Oil Palm Lignin-Derived Laser Scribed Graphene in Neutral Electrolyte for High-Performance Microsupercapacitor Application

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Abstract

Lignin is a renewable natural resource that could be derived from oil palm empty fruit bunches. It has generated significant interest as a precursor in synthesizing graphene as anode and cathode material for supercapacitors. In this paper, we report the synthesis of 3D hierarchical Laser Scribed Graphene (LSG) on a flexible polyimide substrate from lignin extracted from empty fruit bunches (EFB) of oil palm for micro-supercapacitor applications. The intensity and speed of the laser have been tuned to yield densely compacted oil palm lignin LSG at a laser power of 70 % and a speed of 30 % (OPL-LSG 7030). OPL-LSG 7030 possessed lower equivalent series resistance of 60.1 Ω and a larger crystalline size of ~31 nm than the rest of the tested samples. It exhibited exceptional areal capacitance of 30.77 $mFcm^{-2}$ at a current density of 0.08 $mAcm^{-2}$, an energy density of 0.00176 $mWhcm^{-2}$ and a power density of 0.25 $mWcm^{-2}$ when using a unique neutral PAAS/K_2SO_4 gel electrolyte. It achieved excellent capacitance retention of 88.4 % after 5000 charge/discharge cycles and remarkable mechanical stability of 95 % after 400 bending cycles. Furthermore, electrochemical studies revealed the redox properties of readily available quinone/ hydroquinone in the oil palm lignin, which could be inherited in graphene electrodes through a feasible and affordable approach for flexible green energy storage applications.

Keywords: Oil palm lignin, Laser scribed graphene, Microsupercapacitors, Flexible devices

1.0 Introduction

Practical energy storage components for flexible devices are necessary for almost all consumer applications in the 21st century. Supercapacitors are electrical storage devices that utilize either electrolyte adsorption or rapid surface redox reactions for energy storage in flexible electronics [1]. Supercapacitors bridge the gap between an electrolytic capacitor and a battery by combining their extraordinary energy, power density capability and excellent life cycles in an environmentally friendly, sustainable construction [2]. There are three types of supercapacitors: - i.e., electrochemical double layer capacitor (EDLC), pseudo capacitor, and hybrid supercapacitor combining an EDLC and pseudo capacitor. EDLCs are typically constructed from activated carbon, carbon nanotubes, and graphene, while pseudo capacitors comprise metal oxides and conducting polymers [3], [4]. As charges are stored on the anode and cathode surface of EDLCs, increasing the surface area of the material increases the energy densities.

On the other hand, the pseudo capacitor achieves higher energy densities through a faradaic mechanism involving redox reaction in transferring electrons across the electrode [2], [5]. EDLC electrode with limited charge storing capacity is usually paired with redox reactive metal oxides with poor cyclic stability to overcome each of their drawbacks [6], [7]. In recent studies, doping of heteroatoms (O, N, S, B, P) enhanced the electrical, surface, chemical, and mechanical properties of graphene [6], [7]. For instance, Vignesh and his colleagues have successfully synergized nitrogen-doped reduced graphene oxide with cobalt-based metal oxides ($ZnCo_2O_4$) to achieve a capacitance of 950 Fg^{-1} [8].

Eco-friendly green energy storage components free from lithium-ion significantly prevent toxicity and excessive heat to humans, regardless of whether they are used in wearables or disposed of in the future [9]–[11]. Developing energy storage applications using biomass waste has become an area of research for a sustainable carbon source and as a preventive measure against the negative impact of excessive biowaste on the environment [12]–[14].

Lignin is one of the most abundant organic resources on earth and a key source of renewable aromas in nature [15]. It contains more than 60% carbon and is mainly generated as a waste by-product in large quantities in pulp mills, oil palm mills, and

biorefinery facilities [16], [17]. The amount of lignin produced from these industries exceeds 62 million tons annually in the United States, but only 2% are recycled into other valuable applications [17]. Malaysia, a major palm oil producer, generates 22–23 million tonnes of empty fruit bunches (EFB) [18]. Utilizing lignin as a carbon precursor provides access to the functional groups, i.e., carboxyl, hydroxyl, and carbonyl groups present within its cell. For instance, lignin with functional groups such as benzyl and phenolic groups provides ion storage sites for ion storage and active ion storage reaction sites, which promotes high redox activity in supercapacitors [16]. Lignin therefore has a variety of uses when utilized as an energy storage device [16], [19], including lignin sourced from raw wood [20], kraft pulp [21], [22], softwood [17], hardwood [23], [24], crop waste [25], bio-ethanol [26], [27] and cornstalk residue [28].

In recent years, researchers have successfully synthesized oil palm lignin-derived graphene from EFB [29]–[31]. Despite severe downsides such as high energy consumption and complex processes, pyrolysis and carbonisation techniques are alternatives to synthesising graphene from oil palm lignin [17]. Furthermore, graphene produced via those processes have extremely small yields and suffers from significant structural damage as a result of high temperatures (>1000 °C) [1]. The recent discovery of cost-effective laser lithography techniques to produce high-quality 3D graphene from other lignin sources has superseded these conventional techniques, which come with significant drawbacks [7], [21], [23], [32], [33]. The highly porous 3D graphene structure obtained through optimized laser parameters provides a larger surface area for better electrochemical activity and electrolyte diffusion [32]. By optimizing the laser scribing of oil palm lignin-based 3D graphene will improve the quality of graphene with minimal process-induced defects. In this regard, adequate modification of the laser power and speed may result in high-quality graphene in terms of porosity and composition without further compromising the structure of the material [17]. In addition, it is critical to make other decisions, such as selecting the electrolyte and the possible window for the electrode material, to maximize the electrode's capability [5]. It has recently been discovered that the application of a dilute acid electrolyte such as H_2SO_4 on an electrode made of lignin results in protonation of the lignin functionalities and forms cleavage between the linkages found on lignin [34], [35], preventing lignin from performing to its maximum capacity.

This research demonstrates the robust synthesis of graphene derived from novel biowaste oil palm lignin on a flexible polyimide substrate using a low-cost, precise, and single-step laser scribing technique. Independent variables, including laser power and speed, have been well-optimized to synthesize high-quality graphene without further damaging the flexible substrate. To our knowledge, for the first time, synthesized lignin-derived graphene electrode has been assembled employing a neutral electrolyte (K_2SO_4) combined with sodium polyacrylate gel to maximize the redox potential of hydroquinone found in lignin. An outstanding areal capacitance of 30.77 $mFcm^{-2}$ at 0.08 $mAcm^{-2}$, excellent cyclic stability of 88.4 % capacitance retention rate after 5000 charge-discharge cycles, energy density of 0.00176 $mWhcm^{-2}$ and a power density of 0.25 $mWcm^{-2}$ has been achieved by optimized the lignin-derived graphene electrode. Oil palm lignin can be effectively transformed into a functionalized graphene electrode for flexible micro-supercapacitor applications.

2.0 Methodology

2.1 Materials

Lignin was extracted from the oil palm empty fruit bunches and converted as lignosulfonate using a method reported elsewhere [36]. Sodium polyacrylate (PAAS) and Potassium Sulphate (K_2SO_4) were purchased from Sigma-Aldrich. Polyimide film with a thickness of 0.127 mm was purchased from Avantis Laboratory Supply. Deionized (DI) water was used in diluting chemicals and preparing the samples.

2.2 Preparation of OPL-LSG electrodes via laser scribing

Lignin solution (20% v/v) was prepared by adding 20 g of lignin powder into 100 ml of deionized water and stirring for 1 hr at room temperature. The blade coating technique was used to coat the lignin solution on a clean commercial polyimide (PI) film at a blade-to-substrate distance of 500 µm. The lignin-coated polyimide substrate was utilized for laser scribing after being dried in an oven at 50°C for an hour. The CO_2 laser engraving system (V-460, Universal Laser System, Scottsdale, Arizona, USA), with a maximum power of 30 W and a speed of 1000 mm/s, was used to print the interdigitated electrode. In this work, the applied laser power and the moving laser speed were varied from 50 % to 70 % and 30 % to 50 %, respectively. Power and speed beyond this range was not considered since

they are known to either fail to produce graphene or severely damage the flexible polymer substrate. The untreated lignin layer was then dissolved using a water bath, and the remaining water was dried overnight in an oven. OPL-LSG X Y represents oil palm lignin laser-scribed graphene synthesized at a laser power of X% and a speed of Y%. For example, LSG derived at a laser power of 50% and laser speed of 50% is denoted as OPL-LSG 5050. Additionally, this work used an ordinary polyimide derived laser scribed graphene (P-LSG X Y) as a control.

2.3 Assembly of OPL-LSG-based microsupercapacitors

The CorelDRAW software was used to create a CAD drawing of the desired interdigitated electrode before the laser irradiation. The microsupercapacitor's current collectors were coupled with conducting wire and covered with silver paste. PAAS/K_2SO_4 (0.5 M) gel electrolyte was coated on top of 6 pairs of microelectrodes with a total area of 0.5 cm^2 to complete the assembly of microsupercapacitor. Here, PAAS/ K_2SO_4 (0.5 M) was made by mixing 10 ml of 0.5 M K_2SO_4 solution with 1g of sodium polyacrylate salt until a gel-like material was created. Before obtaining the electrical reading, the developed microsupercapacitor was wrapped in polyimide tape and allowed to immerse the microelectrodes in the gel electrolyte for a few hours, to ensure complete diffusion of electrolyte into electrode material.

2.4 Materials characterization

A square sample measuring 1 cm^2 in the area has been laser scribed for each of the samples to measure the surface morphology of the material by Field Emission Scanning Electron Microscopy, FESEM (TESCAN CLARA, UHR SEM), and Scanning Electron Microscopy, SEM (Zeiss Evo LS15) analysis. A few laser-scribed samples were scratched and mixed in ethanol to study the material's microstructure through Transmission Electron Microscopy, TEM (Hitachi, HT7830). The sheet resistance of 1 cm^2 OPL-LSG square samples was measured using van der Pauw (VDP) technique using Ecopia, HMS-5000. The Raman spectra of 1 cm^2 OPL-LSG square samples were obtained from Raman Spectrometer (DXR3 microscope) at 532 nm wavelength. The detailed calculation to identify crystalline size (L_a) for Raman readings is included in the supplementary material. The specific surface area (SSA) was computed using the Brunauer-Emmett-Teller (BET) technique, and pore size distributions were assessed using the Barrett

Joyner Halenda (BJH) model on the N_2 adsorption-desorption isotherms (Tristar 3020 Plus) for the black powder scratched from laser scribed material. The surface material composition of the black powder was examined using X-ray photoelectron spectroscopy (XPS) by Thermo Scientific K-Alpha. The functional groups before and after laser scribing were examined using Fourier-transform infrared spectroscopy (FTIR) by Perkin Elmer, Spectrum One. The X-ray diffraction (XRD) experiments were carried out using an X-ray diffractometer for the black powder by PANalytical X'Pert3 Powder, with an exposure duration of two seconds per step and a step size of 0.02 degrees per step.

2.5 Electrochemical characterization

Cyclic voltammetry (CV), galvanostatic charge-discharge (GCD), and electrochemical impedance spectroscopy (EIS) of fabricated microsupercapacitor were measured in a two-electrode system using an electrochemical workstation by (Metrohm, Autolab). A voltage window of -0.4 V to 0.4 V with a scan rate ranging from 10-200 mVs^{-1} was used in Cyclic voltammetry (CV) measurement. Current densities ranging from 0.08 – 1.0 $mAcm^{-2}$ were used in galvanostatic charge/discharge (GCD) measures. EIS measurements with an amplitude of 10 mV were recorded in the frequency range of 0.01 Hz to 1E+5 Hz. The detailed calculations of electrochemical readings are given in the supplementary material.

3.0 Results and Discussion

3.1 Fabrication and characterization of OPL-LSG electrodes

Fig. 1 depicts the schematic for constructing an OPL-LSG flexible microsupercapacitor, primarily using laser scribing. The lignin solution was first drop-cast onto the polyimide sheet. Following an hour of drying, the sample was laser scribed using a CO_2 laser to generate the desired microelectrode pattern. Before integrating the sample into a pliable micro supercapacitor, untreated lignin molecules were removed using a water bath. The inset on the left indicates that graphene derived from lignin has been functionalized by the assembled electrode. In contrast, the inset on the right displays the FESEM images of highly porous 3D laser-scribed graphene (LSG).

The surface morphology of OPL-LSG electrodes fabricated at various laser speeds and power was observed using a Field Emission Scanning Electron microscope (FESEM). Notably, the highly porous structure of graphene is similar to previously developed lignin [22], [32], [37]. Low-resolution images of the laser-irradiated graphene from OPL-LSG 7030 and OPL-LSG 5050 show a 3D wave-like porous structure (Fig. 2a, d). In contrast, the magnified images reveal a beehive-like structure with numerous macropores and mesopores induced by the discharge of gaseous products such as H_2O and CO_2 (Fig. 2b, e) [21], [32]. The pores enhance the material's surface area, which may promote electrolyte diffusion during the electrochemical process [21], [22]. However, it is important to note that the electrode material produced with a higher laser speed and lower laser intensity has gaps exposing the polyimide layer between them that impede the flow of charge during the electrochemical process, as shown in both views (Fig. 2d-e). Upon examining the cross-sectional view of the electrode material, it is evident that OPL-LSG 7030 has a higher average thickness of 41.7 µm, densely packed with a foam-like material for better electrical continuity (Fig. 2c, f). Identical results were observed in polyimide graphene by varying the laser power and speed (Fig. S1, S2). Further evidence for the transformation of lignin into porous graphene appears in the SEM images of lignin acquired before and after laser scribing on top of plain ITO glasses (Fig. S3, S4).

Fig. 2g shows Transmission Electron Microscopy (TEM) images of single-layer and wrinkled multilayer graphene flakes. OPL-LSG 7030 in Fig. 2g-h, show a variety of pores ranging from meso to macro that would provide a larger surface area for electrolyte ion facilitation [32]. With a d-spacing (interlayer spacing) that correlates to (002) lattice plane of 0.367nm, OPL-LSG 7030 has enhanced the incorporation and removal of electrolyte ions within the graphene electrode (Fig. 2i) [7]. Additionally, the existence of polycrystalline electrode material with a prominent ring reflects the (002) plane, as shown by the selected area electron diffraction (SAED) pattern in Fig. 2i. OPL-LSG 7030 has the lowest average sheet resistance, measuring 368 ohms/square, in line with FESEM findings and Van der Pauw measurements (Fig. 3a). The findings demonstrate that resistance decreases as laser power increase and speed decrease. Polyimide sheet coated with lignin performed as an insulator with a notable resistance level.

Raman spectroscopy was used to examine the impact of varying laser power (fixed speed at 30%) and speed (fixed power at 70%) on the synthesis of graphene. Based on the results (Fig. 3b-c), all the samples exhibit three distinctive peaks located at ~1350 cm^{-1}, ~1585 cm^{-1}, and 2690 $cm^{-1,}$ which are associated with the D (triggered by lattice defects and vacancies of sp^2 carbon bonds), G (sp^2 hybridized carbon), and 2D (stacked layer of graphene) bands, respectively [17], [33]. The adequate thickness and the structural disorder of the graphene were observed using the ratios of the I_G/I_{2D} and I_D/I_G [32], [33]. Although a slight gain can be noticed initially, the I_D/I_G and I_G/I_{2D} ratios of various laser powers and speeds progressively declined and reached saturation (Fig. 3d-e). OPL-LSG 7030 is higher quality graphene than previous variations, with the fewest graphene layers. At the same time, further increases in laser power (80%) and decreases in laser speed (20%) deteriorate the polymer substrate [22]. OPL-LSG 7030 has an average crystalline size of ~31nm, 10 nm greater than the P-LSG 7030 (Fig. S5). In addition, Raman spectra of graphene derived from lignin-coated ITO glasses without polyimide indicate the presence of graphene by showing similar D, G, and 2D peaks with I_G/I_{2D} and I_D/I_G values of 0.998 and 1.007 (Fig. S6). As a result, increasing laser power while lowering laser speed is required to achieve high-quality graphene from OPL.

The FTIR spectra in Fig. 3f represent the bonding states of graphene synthesized from polyimide and oil palm lignin, respectively. The OH stretching is the broadest and most dominating peak at ~3400 cm^{-1}. Compared to polyimide graphene, lignin graphene shows a sharper C-O peak at 1070 cm^{-1}. Moreover, a small peak could be observed at ~1045 cm^{-1}, representing the hydroquinone functionality group derived from lignin [34], [38]. Besides that, carbon-carbon double bond and aromatics could be observed for graphene at 1628 cm^{-1} and 1524 cm^{-1} [23]. The remaining absorption peaks, which are situated at 2945 cm^{-1}, 2352 cm^{-1}, 1731 cm^{-1}, and 1406 cm^{-1}, are associated with C-H stretching, O=C=O (CO2), C=O bonds, and C-O stretching [23], [39]–[41]. Fig. S7 shows that the three peaks of pristine lignin, linked to S=O, C-O, and hydroquinone and situated at 1195, 1112, and 1045 cm^{-1}, were significantly reduced with the conversion of lignin into graphene [37], [40]–[42].

The N_2 adsorption and desorption isotherms were performed on OPL-LSG 7030 to investigate the porosity of the sample (Fig. 4a). The type-III hysteresis loop that is shown on the plotted isotherm confirms the presence of the meso- and macropores in

graphene, which would facilitate ion diffusion at high current densities [43], [44]. Notably, type III does not occur frequently, primarily because of the water vapor on the basal plane of fabricated graphene [45]. Accordingly, OPL-LSG 7030 has a Brunauer-Emmett-Teller (BET) specific surface area (SSA) of 37.13 m^2g^{-1} and a pore volume of 0.125 cm^3g^{-1}. The average pore diameter for OPL-LSG 7030, which consists of a combination of meso and macro pores, is 11.69 nm, as further evidenced in Fig. 4b. A variety of pores, from micro to macro, enhance the overall electrochemical performance of OPL-LSG 7030 by expanding its surface area, accelerating electrolyte ion movement, and retaining ions in a buffer zone [7].

To further examine the elemental compositions and chemical structures, XPS measurements were performed for OP lignin and OPL-LSG 7030. The XPS survey spectra of the samples are shown in Fig. 4c, which revealed three primary peaks at 285.3 eV, 531.3 eV, and 1072 eV, corresponding with C 1s, O 1s, and Na 1s, respectively [22]. The Na peaks are primarily caused by residues from using NaOH during the lignin extraction process [17], [22]. It is apparent that after laser scribing, C 1s intensity increased while Na 1s and O 1s intensity decreased, indicating that laser scribing transforms lignin into high-quality graphene. The high-resolution C 1s XPS peak were further divided into four components, which are located at ~284.5 eV, ~285.2 eV, ~286.2 eV, and ~290.0 eV, respectively, for the C-C/C=C bond, the C-O bond, the C=O bond, and the O-C=O bond [22], [46]. (Fig. 4d, S8). Furthermore, the O 1s XPS spectra of both samples were broken down into three subpeaks, namely C=O (~532.4 eV), C-O (~533.8 eV), and -COOH (~536.2 eV) [7], [47] (Fig. 4e, S9). The greater laser power utilized to carbonize the lignin coated on polyimide increased the local temperature and removed the impurities such as Na that may impact electrochemical performance [32], [48]. Fig. 4e and S9 further demonstrate how the dominance of the C-O bond has weakened and been superseded by the other bonds made available throughout the graphitization process.

XRD analysis was used to examine the structural analysis of fabricated graphene (Fig. 4f). Both samples exhibited one broader peak at 25.9° and a minor peak at 43°, as shown in the previous SAED image. The (002) peak's asymmetry is connected to the OPL-LSG's minor disordered amorphous structure, while the sharp peak confirms the fact that lignin was converted into a highly crystallized graphitic structure [23], [32]. In comparison to OPL-LSG 5050, OPL-LSG 7030 has significantly more evident and

unambiguous (002) and (100) peaks in the analysis [23]. Due to the presence of Na_2SO_4 (a salt produced by the soda pulping process), OPL-L5050 from lignin precursor showed additional peaks at ~ 34°, which is further supported by the presence of Na impurities found in XPS survey spectra [49], [50]. Inadequate graphitization at low laser power has prevented the salt from protruding and being washed away [32].

3.2 Electrochemical performance OPL-LSG-based films

All the prepared samples were assembled into a complete electrode with the addition of PAAS/K_2SO_4 electrolyte and conducting wires, and the electrochemical characteristics of each electrode were assessed. A potential window ranging from -0.4V to 0.4V has been utilized for cyclic voltammetry (CV) and Galvanostatic Charge-Discharge (GCD) curves. Fig. 5a shows the CV curves of a microsupercapacitor with OPL-LSG at various laser power and speed. A pair of distinct redox peaks can be observed from the CV curves due to quinone/hydroquinone groups (aromatics compound) from oil palm lignin in the fabricated laser-scribed graphene observed in FTIR. Hydroquinone offers pseudocapacitance for graphene and enriches its overall functionality by storing and releasing electrons during the electrochemical process, [51], [52]. The estimated areal capacitance (measured in $mFcm^{-2}$) at 50 mVs^{-1} of OPL-LSG 5030, OPL-LSG 6030, OPL-LSG 7030, OPL-LSG 7040, and OPL-LSG 7050 are 1.57, 2.75, 3.15, 2.84 and 1.72, respectively. OPL-LSG 7030 exhibits the highest areal capacitance at 50mVs^{-1} among the different laser powers and speeds considered, indicating that the electrochemical performance has been improved by the densely packed graphene layer from FESEM images.

To further examine the samples, a detailed CV test was performed on the produced samples at various scan rates, ranging from 10 mVs^{-1} to 200 mVs^{-1} (see Fig. 5b, S10). It was apparent that at increased scan speeds, cathodic peak transitions to the positive side and anodic peak transitions to the negative side of the plot were all caused by the internal resistance of the electrode [53]. Fig. 5c depicts the relationship between samples of varying laser power and speed performed at various scan rates with the areal capacitance determined from CV curves. OPL-LSG 7030 surpasses all comparable samples at lower scan rates yet showed average values at higher scan rates due to the limited access time of electrolyte ions entering the thicker graphene

layer. OPL-LSG 7030's excellent areal capacitance value of 8.88 $mFcm^{-2}$ at a scan rate of 10 mVs^{-1} indicates that higher laser power and lower laser speed resulted in higher quality graphene.

Fig. 5d represents graphene's Galvanostatic Charge Discharge (GCD) curves synthesized at various power and speed settings. The GCD curves' deviation from the ideal triangular shape demonstrates a significant contribution from pseudocapacitance, supporting earlier CV test results. OPL-LSG 7030 had the longest discharge time, as indicated by the GCD profile measured at a current density of 0.1$mAcm^{-2}$, which is consistent with the larger area from CV curves. When OPL-LSG 7030 was tested at current densities varying from 0.08 $mAcm^{-2}$ to 1 $mAcm^{-2}$, similar outcomes were seen, with excellent columbic efficiency (Fig. 5e). It is clear that at lower current densities, the electrolyte ions penetrate the interior plane of the electrode material at a higher rate compared to higher current densities. The electrochemical reading of the tested material is enhanced substantially at lower current density, with reactivity from both inner and outer surfaces [53]. Fig. 5f compares the areal capacitance obtained from the GCD profile of assembled graphene electrodes and current density. The calculated areal capacitance (measured in $mFcm^{-2}$) of OPL-LSG at a current density of 0.08 $mAcm^{-2}$ with increasing laser power are 14.8, 25, and 30.77, while 30.77, 24.5, and 13 at increasing laser speeds. Once again, OPL-LSG 7030 proves its remarkable capacitance property at higher and lower current densities at a volumetric capacitance of 7.379 Fcm^{-3}.

Fig. 6a and the inset illustrate the Nyquist plots of laser-scribed lignin at various power and speed settings between 100 kHz and 10 mHz. Equivalent Series Resistance (ESR) measures the surface resistance of the material in the high-frequency region, where the plot intersects the X axis (real axis). In contrast, Warburg resistance is the electrolyte ion diffusion into the pores of electrodes in a low-frequency region [6], [54]. Overall, all the displayed graphs exhibit the standard behaviour of porous carbon, with steeper inclinations of more than 45° without a distinct semicircle appearance [55]. OPL-LSG 7030 recorded the lowest ESR with a value of ~60.1 Ω, indicating capacitor behaviour, while maintaining a close distance from the Y axis (imaginary axis). The ESR values for other samples from OPL-LSG 5030 to OPL-LSG 7050 are 158 Ω, 77.3 Ω, 91.1 Ω, and 133 Ω, respectively. The EIS data and other electrochemical

measurements provided additional evidence of the high conductivity of fabricated graphene.

The electrochemical stability was assessed at a current density of 0.5 $mAcm^{-2}$ to examine the overall performance of synthesized graphene, as shown in Fig. 6b. Notably, OPL-LSG 7030 has a cycling stability of 88.4% capacitance retention over 5000 cycles with an initial boost in capacitance retention due to an "activation process" [56]. OPL-LSG 7030 retained 80.78% of its initial capacitance after 8000 cycles (Fig. S11). The overall decline in the plot was primarily caused by graphene detachment on the fabricated electrode, which reduced the active area of the electrode [57]. Additionally, OPL-LSG 7030 demonstrated the highest areal energy density of 0.00176 $mWhcm^{-2}$ at volumetric energy density 0.422 $mWhcm^{-3}$, and the highest areal power density of 0.25 $mWcm^{-2}$ at volumetric power density 59.95 $mWcm^{-3}$ due to the thicker graphene layer's ability to facilitate greater ionic activity (Fig. 6c).

The fabricated OPL-LSG 7030 electrode was subsequently subjected to a bending test to determine its pliability in a practical application. The CV test for 400 continuous cycles at 50 mVs^{-1} demonstrated remarkable mechanical stability at the 400th bending cycle (Fig. 6d), with capacitance retention of over 95% (Fig. 6e) in the observable region. The Ragone plot in Fig. 6f depicts the relationship between the energy and power density of fabricated graphene OPL-LSG 7030 which is comparable to most previously reported laser-derived planar supercapacitors including 5B-LIG [58], M-PBRV-RGO [59], LSG-P24 [32], and PA-PBI-LIG [60]. At the same time, OPL-LSG 7030 outperforms LIG [61] and 3D-GF [62] in terms of energy and power density. Furthermore, Ph-ddm/LIG [63], KOH-LIG [64] , LIG-B6 [43], LSG-P24-Au [32], sLIG-O/S14 [33] and LWG-ONS 1.5 [7] microsupercapacitors possess an extraordinary electrochemical result either due to the composite material formation, electrode activation or heteroatom doping. Table S1 provides a comparison of the material mentioned above.

4.0 Conclusion

In summary, lignin from oil palm EFB has been effectively transformed into a functionalized graphene using a single-step and cost-effective laser scribing technique. The resulting graphene possesses redox qualities as the laser scribing process retained the properties of the aromatic compounds within the lignin. A high-

quality 3D graphene with a highly porous structure (meso and macro pores), reduced resistance, larger surface area, and smaller pore size has been developed at a laser power of 70 % and laser speed of 30 %. Most importantly, the resulting microsupercapacitor exhibits improved electrochemical properties such as high areal capacitance (30.77 $mFcm^{-2}$), decent energy/power densities (0.00176 $mWhcm^{-2}$, 0.25 $mWcm^{-2}$), and comparable cyclic stability (88.4% for 5000 cycles) than those reported in previous studies. The developed graphene electrode also displayed exceptional mechanical durability at a scan rate of 50 mVs-1 following 400 bending cycles. Doping or incorporating nanocomposites could further enhance this green material's performance for flexible microsupercapacitor applications.

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Figure legends

Fig. 1. Schematic illustration of the procedure to synthesize OPL-LSG electrodes.

Fig. 2. FESEM images of OPL-LSG 7030 at low (a) and high (b) magnification. Cross sectional view of OPL-LSG 7030 (c). FESEM images of OPL-LSG 5050 at low (d) and high (e) magnification. Cross sectional view of OPL-LSG 5050 (f). TEM images of OPL-LSG 7030 at low (g) and high (h) magnification. TEM OPL-LSG 7030 with lattice fringes (The inset represents SAED pattern) (I).

Fig. 3. Sheet resistance of OPL-LSG XY (a), Raman Spectra of OPL-LSG at different laser power (b), and speed (c). Correlations of the ID/IG and IG/I2D obtained at different laser power (d) and speed (e). FTIR of OPL-LSG 7030 and P-LSG 7030 (f).

Fig. 4. N2 adsorption–desorption curves (a) and Pore size distribution (Inset: amplifying pore size distribution) (b), for OPL-LSG 7030. XPS survey of lignin and OPL-LSG 7030 (c), C 1s (d) and O 1s (e) of OPL-LSG 7030. XRD patterns of OPL-LSG 7030 (f).

Figure 5 CV curves of; OPL-LSG XY at a scan rate of $50mVs^{-1}$ (a) and OPL-LSG 7030 at various scan rates (b). Dependence of OPL-LSG XY capacitance at different scan rates (c). GCD curves of; OPL-LSG XY at a current density of $0.1mAcm^{-2}$ (d) and OPL-LSG 7030 at various current densities (e). Correlation of OPL-LSG XY capacitance at different current densities (f).

Figure 6 Nyquist plots of OPL-LSG XY (Inset: close up image of Nyquist plots and EIS equitant fitting circuit) (a). Cyclic stability of OPL-LSG 7030 (b). Correlation of energy density and power density of OPL-LSG 7030 (c). CV curves of OPL-LSG7030 at different bending cycles at $50mVs^{-1}$ (d), Capacitance retention as a function of bending cycle (Inset: Image of fabricated electrode during bending cycle) (e). Ragone plot for comparison of energy density and power density of OPL-LSG 7030 with previously reported supercapacitors (f).