Diverse Interactions of N-Methyl Glycine in Aqueous Paracetamol Solution with the Manifestation of Solvation Consequences

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ARTICLE DETAILS

Article history:
Received 30 November 2018
Accepted 18 December 2018
Available online 05 January 2019

Keywords:
N-Methyl Glycine
Solute-Solvent Interactions
Paracetamol

ABSTRACT

The apparent molar volume ($\phi_0$-) and viscosity B-coefficient of N-methyl glycine of 0.01 M, 0.02 M and 0.03 M aqueous solutions have been estimated in presence of paracetamol at three temperatures namely 298.15 K, 303.15 K and 308.15 K from physicochemical study such as density ($\rho$), viscosity ($\eta$) and refractive index measurements and $^1$H NMR spectroscopy. The volumetric study was employed to evaluate limiting apparent molar volumes ($\phi_0$-) and experimental slopes ($S_\eta$) by using Masson equation for explaining solute–solute and solute–solvent interactions, respectively. The nature of group interactions between the solute, solvent and co-solute have been examined from limiting apparent molar volumes of transfer ($\Delta \phi_0$) values. The viscosity data were employed to determine viscosity A and B coefficients from Jones–Dole equation and the resulting parameters were used to examine the solute–solute and solute–solvent interactions in the solutions. Molar refraction values calculated from refractive indices by applying Lorentz–Lorenz equation were used to depict the intermolecular interactions between N-methyl glycine and paracetamol in their aqueous solution. However, the $^1$H NMR spectroscopy supports the existence of diverse interactions concretely.

1. Introduction

The native conformations of proteins depend on to several non-covalent interactions such as hydrogen bonding, electrostatic and hydrophobic interactions which may originate from surrounding solute and solvent molecules [1, 2]. So physicochemical properties of the proteins are influenced greatly by the presence of surrounding solute and solvent molecules. Physicochemical study of proteins provides many valuable information like hydration, solubility, stability and enzyme activity which are taking place in biochemical and physiological processes of living organism [3-5]. The nature of interaction of drug molecules with protein may also be understood from Physicochemical measurements.

World’s most popular and most commonly used analgesic and anti-inflammatory medicines from cradle to grave is paracetamol which is readily available and inexpensive also [6-8]. Chemical name of paracetamol is N-par-a-methyl aminophenol. It was introduced into the market by an analgesic and anti-inflammatory medicine by McNeil Laboratories mainly for children. After 1961 it became the most frequently sold analgesic medications. Its use as an analgesic is most tolerable than the other non-steroidal drugs (NSAIDs) which should not be used by the people with bronchial asthma, hemophilia, salicylate-sensitized people, peptic ulcer disease, pregnant or breastfeeding women and children under 12 years of age [9, 10]. Currently the use of aspirin as antipyretic and analgesic has been declined due some adverse effects and parallelly the use of paracetamol has been increased. Paracetamol has now been an appropriate analgesic for all age groups.

In continuance of our earlier works [11-15], we attempted to examine the nature of solute-solvent/co-solute interactions of N-methyl glycine in aqueous solutions of paracetamol at 298.15 K, 303.15 K and 308.15 K. The densities, viscosities and refractive indices of 0.01, 0.02 and 0.03 M aqueous N-methyl glycine solutions at 298.15 K, 303.15 K and 308.15 K are reported in Table 1 and densities, viscosities and refractive indices of aqueous N-methyl glycine solutions in presence of paracetamol at 298.15 K, 303.15 K and 308.15 K are reported in Table 2. From the volumetric measurements we calculated limiting apparent molar volume ($\phi_0$-), experimental slopes ($S_\eta$), transfer volume ($\Delta \phi_0$) and from the viscometric measurements we calculated viscosity A and B coefficients to analyse the nature of solute-solute/ co-solute interactions. The refractive index data helps to find the molar refraction (Rn) which also helps to elucidate the interaction between solute and co-solute in aqueous medium.

2. Experimental Methods

2.1 Source and Purity Samples

The studied N-methyl glycine and co-solute paracetamol of purist grade was purchased from Sigma-Aldrich, Germany and was used as purchased. The mass purity of salts was ±0.9%. The salts were dried from moisture at 353.15 K for 48 h, and then they were cooled and store in a desiccator prior to use.

2.2 Apparatus and Procedure

The density ($\rho$) measurements were done by vibrating-tube Anton Paar Density-Meter (DMA 4500M) with an accuracy of 0.00001 x 10⁻³ (kgm⁻³). The density meter was calibrated by using double-distilled water and dry air before taking the densities of our studied solutions [16]. The instrument has temperature monitoring system with the precision ±0.01 K.
The viscosity was determined using a Brookfield DV-III Ultra Programmable Rheometer having spindle size 42 fitted. The Rheometer was fitted with Digital Bath TC-500 which has a precision of ±0.0003 x 10⁻⁶ kg.

Mass measurements for preparation of stock solutions were done by Mettler AG-285 electronic balance with a precision of ±0.0003 x 10⁻³ kg.

Acceptable precautions were followed to minimize evaporation losses during the measurements.

Table 1. Experimental values of density (ρ), viscosity (η) and refractive index (n₀) of different molality of N-methyl glycine solution at 298.15 K, 303.15 K and 308.15 K.

| Concentration (M) | ρ/10³ (kg/m³) | η/mPas | n₀     |
|-------------------|--------------|--------|--------|
| 298.15 K          | 303.15 K     | 308.15 K |
| 0.01 M            |              |        |        |
| 0.009756          | 0.09621      | 0.09460 | 0.0956  | 0.080910 | 0.6604  | 1.3319 |
| 0.02 M            |              |        |        |
| 0.009782          | 0.09649      | 0.09486 | 0.0984  | 0.08116  | 0.6831  | 1.3325 |
| 0.03 M            |              |        |        |
| 0.009817          | 0.09679      | 0.09515 | 0.09016 | 0.01400  | 0.6859  | 1.3327 |

Table 2. Density (ρ) and viscosity (η) and refractive index (n₀) of different molality of aqueous N-methyl glycine in aqueous paracetamol solution at 298.15 K, 303.15 K and 308.15 K.

| Molality (Mol/kg) | ρ/10³ (kg/m³) | η/mPas | n₀     |
|-------------------|--------------|--------|--------|
| 298.15 K          |              |        |        |
| 0.01 M            |              |        |        |
| 0.9981            | 0.99666      | 0.99503 | 0.9053  | 0.8179  | 0.6882  | 1.3321 |
| 0.02 M            |              |        |        |
| 0.99836           | 0.99700      | 0.99538 | 0.9122  | 0.9242  | 0.6947  | 1.3323 |
| 0.03 M            |              |        |        |
| 0.99965           | 0.99772      | 0.99611 | 0.9264  | 0.9373  | 0.7076  | 1.3327 |
| 0.057             | 0.99444       | 0.99811 | 0.9434  | 0.9537  | 0.7231  | 1.3329 |
| 0.072             | 0.99998       | 0.99859 | 0.9594  | 0.8703  | 0.7386  | 1.3331 |
| 0.02 M            |              |        |        |
| 0.99824           | 0.996904     | 0.99527 | 0.9079  | 0.8024  | 0.6909  | 1.3327 |
| 0.023             | 0.99859       | 0.99725 | 0.99562 | 0.9148  | 0.9267  | 0.6974  | 1.3328 |
| 0.045             | 0.999295      | 0.99798 | 0.99635 | 0.9292  | 0.9408  | 0.7110  | 1.3313 |
| 0.057             | 0.99967       | 0.99838 | 0.99676 | 0.9464  | 0.9572  | 0.7262  | 1.3327 |
| 0.072             | 0.998014      | 0.99889 | 0.97263 | 0.9632  | 0.9738  | 0.7423  | 1.3345 |
| 0.03 M            |              |        |        |
| 0.99858           | 0.99791      | 0.99554 | 0.9111  | 0.9228  | 0.6939  | 1.3331 |
| 0.023             | 0.99892       | 0.99754 | 0.99588 | 0.9101  | 0.9297  | 0.7065  | 1.3332 |
| 0.045             | 0.99962       | 0.99829 | 0.99662 | 0.9334  | 0.9442  | 0.7152  | 1.3358 |
| 0.057             | 0.99999       | 0.99869 | 0.99703 | 0.9506  | 0.9608  | 0.7309  | 1.3372 |
| 0.072             | 1.00046       | 0.99921 | 0.99755 | 0.9674  | 0.9782  | 0.7468  | 1.3339 |

3. Results and Discussion

3.1 Density

Apparent molal volumes (φ₀) of N-methyl glycine in aqueous paracetamol solution were determined from the densities of the solution using the following equation [18].

φ₀ = M / ρ - 1000 (ρ - ρ₀) / (m/ρ₀)

where M is the molar mass of N-methyl glycine, ρ₀ and ρ is the density of solvent and solution respectively and M is the molality of the solution. The φ₀ values of N-methyl glycine in aqueous paracetamol solution at 298.15 K, 303.15 K and 308.15 K are shown in Tables 3-5 respectively.

Fig. 3. Molecular interactions between N-methyl glycine with paracetamol in aqueous medium.

The tendency indicates the presence of strong solute-solute interactions which increase with molarity of N-methyl glycine and temperatures. The interaction arises from the hydrophilic-hydrophilic group interaction between solute and co-solute molecules. The interaction of N-methyl glycine with paracetamol in aqueous medium is displayed in Fig. 3. With increasing temperature the secondary solvation layer is released into the bulk solvent leading to the expansion of solution. As a result, the φ₀ values of N-methyl glycine in aqueous paracetamol solutions increase with increase in temperature.

The parameter Sₚ defines the pair-wise interaction of solvated species in solution [20]. The Sₚ values of N-methyl glycine in aqueous paracetamol solution at different temperatures are reported in Table 4. The Sₚ values in our present study is least at 0.03 M N-methyl glycine at 308 K and highest in 0.01 M N-methyl glycine at 298 K. So, Sₚ values decrease with increasing temperature and molarity. This trend is exactly reverse than the φ₀ values explained earlier where φ₀ values increased with increasing temperature and molarity.

Fig. 4. Variation of limiting apparent molar volumes (φ₀) of N-methyl glycine solution in paracetamol solution at different temperatures.

https://doi.org/10.30799/jacs.199.18040403
with increasing concentrations of N-methyl glycine and temperatures. This weakening of $S_m^*$ values signify the presence of poor solute-solute interactions. The smaller $S_m^*$ values than the corresponding $\phi_0$ signifies that the solute-solute interaction is stronger than the solute-solute interaction.

Table 4 Limiting apparent molar volumes ($\phi_0$), experimental slopes ($S_m^*$), viscosity A, B-coefficients of aqueous N-methyl glycin solution in paracetamol at different temperatures

| Temp. [K] | $\phi_0 \times 10^3$ | $\Delta \phi_0 \times 10^3$ | $S_m^* \times 10^3$ | $B$ | $A$ |
|-----------|---------------------|--------------------------|-------------------|-----|-----|
| 298.15 K  | 0.00114             | 0.0021                  | 0.00114           | 0.00114 | 0.00114 |
| 303.15 K  | 0.00154             | 0.0025                  | 0.00154           | 0.00154 | 0.00154 |
| 308.15 K  | 0.00194             | 0.0030                  | 0.00194           | 0.00194 | 0.00194 |

The limiting apparent molar volume of transfer, $\Delta\phi_0$ for N-methyl glycin in paracetamol solution may be expressed as follows:

$\Delta\phi_0 = (\phi_0^\text{paracetamol}) - (\phi_0^\text{aqueous nicotinic acid solution})$

The variation of $\phi_0$ with temperature of N-methyl glycin in paracetamol solution follows the polynomial [21].

$\phi_0 = a_0 + a_1T + a_2T^2$

where $T$ is the temperature in K and $a_0$, $a_1$, and $a_2$ are the coefficients. Values of the coefficients of the above equation for N-methyl glycin in paracetamol solution are reported in Table 5.

The limiting apparent molar expansibilities ($\phi_0^E$) can be determined by the following equation [22].

$\phi_0^E = \frac{\Delta\phi_0}{S_m^*}$

The values of $\phi_0^E$ of N-methyl glycin in paracetamol solution at 298.15 K, 303.15 K and 308.15 K are evaluated and reported in Table 5.

The $S_m^*$ is not the only parameter for estimating the structure-making or breaking nature of any solute [30]. Hepler proposed a different technique to inspect the structure-making and breaking ability of the solute in aqueous solution from the following thermodynamic expression [23].

$S_m^* = \frac{\eta_0}{\rho_0} - \frac{\eta_2}{\rho_2} - \frac{\eta_3}{\rho_3} + \frac{\eta_4}{\rho_4}$

Table 5 Values of empirical coefficients ($a_0$, $a_1$, and $a_2$) of 0.01 M, 0.02 M and 0.03 M N-methyl glycin in paracetamol solution at 298.15 K, 303.15 K and 308.15 K

| Molality of N-methyl glycin | $a_0 \times 10^3$ | $a_1 \times 10^3$ | $a_2 \times 10^3$ | $\phi_0^E$ |
|----------------------------|-----------------|-----------------|-----------------|-----------|
| 0.01 M                     | 2.1618          | 14.59           | -0.2037         | -0.0474   |
| 0.02 M                     | -1951           | 13.19           | -0.2014         | -0.0428   |
| 0.03 M                     | -1473.2         | 9.266           | -0.1518         | -0.0316   |

The limiting apparent molar volume of transfer, $\Delta\phi_0$ for N-methyl glycin in paracetamol solution may be expressed as follows:

$\Delta\phi_0 = N\text{-methyl glycin} - N\text{-methyl glycin in paracetamol} - \phi_0^E$ [in water]

$\Delta\phi_0$ value provide the idea about the nature solute–solvent interactions. The limiting apparent molar volume of transfer may be analyzed in the light of co-sphere overlap model given by Friedman and Krishnan [26]. According to the model positive $\Delta\phi_0$ value signifies the existence of hydrophilic–hydrophilic, ion–ion and ion–ion interactions, whereas the negative $\Delta\phi_0$ value signifies the hydrophobic–hydrophobic interactions [27, 28]. The interactions between N-methyl glycine and paracetamol in aqueous medium may be of following categories.

1. Ionic–ionic interaction of the H+ ion of water and N-methyl glycin with the -COO ion of N-methyl glycin
2. H-bond between -COOH (N-methyl glycin) and -OH (paracetamol) and also with water.
3. Hydrophilic–hydrophilic interaction of polar end of water with -COO ion of N-methyl glycin and -OH group of paracetamol.
4. Ionic–hydrophilic interactions of -COOH (N-methyl glycin) and -OH (paracetamol) with the H+ and OH ion of water.
5. Hydrophobic–hydrophobic interaction of non-polar part of N-methyl glycin and paracetamol.

The interactions of categories (i), (ii), (iii) and (iv) have positive contributions to $\phi_0^E$ values while interaction of type (v) has negative contribution to $\phi_0^E$ values [29-31]. The positive $\phi_0^E$ value indicates that the hydrophilic–hydrophilic and ion–ion interactions are in domination over hydrophobic–hydrophobic and ion–ionic interactions. It is also seen that $\Delta\phi_0$ values are increasing with increase in molality of N-methyl glycin. The intermolecular distance between N-methyl glycin and paracetamol decreases with increasing concentration of N-methyl glycin as a result the hydrophobic–hydrophobic and ion–ionic interactions increase with mobility. Similar result can also be obtained from the following expression given by Franks et al [32].

$\phi_0 = \phi_0^E + \phi_0^E - 4S$

where $\phi_0^E$ is correlated with Von Der Waals volume, $\psi_0$ is the volume correlated with voids or empty space and $\psi_0$ is correlated with shrinkage volume due to electrostriction. The value $\phi_0^E$ and $\psi_0$ will remain same for the same class of solutes in aqueous solutions and only the volume due to electrostriction will vary. The hydrophilic–hydrophilic, ion–ion and ion–ionic interactions will increase with increasing mobility of N-methyl glycin and as a result $\phi_0$ value will decrease [33]. For this reason, $\phi_0$ values increase with increasing mobility of N-methyl glycin.

The volumetric pair wise and triple ion interactions may be estimated from the following equation given by McMillan–Mayer [34].

$\phi_0^E = 2\psi_{vym} + 3Y_{vym}^2$

where $Y_{vym}$ is the volume of transfer respectively and X and Y represent N-methyl glycin and paracetamol respectively. The coefficients $Y_{vym}$ are estimated by putting the $\Delta\phi_0^E$ values at diverse molalities of N-methyl glycin in presence of paracetamol in the above expression and mentioned in Table 6. It is observed that $\psi_{vym}$ values are positive whereas $Y_{vym}$ values are negative for N-methyl glycin in presence of paracetamol in aqueous medium at different temperatures. The positive values of $Y_{vym}$ suggest that existing interactions in our studied solutions are mostly pair wise which arises from hydrophilic–hydrophilic and ion–ionic interactions between solute and co-solute in aqueous medium [35].

Table 6 Pair, $\psi_{vym}$, and Triple, $Y_{vym}$ interaction coefficients of N-methyl glycin in aqueous solution of paracetamol at 298.15 K, 303.15 K and 308.15 K temperatures

| Molarity of N-methyl glycin | $\psi_{vym}$ (m mol/kg) | $Y_{vym}$ (m mol/kg) |
|-----------------------------|-------------------------|----------------------|
| 0.01 M                      | 78.0000                 | -372.23               |
| 0.02 M                      | 99.3100                 | 221.1200              |
| 0.03 M                      | 308.15 K                | 102.3700              |

3.2 Viscosity Calculation

The viscosity data was fit into Jones–Dole equation [36],

$\eta/\eta_0 = 1 / \sqrt{m} = A + B / \sqrt{m}$

where, $\eta_0$ and $\eta$ are the viscosities of the solvent and solution respectively. A plot of $(\eta/\eta_0 - 1) / \sqrt{m}$ against $\sqrt{m}$ gives a straight line with an intercept $A$ and a slope of $B$. The $(\eta/\eta_0 - 1) / \sqrt{m}$ values of N-methyl glycin of different molalities in aqueous paracetamol solution are reported in Table 3. The viscosity coefficients $A$ and $B$ values are reported in Table 4 and the variation of $B$ with temperature of N-methyl glycin is shown in Fig. 4. The viscosity $B$-coefficient signifies solute-solvent interaction and provides valuable information concerning the solvation of the solute in solution [37, 38]. A close inspection reveals that $B$-value is higher for 0.03 M N-methyl glycin solution at 308.15 K and lowest at 0.01 M solution at 298.15 K. So, solute–solute interactions increase with increasing molarity and temperature. Viscosity $A$ coefficient denotes solute-solute interaction. It is reflected from the Table 4 that the values of $A$ coefficient decrease with the increase in molarity and temperature of N-methyl glycin in aqueous solution of paracetamol. Hence solute–solvent interaction diminishes with molarity of N-methyl glycin and also with temperature in K.
3.3 Refractive Index Calculation

The molar refraction, $R_m$ for any compound in its aqueous solution may be determined from the Lorentz–Lorenz relation [39]:

$$R_m = \frac{(nD^2 - 1)}{(nD^2 + 2)} \left[ \frac{M}{\rho} \right]$$

where, $R_m$, $\rho$, $M$ and $n$ are the molar refraction, density of solution, molar mass and refractive index, respectively. The refractive index of a material is defined as $c_2/c_1$, where $c_1$ is the speed of light in any medium and $c_2$ the speed of light in vacuum. The light is refracted more for the substance of higher refractive index [40]. According to Deetles et al. [41] the molar refraction of a substance will be higher when the molecules in any solution are more tightly packed. The values of $R_m$ are shown in Table 3. The increase in molar refraction values with increase in molarity of $N$-methyl glycine in aqueous paracetamol solution indicates close packing of molecules in the mixture resulting in maximum solute–solvent interactions.

3.4 $^1H$ NMR Spectroscopy

Various spectroscopy may be employed to examine the diverse interaction playing in solution of any compound [42–46]. $^1H$ NMR Spectroscopy of pure $N$-methyl glycine, paracetamol and their solution are recorded in D2O at 298.15 K and shown in Fig. 5.

4. Conclusion

The limiting apparent molar volume ($\Phi_V$) and viscosity $B$-coefficient and molar refraction ($R_m$) values indicate the existence of strong solute–solvent interactions between $N$-methyl glycine and paracetamol in aqueous medium. The solute–solvent interactions enhances with increasing molarity of $N$-methyl glycine and temperature. On the other hand, the solute-solute interactions diminish with increasing molarity of $N$-methyl glycine and temperature. The nature of solute–solute interactions was evaluated from the limiting apparent molar volume of $N$-methyl glycine and paracetamol.

The authors are grateful to the UGC supported Major research project, SAP, DRS-III for financial support in order to continue this research work. One of the authors, Prof. M.N. Roy is thankful to University Grant Commission, New Delhi, Government of India for being awarded extra-time grant under Basic Scientific Research via the grant-in-Aid No. F.4–10/2010 (BSR) regarding his active service for augmenting of research facilities to facilitate further research work.

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