Estimation of diffusion coefficients of ascorbate-based ions at infinite dilution through electrolytic conductivity measurements

A N Soriano1, K G R Dollente1, R J D Tabaquero1 and A P Adornado1,2

1School of Chemical, Biological, and Materials Engineering and Sciences, Mapúa University, Manila 1002, Philippines
E-mail: apadornado@mapua.edu.ph/ adonisadornado@yahoo.com

Abstract. Ascorbic acid is a naturally-occurring organic compound that has high water solubility. One significant parameter that can describe the mobility and transport of ascorbic acid and its mineral salts in water systems is the diffusion coefficient. Molar conductivity data were determined from electrolytic conductivity data measured at different concentrations of ascorbate-based ions (dilute region) and temperatures (293.15 to 313.15 K). From these data, the infinite dilution diffusion coefficients of the ascorbic acid and mineral salts in water were estimated using the Nernst-Haskell Equation. The Nernst-Einstein Equation was used to predict the infinite dilution self-diffusion coefficients of the representative ascorbate-based ions ([H+], [Na+], and [Ca2+]). For most temperatures and concentrations considered, the Ca2+ cation had the highest molar conductivity but the [H+] cation had the highest correlated infinite dilution self-diffusion coefficient of $1.14105 \times 10^{-9}$ m$^2$/s at 313.15 K. The [Na+] and [Ca2+] cations had values of correlated infinite dilution self-diffusion coefficient of $2.6263 \times 10^{-9}$ m$^2$/s and $2.4045 \times 10^{-9}$ m$^2$/s at 313.15 K, respectively. The molar conductivity and diffusion coefficient of the ions at infinite dilution were in the order of [H+] > [Na+] > [Ca2+]. It was found that an increase in temperature also increases the diffusion coefficient. This study was able to estimate infinite dilution diffusion coefficient data for ascorbate-based ions. Thus, the results of this study can serve as a basis and reference for future researches to have a better understanding of the fate and transport of ascorbate-based ions in the environment.

1. Introduction
Ascorbic acid, also known as vitamin C, is an organic compound that occurs naturally. It has antioxidant properties and high water solubility. When dissolved in water, it gives mildly acidic solutions. Ascorbic acid is available in mineral ascorbate forms, such as sodium ascorbate and calcium ascorbate. For centuries, it has been used as a treatment for vitamin C deficiencies particularly scurvy. Ascorbic acid can be found in most animals and upper classification of plants. However, only humans and some vertebrates have specific requirements [1]. Vitamin C is necessary for the growth and repair of tissues in all parts of the body. Vitamin C intake helps in faster healing of wounds, and for repairing and maintaining bones and teeth. However, an overdose of vitamin C may cause some adverse effects in the physical and mental health of a person [2]. Aside from knowing the possible health hazards of vitamin C, another major concern nowadays is the dispersion of ascorbic acid in bodies of water after it has been secreted by the body. These particulates may gather in bottom sediments from which they could reactivate and could possibly affect other organisms [3]. Hence, a need to study the mass fate
and transport of these particles in water systems arises.

Diffusion coefficient is a significant physical property that can be used to evaluate the behavior of ionic liquids in environmental systems, particularly the water stream [4]. This is an important parameter indicative of the diffusion mobility of ascorbic acid and various ascorbic acid mineral ascorbates. However, there is limited information on fate and transport properties of ascorbic acid and its mineral salts in literature; but there are some systematic studies concerning molecular diffusion coefficients in the liquid phase [4]. The chief reason for the lack of information is the characteristic slowness of the diffusion process in liquids, which requires time-consuming experiments that normally last for numerous hours or even several days just to obtain single data [5]. Various methods on the determination of diffusion coefficients are accessible. Most of the diffusion coefficients data in literature were acquired using the diaphragm cell method [6] or an optical interference method. Other methods which have also been utilized in previous studies are NMR spin-echo technique [7], capillary tube technique [8], and Taylor dispersion method [9].

However, current studies showed that the determination of diffusion coefficients of electrolytes in dilute aqueous solutions is a much simpler method but has been proven to yield comparable results [10] with methods formerly used. From previous studies, several researchers have successfully correlated diffusion coefficients at infinite dilution from electrolytic conductivity measurements [10-12]. Conductivity measurements can be easily determined and can provide ample information about the transport of electrolytes in a solution. Thus, it can be used to estimate the diffusion coefficients at miniscule concentration.

Thus, to help in understanding the behavior, mobility, and probable impacts of ascorbic acid and the two mineral ascorbates form in the water streams, this study aims to determine the diffusion coefficient of ascorbate-based ions at infinite dilution using conductivity measurements. Specifically, the study intends to: (1) measure the electrolytic conductivity of solutions containing ascorbic acid and mineral ascorbates at infinite dilution, (2) use available correlations to relate the measured conductivity data to the diffusion coefficient of ascorbate-based ions at infinite dilution, and (3) assess the effect of temperature on the diffusion coefficients at infinite dilution.

2. Materials and methods

2.1. Chemicals

The source of ions for ascorbic acid [Ascorbic Acid (Kyle Med)], and the two mineral ascorbate forms – sodium ascorbate (Cigla-Kas Plus), and calcium ascorbate (Ascor-Cee Plus) used in this work were purchased from a local supplier, and were used directly without any further purifications. All solutions were freshly prepared using deionized water prior every run during the experiment. All weight measurements for ascorbic acid and mineral salts during the preparation of solutions were performed on a digital balance. Initial mineral salt solutions concentrations of 0.01 M were prepared, and then aliquots taken were further diluted using deionized water with concentrations recorded correspondingly. The solutions were mixed using a magnetic plate stirrer until the solids are completely dissolved. The same procedures were applied for the succeeding samples with dilutions done infinitely.

2.2. Electrolytic conductivity measurements

The conductivity meter used in this experiment was CON 510 Conductivity Bench Meter provided by Eutech Instruments. A validation of the equipment and calibration systems was done prior to the measurements of the considered systems to ensure that the applied procedures and the apparatus for each property measurements could give acceptably reliable results. For the measurement of conductivity, a 0.1 mol/kg KCl solution and a freshly prepared 0.5 g/L NaOH solution were used as calibration system. The measured values for the KCl and NaOH solutions were compared with the values reported by NIST and Jenway, respectively. Measurements from the conductivity meter gave values of considerable accuracy, with overall absolute average deviation (OAAD) of 4.42%.
This method was proven to be accurate by previous works done by Ribeiro et al [10] and Soriano et al [4]; thus, this method was adapted in this study. A beaker containing 500 mL of sample was properly covered and placed in a water bath. Using the built-in digital thermometer equipped in the conductivity meter, the temperature was monitored and controlled until it reached the desired value. In order to further assure that the system is in thermal equilibrium, a thermocouple was also used. When the equilibrium was reached, the electrolytic conductivity measurement was done by dipping the cell into the solution. The reading was recorded when no further movement in the meter reading was observed. These measurement procedures were done for all samples at increasing temperature on a range of 20-40°C with increments of 5°C and decreasing concentrations. The conductivity cell was washed with deionized water to remove any adhering solution in between measurements, and was dried using a clean, soft, and lignin-free piece of tissue paper. Concentrations were carried out in three independent replicate runs for each mineral salt. The conductivity meter set-up is illustrated in figure 1.

![Figure 1. Conductivity Measurement Set-Up: (1) Water Bath, (2) Thermometer, (3) Sample, (4) Insulator, and (5) Conductivity Meter.](image)

The molar conductivity ($\Lambda$) was evaluated from the measured specific electrolytic conductivity using the relationship given by equation (1),

$$\Lambda = \frac{\kappa}{C}$$

(1)

where $\kappa$ is the specific electrolytic conductivity and $C$ is the total molar concentration.

2.3. Estimation of diffusion coefficient

The diffusion coefficients of the ascorbate-based ions in the dilute solution were computed using the Nernst-Haskell model given by equation (2),

$$D_{AB}^0 = \frac{RT}{F^2} \frac{|z_+| + |z_-|}{|z_+| |z_-|} \frac{\Lambda_+^o \Lambda_-^o}{\Lambda_+^o \Lambda_-^o + \Lambda_-^o}$$

(2)

where $D_{AB}^0$ is the diffusion coefficient of the solute, or in this case the ascorbic acid salt ($A$), in water ($B$) at infinite dilution in m$^2$/s, $R$ is the universal gas constant in J/mol·K, $T$ is the absolute temperature in K, and $F$ is Faraday's constant. Other parameters in the equation are $z_+$ and $z_-$ which designate the charge numbers of the cation and anion, respectively, and $\Lambda_+^o$ and $\Lambda_-^o$ are the molar conductivities of the cation and anion in m$^2$/mol, respectively, at infinite dilution.

The ions of interest were then correlated using the Nernst-Einstein shown in equation (3),

$$F \Lambda = \frac{RT}{z}$$

where $F \Lambda$ is the product of the Faraday's constant and the molar conductivity, $z$ is the charge number of the ion, and $R$ and $T$ are as defined above.
\[ D_{\text{ion}}^o = \frac{RT \Lambda_{\text{ion}}^o}{|z_{\text{ion}}|^2} \]  

(3)

where \( D_{\text{ion}}^o \) is the self-diffusion coefficient of the ion at infinite dilution in \( \text{m}^2/\text{s} \), \( \Lambda_{\text{ion}}^o \) designates the infinite dilution molar conductivity of the ion, and \( z_{\text{ion}} \) is the charge number of the ion.

3. Results and discussion

The molar conductivity (\( \Lambda \)) data for aqueous ascorbate mineral salt solutions at concentration range of 0.01-0.0001 kmol/m\(^3\) and temperature range of 293.15-313.15 K were presented in table 1 and graphically shown in figure 2.

**Table 1.** Calculated molar conductivities (\( \Lambda \)) at infinite dilution of ascorbate solutions at different concentrations and temperatures.

| Ascorbic Acid (HC\(_6\)H\(_7\)O\(_6\)) | C (kmol/m\(^3\)) | 0.01 | 0.00111 | 0.00012 |
|---|---|---|---|---|
| \( T / \text{K} \) | \( \Lambda^o / (\text{S} \cdot \text{m}^2/\text{mol}) \) | \( \Lambda^o / (\text{S} \cdot \text{m}^2/\text{mol}) \) | \( \Lambda^o / (\text{S} \cdot \text{m}^2/\text{mol}) \) |
| 293.15 | 0.000066 | 0.000464 | 0.003375 |
| 298.15 | 0.000067 | 0.000469 | 0.003506 |
| 303.15 | 0.000068 | 0.000476 | 0.003611 |
| 308.15 | 0.000071 | 0.000480 | 0.003722 |
| 313.15 | 0.000072 | 0.000490 | 0.003789 |

| Sodium Ascorbate (NaC\(_6\)H\(_7\)O\(_6\)) | C (kmol/m\(^3\)) | 0.01 | 0.00111 | 0.00012 |
|---|---|---|---|---|
| \( T / \text{K} \) | \( \Lambda^o / (\text{S} \cdot \text{m}^2/\text{mol}) \) | \( \Lambda^o / (\text{S} \cdot \text{m}^2/\text{mol}) \) | \( \Lambda^o / (\text{S} \cdot \text{m}^2/\text{mol}) \) |
| 293.15 | 0.009767 | 0.009363 | 0.010806 |
| 298.15 | 0.009583 | 0.009072 | 0.010764 |
| 303.15 | 0.009363 | 0.008937 | 0.010711 |
| 308.15 | 0.009093 | 0.008766 | 0.010486 |
| 313.15 | 0.008773 | 0.008697 | 0.010481 |

| Calcium Ascorbate (CaC\(_{12}\)H\(_{14}\)O\(_{12}\)) | C (kmol/m\(^3\)) | 0.01 | 0.00111 | 0.00012 |
|---|---|---|---|---|
| \( T / \text{K} \) | \( \Lambda^o / (\text{S} \cdot \text{m}^2/\text{mol}) \) | \( \Lambda^o / (\text{S} \cdot \text{m}^2/\text{mol}) \) | \( \Lambda^o / (\text{S} \cdot \text{m}^2/\text{mol}) \) |
| 293.15 | 0.009497 | 0.010733 | 0.015406 |
| 298.15 | 0.009533 | 0.010523 | 0.014386 |
| 303.15 | 0.009640 | 0.010465 | 0.013856 |
| 308.15 | 0.009543 | 0.010147 | 0.013014 |
| 313.15 | 0.009243 | 0.009805 | 0.012347 |

*Evaluated based on the measured electrolytic conductivity (\( \kappa \)) data as follows: \( \Lambda = \kappa / C \).*

**Figure 2.** Calculated molar conductivities (\( \Lambda \)) at infinite dilution of ascorbate solutions: ascorbic acid (\( \bigcirc \)), sodium ascorbate (\( \Delta \)), and calcium ascorbate (\( \square \)) at different concentrations and \( T = 293.15 \) K [solid lines touching the ordinate are calculated molar conductivity values at infinite dilution].
In order to observe the behavior of the molar conductivity as a function of concentration, an equation to model the relationship of the two was used. Robinson and Stokes (RS) Equation shown in equation (4) is the common model utilized. The molar conductivity data obtained experimentally are fitted in the model to determine the infinite dilution conductivities. As the researchers fitted the experimental data, the overall average absolute deviation (OAAD) shows an average value of 10.97% which is unacceptable, thus, a new correlation is needed. With this, the RS Equation was modified as shown in equation (5). The modified RS Equation correlated well the present data as shown by an overall average absolute deviation (OAAD) of 3.45%. The comparison of the calculation results using the original and modified RS Equation is presented in table 2.

### Table 2. Comparison of parameters calculated using RS Equation and Modified RS Equation.

| T/K  | Ascorbic Acid (HC₆H₄O₆) | Sodium Ascorbate (NaC₆H₄O₆) | Calcium Ascorbate (CaC₆H₄O₁₂) | RS Equation⁵ | Modified RS Equation⁶ |
|------|--------------------------|-----------------------------|-----------------------------|-------------|-----------------------|
|      | A₀                       | α₁                           | α₂                           | AAD        | A₀                    | α₁                           | α₂                           | AAD        |
| 293.15 | 0.0274 | -7.3733 | -0.0098 | 26.42 0.0531 | 0.1920 | 1.3951 | 0.78 |
| 298.15 | 0.0284 | -7.2502 | -0.0105 | 27.15 0.0552 | 0.2003 | 1.4570 | 1.17 |
| 303.15 | 0.0293 | -7.1548 | -0.0111 | 27.58 0.0570 | 0.2068 | 1.5056 | 1.80 |
| 308.15 | 0.0301 | -7.0616 | -0.0117 | 27.71 0.0588 | 0.2140 | 1.5595 | 2.15 |
| 313.15 | 0.0307 | -7.0031 | -0.0121 | 27.75 0.0598 | 0.2177 | 1.5863 | 1.86 |
| 293.15 | 0.0103 | -2.0462 | 0.0136 | 4.89 0.0118 | -0.0996 | 0.7922 | 6.95 |
| 298.15 | 0.0102 | -2.0877 | 0.0129 | 5.90 0.0119 | -0.1171 | 0.9357 | 4.16 |
| 303.15 | 0.0102 | -2.1401 | 0.0116 | 6.15 0.0119 | -0.1220 | 0.9628 | 4.85 |
| 308.15 | 0.0100 | -2.1894 | 0.0109 | 6.00 0.0117 | -0.1176 | 0.9191 | 4.93 |
| 313.15 | 0.0100 | -2.2747 | 0.0083 | 5.98 0.0117 | -0.1200 | 0.9088 | 5.53 |
| 293.15 | 0.0145 | -2.4926 | -0.0191 | 11.01 0.0185 | -0.3036 | 2.1385 | 8.39 |
| 298.15 | 0.0137 | -2.4127 | -0.0123 | 9.42 0.0169 | -0.2514 | 1.7754 | 4.47 |
| 303.15 | 0.0132 | -2.3351 | -0.0082 | 8.41 0.0161 | -0.2208 | 1.5635 | 2.49 |
| 308.15 | 0.0124 | -2.3077 | -0.0034 | 7.47 0.0149 | -0.1874 | 1.3380 | 1.21 |
| 313.15 | 0.0118 | -2.3215 | -0.0013 | 6.87 0.0140 | -0.1660 | 1.1822 | 1.05 |

Overall 10.97 3.45

⑤ \( A = A^0 + (\alpha_1 A^0 + \alpha_2) C^{1/2} \)

⑥ \( A = A^0 + \alpha_1 C^{1/2} + \alpha_2 \left( C^{1/2} \right)^2 \)

\[ \text{AAD} = \left( \frac{100}{n} \right) \times \sum_{i=1}^{n} \left| \frac{\text{Calcd}}{\text{Expt}} - 1 \right| \]

In equations (4) and (5), \( A^0 \) is molar conductivity at infinite dilution and \( \alpha_i \) are the determined empirical parameters.

The present work focuses on infinite dilution of ascorbate-based ions. Using the equation shown in equation (6), the infinite dilution molar conductivities of the anion and cation can be predicted. These positive and negative ions migrate without interionic forces in a low concentration \([4,13]\). Since these ions are assumed to be migrating independently of each other, the total molar conductivity can be
calculated by the addition of the cation and the anions as shown in equation (6),

\[ \Lambda^0 = z_+ A_+^0 + z_- A_-^0 \]  

(6)

In using the Nernst-Einstein Equation in the determination of the self-diffusion coefficient of the ion at infinite dilution of the considered systems, the evaluation of the infinite dilution molar conductivity of the ion is the most essential component. Using equation (6), the infinite dilution molar conductivities of the considered ions were evaluated and are presented in table 3. It was observed that for all temperatures, \([H^+]\) has the highest molar conductivity, while \([Na^+]\) has the lowest. The infinite dilution molar conductivities of all the ions are of the order: \([H^+] > [C_6H_7O_6^-] > [Ca^{2+}] > [Na^+]\).

Comparing the literature values for infinite dilution molar conductivities of the \([Na^+]\) and \([Ca^{2+}]\) ions under study at 298.15 K, \([Ca^{2+}]\) was also found to have a higher molar conductivity than \([Na^+]\). The comparison was only done between \([Na^+]\) and \([Ca^{2+}]\) ions under study since the molar conductivity of \([H^+]\) at 298.15 K were simply taken from the work of Vanýsek [14], while there is no literature value available for \([C_6H_7O_6^-]\).

Table 3. Infinite dilution conductivities of the individual ions \((\Lambda^0_{\text{ion}})\) at different temperatures.

| \(T / \text{K}\) | \([H^+]^e\) | \([Na^+]\) | \([Ca^{2+}]\) | \([C_6H_7O_6^-]\) |
|---|---|---|---|---|
| 293.15 | 3.2494 | 0.8636 | 1.5836 | 1.9445 |
| 298.15 | 3.4965 | 0.8972 | 1.6322 | 2.0281 |
| 303.15 | 3.7122 | 0.9286 | 1.6682 | 2.1486 |
| 308.15 | 3.9051 | 0.9583 | 1.7335 | 2.1704 |
| 313.15 | 4.0802 | 0.9391 | 1.7196 | 2.2147 |

\(^e\)Extrapolated using values taken from Cazes [15] and Vanýsek [14].

\(^f\)Values taken from Cazes [15].

\(^g\)Values taken from Vanýsek [14].

Table 4. Calculated self-diffusion coefficients of the systems under study.

| \(T / \text{K}\) | \(10^9 D_{\text{AB}}^{0}\) (m\(^2\)/s) | \(10^9 D_{\text{cation}}^{0}\) (m\(^2\)/s) | \(10^9 D_{\text{anion}}^{0}\) (m\(^2\)/s) |
|---|---|---|---|
| Ascorbic Acid (H\(C_6H_7O_6\)) | | (Nernst-Haskell Equation) | (Nernst-Einstein Equation) |
| 293.15 | 6.3696 | 8.5068 | 5.0906 |
| 298.15 | 6.8354 | 9.3098 | 5.4001 |
| 303.15 | 7.3687 | 10.0499 | 5.8168 |
| 308.15 | 7.6781 | 10.7465 | 5.9728 |
| 313.15 | 8.0290 | 11.4105 | 6.1936 |
| Sodium Ascorbate (Na\(C_6H_7O_6\)) | | (Nernst-Haskell Equation) | (Nernst-Einstein Equation) |
| 293.15 | 3.1311 | 2.2609 | 5.0906 |
| 298.15 | 3.3124 | 2.3889 | 5.4001 |
| 303.15 | 3.5107 | 2.5140 | 5.8168 |
| 308.15 | 3.6588 | 2.6372 | 5.9728 |
| 313.15 | 3.6885 | 2.6263 | 6.1936 |
| Calcium Ascorbate (Ca\(C_6H_7O_12\)) | | (Nernst-Haskell Equation) | (Nernst-Einstein Equation) |
| 293.15 | 3.4274 | 2.0729 | 5.0906 |
| 298.15 | 3.6120 | 2.1730 | 5.4001 |
| 303.15 | 3.8135 | 2.2581 | 5.8168 |
| 308.15 | 3.9782 | 2.3852 | 5.9728 |
| 313.15 | 4.0606 | 2.4045 | 6.1936 |
Using the Nernst-Einstein Equation shown in equation (3), the infinite dilution self-diffusion coefficient of the ascorbate-based ions was evaluated. Nernst-Einstein Equation relates the molar conductivity of an electrolyte to the diffusion coefficients of its constituent ions. Such equation further illustrates that when ions dissociate and move independently within the system, they diffuse at a unique rate. Correlated values for self-diffusion coefficients of the studied ions are presented in table 4 along with the diffusion coefficients of the studied ascorbate mineral salt systems in water at infinite dilution. Figures 3 and 4 graphically showed these values. As seen in these figures, temperature increase causes rise in the diffusion coefficient of the ions. This may indicate that at high temperatures, ions move faster as vibrational energy is increased and ionic migration is at its highest.

![Figure 3](image1.png)

**Figure 3.** Infinite dilution diffusion coefficients ($D_B^0$) as a function of temperature where (○, △, and □) represent diffusion of cations: [H$^+$], [Na$^+$], and [Ca$^{2+}$], respectively.

![Figure 4](image2.png)

**Figure 4.** Effect of temperature on infinite dilution self-diffusion coefficients where (○, △, and □) represent self-diffusion of cations: [H$^+$], [Na$^+$], and [Ca$^{2+}$], respectively.

4. Conclusion

The estimation of transport parameters of ascorbate-based ions, most importantly the diffusion coefficients, was demonstrated in this study through a simple, yet well-established method involving
electrolytic conductivity data. The measured electrolytic conductivity data for the 0.01 kmol/m³ concentration solutions of ascorbic acid, sodium ascorbate, and calcium ascorbate at 298.15 K were 6.71 µS/cm (lowest), 958.33 µS/cm, and 954.33 µS/cm, respectively. Various models and equations were applied in order to correlate complex parameters from measured electrolytic conductivity data.

For the determination of infinite dilution diffusion coefficients, the Nernst-Haskell equation was used. In addition, self-diffusion coefficients were determined using the Nernst-Einstein equation. Both the infinite dilution coefficients and self-diffusion coefficients were found to increase with increasing temperature. Thus, mobility is greater at higher temperatures. It was found that the [H⁺] ion have the highest self-diffusion coefficient of $9.3098 \times 10^{-9}$ m²/s at 298.15 K. On the other hand, the [Ca²⁺] ion have the least self-diffusion coefficient of $2.1730 \times 10^{-9}$ m²/s at 298.15 K. The [Na⁺] ion has a self-diffusion coefficient of $2.3899 \times 10^{-9}$ m²/s at 298.15 K.

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