

Deep Eutectic Solvent-Based Polymer Electrolyte Films for an Electrochemical Double Layer Cell

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Abstract

Polymer electrolyte films based on DES were prepared with the solution cast technique. DES(ChCl-glycerol) was prepared with two different glycerols: one purchased from the industry and the other fabricated in the lab from waste vegetable oil. Two polymer electrolyte films were prepared with two different DES (industrial and from oil-based glycerol) were named D1 and D2, respectively. Fabricated polymer film (D1) with DES (prepared with industrial glycerol) shows excellent physical and thermochemical properties. The molecular and structural characteristics of the film were studied via FTIR and XRD. The surface morphology, thermal and electric properties of film were studied with SEM, TGA, and impedance spectroscopy techniques, respectively. The obtained films were flexible, strong, and transparent. Its properties are ascribed to a strong coherent hydrogen bond network with the matrix of the polymer. Polymer electrolyte film D1 exhibited good ionic conductivity of $1.25 \times 10^{-3} \text{ Scm}^{-1}$. The prepared polymer film was applied as solid electrolytes in symmetric solid-state supercapacitors. The fabricated EDLC cell exhibits specific capacitance of at voltage with scan rate. Moreover, the prepared electrolyte is biocompatible, environment safe and cost-effective that offers a new path for adaptable electronic devices.

Keywords: Green electrolytes, Supercapacitor, Deep eutectic solvent, Hydrogen bond network, Choline chloride-glycerol.

1. Introduction

Supercapacitors have drawn a lot of interest as effective and widely available secondary energy storage solutions in the development of electronic devices. However, using flammable liquid electrolytes in supercapacitors is a significant challenge as there are conflicting needs for safety and energy density in real-world applications [1]. So, solid polymer electrolytes gained attraction and are considered potential candidates because they have good characteristics like high ionic conductivity, low-cost, high thermal stability, and ease of processability for wide application in energy storage devices [2]. The most recent technique for enhancing the matrix polymer and with broad characteristics is the blending of polymer components. Blending plays a important role in expanding the field of polymer studies with an easy preparation process. [3]. Combining two or more polymers improves the qualities of the polymer that may be employed in a certain application. The proper methods for developing blends are solvent casting technique and melt blending [4]. This is one of the important techniques to increase conductivity. The characteristics of polymer blends are frequently better than those of the individual polymer components [5].

The internal structure of polyvinyl alcohol (PVA) allows for its classification as either semicrystalline or amorphous. Amorphous and crystalline regions can be created in order to synthesize semicrystalline PVA [6]. Moreover, PVA has hydroxyl groups arranged along the carbon chain backbone. The O-H groups serve as a source of hydrogen bond, facilitating in the creation of blending of polymer [7].

At ambient temperature, polyethylene oxide (PEO) is considered as semi-crystalline polymer that exhibits both phases: amorphous and crystalline. PEO has various properties such as strong viscosity, excellent water solubility, and good thermal stability [8]. PEO polymer have etheric links, whereas polymer PVA contains OH groups that combines with other polymer PEO to form the PVA+PEO blend by acting as a source of hydrogen bonds [9].

The current work focuses on solid polymer electrolytes doped deep eutectic solvent (DES). DES are a large family of ionic liquids (ILs) and are considered as a subclass of ILs [10]. The resultant DES has a lower melting point than the melting points of each of its components. DESs often exhibit a significant depression in freezing-point and have liquid state at temperatures below 150 °C [11]. It is noted that the majority of them are in liquids phase between 27 to 70°C.

A DES is typically created by reacting a quaternary ammonium salt that may combine with the quaternary ammonium salt's halide anion to form a complex [11]. DESs have similar properties like ionic liquids such as it exhibits strong ionic conductivity, a broad electrochemical operating window, thermal and chemical stability, and minimal vapor pressure. DESs are also more advantageous than ILs in several ways, including low cost, simple synthesis, environment-safe and biodegradable [12]. DESs based on choline chloride with urea, glycerol and ethylene glycol (EG), etc. have been frequently used as electrolytes or dopants in polymer matrices to produce polymer electrolytes [13]. It has been found that a DES made of glycerol and choline chloride has a great deal of promise for application as a liquid electrolyte. Thus, in order to create a solid-state electrolyte, DESs considered as a conductive medium in a matrix of polymer without losing its conductivity [14].

In this paper, DES has been prepared with choline chloride and glycerol is used as dopants in polymer blends. PVA/PEO-based polymer electrolyte is used to produce solid polymer electrolyte films. The prepared films has been characterized through SEM, XRD, TGA, FTIR and impedance spectroscopy technique. With the optimized film, the EDLC has been fabricated and its electrochemical properties are studied with cyclic voltammetry technique via an electrochemical workstation.

2. Materials and Method

Polymer polyvinylchloride (PVC) with molecular weight~polyethylene alcohol (PEO) with molecular weight~, Distilled water used as a solvent, Deep eutectic solvent produced from choline chloride and Glycerol. All the chemicals were purchased from company Sigma-Aldrich were utilized prior to purification.

2.1 Preparation of polymer blend

A blend of polymers was prepared with two different polymers: PVA and PEO. First, PVA (0.5g) was taken in a capillary tube with distilled water solvent and kept on a magnetic stirrer for 4 hours at 60°C to dissolve it completely. On the other side, PEO (0.5g) was taken and also kept on magnetic stirrer to get homogenous solution. Then, both solutions were mixed together and kept on a stirrer for 24 hours until a clear, transparent, and homogenous solution was obtained. Two samples of polymer blend were prepared to fabricate polymer films.

2.2 Preparation of DES with two different glycerol

The deep eutectic solvent was prepared with components of choline chloride and glycerol. DES was prepared with two different glycerols. Glycerol was obtained directly from industry and named as G1 and other glycerol (G2) was obtained from used vegetable oil. To prepare G2, used vegetable oil was taken from the home. In a clean dry beaker, 100cm³ of used/waste oil (filtered from a fine sieve), 20cm³ methanol, and 4cm³ of 10molar KOH were added to it.

The beaker was placed on a magnetic stirrer for 30 minutes to dissolve all the components. Then mixture of solution was transferred to the centrifuge tube. Centrifugation of the mixture was carried out for 5 minutes. After centrifugation, we see the two separate layers. The above layer was of biodiesel which was carefully extracted with the dropper and the lower layer was of glycerol. This Glycerol was named G2 and used to prepare DES further.

To prepare DES, choline chloride was kept in oven to evaporate moisture from it. After that, ChCl-glycerol was taken in a capillary tube in a molar ratio of 1:2 and placed on the stirrer at 60°C for 120 minutes until a transparent solution was prepared. Two types of DES were prepared from two different types of glycerol (G1 and G2) as discussed above and termed as DES_{1 industrial glycerol} and DES_{2 oil based glycerol}.

2.3 Preparation of solid polymer electrolyte films

With DES_{1 industrial glycerol} and DES_{2 oil-based glycerol} two polymer electrolyte films were prepared. DES₁ and DES₂ were added in already prepared polymer blend solutions as discussed above. Blend composition was taken 1g and DES composition was 0.5g for both films. To obtain homogenous solution, both solutions were kept on magnetic stirrer for 24 hours. After that, homogenous solutions were cast in a petri dish and placed in a vacuum desiccator for one week to obtain free-standing polymer films. Thus, polymer films were obtained successfully and named D1 (prepared with DES_{1 industrial glycerol}) and D2 (prepared with DES_{2 oil-based glycerol}).

2.4 Preparation of electrodes

To prepare the electrode material, 10% conductive Carbon Black, 10% PVDF-HFP, and 80% activated carbon (AC) based on Datura metal peels were combined to produce a homogenous slurry [15]. The final slurry was applied on graphite sheets of 1 cm² in size. The sheets had a thickness of 0.1 mm. These graphite sheets were dried out for 24 hours at 80°C in a vacuum oven to produce AC electrodes. A mass of 1 mg per cm² is loaded on every electrode. DES-based polymer electrolyte was used to create the EDLCs.

3. Experimental Results

3.1 SEM

Morphological studies of polymer electrolyte films were analyzed with a scanning electron microscope. **Figures 1(a) and (b)** show the SEM micrograph of prepared polymeric films D1 and D2, respectively. SEM micrograph of D1 shows better fine and small pores. It can be observed from **Figure 1(a)** that the surface becomes compact and has a dark portion with small grains which shows the amorphous nature of the polymeric film. On the other side, the SEM micrograph of D2 shows the smooth flat, and large grain structure at random positions. It can be observed that the dark portion confirms the amorphous nature and the white portion in **Figure 1(b)** confirms the crystalline nature of the polymer film. Hence, in comparison to both films (D1 and D2), D1 polymer electrolyte film has higher amorphous nature and hence leads to higher value of conductivity.

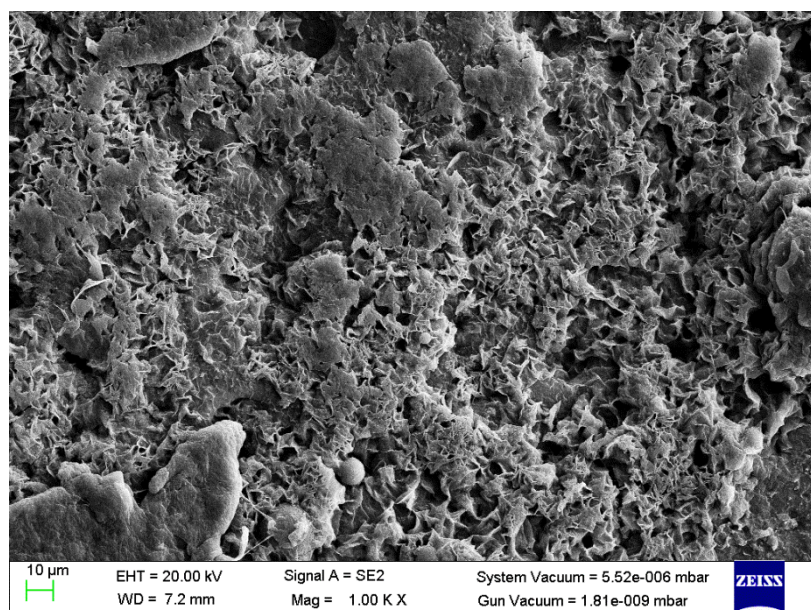


Figure1(a) SEM micrograph of the prepared polymer film (D1).

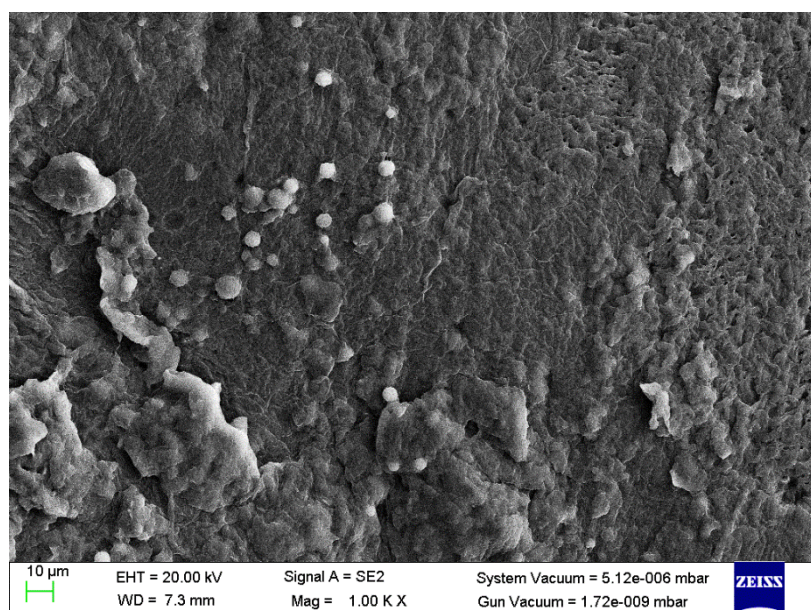


Figure 1(b) SEM micrograph of the prepared polymer film (D2).

3.2 XRD

The amorphous/crystalline nature of prepared polymeric films was confirmed with X-ray diffraction analysis technique. **Figure 2** depicts the XRD pattern of prepared polymeric films D1 and D2. The combination of polymers PVA and PEO shows the semi crystalline nature.

XRD of polymer PVA shows the prominent peaks at $2\theta = 19.5^\circ$, 22.3° , and 40.8° [16,17], corresponding to (101), (200), and (102) planes, respectively. XRD pattern of PEO shows prominent peaks at positions 19.18° and 23.58° corresponding to planes (120) and (112), respectively [18,19]. With addition of DES in the polymeric blend leads to

enhancing the amorphousness of polymer films. In the D1 composition, the peak is observed at 19.5° and 23° which is due to the polymer blend. The peak has a large intensity and confirms the amorphous nature. In D2 composition, the peak is observed at positions 20° and 23° and the intensity and broadness of the film confirms the decrease in crystalline nature of polymeric electrolyte films. In the polymer matrix, this amorphous region also facilitates the random path of ions [20]. Hence, the polymer chain experiences local segmental motion of ions which may increase the conductivity of polymeric films.

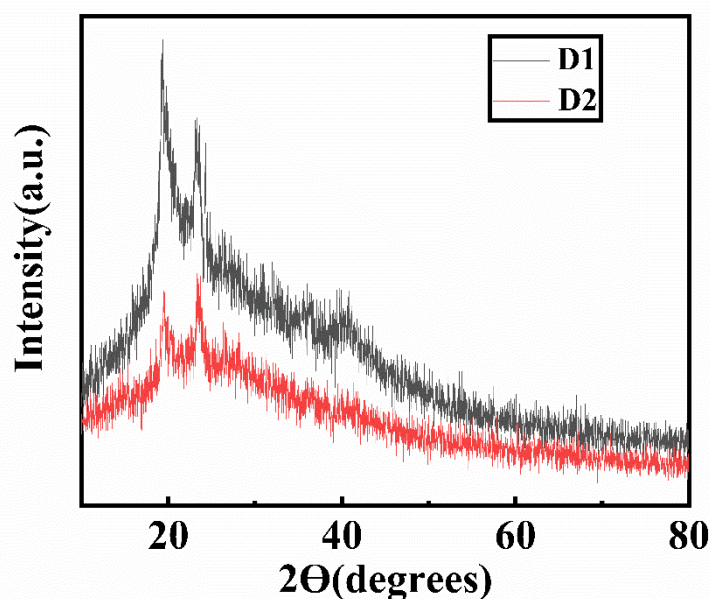


Figure 2. XRD pattern of prepared polymer electrolyte films D1 and D2.

3.3 TGA

The thermo-gravimetric analysis technique was used to calculate the mass loss and thermal stability of prepared polymeric films. **Figure 3** represents the TGA curve of the prepared polymer film D1 and D2. All samples were placed in an oven and heated at $27\text{--}800^\circ\text{C}$ with a heating system rate of 10°Cmin^{-1} . In D1 composition, there are three stages for mass loss of polymer electrolyte film. In the first region, the initial loss for D1 film arises at $27\text{--}86.45^\circ\text{C}$, 9% of mass loss occurs due to evaporation of the solvent and in the second stage, mass loss occurs at 264.28 to 502.5°C due to the degradation of the film. After that, the D1 polymer film was stable up to 502°C , with 8% residue left. In D2 composition, three stages of mass loss appear. The first weight loss of 8% occurs up to 106.60°C due to evaporation of the solvent and the second stage occurs at 106.60 to 321.03°C due to the decomposition of film and the final weight loss occurs at 474.8°C with 7% residue. From the figure, it is observed that the final temperature of D1 is higher than the D2 composition which confirms increase in the thermal stability of polymeric film.

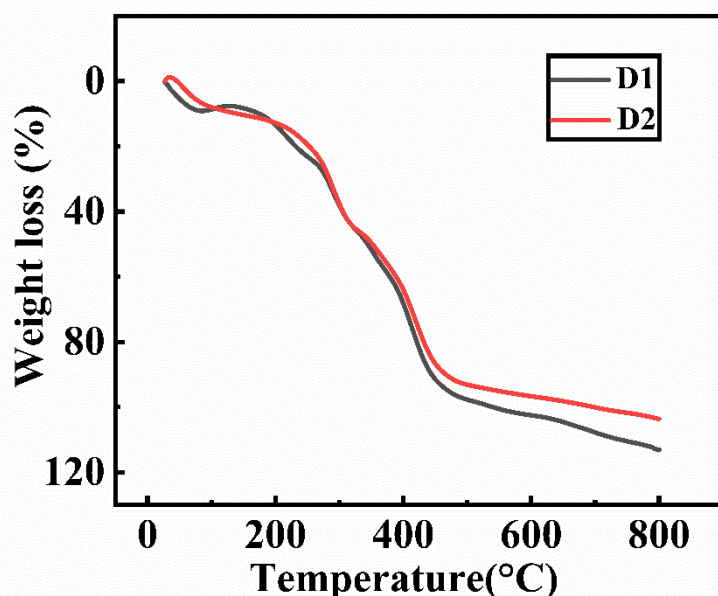


Figure 3. TGA curve of prepared polymer electrolyte films (D1) and (D2).

3.4 FTIR

FTIR spectroscopy was utilized to analyze the vibrational modes and complexation of DES1 and DES2 with a prepared polymeric blend. **Figure 4** depicts the spectra of prepared polymer films D1 and D2 in range of 600-4000 cm^{-1} . Vibrational peaks of DES (ChCl-Glycerol) are observed at 956, 1057, 1218, 1355, 1455, 1750, 2080, 2965 and 3339 cm^{-1} are associated with N-C-C bonding, C-C stretching, CH_2 deformation, NH bending, H-bending, NH bending, CO stretching, CH stretching and NH_2 asymmetrical stretching [21], respectively. The vibrational modes of the PVA+PEO polymer blend are located at 951 cm^{-1} and associated with C=H out-of-plane stretching [22]. A sharp peak was observed at 1096 and associated with C-O-C stretching and observed due to PVA. Peak located at 1279, 1345 and 1463 cm^{-1} is associated with CH_2 symmetrical twisting, C-H deformation and CH_2 scissoring bonding, respectively [22, 23]. **Table 1** shows the vibrational modes of, DES, polymer blend and prepared films D1 and D2. Vibration peaks of blend observed at 1739 and 2878 cm^{-1} are observed because of PEO associated with CO stretching and asymmetric C=H stretching. In composition D1, the vibrational bonds are located at 1096, 1566, 1738, and 2894 cm^{-1} which are correspond to COC stretching, CH_2 scissoring, C=O and asymmetric C=H stretching and are observed due to blending. There is a shift in peaks from 1463, 1739 and 2878 to 1566, 1738 and 2894 cm^{-1} , respectively. The vibrational modes located at positions 1045, 1228, 1363, 1425 and 3289 cm^{-1} are observed due to DES and are associated with C-C stretching, CH_2 deformation, NH bending +CN bending, H- bending and NH_2 asymmetrical bonds, respectively. In composition N2, the vibrational modes located at 953, 1096, 1231, 1346, 1738 and 2880 cm^{-1} corresponding with N-C-C bonding+ CH Bending, COC stretching, CH_2 deformation, CH deformation, CH_2 scissoring, C=H asymmetric stretching and C=O stretching, respectively. The vibrational modes 953 and 1231 and 1462 cm^{-1} are observed due to the complexation of DES and Blend. This suggests that the DES has better complexation with polymer blend.

Table1. Vibrational modes of prepared polymer electrolyte films.

DES		PVA+PEO(1g)		D1		D2	
Wave number	Vibrational bands	Wave number	Vibrational bands	Wave number	Vibrational bands	Wave number	Vibrational bands
956	N-C-C bonding	951	Out of plane CH bending	956	N-C-C bonding	953	N-C-C bonding+ CH Bending
1057	C-C stretching	1096	COC stretching	1045	C-C stretching	1096	COC stretching
1218	CH ₂ deformation	1279	CH ₂ symmetric twisting	1096	C-O-C stretching	1231	CH ₂ deformation
1355	NH bending +CN bending	1345	C-H deformation	1228	CH ₂ deformation	1346	C-H deformation
1455	H-bending	1463	CH ₂ scissoring	1363	NH bending +CN bending	1462	CH ₂ scissoring
1750	NH bending	1739	C=O stretching	1425	H-bending	1738	CO stretching
2080	C-O stretching	2878	CH asymmetric stretching	1566	CH ₂ scissoring	2880	C=H asymmetric stretching
2965	CH stretching			1738	C=O stretching		
3339	NH ₂ asymmetrical			2894	C=H asymmetric stretching		
				3289	NH ₂ asymmetrical		

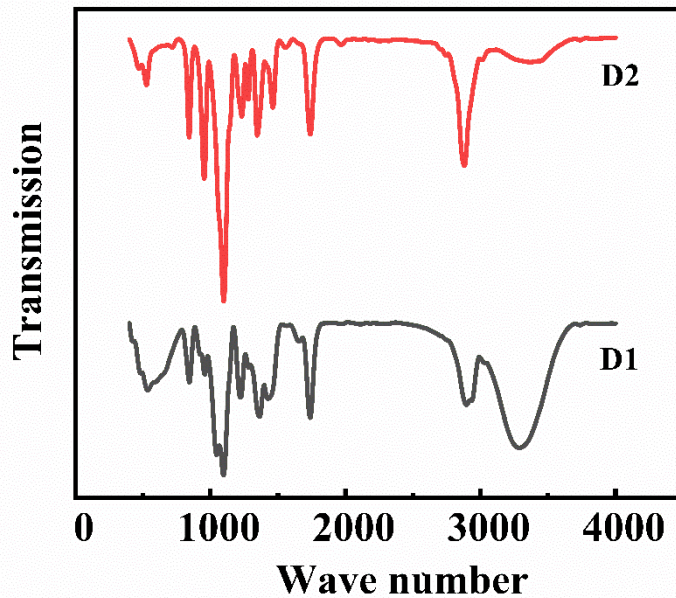


Figure 4. Fourier Transform Infrared Spectra of prepared polymer electrolyte films D1 and D2

3.5 Conductivity

Impedance spectroscopy technique was utilized to calculate the conductivity of produced polymeric films. Two cells were prepared with prepared polymer electrolyte films and with stainless steel electrodes. **Figure 5** shows the Nyquist plot of prepared cells with D1 and D2 composition. The conductivity of DES1 (from industrial glycerol) was measured in the range 10^{-4} Scm^{-1} and conductivity of DES2 (from oil-based glycerol) was measured in the range 10^{-5} Scm^{-1} . Conductivity was calculated by equation 1 as given below.

$$\sigma = \frac{t}{R_b * A} \quad \dots\dots (1)$$

In this equation, t denotes the thickness of polymeric film, R_b denotes resistance (bulk) and A denotes the area of electrode material. The conductivity of prepared film D1 was found to be in the range of $1.25 \times 10^{-3} \text{ Scm}^{-1}$ and the conductivity of D2 composition was in the range of 10^{-4} Scm^{-1} . This represents that D1 is an optimized electrolyte to prepare an Electrochemical double layer cell.

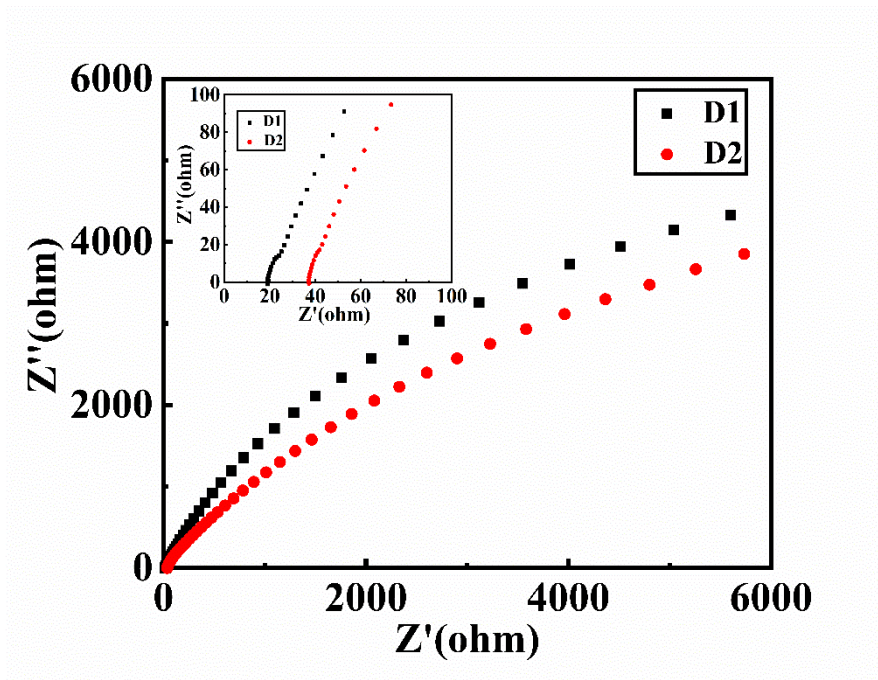


Figure 5. Nyquist plot of prepared cell with D1 and D2 polymer electrolyte films and stainless-steel electrodes.

3.6 Electrochemical Properties

The electrochemical studies of the cell was done with cyclic voltammetry (CV) and impedance spectroscopy techniques.

3.6.1 Cyclic Voltammetry (CV)

Electrochemical properties and capacitance nature of the EDLC cell were studied with the cyclic voltammetry technique via the CH instrument. An electric double-layer cell was prepared with polymer electrolyte film D1 and activated carbon electrode material (datura metel). A $1 \times 1 \text{ cm}^2$ polymeric film was sandwiched in between two carbon electrodes to create the EDLC cell. The CV curve of a prepared cell with N3 film is shown in **Figure 6** at different voltages: 0.8, 1, 1.2, and 1.4V at a fixed scan rate of 0.02V/s. Similarly, **Figure 7** shows the CV curves at various scan rates: 0.01, 0.02, 0.04, 0.06, and 0.08V/s at voltage values of 0.8V, respectively). The EDLC behavior of the cell is confirmed by the rectangular shape of the cyclic curve at 0.8 V at different scan rates, as seen in **Figure 7**. At low voltage, the quasi-rectangular symmetry suggests the presence of a double layer enabling rapid distribution of ions and accumulation of charge. The high resistance prevents diffusion, and the quasi-rectangular form gradually changes as the scan rate rises. It is commonly known that capacitance and charge storage capacity correlate with the area of the CV curve [24]. Specific capacitance has been evaluated using CV curves, which measure the area at various voltages and scan rates. The specific capacitance at different voltage and scan rates is shown in **Table 2** and calculated with equation 2.

$$C_{sp} = \frac{\int IdV}{vmV}$$

where $\int IdV$ represents for the area under the curve, V represents voltage, m denotes the mass of material of activated carbon and v denotes scan rate.

It was found that the bulk resistance of the fabricated cell was 44.7ohm. **Figure 8** shows the Nyquist plot of prepared EDLC with D1 composition. The spike indicates ion absorption at the electrolyte-electrode contact and predicts capacitive behavior in EDLC cells. As the ESR decreases, the EDLC approaches the ideal capacitor. A traditional linear tilting line simultaneously appeared in the low-frequency region which indicates better performance of EDLC.

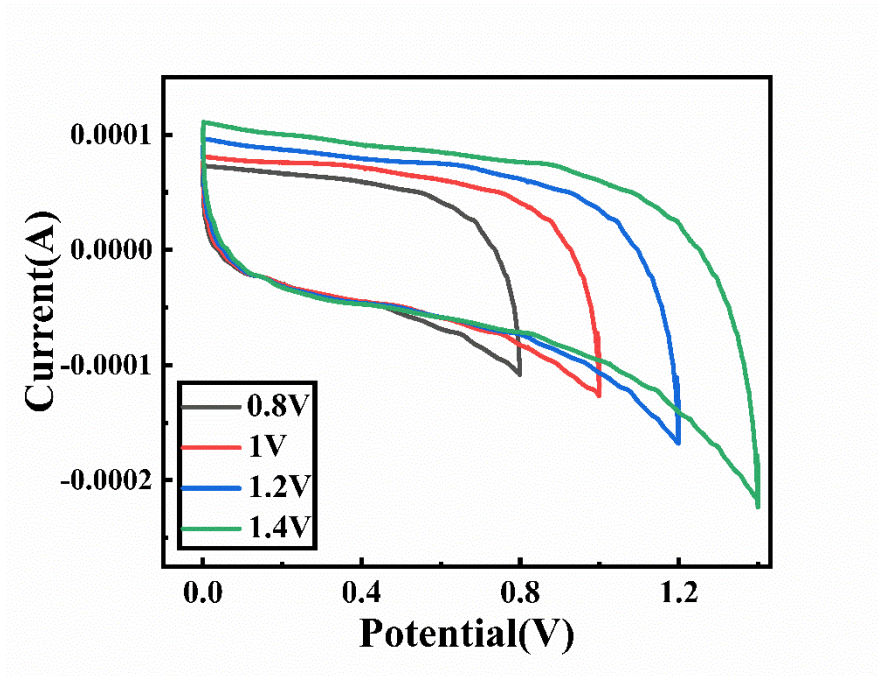


Figure 6. Current versus potential graph of prepared EDLC at different voltage with fixed scan rate 0.02V/s.

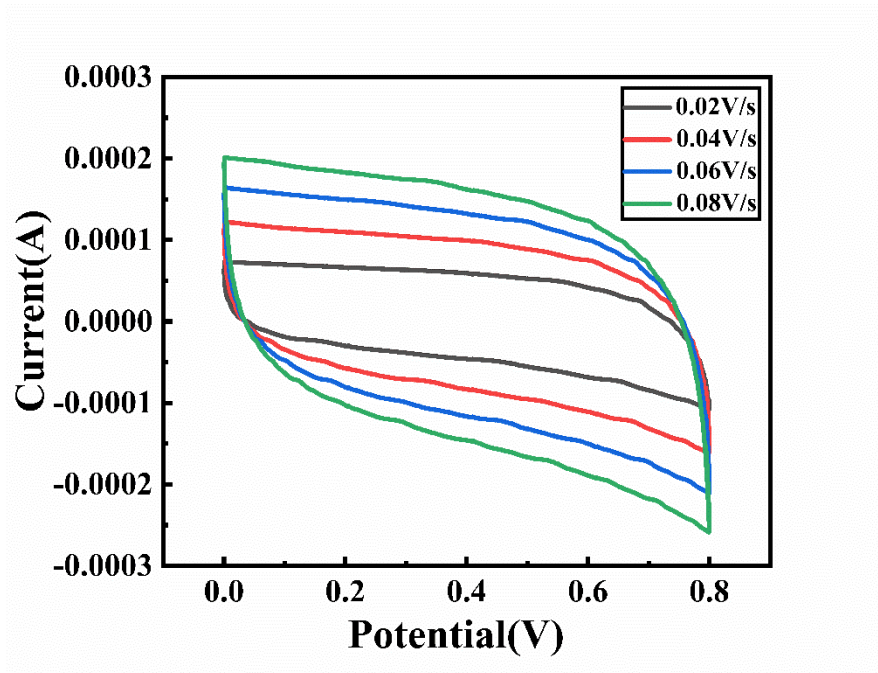


Figure 7. Current versus potential graph of prepared EDLC at different scan rates with fixed voltage 0.8 V.

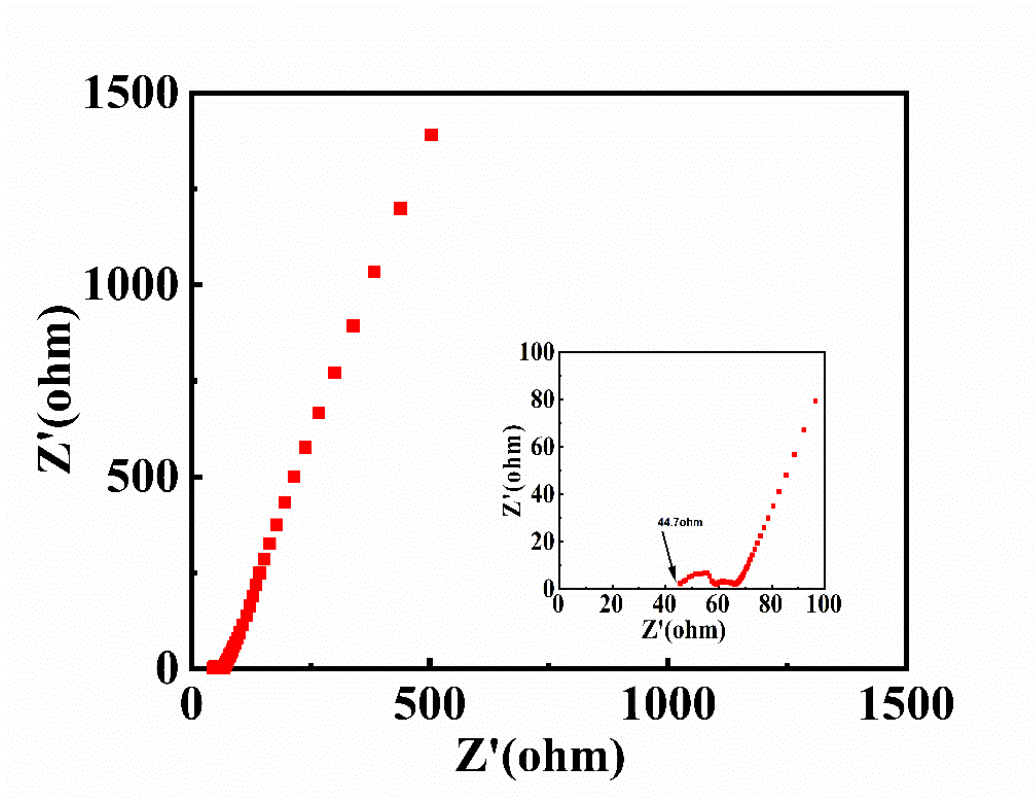


Figure 8. Nyquist plot of prepared EDLC with D1 composition and activated carbon electrode material.

Table 2. The specific capacitance of prepared EDLC cell with optimized film D1 and activated carbon electrodes.

At a fixed voltage of 0.8V		At a fixed scan rate of 0.02V/s	
Scan rate (V/s)	Specific capacitance C_s (Fg ⁻¹)	Voltage (V)	Specific Capacitance C_s (Fg ⁻¹)
0.02	7.67	0.8	5.99
0.04	5.14	1	6.80
0.06	4.67	1.2	7.74
0.08	4.33	1.4	8.71

3.7 Conclusion

We have successfully developed DES-based solid polymer electrolyte film, which has good ionic conductivity, is easy to prepare, and is safe for the environment. SEM micrograph and XRD pattern of prepared film D1 shows an amorphous nature. The prepared film D1 with the DES (industrial glycerol) has better thermal stability and conductivity was found to be in the range of 10^{-3} S/cm. With the prepared film, we have successfully fabricated an EDLC cell. The EDLCs demonstrated a suitable specific capacitance of 8.71 F/g at 0.02V/s with a voltage of 1.6 V. Thus, DES based polymer film considered as a potential candidate for fabrication of device.

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