



IMPACT OF POLYETHYLENE GLYCOL ON CONDUCTIVITY AND DIELECTRIC PROPERTIES OF 2-HYDROXYETHYL CELLULOSE AMMONIUM THIOCYANATE ELECTROLYTES

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ABSTRACT

A sustainable and environmentally friendly approach was used in the development of Solid Biopolymer Electrolyte (SBE), which derived from natural polymers resources. In this study, cellulose-based electrolyte system consists of 2-hydroxyethyl cellulose (2HEC) doped with ammonium thiocyanate (ATC) was further improved by adding polyethylene glycol (PEG) as a plasticiser. The SBE was fabricated via solution casting technique. The addition of PEG into the system remarkably enhanced ionic conductivity up to $1.76 \times 10^{-3} \text{ S} \cdot \text{cm}^{-1}$ at 8 wt.% of PEG, showing an improvement in ion mobility. Electrical properties were investigated through dielectric studies, confirming the ionic nature of the SBE system and the influence of PEG on relaxation behaviour. Temperature-dependent conductivity measurements showed that the system-maintained performance up to 373 K and followed Arrhenius behaviour. These findings highlight the potential of biopolymer-based electrolytes with plasticisers like PEG for sustainable, simple, and environmentally friendly solid-state energy storage applications while demonstrating the importance of material design in achieving high ionic conductivity and thermal stability.

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Introduction

Energy is the capacity to perform work and exists in various forms, including electrical and thermal energy. The principle of energy conservation underpins energy storage technologies which play a critical role in modern energy management by enabling the efficient storage and utilisation of electricity generated from renewable sources. Electrochemical energy storage systems enable the conversion of electrical energy into chemical energy for storage and its subsequent release when required. The increasing use of renewable

energy for green and clean energy has driven batteries to the forefront of scientific research. Since the early development of electrochemical cells in the 1800s, continuous advancements have been made to improve energy density, safety, cost-effectiveness, and environmental compatibility (Dörfler *et al.*, 2021; Sharma *et al.*, 2023).

Electrochemical energy storage systems consist of essential components such as electrolytes where it plays a crucial role in the

overall performance by governing the ionic transport in the device (Rani *et al.*, 2023). Nowadays, Solid Polymer Electrolytes (SPEs) have gained attention due to the advantages in improving safety in terms of mechanical flexibility and elimination of leakage issues associated with liquid electrolytes (Abdulkadir *et al.*, 2022). However, concerns related with fossil resource dependency remained as most of the conventional SPEs are derived from petroleum-based polymers such as poly(ethylene oxide) (PEO) and poly(vinylidene fluoride) (PVDF) (Hossain *et al.*, 2025).

Due to that issues, Solid Biopolymer Electrolytes (SBE) based on renewable materials is currently at the forefront due to its promising alternative in energy storage applications. For example, cellulose-based have been widely used owing to its abundant, biodegradable, and renewable biopolymer, which offers in reducing the environmental impact in comparison with the synthetic polymers (Ghazali & Samsudin, 2022). In addition, the use of cellulose-based materials contributes to improve sustainability as it is more environmentally friendly in disposal pathways. Furthermore, fabrication techniques such as solution casting typically involve relatively low processing temperatures and simple procedures which may reduce energy consumption during material preparation (Manohar & Usha Rani, 2025). This approach is particularly relevant for proton-conducting polymer electrolytes, where minimising processing complexity is desirable for sustainable and scalable production.

Despite its sustainability advantages, cellulose-based SBEs exhibit limited ionic conductivity due to their partial crystallinity, which restricts polymer chain mobility (Ghazali & Samsudin, 2022). Numerous strategies have been taken to optimise the electrolyte's performance through ionic salt doping and incorporation of plasticiser (Majumdar *et al.*, 2021). It is important to choose a suitable ionic salt as it is crucial in providing mobile charge carriers within the polymer matrix. The selection of plasticisers such as ethylene carbonate and propylene carbonate, which are widely used

in the SBE system is also important as it can enhance polymer flexibility, increase free volume, and facilitate ion transport, which leads to the improvement of the system (Ibrahim *et al.*, 2025). In comparison with these plasticisers, polyethylene glycol (PEG) offers additional advantage in polymer chain compatibility due to its ether oxygen groups that can help in coordinating with ionic species and further assist ion migration (Maulana *et al.*, 2021). Studies reported by Samsudin *et al.* (2020) showed that the addition of PEG-plasticised in cellulose-based electrolytes have improved the ionic conductivity up to the range of $\sim 10^{-3} \text{ S} \cdot \text{cm}^{-1}$ at room temperature. However, further study on temperature-dependent electrical behaviour particularly remains limited for 2HEC-ATC SBE system plasticised with various PEG compositions.

The aim of this study is to further examine the effect of PEG as a plasticiser on the electrical properties of 2HEC-based solid biopolymer electrolyte doped with ammonium thiocyanate (ATC) using Electrical Impedance Spectroscopy (EIS), which extends the previous reported study by Ramlli *et al.* (2022). The PEG-enhanced SBE system was evaluated at both room temperature (303 K) and elevated temperature (373 K) to determine the effect of temperature on ionic conductivity. This work aims to provide insight into the role of PEG in influencing electrical performance while contributing to the development of more sustainable cellulose-based solid electrolyte systems.

Materials and Methods

Sample Preparation

2-Hydroxyethyl cellulose (2HEC) was used as the host polymer, ammonium thiocyanate (ATC) as the proton-conducting dopant salt and polyethylene glycol (PEG) as the plasticiser. Detailed chemical compositions, grades, and manufacturer specifications of all materials are summarised in Table 1. All chemicals were analytical or equivalent grade and were used as received without further purification.

Table 1: List of materials

Materials	Chemical Formula/ Specification	CAS No.	Grade	Brand
2-Hydroxyethyl Cellulose (2HEC)	$(C_2H_6O_2)_n$ – substituted cellulose; average molecular weight $\approx 90,000$ g/mol	9004-62-0	Analytical grade (viscosity-based)	ALDRICH
Ammonium Thiocyanate (ATC)	NH_4SCN	1762-95-4	EMSURE [®] ACS/ ISO/Reag. Ph. Eur.	MERCK
Polyethylene Glycol (PEG)	$HO-(CH_2CH_2O)_n-H$; Mn = 400 g/mol	9003-11-6	Analytical grade	ALDRICH

Initially, 2.0 g of 2HEC powder was dissolved in 70 mL of distilled water at room temperature and the solution was stirred until homogeneous mixture was obtained. Subsequently, 36 wt.% of ATC was doped to the solution as this concentration exhibited the highest ionic conductivity in the unplasticised system. For sample preparation, the solution casting technique was utilised based on the method reported by Ramlli *et al.* (2017), with modifications involving variations in the PEG concentration. Following this, PEG was incorporated into the solution using a micropipette and continuously stirred until homogeneity was attained. The PEG content (wt.%) was calculated with respect to the weight of 2-HEC. The corresponding PEG volumes (mL) listed in Table 2 were determined using the density of PEG 400 at room temperature ($\rho = 1.12 \text{ g}\cdot\text{mL}^{-1}$). Six additional samples were prepared using the same procedure with

different PEG contents as summarised in Table 2. The resulting solutions were then cast onto petri dishes, forming thin films and dried in an oven at 328.15 K (55°C) for approximately 10 hours. After drying, the films were stored in desiccators containing silica gel to prevent moisture absorption from affecting the hydrophilic samples.

Sample Characterisation

The ionic conductivity of the SBE was determined through electrical impedance spectroscopy (EIS) across a frequency range of 50 Hz to 1 MHz at temperatures starting from 303 K to 373 K. Each sample was cut into appropriate sizing and was positioned between two stainless steel electrodes in a conductivity cell, which is connected to a computer. The bulk resistance (R_b) value was determined using a Nyquist plot, which depicts imaginary impedance (Z_i) against

Table 2: The composition of PEG for each sample

Labelling	2HEC (g)	ATC (g)	PEG	
			(wt.%)	(mL)
PEG 0	2.000	1.125	0	0
PEG 2			2	0.058
PEG 4			4	0.119
PEG 6			6	0.182
PEG 8			8	0.284
PEG 10			10	0.317
PEG 12			12	0.389

real impedance (Z_r). The ionic conductivity was computed using the following Equation (1). For each composition, measurements were performed on two independent films (different batches), with each film measured five times to ensure reproducibility. The reported ionic conductivity values are presented as mean $\pm$ standard deviation and error bars are included in Figure 5 to reflect measurement variability.

$$\sigma = \frac{t}{AR_b}$$

(1)

A is the electrode-electrolyte contact area of the film (cm^2), R_b is the bulk resistance, and t is the thickness of electrolyte (cm).

Results and Discussion

Figure 1 shows the Nyquist plots for the unplasticised sample (PEG 0) and the highest conducting sample (PEG 8). An inclined spike emerges for both samples, which is attributed to random dipole coordination in the polymer host’s side chains or defined as electrode polarisation due to ion migrations that result in the formation of electrode double layer capacitance (Perumal *et al.*, 2019). The bulk resistance (R_b) is determined from the intercept of the plot on the real axis (Z_r). It is observed that the PEG 8 sample exhibits a smaller intercept, compared to the PEG 0, indicating a lower bulk resistance. This reduction in R_b suggests enhanced ionic mobility and improved charge transport within the polymer electrolyte system upon the addition of PEG, which contributes to higher conductivity of PEG 8 sample. The R_b values

obtained from the Nyquist plots were used to calculate the ionic conductivity of each sample at various temperatures (303 K to 373 K) using Equation (1) and the results are summarised in Table 3. From the table, it is observed that the R_b value decreased alongside the addition of PEG concentration up to an optimum value at 8 wt.%. Upon further PEG addition, the R_b value increased showing an inverse relationship with ionic conductivity.

The ionic conductivity trend of 2HEC-ATC-PEG system can be further understood by considering the plasticisation mechanism. Figure 2 schematically illustrates the effect of PEG on the 2HEC-ATC system, showing the transition from plasticisation at low PEG content [Figure 2 (a)] to over-plasticisation at higher concentrations [Figure 2 (b)]. Previous work reported that the highest ionic conductivity

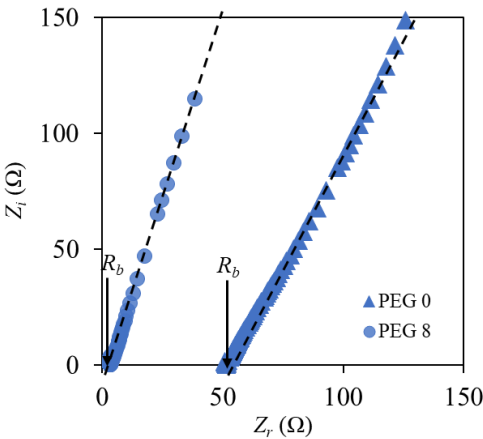


Figure 1: Nyquist plots for PEG 0 and PEG 8

Table 3: Thickness, bulk resistance (R_b), and ionic conductivity (σ) of 2HEC-ATC-PEG samples

Sample	Thickness (± 0.001 cm)	R_b (Ω)	$\sigma \times 10^{-4}$ ($\text{S}\cdot\text{cm}^{-1}$)	Standard Deviation
PEG 0	0.0185	51.3	1.15	3.44×10^{-6}
PEG 2	0.0262	44.1	1.89	2.08×10^{-6}
PEG 4	0.0301	19.9	4.81	3.61×10^{-6}
PEG 6	0.0187	8.6	6.94	5.77×10^{-7}
PEG 8	0.0176	3.2	17.7	9.14×10^{-4}
PEG 10	0.0159	5.0	1.02	5.77×10^{-7}
PEG 12	0.0185	158.0	0.37	2.52×10^{-7}

achieved from doping with 36 wt.% of ATC was $1.16 \times 10^{-4} \text{ S}\cdot\text{cm}^{-1}$ at 303 K (Ramlli *et al.*, 2022). In this work, the ionic conductivity obtained for PEG 0 was $1.15 \times 10^{-4} \text{ S/cm}$, which is close to the value reported in previous work. In this plasticised system of 2HEC-ATC, the conductivity increased when added with 2 wt.% of PEG ($1.89 \times 10^{-4} \text{ S}\cdot\text{cm}^{-1}$) and continued to increase until $1.76 \times 10^{-3} \text{ S}\cdot\text{cm}^{-1}$ at 8 wt.% of PEG.

At lower PEG concentrations [Figure 2 (a)], the increase in conductivity is attributed to the plasticiser enhancing polymer chain flexibility and promoting longer-range segmental motion, which facilitates ion mobility and increases free volume by embedding itself between polymer chains (Hamsan *et al.*, 2020). Additionally, the high dielectric constant of PEG facilitates ion dissociation, generating more free ions and further enhancing conductivity (Sadiq *et al.*, 2022). However, at higher PEG contents [Figure 2 (b)], over-plasticisation occurs, leading to stronger ion-ion interactions that promote the formation of ion pairs and aggregates. These clusters reduce the number of free mobile ions, create localised regions of higher viscosity, hinder ion hopping, and disrupt conductive pathways, collectively lowering conductivity (Ponraj *et al.*, 2020). Based on the schematic, the mechanism highlights how an optimal PEG content balances polymer chain flexibility and ion

dissociation to allow efficient ion transport. This mixed behaviour, where excessive plasticiser leads to ion clustering and reduced conduction has been widely reported in plasticised polymer electrolyte (Aziz *et al.*, 2025).

Figure 3 shows the Nyquist plots for highest conducting sample, PEG 8 at various temperatures (303 K to 373 K). Based on the figure, the R_b values can be observed to decrease as temperature increased. Similar trends were observed for the remaining samples. The ionic conductivity for all samples across the temperatures was calculated and plotted in Figure 4 while the corresponding regression (R^2) values were tabulated in Table 4.

Although SBE system follows Arrhenius behaviour, the R^2 values (Table 4) indicate minor deviations from ideal linearity. This may be attributed to the increased segmental mobility caused by the addition of plasticiser into the system which enhances flexibility of polymer chain that facilitates the ion transport through segmental relaxation processes. As a result, ionic conduction may exhibit characteristics associated with the Vogel–Tammann–Fulcher (VTF) model under such conditions, where ion mobility is linked with polymer motion rather than solely governed by thermally activated hopping (Dennis *et al.*, 2023). Hence, this implies that both Arrhenius-type and segmental

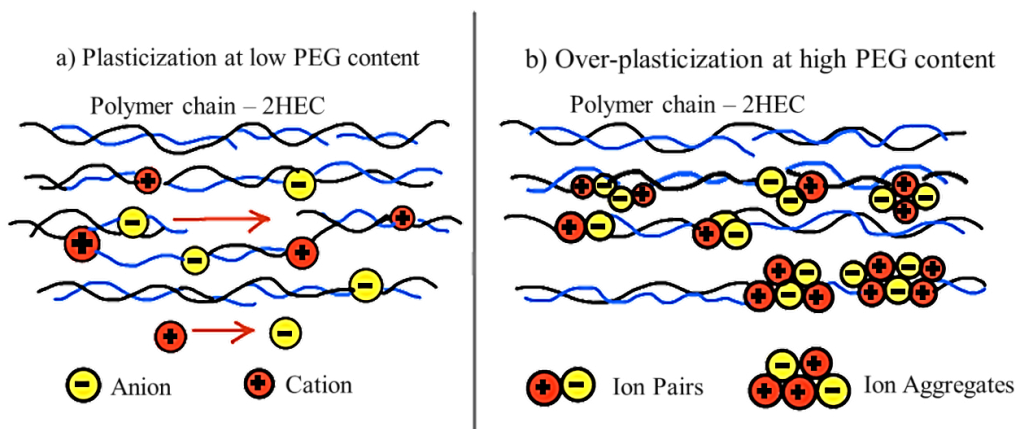


Figure 2: Schematic illustration of PEG plasticisation in the 2HEC–ATC solid biopolymer electrolyte system: (a) Plasticisation at low PEG content (≤ 8 wt.%) and (b) over-plasticisation at high PEG content (> 8 wt.%)

motion type involves in the contribution of the transport mechanism in SBE system. Based on the reported study by Das *et al.* (2024), such behaviour which leads to deviations from ideal linearity is commonly reported in plasticised polymer electrolytes. Despite that, Arrhenius model provides a reasonable approximation to study the understanding of temperature-dependence and activation energy in SBE system (Mazuki *et al.*, 2020).

The experimental temperature range from 303 K to 373 K are taken to consideration in this study. According to Geng *et al.* (2024) and Shin *et al.* (2025), 2HEC is reportedly decomposes within the range of 200°C to 300°C while PEG degradation occurs at higher temperatures approximately around 400°C. In addition, polymer electrolyte membranes containing ATC salt have been reported to remain thermally stable up to approximately 200°C (R. *et al.*, 2025). Hence, the conductivity enhancement at elevated temperatures up to 373 K is considered to arise from thermally activated ion transport processes rather than thermal decomposition.

Further investigation on temperature-dependence analysis was evaluated through the calculation of activation energy, E_a using Arrhenius equation in Equation (2). In polymer system, E_a is referred to the energy required in

breaking the bonds in molecules and the energy needed for its movement to the donor sites (Khan *et al.*, 2020). Table 4 shows the results of the data which were summarised in Figure 5.

$$\sigma = \sigma_0 e^{\left(-\frac{E_a}{kT}\right)} \quad (2)$$

In Figure 5, E_a values can be seen decreased as conductivity increased where the highest conductive sample (PEG 8) possesses the lowest E_a (0.07 eV). This behaviour can be attributed to the introduction of PEG into the polymer matrix, which creates shortcuts in the polymer

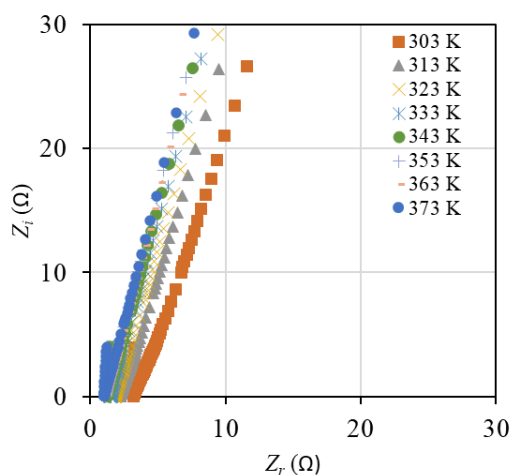


Figure 3: The Nyquist plots for highest conducting sample (PEG 8) at 303 K to 373 K

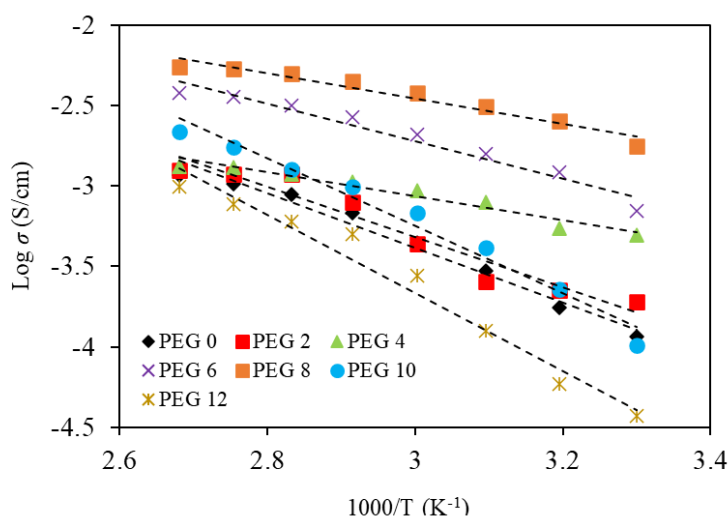


Figure 4: Graph of temperature-dependent for all SBE

Table 4: Regression, R^2 , and E_a values of each sample

Sample	R^2 (Ω)	E_a (eV)
PEG 0	0.98	0.15
PEG 2	0.94	0.13
PEG 4	0.95	0.12
PEG 6	0.96	0.10
PEG 8	0.95	0.07
PEG 10	0.97	0.18
PEG 12	0.97	0.21

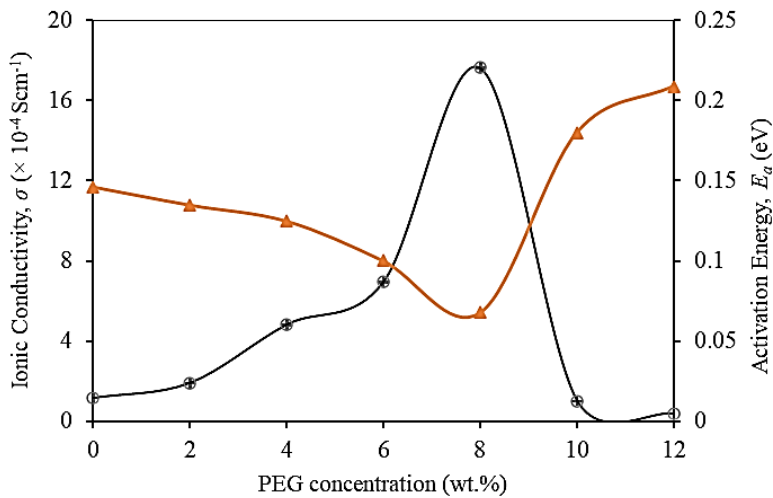


Figure 5: Ionic conductivity and activation energy versus PEG concentration at 303 K

network, reducing the energy required for ions to hop between donor sites (Samsudin & Saadiah, 2018). Additionally, PEG enhances salt dissociation, increasing the number of free ions available for migration, which further facilitates ion transport and contributes to the observed reduction in E_a (Saadiah *et al.*, 2020). These results indicate that PEG not only improves ionic conductivity but also reduces the energy barrier for ion transport, highlighting its effectiveness as a plasticiser in the SBE system.

Dielectric constant, ϵ_r refers to the measure of the system's charge storage capability meanwhile dielectric loss, ϵ_i refers to the measurement of energy losses resulting from ion movements in a fast-varying electric field (Choudhary & Sinha, 2022). Dielectric constant (ϵ_r) and dielectric loss (ϵ_i) were obtained using

Equations (3) and (4). The ϵ_r and ϵ_i values of all SBE samples at different salt compositions were plotted in in Figures 6 and 7, respectively.

$$\epsilon_r = \frac{Z_i}{\omega C_o(Z_r^2 + Z_i^2)} \quad (3)$$

$$\epsilon_i = \frac{Z_r}{\omega C_o(Z_r^2 + Z_i^2)} \quad (4)$$

Based on Figures 6 and 7, both ϵ_r and ϵ_i decreased with increasing frequency with the highest values appeared at low frequency regions. This trend is attributed to the accumulation of charges at the electrode interface, which is more prominent at lower frequencies, allowing ions to respond effectively to the applied field. The increase in free mobile ions resulting from salt dissociation, enhanced by the PEG

plasticiser, leads to a corresponding rise in ionic conductivity, as observed for sample PEG 8, which exhibits the highest ϵ_r and ϵ_i values. As frequency increases, the electric field reverses too quickly for ions to follow, reducing charge accumulation, hence, lowering ϵ_r and ϵ_i , consistent with the decline in conductivity at high frequencies (Asnawi *et al.*, 2021; Fuzlin *et al.*, 2022).

The frequency-dependent analysis of ϵ_r and ϵ_i analysis for selected samples at various temperatures (303 K to 373 K) was carried out and plotted in Figures 8 and 9, which represents the charge transport behaviour of the system. Both ϵ_r and ϵ_i pattern can be seen increased with temperature, reflecting enhanced polymer chain

segmental motion and greater ion mobility, which facilitates long-range ion transport and improves ionic conductivity (Swady & Jawada, 2021). Additionally, relaxation peaks in the ϵ_i spectra (Figure 9), correspond to transitions from long-range to short-range ionic motion, further confirming that the dielectric response is directly linked to the ion hopping mechanism in this PEG-plasticised system (Shetty *et al.*, 2021).

The reciprocal of dielectric components, ϵ_r and ϵ_i are the electrical modulus, real modulus (M_r), and imaginary modulus (M_i). The M_r and M_i values for each sample at temperature from 303 K to 373 K were calculated using Equations (5) and (6). The M_r and M_i values of

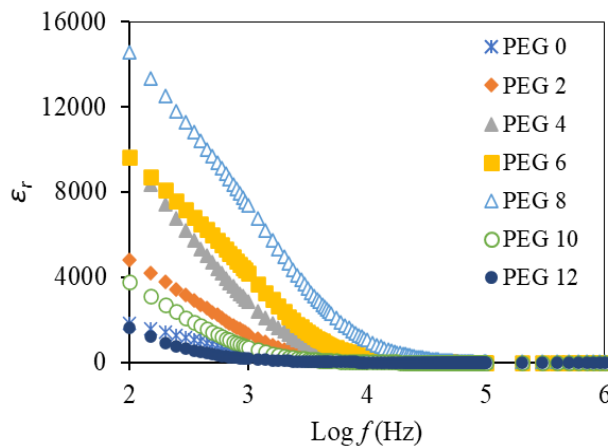


Figure 6: ϵ_r frequency dependence of all SBE at 303 K

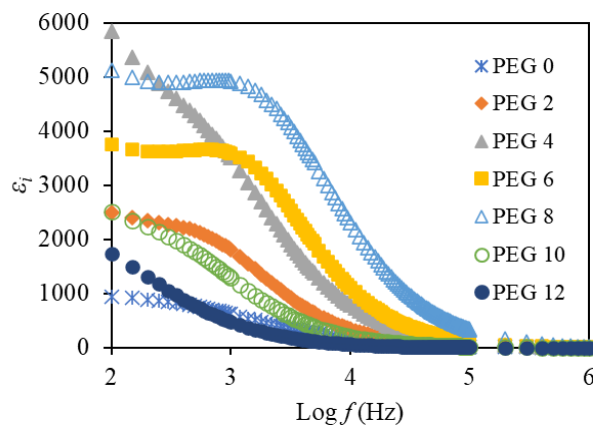


Figure 7: ϵ_i frequency dependence of all SBE at 303 K

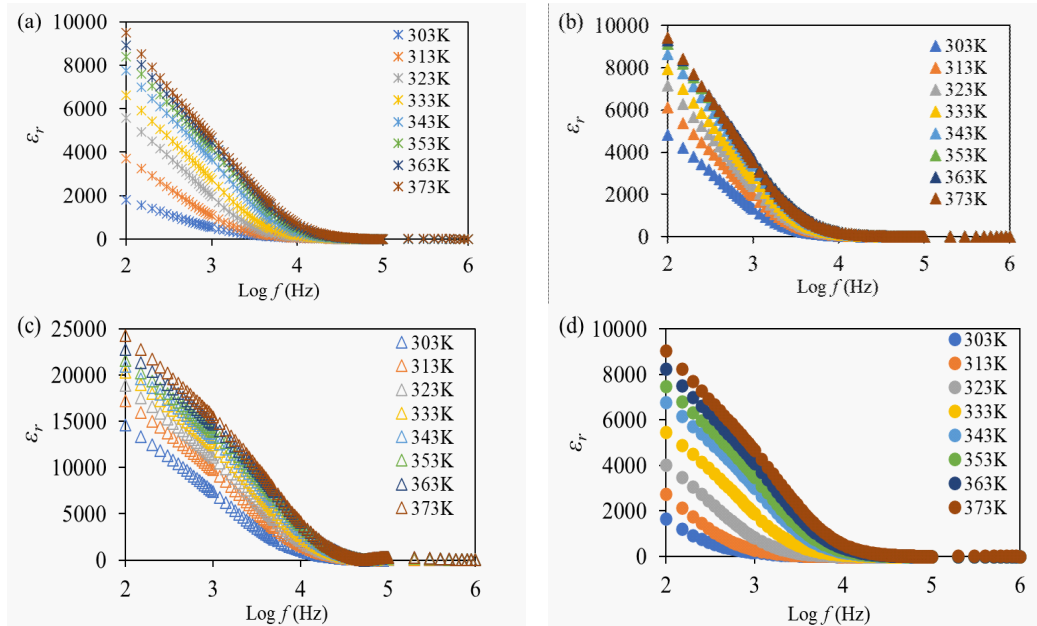


Figure 8: ϵ_r frequency dependence for (a) PEG 0, (b) PEG 4, (c) PEG 8, and (d) PEG 12 at various temperatures

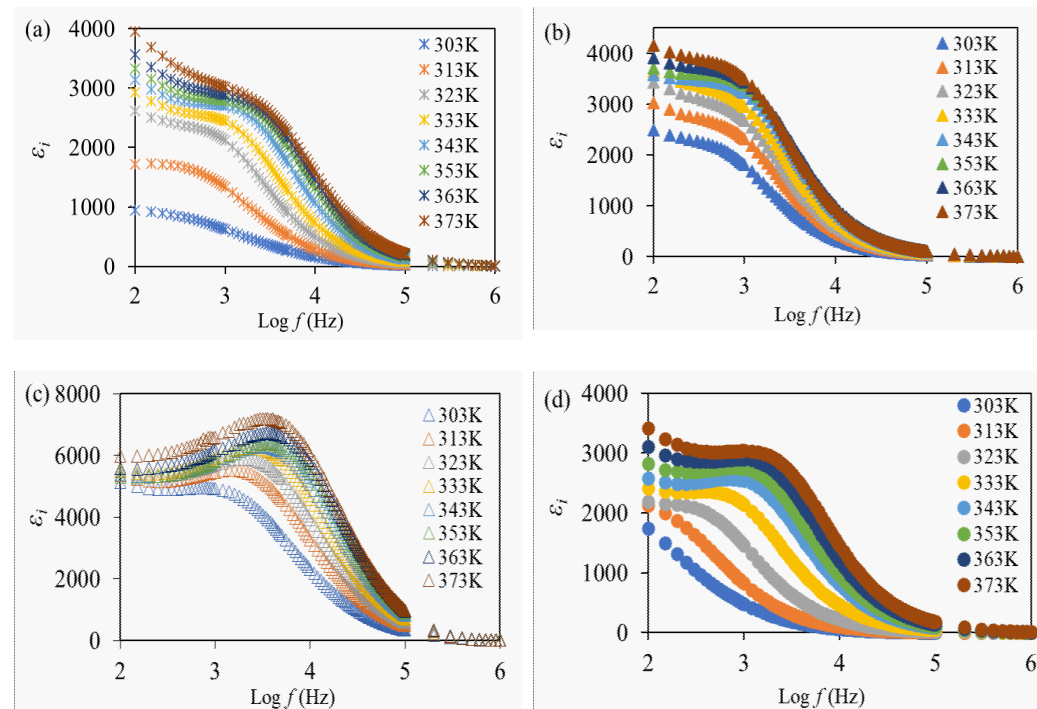


Figure 9: ϵ_i frequency dependence for (a) PEG 0, (b) PEG 4, (c) PEG 8, and (d) PEG 12 at various temperatures

the selected samples at different temperatures were plotted and are presented in Figures 10 and 11, respectively.

$$M_r = \frac{\epsilon_r}{(\epsilon_r^2 + \epsilon_i^2)} \quad (5)$$

$$M_i = \frac{\epsilon_i}{(\epsilon_r^2 + \epsilon_i^2)} \quad (6)$$

Based on Figures 10 and 11, both M_r and M_i values are nearly zero at low frequencies and increase with increasing frequency. The rise in M_r with frequency produces distinct peaks, indicating that at high frequencies, charge carriers are constrained to potential wells and can only move over short distances (Faris *et al.*, 2021). At low frequencies, the long tail observed corresponds to high capacitance at the electrode (Abdullah *et al.*, 2021). The charge carriers were able to move freely over a long distance in the lower frequency region, indicating the successful hopping of charge carriers from one site to another (Sengupta *et al.*, 2020). The increase in M_r and M_i at intermediate to high frequencies reflects restricted ion motion as ions respond less effectively to the rapidly alternating electric

field (Maddu *et al.*, 2022). Furthermore, at higher temperatures, both M_r and M_i decreased, which can be attributed to enhanced polymer segmental motion and increased mobility of charge carriers. This facilitates easier alignment of molecular dipoles and more efficient long-range ion transport, consistent with the observed improvement in ionic conductivity at elevated temperatures (Liu *et al.*, 2019).

Conclusions

This study demonstrated that incorporating PEG as a plasticiser significantly enhances the ionic conductivity of 2HEC-ATC solid biopolymer electrolytes. The system containing 8 wt.% of PEG showed the highest conductivity, increasing from $1.16 \times 10^{-4} \text{ S} \cdot \text{cm}^{-1}$ to $1.76 \times 10^{-3} \text{ S} \cdot \text{cm}^{-1}$, attributed to increased polymer chain segmental motion that facilitates ion hopping. The ionic conduction of the system exhibited Arrhenius behaviour and the most conductive sample had the lowest activation energy ($E_a = 0.07 \text{ eV}$). The factors governing conduction of

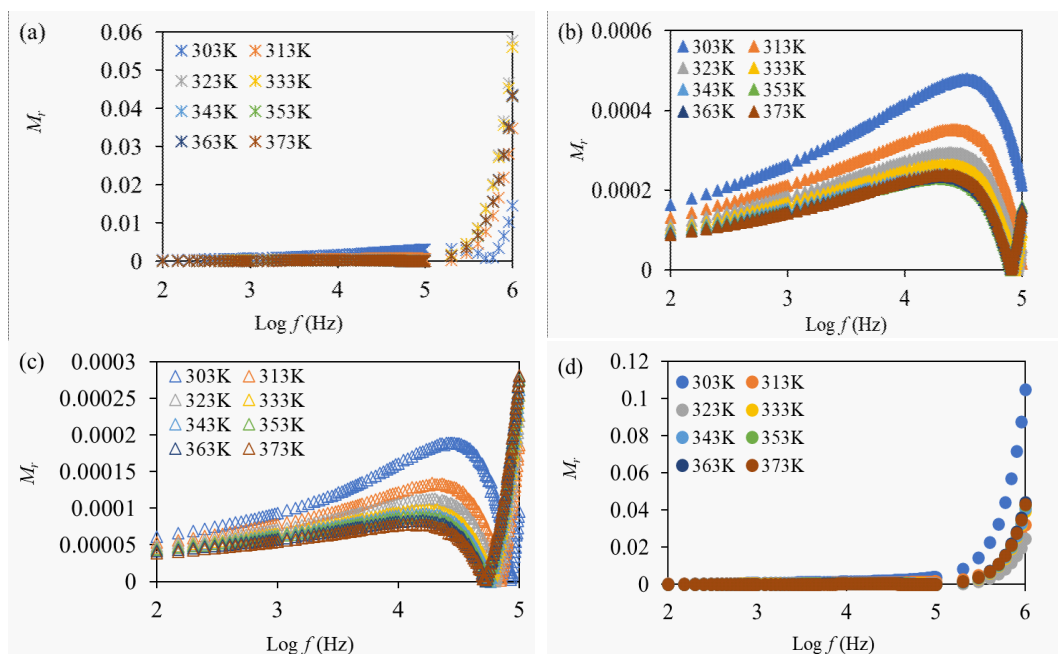


Figure 10: M_r frequency dependence for (a) PEG 0, (b) PEG 4, (c) PEG 8, and (d) PEG 12 at various temperatures

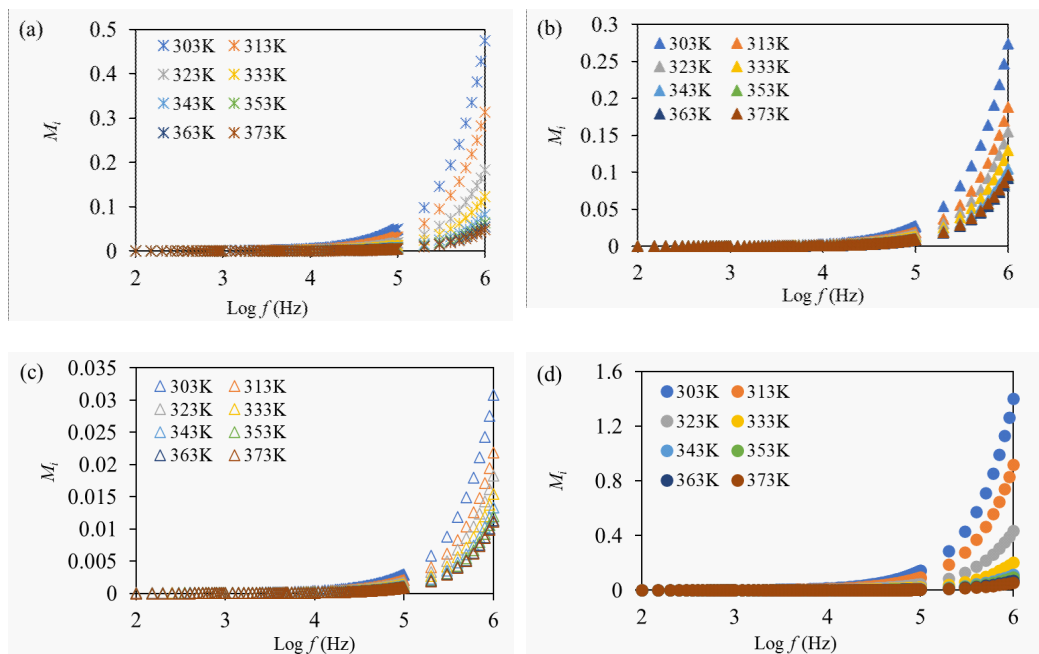


Figure 11: M_i frequency dependence for (a) PEG 0, (b) PEG 4, (c) PEG 8, and (d) PEG 12 at various temperatures

ion was further confirmed through the dielectric and modulus analyses, where ion dissociation and ionic mobility were the dominant factors. Due to the primarily focus on electrical properties of the plasticised SBE system in this current work, further study on mechanical properties such as tensile strength and structural stability is recommended to consider its practical applicability in future study. Overall, these findings contribute to the fundamental understanding of plasticisation effects in cellulose-based solid biopolymer electrolytes and provide useful guidance for the development of advanced solid-state electrochemical energy storage materials.

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Conflict of Interest Statement

The authors declare that there is no conflict of interest in the design of study, in the data collection, analyses, or interpretation data, in the writing of the manuscript, or in the decision in publishing the results.

Author Contributions Statement

NMB and WAM carried out the experimental work, performed data analysis, and prepared the original draft of the manuscript. MNF and MIH assisted in data interpretation, manuscript writing, and revisions. MM provided theoretical guidance and supported result validation. ASS contributed in provided critical revisions and reviewed the manuscript. MINI conceptualised the research idea, designed the research framework, supervised the project, and approved the final version for submission.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author, Mohd Ikmar Nizam Isa, upon reasonable request.

Ethical Conduct

All information communicated in this article does not need ethical approval for research or informed consent because human or animal subjects were not involved.

Declaration of AI-usage

During the preparation of this manuscript, the authors used AI-assisted tools to improve grammar and clarity of the text. The authors reviewed and edited the content thoroughly and take full responsibility for the final content of the manuscript.

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