^a	1.3412 ^a	1.3412	0.0000	0.3891
1.113	1.109 ^a	1.3448 ^a	1.3448	0.0000	0.3607
2.047	2.035 ^a	1.3547 ^a	1.3546	0.0074	0.5897
3.264	3.242 ^a	1.3676 ^a	1.3674	0.0146	0.6786
5.349	5.300 ^a	1.3897 ^a	1.3892	0.0360	0.9245
Water +D-mannitol (at 293.15 K)					
0.111	0.110 ^a	1.3359 ^a	1.3359	0.0000	0.9091
0.226	0.222 ^a	1.3389 ^a	1.3388	0.0075	1.8018
0.448	0.451 ^a	1.3447 ^a	1.3448	0.0074	0.6652
0.563	0.567 ^a	1.3477 ^a	1.3478	0.0074	0.7055
0.674	0.686 ^a	1.3506 ^a	1.3509	0.0222	1.7493
0.789	0.806 ^a	1.3536 ^a	1.3540	0.0300	2.1092
Water + 1-propanol (at 293.15 K)					
0.482	0.496 ^a	1.3357 ^a	1.3358	0.0075	2.8226
0.982	0.987 ^a	1.3385 ^a	1.3385	0.0000	0.5069
1.500	1.474 ^a	1.3414 ^a	1.3413	0.0075	1.7639
2.625	2.595 ^a	1.3477 ^a	1.3475	0.0148	1.1561
3.214	3.223 ^a	1.3510 ^a	1.3510	0.0000	0.2792
3.732	3.838 ^a	1.3539 ^a	1.3545	0.0443	2.7619
Water + D-glucose (at 293.15 K)					
0.221	0.225 ^a	1.3387 ^a	1.3388	0.0075	1.7778
0.512	0.516 ^a	1.3462 ^a	1.3463	0.0074	0.7752
0.698	0.697 ^a	1.3508 ^a	1.3510	0.0148	0.1435
0.934	0.944 ^a	1.3571 ^a	1.3574	0.0221	1.0593
1.060	1.070 ^a	1.3603 ^a	1.3606	0.0221	0.9346
1.310	1.329 ^a	1.3668 ^a	1.3673	0.0366	1.4296

Table 4: The values of calculated molar concentration (by Equation (15)) also, experimental and calculated refractive indices (by Equation (11)), n , for aqueous solutions of some polyol(s) in various molar concentration.

$c_{\text{calcul}}(\text{mol/L})^c$	$c_{\text{exp}}(\text{mol/L})$	n_{exp}	n_{calc}^b	RD (%) for n	RD (%) for c
Water + D-Fructose (at 293.15 K)					
0.279	0.283 ^a	1.3402 ^a	1.3403	0.0075	1.4134
0.508	0.517 ^a	1.3461 ^a	1.3464	0.0223	1.7408
0.806	0.820 ^a	1.3538 ^a	1.3542	0.0295	1.7073
1.178	1.201 ^a	1.3634 ^a	1.3640	0.0440	1.9151
1.566	1.600 ^a	1.3734 ^a	1.3743	0.0655	2.1250
1.833	1.878 ^a	1.3803 ^a	1.3815	0.0869	2.3962
1,3-Propandiol (at 298.15 K)					
1.2875	1.3188 ^d	1.3428 ^d	1.3431	0.0223	2.3734
2.6500	2.6569 ^d	1.3537 ^d	1.3538	0.0074	0.2597
5.4375	5.3950 ^d	1.3760 ^d	1.3757	0.0218	0.7878
8.2250	8.1987 ^d	1.3983 ^d	1.3981	0.0143	0.3208
butane-1, 3-diol (at 298.15 K)					
1.010	1.0282 ^e	1.3430 ^e	1.3432	0.0149	1.7701
2.290	2.3244 ^e	1.3558 ^e	1.3561	0.0221	1.4800
butane-1, 4-diol (at 298.15 K)					
2.340	2.3239 ^e	1.3563 ^e	1.3561	0.0148	0.6928
6.230	6.2848 ^e	1.3952 ^e	1.3957	0.0358	0.8719
Pentaerythritol (at 298.15 K)					
0.364	0.3708 ^f	1.3390 ^f	1.3391	0.0075	1.8339

a) Ref [38]; b) Eq. 26; c) Eq. 30; d) Ref [59]; e) Ref [61]; f) Ref [58]

As can be seen in Table 1, 1-amino-2-propanol has positive deviation from Equation 13 ($K_{\text{ref}(\text{exp})} > K_{\text{ref}(\text{calc})}$). The molecular structure of 1-amino-2-propanol is similar to 1,2-propandiol but there is a difference between structures of these two molecules. 1,2-propandiol has two hydroxyl agents while 1-amino-2-propanol has one hydroxyl and one amine agent. The oxygen atom (in hydroxyl agent) is more electronegative than nitrogen atom (in amine agent). It means that the free electron pair in the amine group is more available (for production of hydrogen bonding) than that of hydroxyl agent. Therefore, the strength of $\text{HB}[(\text{H})_2\text{N}\cdots\text{H-O-H}]$ is more than $\text{HB}[\text{R}(\text{H})\text{O}\cdots\text{H-O-H}]$. It can be the reason for this positive deviation. This deviation is +0.002 for 1-amino-2-propanol.

Also, the area has a positive deviation (+0.0024) from Equation 13 (please see Table 1). This positive deviation

can be due to the existing two amine groups.

On the other hand, Table 1 shows that some solute molecules have a negative deviation from Equation (13) ($K_{\text{ref}(\text{exp})} < K_{\text{ref}(\text{calc})}$). It can be due to having agents which generate $\text{HB}_{(\text{solv-solu})}$ with less strength than hydroxyl agent.

It can be observed in Table 1 that acetone, citric acid, and ethanol have a notable negative deviation from Equation (13). This deviation for acetone is about - 0.001 and it can be due to the existing carbonyl group ($-\text{C}=\text{O}$). In aqueous solutions, hydroxyl (from solute molecules) can form three hydrogen bonds (two $\text{O}_{\text{solute}}\cdots\text{H}_{\text{solvent}}$ and one $\text{H}_{\text{solute}}\cdots\text{O}_{\text{solvent}}$) while carbonyl (from solute molecules) can form two hydrogen bonds (two $\text{O}_{\text{solute}}\cdots\text{H}_{\text{solvent}}$). Therefore, the power of the bond between the carbonyl group and water can be less than that of between hydroxyl group and the water molecule.

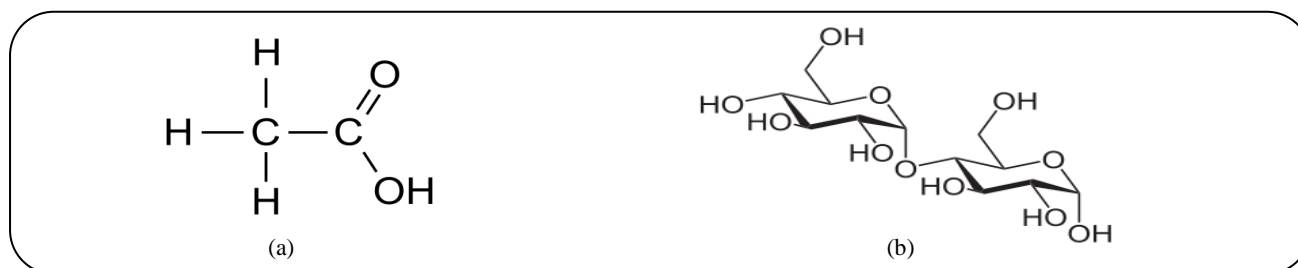


Fig. 3: The molecular structures of acetic acid (a) and maltose (b).

We can see in Table 1 that the acetic acid (Fig. 3a) has a negative deviation from Equation (13) (-0.0016). It can be due to the existing one carbonyl group that is attached to hydroxyl agent (-COOH). It appears that 0.0015 negative deviation (from Equation (13)) occurs per each acidic group (-COOH). Citric acid has three acidic groups and shows -0.0045 deviations from Equation (13). Also, lactic acid has one acidic group and shows about -0.0015 deviation from Equation (13) (please see Table 1). Of course, these are weak acids and it guesses that the deviation from Equation (13) is too high for strong acids.

Also, there is a negative deviation (from Equation (13)) for maltose. The value of this deviation is about -0.01 (Please see Table 1). As can be seen in Fig. 3b, the molecule of maltose has 8 hydroxyl groups as well as 3 oxygen atoms. In this molecule, the intramolecular hydrogen bonds can be formed between various hydrogen and oxygen atoms of maltose. For a solute molecule, the number of hydrogen bonds between solute and solvent molecules can decrease with an increasing number of intramolecular hydrogen bonds.

The relative deviation (*RD*) for refractive indices, concentration, and K_{ref} can be calculated by:

$$\%RD = \frac{X_{\text{experimental}} - X_{\text{calculated}}}{X_{\text{experimental}}} \times 100 \quad \% \quad (20)$$

The acceptable range of relative deviation for high accuracy experiments can be ($0 \leq \%RD \leq 2$).

It must be taken into account in Eq. (13), $N(-O-)$ can be the number of groups and/or atoms of solute molecules that can generate hydrogen bond with solvent molecules.

CONCLUSIONS

In this research work, we used a linear equation for calculation of refractive indices for alkane polyols $[R(OH)_n, n = 2 \text{ to } 6 \text{ -OH,s}]$, and polyols with one ring (monosaccharides) at constant temperature and pressure. The constant of this equation, K_{ref} , can show the power

of interaction between solute and solvent molecules. Two important cases must be noted for better comparison between K_{ref} of different molecules. First, this comparison must be done in a homologous series of chemicals. Second, this comparison must be done in the same concentration of solutes (for instance, in 0.5 or 1 mol/L). Other components show negative or positive deviation from this equation (Equation (13)). Components with negative deviation have agents which generate weaker $HB_{(solute-solv)}$ than hydroxyl group. Components with positive deviation have agents which generate stronger $HB_{(solute-solv)}$ than hydroxyl group. This information can help us to a better understanding of interaction between solutes and solvents in solutions.

Nomenclature

n	Refractive index, -
ρ	Density, kg/m^3
K_{ref}	-, L/mol
M_w	Molar mass, kg/mol
c	Molar concentration, mol/L
T	Temperature, K

Received : Dec. 19, 2017 ; Accepted : Jul. 30, 2018

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