Dielectric Study of Gel Polymer Electrolyte Based on PVA-K₂CO₃-SiO₂

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Abstract. In this study, effect of filler (SiO₂) on dielectric and electrical properties of gel polymer electrolyte (GPE) based on PVA-K₂CO₃ has been investigated and reported. The electrolyte were prepared by incorporating silica particle as a filler into the un-plasticized electrolyte (PVA-K₂CO₃). The prepared electrolyte were characterized physicochemically (FTIR) and electrochemically based on electrochemical impedance spectroscopy (EIS). Based on the impedance spectroscopy, complex permittivity (ε′) (dielectric constant and loss) and complex electrical modulus (M′) (real and imaginary modulus) were calculated. Characterization result indicate that SiO₂ particles has successfully interacts with PVA-K₂CO₃ in the form of a three dimensional polymeric network. At low frequencies, high values of complex permittivity (dielectric constant and dielectric loss) were observed, which increased with increasing temperature, signifying an increase in ionic conductivity of the electrolyte. With the incorporation of filler, the peaks of both ε′ and M′ shifts towards higher frequency side suggesting the speed up the relaxation time. From the electrical modulus, the developed electrolyte is shown to be highly capacitive in nature. Based on the peak shape of the imaginary part of electric modulus, the non-Debye type relaxation predicted. Analysis of both dielectric permittivity and electrical modulus suggest that ionic and polymer segmental motions are strongly coupled. An optimum ionic conductivity of 3.25 × 10⁻⁴ mScm⁻¹ was achieved at ambient temperature at a composition of 15 wt.% SiO₂ (PKS15).

1. Introduction

Polymer Electrolytes (PEs) particularly gel polymer electrolyte (GPEs) has attracted scientific interests nowadays over liquid electrolyte due to the safety nature and environmental friendliness of GPEs [1]. This type of electrolyte (GPEs) was reported to have various advantages such as fast charge and discharge rate at electrode-electrolyte interface, and environmentally safe since they contain no hazardous liquids, hence there is no risk of spillage in electrical energy systems. Moreover, GPE can be used as a thin film both as an electrolyte as well as a separator, long cycle life, have a short charging time, low internal corrosion, simple principles and modes of construction, good flexibility in packaging, wide potential window, high ionic conductivity, and high chemical strength [2-3]. However, the shortcomings of the GPE also are clear that is weak mechanical strength, and sometimes-low dielectric...
constant, which will bring about internal short circuits and a restricted scope of working temperature [4]. To solve this problem and improved the weak mechanical strength, conductivity and dielectric constant of GPE, fillers such as SiO$_2$ are usually added. The addition of filler is reported to be one amongst the foremost approaches to enhance the mobility of ions or/and the interfacial interface among ions and polar groups within the electrolytes [5].

Filler is an additive used to increase mechanical and physical properties of electrolyte. Addition of fillers into the PEs result in the formation of composites [6]. Previous study has reported that, filler is an important factor to manage the conductivity of PEs. The study further confirmed that, addition of fillers improved electrochemical performance of the PEs by increasing the homogeneity and dielectric constant of the samples [7]. Similarly, filler is found not only support ionic conductivity of PEs, but to expand the potential window of polymer electrolyte [8]. Filler likewise has numerous advantages in the preparation of PEs, for examples, increase physical properties of polymer matrix, reduce the crystallinity and increase amorphous degree, decrease glass transition temperature ($T_g$) of polymer membrane and enhance dielectric constant of the PEs [7, 9-10]. Addition of filler similarly permits PEs to preserve its liquid–like features and produced fast mobile ions during charge and discharge process. This is found to improve performance of the cells through reduced capacity diminishing and enhanced potential window [6]. Therefore, this study aims to investigate the important role of dielectric behavior on PVA-K$_2$CO$_3$-SiO$_2$ composite GPEs.

2. Materials and Methods

2.1. Material
The PVA (hydrolyzed 99%), silica material and potassium carbonate were obtained from Sigma-Aldrich (002008792-H), WI, USA. The chemicals are of analytical grade and used as received. For all the experiments preparation, deionized water was used.

2.2. Preparation of GPE (PVA-K$_2$CO$_3$-SiO$_2$)
PVA as host polymer, potassium carbonate (K$_2$CO$_3$) as salt, and silica (SiO$_2$) as filler were used to prepare GPEs. The electrolytes were prepared by dissolution technique with varying amount (ratio) of filler. For the preparation of un-plasticized electrolytes, a fixed ratio of PVA and K$_2$CO$_3$ (70:30) were added into 20 mL of deionized water in a separate glass beaker, heated at 80°C with vigorous stirring until completely dissolved. The two solutions were mixed and heated at 80°C continuously using thermostat at constant stirring until complete dissolution, where a composite of PVA-K$_2$CO$_3$ were obtained as reported in our previous study [11]. For the preparation of electrolyte with filler, an appropriate amount of silica was added to the 70:30 of the obtained un-plasticized electrolytes, heated at 80°C continuously with vigorous stirring until homogeneous, clear and viscous solution were obtained as shown in Figure 1. The optimization study was conducted by varying amount of silica while keeping the polymer-salt constant. The resulting polymer electrolyte were coded as PKS0, PKS5, PKS10, PKS15, PKS20, and PKS25 for electrolyte incorporated with 0, 5, 10, 15, 20 and 25 wt. % of SiO$_2$, respectively. All the samples were stored in a desiccator prior to analysis and testing.

![Figure 1](image.png)
2.3. Physicochemical characterization
The Fourier transform infrared (FTIR) analysis was conducted on an FTIR spectrometer (Bruker Instruments, model Aquinox 55, Germany) in the 4000 – 400 cm\(^{-1}\) range using KBr pellets with the scanning resolution of 4 cm\(^{-1}\).

2.4. Electrochemical Impedance Spectroscopy (EIS)
The Impedance Spectroscopy of the prepared electrolyte was studied using AUTOLAB/AUT51018 (potential/galvanostat) (Metrohm Ltd, Herisau, Switzerland) electrochemical impedance spectroscopy (EIS). A stainless steel cell blocking electrodes sandwiched completely with GPE was used for the analysis [12]. Measurements were conducted over a frequency range of 0.1 Hz to 10\(^5\) Hz and a temperature range of 303.15 – 383.15K.

3. Result and Discussion
In this section, all the results obtained from this study are presented here and followed by discussion.

3.1. Physicochemical characterization
The physicochemical characterization of the prepared electrolytes were characterized physicochemically in order to study the interaction between polymer, salt and the filler.

3.1.1. FTIR (Chemical structure).
To affirm the interaction and coordination between salt, polymer and the filler (silica) in composite polymer electrolytes, FTIR spectroscopy was examined and the result is present in Figure 2. The Figure shows the spectra of blended electrolytes with different weight compositions of filler. The absorption peak of the prepared electrolytes shows various peaks between 4000 cm\(^{-1}\) and 400 cm\(^{-1}\). From the band of PVA/K\(_2\)CO\(_3\) with no filler (PKS0), the absorption spectrum at 3258 cm\(^{-1}\) is allotted to the stretching of O–H vibrations of alcohol group [13-14]. The broad peak appear at 2941 cm\(^{-1}\) is associated with the asymmetric C–H stretching. The sharp peak located at 1647 cm\(^{-1}\) is assigned to the C=O stretching of carboxyl group [35]. The appearance of peaks in the region of 1562 cm\(^{-1}\) could be assign to the strong C = C stretching vibration from α, β-unsaturated ketone of the polymer composites [36]. More so, the peak at 1411 cm\(^{-1}\) belongs to bending vibration of CH\(_2\) while the band 1141 cm\(^{-1}\) occurred because of the CH twitching vibrations. The bands at 1090 cm\(^{-1}\) belong to the C-O broadening of acetyl group present in the PVA structure while the absorption spectrum at 843 cm\(^{-1}\) has been allocated to C-H rocking vibrations. Finally, the small peak at 657 cm\(^{-1}\) denoted to the free CO\(_3\)\(^{2-}\) anions, which designates the high freedom of the solvated K\(^+\), and CO\(_3\)\(^{2-}\) ions in the GPE system [15].
Figure 2. FTIR spectrum of PVA-K$_2$CO$_3$ (PKS0), PKS5, PKS10, PKS15, PKS20 and PKS25 gel polymer electrolytes.

Figures PKS5-PKS25 depict the spectra of polymer electrolytes with 5, 10, 15, 20 and 25 wt.% of SiO$_2$, respectively. It is observed that, the peaks of O–H widening, asymmetric C–H widening, C=O enlarging, C = C broadening and all other peaks are still found with little changes and shifting in the peak intensities [8]. This changes and shifting in the peak intensities are reported to be important in order to establish the complex formation between the host polymer, salt and the filler. For instance, the shifting of hydroxyl band of PKS from 3258 to 3271 cm$^{-1}$ when blended with SiO$_2$ and the formation of new spectrum at 917 cm$^{-1}$ and 1333 cm$^{-1}$ corresponding to symmetric and asymmetric Si–O–Si expanse, respectively, indicating the polymer-salt-silica complexation [16-17]. It is also observed from the spectra of the PKS15 that, the peak of CH wagging at 843 cm$^{-1}$ moved to upper wavenumbers (845 cm$^{-1}$) than the other electrolytes, which designates shortened bond lengths of gel-like structures in the electrolytes [14, 18]. These indicate that SiO$_2$ interacts with PVA-K$_2$CO$_3$ in the form of a three dimensional polymeric. In other words, filler (SiO$_2$) and the electrolytes (PVA-K$_2$CO$_3$) chains existed in the form of GPEs [19].

3.2. Dielectric studies of GPE based on PVA-K$_2$CO$_3$-SiO$_2$

In this section, dielectric constant which indicates the capacitive nature of the PEs observed in this study is presented. It was found that, an increase in dielectric constant with increase in filler amount is ascribed to the increase in number of free mobile ions whereas the decrease could be due to the ion association that results in generating non-conductive ion-pairs.

3.2.1. Dielectric constant ($\varepsilon_r$) and Dielectric loss ($\varepsilon_i$) at ambient temperatures.
Figure 3 (a) and (b) presents the variants of dielectric constant ($\varepsilon_r$) and dielectric loss ($\varepsilon_i$) as a function of log frequency for PVA-K$_2$CO$_3$-SiO$_2$ with different weight percentages of SiO$_2$ at ambient temperatures. From the impedance measurement real and imaginary components, $\varepsilon_r$ and $\varepsilon_i$ were calculated following equations (1) and (2):

$$\varepsilon_r = \frac{Z_r}{\omega C_0 (Z_r^2 + Z_i^2)}$$

(1)
\[ \varepsilon_i = \frac{Z_r}{\omega C_0 (Z_r + Z_i^2)} \]  

(2)

where \( Z_i \) is the imaginary part of impedance, \( Z_r \) is the real part of impedance, and \( \omega = 2\pi f \), where \( f \) is the frequency, \( C_0 = \varepsilon_0 A/t \) which is the vacuum capacitance of the measuring cell, where \( A \) and \( t \) denote area and separation length of the electrode. The quantity \( \varepsilon_0 \) is the dielectric permittivity of free space that is equal to \( 8.854 \times 10^{-12} \text{ F.m}^{-1} \) [20].

From the Figure, change in \( \varepsilon_r \) and \( \varepsilon_i \) with different amount of filler can be clearly observed. It can be observed that, both \( \varepsilon_r \) and \( \varepsilon_i \) are high at low frequencies that could be associated with the charge carriers or polarization effect at the electrodes [21]. Besides, owing to the high periodic reversal of the electric field at the electrolyte-electrode interface at higher frequencies, the influence of charge carriers towards \( \varepsilon_r \) and \( \varepsilon_i \) decreases. This decrease can be associated with the failure of dipoles in PVA-K_2CO_3-SiO_2 system to rotate rapidly, which leads to a lag amongst the frequency of oscillating dipole and that of applied filed. It can be notice that, the increase is more prominent towards low frequencies, and this can be ascribed to the effect of electrode polarization [21].

![Figure 3](image)

**Figure 3.** The plots of (a) \( \varepsilon_r \) and (b) \( \varepsilon_i \) against frequency for PVA-K_2CO_3-SiO_2 samples at ambient temperatures.

The dielectric permittivity of PVA is found to increase with the incorporation of SiO_2 materials and these parameter is considerably inclined by the frequency. The dielectric constant and dielectric loss of PVA-K_2CO_3 rise with the incorporation of SiO_2. The incorporation of SiO_2 is anticipated to rise the degree of salt dissociation and association, which rise the number of mobile ions (K\(^+\)) due to the weak bond formation between hydroxyl groups (–OH) of the polymer, K\(^+\) from the salts and the SiO_2 which result in complex formation. In addition, the partial or weak bond formed between K\(^+\) and SiO_2 with the polar groups of the polymer could aid in speeding the conduction of ions since the ions are in a moveable state [13]. Similarly, silica material was found to affect the changes in the morphological structure of the electrolytes. The silica external surface may chemically coordinate with the ions and subsequently deliver extra site generating favorable tunnels or ways for ion transport within the locality of the electrolytes [22]. The improvement in conductivity based on the hypothesis that the SiO_2 particles act as a supporting matrix. This, consequently, aids to preserve an overall solid structure of the electrolyte with unaffected volume for K\(^+\) transports. The Lewis acid-base surface groups in the polymer relate with cations and anions from the salts and delivers additional sites generating favourable conduction paths for the ion movements, thereby efficiently enhanced ion mobility [21].

3.2.2. **Temperature dependence of \( \varepsilon_r \) and \( \varepsilon_i \) at selected temperatures.**

Figure 4 (a) and (b) show the variations of \( \varepsilon_r \) and \( \varepsilon_i \) as a function of log frequency at different temperatures for high conducting electrolyte sample (PKS15). Both \( \varepsilon_r \) and \( \varepsilon_i \) is observed to be
considerably increase with the increase in temperature as frequency decreases. It is found that temperature significantly affects the dielectric property of the prepared electrolytes in this study. Similar behaviour of increase in \( \varepsilon_r \) and \( \varepsilon_i \) with increase in temperature was reported in previous studies [23]. The increased temperature presents flexibility in the polymer chain segment and thus increased its mobility consistent with the increase in conductivity [21].

3.2.3. Electric modulus analysis at ambient temperature.

The complex electrical modulus \( M^* \) is a technique used widely in analyzing ionic conductivities of an electrolyte material by correlating conductivity relaxation time with the ionic process [12]. In this study, electrical modulus \( (M_{\text{real}} \text{ and } M_{\text{imag}}) \) were investigated at ambient temperatures as a function of log frequency for the sample with the maximum conductivity (PKS15) and the result is presented in Figure 5 (a and b). From the \( \varepsilon^* \), real \( (M_{\text{real}}) \) and imaginary \( (M_{\text{imag}}) \) modulus are calculated using the equations (3 and 4) [24-26]:

\[
M_{\text{real}} = \frac{\varepsilon_r}{\varepsilon_r^2 + \varepsilon_i^2}
\]

\[
M_{\text{imag}} = \frac{\varepsilon_i}{\varepsilon_r^2 + \varepsilon_i^2}
\]

Figure 5 (a and b) show the \( M_{\text{real}} \) and \( M_{\text{imag}} \) of the electric modulus against log \( f \) with respect to the silica content in which the graphs display a substantial decline in strength with increase in silica amount. It can be noticed that both \( M_{\text{real}} \) and \( M_{\text{imag}} \) increase towards high frequencies, which is in accordance with the previous studies earlier reported [23]. The existence of the peak curve in the \( M_{\text{real}} \) at higher frequencies suggests that the synthesized GPEs are good ionic conductors. This could be connected to the bulk effect of the charge carrier and the transformation of the polymer from semi-crystalline to an amorphous region. It is observed that the small values of both \( M_{\text{real}} \) and \( M_{\text{imag}} \) in the low frequency region simplifies the movement of ions. This method of complex dielectric modulus has been used to study the conductivity relaxation phenomena since it has the ability to destroy the effects of electrode polarization to give a clearer picture of electrical property. A long tail observed at low frequency region could be ascribed to the large capacitance that is associated with the blocking electrodes used [25].

It can be observed that, both \( M_{\text{real}} \) and \( M_{\text{imag}} \) increase towards high frequency region. The presence of the peaks at higher frequencies for all the electrolytes suggest that the developed electrolyte are good ionic conductors. Equally, it can be observed that, \( M_{\text{real}} \) and \( M_{\text{imag}} \) decrease with long tail towards low frequencies showing the fact that the electrode polarization phenomena make a negligible contribution.
Similarly, the observed small values for $M_{\text{real}}$ and $M_{\text{imag}}$ in the low-frequency region have been found to simplify migration of ions in the prepared PEs [27].

![Figure 5](image)

**Figure 5.** The plots of (a) $M_r$ and (b) $M_i$ against frequency for PVA-K$_2$CO$_3$-SiO$_2$ samples at ambient temperatures.

### 3.2.4. Temperature dependence of $M_{\text{real}}$ and $M_{\text{imag}}$ at selected temperatures.

Figure 6 (a and b) show the $M_{\text{real}}$ and $M_{\text{imag}}$ of the electric modulus against $\log f$ with respect to temperature in which the graphs display a substantial decline in strength with increase in temperature. From the result in Figure 6 (a), it can be noticed that, at lower frequencies $M_{\text{real}}$ values are very small (almost zero), which indicates the removal of electrode polarization. However, $M_{\text{real}}$ values is observed to be increased with the rise in temperature and frequency reaching a maximum value $M_r$ at high frequency and this reported to be due to the distribution of the relaxation process over a range of frequencies [28].

![Figure 6](image)

**Figure 6.** Temperature dependence of real ($M_{\text{real}}$) and imaginary part ($M_{\text{imag}}$) of $M^*$ for PVA-K$_2$CO$_3$-SiO$_2$ (PKS15) electrolytes at selected temperatures.

Similarly, it is clear that at higher temperature and low frequencies, $M_{\text{imag}}$ displays low value, which could be associated with the large value of capacitance related with the electrode polarization effect due to accumulation of large amount of charge carriers at the electrode-electrolyte interface. The decrease in both $M_{\text{real}}$ and $M_{\text{imag}}$ with further increase in temperature as a function of log frequency comes from the movement of the polymer segments and the carrier charges indicating that the charge carrier has been thermally activated [29].
4. Conclusion

GPEs based on PVA, K$_2$CO$_3$ and SiO$_2$ have been successfully developed in this study. Characterization results based on FTIR analysis confirmed successful interaction and incorporation of salts and silica into the polymer matrix. The study found that, at a region of lower frequency, both $\varepsilon_r$ and $\varepsilon_i$ are observed to decrease with increasing frequency and almost level off at higher frequency region for all electrolytes. The observed improvement in dielectric constant with increase in temperature is primarily due to the decrease in viscosity of the polymeric material. The highest dielectric constant was achieved for electrolyte with 15 wt.% SiO$_2$ (PKS15). Actually, the addition of filler to the polymer matrix creates amorphicity in the polymeric material, hence an increase in $\varepsilon_r$ was observed. Similarly, the presence of relaxation peak in $\mu_{real}$ designates that developed electrolytes in this study are highly ionic conductor. Thus, it can be concluded that dielectric studies is a suitable way to study the dielectric relaxation and charge transport mechanism of electrolyte materials.

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