**Original Research Article**

# DIELECTRIC PROPERTIES OF 1-ETHYL-3-METHYL-IMIDAZOLIUM TETRAFLUOROBORATE (EMIM-BF4) USING COLE-COLE RELAXATION MODEL


## ABSTRACT

The Cole-Cole relaxation equations were derived from the Debye equation. The dielectric constant $\varepsilon'$ and loss factor $\varepsilon''$ of EMIM-BF4 were fitted using the derived equations at temperature range of $5℃\ to\ 65℃$ and frequency range of 0.1GHz to 10GHz. The result obtained shows that the dielectric constant and loss factor of EMIM-BF4 were higher at low frequency (i.e. $f = 0.1GHz$) and decrease as the frequency increases. The dielectric constant also increases with increase in the temperature except at 0.1GHz. At 15$^o$C there was a sudden increase in the dielectric constant especially as the frequency increases beyond 5GHz. This sudden increase in the dielectric constant of EMIM-BF4 may be due to the phase change of EMIM-BF4. The loss factor of EMIM-BF4 was generally small for all frequencies and temperatures. This may be due to the fact that EMIM-BF4 consumed less energy when subjected to an applied field.


## INTRODUCTION

The last decade has witnessed an upsurge in research activities focusing on replacing the abundant used volatile organic solvents (VOC) with a more environmental friendly one. Several alternative methods have been developed and recently ionic liquids have emerged as “green” and environmental friendly solvents. Ionic liquids are a new class of purely ionic, salt-like materials that are liquid at ambient temperatures. In broad sense, this term includes all the molten salts, for instance, sodium chloride at temperatures higher than $800℃$ [1]. Today however, the term “ionic liquids” is used for the salts whose melting point is relatively low (below$100℃$) [2]. A typical ionic liquid (IL) has a bulky organic cation (e.g N-alkylpyridinium, N-N-dialkylimidazolium) that is weakly coordinated to an organic and inorganic anions, such as $BF_4^-, Cl^-, I^-, CF_3SO_3^-, and\ AlCl_4^-$. The big difference in the size of a bulky cation and a small anion does not allow packing of lattice, which happens in many organic salts; instead, the anions are disorganized [3]. Ionic liquids have several advantages compared to commercial organic solvents or electrolyte liquids [4-5]. They are characterised by their non-combustible, non-flammable, display wide electrochemical windows, high inherent conductivity and lack of reactivity in various electrochemical or industrial applications etc. [6-10].

Because their properties, ionic liquids have attracted great attention in many fields, including organic chemistry, electrochemistry, physical chemistry, industrial physics and engineering generally. Today ionic liquids have been thought to be more safe electrolytes materials for electrochemical and energy storing devices, such as lithium batteries for cellular phones, batteries for vehicles, fuel cells, super capacitors, solar cells etc. [11-14].

Due to the special characteristics of ILs, such as wide electrochemical windows, high inherent conductivities, high thermal and electrochemical stability, tuneable physicochemical properties, etc., they are potentially excellent candidates for environmentally sound, green electrolytes in batteries. In order to predict their success in a specific application, it is essential to gain information about their dielectric properties.

In this work, attempt have been made to study the dielectric properties of 1-Ethyl-3-methyl-imidazolium tetrafluoroburate (EMIM-BF4) because of its high ionic conductivity and low viscosity. Therefore, EMIM-BF4 is expected to be a good electrolyte candidate for lithium batteries when compared to organic solvent electrolytes and other ionic liquids.

## MATHEMATICAL DERIVATION OF COLE-COLE EQUATIONS

The Debye equations can be expressed more concisely as

$$\varepsilon^* = \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{1 + j\omega\tau} \qquad (1)$$

But polar dielectrics that have more than one relaxation time do not satisfy Debye equations. An empirical equation for the complex dielectric constant has been suggested as:

$$\varepsilon^* = \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{1 + (j\omega\tau_{c-c})^{1-\alpha}}; \quad 0 \le \alpha \le 1 \qquad (2)$$

$\alpha = 0$ for Debye relaxation, $\tau_{c-c}$ is the mean relaxation time and $\alpha$ is a constant for a given material.

To determine the geometrical interpretation of equation (2), we substitute $1 - \alpha = n$ and rewrite it as

$$\varepsilon' - j\varepsilon'' = \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{1+(\omega\tau_{c-c})^n(cos\frac{n\pi}{2} + jsin\frac{n\pi}{2})}; \quad 0 \le \alpha \le 1 \qquad (3)$$

Where $j^n = cos\,\frac{n\pi}{2} + jsin\frac{n\pi}{2}$ and $\varepsilon^* = \varepsilon' - j\varepsilon''$

Multiply equation (3) by $\frac{1+((\omega\tau_{c-c})^n(cos\frac{n\pi}{2} - jsin\frac{n\pi}{2})}{1+((\omega\tau_{c-c})^n(cos\frac{n\pi}{2} - jsin\frac{n\pi}{2})}$

$$\text{i.e } \varepsilon' - j\varepsilon'' = \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{1+(\omega\tau_{c-c})^n\left(cos\frac{n\pi}{2} + jsin\frac{n\pi}{2}\right)} \times \frac{1+((\omega\tau_{c-c})^n(cos\frac{n\pi}{2} - jsin\frac{n\pi}{2})}{1+((\omega\tau_{c-c})^n(cos\frac{n\pi}{2} - jsin\frac{n\pi}{2})}$$

$$\varepsilon' - j\varepsilon'' = \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty[1+((\omega\tau_{c-c})^n(\cos\frac{n\pi}{2} - jsin\frac{n\pi}{2})}{1+2(\omega\tau_{c-c})^n\cos\left(\frac{n\pi}{2}\right)+(\omega\tau_{c-c})^{2n}} \qquad (4)$$

Equating the real and imaginary part, we have

$$\varepsilon' = \varepsilon_\infty + (\varepsilon_s - \varepsilon_\infty)\frac{1+((\omega\tau_{c-c})^n\cos\,(\frac{n\pi}{2})}{1+2(\omega\tau_{c-c})^n\cos\left(\frac{n\pi}{2}\right)+(\omega\tau_{c-c})^{2n}} \qquad (5)$$

$$-j\varepsilon'' = \varepsilon_s - \varepsilon_\infty\frac{1+((\omega\tau_{c-c})^n(-jsin\frac{n\pi}{2})}{1+2(\omega\tau_{c-c})^n\cos\left(\frac{n\pi}{2}\right)+(\omega\tau_{c-c})^{2n}}$$

$$\therefore\; \varepsilon'' = \varepsilon_s - \varepsilon_\infty\frac{1+((\omega\tau_{c-c})^n(\,sin\frac{n\pi}{2})}{1+2(\omega\tau_{c-c})^n\cos\left(\frac{n\pi}{2}\right)+(\omega\tau_{c-c})^{2n}} \qquad (6)$$

## METHODS

Equations (5) & (6) are called the real and imaginary parts of the Cole-Cole relaxation model. The real part ($\varepsilon'$) represents the dielectric constant while the imaginary part ($\varepsilon''$) represents the loss factor. An algorithm was written using Maple-13 and the dielectric constant $\varepsilon'$ and the loss factor $\varepsilon''$ of 1-ethyl-3-methylimidazolium tetraflouroborate $[EMIM][BF_4]$ were generated (see tables 1 and 3 below). The computations were done within frequency range of 0.1GHz to 10GHz and the temperatures between 5$^o$C and 65$^o$C. These frequency range were chosen because some polarization mechanisms such as atomic and electronic polarization contribution occur at higher frequencies more than megahertz range. The results generated in our computations are discussed below:

## RESULTS AND DISCUSSION

The dielectric constant and the loss factor of ionic liquids were computed using Cole-Cole relaxation method. The dielectric constant $\varepsilon'$ and the loss factor $\varepsilon''$ of EMIM-BF4 were computed within the temperature range of $5°C\ to\ 65°C$ and the frequency of 0.1GHz to 10GHz. The results obtained in this work are shown in table1 and table 2 below and are discussed based on the existing theories.

**Table 1.** The dielectric constant $\varepsilon'$ and loss factor $\varepsilon''$ of ionic liquids (1-ethyl-3-methylimidazolium tetrafluoroborate) within the temperature range of 5°C and 35°C.

| F(GHz) | 5°C | | 15°C | | 25°C | | 35°C | |
|---|---|---|---|---|---|---|---|---|
| | $\varepsilon'$ | $\varepsilon''$ | $\varepsilon'$ | $\varepsilon''$ | $\varepsilon'$ | $\varepsilon''$ | $\varepsilon'$ | $\varepsilon''$ |
| 0.1 | 14.7932 | 0.0459 | 14.6717 | 0.0523 | 14.1658 | 0.0753 | 13.4608 | 0.0870 |
| 0.2 | 14.1668 | 0.0410 | 14.3346 | 0.0481 | 13.9239 | 0.0719 | 13.3343 | 0.0847 |
| 0.3 | 13.7120 | 0.0376 | 13.8837 | 0.0450 | 13.7136 | 0.0689 | 13.2141 | 0.0825 |
| 0.4 | 13.3457 | 0.0350 | 13.6012 | 0.0462 | 13.5236 | 0.0663 | 13.0989 | 0.0805 |
| 0.5 | 13.0409 | 0.0329 | 13.3575 | 0.0405 | 13.3487 | 0.0640 | 12.9879 | 0.0785 |
| 0.6 | 12.7752 | 0.0311 | 13.1419 | 0.0387 | 13.1859 | 0.0618 | 12.8806 | 0.0766 |
| 0.7 | 12.5404 | 0.0295 | 12.9479 | 0.0372 | 13.0331 | 0.0598 | 12.7767 | 0.0748 |
| 0.8 | 12.3297 | 0.0282 | 12.7712 | 0.0357 | 12.8890 | 0.0580 | 12.6759 | 0.0731 |
| 0.9 | 12.1387 | 0.0270 | 12.6088 | 0.0345 | 12.7523 | 0.0563 | 12.5780 | 0.0714 |
| 1.0 | 11.9635 | 0.0259 | 12.4584 | 0.0333 | 12.6223 | 0.0547 | 12.4827 | 0.0699 |
| 1.2 | 11.6524 | 0.0241 | 12.1868 | 0.0313 | 12.3794 | 0.0517 | 12.2998 | 0.0668 |
| 1.5 | 11.2490 | 0.0218 | 11.8361 | 0.0288 | 12.0510 | 0.0478 | 12.0423 | 0.0627 |
| 1.8 | 10.9291 | 0.0200 | 11.5357 | 0.0267 | 11.7571 | 0.0445 | 11.8027 | 0.0590 |
| 2.0 | 10.7356 | 0.0190 | 11.3570 | 0.0255 | 11.5771 | 0.0425 | 11.6518 | 0.0567 |
| 2.5 | 10.3210 | 0.0169 | 10.9675 | 0.0230 | 11.1723 | 0.0382 | 11.3017 | 0.0516 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| 3.0 | 9.9791 | 0.0153 | 10.6400 | 0.0209 | 10.8197 | 0.0347 | 10.9855 | 0.0472 |
| 3.5 | 9.6894 | 0.0140 | 10.3581 | 0.0193 | 10.5082 | 0.0317 | 10.6980 | 0.0434 |
| 4.0 | 9.4390 | 0.0129 | 10.1112 | 0.0179 | 10.2301 | 0.0292 | 10.4352 | 0.0400 |
| 4.5 | 9.2191 | 0.0120 | 9.8920 | 0.0167 | 9.9794 | 0.0269 | 10.1938 | 0.0370 |
| 5.0 | 9.0237 | 0.0112 | 9.6954 | 0.0156 | 9.7519 | 0.0250 | 9.9712 | 0.0344 |
| 5.5 | 8.8482 | 0.0105 | 9.5174 | 0.0147 | 9.5440 | 0.0233 | 9.7652 | 0.0320 |
| 6.0 | 8.6893 | 0.0099 | 9.3551 | 0.0139 | 9.3532 | 0.0218 | 9.5739 | 0.0299 |
| 6.5 | 8.5445 | 0.0093 | 9.6062 | 0.0132 | 9.1771 | 0.0205 | 9.3956 | 0.0280 |
| 7.0 | 8.4115 | 0.0089 | 9.0688 | 0.0125 | 9.0139 | 0.0192 | 9.2292 | 0.0263 |
| 7.5 | 8.2889 | 0.0084 | 8.9414 | 0.0120 | 8.8622 | 0.0181 | 9.0734 | 0.0247 |
| 8.0 | 8.1753 | 0.0081 | 8.8229 | 0.0114 | 8.7207 | 0.0172 | 8.9272 | 0.0233 |
| 8.5 | 8.0696 | 0.0077 | 8.7121 | 0.0109 | 8.5882 | 0.0162 | 8.7896 | 0.0220 |
| 9.0 | 7.9709 | 0.0074 | 8.6083 | 0.0105 | 8.4639 | 0.0154 | 8.6600 | 0.0208 |
| 9.5 | 7.8784 | 0.0071 | 8.5107 | 0.0100 | 8.3470 | 0.0147 | 8.5377 | 0.0197 |
| 10.0 | 7.7915 | 0.0068 | 8.4186 | 0.0097 | 8.2368 | 0.0140 | 8.4219 | 0.0187 |

**Table 2.** The dielectric constant $\varepsilon'$ and loss factor $\varepsilon''$ of ionic liquids (1-ethyl-3-methylimidazolium tetrafluoroborate) within the temperature range of $45°\text{C}\ and\ 65°\text{C}$.

| F(GHz) | $45^0$C | | $55^0$C | | $65^0$C | |
|---|---|---|---|---|---|---|
| | $\varepsilon'$ | $\varepsilon''$ | $\varepsilon'$ | $\varepsilon''$ | $\varepsilon'$ | $\varepsilon''$ |
| 0.1 | 12.8747 | 0.0765 | 12.5661 | 0.1020 | 12.0899 | 01191 |
| 0.2 | 12.7733 | 0.0748 | 12.5278 | 0.1011 | 12.0756 | 0.1186 |
| 0.3 | 12.6805 | 0.0732 | 12.4880 | 0.1002 | 12.0590 | 0.1182 |

| | | | | | | |
|---|---|---|---|---|---|---|
| 0.4 | 12.5935 | 0.0718 | 12.4472 | 0.0993 | 12.0409 | 0.1177 |
| 0.5 | 12.5109 | 0.0705 | 12.4058 | 0.0984 | 12.0216 | 0.1171 |
| 0.6 | 12.4319 | 0.0692 | 12.3640 | 0.0975 | 12.0012 | 0.1165 |
| 0.7 | 12.3559 | 0.0680 | 12.3220 | 0.0965 | 11.9799 | 0.1159 |
| 0.8 | 12.2826 | 0.0667 | 12.2798 | 0.0956 | 11.9580 | 0.1153 |
| 0.9 | 12.2117 | 0.0658 | 12.2375 | 0.0946 | 11.9353 | 0.1147 |
| 1.0 | 12.1429 | 0.0647 | 12.1951 | 0.0937 | 11.9120 | 0.1140 |
| 1.2 | 12.0111 | 0.0627 | 12.1105 | 0.0919 | 11.8640 | 0.1127 |
| 1.5 | 11.8259 | 0.0599 | 11.9843 | 0.0892 | 11.7890 | 0.1106 |
| 1.8 | 11.6531 | 0.0574 | 11.8595 | 0.0865 | 11.7110 | 0.1084 |
| 2.0 | 11.5440 | 0.0559 | 11.7772 | 0.0848 | 11.6579 | 0.1070 |
| 2.5 | 11.2894 | 0.0522 | 11.5755 | 0.0806 | 11.5218 | 0.1033 |
| 3.0 | 11.0567 | 0.0490 | 11.3799 | 0.0767 | 11.3830 | 0.0996 |
| 3.5 | 10.8426 | 0.0462 | 11.1908 | 0.0730 | 11.2427 | 0.0960 |
| 4.0 | 10.6442 | 0.0436 | 11.0083 | 0.0695 | 11.1021 | 0.0924 |
| 4.5 | 10.4595 | 0.0413 | 10.8323 | 0.0662 | 10.9620 | 0.0889 |
| 5.0 | 10.2869 | 0.0392 | 10.6629 | 0.0632 | 10.8229 | 0.0855 |
| 5.5 | 10.1250 | 0.0373 | 10.4997 | 0.0603 | 10.6854 | 0.0822 |
| 6.0 | 9.9727 | 0.0356 | 10.3426 | 0.0575 | 10.5498 | 0.0790 |
| 6.5 | 9.8290 | 0.0339 | 10.1913 | 0.0550 | 10.4164 | 0.0759 |
| 7.0 | 9.6931 | 0.0324 | 10.0457 | 0.0525 | 10.2855 | 0.0729 |
| 7.5 | 9.5643 | 0.0310 | 9.9055 | 0.0503 | 10.1571 | 0.0701 |
| 8.0 | 9.4420 | 0.0298 | 9.7705 | 0.0481 | 10.0324 | 0.0673 |
| 8.5 | 9.3257 | 0.0286 | 9.6404 | 0.0461 | 9.9085 | 0.0647 |

| | | | | | | |
|---|---|---|---|---|---|---|
| 9.0 | 9.2149 | 0.0274 | 9.5150 | 0.0442 | 9.7884 | 0.0622 |
| 9.5 | 9.1091 | 0.0264 | 9.3941 | 0.0424 | 9.6712 | 0.0598 |
| 10.0 | 9.0080 | 0.0254 | 9.2776 | 0.0407 | 9.5568 | 0.0575 |

## DIELECTRIC CONSTANT

The effect of frequency for the dielectric constant of EMIM-BF4 for different temperatures is shown graphically below.

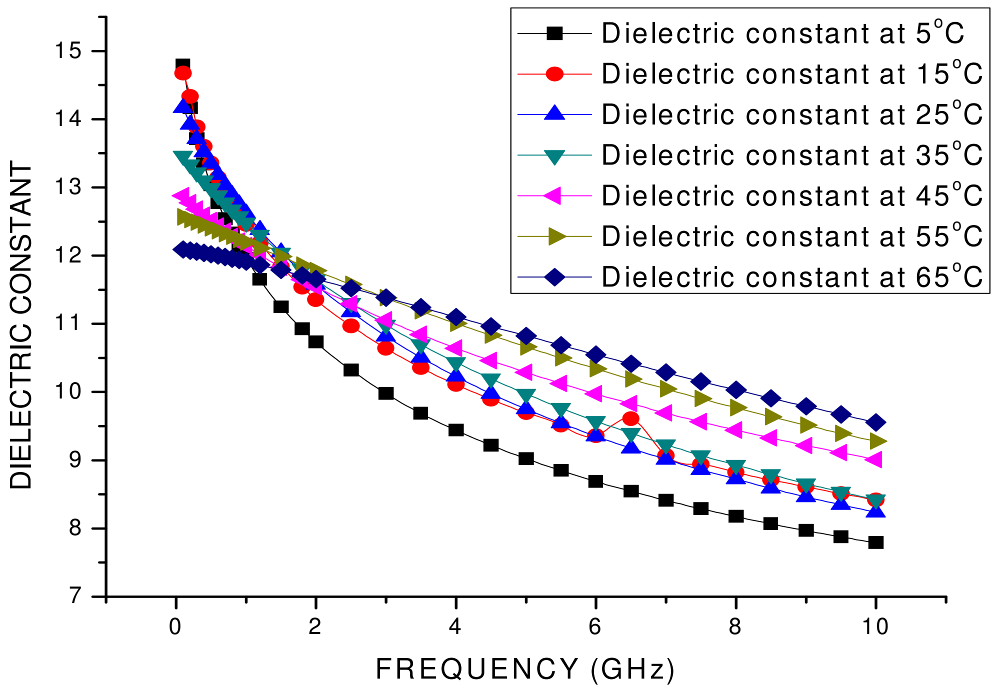


*Fig.1: The graph of dielectric constant against the frequency. The dielectric constant of EMIM-BF4 decrease when the temperature increased beyond 0.1GHz (i.e. 14.733 to 12.090 for 5°C and 65°C respectively). However, as the frequency increase beyond 0.1GHz the dielectric constant increases when the temperature increases (see fig.1 above).*

The dielectric constant $\varepsilon'$ and loss factor of EMIM-BF4 has been studied using Cole-Cole relaxation model. The results revealed that at frequency 0.1GHz the dielectric constant decrease when the temperature increases. This decrease in the dielectric constant as a result of the increase in the temperature at that particular frequency may be due to the relaxation time which has been found to be fast at high temperature and increases dramatically at low temperatures, suggesting a freezing of electric dipole at low temperature [15-17].

The dielectric constant of EMIM-BF4 was also high at lower frequencies. The higher value of dielectric constant $\varepsilon'$ at low frequency may be due to the effect of ionic conductivity which is inversely proportional to frequency or maybe because of the overall conductivity which consists of different conduction mechanisms. The most prevalent one in moist materials is the ionic conductivity [16].

The graph of dielectric constant against frequency in gigahertz (GHz) at various temperatures revealed that the dielectric constant $\varepsilon'$of EMIM-BF4 has high values at low frequency then decreased sharply when the frequency increases. The decrease of dielectric constant at higher frequency range for EMIM-BF4 may be due to the fact that the dipole cannot follow up the applied field. The higher values of dielectric constant $\varepsilon'$ and loss factor $\varepsilon''$ at lower frequencies may be due to the contribution from all the four types of polarization (i.e the space charge, dipole, ionic and electronic polarization) [18]. It is observed that at higher frequencies, only the ionic and electronic polarizations contribute. The decrease in the dielectric constant and loss factor as the result of increase in the frequency may also means that the response of the permanent dipole decreases as the frequency increases and the contribution of the charge carriers (ions) to the dielectric constant decreases [16,19]. It is also observed that at temperature 15℃ and between the frequencies range 5GHz to 7GHz there was a sudden increment in the dielectric constant of EMIM-BF4 (see fig.1 above). This sudden increase in the dielectric constant of EMIM-BF4 at that particular temperature may be due to a phase change of EMIM-BF4 [20]. This is because the dielectric constant strongly dependent on the structure of materials [21].

## CONCLUSION

The Cole-Cole equation and its derivatives have been used to compute the dielectric constant and loss factor of EMIM-BF4. The dielectric constant and loss factor of EMIM-BF4 was higher at lower frequencies and decrease as the frequency increases. The dielectric constant however, increases when the temperature increases, for all frequencies except those at 0.1GHz (see tables 1 and 2 above).

The loss factor of EMIM-BF4 was relatively small for all temperatures studied in this work. This implies that the imaginary part of EMIM-BF4 does not absorb too much heat from alternating field (see tables1 and 2 above)

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