Solvation of Parachloro Meta Xylenol (PCMX) in Alcohols-Aqueous Solutions at 293.15 K

Farid I. El-Dossoki*, E. Abou El-Hasan, Eman G. Abdelrhman

Chemistry Department, Faculty of Science, Port-Said University, Port-Said, Egypt

Abstract: *The densities and the refractive indices of ethanol, methanol and glycerol mixtures with water by ratios of (0.00-0.5104 mole fraction of alcohols) were measured at 293.15 °K. The densities, molal solubility and refractive indices of PCMX in the ethanol, methanol and glycerol–water mixtures by ratios of (0.00-0.5104 mole fraction of alcohols) were measured at 293.15 °K. The apparent molal volume (V_Q), the Wander Wall volume (V_w), the electrostriction volume (V_e) and the solvated radius (r) of PCMX in the above solvent mixtures were calculated and discussed. Also, the molar refraction, the polarizability and the atomic polarization of PCMX in the ethanol, methanol and glycerol–water mixtures by ratios of (0.00-0.5104 mole fraction of alcohols) were measured at 293.15 °K were calculated and discussed. The result indicate that, as the concentrations of ethanol and methanol in alcohols–water mixtures increase, the densities of solvent ($d_\circ$) and PCMX solutions (d) decrease and the refractive indices in both solvent and solution mixtures increase. It also noted that, increase of concentrations of glycerol–water mixtures resulted in increase in the refractive indices and density of the solvents and solutions. The spectra of PCMX in ethanol-water mixtures by ratios of (0.00-0.5104 mole fraction of alcohols) were measured at 293.15 °K. The spectra show a hyper chromic effect as the percentage of ethanol increased.*

Keywords: *Polarizability; Molal volume; Refractive index.*

1. INTRODUCTION

Microorganisms are one of the main sources that have caused food borne diseases and spoilage of products[1]. Some studies have unequivocally demonstrated that processing equipment and the environment can be sources of microbial contamination of food Products[2]. Microbial contamination on environmental surfaces may be transferred the food products directly through surface contact or by vectors such as personnel, pests, air movement or cleaning regimes. Antimicrobial activities process validation is defined as establishing documented evidence that a disinfection process will consistently remove or inactivate known or possible pathogens from inanimate objects.

Parachlorometaxylenole (PCMX) (Dettol) is the name of a commercial liquid disinfectant and as well as antiseptic belonging to a product line of house hold products. The key ingredient which defines its unique antimicrobial property is an aromatic chemical compound known as Chloroxylenol ($C_8H_9 C_{10}$) [3]. This makes up 4.8% of Dettol (PCMX)'s total mixture, with the rest composed of pine oil, isopropanol, castor oil soap, caramel, and water. Because several of the ingredients are insoluble in water, Dettol produces a white emulsion of oil droplets when diluted during use. Apart from its low toxicity and low metal corrosively, it is also relatively cheap and available compare to other disinfectants. Many studies have been reported to explain the solvation process using the apparent molal and refractive index measurements [4-25].

The present work aims to study density, solvation and refractive index of PCMX in alcohols (ethanol, methanol and glycerol)–water mixtures with different ratios (0.00-0.5104 mole fraction of alcohols) at 293.15 °K. Also the work aim to study the spectra of PCMX under the effect of the solvent on at 293.15 °K.

2. EXPERIMENTAL

2.1. Materials and Solutions

Ethanol (C_2H_5OH) (95%), Methanol (CH_3OH) (95%) and Glycerol ($C_3H_8O_3$) (99.5%) are from ELNASR Pharmaceutical chemicals Co.(Egypt). PCMX (Scheme 1) was from Royal Co., Egypt.Alcohols-water mixtures of different ratios (0-0.5104mole fraction of alcohols) were prepared using bidistilled water. Saturated solutions of PCMX were prepared in alcohols-water mixtures of different ratios (0.00-0.5104 mole fraction of alcohols) at 293.15 °K.
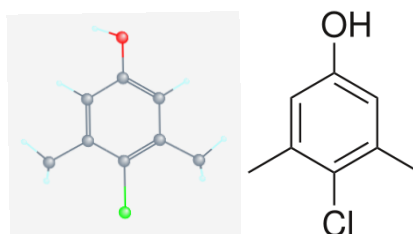
(Scheme 1)

2.2. Apparatus and Procedure

Spectrophotometer of a type Jenway was used to measure the spectra of PCMX and Abe refractometer was used to measure the refractive index (±0.0001) of the solvents and the solutions at 293.15 K. Digital balance of four decimal was used to measure the weight. Ultrathermostate of type Kottermann 4130 was used to adjust the temperature degree at the select degree (±0.01 ºK).After, Alcohols-water mixtures of different ratios (0.0-0.5104 mole fraction of alcohols) were prepared, the density and refractive indices were measured. The density was measured by weighting 1 ml of solvent (±0.0001). Saturated solutions of PCMX were prepared in alcohols-water mixtures of different ratios (0.00-0.5104 mole fraction of alcohols) at 293.15 ºK by adding excess PCMX solid compound for 2 weeks till complete saturation and equilibrium stablishing. After that, filtration was done and one ml of clear filtrate was taken and weight (d value) then evaporated till constant weight.

3. RESULTS AND DISCUSSION

3.1. Solubility Measurements

The density of methanol, ethanol and glycerol-water mixtures of a different ratios (0.00-0.5104 mole fraction of alcohols) and of PCMX in methanol, ethanol and glycerol-water mixtures of a different ratios (0.00-0.5104 mole fraction of alcohols) were measured as explained in the experimental section. The density values were tabulated in Tables 1-3 and represented in Figures 1-3.

Table1. *The density of the solvent mixtures, $d_\circ$, and the density, d, the molality, m, the solvated radius, r , the apparent molal volume, V_Q the Wander Wall volume, V_w the electrostriction volume, V_e and the solvation free energy change (ΔG_s) of the PCMX, in different methanol mole fractions in water mixtures at 293.15 K and Pressure (p = 0.1 MPa)[a]*

Methanol Vol. %	X_1	$d_\circ$ g/cm^3	d g/cm^3	M mol/kg solvent	ΔG_s kJ/mol	r cm	V_Q cm^3/mol	V_w cm^3/mol	V_e cm^3/mol
0.00	0.0000	0.99823	1.00691	0.00448	13.1766	3.336	155.529	102.800	-52.723
0.10	0.0473	0.97790	1.04342	0.00144	15.9419	3.297	150.086	99.207	-50.879
0.20	0.1005	0.95698	1.03381	0.00267	14.4376	3.307	151.479	100.130	-51.351
0.30	0.1607	0.93636	1.01563	0.00409	13.3985	3.327	154.182	101.920	-52.269
0.40	0.2295	0.91574	1.00891	0.00558	12.6416	3.334	155.218	102.600	-52.619
0.50	0.3088	0.89512	1.00791	0.00663	12.2215	3.335	155.371	102.700	-52.670
0.70	0.5104	0.85387	0.98887	0.01144	10.8924	3.357	158.360	104.680	-53.684

[a] *Relative standard uncertainty in the solubilitiesu$_r$(m) = 1.05 %, Relative standard uncertainty in the density u$_r$ (ρ) = 0.04 %.*

Table2. *The density of the solvent mixtures, $d_\circ$, and the density, d, the molality, m, the solvated radius, r , the apparent molal volume, V_Q the Wander Wall volume, V_w the electrostriction volume, V_e and the solvation free energy change (ΔG_s) of the PCMX, in different ethanol mole fractions in water mixtures at 293.15 K and Pressure (p = 0.1 MPa)[a]*

Ethanol Vol. %	X_1	$d_\circ$ g/cm^3	d g/cm^3	M mol/kg solvent	ΔG_s kJ/mol	r cm	V_Q cm^3/mol	V_w cm^3/mol	V_e cm^3/mol
0.00	0.0000	0.99825	1.0069	0.00448	13.1766	3.345	156.776	103.630	-53.147
0.10	0.0332	0.98189	0.98259	0.03908	7.8993	3.464	159.386	105.351	-54.032
0.20	0.0718	0.96865	0.96945	0.06730	6.5749	3.379	161.546	106.780	-54.764
0.30	0.1171	0.95383	0.95473	0.10182	5.5661	3.396	164.037	108.431	-55.608
0.40	0.1710	0.93519	0.93799	0.13799	4.8255	3.416	166.964	110.362	-56.601
0.50	0.2362	0.91385	0.98925	0.21981	3.6912	3.356	158.295	104.632	-53.662
0.70	0.4192	0.86767	1.10290	0.31129	2.8434	3.236	141.906	93.800	-48.106

[a] *Relative standard uncertainty in the solubilitiesu$_r$(m) = 1.05 %, Relative standard uncertainty in the density u$_r$ (ρ) = 0.04 %.*
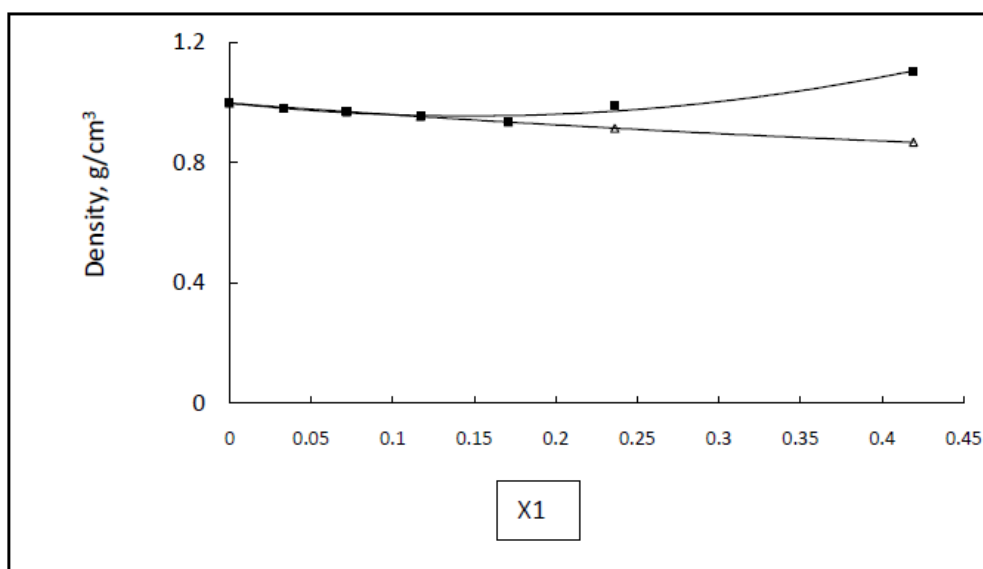
Table3. *The density of the solvent mixtures, d∘, and the density, d, the molality, m, the solvated radius, r , the apparent molal volume, V_Q the Wander Wall volume, V_w , the electrostriction volume, V_e and the solvation free energy change (ΔG_s) of the PCMX, in different glycerol mole fractions in water mixtures at 293.15 K and Pressure (p = 0.1 MPa)^a*

Glycerol Vol. %	X_1	d∘ g/cm^3	d g/cm^3	M mol/kg solvent	ΔG_s kJ/mol	r cm	V_Q cm^3/mol	V_w cm^3/mol	V_e cm^3/mol
0.00	0.0000	0.99823	1.00690	0.00448	13.1766	3.336	155.536	102.810	-52.721
0.10	0.0267	1.01734	1.03745	0.06314	6.7304	3.303	150.955	99.782	-51.174
0.20	0.0581	1.03572	1.04683	0.06843	6.5344	3.293	149.603	98.888	-50.715
0.30	0.0956	1.05592	1.07393	0.10885	5.4035	3.266	145.827	96.392	-49.435
0.40	0.1413	1.07402	1.09841	0.14447	4.7137	3.241	142.575	94.242	-48.333
0.50	0.1980	1.09797	1.13921	0.23977	3.4794	3.202	137.462	90.863	-46.599
0.70	0.3655	1.14145	1.20376	0.34851	2.5682	3.144	130.078	85.981	-44.096

^{a} *Relative standard uncertainty in the solubilities u_r(m) = 1.05 %, Relative standard uncertainty in the density u_r (ρ) = 0.04 %.*
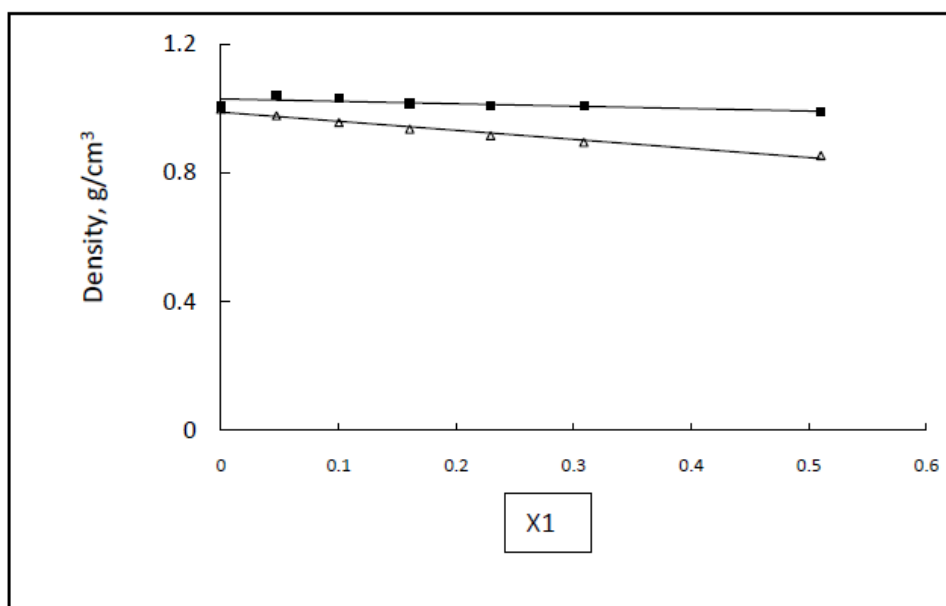
Figure1. *Change of the density, of the solvent mixtures (d˚)(Δ) and the PCMX solution (d) (■) with the mole fraction of methanol in ethanol-water mixtures at 293.15 K*

Figure2. *Change of the density, of the solvent mixtures (d˚)(Δ) and the PCMX solution (d)(■) with the mole fraction of ethanol in ethanol-water mixtures at 293.15 K*
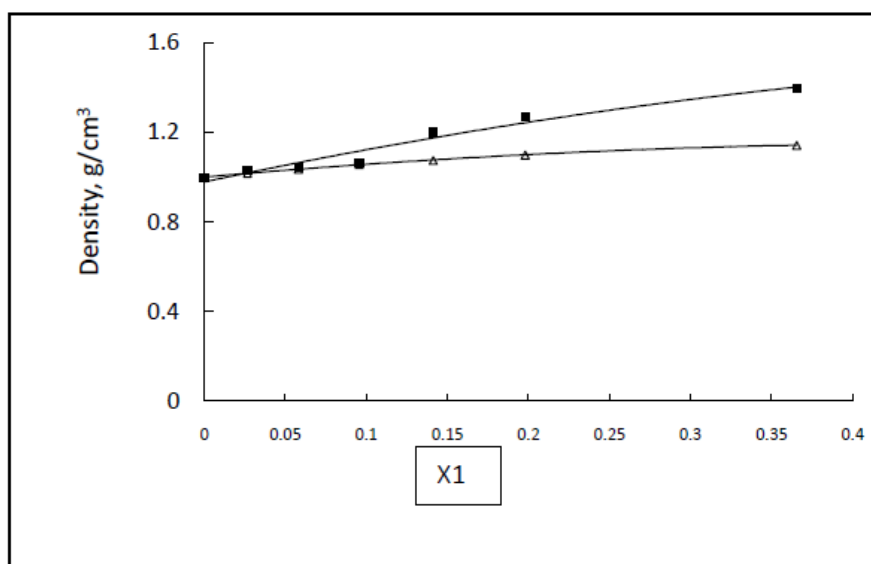
Figure3. *Change of the density, of the solvent mixtures (d˚)(Δ) and the PCMX solution (d) (■) with the mole fraction of glycerol in glycerol -water mixtures at 293.15 K*

The molal solubility of PCMX in methanol, ethanol and glycerol-water mixtures of different ratios(0.0-0.5104 mole fraction of alcohols)was measured as explained in the experimental section. The molalities were calculated applying the following equation:

$$m = w. 1000/ d_\circ M \qquad\qquad (1)$$

Where w is the weight of PCMX dissolved in one ml of solvent, $d_\circ$ is the density of solvent and M is the molecular weight of PCMX.

The free energy change of solvation (ΔG_s) of PCMX in methanol, ethanol and glycerol-water mixtures of different ratios (0.0-0.5104 mole fraction of alcohols) was calculated applying the following equation:

$$\Delta G_s = - 2.303 RT \log m \qquad\qquad (2)$$

From the values of the measured densities and molalities, the values of the solvated radius, the apparent molal volume, the Wander Wall volumes and the electrostriction volume of PCMX were calculated applying a computer program include the following equations:

The apparent molal volume (V_φ) was calculated applying the following equation [26]:

$$V_\varphi = \frac{M}{d} - \frac{d-d_\circ}{m\,d\,d_\circ} \qquad\qquad (3)$$

Where, m and Mare the molality and the molecular weight of PCMX respectively, while d is the density of PCMX solution respectively.

The packing density which isthe relation between the Van der Waals volume and the partial molal volume of the relatively large molecules, is found to be constant [27]. Therefore, it is possible to calculate the Van der Waals volumes (V_w) of the used substances by applying the following equation [27]:

$$\text{Packing density (P)} = V_w / V_\varphi = 0.661 \pm 0.017 \qquad\qquad (4)$$

The electrostriction volume (V_e) which is the volume compressed by the solvent was calculated applying the following equation [28]:

$$V_e = V_w - V_\varphi \qquad\qquad (5)$$

The values of the density of the solvent mixtures, $d_\circ$, the density of PCMX solutions, d, the molality, m, the solvated radius, r , the apparent molal volume, V_Q the Wander Wall volume, V_w the electrostriction volume, V_e and the solvation free energy change (ΔG_s) of the PCMX, in different methanol mole fractions in water mixtures at 293.15 K and Pressure (p = 0.1 MPa) were recorded in Tables 1-3.

As noted from the data in Tables (1-3), the density of the solvent decrease as the mole fraction of methanol and ethanol increase as a result of lower density of methanol and ethanol than that of water. On the other hand, the density of the solvent increase as the mole fraction of glycerol increase as a result of higher density of glycerol than that of water.

Two factors affect the density of PCMX solutions which are the density of the solvent and the solubility of PCMX. The density of the PCMX solution was found to increase as the mole fraction of glycerol increase as a result of the effect of both higher solubility of PCMX in glycerol than that in water and higher density of glycerol than that of water.On the other hand, the density of the PCMX solution decrease as the mole fraction of methanol decrease as a result of the effect of lower density of methanol than that of water. In ethanol-water solvent mixtures, the density of the PCMX solution was found to decrease then increase as the mole fraction of ethanol increase as a result of the competition of the effect of both the density and the solubility. In comparing the density of the solvent and that of the PCMX solutions in the same solvents, Figures (1-3), it was found that difference between the two densities increase as the mole fraction of the alcohols under study increase. This may be due to the effect of higher solubility of PCMX as the mole fraction of the alcohols under study increase.

Also, the data in Tables (1-3), show that is the molality increase as the mole fraction of methanol, ethanol and glycerol increase indicating higher solvation of PCMX as the mole fraction of methanol, ethanol and glycerol increase. In comparing the effect of the solvent on the molality of PCMX (Figure 4), it was found that is the molality change in the following order: Glycerol-water > ethanol-water >methanol-water.
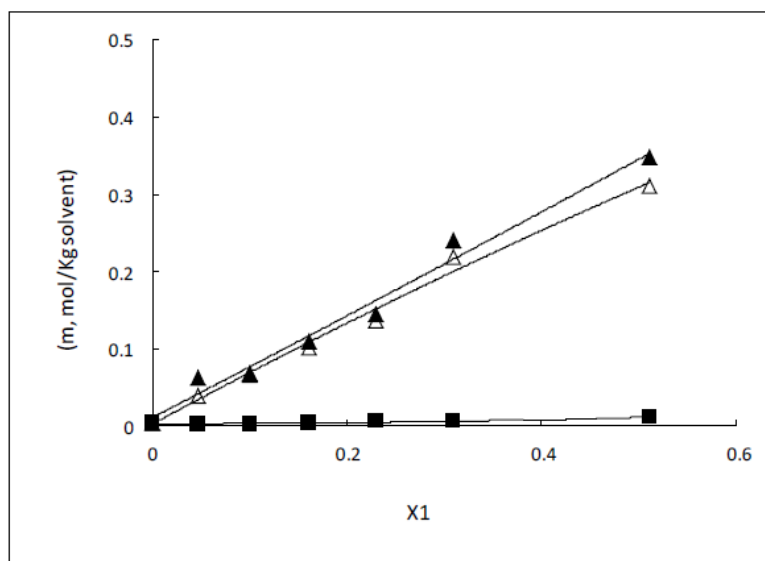
Figure4. *Change of the molality of PCMX with the mole fraction of solvent in glycerol –water (▲), ethanol–water (Δ), and methanol–water (■) mixtures at 293.15 K*

The apparent molal volume was found to increase as the mole fraction of methanol and ethanol increase and as the molality increase as a result of higher solvation of PCMX as the mole fraction of methanol and ethanol increase. On the other hand, the apparent molal volume was found to decrease as the mole fraction of glycerol increase, which may be due to the increase of both the density and solvation of PCMX as the mole fraction of glycerol increase. The change in the electrostriction volume was found to be higher in glycerol-water than that in ethanol-water mixtures, indicating higher solvation of PCMX in glycerol-water than that in ethanol-water mixtures.

The solvation process of PCMX in methanol, ethanol and glycerol-water mixtures of different ratios (0.0-0.5104 mole fraction of alcohols) is a non-spontaneous process as indicated from the positive sign of the free energy change of solvation (ΔG_s).

3.2. Refractive Index Measurements

The refractive indices of methanol, ethanol and glycerol-water mixtures of a different ratios (0.00-0.5104 mole fraction of alcohols) and of PCMX saturated solutions in methanol, ethanol and glycerol-water mixtures of a different ratios (0.00-0.5104 mole fraction of alcohols) were measured as explained in the experimental section.

From the values of the measured refractive indicesand apparent the molal volume, the values of the molar refraction (R), the polarizability (α), and the atomic polarization (P$_A$)of PCMX were calculated applying a computer program include the following equations:

The molar refraction was calculated [29] as follow:

$$R = \frac{n^2 - 1}{n^2 + 2} VQ = P_A + P_E = P_D = P_T \quad \text{................}$$ (6)

Where, (n) is the refractive indices and (V$_Q$) is the molal volume of PCMX solutions. The right hand side of equation (5) is equal to the total molar polarization or the distortion polarization which equal to the summation of both the electron polarization (P$_E$) and the atomic polarization (P$_A$) .The atomic polarization (P$_A$) were calculated[30] from the following equation:

$$P_A = 1.05 \, n^2 \quad \text{.................}$$ (7)

The mean value of the molecular dipole polarizability (α ; dipole moment induced by electric field) can be calculated from the optical refractive index (*n*) of a material containing *N* molecules per unit volume. The refractive index is related to the polarizability of the molecules by Lorenz-Lorenz formulaas shown in the following equation:

$$\frac{n^2 - 1}{n^2 + 2} = \frac{4 \pi \bar{n} \alpha}{3} \quad \text{......................}$$ (8)

where $\bar{n} = \dfrac{N}{V}$, (*N*) is the Avogadro's number and (V$_Q$) is the molal volume..

The solvated radii of PCMX were calculated by considering spherical form of the solvated moleculesapplying the following equation:

$$V_Q = (4/3) \, \pi \, r^3 \quad \text{..........................}$$ (9)

The values of the measured refractive indices, the molar refractions,the atomic polarization, and the polarizabilities, were recorded in Tables (4-6) and represented in Figures 5-7.

Table4. *The refractive indices of the solvent mixtures, n$_s$, and the refractive indices, n, polarizability, α, the molar refraction, R and the atomic polarization, P$_A$ of the PCMX, in different methanol mole fractions in water mixtures at 293.15 Kand Pressure (p = 0.1 MPa)^a*

Methanol Vol. %	X$_1$	n$_s$	n	αx10^{-23}	R	P$_A$
0.00	0.0000	1.33303	1.33310	1.279	32.279	1.865
0.10	0.0473	1.33602	1.33620	1.234	31.135	1.874
0.20	0.1005	1.33831	1.33840	1.253	31.607	1.881
0.30	0.1607	1.34151	1.34157	1.286	32.444	1.889
0.40	0.2295	1.34361	1.34401	1.303	32.868	1.896
0.50	0.3088	1.34370	1.34800	1.318	33.237	1.908
0.70	0.5104	1.40391	1.37237	1.427	35.988	1.977

^{a}*Relative standard uncertainty in the refractive indices u$_r$ (n) = 0.007 %.*

Table5. *The refractive indices of the solvent mixtures, n$_s$, and the refractive indices, n, polarizability, α, the molar refraction, R and the atomic polarization, P$_A$ of the PCMX, in different ethanol mole fractions in water mixtures at 293.15 K and Pressure (p = 0.1 MPa)^a*

| Ethanol Vol. %	X$_1$	n$_s$	n	αx10^{-23}	R	P$_A$
0.00	0.0000	1.33303	1.33310	1.279	32.279	1.865
0.10	0.0332	1.33904	1.34609	1.346	33.939	1.902
0.20	0.0718	1.34609	1.34609	1.364	34.404	1.902
0.30	0.1171	1.35515	1.34609	1.385	34.939	1.902
0.40	0.1710	1.35917	1.35011	1.425	35.935	1.913
0.50	0.2362	1.36622	1.36822	1.413	35.644	1.965
0.70	0.4192	1.36622	1.40241	1.368	34.512	2.065

^{a}*Relative standard uncertainty in the refractive indices u$_r$ (n) = 0.007 %.*

Table6. *The refractive indices of the solvent mixtures, $n_{\circ}$, and the refractive indices, n, polarizability, α, the molar refraction, R and the atomic polarization, P_A of the PCMX, in different glycerol mole fractions in water mixtures at 293.15 K and Pressure (p = 0.1 MPa)^a*

| Glycerol Vol. %	X_1	$n_{\circ}$	n	$\alpha \times 10^{-23}$	R	P_A
0.00	0.0000	1.33303	1.33310	1.2799	32.279	1.865
0.10	0.0267	1.34238	1.35117	1.3041	32.888	1.916
0.20	0.0581	1.35233	1.36219	1.3175	33.226	1.948
0.30	0.0956	1.36272	1.37326	1.3335	33.631	1.980
0.40	0.1413	1.37338	1.40648	1.2683	31.986	2.077
0.50	0.1980	1.38413	1.41748	1.223	30.844	2.109
0.70	0.3655	1.40555	1.43035	1.1378	28.695	2.148

^{a}*Relative standard uncertainty in the refractive indices u_r (n) = 0.007 %.*
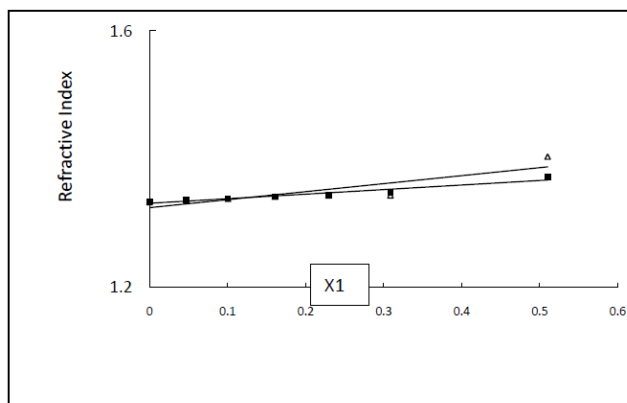
Figure5. *Change of the refractive index, of the solvent mixtures (n1) (Δ) and the PCMX solution (n2) (■) with the mole fraction of methanol in methanol-water mixtures at 293.15 K*
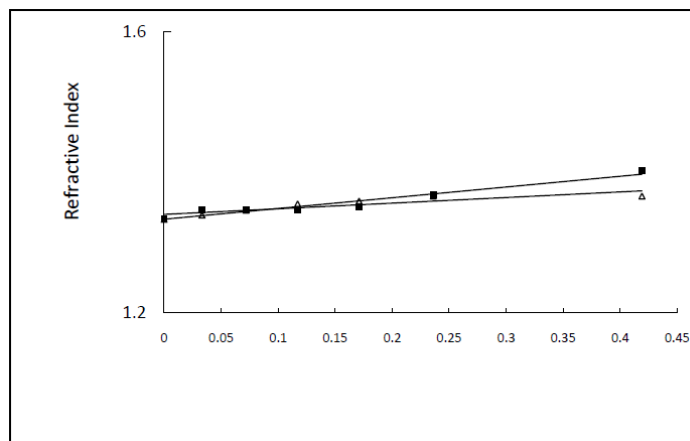
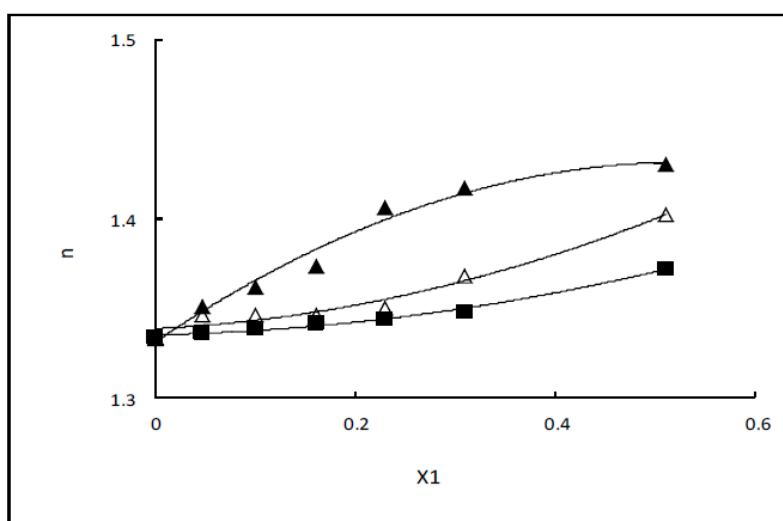**Figure6.** *Change of the refractive index, of the solvent mixtures (n1) (Δ) and the PCMX solution (n2) (■) with the mole fraction of ethanol in ethanol-water mixtures at 293.15 K*
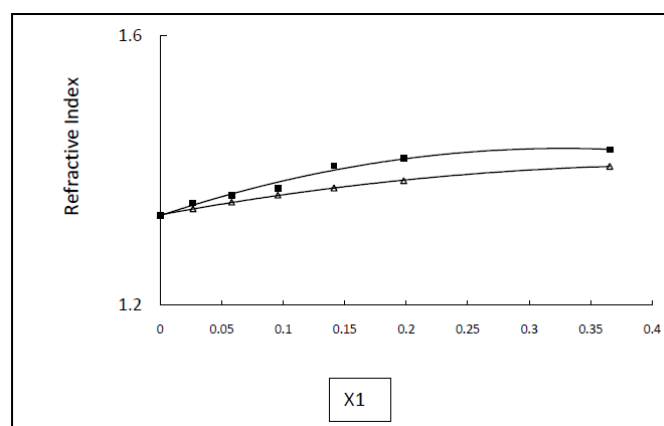
Figure7. *Change of the refractive index, of the solvent mixtures (n1) (Δ) and the PCMX solution (n2) (■) with the mole fraction of glycerol in glycerol -water mixtures at 280.15 K*

As noted from the data in Tables (4-6), and Figures (5-7), the refractive indices, polarizability, molar refraction and atomic polarization are increase as the mole fraction of methanol and ethanol increase. The refractive indices and atomic polarization are increase as the mole fraction of glycerol increase while the polarizability and the molar refraction decrease as the mole fraction of glycerol increase.The increase of the refractive indices, polarizability, molar refraction and atomic polarization as the mole fraction of methanol and ethanol increase may be due to the higher solubility of PCMX in methanol and ethanol than in water.The increase of the, refractive indicesand atomic polarization as the mole fraction of glycerol increase may be due to the higher solubility of PCMX in methanol than in water.

In comparing the refractive indices of the solvent and that the PCMX solutions in the same solvents, Figures (5-6), it was found that difference between the two refractive indices increase as the mole fraction of the alcohols under study increase. This may be due to the effect of higher solubility of PCMX as the mole fraction of the alcohols under study increase.

In comparing the effect of the solvent on the refractive indices of PCMX (Figure 8), it was found that the refractive indices change in the following order: Glycerol-water > ethanol -water > methanol – water.

Figure8. *Change of the refractive indices of PCMX with the mole fraction of solvent in glycerol –water (▲), ethanol–water (∆) and methanol–water (■) mixtures at 293.15 K*

### 3.3. Spectrophotometric Measurements

The spectra of 0.001M PCMX in different ethanol–water ratios mixtures (20-100% (v/v)) at 293.15 K, was measured. The wave length and the absorbance are represented in Figure 9. The results show that there is a small change in the wave length (in a range of 1 nm from 288-287 nm) in transferring from 20% to 100% ethanol-water mixtures. This means that a small blue shift was observed.
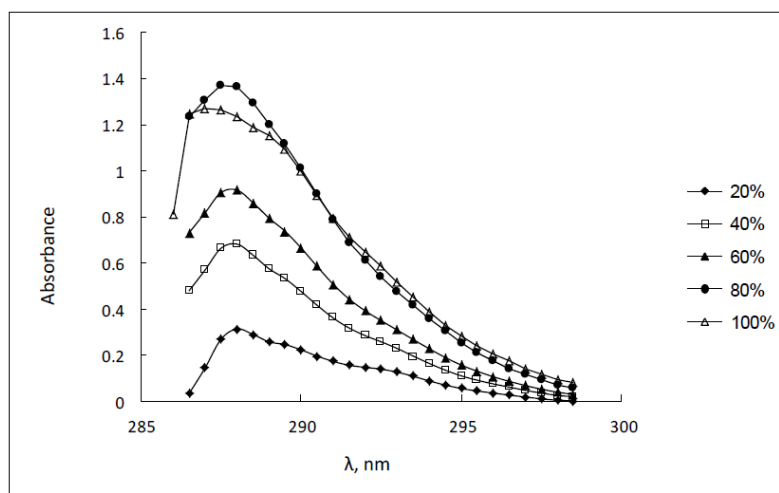

Figure9. *Spectra of 0.001 M PCMX in different ethanol–water ratios mixtures at 293.15 K*

On the other hand the results show that there is a high change in the absorbance (in a range of 0.313-1.275) in transferring from 20% t0 100% ethanol-water mixtures. An increase in the absorption of ultraviolet light by a solution is called a hyperchromic effect. This effect may be due to the disruption of the hydrogen bonds between the PCMX molecules (Scheme 2) as a result of the interaction with ethanol and water solvent molecules. The hydrogen bond limits the resonance of the aromatic ring so the absorbance of the sample is limited as well. The effect of solvent is to disruptthe hydrogen bonds between the PCMX molecules and so enhance the resonance in PCMX molecule between both the enol and keto forms as shown in Scheme 3.
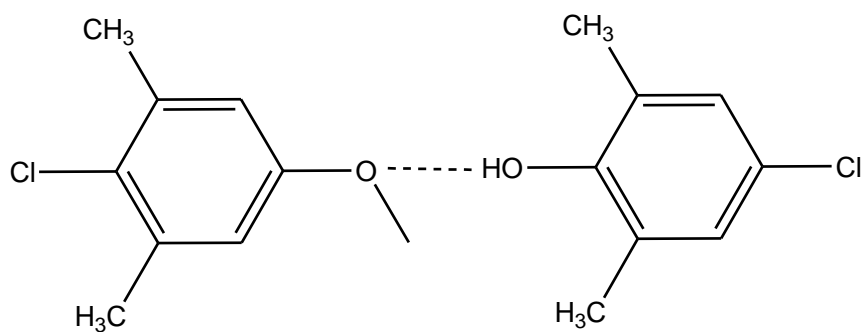
(Scheme 2)
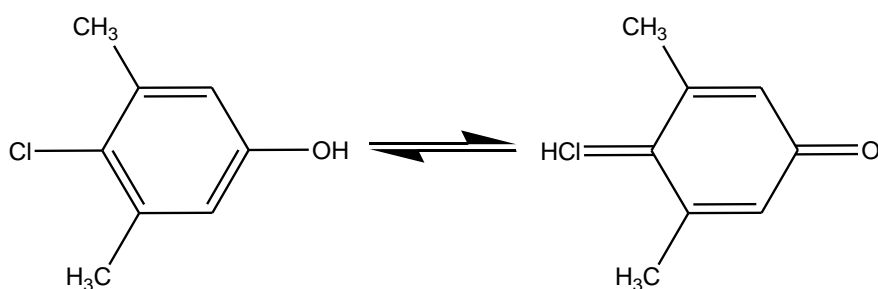
(Scheme 3)

REFERENCES

[1] R.A.N. Chmielewski, J.F. Frank,Comprehensive Review in food science and food safety. 2 **(2003)**22-32.

[2] I.C Blackman, J.F. Frank , Journal Of food Protection.59**(1996)** 827-831

[3] J. M. Ascenzi, Chloroxylenol: an old-new antimicrobial, Handbook of disinfections and antiseptics, New York, M. Dekker, **1996**.

[4] I.Cibulka, J. Chem. Thermodynamics, 80 **(2015)** 41–48.

[5] H.Wang, S.Zhang, J.Wang and Z.Yu, *J. Mol. Liq.,* 209 **(2015)** 563–568.

[6] R.Sadeghi and B.Goodarzi, *J. Mol. Liq.,* 141 **(2008)** 62–68.

[7] M.N.M. Al-Hayan and Maher A.M. Al-Bader, J. Chem. Thermodynamics **(2006).**

[8] H.Rodrıguez, A.Soto, A.Arce and M.K.Khoshkbarchi, *Journal of Solution Chemistry,* 32 (1),**(2003).**

[9] N.saha and B.das, *J. Mol. Liq.,* 89**(2000)**169-174.

[10] F.I.El-Dossoki, *Journal of the Chinese Chemical Society*, *54***(2007)**1129-1137.

[11] F. I. El-Dossoki and E. A.Gomaa, *J.Indian Chem. Soc.,* 82, **(2005)** 219-224.

[12] V.Tiwari and R.Pande, *J. Mol. Liq.,* 128 (1-3)**(2006)** 178-181.

[13] D.R.Delgado, G.A.Rodríguez and F. Martínez, *J. Mol. Liq.,* 177 **(2013)** 156–161.

[14] F. El-Dossoki, *Indian J. of Chem.,* 44A **(2005)** 1594-1596.

[15] E.A.Gomaa, M.M.Mostafa and F.I.El-Dossoki, *Al-Azhar Bull. Sci*. 14 (2) **(2003)** 97-107.

[16] F.I. El-Dossoki, *Egpt. J. Chem.*45 (3) **(2002)** 451-461.

[17] F. I. El-Dossoki, *International Research Journal of Pure & Applied Chemistry (IRJPAC)* 10 (3)**(2016)** 1-18.

[18] M.S. Calado, A. S.H. Branco, J.F. Diogo, j.M.N.A. Fareleira and Z. P. Visak, J. Chem. Thermodynamics, 80 **(2015)** 79–91.

[19] F. I. El-Dossoki, *Journal of Solution Chemistry*, 44 (2)**(2015)**264-279.

[20] V. Singh, P. K. Banipal, T.S. Banipal and R.L. Gardas, *J. Mol. Liq.,* 209 **(2015)** 352–357

[21] R. Kolhapurkar and k.Patil, *J. Mol. Liq.,* 178 **(2013)** 185–191.

[22] A. Bald, Z.Kinart and R.Tomaš, *J. Mol. Liq.,* 178 **(2013)** 94–98.

[23] D. Matkowska and T. Hofman, *J. Mol. Liq.,* 177 **(2013)** 301–305.

[24] B. Giner, C.Lafuente, A.Villares, M.Haro and M.C.Lopez, *J. Chem. Thermodynamics,* 39 **(2007)** 148–157.

[25] E. J. King, *J. Phys. Chem.,* 73 **(1961)** l220.

[26] R. Copal M. A. Siddiqui, *J. Phys. Chem.,* 73 **(1969)** 3390.

[27] G.J. Moody, J.D.R. Thomas , Dipole Moments in Inorganic Chemistry, Edward Arnold , Aberal, Great Britain **(1971).**

[28] E.R. Mongnaschi, L.M.Laboranti , *J.Chem . Soc. Faraday Trans* , 92 (18) **(1996)** 3367.

[29] J.B.Hasted , Aqueous Dielectrics, Chapman and Hall London, **(1973).**

[30] J.I. Kirn, *Z.Phys . Chem. Neue Folge*, 113 **(1978)** 129.