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Scientific Paper
ISSN 0351-9465, E-ISSN 2466-2585
https://doi.org/10.62638/ZasMat1050

Zastita Materijala 65 (4)
703 – 711 (2024)

Structural, Electrochemical, and Dielectric Studies of Phytagel and 1-ethyl-3-methylimidazolium Tricyanomethanide-based Bio-polymer Electrolytes

ABSTRACT

The present work is focused on the synthesis and detailed study of biopolymer phytagel and ionic liquid 1-ethyl-3-methylimidazolium tricyanomethanide (EMIm[TCM]) blended polymer electrolyte films for energy applications. Here, biopolymer phytagel-based polymeric films are synthesized with different concentrations of ionic liquid (EMIm[TCM]) using the solution cast technique. The synthesized films are characterized for their structural, electrochemical, and dielectric properties using different characterization tools i.e., XRD, FTIR, Electrochemical Impedance Spectroscopy, Linear Sweep Voltammetry, and Wagnor polarization technique. The film with 30wt% EMIm[TCM] shows a maximum conductivity of 3.64 × 10-4 S cm-1 and an electrochemical stability window of 3.1 V. The dielectric properties such as dielectric constant (κ), dielectric loss tangent (tanδ), relaxation time, and frequency are also studied for the prepared pure phytagel and phytagel/EMIm[TCM] polymeric films.

Keywords: *Biodegradable polymers, Phytagel, Ionic liquid, 1-ethyl-3-methylimidazolium tricyanomethanide, Polymer electrolyte*

1. INTRODUCTION:

Electrochemical devices such as batteries, supercapacitors, dye-sensitized solar cells (DSSCs), etc. mostly consist of electrodes, electrolytes, and separators [1]. Electrolyte is one of the key components which is responsible for the performance of any electrochemical device [2]. Traditionally, liquid electrolytes such as H_2SO_4, KOH, $ZnCl_2$, $ZnSO_4$, NaCl, etc are being used in electrochemical devices that have certain limitations such as – leakage, evaporation, bulky design, etc. which affect the performance of the devices [3, 4]. To overcome such limitations, polymer electrolytes can be taken into consideration. Polymer electrolytes are mostly containing a host polymer, ionic species (salts, ionic liquids, fillers, etc.), and a plasticizer [5–7]. Based on their origin, polymers can be divided into two types i.e., synthetic polymers and natural polymers. Most of the research is done on polymer electrolytes based on synthetic polymers like – PVDF-HFP, PEO, PMMA, PEMA, etc. which are made up of petroleum resources and take a lot of time to degrade [8]. Taking this limitation of synthetic polymers into consideration, natural bio-degradable polymers such as corn starch, agarose, phytagel, etc. become a new interest for researchers [9]. Biopolymers are environment friendly and abundant in nature that is why a lot of research is going on to develop solid or gel polymer electrolytes using natural polymers as a host material. The biopolymer-based electrolytes which are doped with only salts show less ionic conductivity, mostly in the order of ~10^{-5} S cm^{-1} [10]. To increase the ionic conductivity of the biopolymer-based electrolytes certain approaches can be followed such as making blends or composites, doping of fillers or

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Paper received: 30.03.2024.
Paper accepted: 5.06.2024.
Papir is available on the website: www.idk.org.rs/journal

nanofillers, co-polymerization, or using ionic liquids as ionic species in polymers, etc [10–14]. Ionic liquids are the salt that have a low vapor pressure and they are non-flammable in nature. Mostly ionic liquids are present in molten state at temperatures less than 100°C [15]. Along with high ionic conductivity they possess high chemical and electrochemical stability [16]. Ionic liquids behave as ionic species as well as a plasticizer [17]. Various biopolymers have been used as a host-polymer for the preparation of a bio-polymer-based polymer electrolyte such as arrowroot [18], corn starch [19], chitosan [20], agar-agar [21], carrageenan [22], gellan gum [23], pectin [24], methyl cellulose [25], etc. In the present study, a biopolymer phytagel is chosen as a host polymer because of its low cost, environment-friendly and non-hazardous nature, thermal resistance, easy chemical modifications, and ability to form films [26]. Ionic liquid- 1-ethyl-3-methylimidazoliumtricyanomethanide (EMIm[TCM]) has been used as an ionic species as well as a plasticizer to increase the ionic conductivity of the polymeric film. The polymer electrolyte films are synthesized by blending phytagel with different weight percentages of EMIm[TCM] and then characterized for structural, optical, and electrochemical properties using different characterization tools such as X-ray diffractometer (XRD), Fourier transform infrared spectroscopy (FTIR), Electrochemical impedance spectroscopy (EIS), Linear sweep voltammetry (LSV), Wagnor polarization technique (t_{ion}). Dielectric studies such as dielectric constant, dielectric loss tangent (tanδ), relaxation time and frequency, etc. are also done.

2. EXPERIMENTAL:

2.1. Preparation of polymer electrolytes.

Phytagel and Dimethyl sulfoxide (DMSO, purity ≥ 99.9%) are procured from Sigma-Aldrich and EMIm[TCM] is procured from Tokyo Chemical Industry Co. Ltd. First, host biopolymer phytagel (200 mg) is dissolved into DMSO with constant stirring using magnetic stirrer for 12 hours at 60°C, then different weight percent of ionic liquid EMIm[TCM] is added to the solutions and again stirred for 12 hours at room temperature. After complete dissolution, all the solutions are poured into glass petri dishes and kept in a vacuum oven at 60°C until the complete evaporation of common solvent (DMSO). After the complete evaporation of the solvent, the polymeric films are peeled out from the petri dishes and stored in a vacuum for further characterization. After 40wt% concentration, the ionic liquid starts to come out from the polymeric films that's why the films are only synthesized with 0wt% to 40wt% blending of ionic liquid.

2.2. Instrumentation.

X-ray differentiation (XRD) patterns of the phytagel-based polymeric films were performed using XPERT-Pro X-ray diffractometer with Cu-Kα radiation (λ = 1.54 A°) in the range from 10° to 80°. FTIR spectroscopy was performed by PerkinElmer Spectrum version 10.4.00. The electrochemical impedance spectroscopy (EIS), Linear sweep voltammetry (LSV), Wagnor polarization technique, and dielectric studies of the polymeric films were carried out by using an electrochemical workstation (CHI-604D, USA).

3. RESULT AND DISCUSSION.

3.1. Structural Studies

X-ray diffraction-

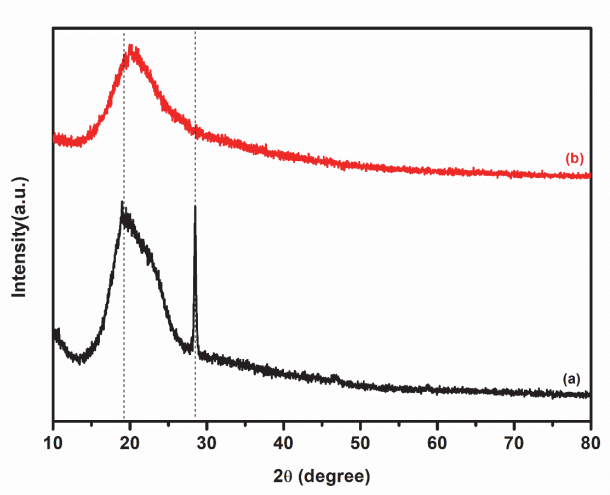

Figure 1. XRD plots of: (a) pure phytagel and (b) phytagel/30wt%EMImTCM film

To study the structural properties of the polymeric films, X-ray differentiation is performed using XPERT-Pro X-ray diffractometer with Cu-Kα radiation (λ = 1.54 A°) in the range from 10° to 80° as shown in figure 1. The XRD patterns for pure phytagel show two predominant peaks, a broad peak at 19.14° and a sharp peak at 28.5° which indicates the semicrystalline nature of the host polymer phytagel [26]. From the XRD patterns of phytagel/EMIm[TCM] it can be seen that the sharp peak present in phytagel at 28.5° disappears and the peak at 19.14° becomes more broad which shows the decrease in the crystalline nature of phytagel on adding of EMIm[TCM] ionic liquid in it.

Fourier Transform Infrared Spectroscopy (FTIR):

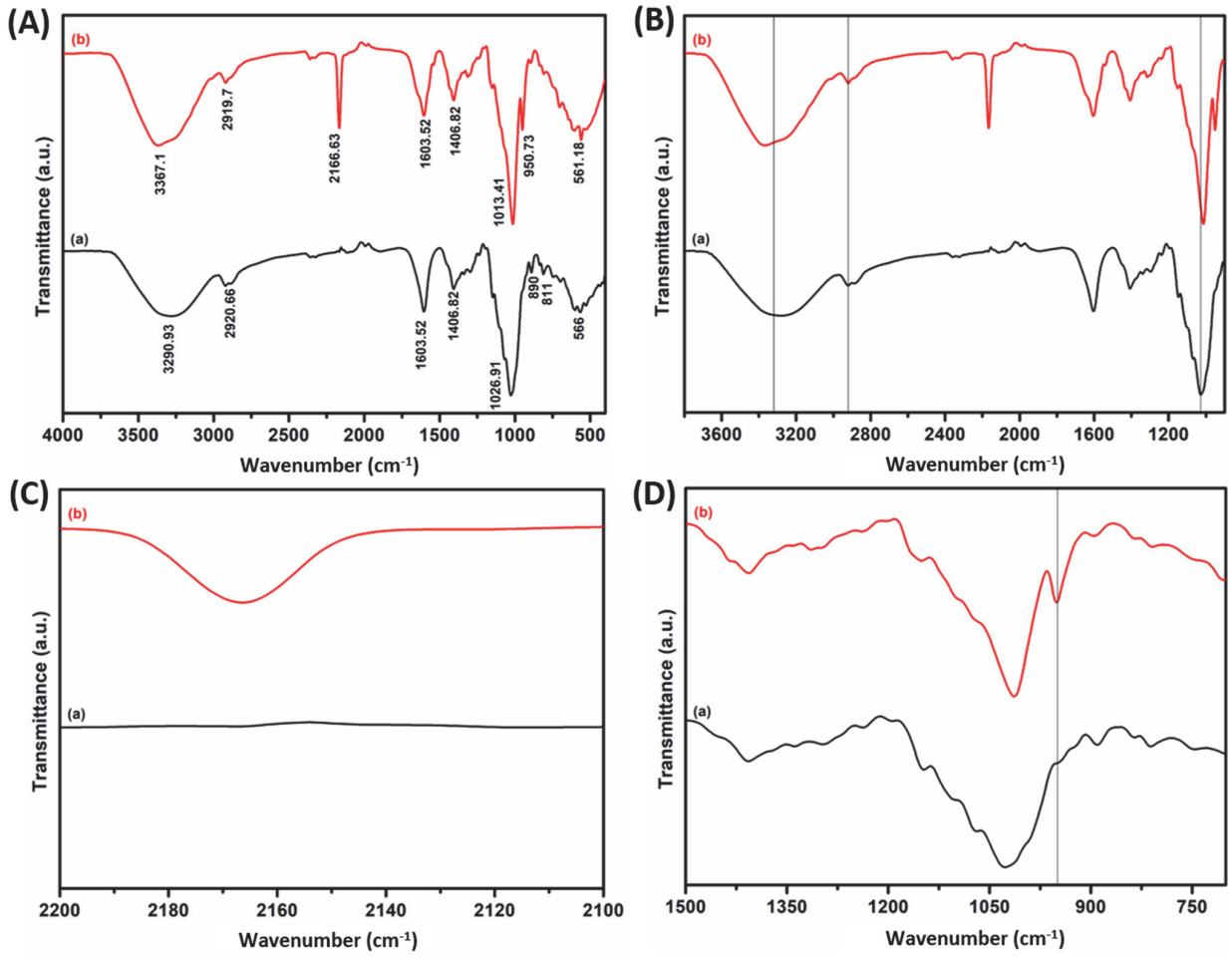

Figure 2. (A) FTIR spectra and its expanded representation in (B) 900 cm^{-1} to 3800 cm^{-1}, (C) 2100 cm^{-1} to 2200 cm^{-1}, and 800 cm^{-1} to 1500 cm^{-1} for (a) pure phytagel and (b) phytagel/30wt%EMIm[TCM]

FTIR spectroscopy has been used to determine the complexation and interaction between biopolymer and ionic liquid doped biopolymer. For this a FTIR spectroscopy of pure phytagel and maximum conducting EMIm[TCM] blended phytagel polymer films has been caried out using PerkinElmer Spectrum version 10.4.00 as shown in figure 2(a). Table 1 shows some important peaks related to host polymer phytagel and ionic liquid EMIm[TCM]. Pure phytagel shows two characteristic peaks related to O-H starching of alcohol at 3290, 2920 cm^{-1}, also it shows two peaks representing the C=O stretching and C=C bending of alkene in phytagel at 1603 and 1026 cm^{-1} respectively and at 1406 it shows a sharp medium peak showing the C-H bending of alkanes. It can be seen from the graph that the peaks at 3290, 2920 and 1026 cm^{-1} are shifted to 3367, 2919 and 1013 cm^{-1} in phytagel/EMIm[TCM] (figure 2B) along with this it shows two new peaks at 2166 and 950 cm^{-1} showing the C≡N of anion [C(CN3)]$^{-}$ and C=C bending of cation EMIm^{+} of ionic liquid respectively [28, 29]and the archetypal channel material is poly(3,4-ethylenedioxythiophene (figure 2 C&D). These observations clearly state that almost all the FTIR peaks of host polymer phytagel are present in phytagel/EMIm[TCM] films without disappearing of any major peaks also showing presence of two significant peak related to the ionic liquid EMIm[TCM], which clearly shows the complex nature of the samples and the considerable changes occurs due to interaction of ionic liquid ions [30].

Table 1. Functional groups corresponding to the different FTIR bands present in FTIR spectra of pure phytagel and phytagel/30wt%EMIM[TCM].

IR bands (cm^{-1})		Functional group (Mode of Vibration/Chemical bond)
Phytagel	3290	O-H stretching, alcohol
	2920	O-H stretching, alcohol
	1603	N-H bending, amine
	1406	C-H bending, alkane
	1026	C-N stretching, amine
$EMIm^+$	1013	C=C bending, alkene
	561	Ring in-plane sym. bending
$[C(CN_3)]^-$	2166	C≡N

3.2. Dielectric and Electrochemical Studies

Electrochemical Impedance Spectroscopy- To study the electrochemical properties of prepared polymer electrolytes, electrochemical impedance spectroscopy (EIS) measurement has been done for the prepared polymeric films consisting different concentration of ionic liquid (0-40 wt%). The impedance spectroscopy of the cell: SS | Phytagel/EMImTCM | SS (SS is stainless steel electrodes) has been done in the frequency range of 100 Hz to 1 MHz. Using the EIS data, Nyquist plots are drawn for all the polymeric films with 0wt% to 40wt% ionic liquid. The Nyquist plot for phytagel/30wt%EMIm[TCM] is shown in figure 3A. Ionic conductivity for all the films has been calculated from Nyquist plot using the following equation (Eq. 1)-

$$\sigma = \frac{1}{R_b} \times \frac{t}{A} \qquad (1)$$

Where, σ is ionic conductivity, Rb is bulk resistance, t is thickness of the polymeric film and A is the area of contact. The calculated ionic conductivity of pure phytagel film, ionic liquid and phytagel/EMIm[TCM] film is shown in figure 3B and listed in table 2. The ionic conductivity increases with the increase in the concentration of ionic liquid in polymer films and it attains a maximum at 30wt% of IL and after that it starts to decrease (figure 3B). It is well known that the expression for conductivity is $\sigma = nq\mu$ where, σ is conductivity, n is number of charge carriers, q is electronic charge and μ is mobility, in present case the ionic conductivity increases because the n and μ are increasing as ionic liquid is working as an ionic species as well as plasticizer (which provides easy passes to ions). The decrease in conductivity beyond blending of 30wt% ionic liquid is may be due to the steric hindrance in between ions which causes obstacle in ionic movement on the way to the respective electrodes [31].

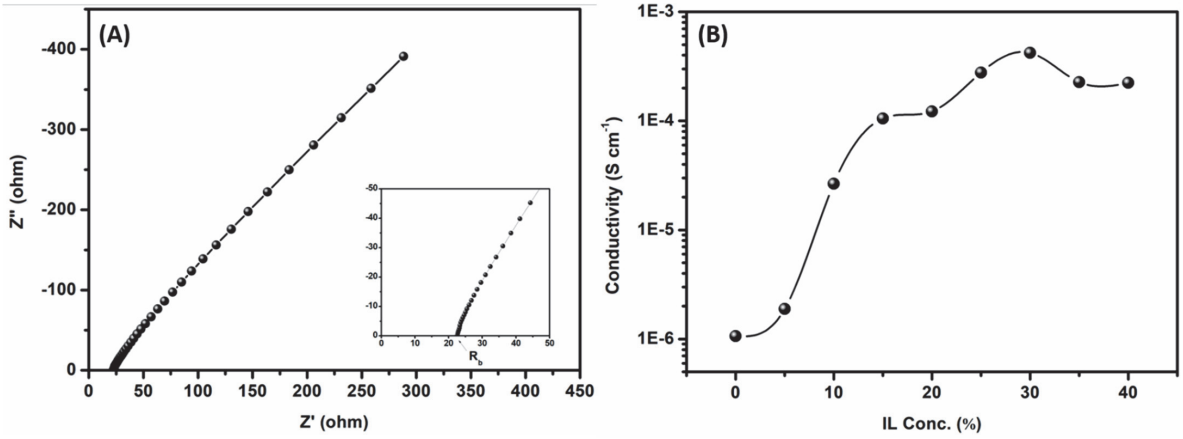

Figure 3. (A) Nyquist plot of phytagel/30wt% EMIm[TCM], (B) Ionic conductivity vs EMIm[TCM] concentration

Table 2. The ionic conductivity, Relaxation frequency, and Relaxation time corresponding to polymeric films consist of different weight percent of ionic liquid in phytagel.

Polymeric film	Ionic conductivity (S cm^{-1})	Relaxation frequency (f_{max}, Hz)	Relaxation time ($\tau = \frac{1}{2\Pi f_{max}}, sec$)
Pure Phytagel	1.06 × 10^{-6}	227	7.01×10^{-4}
Phytagel/EMImTCM (5wt%)	1.88 × 10^{-6}	16883	9.43×10^{-6}
Phytagel/EMImTCM (10wt%)	2.65 × 10^{-5}	147186	1.08×10^{-6}
Phytagel/EMImTCM (15wt%)	1.05 × 10^{-4}	560389	2.84×10^{-7}
Phytagel/EMImTCM (20wt%)	1.22 × 10^{-4}	191147	8.33 ×10^{-7}
Phytagel/EMImTCM (25wt%)	2.77 × 10^{-4}	354441	4.49×10^{-7}
Phytagel/EMImTCM (30wt%)	3.64 × 10^{-4}	311648	5.10×10^{-7}
Phytagel/EMImTCM (35wt%)	2.26 × 10^{-4}	357695	4.45×10^{-7}
Phytagel/EMImTCM (40wt%)	2.23 × 10^{-4}	429211	3.70×10^{-7}

Dielectric studies: Dielectric studies are necessary to explore the dissipation of electric energy in various optical and electronic devices. The dielectric constant values are calculated at three different frequencies i.e., 825200, 99610, and 8301 Hz using the formula $\kappa = \frac{C}{C_o}$, where, κ is dielectric constant, C is capacitance and C_o is the capacitance at vacuum. The variation of dielectric constant for all polymeric film with respect to different wt% of ionic liquid shown in figure 4. It can be clearly seen from figure 4 that dielectric constant is also following almost the same trend as ionic conductivity with respect to concentration of ionic liquid. The increase in dielectric constant trend with respect to increase in ionic liquid concentration is indicating that the major charge carriers in the polymer electrolyte films are ions.

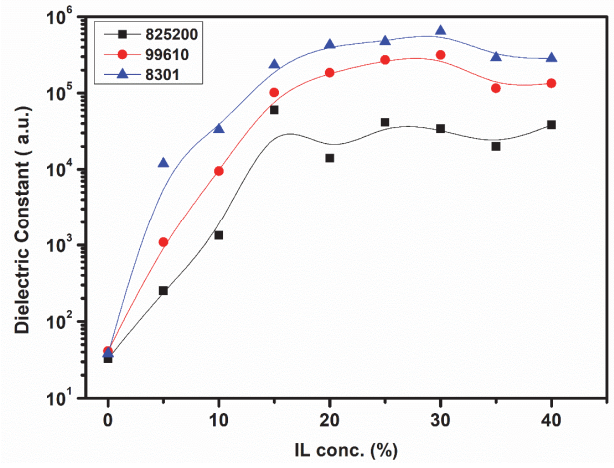

Figure 4. Variation of dielectric constant with respect to EMIm[TCM] concentration at 825200, 99610, 8301 Hz.

The complex permittivity (ε*) provide a detail information about the polarized mechanism which is expressed by the following equation (Eq. 2)-

$$\varepsilon^* = \varepsilon' - j\varepsilon'' \quad (2)$$

Where ε' and ε'' are representing the real and imaginary parts of complex permittivity, which is expressed by Eq. (3 & 4)

$$\varepsilon' = \frac{-Z''}{\omega C_o(Z'^2+Z''^2)} \quad (3) \text{ and } \varepsilon'' = \frac{-Z'}{\omega C_o(Z'^2+Z''^2)} \quad (4)$$

Where, z' and z'' are the real and imaginary part of impedance, ω = 2π*f* is the angular frequency, C_o= (ε_oA/t), A is cross-section area, t is the thickness of polymer electrolytes. Figure 5 represents the variation of ε' and ε'' with frequency for polymeric films with Ionic liquid concentration of 0wt% to 40wt%. Figure 5 A&B reveals that and has high values at lower frequencies, which may be due to the alignment of dipoles with applied field and accumulation of charges near blocking electrode-electrolyte interface (space charge effect). The ε' and ε'' starts to decrease with increasing in frequency, since the dipoles are not able to align with the rapid changing of applied AC field also, with the increasing frequency the diffusion of ions does not take place at available sites [32]. The value for ε' and ε'' is also vary with different concentration of EMIm[TCM], the highest value for ε' and ε'' is comes out for the polymer electrolyte film with 30wt% of EMIm[TCM]. It may be because of the increase in charge carriers and mobility of ions with increase in concentration of ionic liquid.

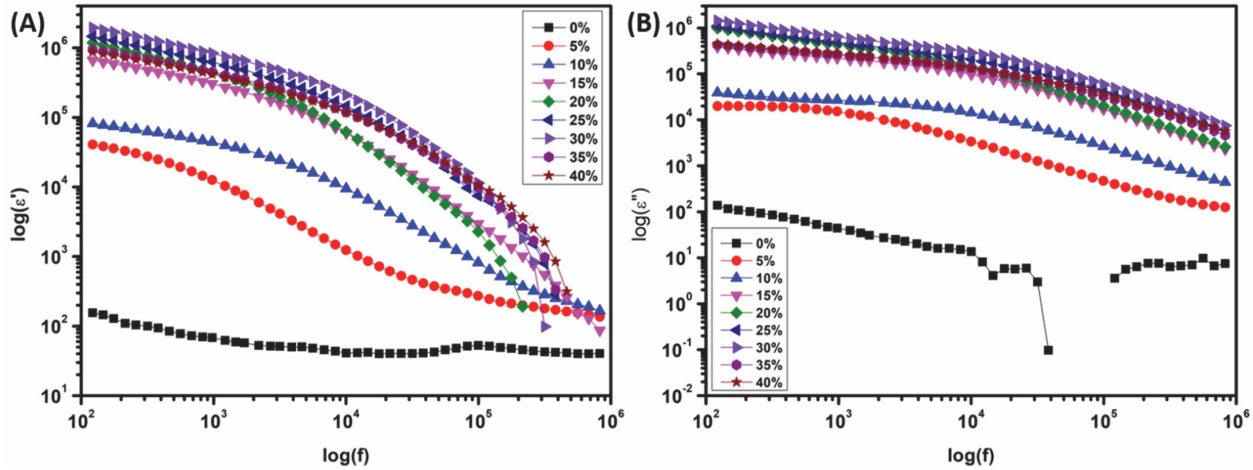

Figure 5. Variation of ε′ and ε″ with respect to frequency for all polymeric films.

Dielectric loss or loss tangent is also calculated for the polymeric films which is represented by tan(δ) and expressed mathematically as (Eq. 5)-

$$\tan(\delta) = \frac{\varepsilon''}{\varepsilon'} \quad (5)$$

Where, ε' and ε'' are real and imaginary part of dielectric constant. The variation of tan(δ) with frequency for all prepared polymeric films is shown in figure 6. The loss tangent (tan(δ)) decreases with the increasing frequency and attains a maximum point at certain frequency known as relaxation frequency, in the presence of restful dipoles and again starts to decrease. Relaxation time (τ), which is reciprocal of relaxation frequency is also calculated for the phytagel and phytagel/EMIm[TCM] polymer films, which are listed in table 2. The phytagel based films consisting of ionic liquid more than 10% are showing relaxation time in the order of 10^{-7} seconds which shows the fast ionic movement in them.

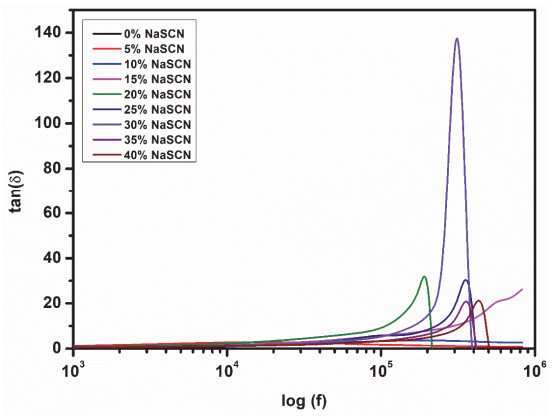

Figure 6. Variation of dielectric loss (tan δ) with respect to frequency for pure phytagel and EMIm[TCM] blended phytagel films.

Linear Sweep Voltammetry:

The Electrochemical stability window (ESW) i.e., working potential is a very important parameter of any electrolyte for their application practical devices. To determine the working potential window of the maximum conducting (phytagel/30wt%EMIm[TCM]) film the linear sweep voltammetry is performed shown in figure 7. The working potential range of the phytagel/30wt%EMIm[TCM] is comes out from -1.69 V to 1.42 V i.e., ~3.1 V which is sufficient for their application in electrochemical energy devices such as supercapacitors, batteries, dye sensitized solar cells etc.

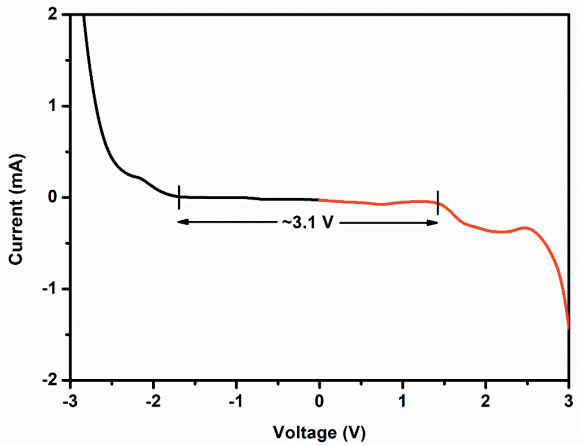

Figure 7. Linear Sweep Voltammetry (LSV) plot for phytagel/30wt% EMIm[TCM]

Ionic transport number:

Wagner polarization technique is used to calculate the total ionic transport number (t_{ion}) of the polymer electrolyte film phytagel/30wt%EMIm[TCM].

Cell SS|phytagel/30wt%EMIm[TCM]|SS is polarized with a 7.5V potential. The current vs time plot for phytagel/30wt%EMIm[TCM] is shown in figure 8. The total ionic transport number (t_{ion}) has been calculated using the following expression (Eq. 6)-

$$t_{ion} = 1 - \frac{i_e}{i_t} \quad (6)$$

Where, i_e is the remaining current and i_t is the total current. The value of t_{ion} is comes out to be ~0.99 which shows that the ionic charge carriers are predominant in the prepared polymer electrolyte film.

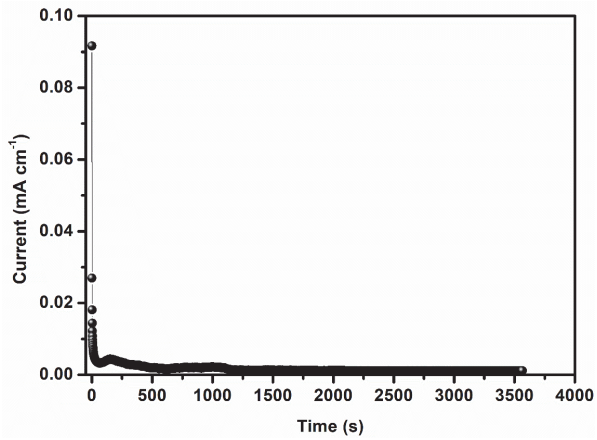

Figure 8. Potentiostatic current vs time plot (t_{ion}) for phytagel/30wt%EMIm[TCM].

Conclusion. Ionic liquid 1-ethyl-3-methylimidazolium tricyanomethanide and biopolymer phytagel blended polymer electrolyte films are successfully synthesized using solution cast technique. The XRD studies shows the semi-crystalline nature of host polymer phytagel and also evident the increase in amorphous phase with blending of ionic liquid. FTIR reveals the presence of different functional groups and interaction of ionic liquid ions with polymer chain of phytagel. The polymer electrolyte film with 30wt% of ionic liquid concentration shows the maximum conductivity value of 3.64×10^{-4}. The dielectric plots follow the same trend as ionic conductivity. The relaxation time calculated from dielectric loss tangent graph is comes out in the order of 10^{-7} for the films >10wt% of EMIm[TCM] indicating the fast ionic movement. The t_{ion} value of maximum conducting film, calculated using DC polarization is comes out to be ~0.99 which shows the predominant ionic charge carriers in prepared polymer electrolyte films. The maximum conducting film is electrochemical stable upto ~3.1V.

Conflict of Interest:

The authors declare no conflict of interest.

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IZVOD

STRUKTURALNA, ELEKTROHEMIJSKA I DIELEKTRIČNA ISTRAŽIVANJA FITAGELA I 1-ETIL-3-METILIMIDAZOLIJUMA BIO-POLIMER ELEKTROLITA NA BAZI TRICIJANOMETANIDA

Ovaj rad je fokusiran na sintezu i detaljno proučavanje biopolimernih fitagela i jonske tečnosti 1-etil-3-metilimidazolijum tricijanometanida (EMIm[TCM]) mešanih polimernih elektrolitnih filmova za energetske primene. Ovde se polimerni filmovi, na bazi biopolimera fitagela, sintetišu sa različitim koncentracijama jonske tečnosti (EMIm[TCM]) korišćenjem tehnike livenja rastvora. Sintetizovani filmovi su okarakterisani zbog svojih strukturnih, elektrohemijskih i dielektričnih svojstava korišćenjem različitih alata za karakterizaciju, kao što su XRD, FTIR, spektroskopija elektrohemijske impedanse, linearna voltametrija i Vagnorova tehnika polarizacije. Film sa 30vt% EMIm[TCM] pokazuje maksimalnu provodljivost od 3,64 × 10-4 S cm-1 i prozor elektrohemijske stabilnosti od 3,1 V. Dielektrična svojstva kao što su dielektrična konstanta (k), tangenta dielektričnog gubitka (tanδ), vreme relaksacije i učestalost su takođe proučavani za pripremljene čiste fitagel i fitagel/EMIm[TCM] polimerne filmove.

Ključne reči: biorazgradivi polimeri, fitagel, jonska tečnost, 1-etil-3-metilimidazolijum tricijanometanid, polimerni elektrolit

Naučni rad

Rad primljen: 30.03.2024.

Rad korigovan: 27.05.2024.

Rad prihvaćen: 5.06.2024.

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