

Microbial Cellulose-Derived Interconnected Carbon Nanotube Networks as Binder- and Metal-Free Electrodes for Electrochemical Capacitor

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Microplastic and metal waste from electronic industries are now a major global problem due to high demand in smart technology. This work aims to fabricate a green electrical double-layer capacitor (EDLC) using natural resources. An eco-friendly electrodes film are produced by the presence of green polymers called microbial cellulose from symbiotic culture of bacteria and yeast (SCOBY) via harmless, inexpensive and simple procedures. The microbial cellulose electrodes (MCE) are paired with a thin layer of green polymer electrolyte (GPE). Glycerol serves as the plasticization agent for alternative pathways for ionic migration. The most optimum GPE possesses good ionic conductivity of $\sim 10^{-3}$ S/cm. From transference number analysis (TNM), ions have found to be the dominant charge carrier in the GPE as the TNM number of ion is close to unity. Linear sweep voltammetry (LSV) analysis illustrated that the GPE is electrochemically stable up to 2.4 V. The GPE is sandwiched between two identical MCE to form an EDLC. Cyclic voltammetry (CV) analysis showed that the green EDLC stores energy through non-Faradaic mechanism and the specific capacitance is influenced by the sweep rates. The EDLC can be charged and discharged up to 2 V and it showed a great cyclability.

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Introduction

The demand for the storage as well as power backup system has increased the use and development of energy storage devices that can accumulate the electrical energy produced. The most popular and common energy storage devices are batteries e.g. lithium-ion batteries and lead acid batteries [1]. There is no doubt that the conventional lithium-ion batteries have outstanding performance and have been used in various gadgets such laptops, tablets, smartphones and camera. However, lithium-ion batteries possess cobalt in it which is a threat to the environment. Other than that, lithium can be explosive [2]. Supercapacitors (SCs) are one of the alternative to this drawback due to its simplicity, low cost and safe preparation procedures [3]. SCs can be categorized into three major types which are electrochemical double layer capacitor (EDLC), pseudocapacitor and hybrid capacitor. Each of these capacitors have different energy storage mechanism due to the different choices of electrode materials [4].

The energy storage mechanism of EDLC includes non-Faradaic process as ions from the polymer electrolyte form double-layer on its carbon-based electrodes [5]. EDLC has some unique characteristics such as good thermal stability,

large cycling stability, excellent reversibility, high power density, rapid charge-discharge rate, cost efficient, safe and easy fabrication method [6]. One of the most suitable materials for EDLC applications is carbon, owing to its high mechanical strength, electrical conductivity, abundance, controllable structure and low density. Numerous forms of carbon e.g. carbon black, carbon nanotubes (CNTs), natural or artificial graphite, graphene, and activated carbon, are used in commercial SCs as active materials or conductive additives form ionic polarization process [7-9]. CNT has attracted so many attentions due to their electrical conductivity, intrinsic high-aspect ratio, excellent mechanical strength and chemical stability [10]. According to the work by Deng et al. [11], the inclusion of 6 wt.% CNT into a fibrous cellulose acetate network has increased both surface area and the electrical.

Microplastics are currently a global problem that can seriously affect human's health and ocean [12]. This plastics travel through the domestic water supply. Microplastics are discovered in marine ecosystem in a form of pellet, scrubber, fibers or fragment that are consisted of various non-biodegradable synthetic polymers e.g. polyvinylidene fluoride (PVdF), polyacrylonitrile (PAN),

polyurethane (PUR), polyvinyl chloride (PVC) and polymethyl methacrylate (PMMA) [13, 14]. Commonly, commercial batteries and SC contain PMMA or PVdF as the polymer electrolyte or binder in the electrode, respectively. Thus, to replace the expensive, toxic and non-biodegradable binders, microbial cellulose from symbiotic culture of bacteria and yeast (SCOBY) is used as the binder for the electrodes. Microbial cellulose has an amazing polymeric networks in their structure that can enable us to form a metalless electrodes. Most of the time carbon paste is coated on a metal foil, in the case of commercial SC. Other unique characteristics of microbial cellulose are bendable, environmental friendly, excellent mechanical strength and excellent film forming properties [15, 16]. The weblike structure of microbial cellulose enables the doping of active material like CNT to form a high surface area electrode.

Apart from electrodes, polymer electrolytes are a key component in achieving green, safe and biodegradable supercapacitors. Most commonly used polymers in the polymer electrolyte or electrode separators of commercial energy storage devices are PAN, PVdF and PMMA. Another safe option to eliminate these problems is to use natural polymers. Natural polymers are abundance, inexpensive and harmless to human some of it can be found in our kitchen [17]. There are two major sources of natural polymers which are from food waste and plant-based such as proteins (gelatin, zein, soy protein isolates etc.) and polysaccharides (chitosan, cellulose, pectin, carrageenan, dextran, starch, agar, etc.), respectively. Methylcellulose is one example of cellulose derivative which is extracted from methylation of alkali cellulose. Methylcellulose has good film forming properties, good solubility in water as well as good electrical and mechanical properties [18]. Starch is another great abundance and cheap natural polymer. The structure of starch has linear amylose (poly- α -1,4- D-glucopyranoside) and branched amylopectin (poly- α -1,4-D-glucopyranoside and α -1,6-D-glucopyranoside) [19].

Polymer blending is an effective method to gain desired combination of electrical, thermal and structural properties [20]. Both methylcellulose and potato starch have electron lone pairs in their oxygen-containing functional groups. These electron lone pairs are very crucial for ionic transportation as it form dative bond with each other [21]. In order to make the methylcellulose-potato film to be conductive, it must be doped with ionic sources. Nowadays, protonic (H^+) based polymer electrolyte has gained more attention to replace lithium. H^+ can be obtained from inorganic acids such as sulfuric acid (H_2SO_4) and phosphoric acid (H_3PO_4). Nevertheless, these inorganic acids cause chemical degradation that can leads to incompatibility with practical applications. Another great source of H^+ is ammonium salt. Ammonium iodide (NH_4I) are chosen in this work due to its low lattice energy, U of 637 kJ/mol [22]. Other ammonium salts also can be used e.g. ammonium bromide (NH_4Br , $U = 682.0$ kJ/mol), ammonium acetate ($NH_4CH_3CO_2$, $U = 703.1$ kJ/mol),

ammonium fluoride (NH_4F , $U = 834.0$ kJ/mol), ammonium sulfate ($(NH_4)_2SO_4$, $U = 1754.7$ kJ/mol) and ammonium phosphate ($(NH_4)_3PO_4$, $U = 3334.0$ kJ/mol).

In this work, methylcellulose-potato starch blend, NH_4I and glycerol are chosen as the polymer host, ion provider and plasticizing agent, respectively. The polymer electrolyte will be used in the preparation of the EDLC. The electrodes of the EDLC are prepared with microbial cellulose as the binder and CNT as the active material for charge double-layer process. Past EDLC has metal foil and non-biodegradable binder e.g. PVdF in their electrodes. Our EDLC eliminates both metal foil and binder with unique structure of microbial cellulose.

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Experimental

Materials/chemicals details

Potato starch, glycerol and ammonium iodide were purchased from SYSTERM. Methylcellulose, carbon nanotubes and sodium dodecyl sulfate were procured from SYSTERM while acetic acid was from Friedemann Schmidt. Source of cellulose is a type of microbial cellulose (BC) hydrogel obtained from local market (Malaysia).

Material synthesis

Preparation of polymer electrolytes

An amount of 0.4 g of potato starch was dissolved in 100 mL of 1% acetic acid at 80 °C. Methylcellulose (0.6 g) was poured into the potato starch solution that was cooled down to room temperature and stirred until a homogeneous

solution was obtained. Next, 0.43 g of ammonium iodide (NH₄I) was included into the potato starch-methylcellulose solution. To plasticize the film, 0.29 g of glycerol was added. The solution was casted in Petri dishes and left to dry in a drying cabinet at room temperature.

Preparation of microbial cellulose-MWCNT electrodes

A total of 3 g of microbial cellulose was filtered using manual filtration. Then, an amount of 0.05 mg of hydroxyl multiwalled carbon nanotube (MWCNT-OH) was stirred in 20 mL of deionized water. Microbial cellulose and MWCNT-OH solution were then mixed together with deionized water for 1 day at 45°C. The homogenous solution was vacuum-filtered through a Buchner funnel. Lastly, the microbial cellulose-MWCNT electrodes were stripped off and dried at room temperature. More information regarding this electrode was described in our previous report [23].

Characterizations

The ionic conductivity (σ) of the polymer electrolyte was studied using HIOKI 3532–50 LCR. The thickness of potato starch-methylcellulose-NH₄I-glycerol film, x , was measured using Mitutoyo micrometer screw gauge. Impedance measurement was carried out in a frequency range from 50 Hz to 5MHz. The polymer electrolyte film was placed in a Teflon holder with two stainless steel electrodes. The σ was calculated using the following equation:

$$\sigma = \frac{x}{AR_b} \quad (1)$$

where R_b stands for the bulk resistance of the electrolyte. The transference number analysis (TNM) was performed using V&A instrument DP3003 digital DC power supply that was connected to a multimeter and to the polymer electrolyte film. The polymer electrolyte film was subjected to a working voltage of 0.2 V. TNM of ions (t_{ionic}) and electrons (t_{elec}) were calculated using the given equations:

$$t_{ionic} = \frac{I_b - I_c}{I_b} \quad (2)$$

$$1 - t_{ionic} = t_{elec} \quad (3)$$

where the beginning and constant current were set as I_b and I_c , respectively. The electrochemical steadiness of the polymer electrolyte film was determined using Digi-IVY DY2300 potentiostat via linear sweep voltammetry (LSV) measurement. The polymer electrolyte film was subjected to a linear voltage up to 3 V at a sweeping rate of 1 mV/s.

Device fabrications and measurements

Microbial cellulose-MWCNT electrodes were cut into an area of 2.01 cm². The glycerolized polymer electrolyte was placed in between two identical microbial cellulose-MWCNT electrodes to form a simple EDLC. The EDLC was packed in CR2032 coin cell which then pressed using a battery clasper.

Characterization of the EDLC

Cyclic voltammetry analysis was employed to verify the capacitive behaviour of Microbial cellulose-MWCNT based EDLC. The coin cell was packed in a Teflon holder and connected to a potentiostat (Digi-IVY DY2300). The sweeping rate of the CV analysis was from 10 to 100 mV/s. The specific capacitance (C_{cv}) of the constructed EDLC was calculated using following equation:

$$C_{cv} = \int_{V_i}^{V_f} \frac{I(V)dV}{2(V_f - V_i)mx} \quad (4)$$

where V_f and V_i are the last and beginning voltages, respectively. $I(V)$ stands for the area of CV plot which was extracted using OriginPro 9.0 software. The mass of MWCNT is set as m while x is the sweeping rate. The charge-discharge measurements were analyzed with the current density of 0.75 mA cm⁻² using a battery cyler (NEWARE). Important parameters e.g. specific capacitance (C_{cd}), internal resistance (ESR), power density (P_{den}), energy density (E_{den}) and efficiency were obtained via these following equations:

$$C_{cd} = \frac{i}{ym} \quad (5)$$

$$ESR = \frac{V_d}{i} \quad (6)$$

$$E_{den} = \frac{C_{cd}V^2}{2} \quad (7)$$

$$P_{den} = \frac{V^2}{4mESR} \quad (8)$$

$$Efficiency = \frac{t_d}{t_c} \times 100\% \quad (9)$$

where y is the gradient of discharge curve, V_d is the potential drop and i is the applied current (1.5 mA).

Results and discussion

Conductivity analysis

Ionic conductivity, σ is the heart of polymer electrolyte study. It is very crucial for an electrolyte to have a high value of σ , for it to be useful enough for energy storage

application. **Table 1** shows the room temperature σ of the polymer electrolyte system. The film with potato starch and methylcellulose possesses a very low ionic conductivity of 10^{-11} S/cm. The low value of σ is due to the low ionic density in the polymer matrix. There is a small amount of H^+ in potato starch-methylcellulose film due to the presence of 1 wt.% acetic acid (CH_3COOH). The inclusion of 30 wt.% NH_4I increases the conductivity up to six orders. One of the hydrogen in NH_4^+ is loosely bound and can be used for ionic conduction in the presence of electric field [24]. The σ is optimized at $(1.47 \pm 0.10) \times 10^{-3}$ S/cm with the existent of plasticizer, which in this case glycerol. There are several reasons why addition of glycerol improves the ionic conductivity: i) due to its high dielectric constant (46.5), more ions can be dissociated into mobile ions [25] and ii) molecules of glycerol are smaller than polymer molecules. This situation enables the plasticizer molecules to travels in between polymer chains which reduces the cohesive forces that holds the polymer structure, resulting in easier ionic conduction [26, 27].

Table 1. Polymer electrolyte with their respectively ionic conductivity at room temperature

Polymer Electrolyte	σ (S/cm)
potato starch-methylcellulose	$(2.55 \pm 2.10) \times 10^{-11}$
potato starch-methylcellulose- NH_4I	$(6.72 \pm 1.25) \times 10^{-5}$
potato starch-methylcellulose- NH_4I -glycerol	$(1.47 \pm 0.10) \times 10^{-3}$

Transference number analysis

Common energy storage mechanism of an EDLC is via charge double-layer. Thus it requires a polymer electrolyte with high t_{ionic} for an efficient polarization process. TNM measurement is performed with a working voltage of 0.2 V at room temperature. **Fig. 1a** depicted the polarization plot of potato starch-methylcellulose- NH_4I -glycerol electrolyte. A high current of $6.9 \mu A$ can be observed at the beginning of plot. During this stage, ions in the bulk of the electrolyte instantly migrate towards the stainless electrodes and started to form electrostatic force with the electrons in the electrode. After a while, the current reduces and become constants. The stable and low current value signify the polarization process is completed and only electrons flow into the electrode. Ions are verified as the main charge carrier as the calculated t_{ionic} and t_{elec} are 0.99 and 0.01, respectively. The outcome is comparable with other protonic-based study. A glycerolized polymer blend of starch and chitosan with NH_4Cl as the ion source is reported to achieve t_{ionic} ranging from 0.91 to 0.98 [28]. As reported by Vijaya et al. [29], the t_{ionic} for polyvinylpyrrolidone (PVP)- NH_4Cl based electrolytes is 0.93–0.97. NH_4Cl has

higher lattice energy than NH_4I which 708 kJ/mol. According to Yusof [30], NH_4I based polymer electrolyte with σ of $\sim 10^{-3}$ S cm^{-1} has t_{ionic} of 0.99.

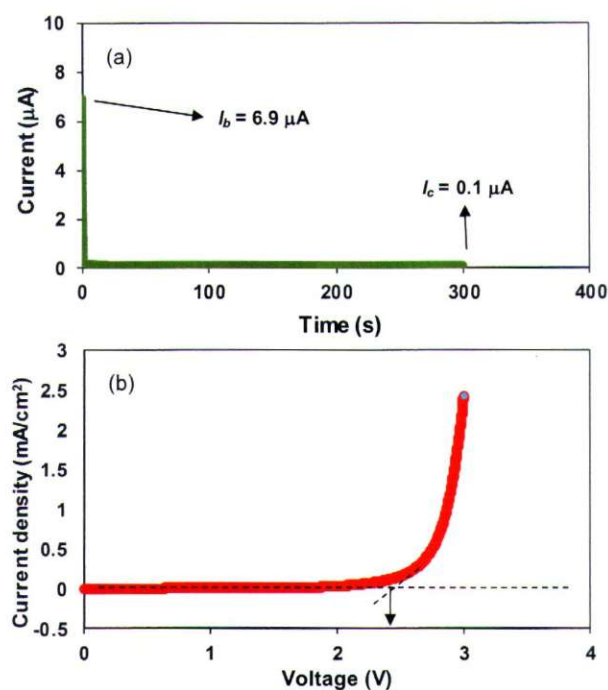


Fig. 1. (a) TNM plot of potato starch-methylcellulose- NH_4I -glycerol with working voltage of 0.2 V. (b) LSV plot of potato starch-methylcellulose- NH_4I -glycerol with scan rate of 1 mV/s.

Linear sweep voltammetry analysis

Apart from high σ and t_{ionic} values, a polymer electrolyte must be electrochemically stable in the potential range depending on the types of applications. The electrochemical stability of the polymer electrolyte can be determined using LSV measurement. In this analysis, the potential is swept linearly and the current response is monitored. **Fig. 1b** shows the LSV plot of potato starch-methylcellulose- NH_4I -glycerol electrolyte. When the electrolyte is subject to a voltage at a sweeping rate of 1 mV/s, it can be observed that there is absence in current response at potential below 2.0 V. The constant current is related with the capacitive current as ions-electrons double layer formed on the contact surface area between the electrodes and the electrolytes. The plot verified that the starch-methylcellulose- NH_4I -glycerol electrolyte has degraded after the voltage surpasses 2.4 V. The sharp increase in current beyond that threshold value is attributed to the oxidation of oxide and other functional groups in the polymer chains which causing instability in current flow. From this outcome, it is surmised that potato starch-methylcellulose- NH_4I -glycerol electrolyte can be used in

any suitable electrochemical application with voltage lower than 2.4 V.

Cyclic voltammetry measurement of the microbial cellulose-based EDLC

The verification of charge double layer in the fabricated EDLC with the cell arrangement of microbial cellulose-electrode | potato starch-methylcellulose-NH₄I-glycerol electrolyte | microbial cellulose-electrode is done via CV measurements.

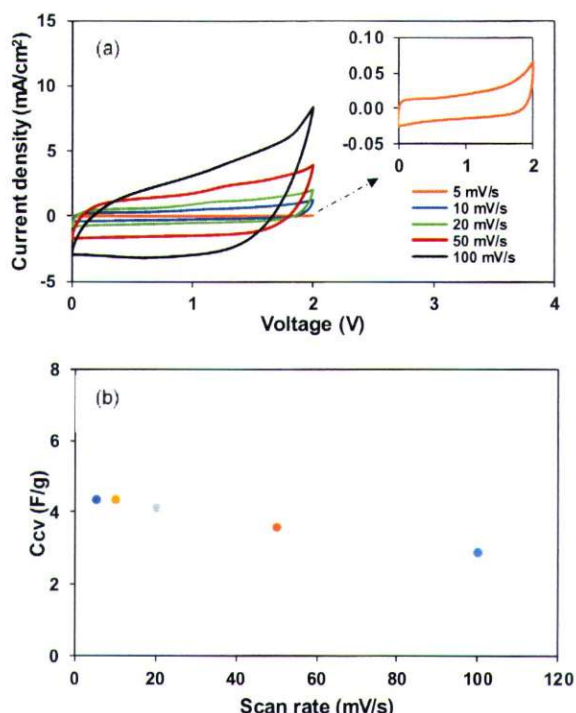


Fig. 2. (a) CV curves the constructed EDLC at various scan rates and (b) The influence of scan rate on C_{cv} value.

A capacitor is a scan rate dependent devices hence different scan rates are applied to the EDLC ranging from 10 to 100 mV/s. The form of CV plot in **Fig. 2a** varies from rectangular and leaf-like shapes with absence of large redox peaks. According to Jäckel et al. [31] a peak in CV curve indicates the existent of Faradaic current corresponds to the intercalation/deintercalation mechanism which does not occur in a double-layer capacitor. It is noticeable that the form of CV plot at high scan rate is a leaf-like shape rather than a perfect rectangular. At high scan rate, the conduction of mobile ions occurs at rapid rate causing the recombination of ions or improve buildup of charge double-layer. Furthermore, this is due to the internal resistance as well as the porosity of the carbon which creates current dependence of voltage [32]. This explains the low value of C_{cv} (2.9 F/g) in **Fig. 2b**, when 100 mV/s is applied. A high value of C_{cv} (4.4 F/g) is obtained when 5 mV/s is used.

During slow scan rate, nearly perfect plateau region is observed in the CV plot of 5 mV/s which signifies that the free ions moves at a constant rate and hence developing the accumulation of ions at the electrode's surface with low ohmic resistance [33, 34].

Charge-discharge measurement of the microbial cellulose-based EDLC

Fig. 3 shows the charge-discharge curve of the constructed EDLC at selected cycles. The EDLC is charged up to 2 V and discharged down to 0 V. A typical capacitor will show a triangle plot due to electrostatic force between ions of the electrolyte and electrons of the electrodes. Unlike conventional batteries where the discharge curve is non-linear due to the process of inter- and de-intercalation.

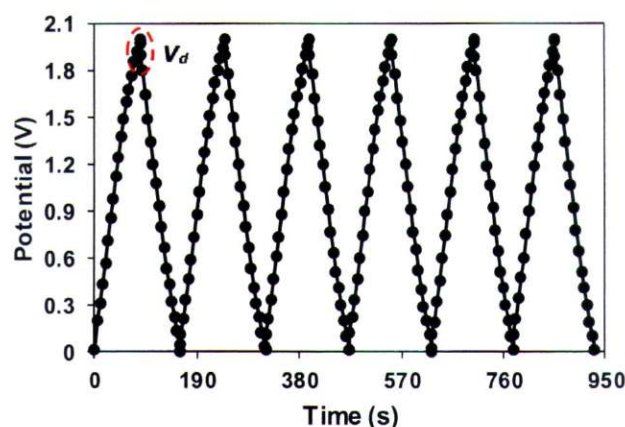


Fig. 3. Charge-discharge plot of the fabricated microbial cellulose-based EDLC at selected cycles.

Fig. 4 illustrates the C_{cd} and efficiency as a function of cycle number for the constructed microbial cellulose-based EDLC at 0.75 mA/cm². The C_{cd} at the 1st cycle is found to be 3.7 F/g where this value is harmonized with the one in CV analysis. C_{cd} value decreases to 3.3 F/g at the 50th cycle, followed by almost consistent value after the 100th cycle upwards and achieved an average value of 3.0 F/g as 1000 cycles are completed. The decrement of C_{cd} at the beginning stage could be due to several pores of the electrodes could be permanently congested or blocked after several charge-discharge processes or formation of ion aggregates and pairs [35].

The large value of efficiency indicates that the discharging part is almost similar to or longer than the charging time. The efficiency of 88.4% is noticeable at the 1st cycle. Usually, the charging process lasts longer in comparison with discharging at the 1st cycle before stabilization can be obtained. By applying the current, anions and cations start to approach the electrode and become familiar with the conduction pattern. The efficiency is varied in the range of 90–97% beyond 100th cycle. This outcome of the current

work discloses that the EDLC has excellent cycling stability.

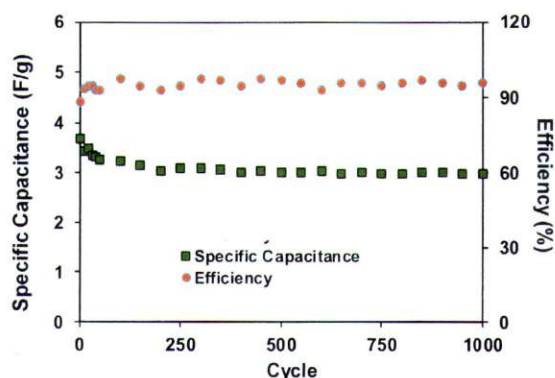


Fig. 4 C_{cv} and efficiency values of the microbial cellulose-based EDLC throughout 1000 charge-discharge cycles.

The equivalent series resistance (*ESR*) is another important parameter need to be analyzed to understand the electrolyte-electrodes contact along with internal resistance of the EDLC. As seen in **Fig. 3**, there is a small voltage drop (V_d) prior discharging process. The average of voltage drops of the EDLC ~ 0.0997 V. **Fig.5** shows the *ESR* values of the EDLC throughout the 1000 complete charge-discharge cycles. The average *ESR* value is 66 Ohm. The existent of *ESR* is attributed the components of the EDLC e.g. polymer electrolyte and the space between electrodes and the electrolyte [36]. Small values of *ESR* verify that microbial cellulose-based EDLC has good contact between the electrodes and electrolytes, evidencing the facility with which charge carrier conduction towards the surface of the electrodes can take place, therefore creating an electrical double-layer for via electrostatic force [37].

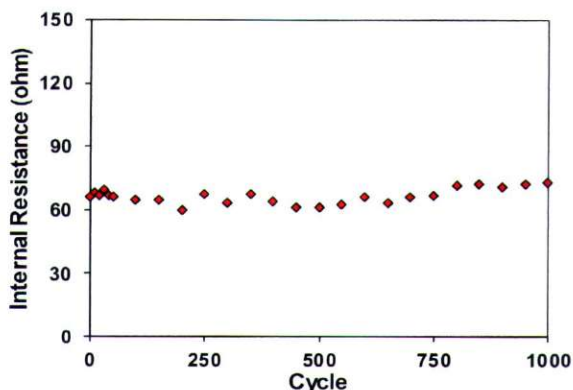


Fig. 5. *ESR* values of the microbial cellulose-based EDLC throughout 1000 charge-discharge cycles.

Fig. 6 depicts the variation of E_{den} and P_{den} over 1000 cycles. The stored energy for the 1st cycle is calculated to be 1.84 Wh/kg and reduced to 1.61 Wh/kg at 100th cycle. As the cycle number exceeded 100 cycles, E_{den} is clearly started to stabilize at an average value of 1.52 Wh/kg throughout the 1000 charge-discharge cycles. The stabilization of E_{den} signifies that NH_4^+ or H^+ and I^- in the polymer matrix of potato starch-methylcellulose blend travel toward the electrodes with almost similar energy barrier [38]. According to Winie et al. [39], a biopolymer-based EDLC has energy density from 0.57 to 2.8 Wh/kg as the current density changes from 2 to 0.6 mA/cm², respectively. The stability in energy density and capacitance trends indicate the low occurrence of ionic aggregation. By referring to Equation (8), P_{den} of the EDLC is highly correlated with *ESR*. As the charge transfer is related to the gap between electrolyte and electrodes [40], P_{den} value at the 1st cycle is found to be 226 W/kg and fluctuated in a small range. The average P_{den} of the EDLC is 225 W/kg.

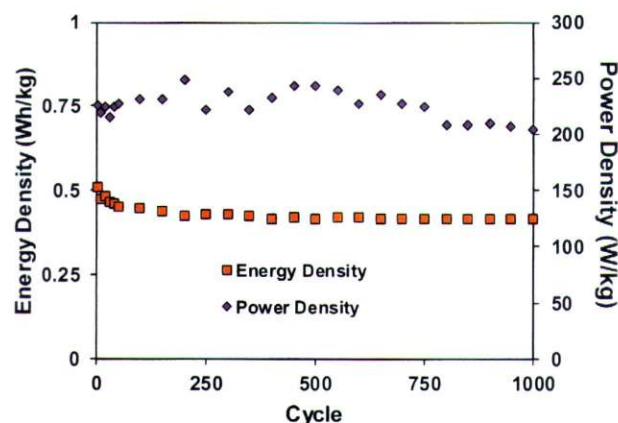


Fig. 6. Energy and power density values of the microbial cellulose-based EDLC throughout 1000 charge-discharge cycles.

Conclusion

A binderless, metalless and biodegradable electrode for EDLC based on microbial cellulose-multiwalled carbon nanotube (MWCNT) was successfully synthesized via harmless, inexpensive and straightforward method. This to ensure the EDLC follows the guideline in “Sustainable Development Goals”. The microbial cellulose-MWCNT electrodes were used to sandwich a green polymer electrolyte with potato starch and methylcellulose as the polymer host. Ammonium Iodide and glycerol were employed as ion provider and plasticization agent, respectively. The prepared potato starch-methylcellulose- NH_4I -glycerol has high ionic conductivity, transference number and oxidation voltage of $(1.47 \pm 0.10) \times 10^{-3}$ S/cm, 0.99 and 2.54 V, respectively, making it beneficial for the

EDLC application. From CV measurement, no obvious redox peaks were observed thus indicated the capacitive behavior of the constructed EDLC. The EDLC was found to be scan rate dependent where from CV, the specific capacitance achieved was from 4.4 to 3.0 F/g as the EDLC was swept with 5 to 100 mV/s. The charge-discharge measurement revealed that the microbial cellulose-based EDLC has specific capacitance, ESR, energy density and power density of 3.7 F/g, 66 Ohm, 1.61 Wh/kg and 226 W/kg, respectively. The EDLC showed a high cycling ability for 1000 complete charge discharge cycles. The performance of the EDLC can be further enhanced by improving the electrodes as well as the electrolyte, such as the inclusions of nano-filler, polymer grafting, using different types for carbon, salt and polymers. The main key is to obtained a really tight contact between the electrodes and the electrolytes. The total performance of the microbial-based EDLC has proven that it is appropriate for low voltage applications.

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Conflicts of interest

There are no conflicts to declare.

Keywords

Microbial Cellulose, Bacterial Cellulose, EDLC, Green Energy Storage, Biodegradable

Supporting information

Supporting informations are available online at journal website.

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(a) Scientific article

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Graphical Abstract

