

Electrolyte for SC

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Nonflammable supramolecular polymer electrolytes for flexible and high-performance supercapacitor application

Abstract

The present work reports on the high-performance and flexible supercapacitors that were assembled by using a cost effective and nonflammable electrolyte with activated carbon electrodes. The electrolyte was prepared by blending of ternary system of poly(vinyl alcohol)(P)/Glycerol (G)/Boric acid (B) hydrogel doped with LiNO₃ (Li) that was quickly formulated at ambient temperature. Hence, the partially in situ crosslinking reaction occurred in the polymer via reaction of P and B electrolyte and the polymer electrolytes were cast onto the carbon electrodes. The doped PGBLi hydrogel electrolyte retained excellent flexibility and high ionic conductivity in a broad temperature domain. The performance of assembled supercapacitor devices was measured at low and high temperature regions confirming the stability of the device. The specific capacitance of 394 F g⁻¹ at 1A g⁻¹ was provided by the supercapacitor, providing promising cyclic stability that 95.4% capacitive performance was maintained after 10,000 cycles. The device showed a specific energy of 55.8 Wh kg⁻¹ at a power of 233 W kg⁻¹. The doped PGBLi hydrogel electrolyte is cheap, biocompatible, nonflammable, easily scale up and can be anticipated to use for flexible wearable electronics.

Keywords: polymer electrolyte, boric acid, supercapacitor, glycerol, energy storage.

1. Introduction

With the rising attention to environmental and energy concerns, supercapacitors (SCs) are highly valued by the scientific community throughout the world^{1,2}. SCs exhibit more capacity than conventional capacitors and a faster charging/discharging rate than batteries³. Various applications of SCs in low-load conditions include video recorders, mobile phones, and notebook computers⁴, whereas, in high-power-load conditions, they can be used in electric vehicle power systems⁵. To evaluate the energy storage performance, electrolytes are considered the key component of SCs⁶.

However, conventional liquid electrolytes possessing high ionic conductivity face the problem of difficult packaging, easy leakage, less reliability, corrosion, toxicity, and poor low-temperature performance, significantly reducing their applications in market^{7,8}. Even though solid electrolytes have sorted out some of these problems, the poor ionic conductivity affects the electrochemical function of the whole device⁹⁻¹¹.

To address these concerns, the researchers have proposed flexible electrolytes by doping different ionic salts (NaCl and LiBr) into gel-based polymer matrix (PAAm, PVA, and PHEAA/gelatin)¹².

Such gel polymer electrolytes (GPEs) are considered the best candidates in SCs as they exhibit

higher ionic conductivity compared to solid electrolytes without any safety issues of liquid electrolytes¹³. PVA based hydrogels are one of the most used polymers during the preparation of GPEs due to their good film forming property, non-toxicity, biodegradable nature, and cost effectiveness, but they suffer from the weak ionic conductivity¹⁴⁻¹⁶. To design a novel GPE, it has

been reported that addition of few ions from inorganic salt into the polymer matrix, may lead to fast mobility of ions¹⁷. The amorphous structure of polymer provides easy paths for ions dissociation¹⁸, which makes them quite suitable for use in SCs by enhancing the electrochemical potential window¹⁹. Recently, the inorganic neutral salts of lithium like LiBF₄, LiPF₆, LiCF₃SO₃,

LiClO4, LiN(SO2CF3)2, have been proved to be highly conductive medium, when used with PVA, during synthesis of GPEs²⁰.

One problem related to hydrogel electrolytes is their less tolerance for subzero temperature, in coldest areas, which limits their application in supercapacitors^{21,22}. Researchers have introduced
³⁸ an effective way to improve the anti-freezing property of hydrogel electrolytes by introducing glycerol (a nontoxic, biodegradable, nonflammable and eco-friendly agent) into the polymer matrix^{23–25}. In the meantime, PVA based borate complexes (PVA/H₃BO₃) for the preparation of electrolytes have drawn tremendous interest of researchers from all over the world, owing to distinguished physical and chemical properties. H₃BO₃ possess several unique features including chemical stability and non-combustibility²⁶. Furthermore, scientists have reported that boron based GPEs (B-GPE) depict exceptional thermal stability, mechanical strength as well as remarkably low flammability²⁷.

¹ In this work, we developed a stable ionic conductive gel polymer electrolyte possessing excellent mechanical strength and flexibility (at ambient as well as subzero temperature), through a cost-effective one-pot synthesis route by incorporating lithium nitrate (Li), glycerol (G), boric acid (B)
⁶³ into the polymer matrix (P). The electrochemical performance of the synthesized PGBLi was investigated in a symmetric SC cell configuration using activated carbon electrodes. Various tests including cyclic voltammetry, galvanostatic charge-discharge, flexibility analysis and operational life of SC device were conducted to evaluate the electrochemical properties of synthesized GPE.
¹³ ³ All obtained results exhibited that PGBLi GPE is a promising electrolyte candidate for next generation supercapacitors.

2. Experimental

2.1. Materials

Glycerol and boric acid were obtained from Sigma Alrich. Lithium nitrate (LiNO_3) and polyvinyl alcohol purchased from Loba Chemie. Activated carbon (AC), N-Methyl-2-pyrrolidone (NMP), Timical super C65 conductive carbon (CC), and polyvinylidene fluoride (PVDF) were all acquired from MTI Corp. Split cell assembly were purchased from MTI Corp. Deionized water (DI) was supplied by Merck Milli-Q system.

16 2.2. Preparation of carbon electrodes

A specially designed mixture of activated carbon (AC) at 80% (w/w), PVDF binder at 10% (w/w), and conductive carbon (CC) at 10% (w/w) was used to make the electrodes for the supercapacitors. These ingredients were combined with a magnetic stir bar in NMP solvent at 70 °C to create a paste texture. An automated coater was used to apply the resulting slurry to the aluminum current collector (MRX Shenzhen equipment). The vacuum oven was used to dry the carbon-coated current collectors for several hours at 80 °C .

65 2.3. Preparation of the gel polymer electrolyte

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Gel polymer electrolyte was prepared by mixing 0.5 g of P in 5 mL of DI at 80 °C until it became homogeneous. After that it was cooled down to room temperature 1 g of glycerol was added into the solution and stirred at 300 rpm for 15 min. Boric acid (0.12 g) was dissolved in 2 mL of DI, separately than it was added to the mixture. Finally, lithium nitrate at different molar ratios were added to the prepared solution (0.5 M and 1 M) and mixed until a homogeneous mixture was obtained.

Scheme 1: Schematic diagram for the synthesis of PGBLi electrolyte and its different variants.

2.4. Experimental evaluation

Using Perkin Elmer Spectrum TwoTM, the FT-IR spectra of PGLi and PGBLi electrolytes are analyzed in the 400–4000 cm⁻¹ range. By heating the sample under inert air circumstances (10 °C min⁻¹), the PerkinElmer Pyris 1 was used to perform thermogravimetric analysis (TGA). Glass transition temperatures of the electrolytes were investigated by using DSC (Differential scanning calorimetry), Hitachi DSC 7000X. The samples were treated at a heating rate of 10 °C min⁻¹ under an inert atmosphere. The ion conductivity of the electrolytes was studied by Novocontrol dielectric-impedance analyzer with respect to Frequency (0.1 Hz to 3 MHz) and temperature. The thin films with a diameter of 5 mm and a thickness of around 100 μm were placed between platinum electrodes and their conductivities were measured under inert atmosphere. The DC conductivities (σ_{dc}) were derived from AC conductivity (σ_{ac}) and plotted with respect to temperature.

Studies were acquired from the electrochemical workstation using cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) (CorrTest CS3004). Using the MTI Battery Analyzer, galvanostatic charge/discharge (GCD) electrochemical tests of the produced cells were carried out (1–5 mA and up to 2 V).

2. Results and discussion

Figure 1: **a)** FT-IR spectra of the electrolytes PGB1Li, PGB0.5Li, PG0.5Li, PG1Li at various doping ratios. **b)** TGA curves of PGB1Li, PGB0.5Li, PG0.5Li, PG1Li **c)** DSC profiles of the PG, PG0.5Li, PG1Li. **d)** DSC profiles of the PGB1Li, PGB0.5Li. **e)** AC conductivity of PGB1Li **f)** DC conductivity of PGB1Li.

In all spectrum, Glycerol (G) displays intense and sharp absorptions at $1150\text{-}930\text{ cm}^{-1}$ belonging to C-O stretching. The C-H stretchings of G and P are illustrated by bands in the range of 3000– 2850 cm^{-1} . The broad peak at $1250\text{-}1480\text{ cm}^{-1}$ can be attributed to C-H and C-C-O vibrations of P.²⁸

The strength of these peaks slightly increased after doping with boric acid (PGB1Li and PEGB0.5Li) which may be due to partial crosslink as confirmed by the insolubility of these products. Typical O-H stretching at 3400 cm^{-1} is associated with O-H vibrations of P and G (Fig. 1a). Thermogravimetric analysis (TGA) demonstrated that all the electrolytes are thermally stable up to at least $200\text{ }^{\circ}\text{C}$ (Fig 1b). For boric acid doped electrolytes, 10% the weight decay up to the degradation temperature is related to humidity loss, i.e., PGB1Li and PGB0.5Li. DSC profiles of the electrolytes illustrated that they have low glass transition temperatures, T_g ranging from -20 to $-5\text{ }^{\circ}\text{C}$ which can be attributed to softening effect of Glycerol. This indicated the possible utilization of boron doped PGB0.5Li and PGB1Li whithin a broad temperature domain up to at least $200\text{ }^{\circ}\text{C}$. The onset of melting temperature for PG and PG0.5Li are 100 and $120\text{ }^{\circ}\text{C}$, respectively. However, PG1Li has no melting transition which may be due to complexation in the

presence of excess lithium salt. Similarly, partially chemical crosslinked samples PGB1Li, PGB0.5Li have weak melting transition in the same domain.

The σ_{ac} ($= \sigma'(\omega)$) versus frequency at temperatures from 0 to 90 °C are illustrated in Fig 1e. σ_{ac} was calculated according to Eq 1²⁹⁻³³

$$\sigma'(\omega) = \sigma_{ac}(\omega) = \varepsilon''(\omega) \omega \varepsilon_0 \quad (1)$$

Real part of the conductivity is $\sigma'(\omega)$, angular frequency is $\omega = 2\pi f$, vacuum permittivity ($\varepsilon_0 = 8.852 \times 10^{-14}$ F cm⁻¹) is ε_0 and imaginary part of the complex dielectric permittivity ($\varepsilon^* = \varepsilon' - i\varepsilon''$) is ε'' . The curve includes frequency dependent conductivity lower frequencies domains due to electrode polarizations and frequency independent plateau regions (at intermediate or higher frequencies). The σ_{dc} corresponding to lithium ion conductivity of the biopolymer electrolytes was obtained from the σ_{ac} data according to Eq. 2.²⁹⁻³³

$$\sigma(\omega) = \sigma(0) + A\omega^n \quad (2)$$

where $\sigma(0) = \sigma_{dc}$ and A, n are constants where $0 < n < 1.0$.

Fig. 1b illustrates the σ_{dc} value of the as a function of temperature which can be interpreted with Vogel tamman Fulcher (VTF)²⁸

Clearly, the Li⁺ ion conductivity of PGB1Li illustrates the curved line which increased with a temperature activated process. The curvature is a proof of concept for faster ion migration via contribution of segmental relaxation.

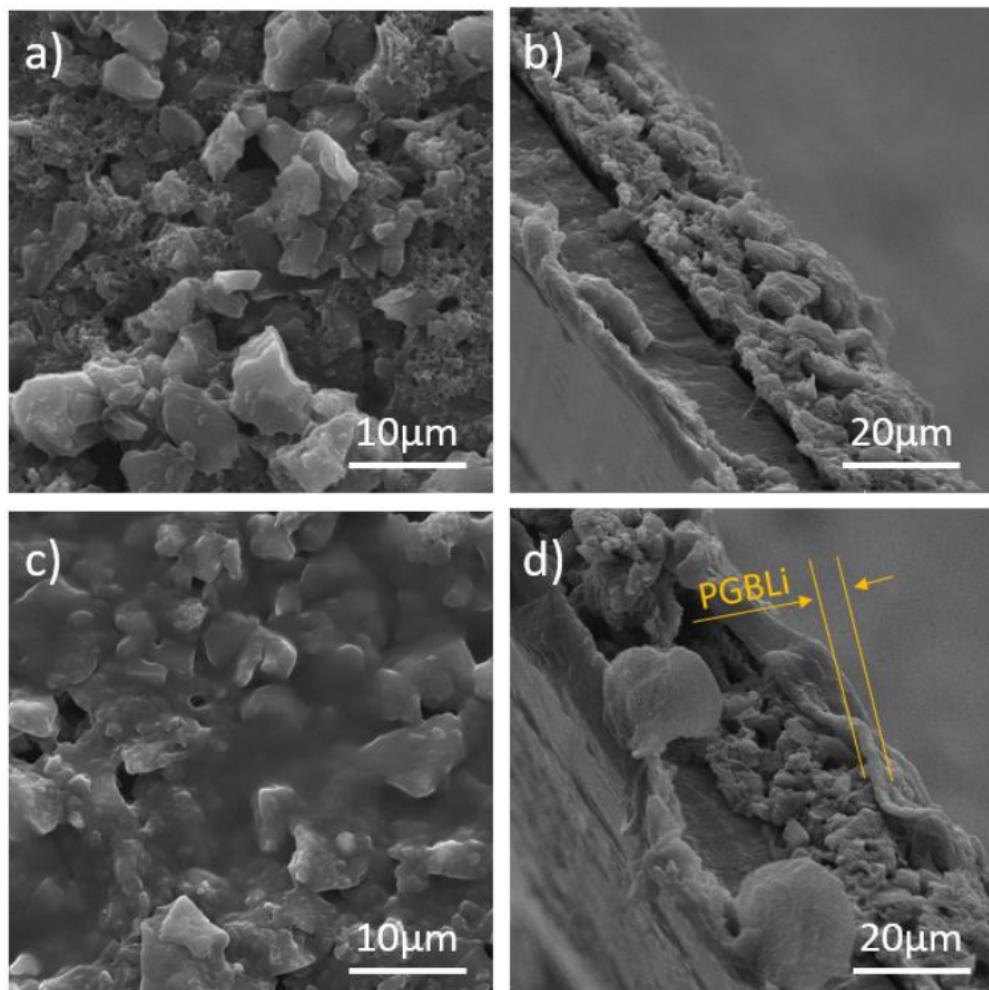


Figure 2: a) Surface , b) cross section of the uncoated carbon electrode, c) surface of PGB1Li casted d) cross sectional images of PGB1Li casted carbon electrode.

Fig. 2a show the surface morphology of carbon composite electrodes with a uniformly distributed active carbon and conductive carbon particles created porous network. An average thickness of the coating ($20 \mu\text{m}$) is obtained by cross-sectional SEM images of the carbon composite electrode. A thin electrolyte film is coated on the electrode by a simple casting method which covered the surface of electrode as shown in Fig. 2. The electrolyte directly diffuse porous networked carbon

electrode onto the current collector. The cross-section of the electrode coated with electrolyte shows the electrolyte thickness and it observed that a consistently thin electrolyte coating formed,
5 covering the entire electrode surface.

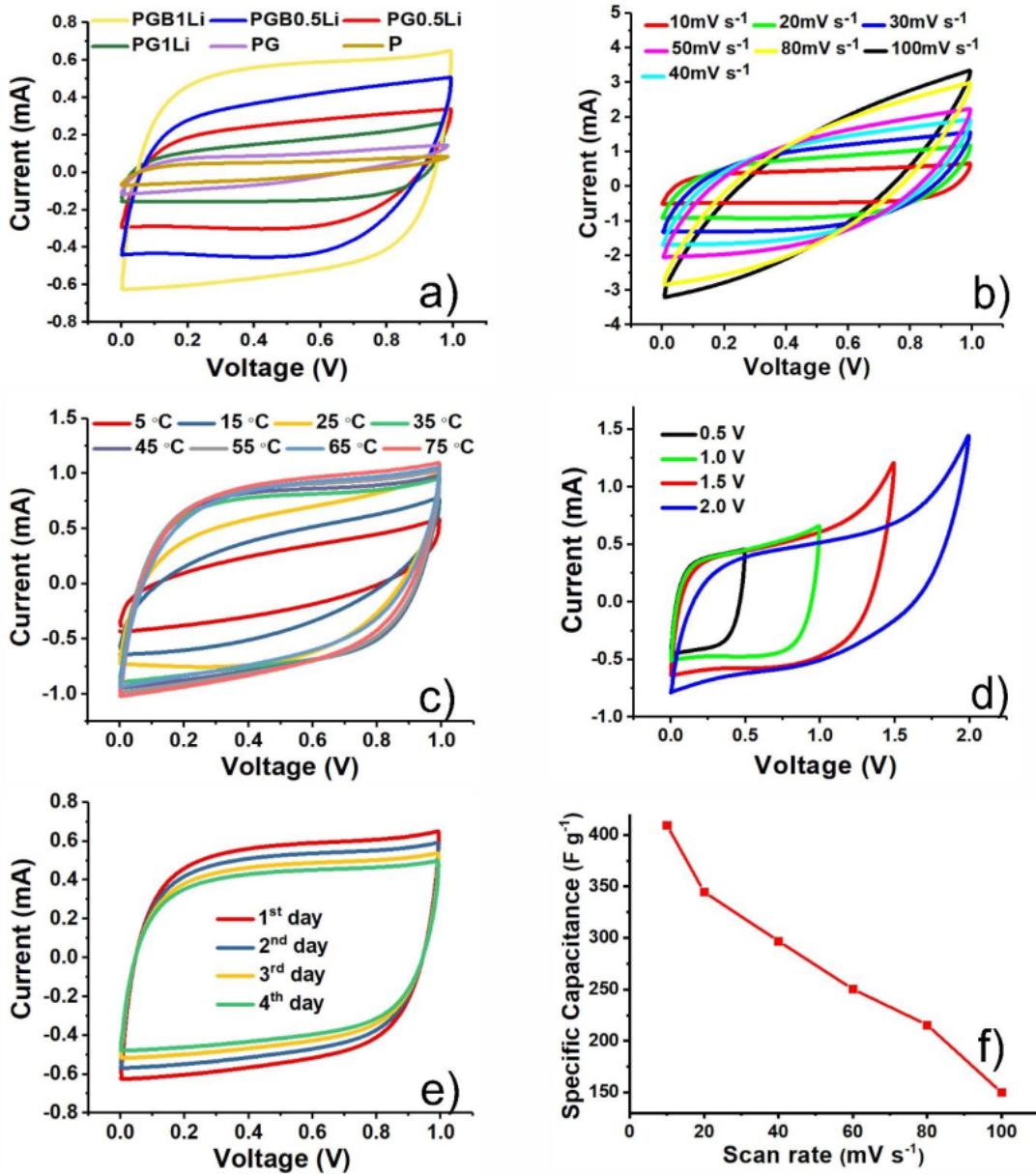


Figure 3a) Comparison of CV of PGB1Li, PGB0.5Li, PG1Li, PG0.5Li, PG, P **b)** CV of PGB1Li at different scan rates $10 - 100 \text{ mV s}^{-1}$, **c)** CV of PGB1Li at different temperature ($5 - 75^\circ\text{C}$), **d)** CV curves of PGB1Li based supercapacitor at different potential windows ($0.5 \text{ V} - 2.0 \text{ V}$) **e)** CV curves of PGB1Li during different days **f)** Specific capacitance of PGB1Li based device at different scan rates.

Figure 3a depicts the CV measurements of supercapacitors containing PGB1Li, PGB0.5Li, PG1Li, PG0.5Li, PG and P at a potential window of 0 to +1 V and a scan rate of 10 mV s^{-1} . The supercapacitor prepared from PVA and PVA-Gly electrolytes showed low capacitance. CV profiles demonstrated a quasi-rectangular shape, indicating a suitable electric double layer capacitor (EDLC) behavior, the capacitance that is stored at the electrode/electrolyte interface by an electrolyte ion accumulation³⁴.

While the composition of supercapacitor exhibited the improved specific capacitance with Li ions. Li^+ transport in supercapacitors affected by the homogeneous-sized pores of the electrode³⁵. The ionic size of Li and the mobility of the electrolyte used could play a significant role in adsorb and thus specific capacitance of the device³⁶. Increasing the concentration of Li^+ ions in the electrolyte structure improved the supercapacitor's capacitance behavior. The ionic transport mechanism in a polymer electrolyte depends on the size, ion pairing, the nature of the amorphous phase, the dielectric constant of the polymer host, and the velocity of charge carriers (both anions and cations)³⁷.

The forward-reverse scan of the CV plot of PGB1Li electrolyte was the optimum, where the PGB1Li-based system had the highest specific capacitance C_s of 409 F g^{-1} at 10 mV s^{-1} compared to only 104.4 F g^{-1} of PG1Li. This could be attributed to the additional contribution of boric acid,

⁵ indicating a suitable environment after partial crosslinking between PVA and boric acid, and ⁴⁸ attributed to the PGB1Li's high ion mobility.

⁴⁸ Figure 3b showed a significant improvement in the electrochemical performances of the supercapacitor when measured with the PGB1Li system at potentials ranging from 0 to 1 V and ¹⁷ scan rates ranging from 10–100 mV s⁻¹. It is observed that the current density rises as the scan rate ³ is steadily raised. By maintaining the CV forms at low and high scan speeds in forward and reverse ⁵ scans, the PGB1Li curves demonstrate that the material may offer ion transitions in both situations ⁷⁴ (fast and low current rates)^{38,39}, leading to better diffusion-controlled ion transfer. By facilitating ion transport between electrodes within the supercapacitor, this characteristic exhibits the capacity to hold charge.

¹¹ The thermal behavior of the fabricated device (PGB1Li-based supercapacitor) was investigated using CV measurements at temperatures ranging from 5 to 75 °C (Figure 3c). With increasing ⁵ temperature, CV plots present further enhanced capacitive performance with excellent cyclic ¹¹ stability under the forward-reverse scans at a scan rate of 10 mV s⁻¹. When the temperature rises to 25 °C, the supercapacitor's capacitance increases gradually. Clearly, no significant specific capacitance increase was noticed at further variation of temperatures (35 to 75 °C), which can be shown by the high stability of the system through the crosslinking structure of the electrolyte and the reach of ion saturation.

⁴⁷ Figure 3d exhibits the CV curves of PGB1Li based supercapacitor cells in different potential windows at the scan rate of 10 mV s⁻¹. The trend of CV curves shows almost rectangular behavior up to 1 ⁵⁵ indicating that the supercapacitor can be safely operated, reversibly cycled, and is stable in different voltage ⁴⁰. However, for potential exceeding 1 V, the CV curve deviated from a semi

rectangular form. This limited operating potential of device is mainly due to limited ESW (electrochemical stability window) of the electrolyte, which is restricted⁴¹. After 4 days, PGB1Li based supercapacitor displayed stable performance with a near perfect CV curve as shown in Figure 3e. The variation of specific capacitance with scan rates different was calculated in PGB1Li electrolyte (Figure 3f). As seen in the figure, the specific capacitance is 380 F g^{-1} at a low scan rate (10 mV s^{-1}), then reduces with increase in the scan rate due to insufficient time for the electrolyte to adsorb and desorb on the electrode surface³⁶.

3.2 Electrochemical measurements

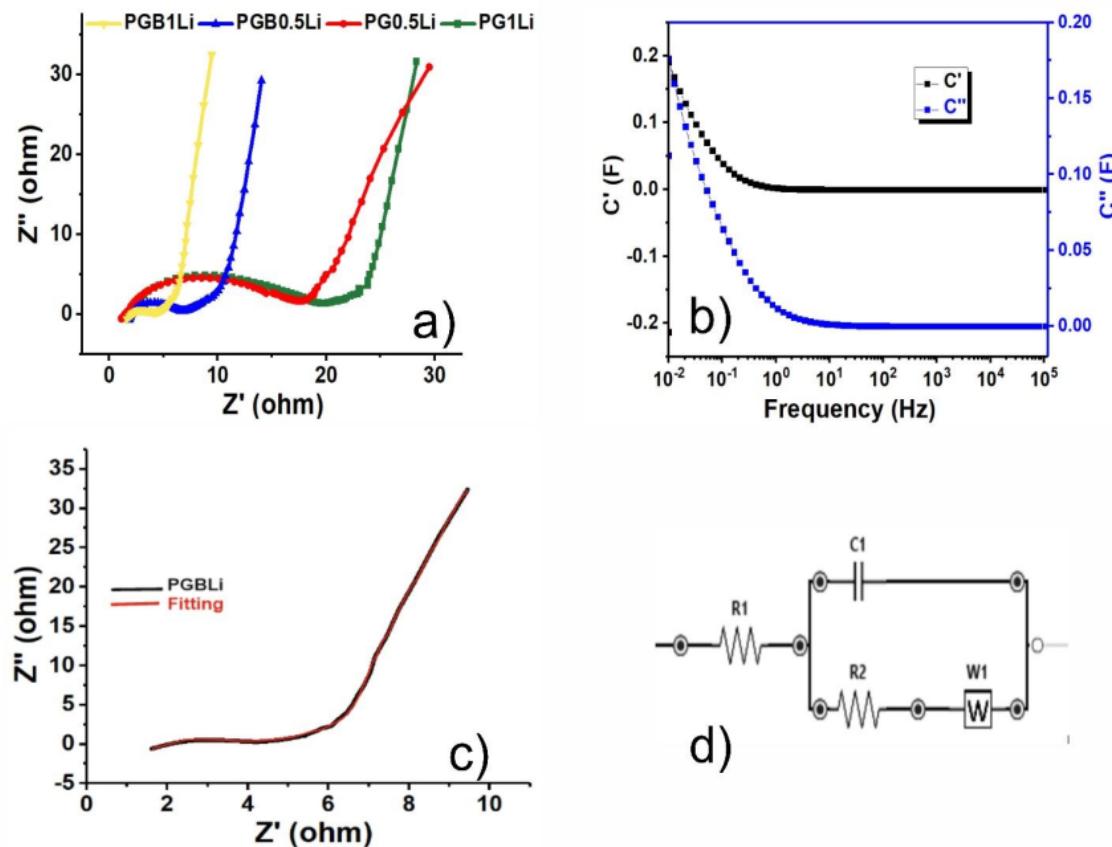


Figure 4a) EIS curves of supercapacitors including different electrolytes **b)** Complex real (C') complex imaginary capacitance (C'') vs. frequency (Hz), **c)** AC conductivity of PGB1Li d) DC conductivity of PGB1Li.

Table 1. ESR and R_{ct} of the PGB1Li, PGB0.5Li, PG0.5Li, PG1Li Supercapacitors

Electrolyte	ESR (Ω)	R_{ct} (Ω)
PGB1Li	1.68	3.32
PGB0.5Li	1.81	4.92
PG0.5Li	1.96	5.19
PG1Li	2.01	16.97

Nyquist plots of all types of electrolytes were studied of 10 mHz to 100 kHz as it shows in Figure 4a. The behavior of the supercapacitor can be displayed in the low-frequency region in vertical rising shapes of EIS curve parallel to the Z-axis , the charge transfer resistance (R_{ct}) and conductivity of the device can be shown in the high-frequency region of the diameter of the semicircles [34], and the electrolyte resistances can be also presented in high-frequency region as equivalent series resistance (ESR) [35,36]. The results show an ESR value of 1.68 ohm was obtained for the PGB1Li solid electrolyte, which is lower than the 1.81, 1.96, and 2.01 ohms for the PGB0.5Li, PG0.5Li, and PG1Li electrolytes, respectively which is showed in Table 1. The Warburg impedance is defined as a straight line below-frequency frequency and real axes [37].

The complex real capacitance (C') and complex imaginary capacitance (C'') of the CAG- Co based supercapacitor are shown in Fig 4b. The (C') and (C'') component of the impedance acquired from the EIS analysis, are computed using the equations [38] .

$$C' = -\frac{Z''(\omega)}{\omega|Z(\omega)|^2}$$

$$C'' = -\frac{Z'(\omega)}{\omega|Z(\omega)|^2}$$

The significant increase in C' values at 10 Hz frequency for CAG- Co based supercapacitor indicating the relaxation type behaviour.

The values are derived by fitting the experimental data using the analogous circuit as shown in

Fig 4c . The resistor connected in parallel with the capacitance (C) is used to exact fit in the

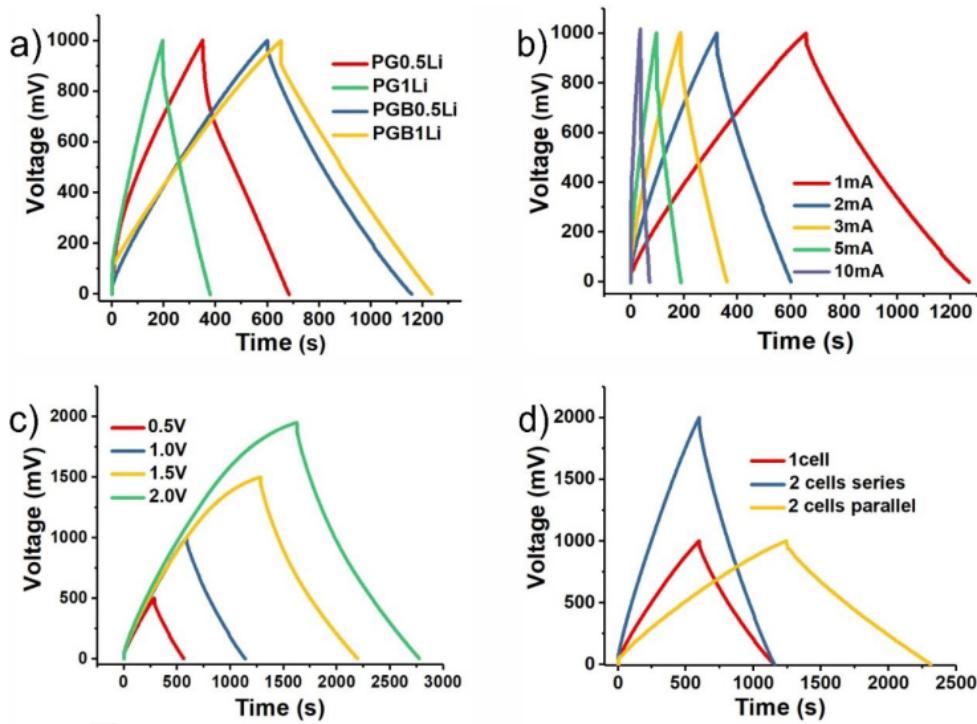
equivalent circuit model (Fig. 4d). In Fig 4c, a typical analysis of the PGB1Li based

supercapacitor is represented as black squares, and the fitting data is plotted as red line. A

straight line parallel to the Y-axis represents capacitance behavior in the low-frequency range,

suggesting that excellent chemistry between PGB1Li electrolyte and the electrode has been

accomplished, which contributes to charge storage capacity.



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Figure 5: a) Comparison of GCD profiles at 1 mA specific current, b) PGB1Li at different
 5 current densities c) GCD profiles of PGB1Li at different voltages d) GCD profiles of PGB1Li
 based supercapacitor connected in series and parallel.

In a typical GCD test, the system is charged and discharged based on the operating voltage window and constant current, while the change in voltage versus time is recorded. Fig.5a shows a comparative GCD of the PG_xLi, and PGB_xLi (x shows molarity of Li) electrolytes at a specific current of 1 mA and potential window from 0 to 1 V. Optimizing the concentration of the Li in the GPE and adding the boric acid can provide enhanced ion access in the electrolyte and a faster charge transfer rate. The high discharge time may refer to form the BO₃²⁻ in the electrochemical processes and by charging and discharging the crystal growth of BO₃²⁻ modified ⁴². In addition, increase the concentration of the Li additive with boric acid led to longer charge-discharge periods since the boric acid modified electrolyte inhibits the lithium dendrite growth and promote the

dissociation of lithium salts and increase the Li⁺ concentration base on the Lewis acid-base combination ⁴³. Therefore, the PGB1Li was determined ⁷ A series of GCD experiments were conducted to analyse the stability and investigate the electrochemical performance. The PGB1Li electrolyte was tested at various current density in voltage range 0 to 1 V as shown in Fig.5b. The discharge time which is proportional to specific capacitance and as current density increases, discharge time decreases owing to delay for accumulation of the charges on the surface of electrode.

⁷ As illustrated in Fig.5c, the stability window of the device was tested at various voltage sweeps of ¹⁰ 0 – 0.5 V, 0 – 1 V, 0 – 1.5 V and 2V ⁷ at constant current density of 1 mA. All supercapacitors ⁶¹ exhibit a triangular shape corresponding to an electrical double layer capacitor (EDLC). The ¹⁰ assembled supercapacitor is stable within 2 V domain which can be selected as the cell potential.

Fig. 5d exhibits the devices in series connection having the potential of 2.0 V at a current density of 1 mA while maintaining its original shape. ¹⁰ Additionally, the discharge time in the parallel connected devices was doubled as compared to the single cell that related to enhancement of the capacitance of the supercapacitors, indicating the improvement in the overall capacitance.

² To show the rate capability of devices within the voltage range of 0–1 V, Figure 6a shows the ²⁶ specific capacitances (C_s) of manufactured supercapacitors, measured against different specific ² currents of 1, 2, 3, 5, and 10 A g⁻¹. The following Eq. (1) was used to evaluate the C_s ,

$$C_s = \frac{2I\Delta t}{w\Delta V} \quad (1)$$

Where I , Δt , w , and V , respectively, stand for the discharge current, discharge time, active material ² mass on an electrode, and discharge potential window. For the other fabricated supercapacitors

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Figure 6: a) Specific capacitance, b) Ragone plot f the PGB1Li, PGB0.5Li, PG0.5Li and PG1Li based supercapacitor devices, c) Cycle number, d) Number of Days of the PGB1Li based supercapacitor devices.

(PG1Li, PG0.5Li, and PGB0.5Li), the observed C_s values were 97,199, and 300 F g^{-1} at 1 A g^{-1} current, respectively. A supercapacitor can have a maximum 394 F g^{-1} of C_s when using electrolyte PGB1Li. Clearly, the insertion of boric acid in the polymer electrolyte resulted in the improvement of C_s . It was reported that the positive contribution of the boric acid derivatives could be due to direct contribution of the additive in the electrochemical reaction where the performance of the energy storage system progressively improved by the formation of boron based intermediates⁴².

The energy and power density performances of all manufactured supercapacitors are shown in Figure 6b after being derived from GCD profiles using Eqs. (2) and (3), respectively.

$$E = 1/2 \times C_s \times \Delta V^2 / 3.6 \quad (2)$$

$$P = 3600 \times E / \Delta t \quad (3)$$

where E and P stand for, respectively, energy density and power density. The PGB1Li device outperformed all other supercapacitor devices in terms of highest energy density, reaching 55.8 Wh kg^{-1} at a matching power density of 233 W kg^{-1} . However, with a power density of 510 W kg^{-1} , a minor fall was seen, dropping to 44.98 Wh kg^{-1} . This shows that the device performs admirably at large discharge currents. 10000 GCD cycles at 1 A g^{-1} current were used to test the PGB1Li-based device's cyclic charge-discharge performance. Figure 6c shows the device specific capacitance and coulombic efficiency. The device yielded a remarkable cyclic stability was

confirmed after 10,000 cycles, during which time it maintained almost 95.4% of its initial capacitance. In addition, the supercapacitor device showed a columbic efficiency of 94% after 10000 charge-discharge cycles which may be the result of the electrode-electrolyte interaction allows ion transportation without a loss⁴⁴. Figure 6d demonstrates the stability of the same device obtained from GCD analysis at different period. It was observed that the supercapacitor device containing PGB1Li retained approximately 94% of its initial capacitance after 30 days of measurements.

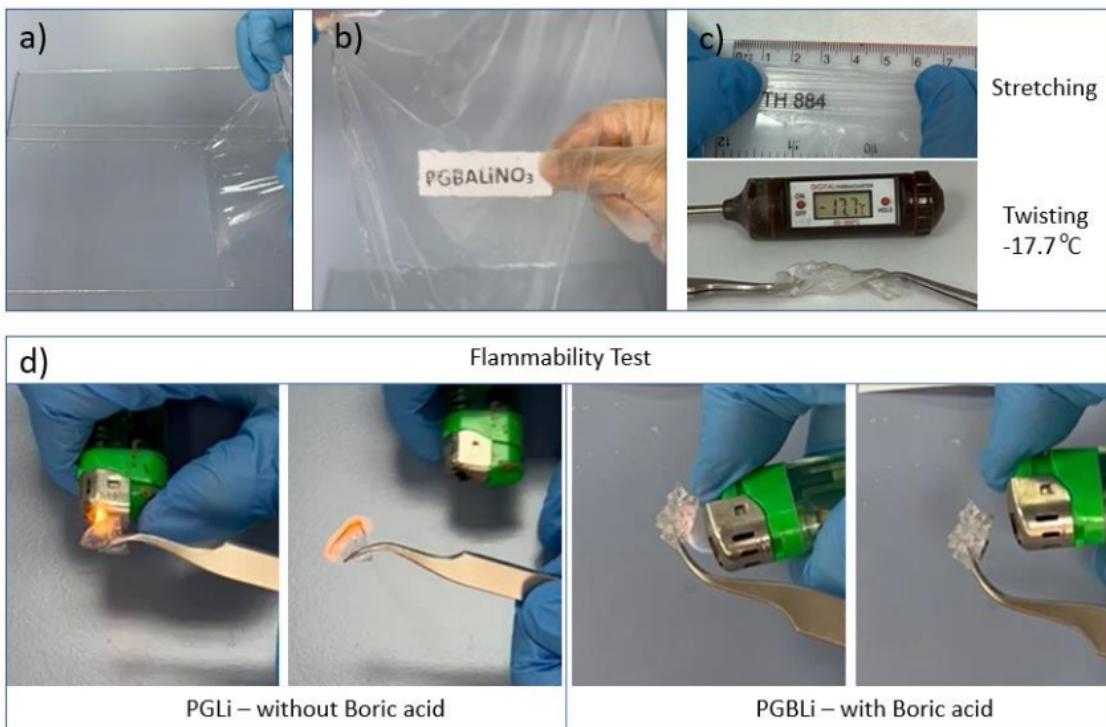


Figure 7. **a – b)** Flexible and transparent PGB1Li electrolyte, **c)** Stretching and low temperature flexibility of PGB1Li, **d)** Comparative flammability test with and without boric acid loaded electrolyte.

Electrolyte properties such as ease of preparation, scalability, flexibility and mechanical robustness are key properties for energy storage device applications. Figure 7a shows the easy removal of the PGLi electrolyte from the glass substrate poured during preparation. Scalability and ease of use are illustrated in Figure 7b as a transparent electrolyte in A4 paper size. The same electrolyte performed very well under mechanical tests under different stretching and bending conditions. In particular, electrolyte maintains its flexibility performance at -17.7 °C as shown in Figure 7c.

The flame-retardant property of electrolytes is a very important property in the operational uses of energy storage systems. Figure 7d compares the flammability performance of boric acid doped and undoped PGLi electrolytes. The PG1Li electrolyte, which does not contain boric acid, appears to catch flame when the temperature increase. However, it was observed that the rapid temperature changes have no effect on the structure of boric acid loaded PGB1Li electrolyte and the system does not catch flame under the same conditions.

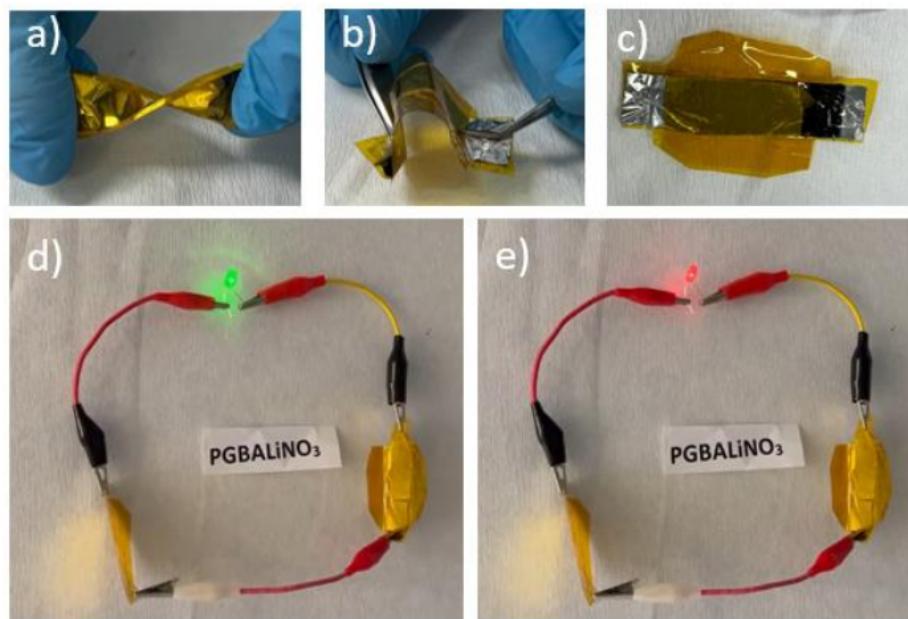


Figure 8. a-c) flexibility test of supercapacitor, d-e) operational stability of two supercapacitors connected in series.

The flexibility of a PGB1Li-based supercapacitor was examined under various bending, twisting, conditions, as depicted in Figures 7 a-c. We noticed no physical or mechanical deterioration in bending situations and excellent mechanical properties were obtained for the device at all bending angles from free state to 150° bending. Operational stability of two supercapacitor devices connected in series was tested by RGB light emitting diodes. The RGB led has been successfully operated after charged at 2.5 V for 3 min. (in the supplementary video).

Table 1. Comparison of the Specific Capacitance, Energy density, Power density, and cycles of Bio-Electrolyte-Based Supercapacitors with the Present Report.

Electrode	Electrolyte	8 Capacitance	Energy density	Power density	Cycle stability	Ref.
		F g ⁻¹	Wh kg ⁻¹	W kg ⁻¹	Cycle	
AC	PGBLi	394	55.8	233	10000	This work
Carbon nanofibers	PVA-H ₂ SO ₄ -H ₃ BO ₃	134	67	1000	1000	[51]
AC	PVA/H ₃ PO ₄ /(EMIM) BF ₄	271	54.3	23880	3000	[47]
AC	PVA-H ₂ SO ₄ -P- benzenediol	474.29	11.31	1000	3000	[55]
AC	CMC-PVA/LiNO ₃	~100	3.2	400	5000	[53]
AC	PVA/ Na ₂ SO ₄	130	13	10000	8000	[52]
AC	PVA/LiClO ₄ / SPEEK(Li)	146.96	18.26	1100	3000	[50]
AC	PVA/NaCl/glycerol	211	7.4	250	2500	[49]
Carbon composite	(Gly)/(BA)/ KOH	327	45.4	920	10000	[45]
JC	Glycerol/KOH	150	20	500	10000	[56]
	PVA/ λ -carrageenan / EG	113.6	24.3	24.3	10000	[57]

AC	EtOPC/glycerol/LiTFSI	162	14.2	7500	2400	[58]
AC	PVA-H ₂ SO ₄ -ARS	4 ₇₁	39.4	400	1000	[59]
AC	PVA-PVP-H ₂ SO ₄ -MB	328 F g ⁻¹	10.3 Wh kg ⁻¹	246 W kg ⁻¹	2000	[60]
AC	Gly/KOH/Mo	328	45.6	497	25000	[61]
AC	Gly/H ₃ PO ₄ /Co	349	47	420	15000	[62]
AC	HSS-S-F/PVA	147	20.40	-	2250	[63]

Table 1 compares various metrics, including device capacitance, current density, cycle numbers, and energy density, used in the construction of supercapacitors employing various electrode and electrolyte materials as described in the literature. It is evident from Table 1 that the manufactured supercapacitor indicates outstanding cycle stability at high energy density and is therefore very competitive among other supercapacitors reported recently in the literature. The device constructed in this study has proven its stability up to 10,000 cycles, and the capacitance loss of the device is quite modest based on the decline in coulombic efficiency. As compared to previously published literature, the precise capacitance values showed that the device has a promising dynamic potential in electrochemical applications.

Conclusion

In conclusion, a novel partially crosslinked non-flammable and temperature tolerant electrolytes were used to assemble the flexible supercapacitor with the activated carbon as electrode. Li doped PBG based supramolecular polymer electrolyte was employed as both separator and electrolyte. A facile fabrication of the electrolyte, high conductivity, cost effectiveness, and easy assembly process can encourage the use of this polymer electrolyte for various energy storage systems. The doped PGB electrolyte exhibited an excellent electrochemical performance with high specific

capacitance of 394 F g^{-1} at 1 A g^{-1} . After 10,000 GCD cycles at a current density of 1.0 A g^{-1} , that maintained 95.4% of initial capacitance at various angles proving the flexibility for the device. Thus, these multifunctional electrolytes can be scaled up and can be easily recycled and reused. They illustrated high performance in the quasi-solid-state supercapacitor and can be endowed for the development of facile energy storage devices within a broad temperature domain.

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