

Contents lists available at ScienceDirect

Energy Conversion and Management





Research Paper Coal-based graphitized activated carbon for solar energy powered supercapacitor IoT applications

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ARTICLE INFO

Keywords: Low-grade coal Graphitized activated carbon Electrode material Supercapacitor Solar cell IoT

ABSTRACT

Solar cells show promise as energy conversion gadgets, but their extended use is limited by intermittent sunlight. Self-powered solar cell integration with an electrical energy storage system could be one solution to this problem. Therefore, we have developed a coin cell supercapacitor (SC) device based on coal-based graphitized carbon materials and integrated with solar cells to assess its efficiency for energy transformation and storage for practical validity. Supercapacitor carbon electrode was developed from a naturally abundant carbon precursor i. e. low-grade coal using molasses as binder, for the fabrication of high-quality SC devices. The fabricated supercapacitor exhibits a notable 127 F g^{-1} specific capacitance, maintaining 92 % of its initial value for up to 10,000 charge/discharge cycles at 5 A g^{-1} , and a maximum energy density of 70.57 Wh k g^{-1} and power density of 2000.17 W kg⁻¹. In order to create a self-sustaining power pack, the SCs (4 V) are integrated with a commercial solar cell (6 V, 750 mAh). Importantly, in the presence of natural sunlight, the solar-powered as-fabricated supercapacitor bank can significantly power a commercial IoT module (4-6 V, 600 mAh) demonstrating its potential for successful solar-to-electric energy transformation and storage. The findings demonstrate that solarpowered coal-based SC can be explored as an incredibly rapid future-proof energy storage technology that can help reduce energy demand in the foreseeable future.

1. Introduction

Electrochemical energy storage devices have become crucial in today's energy-dependent universe to address the fast-depleting fossil fuel supply. Photovoltaic (PV) solar cell devices have been researched and commercialized extensively for their ability to harvest solar energy due to their optimal power conversion efficiency, cost-effective, and customizable design [1-4]. Conversely, the short-term variability of solar radiation causes fluctuations in the electrical output from photovoltaics, making it impractical to use that method for extended applications [1]. An alternative to address this issue could be the use of integrated photovoltaic solar cells and supercapacitor devices. There are very few articles that thoroughly investigate the global research on solar energypowered supercapacitors for Internet of Things (IoT) applications [5,6]. In a study, the integration of solar cells with supercapacitor (SC) devices was performed and evaluated their performance for energy conversion and storage to assess practical viability [1]. Narayanan et al. [7]

introduced a solar-powered supercapacitor combining a plasmonic quantum dot solar cell (ODSC) and an MWCNT supercapacitor, offering a cost-effective solution for future power sources. Most recently, an investigation of micro-supercapacitors (MSCs) utilizing graphene nanowalls (GNWs) was reported [8]. Their study encompasses the synthesis of materials, device fabrication, and demonstrations of Internet of Things (IoT) applications. It has also been observed that integrating solar cells with an electrical energy storage unit not only enables the storage of solar energy but also reduces fluctuations in the output power caused by variations in solar irradiation. On the other hand, the development of supercapacitors has drawn the attention of many researchers and has been in the spotlight due to their durable cycling life with slight changes in performance when compared to batteries. Supercapacitors have high power density, wide-ranging temperature, and voltage compatibility, minimal internal resistance, high amperage capability, fast charge-discharge, maximum efficiency, flexible packaging, lightweight, and environment-friendly characteristics, positioning them as

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https://doi.org/10.1016/j.enconman.2024.118964

Received 9 April 2024; Received in revised form 6 August 2024; Accepted 19 August 2024 Available online 24 August 2024 0196-8904/© 2024 Elsevier Ltd. All rights are reserved, including those for text and data mining, AI training, and similar technologies. potential frontrunners for the most efficient energy storage devices in the future, suitable for both commercial and residential applications [9–11]. It is therefore used to replace batteries in a variety of applications, including electronic goods, transportation, power backup, grid balancing, IoT, and heavy-duty applications [12,13]. It is also used in combination with secondary batteries to provide the additional power required in these applications [12]. However, it is to be mentioned that the upgradation and optimization of supercapacitors are heavily contingent upon the selection of appropriate electrode materials, electrolyte mediums, current collectors, binders, and separators [11].

The advancement of supercapacitors is greatly impacted by the interaction between the electrical double layer (EDL) and the surface area, texture, dimensions, and morphology of carbon electrode materials [14]. This has unveiled new avenues for discovering and developing advanced materials for high-performance energy storage systems (EES). Advanced materials such as activated carbon (AC), reduced graphene oxide (rGO), fullerene, carbon nanotubes (CNTs), porous carbon (PC), biochar, carbon aerogel, metal-organic frameworks (MOFs) and their derivatives [15–17], and CoS₂ nanoneedles (Cu-CoS₂ NNs) [18], exhibit better performance as electrodes in energy storage systems (EES). Their better performance is attributed to their large specific surface areas (SSA), cost-effectiveness, excellent electrical conductivity, and mechanical properties, making them highly promising for energy storage as well as solar energy-powered supercapacitor IoT applications [14,19–21]. Although high-performance, carbon materials like carbon nanotubes and graphene offer exceptional capabilities, their synthesis process is limited by cost and environmental concerns. Thus, developing affordable, eco-friendly porous activated carbon from low-cost, sustainable, and abundant precursors is crucial. In recent decades, significant research has been dedicated to optimizing precursor materials for the synthesis of advanced porous activated carbon [10]. Coal feedstock has emerged as an especially promising precursor due to its ability to generate tailor-made activated carbon suitable for energy applications [11]. Currently, numerous studies investigated the synthesis of activated carbon from various ranks of coal, including lignite, bituminous, and anthracite, using a range of activation techniques [21,22]. Utilizing lowgrade subbituminous coal offers economic benefits due to its lower extraction costs and widespread availability. Furthermore, the smaller graphite-like crystalline domains inherent in low-grade subbituminous coal facilitate easier exfoliation compared to the larger crystallites present in high-grade coal [9,10,22]. There are a few such studies on coalderived carbon electrodes used in supercapacitor applications [10,21,23]. Wang et al. [23] developed a novel melt infiltration process to prepare highly porous activated carbon with a SSA of 1872 m² g⁻¹ from low-rank coal. This method resulted in a supercapacitor electrode material exhibiting a specific capacitance of 211 F g-¹, demonstrating its potential for high-performance supercapacitor applications. Guo et al. [24] developed hierarchical porous carbon spheres from coal using a simple one-pot spray pyrolysis method. The resulting electrode materials, with a SSA of 948.55 m²g⁻¹, demonstrated good performance in supercapacitors, achieving a specific capacitance of 227 F g⁻¹. While the above-mentioned studies have demonstrated that the development of activated carbons (ACs) is critical for high-performance EDLCs, it is equally important to harness the abundant low-grade coal feedstock through innovative strategies. Such approaches are crucial for addressing our significant energy needs effectively and efficiently.

It is necessary to take into account aspects like safety, compatibility between various components, and cost-effectiveness in addition to electrochemical performance [11]. The energy-storage efficiency of supercapacitors depends on the binder, an inactive component within the electrode paste that facilitates the binding of conductive additives and activated material together [9]. The binder serves as a cohesive agent, binding the slurry to the substrate (current collector), and ensuring that the activated materials do not detach during the operation of the electrodes [9]. The development of a greener process for electrode manufacturing has been proposed, with the avoidance of organic solvents and their replacement by water as the solvent medium, utilizing aqueous-based electrolytes [25,26]. A lot of efforts have been implemented to explore alternative water-based binders and enhance electrochemical performance [26,27]. Considering recent proposals for alternatives, carboxymethyl cellulose (CMC) stands out as the forefront choice for aqueous, non-fluorinated binders in electrochemical double-layer capacitors (EDLC) [28-30]. Even though this material satisfies the requirements outlined above, it has the major drawback of relatively lower thickness of coatings (or mass loading). The reason behind the issue is the contraction of CMC coatings during the drying phase, which may lead to cracks in the coating. Additionally, cracking occurs when rolling after drying, for example, due to the coating's low flexibility at high mass loadings [25]. The system based on Styrene-Butadiene Rubber (SBR) and CMC is the most studied water-based binary binder combination [1,25,26]. Also, the efficiency and reliability of supercapacitors are impacted by the interactions between electrolytes and both active and inactive components like solvent binders and current collectors [31].

In light of the above, the present research work intends to introduce the integration of fabricated coal-based supercapacitors (coin cell configuration) with photovoltaic (PV) solar cells for using in the Internet of Things (IoT) modules/devices of commercial/industrial applications. Also, the biodegradable molasses was efficiently utilized as a novel binder through a modified chemical activation method to create graphitized activated carbon from abundant low-grade coal feedstock, which is then utilized to fabricate high-performance symmetric supercapacitor coin cells. Additionally, our objective is also to develop a green energy storage device i.e. supercapacitor by using low-cost neutral electrolytes with stable long-term performance. Therefore, in this work, we compare the electrochemical properties of coal-derived graphitized activated carbon with molasses, a novel biodegradable water-soluble binder, and carboxymethyl cellulose-styrene-butadiene rubber (CMC-SBR), a commercial binder with neutral electrolytes (i.e., 1 M Na₂SO₄ and 1 M Li₂SO₄), and three different Aluminium current collectors in two-electrode system (SwagelokTM cell configuration). Furthermore, by combining our manufactured coin cell supercapacitor with a commercial PV solar cell, we demonstrate an applicable strategy for integrating a self-sustaining novel coal-based supercapacitor power bank. This solar-powered supercapacitor can power an IoT module in the presence of natural sunlight, demonstrating its potential for efficiently and continuously converting solar energy into electrical energy storage.

2. Materials and methods

2.1. Raw materials

For this work, low-grade (subbituminous) coal feedstock was chosen as a carbon precursor for the synthesis of graphitized activated carbon. The raw coal sample was collected from the Meghalaya Coalfield, Northeast Region of India. Molasses (a waste generated from sugarcane or sugar beet processing) was collected from Jorhat locality (Northeast Region of India) for using as a novel binder in this study. The chemicals used in the current research work were: H2O2 (20-30 %, Merck), KOH (Merck), HCl (Rankem; LR), HF (Molychem), CMC (Sigma-Aldrich), SBR (MatLabs Technologies), Na2SO4 (HIMEDIA), Li2SO4 (Sigma Aldrich), C3H6O (Alfa Aesar), C2H5OH (98 %, Molychem), and distilled water for cleaning purposes in the experimental procedures. Commercial activated carbon (MatLabs Technologies) and Acetylene black (MatLabs Technologies) were used as electro-active material and additive, respectively in this study. Whatman glass microfiber filters (GF/A, 8*10 ins.) were used as separators for all the types of cells. Etched Aluminium foil (thickness: 30 µm), formed Aluminium foil (thickness: 30 µm), and Aluminium foil (thickness: 16 µm) purchased from MatLabs Technologies were used as current collectors in Swagelok[™] cell. For making supercapacitor coin cell devices, components of stainless-steel coin cell 2032-positive case (Model: CR2032E, material:

Compositions of the electroactive materials, binder, and additives to fabricate electrodes.

Sl. No.	Sample code	Electro-active materials	Binder	Additives
1	JCAC-CMC- Molasses	Coal-derived AC (JCAC) : 89 wt%	CMC (2 wt%) + Molasses (4 wt%)	Acetylene black (5 wt%)
2	JCAC-CMC- SBR	Coal-derived AC (JCAC) : 89 wt%	CMC (2 wt%) + SBR (4 wt%)	Acetylene black (5 wt%)
3	MtAC-CMC- Molasses	Commercial AC (MtAC) : 89 wt%	CMC (2 wt%) + Molasses (4 wt%)	Acetylene black (5 wt%)
4	MtAC-CMC- SBR	Commercial AC (MtAC) : 89 wt%	CMC (2 wt%) + SBR (4 wt%)	Acetylene black (5 wt%)

316#), negative case (Model: CR2032 negative, material: 316#), spacers (Gasket-316#, F15.8 \times 1.0 mm), and springs (Model: Three wheel, Sharpnel-316#) were used. PV solar cell (6 V, 750 mAh) and IoT module NodeMCU (4 V-6 V, 600 mAh) were purchased from Amazon India.

2.2. Chemical modification of low-grade coal into graphitized activated carbon

Graphitized activated carbons (GAC) were synthesised from the subbituminous coal samples using ultrasonic-aided oxidation and a subsequent thermochemical activation method. In brief, 100 g of raw coal (JCR) (sizes $\leq 210 \,\mu$ m) was oxidized using an environment-friendly hydrogen peroxide solution (20–30 % H₂O₂). The oxidized coal was then subjected to thermochemical activation with potassium hydroxide

(KOH) as the activating agent, with a weight ratio of oxidized coal to KOH of 1:2. The detailed synthesis methods of graphitized ACs were reported in our previous studies [10,11,14]. The raw coal and synthesized GAC are denoted as JCR and JCAC, respectively in the subsequent sections.

2.3. Mechanism of modified KOH activation method

In this study, coal sample was chemically activated using a KOH solution in presence of ultrasonic energy. Therefore, during the ultrasonic-assisted activation, KOH molecules interact with active sites on the coal surface, facilitating the formation of K^+ ions and the development of ordered and disordered layers that penetrate the carbon pores. The reaction mechanism involves: (i) cavitation and mechanical forces that create partial cracks and improve pore connectivity while reducing surface impurities and (ii) the ultrasonic capillary effect (UCE) that enhances liquid penetration into pores, increasing ion diffusion and surface modification of the produced activated carbon [10,11,14]. The details of the steps of the mechanism of graphitized ACs formation was reported in our previous studies [10,11,14].

2.4. Fabrication of carbon electrode in SwagelokTM cell and coin cell assembly

The electro-active materials i.e. the activated carbon samples (89 wt %) were mixed separately with CMC (2 wt%), SBR (4 wt%), and Molasses (4 wt%) using Acetylene black as an additive (5 wt%) in each composition and dissolved in water for homogeneous mixing to fabricate the coal-derived graphitic carbon electrode and commercial activated carbon (Matlabs) electrodes, respectively. The details of the compositions of each electroactive materials are shown in Table 1.

The schematic diagram of the fabrication process of the SwagelokTM



Fig. 1. Schematic diagram showing the electrode compositions using coal-derived/commercial activated carbon samples for fabrication of Swagelok[™] cell and coin cell assemblies.



Fig. 2. (a) XRD spectra of raw coal (JRC) and coal-derived graphitic porous AC (JCAC) showing the presence of gypsum (G) and quartz (Q); and the turbostratic graphitic peaks at *002* and *100* planes; (b) Raman spectra of raw coal (JRC) and coal-derived graphitic porous AC (JCAC) showing the formation of D and G bands in graphitic porous carbon after activation; (c) FTIR spectra of raw coal (JRC) and synthesized AC (JCAC) demonstrating the occurrence of surface functional groups (-OH, -CH, -C=O, $-CH_3$, and -C-O-C); (d) XPS survey spectra synthesized AC (JCAC) indicating the presence of C_{1s} , O_{1s} , N_{1s} , and S_{2p} valence state; (e) deconvoluted C_{1s} spectra of JCAC revealing the presence of chemical bonding (C=O, C-OH, C-O-C, C=C); (f) deconvoluted O_{1s} spectra of JCAC revealing the occurrence of chemical bonding (C=O).

cell and coin cell assemblies using coal-based graphitic activated carbon electrodes and commercial activated carbon is depicted in Fig. 1. The processes of fabricating electrodes for SwagelokTM cell and coin cell assemblies are described in the supplementary materials (SM).

2.5. Analytical characterization and electrochemical analysis

The detailed analytical characterization techniques (proximate and ultimate analysis, FTIR, XRD, XPS, SEM, TEM, and Raman) and electrochemical methods used in the study were performed following our previous works [10,11,32–34].

3. Result and discussion

3.1. Physicochemical properties

3.1.1. Structural parameters of the raw coal, electroactive, and binder materials

The amorphous/crystallinity of the carbon structure present in the raw coal (JRC) and as-synthesized AC (JCAC) was analyzed by XRD, and as seen in Fig. 2a, the raw coal sample contain the minerals, including quartz (SiO₂) and gypsum (CaSO₄·2H₂O) along with amorphous constituents [35].

The XRD peak analysis of raw coal clearly shows the presence of an amorphous structure, as indicated by a broad protrusion in the $5-35^{\circ}$ range, with no distinct peak [10,32]. However, the coal-derived AC (i.e., JCAC) exhibits distinct peaks at 26.13° and 43.60° associated with the diffraction of the turbostratic carbon layer structure with (*002*) and (*100*) planes [10,11,36]. Thus, the synthesised AC has a graphite-like

Physicochemical parameters of raw coal, synthesized graphitic porous AC, and molasses binder sample (wt. %).

Sl. No.	Samples	Sample codes	Proximate	Proximate analysis (%)			Elemental	Elemental analysis (%)		
			М	VM	Ash	FC	С	н	0	TS
1	Raw coal	JRC	11.98	42.01	2.78	43.23	67.16	7.24	22.63	2.97
2	Coal-derived AC	JCAC	30.81	10.29	2.76	56.14	87.4	1.88	9.93	0.79
3	Molasses binder	MB	2.63	77.24	2.15	17.98	39.68	6.72	53.47	0.13

(M=moisture content; VM=volatile matter content; Ash = ash content; FC=fixed carbon content; C=Carbon content, H=Hydrogen content, TS=total sulfur, O=Oxygen content: obtained by difference).

structure, demonstrating the electrode material appropriate for supercapacitor efficiency [10,36–38].

The data from elemental and proximate analysis, presented in Table 2, provide additional support for the findings of the XRD analysis. Coal-derived activated carbon (JCAC) has significantly lower volatile matter, ash content, sulphur content, hydrogen content, and oxygen content than the raw coal samples. The fixed carbon (FC) content in activated carbons (ACs) is higher than in the corresponding raw coals due to lower volatile matter (VM) and moisture (M). Sulfur content is significantly reduced after oxidation and activation, as shown by total sulfur analysis (Table 2). The H_2O_2 oxidation method converts coal sulfides into water-soluble sulfates, which are removed during filtration, further decreasing sulfur content [11]. Through the vibration and cavitation mechanism during ultrasonic-assisted treatment, this process effectively reduces the ash contents (mineral matter) from raw coal (JCR) [10.39]. Organic residues containing ash and volatile matter were removed during thermal treatment and acid treatment, resulting in the production of activated carbon. Furthermore, both proximate and elemental data also confirm that the JCAC contains significantly more carbon than the corresponding raw coal [10,11].

In order to extract more graphitic structural information, the Raman spectroscopic data of raw coal (JRC) and synthesized AC (JCAC) were performed (see Fig. 2b). Because of the amorphous nature of the raw coal, it has a weak and broad D absorption band at approximately 1360 cm⁻¹. Conversely, the raw coal also displays a weak G absorption band at around 1622 cm⁻¹, signifying the occurrence of several surface functional groups containing oxygen [10,40]. But, the graphitic porous carbon (JCAC) exhibits two well-defined peaks, with the D band occurring at approximately 1351 cm⁻¹ and the G band at around 1610 cm⁻¹. These peaks are closely related to the turbostratic structure (sp³C/C–C) and graphite crystallite (sp²C/C=C), respectively [10,37,41].

The basic chemistry of the indigenous molasses sample is presented in Table 2. The molasses sample has significantly lower content of moisture, ash, hydrogen, and sulfur, which is conducive to good electrode adhesion. On the other hand, the high carbon and oxygen contents in the molasses sample indicate the presence of a high oxygencontaining functional group, which acts as a potential binder for electrode fabrication in our process [44]. Also, the viscosity of the binders is enhanced by the reduction of water contents in molasses, which raises the resistance to heat and the surface tension among molecules [44]. As a result, the lower moisture content of molasses binder has a positive effect on electrode formulation and calendering.

3.1.1.1. Functionality and chemical compositions of the raw coal, electroactive, and binder materials. The surface functional groups present in the raw coal and synthesized graphitic porous AC material were evaluated qualitatively using FTIR spectroscopic techniques [11,44–46]. The major surface functional groups observed in both the samples (JCR and JCAC) are illustrated in Fig. 2c. The broad spectrum of absorption viewed at 3445 cm⁻¹ (JCR) and 3426 cm⁻¹ (JCAC) is assigned to the alcoholic/phenolic (O–H) stretching (intermolecular) mode. The C–H stretching mode is responsible for the absorption peaks at 2927 cm⁻¹ (JRC) and 2924 cm⁻¹ (JCAC). The absorption bands at 1626 cm⁻¹ (JRC) and 1608 cm⁻¹ (JCAC) are attributed to the vibrational stretching mode of carboxylic (C=O) groups. In addition, the CH₃ symmetric bending absorption peaks are observed at 1399 cm⁻¹ (JRC) and 1386 cm⁻¹ (JCAC). The absorption bands at 1120 cm⁻¹ (JRC) and 1117 cm⁻¹ (JCAC) are ascribed to the stretching of carboxylic C–O in the C–O–C group [10,14]. During the oxidation process, H₂O₂ breakdown the covalent bonds (e.g., Ar–OH, Ar–O–Ar), facilitates carbon substitution, and generates oxygen-containing functional groups such as –COOH, C–O–C, and R–OH in low-grade coal chemical structure [11,47]. This results in the creation of oxygen-rich functional groups in the synthesized activated carbon.

Thus, the extensive FTIR analysis of the AC derived from coal revealed the development of significant surface functional groups containing oxygen and sp^2 (aromatic) structural units during the subsequent processes of oxidation and ultrasonic-assisted KOH activation. This is to be mentioned that the occurrence of oxygen-containing surface functional groups significantly impacts the wettability of the electroactive material, thereby contributing to the favorable electrochemical properties of the supercapacitor.

XPS spectroscopy was employed to validate the occurrence of functional groups and elucidate their chemical bonding properties, with the outcome displayed in Fig. 2def. The XPS survey spectrum (Fig. 2d) revealed the strong peaks at 164, 285, 400, and 532 eV, which are assigned to the S_{2p}, C_{1s}, N_{1s}, and O_{1s} valence states, respectively. For a more comprehensive understanding of the chemical binding states within various surface functional groups, we also examined the deconvoluted high-resolution spectra of C_{1s} and O_{1s}. Four peaks at 284.6, 285.3, 286.7, 290.2, and 293.8 eV are found to be visible in the highresolution C_{1s} spectra (Fig. 2e) for the JCAC sample suggesting the existence of the chemical bonds C–C, C–O–C, C–OH, C=O, and C=O, respectively. Furthermore, the deconvoluted O_{1s} XPS spectra (Fig. 2f) of JCAC show that there are two primary peaks for its organic C=O at 532.1 eV and organic C-O at 534.1 eV [37,42,44,46].

Figure S1a provides the additional details regarding the molasses binder through FTIR spectroscopic analysis. The broad spectrum of absorption is attributed to the mode of intermolecular stretching; which includes interactions with alcoholic/phenolic (O–H) groups viewed at 3377 cm⁻¹. The absorption peak at 2931 cm⁻¹ is assigned to the C–H stretching mode. The absorption band at 1641 cm⁻¹ is indicative of the carboxylic (C=O) stretching mode of vibration. Additionally, the CH₃ symmetric bending absorption peaks are observed at 1427 cm⁻¹. The absorption bands at 1055 cm⁻¹ are due to carboxylic C–O stretching in the C–O–C group [43]. From FTIR analysis, the current molasses sample is found to have the highest content of surface functional groups and aromatic sp² moieties. The presence of oxygen-containing functional groups in molasses enhances its binding property that binds electroactive material to the current collector and results in good electrochemical performance.

The particle size distribution of the molasses sample (Figure S1b) was also analysed and found to consist of a smaller average particle size (d0.5 = 87.579 μ m) due to which it can be processed more easily to fabricate superior electrodes with uniform mass loading and smooth surfaces.

Thermogravimetric analysis (TGA) of molasses and styrenebutadiene rubber (SBR) is necessary for understanding the thermal properties of binder materials and degradation of the binder during drying that can impair the coating's integrity [25]. Molasses and SBR



Fig. 3. (a) Nitrogen adsorption–desorption isotherm curve of coal-derived AC (JCAC) demonstrating the standard Type-I isotherm graph with both microporous and mesoporous configurations, (b) BJH pore size distribution of JCAC, (c) DFT pore size distributions of JCAC, and (d) particle size distribution of JCAC and commercial AC (MtAC).

were heated separately, at 5 °C min⁻¹ in a TGA as shown in **Figure S1c**. Both binders were preheated at 80 °C for 1 h due to their substantial amounts of water absorption at ambient temperature [25,46]. As can be observed, molasses dehydrated insignificantly slower than SBR at 180 °C in **Figure S1c**. The electrodes would finish the drying stage within a short timeframe on an industrial production line [48]. Therefore, at the optimum temperatures for electrode formation in the air, molasses was relatively stable than SBR. Molasses binders efficiently release almost all of the water during the preheated stage at 80 °C, while SBR does not, which makes them considerably easier to dry than SBR.

3.1.1.2. Textural properties of the raw coal, electroactive, and binder materials. Nitrogen adsorption-desorption isotherms were examined for the surface microstructural properties of the as-synthesized activated carbon (JCAC). It displays the typical type-I isotherm exhibiting H4 hysteresis, as illustrated in Fig. 3a, demonstrating the coexistence of microporous as well as mesoporous surfaces with a large surface area [10,32]. The porosity properties of JCAC from the N₂ adsorption-desorption isotherm study of the derived activated carbon (JCAC) (synthesized at an activation temperature of 800 °C with coal: KOH ratio of 1:2) reveals a high specific surface area (SSA) of 704.82 $\text{m}^2 \text{g}^{-1}$ with a pore volume of 0.26 cm³ g⁻¹ (Table S1). It was reported that an activation temperature of 800 °C represents the optimum condition for obtaining a high SSA in activated carbon derived from coal [10,49]. The presence of mesopores in the AC sample (JCAC) is also evident from the BJH pore size distribution curve (Fig. 3b) with the mode centred at 5 nm. On the other hand, the DFT pore size distribution of the synthesised AC indicates the existence of a significant amount of micropores (see Fig. 3c). Thus, the simultaneous presence of mesopores and micropores in the coal-derived AC significantly influences the specific capacitance and high-power performances in the supercapacitor cell for power applications.

The particle size distribution of ACs greatly influences the efficiency of the electroactive material employed for supercapacitor applications. At smaller sizes, electrodes of superior quality with even surfaces and consistent mass loading can be produced more easily from activated carbon powder [10]. We observed that the particle size distribution profile (Fig. 3d) of coal-derived AC (JCAC) is smaller, featuring a smaller average particle size (d0.5 = 49.922 μ m) in comparison to the commercial activated carbon (MtAC) (d0.5 = 69.924 μ m). As a result, the coal-derived AC (JCAC) performs significantly better than commercial AC (MtAC) in terms of electrode composition and calendering.

3.1.1.3. Results from the coating and bending tests of the binders. The collective findings from adhesion tests performed on the fabricated electrodes under various compositions are summarized in Table S2 and good adhesion was observed during the analysis. It is seen that when the optimal processing circumstances are followed, electrodes made with a water-based indigenous binder molasses exhibit the identical adhesion level as electrodes made with commercial binder i.e. CMC+SBR. Thus, the substitution of easily available water-based binders for super-capacitor fabrication represents a significant step toward the realization of an eco-friendly, cost-effective, and safer method of electrode fabrication [25,26].

The electrodes were cut into strips about 13 cm in width and encircled around steel pins (10 cm in diameter) to test the coatings' flexibility based on the new binders. It should be mentioned that, given that rolls in coating lines normally have a diameter of several centimeters, the flexibility test that was applied was fairly conservative [25]. Nonetheless, this makes it possible to objectively assess the relative flexibility/bendability of the electrodes with various binders. Figure S2 shows that JCAC-CMC-Molasses have a homogeneous surface without any cracks or wounds even at 2 mg cm⁻² mass loading, indicating significantly greater flexibility.

3.1.1.4. Morphological characteristics of the activated carbon sample and different fabricated electrodes. The microtexture or surface appearance of the activated carbon derived from coal (i.e., JCAC) and their electrodes made using different types of binders (JCAC-CMC-Molasses and JCAC-



Fig. 4. FE-SEM micrographs of the coal-derived AC (JCAC); (a) microscopic images at 20 µm, (b) Zoom-in view at 2 µm, (d) at 200 nm showing microporous and mesoporous surface, and (c) SEM-EDS atomic analysis.

CMC-SBR) were studied by employing electron beam (FE-SEM) analysis as illustrated in Fig. 4. The FE-SEM microscopic images (Fig. 4**abd**) show a significant quantity of tiny and sizable pores (abundant micropores with mesopores) on the surface of the coal-based AC as a result of ultrasonic-aided KOH chemical activation. These interlinked micropores are found to be beneficial for charge storage capacity boosting the electrode's electrochemical performance in the supercapacitor [10,39]. The atomic compositions of chemically activated JCAC were also evaluated via SEM-EDS elemental analysis (see Fig. 4 c), where C, O, F, Si, S, and K were detected with atomic compositions of 87.58, 6.20, 4.72, 0.40, 0.39, and 0.71 %, respectively.

Figures S3ac (see the supplementary materials) demonstrates a series of FE-SEM micrographs comparing the two types of electrodes i.e., JCAC-CMC-SBR and JCAC-CMC-Molasses, respectively. It is observed that the SBR-based electrodes exhibit more cracks in terms of both quantity and size than those made with molasses binder. SEM-EDS atomic analysis was employed to ascertain the elemental composition of the two types of electrodes. It was found that JCAC-CMC-SBR has C and O with atomic compositions of 96.28 and 2.72 %, respectively and JCAC-CMC-Molasses has C and O with atomic compositions of 95.72 and 4.28 %, respectively (**Figures S3bd**).

The TEM analysis of JCAC (Fig. 5**ab**) illustrates a surface with many pores and a sheet-like structure that tally with the observations obtained from FE-SEM analysis. The creation of a porous/lattice framework containing abundant micropores and mesopores is depicted in the HR-TEM images (Fig. 5c). The turbostratic lattice structure is represented by the standard diffused diffraction pattern over the selected area electron diffraction (SAED) (see Fig. 5d) of JCAC. Two bright rings, correlating with XRD analysis, were noticed in the SAED pattern, matching the crystallographic planes (002) and (100) [10]. The atomic composition of JCAC was also evaluated by TEM-EDS analysis (Fig. 5e), which reveals the presence of C, O, F, S, and K with 82.50, 9.24, 0.98, 1.10, and 6.18 %, respectively and those were also electronically mapped (in the JCAC product (Fig. 5f), respectively.

Another important finding from the HR-TEM analysis is that the coalderived AC electrodes JCAC-CMC-SBR (Fig. 6a-c) and JCAC-CMC-Molasses (Fig. 6d-f) show a sheet-like structure with a porous surface. Two bright rings that correlated with XRD analysis were observed in the SAED pattern aligning with the crystallographic planes (002) and (100) [10]. The atomic composition of JCAC-CMC-SBR and JCAC-CMC-Molasses (Fig. 6gh) were also evaluated by TEM-EDS analysis, where C, O, F, S, Cl, and K were detected along with their mapping images (Fig. 6i-p).

Thus, it is found that the coal-derived AC electrodes with two different types of binders have relatively good homogeneity along their thickness examined by FE-SEM and HR-TEM analysis, revealing that molasses is a viable binder for the fabrication of electrodes for supercapacitors.

3.2. Electrochemical performances of coal-derived activated carbon electrodes with different binders, current collectors, and neutral electrolytes

In order to investigate the practical applications of coal-derived activated carbon electrodes using the different types of binders in supercapacitor devices, two symmetrical coin cell supercapacitors (SC) were made with 1 M Na_2SO_4 as the green electrolyte. The prime focus was given on the effects of binder and electroactive material on



Fig. 5. TEM/HR-TEM analysis of coal-derived AC; (a) Low-magnification TEM image, (b) and (c) HR-TEM images showing the creation of a porous/lattice framework, (d) selected area electron diffraction (SAED) pattern, (e) TEM-EDS, and (f) mapping of elements (C, O, K, S, and F).



Fig. 6. TEM/HR-TEM analysis of the two coal-derived AC electrode materials. (a) Low-magnification TEM image, (b) HR-TEM images, (c) SAED pattern, (g) TEM-EDS, and elemental mapping of (i) C, (j) O, (m) S, and (n) K element of JCAC-CMC-SBR electrode; (d) Low-magnification TEM image, (e) HR-TEM images, (f) SAED pattern, (h) TEM-EDS, and elemental mapping of: (k) C, (l) O, (o) S, and (p) Cl element of JCAC-CMC-molasses electrode.



Fig. 7. Electrochemical analysis of AC electrodes (JCAC-CMC-Molasses, JCAC-CMC-SBR, MtAC-CMC-Molasses, and MtAC-CMC-SBR cast on three types of current collectors i.e., Al, formed Al, and etched Al current collector) in 1 M Li₂SO₄ electrolyte: (a, g, and m) CV curves at 5 mV s⁻¹; (d, j, and p) CV curves of JCAC-CMC-Molasses at different scan rates from 5 to 100 mV s⁻¹; (b, h, and n) GCD curves at current density of 0.5 A g⁻¹; (e, k, and q) GCD curves of JCAC-CMC-Molasses at different current densities from 0.5 to 10 A g⁻¹; (c, i, and o) The Nyquist plots; (f, l, and r) The Nyquist plot of JCAC-CMC-Molasses.

Comparative electrochemical outcomes of AC electrodes with different binders and current collectors evaluated from GCD and PEIS tests in 1 M $\rm Li_2SO_4$ electrolyte.

Sample codes	Current collector (C. C)	1 M Li ₂ SO ₄ electrol Specific capacitance (F g ⁻ ¹) at 0.5 A g ⁻¹	yte ESR _{DC} (Ω) measured from IR drop	ESR _{AC} (Ω) at 1 kHz
JCAC-	Aluminium C.	80	2.07	1.69
Molasses	C Formed aluminium C.	83	1.29	0.89
	Etched aluminium C.	117	0.80	0.79
JCAC-	Aluminum C.	80	2.11	1.90
SBR	Formed aluminium C.	71	1.39	1.32
	Etched aluminium C.	109	1.19	0.96
MtAC- CMC-	Aluminium C.	70	2.95	2.63
Molasses	Formed aluminium C.	66	1.64	1.39
	Etched aluminium C.	100	1.20	1.39
MtAC-	Aluminium C.	65	3.54	3.10
SBR	Formed aluminium C.	49	1.74	1.60
	Etched aluminium C. C	91	3.02	1.49

electrochemical characteristics, as well as the variability in specific capacitance, energy, and power density concerning different binders (CMC+molasses and CMC+SBR), current collectors (etched, formed, and aluminium current collectors), and electrolytes (1 M Li₂SO₄ and 1 M Li₂SO₄) which are elaborated in the subsequent sections. First, we have created a benchmark 2-electrode SwagelokTM cell using the commercially accessible lab-scale standard electrode formulation. For the sake of comparison, the coal-derived AC's (JCAC) electrochemical performances were compared with the commercial activated carbon (MtAC) as well as indigenous molasses binder with commercial binder.

3.2.0.1. Neutral electrolyte: 1 M Li₂SO₄

The comparative electrochemical performances obtained for the four coal-based graphitized porous electrode materials (JCAC-CMC-Molasses, JCAC-CMC-SBR, MtAC-CMC-Molasses, and MtAC-CMC-SBR) with three types of current collectors in 1 M Li₂SO₄ neutral electrolyte are demonstrated in Fig. 7. Fig. 7agm illustrate the comparative cyclic voltammetry (CV) profiles of the above-mentioned electrode materials in the potential window of 0-2 V at a scan rate of 5 mV s⁻¹ with three types of current collectors i.e., Al, formed Al, and etched Al current collector, respectively. A perfect double-layer capacitor with charge-discharge functionality was observed in the JCAC-CMC-Molasses electrochemical performance, matching the commercial electrode with a nearly perfect rectangular shape and no redox peaks. Fig. 7djp depict the CV profiles of JCAC-CMC-Molasses electrode obtained in a potential window of 0-2 V at various scan rates of 5, 10, 20, 50, and 100 mV s⁻¹ in a 1 M Li₂SO₄ electrolyte with three types of current collectors i.e., Al, formed Al, and etched Al current collector, respectively. At a higher scan rate, the CVs of the JCAC-CMC-Molasses electrode retained the rectangular pattern, indicating lower internal cell resistance [50]. In comparison to other electrode samples, the JCAC-CMC-Molasses electrode with etched Al current collector exhibited increased capacitance and superior reversibility even under higher scan rates, indicating the electrode with the ideal active mass loading (2 mg cm⁻²) has an outstanding rate performance. Hu et al. [51] highlighted the significance of using the proper thickness and mass of the active material for practical applications in order to obtain significant values for specific capacitance and energy density [1]. The study found that the ideal active electrode mass loading is 2 mg cm⁻², due to the superior binding ability of the molasses binder to activated carbon compared to the commercial binder like styrene-butadiene-rubber (SBR), which has an active mass loading of approximately 3 mg cm⁻².

In order to examine the specific capacitance value and cycling performances of the coal-derived AC electrode, galvanostatic charge-discharge (GCD) analysis was conducted at various current densities in the 1 M Li₂SO₄ electrolyte. The comparative GCD profiles of ACs at a current density of 0.5 Ag⁻¹ were demonstrated in Fig. 7bhn with three types of current collectors i.e., Al, formed Al, and etched Al current collector, respectively. The GCD curves for JCAC-CMC-Molasses with three distinct current collector types-Al formed Al, and etched Al current collector-are shown in Fig. 7ekq. These curves are obtained in the potential range of 0-2 V at various current densities of 0.5, 1, 2, 3, 5, and 10 Ag⁻¹. The GCD patterns of JCAC-CMC-Molasses were measured at both low and high current density, demonstrating a consistently linear potential change with an extremely lower iR drop in comparison to other electrode materials. Moreover, each electrode demonstrates a stable capacitive cycle with a symmetric triangular shape [10,50]. The maximum specific capacitance of JCAC-CMC-Molasses with etched Al current collector can reach 117 F g^{-1} at 0.5 A g^{-1} . This is on the higher side than that of commercial and other AC materials showing a specific capacitance of approximately in the range of 65–109 F g^{-1} (see Table 3). Thus, the JCAC-CMC-Molasses electrode outperforms commercial activated carbon in terms of capacitance. This suggests that low-grade coal, when used as an AC precursor, and molasses binder with numerous oxygen-containing functional groups, has better prospects in energy storage applications than the most widely used expensive precursor.

The ESR_{DC} of the electrodes calculated from the GCD curve is illustrated in Table 3. The ESR_{DC} of the JCAC-CMC-Molasses electrode is considerably lower at 0.80 Ω compared to other electrodes which is due to another factor like using an etched aluminium current collector.

Etched aluminium foil, in which an alkaline solution is used to roughen the foil's surface to enhance electrode material adhesion and reduce interface resistance [50]. Therefore, JCAC-CMC-Molasses cast on etched al current collector is a promising carbon electrode for the electrochemical energy storage device with promising specific capacitance.

In order to ascertain the electrolyte ion-diffusion resistance and internal charge-transfer kinetics of the AC electrodes, the potentioelectrochemical impedance spectroscopy (PEIS) test was performed in the SwagelokTM cell. Fig. 7cio illustrate the Nyquist plots of all the AC electrodes with the Al, formed Al, and etched Al current collectors, respectively. PEIS testing of JCAC-CMC-Molasses was performed before and after the galvanostatic charge-discharge process using a signal amplitude of 10 mV across the open circuit voltage and a frequency range of 10 mHz to 200 kHz (inset indicates the wider high-frequency area of the Nyquist plot) (Fig. 7flr). The Nyquist plots of JCAC-CMC-Molasses, utilizing an etched Al current collector, demonstrate an equivalent series resistance (ESR_{AC}) as low as 0.79 Ω at 1 kHz in a 1 M Li₂SO₄ electrolyte (Table 3). This emphasizes the outstanding electrical conductivity of coal-derived AC [10]. The nearly vertical line observed in the low-frequency area indicates the characteristic of an ideal doublelayer capacitor [52,53]. Thus, according to Warburg impedance, electrolyte ion diffusion occurs in active material pores in the middlefrequency section [10,52,54]. A small semicircle within the highfrequency area can be observed in the inset of Fig. 7flr, indicating



Fig. 8. Electrochemical analysis of AC electrodes (JCAC-CMC-Molasses, JCAC-CMC-SBR, MtAC-CMC-Molasses, and MtAC-CMC-SBR cast on three types of current collectors i.e., Al, formed Al, and etched Al current collector) in 1 M Na₂SO₄ electrolyte: (a, g, and m) CV curves at 5 mV s⁻¹; (d, j, and p) CV curves of JCAC-CMC-Molasses at different scan rates from 5 to 100 mV s⁻¹; (b, h, and n) GCD curves at current density of 0.5 A g⁻¹; (e, k, and q) GCD curves of JCAC-CMC-Molasses at different current densities from 0.5 to 10 A g⁻¹; (c, i, and o) The Nyquist plots; (f, l, and r) The Nyquist plot of JCAC-CMC-Molasses.

Comparative electrochemical outcomes of AC electroc	des with	different	binders
and current collectors evaluated from GCD and PEIS te	st 1 M Na	2SO4 elec	trolyte.

Sample codes	Current collector (C. C)	1 M Na ₂ SO ₄ electroly Specific capacitance (F g ⁻¹) at 0.5 A g ⁻¹	yte ESR _{DC} (Ω) measured from IR drop	ESR _{AC} (Ω) at 1 kHz
JCAC- CMC-	Aluminium C.	84	0.99	0.78
Molasses	Formed aluminium C.C	115	0.79	0.78
	Etched aluminium C.C.	127	0.82	0.47
JCAC- CMC-	Aluminium C.	83	1.00	1.18
SBR	Formed aluminium C.C.	101	0.88	1.03
	Etched aluminium C.C.	113	1.05	0.64
MtAC- CMC-	Aluminium C.	81	1.37	3.66
Molasses	Formed aluminium C.C.	97	1.12	1.29
	Etched aluminium C C	107	2.01	1.16
MtAC-	Aluminium C.	71	3.19	5.87
SBR	Formed	96	1.35	1.54
	Etched aluminium C.C	98	2.20	1.55

extremely low interfacial resistance because of the excellent electrical conductivity and microporous structure obtained for coal-derived AC [10,55].

3.2.0.2. Neutral electrolyte: 1 M Na₂SO₄

The electrochemical analysis of developed AC electrodes was thereafter studied in 1 M Na₂SO₄ electrolyte using the same 2-electrode configuration (SwagelokTM cell). Fig. 8 demonstrates the comparative electrochemical performances obtained from the four cells fabricated using JCAC-CMC-Molasses, JCAC-CMC-SBR, MtAC-CMC-Molasses, and MtAC-CMC-SBR with three types of current collectors in the presence of 1 M Na₂SO₄ neutral electrolyte.

Fig. 8agm illustrate the comparative cyclic voltammetry (CV) profiles of the above-mentioned electrode materials with a potential range of 0–2 V at a scan rate of 5 mV s⁻¹ which reveals a perfect double-layer capacitor with charge–discharge functionality in the JCAC-CMC-Molasses, matching the commercial electrode with nearly perfect rectangular shape and no redox peaks is observed. The CV profiles of JCAC- CMC-Molasses electrode obtained in a potential range of 0–2 V at various scan rates of 5, 10, 20, 50, and 100 mV s⁻¹ in a 1 M Na₂SO₄ electrolyte is depicted in Fig. 8**djp** and it is found that at a high scan rate, the CVs of JCAC-CMC-Molasses electrode maintained the rectangular shape, indicating low internal cell resistance [50]. Compared to other electrodes, the JCAC-CMC-Molasses electrode exhibited superior capacitance and enhanced reversibility even under elevated scan rates.

Fig. 8ekq represent the GCD curves of JCAC-CMC-Molasses for neutral electrolytes with a potential window of 0–2 V at different current densities of 0.5, 1, 2, 3, 5, and 10 A g⁻¹ indicating that the GCD patterns of JCAC-CMC-Molasses recorded at low and high current densities have ideal linear potential change with an extremely lower iR drop compared with other electrode materials. At a low current density of 0.5 A g⁻¹, the maximum specific capacitance of JCAC-CMC-Molasses with etched Al current collector can reach 127F g⁻¹ which is larger than that of commercial and other AC electrodes having a specific capacitance in the range of 71–113F g⁻¹ (see Table 4). The ESR_{DC} of JCAC-CMC-Molasses electrode is also calculated to be significantly lower with 0.82 Ω compared to other electrodes.

The corresponding PEIS results are shown in Fig. 8cio. This is to be mentioned that the PEIS testing was done both before and after the galvanostatic charge-discharge process, using a signal amplitude of 10 mV across the open circuit voltage within a frequency range of 10 mHz to 200 kHz. The inset of the Nyquist plot (inset widened high-frequency area) (see Fig. 8cio) shows a considerably small semicircle of JCAC-CMC-Molasses at high-frequency area which indicates the better movement of electric charge occurring at the interface between the electrode and electrolyte as compared with other electrode materials (JCAC-CMC-SBR, MtAC-CMC-SBR, and MtAC-CMC-Molasses). The Nyquist plots (Fig. 8flr) of the JCAC-CMC-Molasses electrode show an ESR_{AC} at 1 kHz as low as 0.47 Ω , indicating the excellent electrical conductivity of the coal-derived AC electrode (Table 4) [10]. The smaller ESR value in Na₂SO₄ electrolyte indicates that water-soluble CMC+molasses could expedite ion penetration into coal-derived porous graphitized activated carbon structures.

Thus, among the four electrodes, JCAC-CMC-Molasses with etched Al current collector showed a low ESR value, indicating good ionic conductivity at the electrode/electrolyte interface.

3.3. Electrolyte performance and comparative results

In order to establish proper baselines of the coal-derived ACs electrodes with molasses binder, the electrochemical properties of neutral electrolytes (1 M Li₂SO₄ and 1 M Na₂SO₄) were investigated and compared using the commercial electrode under similar conditions. The comparative data of Li₂SO₄ and Na₂SO₄ electrolytes with respect to different current collectors for the JCAC-CMC-Molasses sample is



Fig. 9. (a) Typical Ragone plot (Energy density vs. Power density) of the supercapacitor cell based on JCAC-CMC-Molasses in 1 M Na₂SO₄ electrolyte; (b) Cycling stability test of the supercapacitor cell based on JCAC-CMC-Molasses in 1 M Na₂SO₄ electrolyte showing the excellent capacitance retention of 92 % (at 5 A g⁻¹).

Comparative evaluation of our developed coal-derived AC's electrochemical properties with those reported in existing supercapacitor research.

Electrode material	Electrolyte	Cell voltage (V)	Power density (W kg ⁻¹)	Energy density (W h kg ⁻¹)	Reference
Mesoporous MnO ₂	$1 \text{ M Na}_2 \text{SO}_4$	1	~70	~28.40	[59]
Mesoporous MnO ₂	$1~{\rm M}~{\rm Li}_2{\rm SO}_4$	1	~70	~28.80	[59]
Microporous carbon	0.5 M Na₂SO₄	1.8	~40	~7	[60]
Activated carbon	0.5 M Na ₂ SO ₄	1.6	-	~10	[61]
3D flower-like and hierarchical porous carbon (FHPC)	1 M Na ₂ SO ₄	1.8	317.5	15.9	[62]
Highly porous graphene planes	6 М КОН	1	~50	6.5	[63]
Phosphorus- enriched carbons	1 M H ₂ SO ₄	1.3	33	16.3	[64]
Coal-derived activated carbon	1 M Na ₂ SO ₄ and 1 M Li ₂ SO ₄	2	2000.17	70.57	This work

illustrated in Figure S4.

The ionic conductivity of Na_2SO_4 is greater than that of Li_2SO_4 , which can be attributed to cation differences [56,57]. Although the current responses in the CVs of the Li_2SO_4 and Na_2SO_4 (Figures S4adg) differ, Na^+ exhibits higher current responses in both water decomposition reactions than Li⁺, most likely due to the stronger solvation-induced ion-water bond of Li⁺ [56,57]. The electrochemical properties of Na₂SO₄ electrolytes were superior to those of Li₂SO₄, therefore both electrolytes showed a similar 2 V potential window from the near-rectangular CVs at 5 mV s⁻¹. In addition, GCD curves of all the electrodes show a symmetric triangular shape with a stable capacitive cycle (**Figures S4beh**). The PEIS spectra (**Figures S4cfi**) of both electrolytes demonstrated near-vertical lines with comparatively small interfacial resistances, further supporting the capacitive behavior [58]. This suggests that despite the difference in their cation radii, Li⁺ and Na⁺ can still access the micropores within the coal-derived AC structure [56,58]. However, due to the higher ionic conductivity of the Na₂SO₄ electrolyte, the ESR of the cells with that electrolyte was lower than that of the Li₂SO₄ (0.47 vs. 0.79 Ω) (Tables 3 and 4).

In the Ragone plot of JCAC-CMC-Molasses electrode material, it is observed that even with an increase in the maximum power density up to 2000.17 W kg⁻¹, JCAC-CMC-Molasses still maintains a maximum energy density of 70.57 Wh kg⁻¹ in 1 M Na₂SO₄ (see Fig. 9a). The cycling stability of JCAC-CMC-Molasses is demonstrated in Fig. 9b, which revealed that it exhibits excellent cyclic stability with capacitance retention of 92 % after 10,000 galvanostatic charge–discharge cycles at 5 A g⁻¹.

As expected, the developed coal-based AC electrode with a biodegradable molasses binder in the neutral electrolyte has the maximum energy and power density when compared to the prior studies outlined in Table 5. We aimed to compare our findings to those from other active and binder materials for various electrolytes as shown in Table 5. Thus, coal-derived AC material has a higher energy and power density than the materials identified in previous studies, which is promising for future research and development.

In the study, the coal-derived AC with an indigenous binder like molasses, etched Al current collector, and Na_2SO_4 electrolyte-based material compositions are found to be very promising for more affordable supercapacitor cells that perform as well as or better than Li_2SO_4 electrolyte-based supercapacitor cells with similar components. For our further application purposes, the JCAC-CMC-Molasses electrode was



Fig. 10. Electrochemical analysis of coin cell supercapacitors (10 numbers) in Na₂SO₄ neutral electrolyte: (a) CV profile at 5 mV/s; (b) GCD profile at 0.5 A/g, (c) The Nyquist plots (d) Bode impedance.



Fig. 11. Application of coal-derived AC carbon-based coin cell supercapacitors powered by solar PV in IoT module; (a) Coin cell supercapacitor, (b) coin cell supercapacitor in series connection, (c) coin cell supercapacitor in parallel connection with charge board, supercapacitor bank in (d) front view and (e) back view, (f) Supercapacitor charging from PV cells and powering an IoT module from Supercapacitor bank, and (g) solar-powered supercapacitors in IoT application (inset showing the Wi-Fi connection to the mobiles during testing).

assembled to make a symmetric coin cell supercapacitor using etched a luminium current collector and 1 M Na_2SO_4 electrolyte.

3.4. Electrochemical testing of coin cell supercapacitor cell

Ten numbers of coin cell supercapacitors containing coal-derived graphitized AC, molasses binder, and etched aluminium current collector with 1 M Na₂SO₄ electrolyte solution was fabricated for further testing and application. Interestingly, the electrochemical properties of the coin cell supercapacitor (Fig. 10) have shown similar electrochemical properties as observed with the SwagelokTM cell testing. Fig. 10a illustrates the comparative cyclic voltammetry (CV) profiles with a rectangular pattern without oxidation-reduction peaks of the coin cell supercapacitors in the potential range of 0–2 V at 5 mV $\ensuremath{\text{s}}^{-1}$ indicating a perfect double-layer capacitor with charge-discharge functionality. The comparative GCD profiles of coin cell supercapacitors at 0.5 A g^{-1} were demonstrated in Fig. 10b and the total capacitance of the ten-coin cell supercapacitors is calculated to be 1.12 F. The ESR_{AC} for coin cell supercapacitor at 1 kHz is as low as 1.51 Ω which is calculated from the Nyquist plots (Fig. 10c). This shows that the fabricated coin cell supercapacitors have low electrical resistance and good conductivity between the electrodes and the current collector (Table S3).

The Bode impedance of the coin cells illustrated in Fig. 10d shows that the coin cells have good capacitive behavior since the phase angle was approximately 70 °C at 0.01 Hz. Remarkably, the point of intersection at a phase angle of 45 °C indicates the characteristic frequency (f_o) at which capacitive impedance equals resistive impedance[10]. Also, due to the AC's high specific surface area and hierarchical porosity, JCAC-CMC-Molasses demonstrated improved ion transport behavior [10].

3.5. Application of coin cell supercapacitor bank powered by solar energy in IoT module

The fabricated coin cell supercapacitors were integrated with the PV solar cell to power the IoT module, as shown in Fig. 11. In this setup, to boost capacitance and voltage, a set of coin cell supercapacitors were connected in series and parallel as shown.

Two coin-cell supercapacitors were connected in series, and then five sets of two coin-cell supercapacitors (total number of cells: 10) were connected in parallel to achieve a high capacitance value (Fig. 11 **a-e**). The schematic diagram of a supercapacitor charging from PV solar cells and powering an IoT module from a supercapacitor bank is shown in Fig. 11f.

A practical approach for combining the symmetric coin cell supercapacitor (4 V, 1.12 F) with a commercial solar panel (6 V, 750 mAh) to use in the IoT module for a self-sustaining power source, as shown in Fig. 11g. Table S4 represents the recorded output voltage (V) data during coal-based supercapacitor charging from PV cells and Table S5 displays that the IoT device functions correctly when the output power is approximately 3 W. Also, it has been noted that the device stops functioning and becomes non-functional when the output power drops to 1.3 or 1.4 W. Therefore, the solar-charged supercapacitor bank can successfully convert solar energy into electrical energy when exposed to sunlight, as demonstrated by its ability to continuously power a commercial IoT module (4–6 V, 600 mAh) (Table S4 and S5).

The coal-based supercapacitor cells are effective in capturing and converting solar energy into electrical energy, followed by its storage, has demonstrated its suitability to use renewable energy sources and the ability to generate electricity at a reasonable cost. Thus, these findings demonstrate the coin cell supercapacitors made using the novel coalderived graphitized activated carbon can be satisfactorily combined with solar power and are regarded as an ultrafast advanced energystorage system that can used to continuously power the IoT modules and will lessen the demand for energy and lower the price of electricity shortly.

4. Conclusions

High-specific surface area coal-derived activated carbon and biodegradable molasses were used to develop a high-efficiency symmetric supercapacitor electrode for coin cell supercapacitors. The electrochemical performance of these symmetric supercapacitors was investigated using a green aqueous Na₂SO₄ electrolyte at an operating potential of 2 V, nearly double that of other aqueous electrolytes like KOH, HCl, and H₂SO₄. The fabricated JCAC-CMC-Molasses electrode demonstrates a remarkable specific capacitance of 127 F g⁻¹ at 0.5 A g⁻¹, with excellent cyclic stability, exhibiting an impressive capacitance retention of approximately 92 % after 10,000 charge-discharge cycles. Additionally, it features a high energy density of 70.57 Wh kg⁻¹, coupled with a power density of 2000.17 W kg⁻¹, surpassing the performance of other carbonbased supercapacitor electrodes. Thus, this study integrates coal-based coin cell supercapacitors with a PV solar cell to effectively power an IoT module revealing it's the effectiveness in converting solar energy into a reliable power supply for practical applications in electronics sector. Furthermore, these results open up a new avenue for sustainable conversion of coal feedstock for advanced and eco-friendly energy storage devices suitable for applications in IoT modules, potentially reducing energy demand and electricity costs in the near future.

CRediT authorship contribution statement

Mousumi Bora: Writing – original draft, Validation, Methodology, Investigation, Formal analysis, Data curation. Debashis Sarmah: Writing – original draft, Methodology, Investigation, Formal analysis. Santhi Maria Benoy: Methodology, Investigation, Formal analysis. Abhishek Hazarika: Investigation, Formal analysis. Akhil Rajbongshi: Writing – review & editing, Validation, Methodology. Binoy K Saikia: Writing – review & editing, Visualization, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

Acknowledgments

The authors would like to thank the Director of CSIR-NEIST for the interest in our energy and environment research. We gratefully acknowledge the suggestions provided by Dr. Dhrubajyoti Bhattacharjya during the initial stages of this work. The authors would like to express their appreciation for the financial support (GPP348) received from MeitY, Government of India. The author (MB) would like to thank the UNIVERSITY GRANTS COMMISSION (UGC) for Savitribai Jyotirao Phule Fellowship for Single Girl Child (SJSGC) with fellowship grant (QSP/MR/11/06/2023(UGC-SJSGC)/6333).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.enconman.2024.118964.

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