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STUDY OF THE STRUCTURAL AND ELECTRICAL PROPERTIES OF THE PVA–NH_4SCN MEMBRANE FOR ITS APPLICATION IN ELECTRIC DOUBLE LAYER CAPACITORS

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The PVA–NH_4SCN polymer membranes were prepared by simple solution casting technique by passing ultrasound waves during the preparation. The polymer membranes were subjected to X-ray diffraction analysis and scanning electron microscopy. The X-ray diffraction pattern confirmed the incorporation of a salt into the polymer matrix. The scanning electron microscopy images showed the morphological changes of the polymer membrane. The polymer electrolyte (designated as UPVA20) incorporated with the 20 wt.% of the salt had the highest electrical conductivity in the order of 10^{-4} S cm^{-1}. It was concluded from the dielectric, tangent and modulus spectra that the UPVA20 membrane was good at its properties. Thus, electric double layer capacitor was constructed with UPVA20 membrane as the separator. The capacitance value of the electric double layer capacitor determined from cyclic voltammetry was found to be 1652 mF g^{-1}. The ultrasound assisted preparation of polymer membranes were good at performance when compared with the polymer membranes of ultrasound unassisted preparation. Among all the polymer electrolytes, UPVA20 polymer membrane had high conductivity, potential stability and capacitance.

Keywords: electric double layer capacitor, poly(vinyl alcohol), NH_4SCN, blend polymer electrolyte, ultrasound.

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Introduction

The water-soluble polymer membranes like poly(ethylene oxide) (PEO), poly(acrylamide) PAM, poly(vinyl alcohol) (PVA), poly(vinyl pyrrolidone) (PVP), poly (hydroxyethyl acrylate) PHEA etc. are widely used in pharmaceutical, cosmetic, biomedical and in personal care products. Among the above, PVA is a promising biomaterial used for tissue mimicking and vascular implanting [1]. The polymer PVA is also well explored for its proton conductivity and as a membrane for energy devices like proton battery, electric double layer capacitor (EDLC), fuel cells etc. Only scarce reports are seen on the proton conductivity of NH_4SCN added PVA system as mentioned below. Kulshrestha et al. [2] reported a conductivity of the order of 10^{-3} S cm^{-1} at room temperature for PVA (molecular weight (M.Wt) 125,000)/NH_4SCN prepared with triple distilled water as the solvent. Shukla et al. [3] reported a proton conductivity of 7.4×10^{-4} S cm^{-1} for $(PVA_{0.15}:NH_4SCN)_{0.12}$:PVAc system with PVA (M.Wt=78,000), using DMSO as the solvent. Agrawal et al. [4] reported a proton conductivity of 2.58×10^{-3} S cm^{-1} for PVA (average M.Wt=124,000–186,000)/NH_4SCN gel electrolyte system made with DMSO solvent. The above reports show that the PVA:NH_4SCN systems are promising and have to be further explored well for their properties.

The passing of ultrasound during the making of membranes influences the properties of the membranes and the electrochemical performances of the devices. Recently, our group reported on the blending effect of poly(acrylamide) (PAM) in the PVA:PAM:NH_4SCN system prepared by ultrasound

assisted solution casting method [5]. The present report is on the influence of ultrasound on the proton conductivity and dielectric nature of the different wt.% of ammonium thiocyanate (NH_4SCN) added PVA (M.Wt 140,000) electrolyte, with distilled water as the solvent. The amount of salt content and the presence of ultrasound greatly influenced the electrical properties. The effect of passing ultrasound on the characteristics of the solid polymer membrane is investigated. Among the different energy devices as mentioned above, the EDLC is a clean and rechargeable energy storage system with a wide range of applications. The EDLCs with polymer electrolytes have the great advantage of device flexibility [6]. Hence, in the present work, the best membrane based on electrical property was subjected to EDLC studies.

Experimental

Materials

The poly(vinyl alcohol), $[-CH_2CHOH-]_n$, HIMEDIA (M.Wt: 140,000), was used as the host polymer and the ammonium thiocyanate (NH_4SCN) HIMEDIA of high purity (99%) was used as the salt. The double distilled water was used as the solvent. The activated carbon used was purchased from Merck of high quality. The carbon cloth used was purchased from the Sainergy Fuel Cell India Pvt. Ltd.

Preparation

The polymer membranes were prepared by incorporating 15, 20 and 25 wt.% of NH_4SCN with PVA. The membranes were prepared with and without ultrasound by the simple solution casting method, which is mentioned by W and U series, respectively. The 100 wt.% of PVA and 15 wt.% of NH_4SCN dissolved in 50 ml of distilled water were exposed to ultrasound for 20 min and left for slow stirring for 24 hrs, then it was casted on the Petri plate which was maintained at 40^{0}C for 48 hrs. It was peeled off when the casted solutions were completely dried and the sample code was given as UPVA15. The same method of preparation was repeated for the solutions and it was not exposed to ultrasound, then the sample code was given as WPVA15. Similarly, the other films with 20 wt.% and 25 wt.% of salt were prepared and the sample codes are given as UPVA20, UPVA25 (with ultrasound) & WPVA20, WPVA25 (without ultrasound), respectively.

Characterization techniques

The polymer membranes were exposed to X-ray diffraction studies (XRD), using X-ray diffractometer (Shimadzu, Japan). The Fourier transform infrared spectra (FTIR) were recorded in the wave number range of 400–4000 cm^{-1} using Spectrum Two Model IR spectrometer at a resolution of 4 cm^{-1}. The morphology of the films was taken by Scanning Electron Microscopy (SEM) (JEOL, JSM6390, Japan). The ultrasonic processor (UP400S, Germany) was used for membrane preparation. AC impedance spectroscopy studies were made in the frequency range of 1 Hz to 1 MHz at an amplitude voltage of 5 mV. Linear sweep voltammetry (LSV) was done for the films in the voltage interval of 0–5 V. The above electrical studies were made using CH electrochemical workstation CHI6008E.

Fabrication of EDLC

The EDLC was constructed by alignment of carbon electrode//polymer membrane//carbon electrode as the device design.

The procedure of electrode making was as follows. The slurry was prepared with 75% of activated carbon, 15% of carbon black, 10% of PVDF (binder) in ethanol (solvent). The slurry was coated on the carbon cloth and evaporated for solvent at 80^{0}C for 12 hrs. The EDLC was fabricated by making a sandwich of above electrodes with the polymer electrolytes.

Results and discussion

X-ray diffraction analysis of polymer membranes

The XRD patterns of the PVA–NH_4SCN polymer membranes are shown in Fig. 1. The peak around 2θ~22^0 corresponding to (110) plane of the polymer PVA confirms the semi-crystalline nature of the membranes. Similar peak was observed by Patel et al. [7] in the PVA:NH_4SCN system. The intensity of the above peak was disturbed both by the preparation route and by the amount of salt content. The absence of peaks corresponding to the salt indicates good complex formation between the salt and the polymer matrix.

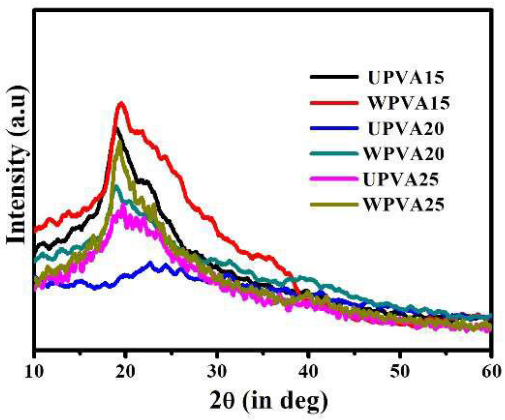


Fig. 1. XRD patterns of the PVA–NH_4SCN polymer membranes

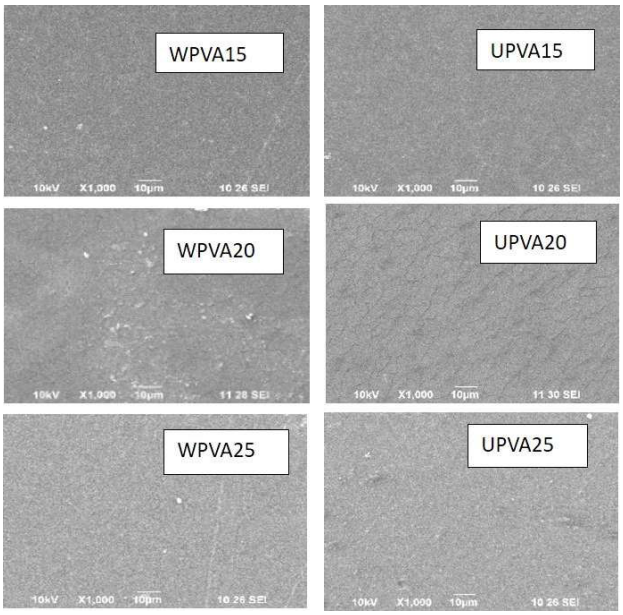


Fig. 2. SEM images of the PVA–NH_4SCN polymer membranes

SEM analysis of polymer membranes

The SEM images for the polymer electrolytes are given in Fig. 2. The SEM images clearly show that the all the ultrasound assisted polymer membranes have smooth surface while the membranes made without ultrasound were found to have roughness in their surface.

The smoothness of the surface in the polymer membranes helps in the formation of better interface with the electrodes in EDLC.

XRD and SEM analysis of electrodes

The texture of the carbon cloth current collector and the smooth coating of the active slurry over the carbon cloth play a major role in good interface between the electrode and the polymer electrolyte. Smoother the slurry is coated over the current collector, a good interface is expected between the electrode and the polymer electrolyte.

Figure 3,a shows the XRD pattern of the bare uncoated carbon cloth and the slurry coated carbon cloth. As can be observed from Fig. 3,a (i), the peaks at $2\theta=26^0$ and 43^0 represent the (002) and (101) hkl planes of the carbon cloth current collector [8]. After coating the slurry (Fig. 3,a (ii)), the intensity of the peaks corresponding to the carbon cloth was found to decrease in the intensity, which is by the coating effect of the activated carbon/carbon black mixture. There was no shift in the peaks and no new peaks were observed. The decrease in intensity was due to the fact that the activated carbon, which forms the major part of the slurry, is commonly known to be more amorphous or less crystalline in its nature than the carbon black, and it is also observed in XRD pattern as discussed below.

The XRD pattern of the activated carbon, carbon black and the mixture of activated carbon, carbon black (15% of carbon black in activated carbon) are shown in Fig. 3,b (i–iii). To obtain XRD patterns, the 0.2 g activated carbon and 0.03 g of carbon black were mixed together and ground well as done in the slurry making process. The peaks at $2\theta=26^0$ and 43^0 represent the (002) and (101) hkl planes of the activated carbon/carbon black. The peaks at $2\theta\sim39^0$ and 65^0 also belong to the carbon phases, which were not reflected in slurry coated carbon cloth. This may be due to the changes happened during the slurry making process, where the materials were mixed with binder and solvent and ground well before coating on the carbon cloth current collector. As seen in the XRD patterns, the carbon black is more crystalline than the activated carbon. The XRD pattern of the mixture of activated carbon and carbon black is also less crystalline than

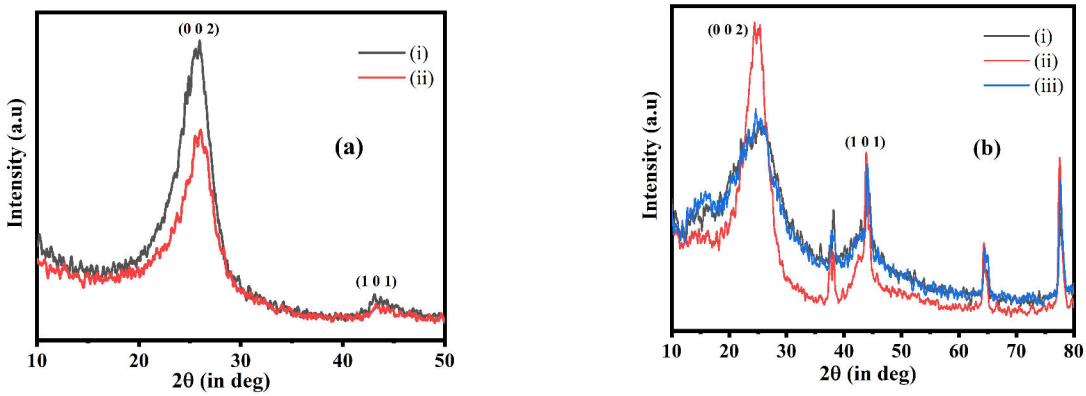


Fig. 3. (a) – XRD patterns of (i) carbon cloth and (ii) slurry coated carbon cloth; and (b) – XRD patterns of (i) activated carbon, (ii) carbon black and (iii) mixture of activated carbon and carbon black

carbon black due to the addition of carbon black only at less percentage.

Figure 4 shows the SEM images at different scales and magnification of the bare carbon cloth and the slurry coated carbon cloth. The bare carbon cloth shows the threads of nanofibers (Fig. 4, a and c). The coated carbon cloth (Fig. 4, b and d) shows the presence of slurry made with mixed activated carbon and the carbon black made as discussed under experimental section. The presence of the particles is seen well in the inset of Fig. 4, b. The carbon cloth surface after coating was smooth, as the physical mixture of activated carbon and the carbon black were made as smooth slurry using PVdF binder in presence of NMP. The SEM images of activated carbon, carbon black and activated carbon with carbon black as a physical mixture are shown in

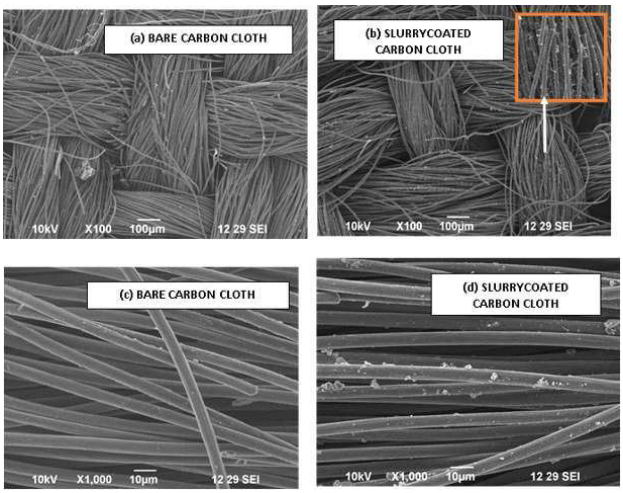


Fig. 4. SEM images of (a–c) bare carbon cloth and (b–d) slurry coated carbon cloth at two different scale and magnification

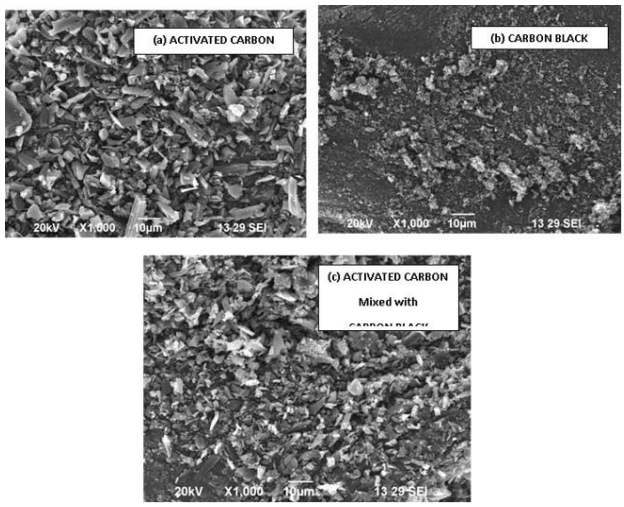


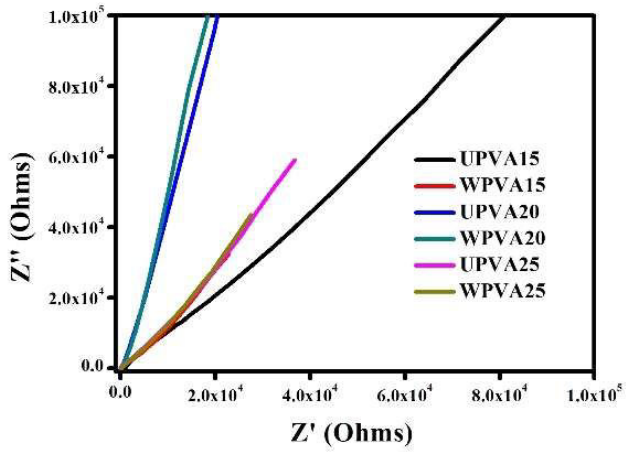
Fig. 5. SEM images of (a) activated carbon, (b) carbon black and (c) mixture of activated carbon and carbon black

Fig. 5, which exhibits the presence of particles with irregular morphology. The activated carbon used has a surface area of 2475 m^2 g^{-1}, as reported in our previous study [5].

Electrical properties of the polymer membranes

Conductivity analysis

The Nyquist plot and AC conductivity of the polymer membranes are shown in Fig. 6, a and b, respectively. From the Nyquist plot, the ionic conductivity of the polymer membrane was calculated using the relation $\sigma=(1/R)\times(L/A)$, where «R» is the resistance, «L» is the thickness and «A» is the area of the polymer membrane [9]. The ultrasound assisted polymer membranes have high conductivity of the order of 10^{-4} S cm^{-1} in which UPVA20 was found to have a conductivity of 1.4×10^{-4} S cm^{-1}. Comparatively, the polymer membranes without ultrasound have low conductivity in the order of



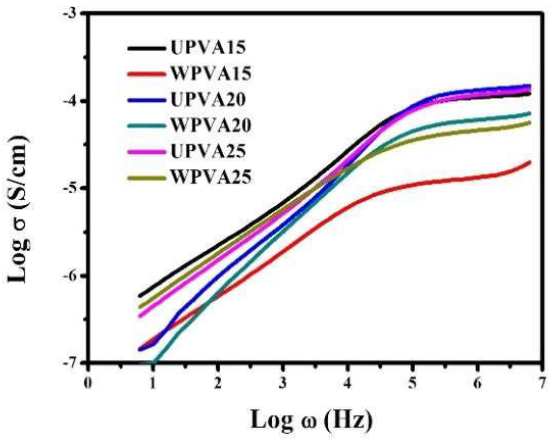


Fig. 6. (a) – Nyquist plot and (b) – conductivity plot of the PVA-NH_4SCN polymer membranes

Electrical and electrochemical data

Membrane	Conductivity, S cm^{-1}		LSV, V	Capacitance CV, mF g^{-1}
	from conductivity plot	from Nyquist plot		
UPVA20	1.4×10^{-4}	2.9×10^{-4}	1.8	1652
UPVA25	1.2×10^{-4}	3.4×10^{-4}	1.7	890
UPVA15	1.1×10^{-4}	1.2×10^{-4}	1.5	409
WPVA20	3.6×10^{-5}	3.3×10^{-5}	2.0	282
WPVA25	2.9×10^{-5}	1.2×10^{-5}	2.1	192
WPVA15	1.4×10^{-5}	1.1×10^{-5}	1.9	99

$\times10^{-5}$ S cm^{-1}. The conductivity plot shows the polarization at low frequency and the DC conductivity plateau at the mid frequency region. The conductivity values from Nyquist and AC conductivity plot are presented in Table. The difference in the electrical properties of the membranes under study is due to the enhanced dissociation of ammonium thiocyanate with PVA in presence of ultrasound. More dissociation tends to increase the mobility of the charge carriers and hence the ionic conductivity also increases [10].

Dielectric loss

The dielectric permittivity of the membranes is shown in Fig. 7. The dielectric loss of the membranes were determined by real part (ε') and imaginary part (ε'') of the complexes with the relation $\varepsilon^*(\omega)=\varepsilon'(\omega)-j\varepsilon''(w)$. The dielectric permittivity decreases with the increase in frequency because of the long motion of the ions at lower frequencies [11]. Here, the polymer membrane UPVA20 has high dielectric permittivity at all the frequencies. It was due to the good number of dissociated ions in the membrane.

The decrease in ε' and ε'' at lower values of frequency is due to the accumulation of the charges. In vice versa, the loss tangent spectra increases with the increase in frequency [12] and the tangent spectra depend on the dielectric spectra with the relation $\tan\delta=(\varepsilon'/\varepsilon'')$. The relation between the modulus spectra and dielectric spectra is given with the equation $M'=\varepsilon'/(\varepsilon'^2+\varepsilon''^2)$ and $M''=\varepsilon''/(\varepsilon'^2+\varepsilon''^2)$ and the complex form of the modulus is $M^*=M'+j\cdot1/M''$ [13]. The modulus spectra help to determine the electrode polarization effect on polymer membrane. While changing the wt.% of the salt, the intensity of peaks in the modulus spectra varies. Among those,

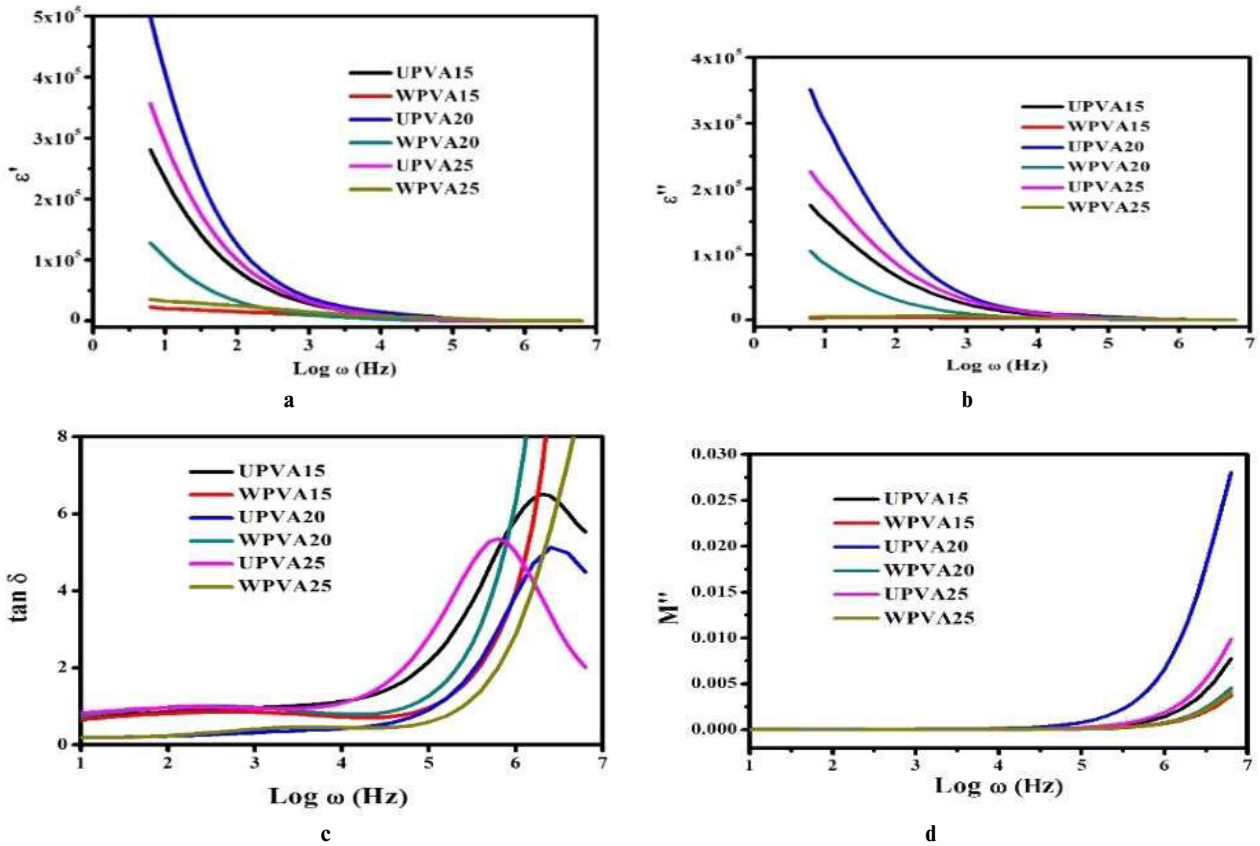


Fig. 7. (a) – dielectric permittivity, (b) – dielectric loss, (c) – loss tangent and (d) – modulus spectra of the PVA–NH_4SCN polymer membranes

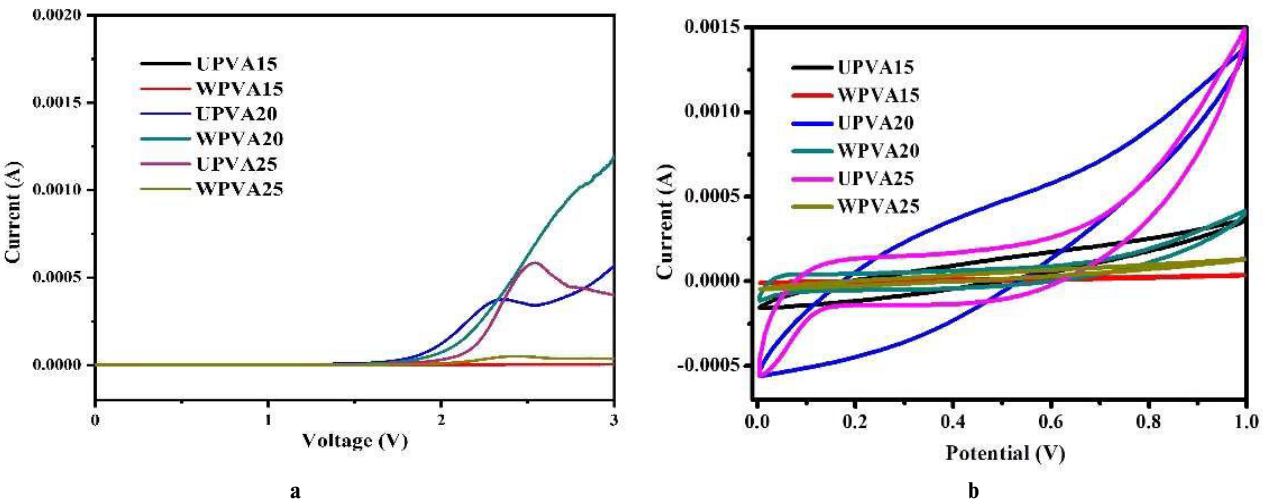


Fig. 8. (a) – Linear sweep voltammetry and (b) – cyclic voltammetry of EDLC at the scan rate of 100 mV s^{-1} of the PVA–NH_4SCN polymer membranes

UPVA20 has higher peak intensity tyan other membranes, indicating lower relaxation time for the ion movement. Lower the relaxation time for charge carriers, higher will be the conductivity, as indicated in conductivity analysis.

Electric double layer capacitor

The electrochemical studies such as LSV and CV studies were done as shown in Fig. 8. The potential stability for the polymer membranes between 1–2 V shows the membrane UPVA20 to have good voltage stability as in Table. The slight decreases in voltage stability of membranes made with ultrasound are due to the increase in their flexibility. After checking out the potential stability, the device was fabricated with the polymer electrolytes as mentioned in experimental part. The CV's run at the scan rate of 100 mV were compared for the EDLC's made with different membranes under study and the capacitance value were calculated. The electrochemical data from the CV and LSV plots are presented in Table. The EDLC made with UPVA20 has a high capacitance value of 1652 mF g^{-1}. This was comparable with the literature reports, where Aziz et al. [14] reported on the PVDX 20 PVA:Dextran:NH_4I (20 wt.%) blend membrane with an electrical conductivity of 2.08×10^{-5} S cm^{-1}, and the EDLC made with activated carbon delivered a capacitance of 4.2 F g^{-1} in the 0–1 V range. Shuhaimi et al. [15] reported on the carboxymethyl cellulose (CMC)–NH_4NO_3 (25 wt.%)-based solid polymer electrolyte with an electrical conductivity of 2.10×10^{-6} S cm^{-1}, and the EDLC made with activated carbon delivered a capacitance of 1.67 F g^{-1} in the 0–1 V range. It was observed that EDLC constructed with the ultrasound assisted polymer membranes has good capacitance when compared with EDLC made with the ultrasound unassisted polymer membranes.

Conclusions

The PVA–NH_4SCN polymer membranes with different salt concentration were prepared by simple solution casting technique under the influence of ultrasound. The polymer membranes were subjected to XRD and SEM analysis. The XRD pattern confirms the PVA–NH_4SCN complex formation. The SEM images show the morphological changes of the polymer membrane in the presence of ultrasound. The conductivity of the polymer membranes were compared, from which UPVA20 membrane was found to have the highest conductivity in the order of 10^{-4} S cm^{-1}. From the dielectric spectra, tangent and modulus spectra, it was concluded the UPVA20 film was good at its electrical properties. The potential stability was observed for the polymer membranes to be good. From CV, it was concluded that EDLC made using membrane UPVA20 has 1652 mF g^{-1} as the highest specific capacitance. The ultrasound assisted polymer membranes are good at properties when compared with the ultrasound unassisted polymer membranes. Hence, UPVA20 polymer membrane has high conductivity, potential stability and has the suitability for EDLC applications.

Acknowledgements

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ВИВЧЕННЯ СТРУКТУРНИХ ТА ЕЛЕКТРИЧНИХ ВЛАСТИВОСТЕЙ МЕМБРАНИ ПВС–NH_4SCN ДЛЯ ЇЇ ЗАСТОСУВАННЯ В СУПЕРКОНДЕНСАТОРАХ

А. Свамінатхан, Р. Раві, С. Айясамі, В. Бходжан, Р. Свамінатхан, Р. Нандхакумар

Полімерні мембрани полі(віниловий спирт)–NH_4SCN були виготовлені простим методом лиття розчину шляхом проходження ультразвукових хвиль під час приготування. Полімерні мембрани піддавали рентгеноструктурному аналізу та скануючій електронній мікроскопії. Рентгенівська дифрактограма підтвердила включення солі в полімерну матрицю. Зображення скануючої електронної мікроскопії показали морфологічні зміни полімерної мембрани. Полімерний електроліт (позначений як UPVA20), включений з 20 мас.% солі, мав найвищу електропровідність порядку 10^{-4} См/см. Зі спектральних даних було зроблено висновок, що мембрана UPVA20 має хороші властивості. Таким чином, був сконструйований суперконденсатор з мембраною UPVA20 в якості сепаратора. Значення ємності суперконденсатора, визначене за допомогою циклічної вольтамперометрії, було рівним 1652 мФ/г. Підготовка полімерних мембран за допомогою ультразвуку показала задовільні показники в порівнянні з полімерними мембранами без використання ультразвуку. Серед усіх полімерних електролітів полімерна мембрана UPVA20 мала високу провідність, стабільність потенціалу та ємність.

Ключові слова: суперконденсатор; полі(віниловий спирт); NH_4SCN; змішаний полімерний електроліт; ультразвук.

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