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# Electrical properties of 1-Butyl-1-methylpyrrolidinum Hexafluorophosphate Ionic Liquid doped Polymer Electrolyte

## **ABSTRACT**

This paper reports the recent developments in electrolyte containing lonic liquids with Poly(VinylideneFluorodine-co-Hexafluoropropylene) PVDF-HFP based electrolytes for Supercapacitor applications. The electrolyte was examined using various tools. At 10wt% ionic liquid the polymer electrolyte shows an ionic conductivity of 7.7×10<sup>-3</sup> S/cm as obtained from impedance analysis. The electrochemical stability window was determined by linear sweep voltammetry and found to be 3.91V. To confirm the role of ions in charge transport, the ionic transference number was measured, which indicated the ions were the main contributors. Optical microscopy observations revealed that the crystallinity of the polymer decreased with increasing ionic liquid. The electrolyte film with the highest conductivity was used to assemble an electric double layer capacitor (EDLC). The device delivered a specific capacitance of 87.6F/g.

Keywords: Polymer, Polymer electrolyte, Ionic liquid, Ionic conductivity, Supercapacitor, SDG-7

# 1. INTRODUCTION

The two most concerning environmental issues are the impending exhaustion of fossil energy sourcesand the widespread creation of greenhouse gases as a result of their rapid usage[1]. These problems have accelerated the hunt for energy storage technologies as well as green and alternative energy sources [2], [3].

Energy storage devices and power backup are essential for ensuring consistent and reliable energy supply, allowing for future consumption whenever desired. Reducing reliance on fossil fuels and carbon emissions promotes cleaner energy production and consumption.

Batteries are essential for storing energysystems due to their high specific energy densities and ability to deliver significant power. As of now, batteries are already a part of our daily lives, but there are still certain features which display their limitations such aslower power density,safety issues,toxic electrolytes that are not environmental friendly,handling difficulty,heating

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Paper received: 06.06.2025. Paper corrected: 14.09.2025. Paper accepted: 17. 09.2025. effect whereas capacitor has higher power density but low energy density. Supercapacitor is a device that combines the benefits of batteries as well as capacitor. Comparative overview is shown in table

The supercapacitor is a promising energy storagedevice also known as Ultracapacitor are used as additional energy storage devices in electric vehicle in order to store energy from regenerative braking. Also it is used where lots of power needed in less time, like in quick accelerations. Compared to a secondary rechargeable battery, a supercapacitor has a number of advantages, including a longer cycle life, a higher power density, faster energy delivery, a shorter charging time, and environmental friendliness [6].

Electrolytes are the essential components of energy storage devices- such as batteries and supercapacitors[7], [8]. These devices rely heavily rely on the physical behavior and chemical nature of electrolyte, because the device's capacity, safety, power density, cyclabilityand rate performance. are all influenced physiochemical behavior [9], as they are essential to the movement and equilibrium of charges between the two electrodes [9]. Battery and supercapacitor performance can be greatly influenced by electrolytes[10].

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Characteristics	Battery	Capacitor	EDLC	Ref.
Coulombic efficiency (%)	70-85	~100	~99	[4]
Charge period	1-5 h	10 <sup>-6</sup> -10 <sup>-3</sup> s	sec - min	[4]
Discharge period	0.03 to 3 h	10 <sup>-6</sup> -10 <sup>-3</sup> s	Sec - min	[4]
Charge cycle count	~1000	Almost infinite	> 500000	[4]
Specific energy Wh kg <sup>-1</sup>	~ 1606	< 0.1	~ 1091	[5]
Specific power Wh kg <sup>-1</sup>	< 1000	> 10000	~ 19,6000	[5]

Table 1. Comparison among Batteries, Capacitor and EDLC(Electric double layer capacitor) on the basis of their performance

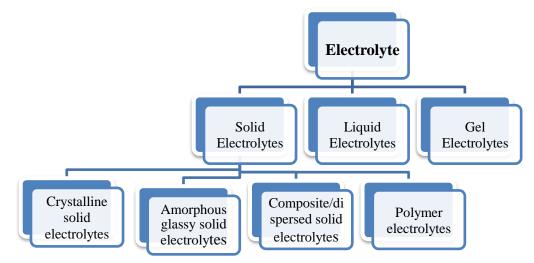


Figure 1. Schematic representation of various types of Electrolytes

To ensure long cycling life, safety, and a high power and specific energy of the supercapacitive devices-electrolytes are essential. Conductivity and ionic mobility are crucial components for determining electrolyte performance [11]. Electrolytes- The ion conductors in electrochemical devices play a vital role in determining their overall functionality and durability. Polymer electrolytes are key to supercapacitor performance. They are safer, more flexible and could be used in leak-proof operating conditions.

The drawback of polymer electrolytes is generally low ionic conductivities that restrict their exploitation technological applications. in Commonly used liquid electrolytes offering high ion transport mobility but they have a number of downsides like leakage, flammability and narrow electrochemical windows. Conversely Solid polymer electrolytes (SPEs) provides improved flexibility and safety but often compromise on ionic conductivity and mechanical properties. Numerous methods have been explored to address these including polymer blending, plasticizers, adding ionic liquid.

lonic liquids(ILs) are liquid salts, ionic liquid are the salts whose melting point is lower than the boiling point of water - consists of dissimilar size of cations and anions[12], [13] leads to the formation of ionic liquid. In fact, ILs are not a solvent but rather a mobile matrix of ions[14]. The electrolyte's

ion transport capability, one of the major key factor to construct electrochemical devices, is greatly affected by the physical characteristics of ILs, including ion size and viscosity[15], [16]. since high IL viscosity reduces energy storage device power density, especially that of supercapacitors[17].

For an ionic liquid to be liquid at room temperature those cations should be opted which are less symmetrical because the greater is symmetry the more would be the efficient packing of the cations in the lattice, and the more would be the efficient packing the higher would be the melting point. Also the greater the size of the anion, the lower would be the melting point[18]. Because the greater would be the size of the anion the more would be the reduction of force of attraction[18]. In Table 2, few electrochemical devices using examples are given where ionic liquids have been used and tabulated in table 2.

Table 2. Electrochemical Devices using Ionic Liquid

Electrochemical Devices	Ionic Liquid	Ref.
Batteries	[EMIM][TFSI] LiTFSI [DMPIM][TFSI] LiTFSI	[19]
Electric Double Layer Capacitor	[DEME][BF4]	[19]
Dye Sensitihzed Solar Cells	[PMIM][I] [EMIM][TCN]	[19]

#### 2. METHODOLOGY

To synthesize ionic liquid doped polymer electrolyte filmthe solution casting technique is employed to create polymeric electrolytes based on ionic liquids. After separately weighing the proper amounts of polymer and salt, they were dissolved in solvent and agitated at ambient temp tilla homogeneous solution was formed. Then dense solution was transferred in to petri plates, dry solid polymer electrolytes films were produced by gradually evaporating the solvent at room temperature. Prior to characterization, the polymer film was kept in a desiccator to take out moisture and eliminate any leftover solvent. Generally in this method the solvent is dissolved a predetermined amount of polymer electrolyte for four hours at room temperature, the solution should be continually stirred, adding specific amount of salt in the polymer solution and continuously stirring this till homogeneous solution is obtained. Pouring this solution into a petri dish, covering it and allowing the solvent to evaporate at RT.After drying a free standing polymer electrolyte film is obtained and the film can be peeled off for further applications and characterizations [20], [21].

#### 3. RESULTS AND DISCUSSION

Electrochemical Impedance Spectroscopy

The Electrochemical impedance spectroscopy (EIS) analysis reveals important characteristics of the electrolyte's ionic conductivity. The Nyquist plot exhibited a typical half-circle in high frequency domain with a straight portion observed at lesser frequencies. This bulk impedance of the prepared polymer electrolyte was reduced to a large extent

after incorporating ionic liquid.lonic mobility inside the polymer matrix is suggested by this comparatively low bulk resistance [22]. The values are given in table 3 and shown in figure 3.

Table 3. Effect of ionic liquid concentrations on polymer electrolyte conductivity

Composition	σ
0wt%	×10 <sup>-5</sup>
5wt%	3.90×10 <sup>-3</sup>
10wt%	7.78×10 <sup>-3</sup>
15wt%	1.11×10 <sup>-4</sup>
20 wt%	1.40×10 <sup>-4</sup>

To calculate the ionic mobility of the film, given formula is applied:

Ionic conductivity 
$$\sigma = \frac{l}{Rb \times A}$$
..... (1)

here I is the film thickness

A denotes the area

R<sub>b</sub> stands for films's bulk resistance

It is clear from figure 3 along with table 3 that mixing on mixing the ionic liquid the conductivity values get increased, attain maxima and then decreases[23]. This is a common phenomenon occurs in most of polymer IL electrolyte and widely available in literature. The equation is generally defines in increase in  $\sigma$ .

$$\sigma = nq\mu$$
 (2)

where n is the number of mobile charge carriers. q is the coulombic charge, while  $\mu$  is the ion's mobility.

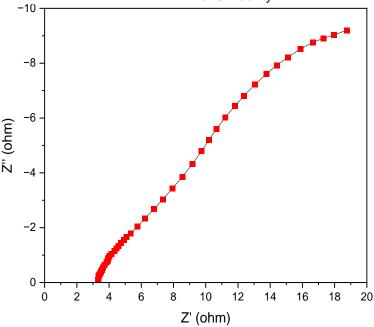


Figure 2. A typicalImpedancespectroscopy of PVDF-HFP:Nal:IL polymer matrix

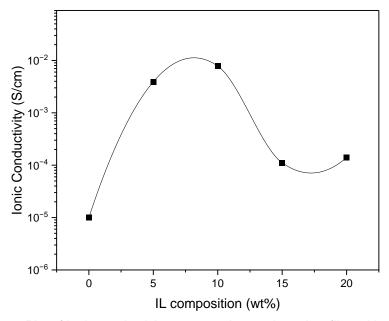


Figure 4. Plot of ionic conductivity verses polymer electrolyte filmswith different concentrations of ionic liquid

# Linear Sweep Voltammetry

The electrochemical stability of the polymeric film was analyzed with the help of LinearSweep Voltammetry. An electrolyte's potential stability is a critical factor that establishes the operating voltage for electrochemical devices. The test was done using a stainless steel cell at room temperature. The voltage was increased from 0 V to 5 V at the exposure between -3 V to 3 V. Fig. 5 shows that system began to oxidize at -1.65 V and reduction occured at 2.26V at a scan rate of 10mV/s.

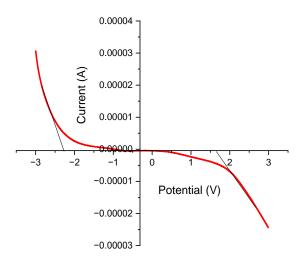


Figure 5. A Maximum conducting IL-doped polymer electrolyte's potential against current is plotted via a linear sweep voltammeter

The polymeric electrolyte film with maximum conductivitiy exhibits an ESW of 3.91 V for the system PVDF-HFP:Nal:IL1-Butyl-1-methylpyrro-

lidinium Hexaflourophosphate, when a potential is supplied over 2.26 V to -1.65 V, it increases the number of free ions, which increases current and heat and can cause the electrolyte to fail which seems suitable for most of electrochemical devices[24][25].

#### Ionic transference number

The ionic transference number(tion), also known transport number, represents percentage of the total electric current carried by a specific ionic species in an electrolyte solution or solid electrolyte material. It represents the proportion of the charge transported by the mobile ions relative to the total charge transport in polymer electrolyte context. A steady voltage was applied for about 4000 seconds throughout the material to study the electronic and ionic behavior of the film. Fig 5 shows thecurrent versus time graph. The starting current in the current as opposed to time plot comes from ions as well as electrons, while the steady remaining current is only due to the electrons[26], [27]. The ionic transference number can be calculated using the formula

$$t_{ion} = \frac{initial\ current - finale\ current}{initial\ current} \qquad .....(3)$$

Using equation we have evaluated the  $t_{\text{ion}}$ value which is 0.97, this means that 97% of the current is carried by ions which shows that our system is predominantly ionic in nature. Similar results we have obtained in almost all samples which shows that our IL mixed polymer electrolyte is predominant by ions not by electrons [28].

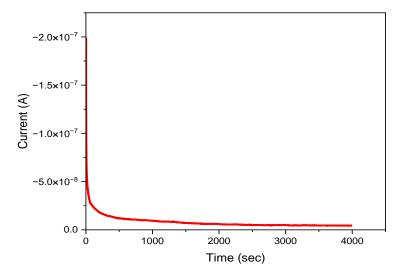


Figure 6. Curve of time against current to assess t<sub>ion</sub>value of the IL-doped polymer electrolyte with the highest conductivity

Using equation we have evaluated the  $t_{\text{ion}}$ value which is 0.97, this means that 97% of the current is carried by ions which shows that our system is predominantly ionic in nature. Similar results we have obtained in almost all samples which shows that our IL mixed polymer electrolyte is predominant by ions not by electrons [28].

## Polarized Optical Microscopy

POM was employed to study the morphological characteristics and crystalline nature of the PVDF-HFP polymer electrolyte films. Figure 7 shows the

POM images of (a) pure PVDF-HFP, (b) PVDF-HFP + Nal, and (c) PVDF-HFP + Nal + [BmPy][PF6]. Pure PVDF-HFP film shows (Fig. 7a) bright regions under polarized light, indicating its semi-crystalline nature of polymer. However, the brightness diminishes when salt is added (Fig. 4b) andfurther decreases on addition of ionic liquid (Fig. 4c). This increase in dark region indicates that mixing IL produces more amorphous region which is well known favorable condition for conductivity enhancement.

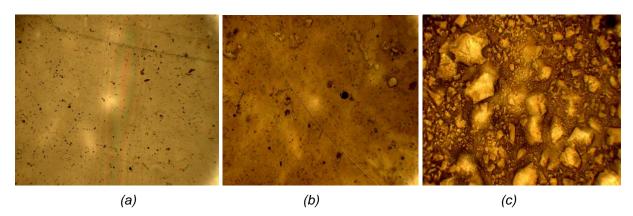


Figure 7. POM images of (a) pure PVDF-HFP, (b) Nal doped PVDF-HFP, (c) PVDF-HFP+Nal+IL doped polymer electrolyte film

# Cyclic Voltammetry

A sandwich type EDLC was made using two identical electrodes coated with carbon, between them a polymer electrolyte mixed conductive ionic liquid was placed to help ionic move easily and improve overall performance[29]. Measurements using cyclic voltammetry were conducted between -1 to +1 Voltage, with various scan rate of 10, 20, 30, 40, 50 and 100V/s were used and shown in figures 7 and 8. The evaluated specific Capacitance found was 87.6 F/g.

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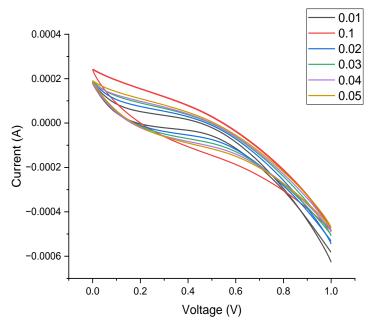


Figure 8. CV depiction of assembled EDLC within the potential range of 0 to 1V, at 0.01mV, 0.02mV, 0.03mV, 0.04mV, 0.05mV, 0.1mV scan rate.

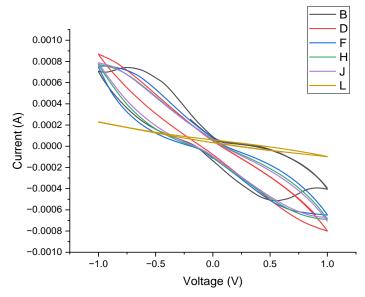


Figure 9: CV depiction of fabricated EDLC within the potential range -1 to 1 V, at 0.01mV, 0.02mV, 0.03mV, 0.04mV, 0.05mV, 0.1mV scan rate.

The following formula is used to calculate the specific capacitance

$$Cs = \frac{\int Idv}{m \, v \, \Delta V} \tag{4}$$

#### 4. CONCLUSION

The aforementioned observations and discussions support the notion that the inclusion of ionic liquids facilitates better ion transports within the electrolyte matrix. It also reduces the crystallinity and improves conductivity values of polymer electrolytes significantly. A significant

decrease in crystallinity can be achieved by adding different inorganic fillers, mixing, or low viscosity molten salts (ionic liquids), which will boost conductivity. The impedance spectroscopy confirmed a high ionic conductivity that was 7.7×10<sup>-3</sup> S/cm which indicates efficient ion transport within the polymer matrix. Linear sweep voltammetry was used to determine electrochemical stability window, extended up to 3.91V making the material suitable for high voltage operation. The ionic transference number of 0.97 confirmed that conductivity is majorly due to the ionic movement. The POM study

revealed the reduction in crystallinity of polymer film on adding ionic liquid. The optimized polymer electrolyte film was used to fabricate the EDLC, which delivers a specific capacitance of 87.6 F/g.

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#### **IZVOD**

# ELEKTRIČNA SVOJSTVA JONSKE TEČNOSTI DOPIRANOG POLIMERNOG ELEKTROLITA 1-BUTIL-1-METILPIROLIDINIJUM HEKSAFLUOROFOSFATA

Ovaj rad izveštava o najnovijim dostignućima u oblasti elektrolita koji sadrže jonske tečnosti sa elektrolitima na bazi poli(vinilidenfluorodin-ko-heksafluoropropilena) PVDF-HFP za primenu u superkondenzatorima. Elektrolit je ispitan korišćenjem različitih alata. Pri 10 težinskih% jonske tečnosti, polimerni elektrolit pokazuje jonsku provodljivost od 7,7×10-3 S/cm, dobijenu analizom impedanse. Prozor elektrohemijske stabilnosti je određen linearnom voltametrijom sa promenom i utvrđeno je da iznosi 3,91 V. Da bi se potvrdila uloga jona u transportu naelektrisanja, izmeren je broj jonskog prenosa, što je pokazalo da su joni glavni doprinosioci. Posmatranja optičkom mikroskopijom pokazala su da se kristalnost polimera smanjuje sa povećanjem jonske tečnosti. Film elektrolita sa najvećom provodljivošću korišćen je za sastavljanje dvoslojnog električnog kondenzatora (EDLC). Uređaj je isporučio specifični kapacitet od 87,6 F/g.

**Ključne reči:** Polimer, Polimerni elektrolit, jonska tečnost, jonska provodljivost, superkondenzator, SDG-7

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