IEEE Access
Multidisciplinary | Rapid Review | Open Access Journal

Received October 26, 2021, accepted November 5, 2021, date of publication November 15, 2021, date of current version November 24, 2021.

Digital Object Identifier 10.1109/ACCESS.2021.3128320

Laser Scribed Graphene-Based Flexible Microsupercapacitors With Fractal Design

AASHIR WAHEED SYED AND MOHAMMAD ALI MOHAMMAD
School of Chemical and Materials Engineering, National University of Sciences and Technology, Islamabad 44000, Pakistan
Corresponding author: Mohammad Ali Mohammad (dr.ali@nust.edu.pk)

This work was supported by the National University of Sciences and Technology (NUST) Internal Research Funding.

ABSTRACT In recent times, there has been a great increase in the demand of flexible micro-supercapacitors for use in many commercial applications. Flexible capacitors have the potential to replace conventional batteries as they can provide competitive energy storage while being much smaller in size and having greater lifetime than average lithium-ion batteries. The most used design for in-plane flexible micro-supercapacitors is the interdigitated design. However, from findings of various recent studies, it is believed that the capacitance of in-plane EDLC micro-supercapacitors can further be improved by changing electrode designs. Different fractal designs have shown great promise to enhance the electrochemical performance of in-plane micro-supercapacitors by maximizing the active surface area and minimizing the energy lost during ion-transport. In this work, we successfully fabricate graphene-based, laser-scribed flexible micro-supercapacitors and study the effects of change in geometry parameters on total capacitance. Four different geometries for electrode were designed i.e., (1) interdigitated design, (2) fractal F1 (Hilbert) design, (3) fractal F2 (Peano) design, and (4) fractal F3 (Moore) design, and were patterned onto the thin layer of graphene oxide using laser scribing technique. Detailed analysis was presented for the change in capacitance using cyclic voltammetry, galvanic charge-discharge, electrochemical impedance spectroscopy and electrostatic simulation using COMSOL Multiphysics. It was found that fractal design micro-supercapacitors give better performance with higher capacitance and energy density values in comparison to conventional design interdigitated micro-supercapacitors. Among all, Fractal F3 design showed the highest capacitance and energy density value in comparison to other devices. It was found that there were two factors affecting the performance of devices, i.e., (1) a higher active surface area for fractal designs as compared to interdigitated designs with the same unit area, and (2) in addition to electric double layer capacitance, there was also electric field being generated on the edges of electrodes, defined as edging effect. Among both factors, the latter was the major enhancer of capacitance in the fractal design devices.

INDEX TERMS Fractal, electric double layer capacitor, flexible electronics, graphene, laser scribing, COMSOL multiphysics, micro-supercapacitor.

I. INTRODUCTION

In the recent times, there has been an increase in demand for small flexible electronic devices, such as microrobots, implantable medical devices and wearable sensors, that require an efficient and compact device solution for energy supply and storage [1], [2]. Solutions like 3D micro-batteries and thin films have been proposed, but because of their short life cycle and poor rate performance, they have failed to achieve the requirements [3]. In comparison, micro-supercapacitors with their long-life cycle, high power density and charge-discharge rate have shown promise for such applications [4]. Moreover, micro-supercapacitors can draw the maximum current that a source can apply and charge most efficiently, irrespective of the voltage. The aforementioned properties, therefore, make supercapacitors the most efficient solution for self-powered system applications [5]. In terms of charge storage mechanisms, there are two main types for supercapacitors: (1) electric double layer capacitor (EDLC) and (2) pseudocapacitor [6]. In EDLC, charge is stored electro-statistically (non-Faradically) i.e., charges don't shift between electrolyte and electrode like conventional capacitors rather they are collected at electrolyte-electrode interface. In contrast, pseudocapacitors

The associate editor coordinating the review of this manuscript and approving it for publication was Jesus Felez.

store charge through fast and reversible oxidation-reduction reactions, electrosorption and intercalation mechanisms on the surface of electrode (Faradic process) [7]. EDLCs are known to have high power density, whereas pseudocapacitors are usually known for higher energy density than EDLCs [8].

Many materials with good porosity and high surface area like MXenes, metal-organic frameworks (MOFs), graphene etc. have been under study for energy applications [9]–[11]. Among them, graphene, a two-dimensional one-atom-thick single layer of sp^2-bonded carbon is considered to have a great potential for energy storage [12]–[15]. It can produce supercapacitors with great power because of its high electrical conductivity and theoretical surface area (2630 m^2g^{-1}). Graphene-based sandwiched supercapacitors were initially fabricated by many researchers and their results showed a high-power performance with better frequency response, [12], [14], [16]–[18] but because of their structure, their integration into electronics systems was limited. Yoo *et al.* [19] was the first to study and report the increase in specific capacitance of in-plane graphene-based supercapacitors. They also engineered a method for direct pattering of hydrated graphene oxide (GO) films, to fabricate in-plane micro-supercapacitors using laser reduction [20]. After this discovery, they used an interesting technique to fabricate supercapacitors while using hydrated GO as both separator and electrolyte. Even though this was a promising technique, the high internal resistance and poor frequency response caused this device to fail for practical applications. While keeping all these problems into consideration, El-Kady *et al.* [21] fabricated interdigitated flexible in-plane graphene-based micro-supercapacitors using a standard market-grade LightScribe DVD Burner. They developed micro-supercapacitors on flexible polyethylene terephthalate (PET) substrate. They were able to fabricate more than 100 micro-supercapacitors on a single disc in less than 30 minutes with volumetric power density of 200 Wcm^{-3}. This study revolutionized the approach and study for in-plane graphene-based micro-supercapacitors. All the fabricated devices were highly flexible and exceptionally thin.

Micro-supercapacitors are generally fabricated using photolithography [22]–[25]. However, the above mentioned, facile and inexpensive technique of laser scribing has shown great promise for preparation of high-performance micro-supercapacitors on flexible substrates [26]–[28]. Light scribing is a direct writing technique that is used to burn graphics and text onto dye coated DVD or CD surface [26]. El-Kady *et al.* were the first who, instead of printing on special dye, coated the disk with GO film and reduced it by direct laser writing onto the surface. As a result, they discovered an unusual photothermal effect that converted GO to laser scribed graphene (LSG) [28], [30], [31].

Commonly known geometries for ELDCs are sandwiched and in-plane interdigitated electrodes (IDE). Interdigitated micro-supercapacitors show higher operating voltage, higher power density, longer lifetime, and faster charge/discharge in comparison with sandwiched structure micro-supercapacitor devices. Although interdigitated micro-supercapacitors show better performance than stacked geometry, some theoretical studies suggest even further improvement in performance of micro-supercapacitors by change in design.

Recently, Pei *et al.* [32] fabricated nanosupercapacitors with fractal designs using focused ion beam and studied their improvement in performance, based on electrochemical testing and simulation, in comparison to conventional interdigitated supercapacitors. The basic concept of fractal applies to any object that has fragmented or self-similar form [33], [34]. From lines to loops, numerous topologies of fractal designs are available, that can be modified for electronic applications. Generally, a fractal design is created by initially designing a unit cell, using an algorithm and then they are repeated many times in different directions, orientations, and scales to generate the resulting structure [35], [36]. Based on the possible superior performance of fractal designs, recently many researchers have started working on fractal design electrodes for various applications. Different research groups worked on RF-MEMS capacitors [36], [37], stretchable electronics [38], biosensors [39], optoelectronics devices [40], [41], high density CMOS capacitors [42], neural simulation [43], and in characterization of complex irregular surfaces [44] with fractal designs. Theoretical studies have also supported that fractal designs maximize the electrochemically active surface area and enhance electrochemical system performance, whereas losing minimum energy during ion transport through fractal design network [34]. Also, an increase in area can be accomplished while using fractal design electrodes, that may cause increase in overall capacitance [45]–[48].

In this paper we systematically studied multiple fractal designs to improve the performance of micro-supercapacitors. Three different fractal designs were simulated, fabricated, and their electrochemical performance was compared with conventional interdigitated design micro-supercapacitors. The fractal designs that were used by us are (1) fractal Hilbert (F1) [49], fractal Peano (F2) [50], and fractal Moore (F3) [51]. The objective of this study was to fabricate graphene-based, flexible micro-supercapacitor devices with fractal designs and compare their electrochemical performance with interdigitated electrode designs. We also used COMSOL Multiphysics to perform an electrostatic study and investigate the behavior of an electric field around the edges of micro-supercapacitor electrodes.

II. FABRICATION

Graphene was synthesized using modified Hummers method [52] and polymer gel electrolyte, H_3PO_4/PVA, was synthesized according to literature [53] (discussed in supporting information).

Structure for micro-supercapacitors consisted of four different designs, each of which had same electrode width and gap between them. The structure designs shown in Figure 1 (a-d) were all first drawn on AutoCAD with electrode width of 400μm and a gap of 185μm between them. After

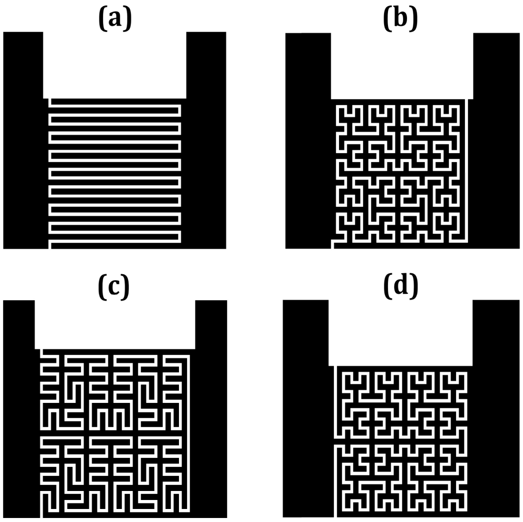

FIGURE 1. Computer generated designs of (a) IDE, (b) F1 (Hilbert), (c) F2 (Peano) and (d) F3 (Moore) designs for supercapacitors.

designing, all these patterns were converted into. BMP files to allow importing of images into the laser scribing software.

For the patterning of laser scribed graphene, a LightScribe DVD drive was used to pattern the desired designs on GO films. Initially, the dispersed solution of synthesized GO was prepared with the concentration of 5 mg ml^{-1} in DI water. A Kapton polyimide film (0.25mm) was attached using adhesive spray on the surface of LightScribe DVD disc. Dispersed GO solution was then drop-casted on Kapton polyimide film and then left to dry for almost 24 hours under ambient conditions. After complete drying, the disc was inserted into the drive for laser scribing of GO film. All four different patterns were inserted separately into laser scribing software. All four patterns were laser scribed on to GO film, converting insulating golden-brown GO into highly conductive black laser scribed graphene. It took about 20 minutes for each pattern to be patterned precisely on to the GO film. In this case, laser scribed graphene serves as both current collector and active electrode, so laser scribed graphene pattern is used directly as a micro-supercapacitor.

After completion of the patterning process, active electrode pattern area is defined using Kapton (polyimide) tape to avoid contact of electrolyte with current collector area. The protected pattern is then coated with electrolyte. For electrochemical testing of the device, copper tape was applied to current collector edges to ensure a good contact between micro-supercapacitor and electrochemical workstation. To improve the contact, silver paste was applied at the edge of the copper tape in a way that it covers half on copper tape and half on current collector, working as a contact between the two. Then micro-supercapacitor device is left under ambient conditions to allow electrolyte to fully access all the parts of the electrodes and also evaporate any amount of excess water. After drying, the resulting device is a fully functional micro-supercapacitor and can be characterized and tested for any application. Figure 2 shows the schematic diagram the fabrication process. Figure 3 shows snapshots of the actual fabricated micro-supercapacitor devices.

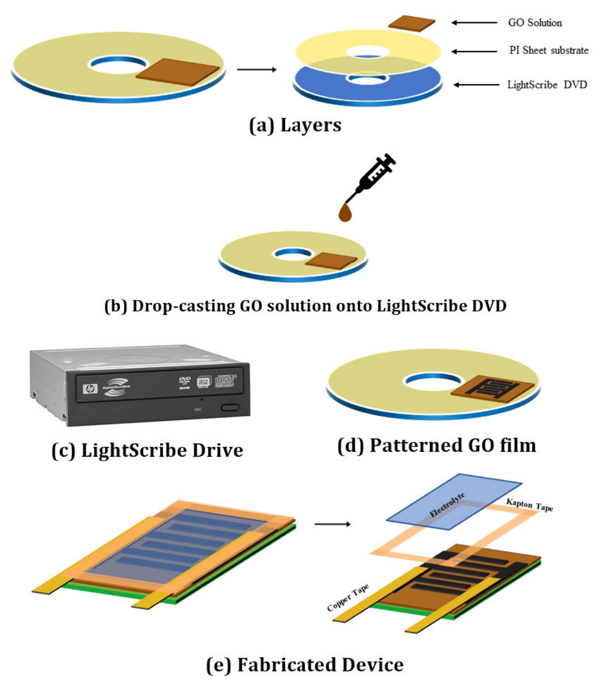

FIGURE 2. Step-by-step fabrication process of micro-supercapacitor devices.

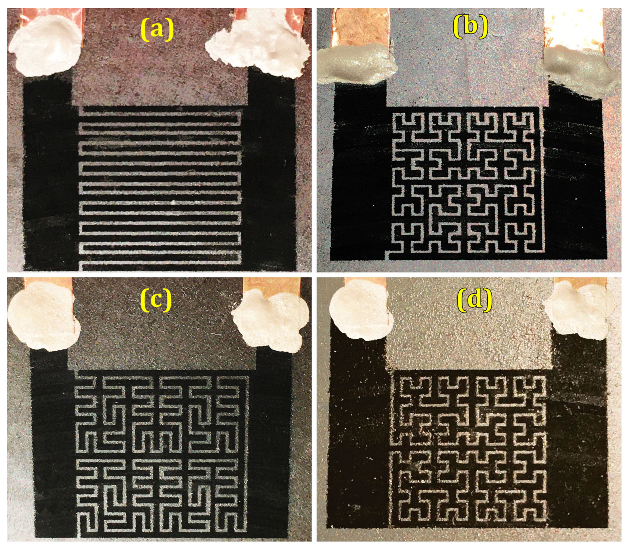

FIGURE 3. Fabricated (a) IDE, (b) F1, (c) F2 and (d) F3 design micro-supercapacitor devices.

III. RESULTS & DISCUSSIONS

Before the fabrication of micro-supercapacitor devices, graphene as a material was characterized to study the changes from GO to LSG after reduction. Characterization techniques of X-ray Diffraction (XRD), Raman Spectroscopy, Scanning Electron Microscopy (SEM), and Optical Profilometry were used for this study. Fourier Transform Infrared Spectroscopy (FTIR) was used to characterize the synthesized electrolyte (H_3PO_4). These results are discussed in supporting information (Figure S1-S5).

Next, micro-supercapacitors of all four different designs were electrochemically characterized using Cyclic Voltammetry (CV), Galvanic charge-discharge (GCD), and Electrochemical impedance spectroscopy (EIS).

Figure 4 (a-d) shows Cyclic voltammetry (CV) curves for all four (IDE, F1, F2, F3) micro-supercapacitors at four different scan rates (10 mV s^{-1}, 20 mV s^{-1}, 50 mV s^{-1}, 100 mV s^{-1}). Figure S9 - S12 shows CV curves of all four micro-supercapacitor devices at all various scan rates (5 mV s^{-1}, 10 mV s^{-1}, 20 mV s^{-1}, 30 mV s^{-1}, 40 mV s^{-1}, 50 mV s^{-1}, 100 mV s^{-1}, 200 mV s^{-1}, 400 mV s^{-1}, 600 mV s^{-1}, 800 mV s^{-1}, 1000 mV s^{-1}). The CV was performed at a potential window of 0 to 1 V. Cyclic voltammogram shows rectangular (more like lens-shaped) characteristics with the lack of any oxidation or reduction peaks. This behavior clearly indicates electrochemical double layer capacitor (EDLC) behavior micro-supercapacitors. The value of current density in response to the applied potential difference increases with the scan rate. Figure S6 (Supporting information) shows the comparison of CV curves at scan rate of 1000 mV s^{-1}. It is observed from the graphs that current density of fractal electrode design micro-supercapacitors is a bit higher than IDE electrode design micro-supercapacitor and fractal design CV graph also show more area under the curves, indicating higher capacitance value for fractal design supercapacitors.

After studying the capacitive nature of the device through cyclic voltammogram, we characterized with galvanic charge-discharge (GCD) testing for further study. Figure 5 (a-d) shows GCD curves for all designs of micro-supercapacitors at four different current densities (3 μA.cm^{-2}, 5 μA.cm^{-2}, 7 μA.cm^{-2}, 9 μA.cm^{-2}). Figure S13 - S16 shows GCD curves of all four micro-supercapacitor devices at all various current densities (3 μA.cm^{-2}, 4 μA.cm^{-2}, 5 μA.cm^{-2}, 6 μA.cm^{-2}, 7 μA.cm^{-2}, 8 μA.cm^{-2}, 9 μA.cm^{-2}, 10 μA.cm^{-2}, 11 μA.cm^{-2}, 12 μA.cm^{-2}, 13 μA.cm^{-2}). GCD results show triangular curves, confirming the EDLC behavior. The voltage drop observed at start of each discharge curve, also known as iR drop, is the measure of overall resistance of the device. Discharge curve and iR drop value are in direct proportion to each other. Small iR drop is observed in the galvanic discharge curves for all devices, indicating low resistance for tested micro-supercapacitor devices which confirms a good power density of devices. Figure S7 (Supporting information) shows the comparison of GCD curves at current density of 3 μA.cm^{-2}. It is also observed that there is a significant increase in discharge time for fractal design micro-supercapacitors, indicating higher capacitance value.

Electrochemical impedance spectroscopy (EIS) was performed to analyze the internal resistance of micro-supercapacitor devices. Testing frequency range was set from 100 mHz to 1 MHz. The Nyquist plots calculated from EIS are shown in Figure S8 (Supporting Information) from all four micro-supercapacitors. The equivalent series resistance calculated from all different micro-supercapacitor Nyquist plots ranged between 2Ω and 7Ω, which is a low resistance value and indicative of a good ion diffusion. LSG micro-supercapacitors are known to have good frequency response with very little relaxation time [21].

Flexibility of electronic devices is an important aspect because of their potential applications in wearable electronics, roll-up displays, smart sensors etc. F3 design micro-supercapacitor device was electrochemically tested for CV and GCD curves, while bending the device at 30° angle. The results showed similar behavior as in comparison to behavior of F3 device at 0°. Capacitance of the device was also calculated from GCD curves and results showed the same value at 30° as it showed on 0° (Figure S17 shows CV and GCD graph of bent F3 device at 30° angle). The cyclic stability was evaluated using F3 design micro-supercapacitor (as shown in Figure 6) by continuous charge discharge for 5000 cycles. Results showed 96% retention of initial capacitance value after 5000 cycles.

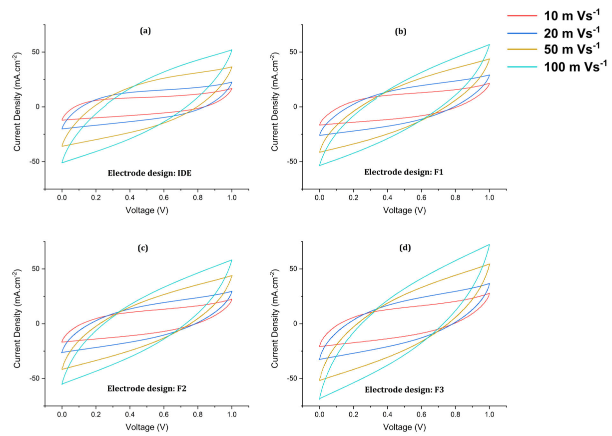

FIGURE 4. Cyclic voltammetry of all four micro-supercapacitor electrode designs. (a) IDE, (b) F1, (c) F2, and (d) F3 electrode design.

A. COMPARISON

After a detailed characterization and study of devices, we have observed mannerism of improvement in performance of the devices. Areal capacitance of all micro-supercapacitors was calculated from GCD curves as discussed in the Supporting Information. Figure 7 shows the comparison of areal capacitance calculated from charge discharge curves. It is observed that areal capacitance increases with decreasing applied current density. Another thing that can be seen clearly from the graph is that fractal designs have higher areal capacitance value than IDE design. The maximum capacitance value obtained was $\approx$1.7 mF.cm^{-2} at 3μA from F3 fractal design micro-supercapacitor, whereas maximum capacitance obtained from IDE design micro-supercapacitor was $\approx$0.8 mF.cm^{-2} at the same value of current density. The calculated capacitance value for F3 fractal design is more than twice the value of capacitance calculated from IDE design micro-supercapacitor.

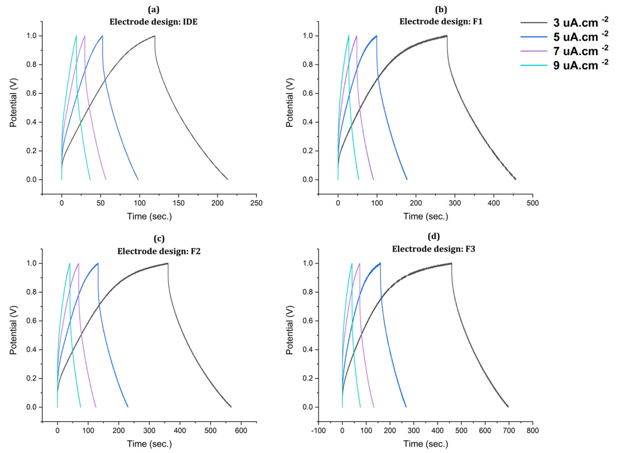

FIGURE 5. Galvanic charge-discharge of all four micro-supercapacitor electrode designs. (a) IDE, (b) F1, (c) F2, and (d) F3 electrode design.

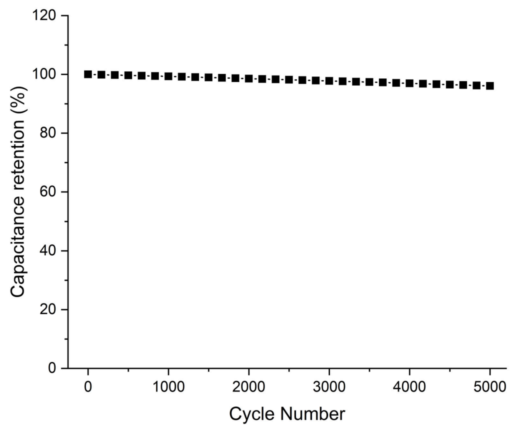

FIGURE 6. The capacitive retention (stability) of F3 design micro-supercapacitor at 5000 cycles.

Energy and power density for all micro-supercapacitors was calculated as discussed in the Supporting Information. Figure S8 (supporting information) shows Ragone plot for comparison of all micro-supercapacitors. A similar mannerism is observed that fractal design micro-supercapacitors show higher energy density than IDE design micro-supercapacitor at same value of power density. It is also worth noting that the energy densities decrease gradually as the power density is increased.

B. DISCUSSION

After these results and observations, it is very important to understand that why fractal design micro-supercapacitor devices were able to perform better than conventional IDE design supercapacitor and what are the factors that affect such improvement. Evaluating the factors, the first thing noted is the area of the device. The material and electrolyte used were the same for all the micro-supercapacitor devices and the distance between the electrodes and electrode width were also kept constant while designing the patterns. When the area of different devices was calculated, F3 design area was 71.4560 mm^2 whereas IDE design area was 71.3643 mm^2.

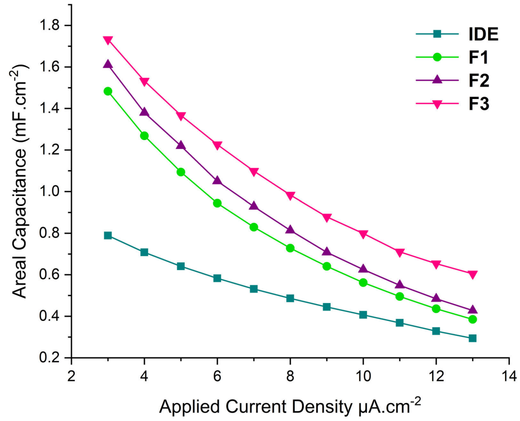

FIGURE 7. Comparison of areal capacitance of all four designs of micro-supercapacitor.

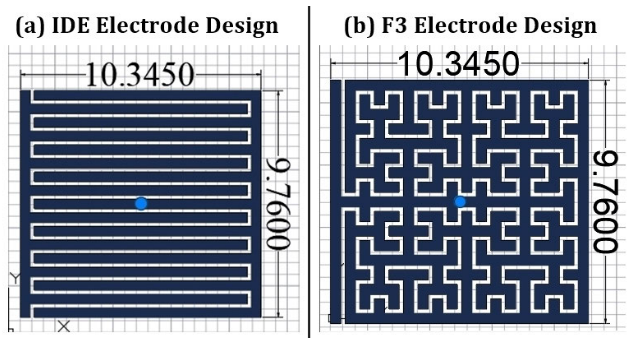

FIGURE 8. Electrode area comparison of (a) IDE and (b) F3 design micro-supercapacitors.

Results showed that F3 electrode design have 0.13% more area of electrode than IDE electrode design for the same unit base area, which is not much, but one of the factors for fractal design micro-supercapacitor's better performance (as shown in Figure 8).

The other factor that seemed to be improving capacitance of fractal design micro-supercapacitor over IDE design is the edging effect. Edging effect is a well-known phenomenon for IDE design capacitors. In comparison with any round-shaped or continuous electrode, an edge-intensive electrode will have increased electric field lines of force or lateral electric flux, known as fringing fields. These fields will result in a greater accumulation of charge (electrolyte ions) on or near the electrodes, that will induce an enhanced electrostatic capacitance [4], [36], [42], [45], [54]–[56]. In case of fractal design electrodes, they have higher number of open and rough edges that leads to enhanced lateral electrical flux and more efficient accumulation of electrostatic charge because of their unique designs, causing better edging effect and increase in capacitance than IDE design electrodes (as shown in Figure 9) [32]. However, in terms of fractal design electrodes, the number of open edges depend upon its geometry and its order number. Fractal designs with different geometries have different number of edges, and fractal designs with same geometry but different order number will have different count of edges, hence explaining different capacitance values for different fractal design micro-supercapacitors.

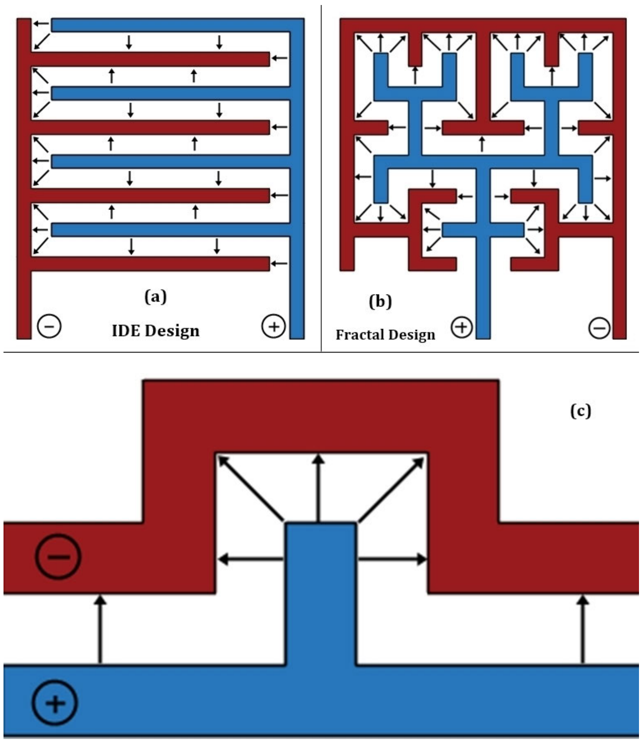

FIGURE 9. Schematic comparison of edging effect of (a) IDE design and (b) fractal design micro-supercapacitor. (c) Electrical lines of force on edge in fractal design.

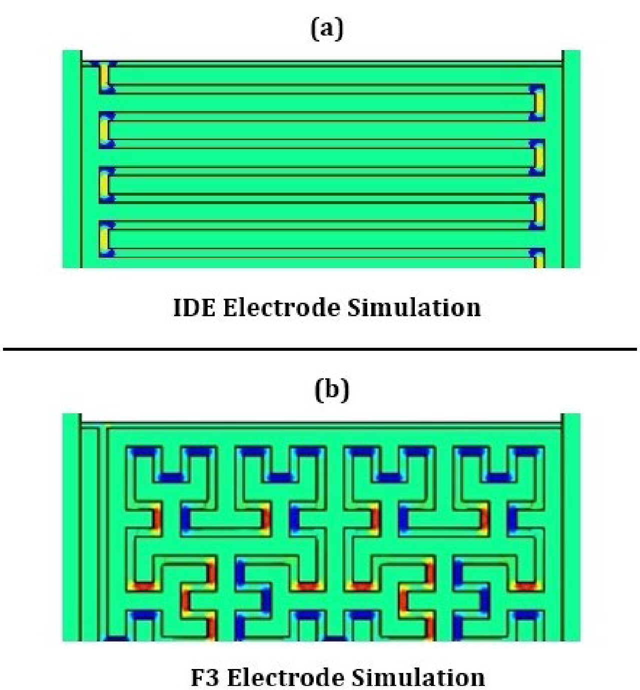

FIGURE 10. Comparison of electric field of (a) IDE design and (b) fractal design micro-supercapacitor obtained from COMSOL multiphysics simulation.

Figure 10 shows distribution of electric field in IDE design micro-supercapacitor and fractal design F3 micro-supercapacitor. COMSOL Multiphysics simulation tool was used to get these images. It is very clear from the images

TABLE 1. Comparison of our work with similar previous work.

Electrode Material	Electrolyte	Capacitance	Ref.
Graphite	SiWA/H_3PO_4 /PVA	1 mF cm^{-2} at 1V s^{-1}	[58]
rGO	1.0 M Na_2SO_4 / 1.0 M $TEABF_4$	0.51 mF cm^{-2} at 40 mV s^{-1}	[20]
CNT-graphene hybrid fibers	1M Na_2SO_4 Aqueous Solution	1.3 mF cm^{-2} at 10µA cm^{-2}	[59]
LSG	H_2SO_4/PVA gel	1.35-2.32 mF cm^{-2} at 1.68 x 10^4 mA cm^{-3}	[21]
LSG	H_3PO_4/ PVA gel	0.8 mF cm^{-2} at 10mV s^{-1}	[27]
rGO/Au nanoparticle	H_2SO_4/PVA gel	0.77 mF cm^{-2} at 1V s^{-1}	[60]
LSG-PANI	H_2SO_4/PVA gel	0.8 mF cm^{-2} at 0.5mA cm^{-2}	[61]
LSG	[BMIM] [NTf2] / Silica powder	0.181 mF cm^{-2} at 5mA cm^{-2}	[15]
LSG	H_3PO_4 /PVA	1.7 mF cm^{-2} at 3µA cm^{-2}	This Work

that strength of electric field increases near the edges in both electrode designs and since the fractal design has more open edges, so they collect more charge and hence it gives a higher capacitance.

Table 1 shows the comparison of our work with similar research done in the past. The capacitance value achieved by F3 design micro-supercapacitor device was found to be much higher, in comparison to the research work done on graphene-based supercapacitors in the past. Beside the works referenced in the table, we would also like to call the attention of the community towards the paper by Thekkekara *et al.* [48] in which they have demonstrated 300 times increase in capacitance compared to IDE structure. This paper does not seem to be in line with other papers comparing fractal and non-fractal structures and since the trend may be questionable, it is not included in the comparison.

IV. CONCLUSION

In this work, we successfully fabricated laser scribed flexible micro-supercapacitors and studied the effects of change in geometry parameters on the total capacitance. Four supercapacitors, i.e., one conventional interdigitated and three fractal designs, were fabricated and studied using electrochemical testing. We found that fractal design supercapacitors show better performance in comparison to conventional IDE design micro-supercapacitors. Fractal design micro-supercapacitors showed better areal capacitance and higher energy density than IDE design micro-supercapacitor. Among all, F3 design showed the highest capacitance value of $\approx$1.7 mF.cm^{-2} which is more than twice the value of capacitance of IDE design micro-supercapacitor (i.e., $\approx$0.8 mF.cm^{-2}). We also discovered that fractal design micro-supercapacitors had a higher active surface area. F3 design showed 0.13% increased electrode area in comparison to IDE design. Even though it caused an improvement in performance of fractal design micro-supercapacitors, but with this small variation, its contribution to the cause was not as much, in comparison to increase in capacitance value. To investigate it further, we studied the effect of fringing fields in the literature and then COMSOL Multiphysics simulation was also performed. The simulations helped to verify the "edging effect" and how the force of electric field lines along the edges of in-plane supercapacitors improve and how their impact increase further in case of high order fractal design supercapacitors, that results in higher capacitance for fractal designs.

ACKNOWLEDGMENT

The authors wish to acknowledge Farheen Nasir (currently at the University of Waterloo) for all the useful discussions and guidance with electrolyte synthesis, simulations, and electrochemical calculations. They also thank Sabeen Fatima and Dr. Syed Rizwan from the National University of Sciences and Technology (NUST) for their support and guidance.

REFERENCES

[1] B. Wang, M. Ahmed, B. Wood, and F. Iacopi, "All-solid-state supercapacitors on silicon using graphene from silicon carbide," *Appl. Phys. Lett.*, vol. 108, no. 18, May 2016, Art. no. 183903.

[2] J. Li, V. Mishukova, and M. Östling, "All-solid-state micro-supercapacitors based on inkjet printed graphene electrodes," *Appl. Phys. Lett.*, vol. 109, no. 12, Sep. 2016, Art. no. 123901.

[3] Q. Jiang, N. Kurra, C. Xia, and H. N. Alshareef, "Hybrid microsupercapacitors with vertically scaled 3D current collectors fabricated using a simple cut-and-transfer strategy," *Adv. Energy Mater.*, vol. 7, no. 1, Jan. 2017, Art. no. 1601257.

[4] X. Tian, M. Shi, X. Xu, M. Yan, L. Xu, A. Minhas-Khan, C. Han, L. He, and L. Mai, "Arbitrary shape engineerable spiral micropseudocapacitors with ultrahigh energy and power densities," *Adv. Mater.*, vol. 27, no. 45, pp. 7476–7482, 2015.

[5] N. Kurra, B. Ahmed, Y. Gogotsi, and H. N. Alshareef, "MXene-on-paper coplanar microsupercapacitors," *Adv. Energy Mater.*, vol. 6, no. 24, Dec. 2016, Art. no. 1601372.

[6] E. Frackowiak and F. Béguin, "Carbon materials for the electrochemical storage of energy in capacitors," *Carbon*, vol. 39, no. 6, pp. 937–950, 2001.

[7] Z. Niu, L. Zhang, L. Liu, B. Zhu, H. Dong, and X. Chen, "All-solid-state flexible ultrathin micro-supercapacitors based on graphene," *Adv. Mater.*, vol. 25, no. 29, pp. 4035–4042, 2013.

[8] X. Zhang, X. Liu, S. Zeng, J. Fang, C. Men, X. Zhang, and Q. Li, "Reducing and uniforming the Co_3O_4 particle size by sulfonated graphenal polymers for electrochemical applications," *Nanosc. Res. Lett.*, vol. 12, no. 1, p. 165, Dec. 2017.

[9] X. Li, X. Yang, H. Xue, H. Pang, and Q. Xu, "Metal–organic frameworks as a platform for clean energy applications," *EnergyChem*, vol. 2, no. 2, May 2020, Art. no. 100027.

[10] K.-B. Wang, Q. Xun, and Q. Zhang, "Recent progress in metal-organic frameworks as active materials for supercapacitors," *EnergyChem*, vol. 2, no. 1, Jan. 2020, Art. no. 100025.

[11] Z. Liang, R. Zhao, T. Qiu, R. Zou, and Q. Xu, "Metal-organic framework-derived materials for electrochemical energy applications," *EnergyChem*, vol. 1, no. 1, Jul. 2019, Art. no. 100001.

[12] M. F. El-Kady, V. Strong, S. Dubin, and R. B. Kaner, "Laser scribing of high-performance and flexible graphene-based electrochemical capacitors," *Science*, vol. 335, no. 6074, p. 1326, 2012.

[13] Y. Zhu, S. Murali, M. D. Stoller, K. J. Ganesh, W. Cai, P. J. Ferreira, A. Pirkle, R. M. Wallace, K. A. Cychosz, M. Thommes, D. Su, E. A. Stach, and R. S. Ruoff, "Carbon-based supercapacitors produced by activation of graphene," *Science*, vol. 332, no. 6037, p. 1537, 2011.

[14] J. R. Miller, R. A. Outlaw, and B. C. Holloway, "Graphene double-layer capacitor with AC line-filtering performance," *Science*, vol. 329, no. 5999, p. 1637, 2010.

[15] L. V. Thekkekara, X. Chen, and M. Gu, "Two-photon-induced stretchable graphene supercapacitors," *Sci. Rep.*, vol. 8, no. 1, p. 11722, Dec. 2018.

[16] J. R. Miller, R. A. Outlaw, and B. C. Holloway, "Graphene electric double layer capacitor with ultra-high-power performance," *Electrochim. Acta*, vol. 56, no. 28, pp. 10443–10449, 2011.

[17] K. Sheng, Y. Sun, C. Li, W. Yuan, and G. Shi, "Ultrahigh-rate supercapacitors based on eletrochemically reduced graphene oxide for AC line-filtering," *Sci. Rep.*, vol. 2, no. 1, p. 247, Dec. 2012.

[18] T. Malik, S. Naveed, M. Muneer, and M. A. Mohammad, "Fabrication and characterization of laser scribed supercapacitor based on polyimide for energy storage," *Key Eng. Mater.*, vol. 778, pp. 181–186, Sep. 2018.

[19] J. J. Yoo, K. Balakrishnan, J. Huang, V. Meunier, B. G. Sumpter, A. Srivastava, M. Conway, A. L. M. Reddy, J. Yu, R. Vajtai, and P. M. Ajayan, "Ultrathin planar graphene supercapacitors," *Nano Lett.*, vol. 11, no. 4, pp. 1423–1427, Apr. 2011.

[20] W. Gao, N. Singh, L. Song, Z. Liu, A. L. M. Reddy, L. Ci, R. Vajtai, Q. Zhang, B. Wei, and P. M. Ajayan, "Direct laser writing of micro-supercapacitors on hydrated graphite oxide films," *Nature Nanotechnol.*, vol. 6, no. 8, pp. 496–500, 2011.

[21] M. F. El-Kady and R. B. Kaner, "Scalable fabrication of high-power graphene micro-supercapacitors for flexible and on-chip energy storage," *Nature Commun.*, vol. 4, no. 1, p. 1475, Jun. 2013.

[22] D. Pech, M. Brunet, H. Durou, P. Huang, V. Mochalin, Y. Gogotsi, P.-L. Taberna, and P. Simon, "Ultrahigh-power micrometre-sized supercapacitors based on onion-like carbon," *Nature Nanotechnol.*, vol. 5, no. 9, pp. 651–654, 2010.

[23] M. Beidaghi and C. Wang, "Micro-supercapacitors based on interdigital electrodes of reduced graphene oxide and carbon nanotube composites with ultrahigh power handling performance," *Adv. Funct. Mater.*, vol. 22, no. 21, pp. 4501–4510, 2012.

[24] M. S. Kim, B. Hsia, C. Carraro, and R. Maboudian, "Flexible micro-supercapacitors with high energy density from simple transfer of photoresist-derived porous carbon electrodes," *Carbon*, vol. 74, pp. 163–169, Aug. 2014.

[25] J. Lin, C. Zhang, Z. Yan, Y. Zhu, Z. Peng, R. H. Hauge, D. Natelson, and J. M. Tour, "3-dimensional graphene carbon nanotube carpet-based microsupercapacitors with high electrochemical performance," *Nano Lett.*, vol. 13, no. 1, pp. 72–78, 2013.

[26] G. Wang, V. Babaahmadi, N. He, Y. Liu, Q. Pan, M. Montazer, and W. Gao, "Wearable supercapacitors on polyethylene terephthalate fabrics with good wash fastness and high flexibility," *J. Power Sources*, vol. 367, pp. 34–41, Nov. 2017.

[27] J. B. In, B. Hsia, J.-H. Yoo, S. Hyun, C. Carraro, R. Maboudian, and C. P. Grigoropoulos, "Facile fabrication of flexible all solid-state micro-supercapacitor by direct laser writing of porous carbon in polyimide," *Carbon*, vol. 83, pp. 144–151, Mar. 2015.

[28] V. Strong, S. Dubin, M. F. El-Kady, A. Lech, Y. Wang, B. H. Weiller, and R. B. Kaner, "Patterning and electronic tuning of laser scribed graphene for flexible all-carbon devices," *ACS Nano*, vol. 6, no. 2, pp. 1395–1403, 2012.

[29] D. G. Stinson and M. Maguire, "LightScribe direct disc labeling," in *Proc. Int. Symp. Opt. Memory Opt. Data Storage*. Honolulu, HI, USA: Optical Society of America, 2005, doi: 10.1364/ISOM_ODS.2005.MA4.

[30] S. Gilje, S. Dubin, A. Badakhshan, J. Farrar, S. A. Danczyk, and R. B. Kaner, "Photothermal deoxygenation of graphene oxide for patterning and distributed ignition applications," *Adv. Mater.*, vol. 22, no. 3, pp. 419–423, 2010.

[31] L. J. Cote, R. Cruz-Silva, and J. Huang, "Flash reduction and patterning of graphite oxide and its polymer composite," *J. Amer. Chem. Soc.*, vol. 131, no. 31, pp. 11027–11032, 2009.

[32] L. Pei, P. Zhuang, Y. Sun, X. Zhang, H. Qiao, K. Zhu, P. M. Ajayan, M. Ye, and J. Shen, "Nanosupercapacitors with fractal structures: Searching designs to push the limit," *J. Mater. Chem. A*, vol. 9, no. 32, pp. 17400–17414, 2021.

[33] L. Nottale and J. Schneider, "Fractals and nonstandard analysis," *J. Math. Phys.*, vol. 25, no. 5, pp. 1296–1300, 1984.

[34] B. B. Mandelbrot, "Les objets fractals: Forme, hasard et dimension," in *Nouvelle Bibliotheque Scientifique*. Paris, France: Flammarion, 1975.

[35] H. Jürgens, H.-O. Peitgen, and D. Saupe, "The language of fractals," *Sci. Amer.*, vol. 263, no. 2, pp. 60–67, 1990.

[36] A. M. Elshurafa, A. G. Radwan, A. Emira, and K. N. Salama, "RF MEMS fractal capacitors with high self-resonant frequencies," *J. Microelectromech. Syst.*, vol. 21, no. 1, pp. 10–12, 2012.
[37] A. M. Elshurafa, P. H. Ho, M. H. Ouda, A. G. Radwan, and K. N. Salama, "Low-voltage puzzle-like fractal microelectromechanial system variable capacitor suppressing pull-in," *Micro Nano Lett.*, vol. 7, no. 9, pp. 965–969, Sep. 2012.
[38] J. A. Fan, W.-H. Yeo, Y. Su, Y. Hattori, W. Lee, S.-Y. Jung, Y. Zhang, Z. Liu, H. Cheng, L. Falgout, M. Bajema, T. Coleman, D. Gregoire, R. J. Larsen, Y. Huang, and J. A. Rogers, "Fractal design concepts for stretchable electronics," *Nature Commun.*, vol. 5, no. 1, p. 3266, May 2014.
[39] Y. Mashraei, S. Sivashankar, U. Buttner, and K. N. Salama, "Integration of fractal biosensor in a digital microfluidic platform," *IEEE Sensors J.*, vol. 16, no. 24, pp. 8775–8783, Dec. 2016.
[40] F. Afshinmanesh, A. G. Curto, K. M. Milaninia, N. F. Van Hulst, and M. L. Brongersma, "Transparent metallic fractal electrodes for semiconductor devices," *Nano Lett.*, vol. 14, no. 9, pp. 5068–5074, 2014.
[41] D. Zuani, T. Reindl, M. Rommel, B. Gompf, A. Berrier, and M. Dressel, "High-order Hilbert curves: Fractal structures with isotropic, tailorable optical properties," *ACS Photon.*, vol. 2, no. 12, pp. 1719–1724, 2015.
[42] H. Samavati, A. Hajimiri, A. R. Shahani, G. N. Nasserbakht, and T. H. Lee, "Fractal capacitors," *IEEE J. Solid-State Circuits*, vol. 33, no. 12, pp. 2035–2041, Dec. 1998.
[43] L. Golestanirad, B. Elahi, A. Molina, J. R. Mosig, C. Pollo, R. Chen, and S. J. Graham, "Analysis of fractal electrodes for efficient neural stimulation," *Frontiers Neuroeng.*, vol. 6, p. 3, Jul. 2013.
[44] B. B. Mandelbrot, "Stochastic models for the earth's relief, the shape and the fractal dimension of the coastlines, and the number-area rule for islands," *Proc. Nat. Acad. Sci. USA*, vol. 72, no. 10, pp. 3825–3828, 1975.
[45] R. Aparicio and A. Hajimiri, "Capacity limits and matching properties of integrated capacitors," *IEEE J. Solid-State Circuits*, vol. 37, no. 3, pp. 384–393, Mar. 2002.
[46] L. Yang, F. Yang, K. Li, W. Shen, T. Xu, X. Xu, Y. Zhou, M. Zhong, M. Zheng, and D. Wei, "Screen-printable and flexible in-plane microsupercapacitors with fractal electrode design," *Flexible Printed Electron.*, vol. 6, no. 2, Jun. 2021, Art. no. 025008.
[47] B. Barnes, O. Suleiman, J. Badjo, and K. S. Das, "Fractal-based electrolytic capacitor electrodes: Scaling behavior with respect to fractal order and complexity," 2018, *arXiv:1810.00221*.
[48] L. V. Thekkekara and M. Gu, "Bioinspired fractal electrodes for solar energy storages," *Sci. Rep.*, vol. 7, no. 1, p. 45585, May 2017.
[49] D. Hilbert, "Über die stetige abbildung einer Line auf ein flächenstück," *Math. Ann.*, vol. 38, no. 3, pp. 459–460, 1891.
[50] G. Peano, "Sur une courbe, qui remplit toute une aire plane," *Math. Ann.*, vol. 36, no. 1, pp. 157–160, 1890.
[51] G. A. Bliss, "The scientific work of Eliakim Hastings Moore," *Bull. Amer. Math. Soc.*, vol. 40, no. 7, pp. 501–514, 1934.
[52] N. I. Zaaba, K. L. Foo, U. Hashim, S. J. Tan, W.-W. Liu, and C. H. Voon, "Synthesis of graphene oxide using modified hummers method: Solvent influence," *Proc. Eng.*, vol. 184, pp. 469–477, Jan. 2017.
[53] S.-K. Kim, H.-J. Koo, A. Lee, and P. V. Braun, "Selective wetting-induced micro-electrode patterning for flexible micro-supercapacitors," *Adv. Mater.*, vol. 26, no. 30, pp. 5108–5112, 2014.
[54] F. Nasir and M. A. Mohammad, "Investigation of device dimensions on electric double layer microsupercapacitor performance and operating mechanism," *IEEE Access*, vol. 8, pp. 28367–28374, 2020.
[55] B. Y. Park, R. Zaouk, C. Wang, and M. J. Madou, "A case for fractal electrodes in electrochemical applications," *J. Electrochem. Soc.*, vol. 154, no. 2, p. P1, 2007.
[56] M. J. Kirkby, "The fractal geometry of nature," *Earth Surf. Processes Landforms*, vol. 8, no. 4, p. 406, 1983.
[57] H. Gao and K. Lian, "High rate all-solid electrochemical capacitors using proton conducting polymer electrolytes," *J. Power Sources*, vol. 196, no. 20, pp. 8855–8857, 2011.
[58] H. Cheng, Z. Dong, C. Hu, Y. Zhao, Y. Hu, L. Qu, N. Chen, and L. Dai, "Textile electrodes woven by carbon nanotube–graphene hybrid fibers for flexible electrochemical capacitors," *Nanoscale*, vol. 5, no. 8, pp. 3428–3434, 2013.
[59] R.-Z. Li, R. Peng, K. D. Kihm, S. Bai, D. Bridges, U. Tumuluri, Z. Wu, T. Zhang, G. Compagnini, Z. Feng, and A. Hu, "High-rate in-plane micro-supercapacitors scribed onto photo paper using *in situ* femtolaser-reduced graphene oxide/Au nanoparticle microelectrodes," *Energy Environ. Sci.*, vol. 9, no. 4, pp. 1458–1467, 2016.
[60] L. Li, J. Zhang, Z. Peng, Y. Li, C. Gao, Y. Ji, R. Ye, N. D. Kim, Q. Zhong, Y. Yang, and H. Fei, "High-performance pseudocapacitive microsuper-capacitors from laser-induced graphene," *Adv. Mater.*, vol. 28, no. 5, pp. 838–845, 2016.

AASHIR WAHEED SYED received the B.S. degree (Hons.) in electrical engineering (electronics) from Air University, Pakistan, in 2017, and the M.S. degree (Hons.) in nanoscience and engineering from the School of Chemical and Materials Engineering of the National University of Sciences and Technology, Pakistan, in 2021.

His research interests include supercapacitors, surface acoustic wave devices, MEMS sensors, sound emission devices, and flexible electronics.

MOHAMMAD ALI MOHAMMAD received the B.Sc. degree in electrical engineering and the Ph.D. degree in electrical and computer engineering from the University of Alberta, Edmonton, AB, Canada, in 2008 and 2013, respectively. He majored in micro systems and nanodevices with research focus on electron beam lithography. He worked with Tsinghua University, China, as a Postdoctoral Fellow, from 2013 to 2016, where his research interests included the fabrication and testing of resistive memory devices and surface acoustic wave devices. In 2016, he joined the National University of Sciences and Technology (NUST), Islamabad, Pakistan, a Faculty Member. He then served as the Director (Research) of NUST from 2018–2021 and is presently with the Special Technology Zones Authority (STZA) as the Director (policy, research, and market intelligence). He remains affiliated with NUST as an Adjunct Faculty and is interested in flexible electronics, microfabricated devices, and technology policy.