

REVIEW

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# Supercapacitors with alternating current line-filtering performance

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## Abstract

Alternating current (AC) line filters have been widely used to smooth the leftover AC ripples on direct current voltage. Currently available commercial aluminum electrolytic capacitors (AECs) are primarily used for this application. However, the bulky volume and low capacitance of AECs have become incompatible with the rapidly developed intelligent electronic devices and industry dynamics. Supercapacitors with high specific capacitance and AC line-filtering performance could become the next-generation candidates to replace AECs for smoothing leftover AC ripples. Thus, most conventional supercapacitors behave like a resistor and not a capacitor at 120 Hz mainly because complex pore structures of electrode materials prevent the diffusion of electrolyte ions. Various electrode materials have been reported to reveal supercapacitors with AC line-filtering performance; however, the balance of high specific capacitance and an excellent filtering efficiency is a prodigious challenge. This review summarizes recently reported supercapacitors based on different types of electrode materials with AC filtering performance and attempts to develop the relationship between different influencing factors and features of functional materials.

**Keywords:** Supercapacitor, Micro-supercapacitor, Alternating current line filtering, Phase angle, Electrode

## Introduction

Direct current (DC) and alternating current (AC) voltages are used for the conduction and transmission of electrical energy. Compared with DC, AC can be boosted or lowered through transformers [1]. Large-scale electricity applications, such as long-distance transmission, enterprise electricity, and household electricity, generally use AC, whereas some small electronic daily-use devices, such as mobile phones, flashlights, and remote controls,

use DC. Some require the conversion of AC into constant DC. To produce constant DC voltage, a rectifier is required to convert AC voltage into unidirectional rectified DC voltage. The rectified DC voltage ripples must be further smoothed using electronic filters to produce stable and constant DC voltage [2]. The AC line filters are used to smooth residual AC ripples generated on DC voltage supply equipment, and to satisfy the current large-scale power demand, the AC filter must be installed in a capacitor with large capacitance. Aluminum electrolytic capacitors (AECs) are widely used as power devices for AC filtering applications [3]. However, the capacitance of AECs is of a lower order of magnitude than that of electrochemical capacitors (ECs); meanwhile, the rigid shape and bulky volume are increasingly incompatible with rapidly miniaturized electronics and flexible systems [4]. Therefore, the development of capacitors with high specific capacitance that can replace AECs with AC

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filter performance is essential for the future development trend.

Among various types of electrochemical energy storage devices, supercapacitors have received particular attention of scientific and industry communities in recent years because of excellent superior power density, rapid charge/discharge rate, and outstandingly long lifecycle [5–7]. Supercapacitors can be categorized into two types, namely electrical double-layer capacitors (EDLCs), where charge storage occurs through physical adsorption of ions, and pseudocapacitors (PCs), where the chemical adsorption of ions is attained through redox active materials, such as many metal oxides [8–11]. As energy storage devices, they depend on DC power. Because of a complex microporous structure in a carbon-based electrode, the EDLC-type supercapacitors conventionally exhibit large electrochemical resistance and can be rarely charged and discharged at a frequency higher than 1 Hz [12, 13]. The porous electrodes lead to poor performance for AC line filtering and thus hinder the elimination of residual AC ripple from rectified current [14]. For PCs, because of inherently low charge transfer on the electrolyte/electrode surface of the electrode material [15], capacitors behave like resistors and not capacitors in 120 Hz, which impedes their applications in AC line filtering [4, 16–18].

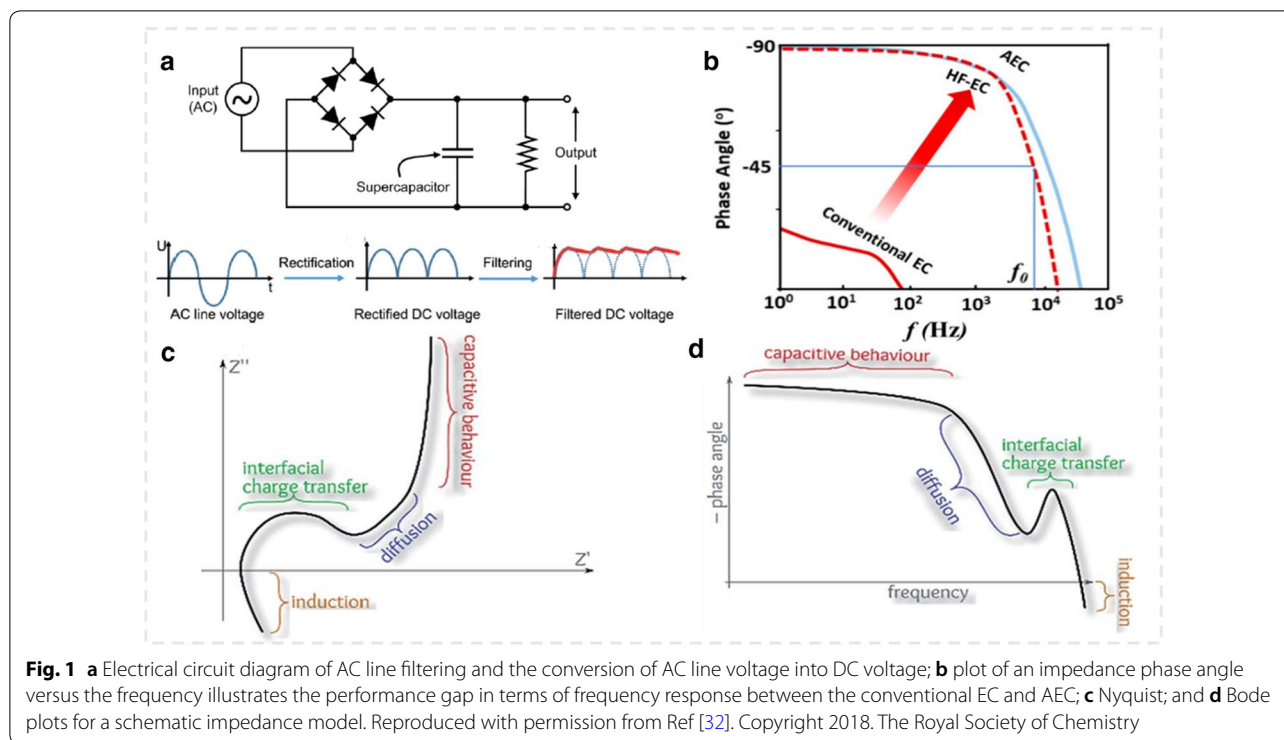
The unsatisfactory frequency response of supercapacitors is primarily attributed to the large series resistance of electrode materials and complex microporous structures. In 2010, Miller and coworkers reported that vertically oriented graphene nanosheet (VOGN) electrodes prepared through chemical vapor deposition (CVD) show excellent AC filtering performance [16]. It became the beginning of a new generation of capacitors with AC filtering performance, which then inspired numerous researchers to develop electrode materials with AC filtering performance, such as various types of graphene, electrochemically reduced graphene oxides (ErGOs), ordered mesoporous carbon (OMC), carbon derivatives, conducting polymers, and metal oxides [1–28]. This review aims to summarize the recently developed supercapacitors based on different types of new materials with AC filtering performance and attempts to understand the relationship between these materials and AC line-filtering performance.

To promote the application of supercapacitors in the AC line-filtering frequency response, the reported supercapacitors were classified based on electrode materials. It is hoped that classification can provide convenience for the development of new materials for AC filtering. "Key factors of an AC line filter" section presents the main factors of the AC line filter. The carbon-based electrode materials, conductive polymer-based electrode

materials, and transition metal-based nanomaterials are summarized in "Carbon-based electrodes", "Conducting polymer-based electrodes" and "Transition metal-based electrodes" sections. The prospect and summary of supercapacitors with AC filtering performance are illustrated in the final section.

### Key factors of an AC line filter

Figure 1a shows the full-wave bridge alternating current through AC line filtering to convert AC power into DC power. First, the AC input signal comprises a function generator. Second, a full-bridge square array comprising four diodes converts AC input signals into pulsating DC signals. Finally, the supercapacitor converts the rectangular pulse signal into a smooth DC output. The fundamental principle of frequency response is examined, and the review provides the primary reference guide for the future research and design of supercapacitors with AC filtering performance. Electrochemical impedance spectroscopy is one of the main methods to study capacitors with AC filter performance, wherein a low frequency response represents capacitance characteristics [29, 30]. The overall impedance models of PCs and double-layer capacitors (DLCs) are similar, although they show different behaviors because of different frequency ranges [31, 32]. The curve of the impedance phase angle with frequency illustrates the performance gap in terms of frequency response between the conventional EC and commercial AEC (Fig. 1b). Figure 1c shows the general capacitor impedance behavior, including three major regions, namely a high-frequency semicircle, an intermediate frequency Warburg impedance, and a low-frequency capacitance behavior. The semicircle represents the charge transfer resistance formed at the interface between the electrode and electrolyte at the beginning of the electrochemical reaction [33]. The medium-frequency region represents the characteristic of the rapid diffusion of ions to the electrode, and the high-frequency region represents the impedance of charge transfer associated with the Faraday redox reaction of the electrode material [34]. Figure 1d describes the relationship between the phase angle of impedance and frequency, which is termed the Bode phase diagram. The impedance diffusion behavior of the electrode material corresponds with the Bode diagram on the right [35, 36]. A gradual transition is observed from the second region to the third region in the impedance spectrum, which corresponds to the gradual increase in the phase angle in the Bode diagram to reach a near optimally stable value of approximately 90° [37]. The phase angle at 120 Hz was the primary indicator for measuring AC line-filtering performance.



**Fig. 1** a Electrical circuit diagram of AC line filtering and the conversion of AC line voltage into DC voltage; b plot of an impedance phase angle versus the frequency illustrates the performance gap in terms of frequency response between the conventional EC and AEC; c Nyquist; and d Bode plots for a schematic impedance model. Reproduced with permission from Ref [32]. Copyright 2018. The Royal Society of Chemistry

The resistance capacitance (RC) time constant ( $\tau_{RC}$ ) produced using the complete resistance measured under 120 Hz, which was the result of charge (electrons and ions) mobility limitation. The time constant was obtained using the equation ( $\tau_{RC} = C \times Z'$ ), where  $Z'$  is the resistance derived from the real part of the impedance, and  $C$  is the specific capacitance calculated from the equation  $C = -1/(2\pi fZ'')$ , with  $f$  and  $Z''$  representing frequency (Hz) and imaginary part of impedance ( $\Omega$ ), respectively [38]. The RC time constant of conventional supercapacitors is generally 1 s, which is excessively high for 120-Hz AC filtering with a time period of 8.3 s [39]. Such a high time constant can be attributed to the complex pore structure of supercapacitor electrode materials, which hinders the high-speed diffusion of ions and increases the effective charge transfer resistance [40, 41]. Therefore, to fabricate supercapacitors that meet the performance of AC filtering, electrode materials with a high specific surface area, high conductivity, and highly ordered pore structure, must be designed. Another crucial parameter to detect the performance of AC filtering is the relaxation time constant ( $\tau_0 = 1/f_0$ ), which is the minimum time required to discharge all energy from the device with an efficiency of more than 50% [42]. Characteristic frequency  $f_0$  is the value at a phase angle of 45°. Lower  $\tau_0$  corresponds to the prominent rate performance of the supercapacitor, and the ability to maintain

capacitive behavior at a high frequency is also reflected when the phase angle is close to the optimal value of 90° [11]. In the design of supercapacitors with AC filtering performance, the ultrahigh rate performance of electrode materials is a necessary characteristic to eliminate high-order harmonics to protect electronic devices from damage [43]. In summary, three primary factors for the preparation of supercapacitor electrode materials with AC filtering properties are as follows: (1) high specific surface area, (2) high electrical conductivity, and (3) ordered pore structure distribution.

### Carbon-based electrodes

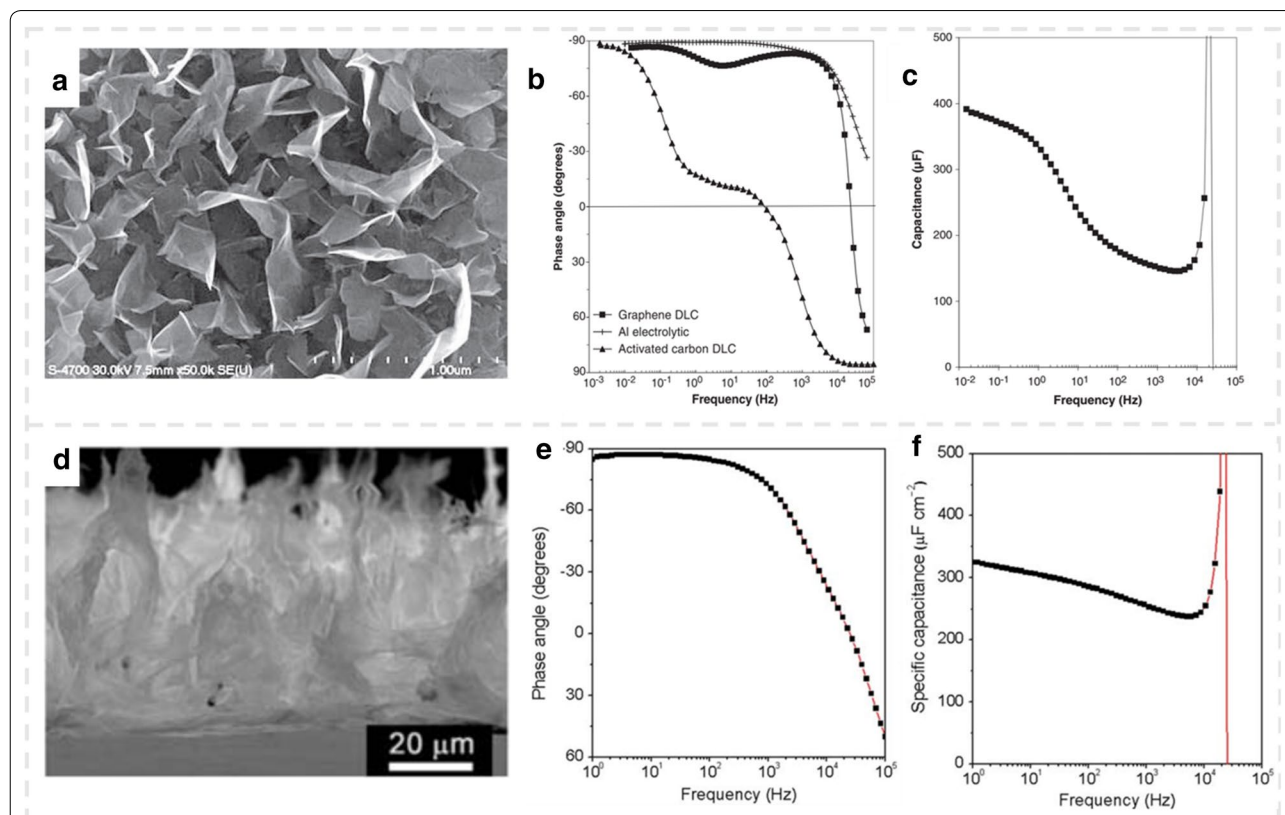
Carbon-based materials including activated carbon [44], onion-like carbon [41], carbon derivatives carbon [45, 46], carbon nanotubes (CNTs) [17, 30, 47–50], and graphene [51–53] have been intensively applied as electrode materials for supercapacitors. Typical supercapacitors based on these porous carbon electrode materials show a low frequency response and a phase angle of approximately 0° at 120 Hz, which is considerably lower than the optimal 90° for AC line filtering [54]. The main reason is the complex and disordered porous structure of electrode materials, which hinders the transfer of electrons and mass [13]. Therefore, various new carbon-based electrode materials have been developed for a high-frequency response to satisfy the requirements for AC line filtering.

### Graphene-based electrode

Graphene-based electrodes have led to extensive research on the application of supercapacitors in recent years because of its inherent high specific surface area and high conductivity. A large number of research results have shown that graphene has great potential in energy storage. Graphene can be used in devices such as micro-capacitors, transparent batteries, and rapid charge–discharge electronic equipment [55–60]. In 2010, for the first time, Miller reported that a VOGN electrode-based capacitor grown on metal current collectors through CVD can be used for AC line filtering [12]. The as-produced capacitor had an RC time constant of 1 s. Figure 2a shows a plain view of a VOGN electrode-based capacitor. The irregular surface is caused by the defect due to external stress and incorporation of hydrogen. Figure 2b shows the impedance phase angle versus the frequency of the VOGN electrode-based capacitor, activated carbon electrode-based capacitor, and commercial aluminum electrolytic capacitor. At 120 Hz, the phase angle of the capacitor based on the graphene nanosheet was

83°, which was close to the phase angle 85° of the AEC, whereas the impedance phase angle of the capacitor based on activated carbon was almost 0°. This difference was almost entirely caused by the complex pore structure of activated carbon. The frequency-dependent capacitance of the graphene nanosheet capacitor shows that the capacitance value was 175 μF and the RC time constant was less than 200 ms at 120 Hz (Fig. 2c). Moreover, some graphene-based electrode materials prepared using the CVD method have been used to increase the frequency of the corresponding supercapacitors [20, 24, 61].

Sheng [9] reported a similar frequency response based on a three-dimensional graphene electrode prepared by the electrochemical reduction of graphene oxide. The as-prepared ErGO electrode comprised a porous 3D interpenetrating layer with a thickness of approximately 20 μm (Fig. 2d). At 120 Hz, the phase angle of ErGO-DLC (−84°) was comparable to that of commercial aluminum electrolytes (−85.5°) at 120 Hz (Fig. 2e). The maximum area capacitance of the capacitor was 325 μF cm<sup>−2</sup> at the lowest frequency of 1 Hz (Fig. 2f). However,

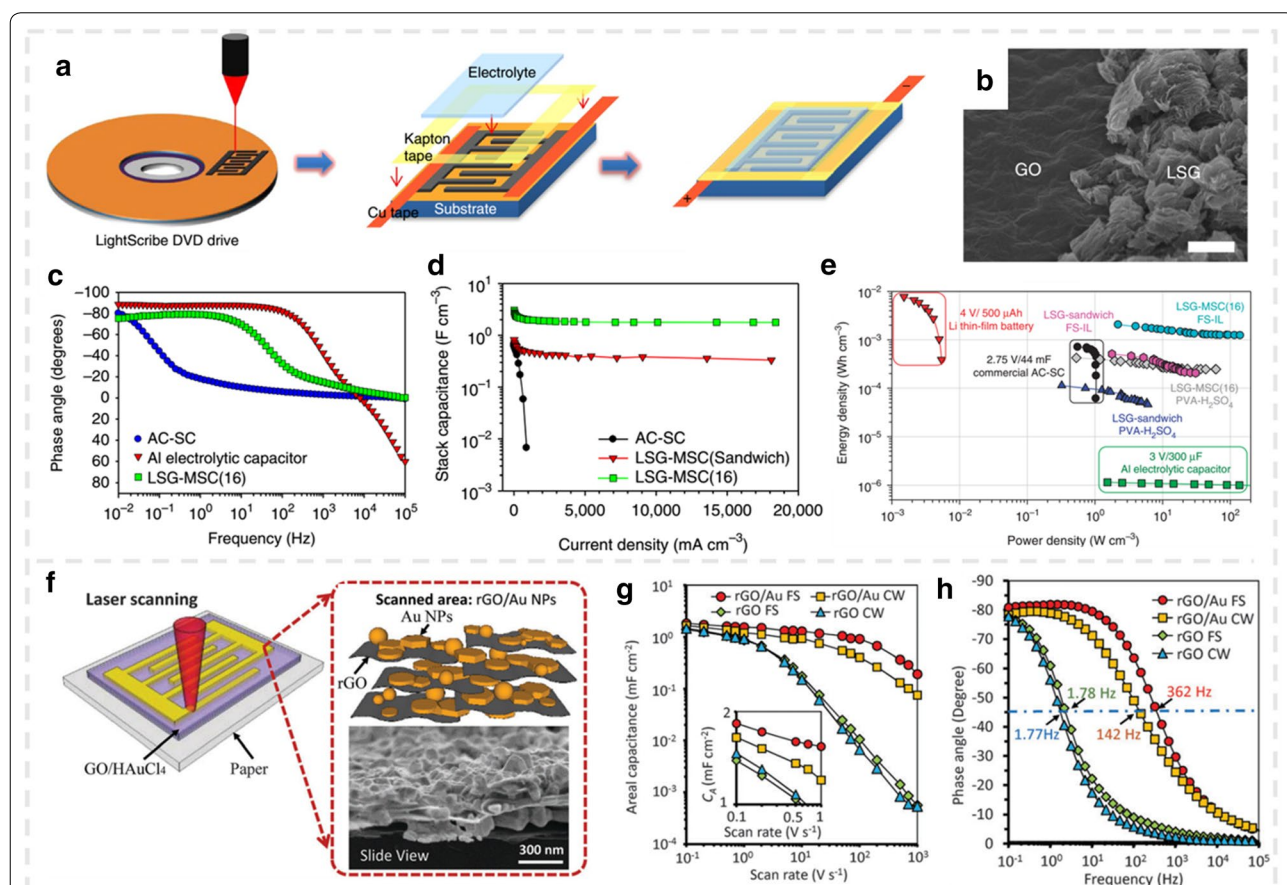


**Fig. 2** a Scanning electron microscope (SEM) micrograph vertical aligned graphene on the Ni surface; b impedance phase angle versus the frequency for a graphene nanosheet-based capacitor, commercial aluminum electrolytic capacitor, and activated carbon-based capacitor; c capacitance versus the frequency of the graphene nanosheet capacitor; Reproduced with permission from Ref [12]. Copyright 2019. AAAS. d cross-sectional SEM image of the ErGO electrode; e plot of the impedance phase angle versus the frequency; and f plot of specific capacitance versus the frequency. Reproduced with permission from Ref [9]. Copyright 2012. Macmillan Publishers Limited

the excellent frequency response caused by low specific capacitance at the cost of a low material density load could not satisfy practical requirements. The primary reason is that supercapacitors with graphene-based materials as electrode materials are frequently affected by the accumulation of graphene sheets during processing. In response to the aforementioned problems, Yoo and coworkers [62] fabricated in-plane and ultrathin supercapacitors, wherein the surface of each layer of graphene could be used to store energy. Compared with the conventional two-dimensional stacked electrode, the planar design resulted in a dramatic increase in capacitance.

On the basis of the design of this type of planar electrodes, with the rapid development of miniaturized electronic equipment, the demand for compact energy storage on-chip is increasing day by day. Compared with conventional AECs, the in-plane micro-supercapacitors

(MSCs) have higher volume energy density and a more compact structure. Therefore, the development of planar MSCs with an AC line-filtering function is an ideal alternative to replace AECs and integrate with other electronic units. Kaner and coworkers [63] reported the scalable fabrication of graphene MSCs, which could be developed on a large scale by using a standard Light-Scribe DVD recorder for direct laser writing on graphite oxide films. The laser-scribed graphene (LSG)-based MSCs exhibited a suitable frequency response in a short relaxation time (Fig. 3) compared with commercial AC supercapacitors and AECs. Figure 3a–c show the preparation process of the photolithographic graphene electrode and the assembly of the device. Figure 3b shows the morphology of the film after the laser treatment. The film expanded, and thus, the film could be in complete contact with the electrolyte, which was essential for the



**Fig. 3** a Schematic of the fabrication process for an laser scribed graphene (LSG) micro-supercapacitor; b a tilted-view (45°) SEM image shows the direct reduction and expansion of the GO film after exposure to the laser beam, scale bar (10 μm); c the impedance phase angle versus the frequency for LSG-MSC compared with a commercial AC-supercapacitor and aluminium electrolytic capacitors; d volumetric capacitance of an LSG-MSC and a commercial AC-supercapacitor; e Ragone plots of LSG-MSCs compared with commercially available energy-storage systems; Reproduced with permission from Ref [63]. Copyright 2013. Macmillan Publishers Limited. f Direct femtosecond laser writing of rGO/Au microelectrodes; g areal capacitance as a function of the scan rate (inset shows a magnified low-scan-rate region); and h Bode plot of impedance of the rGO-based MSCs. Reproduced with permission from Ref [64]. Copyright 2016. Macmillan Publishers Limited

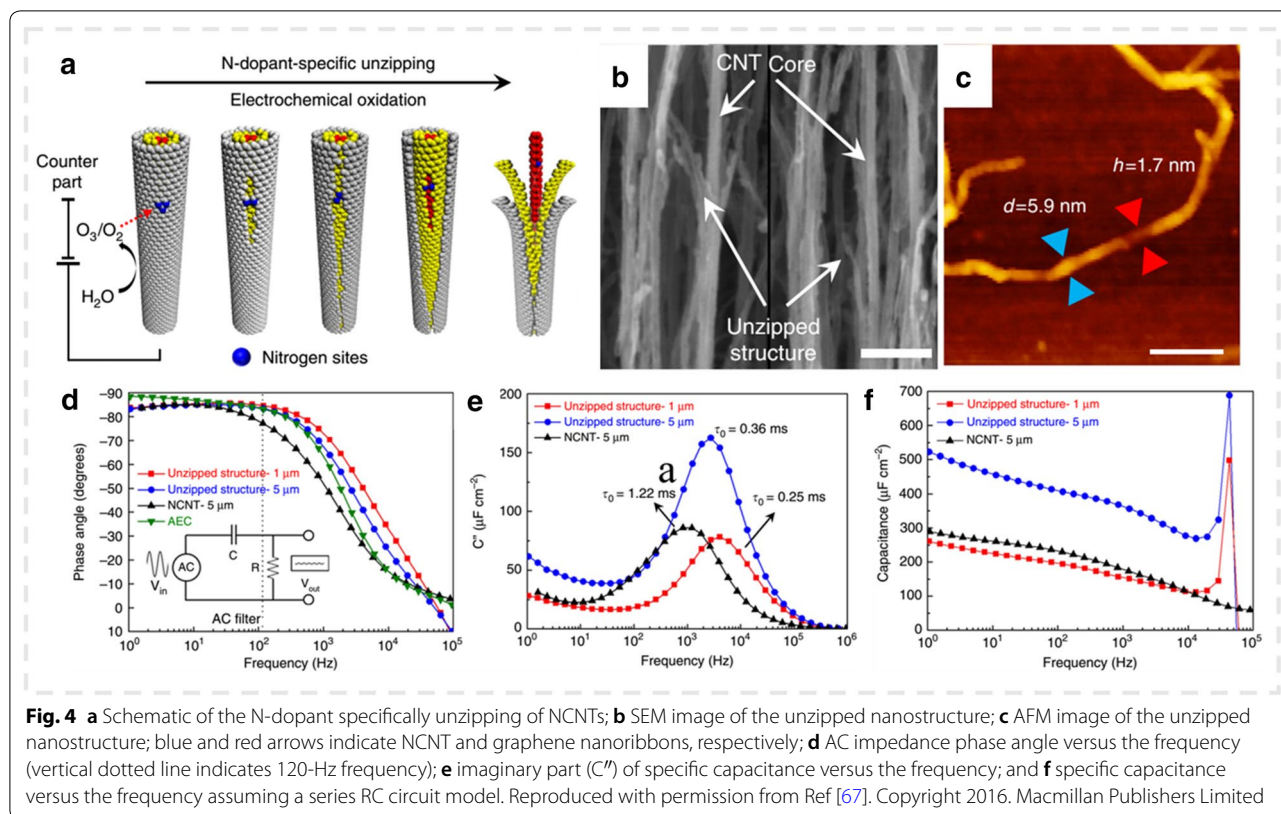
charge–discharge performance of the electrode. Furthermore, such LSG-MSCs have an extremely small relaxation time (Fig. 3c), thus showing an excellent frequency response. Figure 3d shows the volume capacitances of the LSG-MSC, AC supercapacitors, and AECs. At a current density of  $16.8 \text{ mA}\cdot\text{cm}^{-3}$ , the stack capacitance of LSG-MSC was  $3.05 \text{ F cm}^{-3}$ , which was approximately three orders of magnitude higher than the conventional discharge current density used to evaluate conventional supercapacitors. The LSG microdevice showed three times higher energy density and 200 times higher power density compared with the Ragone plots of carbon-based supercapacitors (Fig. 3e).

The LSG-MSC has a power density equivalent to AECs and provides an energy density of more than three orders of magnitude. Figure 3f shows that the femtosecond laser (FS) reduced the  $\text{HAuCl}_4$  coating on Au nanoparticles and inserted it into an RGO to form a stacked structure for a RGO-Au microelectrode, which significantly improved the conductivity of the electrode reported by Hu and coworkers [64]. The area capacitance of RGO-Au FS-MSCs was evidently higher than that of other electrodes prepared using the FS method at the same scanning speed (Fig. 3g). This advantage was more evident at high scanning rates. Furthermore, the as-fabricated rGO/Au FS-MSCs satisfied the requirement of AC line filtering in the high-frequency region. The as-produced RC time constant was 2.76 ms when the characteristic frequency at the phase angle was  $45^\circ$  (Fig. 3h). The RGO/Au electrode obtained using the FS etching increased the conductivity of the electrode by two orders of magnitude to  $1.1 \times 10^6 \text{ S m}^{-1}$ , and the planar inter-digital electrode shortened the ion diffusion distance. The fabricated supercapacitor has a high-frequency response, large specific capacitance ( $0.77 \text{ mF cm}^{-2}$  at  $1 \text{ V s}^{-1}$ ), and outstanding cycle stability. These improvements in the electrode design are expected to lead to an ideal energy density for supercapacitors.

Nitrogen doping for graphene was reported to significantly improve the specific surface area and electrical conductivity of electrode materials, which were the most crucial features of energy storage materials. The radius of N is close to C, making N easy to replace carbon atoms in the atomic lattice, and further resulting in the formation of N-doped carbon materials. The nitrogen atom has one more extra-nuclear electron than the carbon atom and has a high electron affinity. Therefore, C atom adjacent to N in the nitrogen-doped carbon material has high positive charge density. At the same time, there is a conjugation channel between N lone pair electrons and large  $\pi$ -bonds in carbon lattice, rendering N-doped carbon materials with excellent electrochemical properties. [65, 66] As a conventional application, the research

on ultrahigh power DLCs for AC line filtering was presented. Kim and coworkers [67] proposed a controllable method to customize the complete crystalline graphene nanostructure through the specific decompression of CNT heteroatom dopants. The cooperative nanostructure exhibited a large specific surface area and strong electrical connectivity and showed the performance of ultrahigh power supercapacitors, which could be used for AC line filtering with a record high-speed capability of a phase angle of  $85^\circ$  at 120 Hz. Figure 4a shows the electrochemical decompression process of a nitrogen-doped CNT (NCNT) forest in  $1 \text{ M H}_2\text{SO}_4$  at a potential of 0.6–0.8 V. The SEM image validated that the carbon nanostructure comprising unzipped graphene nanoribbons wrapped around the NCNT core (Fig. 4b). The atomic-force microscopy (AFM) image showed that the 1.7-nm-thick three-layered graphene sheets were produced from 5.9-nm-diameter triple-walled NCNTs (Fig. 4c). The impedance phase angle spectrum further validated that the unzipped structure exhibited excellent AC filtering performance. Figure 4d compares the impedance phase angles of DLCs at different frequencies of unzipped, original NCNT forests and conventional AECs at different frequencies. Moreover, a 1-mm-high unzipped nanostructure of DLCs could reach up to a phase angle of  $45^\circ$  at 8.1 kHz and exhibited the impedance phase angle of  $85^\circ$ , which was comparable to that of the AEC or other reported DLCs. Figure 4e presents the specific capacitance and RC time constant. Moreover, a 5-mm-unzipped structure-based DLC exhibited  $402 \text{ mF cm}^{-2}$  at 120 Hz, which was considerably higher than  $230 \text{ mF cm}^{-2}$  of the 5-mm-NCNT-DLC (Fig. 4f). Furthermore, the RC time constants of DLCs fabricated using a 5-mm-NCNT forest, 5-mm-unzipped structures, and 1-mm-unzipped structures were 616, 468, and  $212 \mu\text{s}$  at 120 Hz, respectively. These values were adequately smaller than the standard period of 8.3 ms, which was crucial for effective AC line filtering at 120 Hz. The unzipped nanostructures showed a small relaxation time constant ( $\tau_0 = 0.25 \text{ ms}$ ) at 1 mm, which indicated that DLCs constructed with the customized unzipped structures have not only ultrahigh power performance but also a large specific capacitance at 120 Hz. Although heteroatom doping could increase the conductivity of the material to a certain extent, through some research attempts, heteroatom doping is not suitable for eliminating AC filtering. This could be the induction of active scattering centers by heteroatoms in the carbon lattice, and therefore the electrons dispersing and the discharge rate of the charge are both hindered [66].

To satisfy the growing demand for portable, wearable, and implantable electronic devices, high-performance and flexible MSCs have a prominent application



prospect. Direct laser writing (DLW) technology provided a favorable protocol for the fabrication of flexible, high-performance miniature supercapacitors that satisfied the requirements. Zhu and coworkers [68] developed graphene-based MSCs (MG-MSCs) through the DLW of stacked graphene films made through industry-scale CVD. Figure 5a shows the fabrication process of flexible all-solid-state MG-MSCs. Furthermore, the MG-MSCs demonstrated an outstanding AC line-filtering performance, which indicated the phase angle of  $76.2^\circ$  and a RC constant of 0.54 ms at 120 Hz. Figure 5b compares the parameters of AC filter performance with those of the reported supercapacitors, which further illustrated the application prospects of MG-MSCs in a flexible AC line changer.

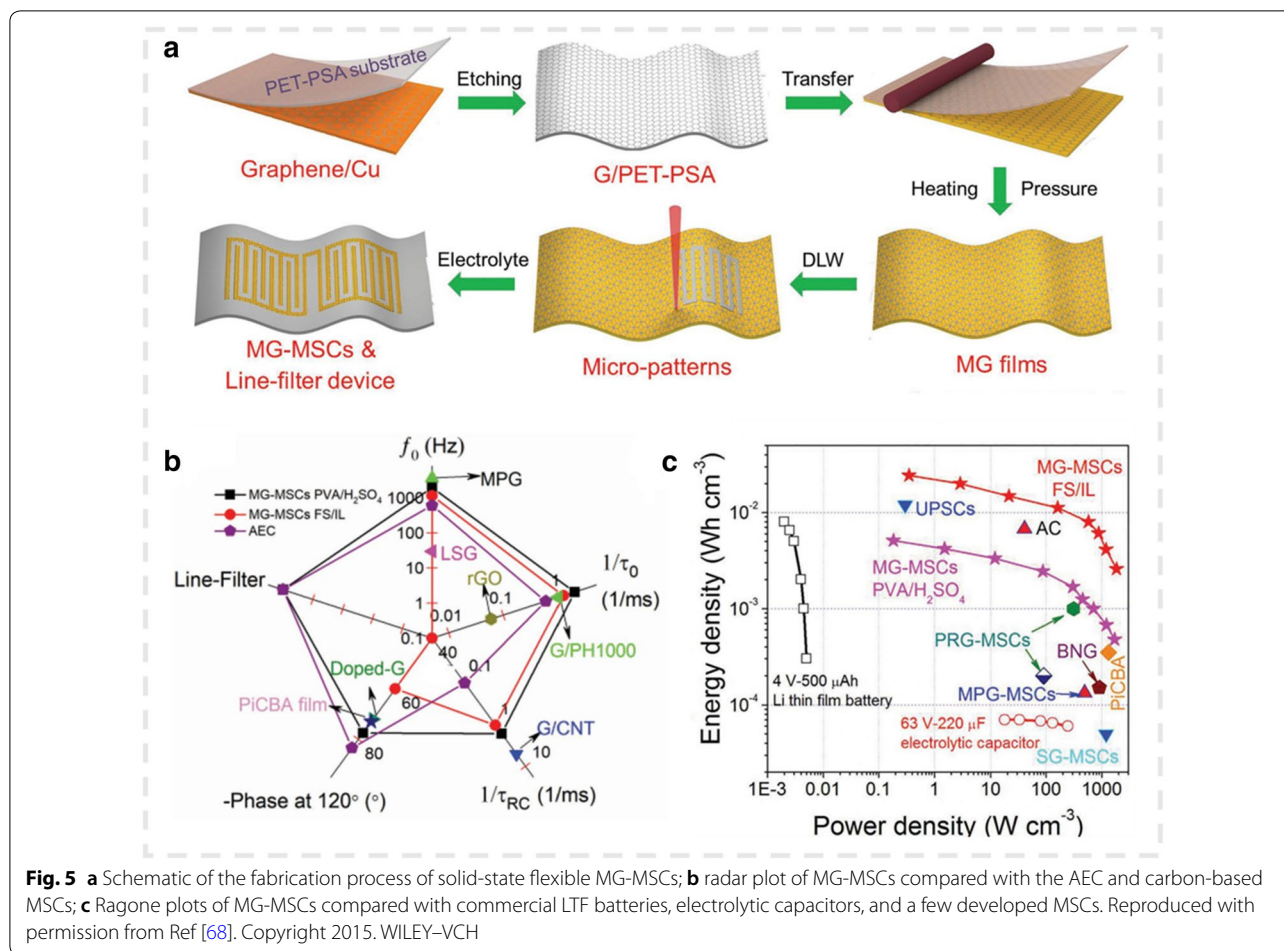
The Ragone graph, which compared the power density and energy density performance of MG-MSCs with those of different types of MSCs is presented in Fig. 5c. The MG-MSCs exhibited an exceptional energy density of  $23 \text{ mWh cm}^{-3}$  at  $0.32 \text{ W cm}^{-3}$  in an ion-gel electrolyte, which was higher than those of other carbon-based thin-film MSCs, such as doped graphene (sulfur-doped graphene, SG-MSCs,  $12.4 \text{ mWh cm}^{-3}$ ; nitrogen and boron codoped graphene, BNG-MSCs,  $16.9 \text{ mWh cm}^{-3}$ ) [69], MPG-MSCs ( $7.5 \text{ mWh cm}^{-3}$ ) [70], photochemical reduction of GO/TiO<sub>2</sub> composite (PRG-MSCs,  $\approx 13.2$

$\text{mWh cm}^{-3}$ ) [71], and ultrathin printable MSCs (G/PH1000, UPSCs-25,  $12 \text{ mWh cm}^{-3}$ ) [9]. After the function generator inputs the AC signal, the AC signal can be converted into the pulse DC signal by full-bridge rectifier, and finally, the DC output is smoothed by the miniature supercapacitor prepared in the experiment.

### CNT-based electrode

Another effective strategy to improve the capacitance of supercapacitors was to add spacers, such as CNTs between graphene sheets to prevent their restacking. Thus, Wang and coworkers [50] developed ultrahigh power density MSCs based on reduced graphene oxide (rGO) and CNT composite electrodes. Figure 6a illustrates the process of fabricating an MSC by integrating electrode materials into a digital inter-Ti/Au microelectrode. Figure 6b shows the SEM image of the rGO-CNT hybrid electrode, which evidently revealed the uniformly filled thin film with the appearance of CNTs between the rGO sheets through the controllable range of deposition, and no evident stacking phenomenon was observed.

As shown in Fig. 6c and d, the specific capacitance increased compared with that of the reported capacitor; however, frequency response performance was poor, which could not satisfy the requirements of AC filtering. Hata and coworkers [48] fabricated MSCs



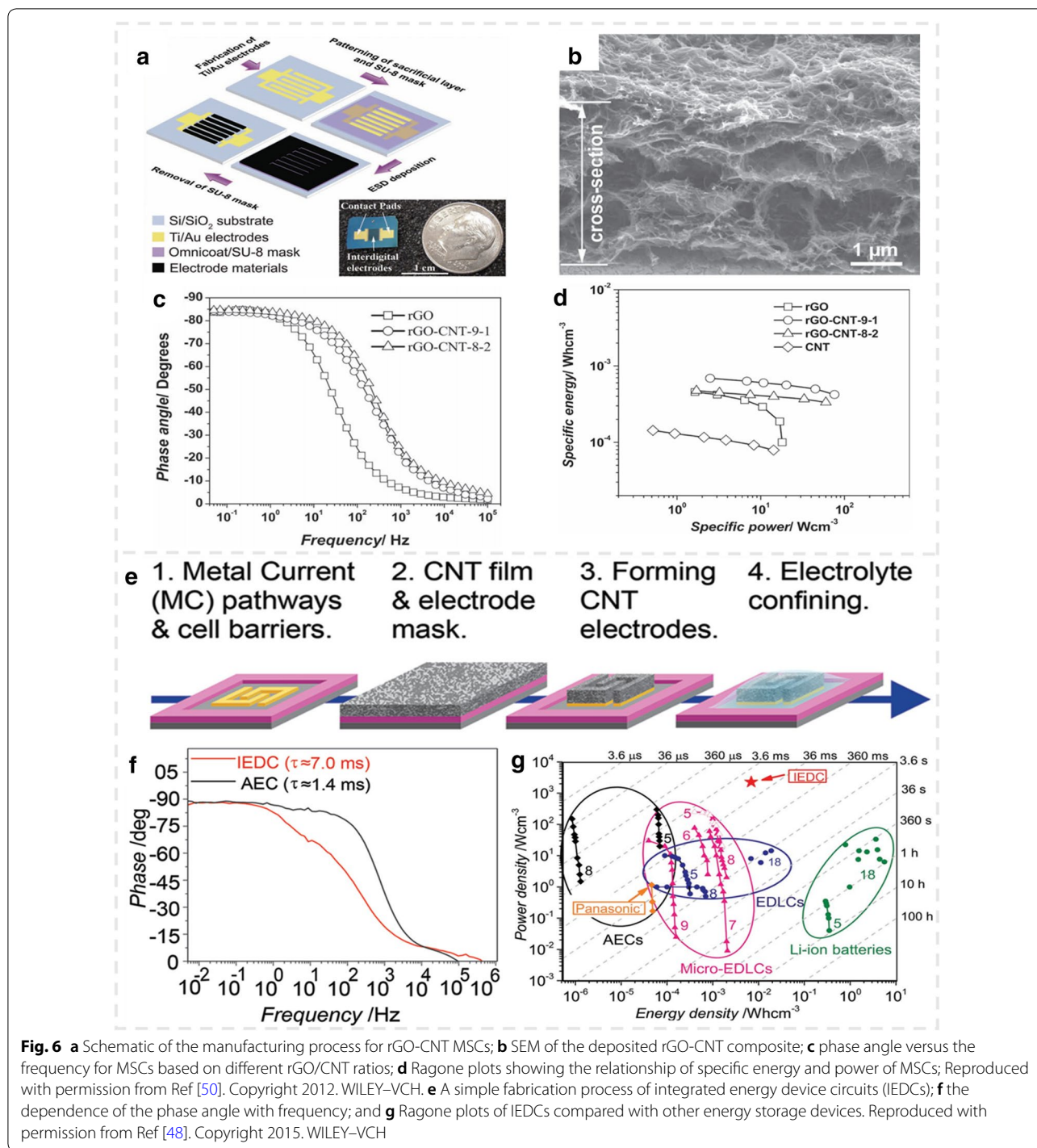
based on the interdigital single-walled CNT (SWCNT) electrode, which solved the large volume problem of AECs in microdevices. The fabrication process of the IEDC is presented in Fig. 6e. From Fig. 6f and g, it can be concluded that the power density of IEDC (2271 W cm<sup>-3</sup>) was two orders of magnitude higher than that of commercial capacitors (14 W cm<sup>-3</sup>) and one order of magnitude higher than that of commercial AECs (300 W cm<sup>-3</sup>). The resulting relaxation time constant (7 ms) could be used in AC filtering for AC–DC conversion.

Flexible and wearable portable electronic devices were inevitably developed to meet increasing social requirements. Flexible microdevices with AC filtering performance were also fabricated to meet these requirements. Xie and coworkers [72] reported an in-plane MSC with AC line-filtering performance based on the SWCNT film and TiO<sub>2</sub> NPs. Figure 7a shows the fabrication process of the microdevice. The SWCNT enhanced the stretchability of SWCNT microelectrodes. Figures 7b and c compared the optical photographs of the microdevices at a telescopic pressure of 200 and without any pressure,

respectively. Figure 7e presents the phase angles under different telescopic pressures.

At present, flexible supercapacitors with AC filtering performance are limited by low energy density. One of the reasons is that they are limited by narrow voltage window in aqueous-solution-based gel electrolytes [17]. Design the 3.0 V flexible supercapacitor for AC line filtering based on an ionic-liquid-based polymer gel electrolyte and carbon nanotube electrode material become emerging. Such flexible supercapacitor can exhibit 20 times higher areal energy density than that of the previously demonstrated 1.0 V flexible supercapacitor. Kang et al. [17] designed a 3.0 V flexible supercapacitor with CNT electrode materials based on a ionic-liquid-based polymer gel electrolyte that could be used for AC line filtering (Fig. 7f–i). The as-resulted supercapacitor had an approximately 20 times higher capacitance than the previously developed 1.0 V supercapacitor (0.66 vs 0.03 μWh/cm<sup>2</sup>). The frequency response of flexible supercapacitors based on CNTs was closely related to the thickness of electrode materials. The phase angle increased from 70.6° to 82.7°, with the thickness of the

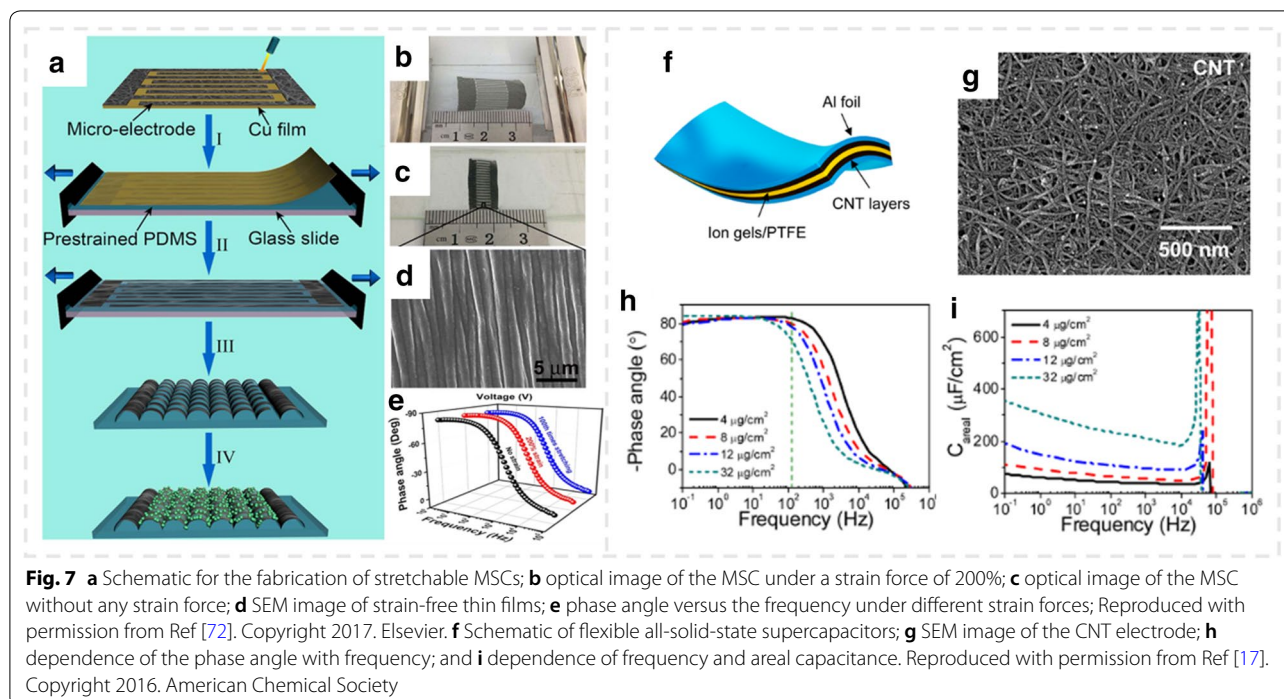




**Fig. 6** **a** Schematic of the manufacturing process for rGO-CNT MSCs; **b** SEM of the deposited rGO-CNT composite; **c** phase angle versus the frequency for MSCs based on different rGO/CNT ratios; **d** Ragone plots showing the relationship of specific energy and power of MSCs; Reproduced with permission from Ref [50]. Copyright 2012. WILEY-VCH. **e** A simple fabrication process of integrated energy device circuits (IEDCs); **f** the dependence of the phase angle with frequency; and **g** Ragone plots of IEDCs compared with other energy storage devices. Reproduced with permission from Ref [48]. Copyright 2015. WILEY-VCH

film decreasing from 300 to 50 nm. The corresponding areal capacitance was reduced from 233 to 43 μF cm<sup>-2</sup> at 120 Hz. Thus, the potential application of supercapacitors in flexible or portable electronic products was considerably improved by increasing the device voltage and simultaneously maintaining the rapid response ability.

In addition, most supercapacitors with AC line filtering performance take advantage of their high ionic conductivity in water, but limit the operating voltage to less than 1.0 V, and lead to the low power density of supercapacitors [17]. The basic requirement for expanding the voltage window is to design appropriate electrode

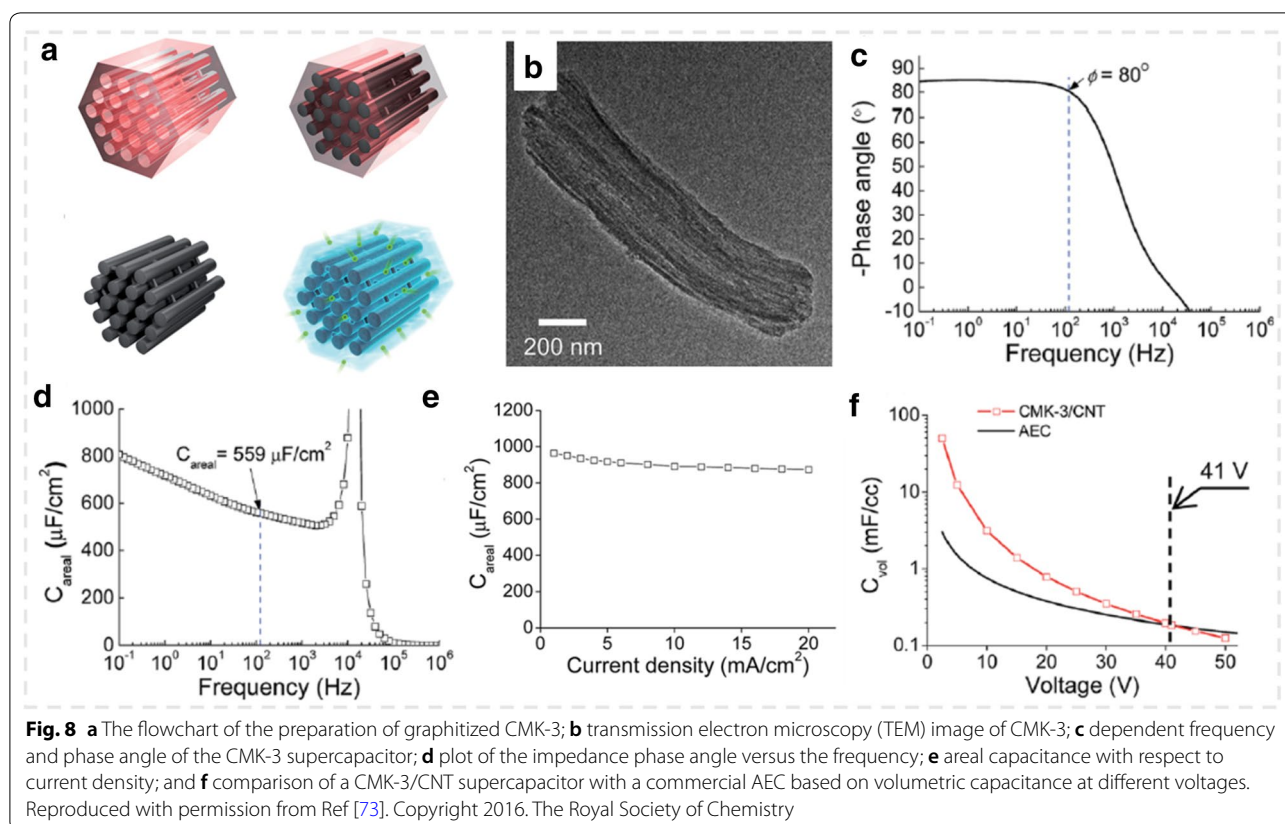


materials with a porous structure. Yoo [73] reported a supercapacitor with AC line filtering by using graphitic-OMC (GOMC or CMK-3) as the electrode material to increase the voltage window. The orderly network structure was formed by adding a small amount of CNTs into CMK-3 to meet the requirement of AC line-filtering performance. The CMK-3 particles with well-defined, wide-open pore structures are obtained by infiltrating and carbonizing the SBA-15 silica template (Fig. 8a). As can be seen from the TEM image in Fig. 8b, CMK-3 has a straight pore channel. Figure 8c and d show the frequency response of the CMK-3/CNT supercapacitor. The supercapacitor based on an organic electrolyte has an excellent frequency response and high area capacitance, and the phase angle could reach  $80^\circ$  at 120 Hz. The near-vertical Nyquist curve shows that the CMK-3/CNT supercapacitor had a lower equivalent series resistance ( $\sim 0.25 \Omega$ ). Furthermore, the supercapacitor exhibited a high-area capacitance of  $559 \mu\text{F cm}^{-2}$  at 120 Hz. Furthermore, the supercapacitor exhibited a very short relaxation time constant of 1 ms. The maximum areal capacitance reached up to  $963 \mu\text{F cm}^{-2}$  at current density  $1 \text{ mA cm}^{-2}$  (Fig. 8e). The voltage value of CMK-3/CNT supercapacitors was 41 V compared with that of commercial AECs (Fig. 8f). The volume capacitance of the CMK-3/CNT supercapacitor at 2.5 V was 16.7 times of commercial AECs ( $50.2 \text{ mF cm}^{-3}$  vs  $3.0 \text{ mF cm}^{-3}$ ). Compared with the previously reported supercapacitors, the maximum voltages could reach only 3.4 V (VOGNs) [74]

and 21 V (single walled nanotubes) [31]. The reason for the high voltage value of the CMK supercapacitor (41 V) compared with that of commercial AECs was the rationally designed electrode materials. The results showed that the high-frequency applications of supercapacitors could be developed through the optimization of the pore structure and increasing the conductivity of carbon electrode materials.

#### Other carbon-based electrodes

In addition to graphene and CNTs, other carbon-based electrode materials are used in the study of supercapacitors with AC line-filtering performance. The rapid frequency response is usually achieved by controlling the ordered pore structure of carbon materials. Kossyrev [75] examined carbon solutions for high-frequency-response applications of supercapacitors. The supercapacitors with carbon black as an electrode could meet the frequency response operation of AC line filtering because of the short length of the pore in the carbon black material. Furthermore, three samples S-1, S-2, and S-3 were prepared using different concentrations of the surfactant. The phase angle reached  $-75^\circ$  at 120 Hz, and the RC time constant reached 0.354 ms. Medium/large pore camphore-derived carbon sponge-assembled button supercapacitors have been applied to AC line filtering with an ultrafast frequency response having a phase angle of  $78^\circ$  and a RC time constant of 319 ms at 120 Hz [76]. Figure 9a shows the SEM images of carbon sponges



of lyophilized S-3 that exhibited larger pores and a high degree of meso/macroporosity with sodium dodecyl sulfate (SDS) surfactant concentrations at 1 wt %. Figure 9b compares the frequency and phase angle of S-3, active carbon-based supercapacitor, and AEC. The phase angle of the S-3 carbon sponge ( $-78^\circ$ ) was considerably larger than that of the activated carbon-based capacitance ( $0^\circ$ ), which was comparable with that of the AEC ( $-85.5^\circ$ ). The capacitance was  $172 \text{ mF cm}^{-2}$ , and the RC time constant was 319 ms at 120 Hz (Fig. 9c, d).

### Conducting polymer-based electrodes

In addition to the development of EDLCs with carbon-based electrodes for AC line filtering, various interesting conductive electrode material-based supercapacitors with AC line filtering have been reported. The electrochemical conductivity and high capacitance inherent of conductive polymer bridged the performance gap of the low-energy-density double-layered capacitors with carbon-based electrodes. Alshareef and coworkers [77] reported a polymer-based MSC with AC line-filtering performance other than a carbon-based electrochemical double-layer capacitor. Figure 10a shows the electropolymerization process for the preparation of a porous conductive polymer (PEDOT) electrode with

an anionic surfactant at a lower anode potential. Figure 10b shows the nanomorphology of PEDOT with an open porous structure during electrochemical deposition. At 120 Hz (Fig. 10c), the impedance phase angle was  $65^\circ$  for the PEDOT-MSC. The PEDOT-MSCs exhibited a maximum areal capacitance of  $9 \text{ mF cm}^{-2}$  in 1 M  $\text{H}_2\text{SO}_4$  and a volumetric stack capacitance of  $50 \text{ F cm}^{-3}$  (Fig. 10d). The Ragone plot (Fig. 10e) shows that the maximum energy density and power density of PEDOT-MSC based on the ionic gel were  $7.7 \text{ mWh cm}^{-3}$  and  $850 \text{ mW cm}^{-3}$ , respectively. Moreover, this energy density was significantly higher than that of carbon-based materials ( $E = 0.15\text{--}6 \text{ mWh cm}^{-3}$ ). Furthermore, Shi and coworkers [78] reported an EC-based on highly conductive  $\text{H}_2\text{SO}_4$ -treated PEDOT:PSS porous electrodes with a flexible graphite foil as a current collector (denoted as AT-PEDOT:PSS) with AC line-filtering performance. Figure 10f shows that the manufacturing process of AT-PEDOT:PSS on the graphite foil [78]. The PEDOT was doubled on cellulose, and the PEDOT:PSS solution penetrated paper fibers through the inherent microporous structure of the cellulose paper. All cellulose fiber frameworks were completely dissolved in  $\text{H}_2\text{SO}_4$  solution, and moreover, the residual AT-PEDOT:PSS membrane inherited its porous structure. The AT-PEDOT:PSS SEM

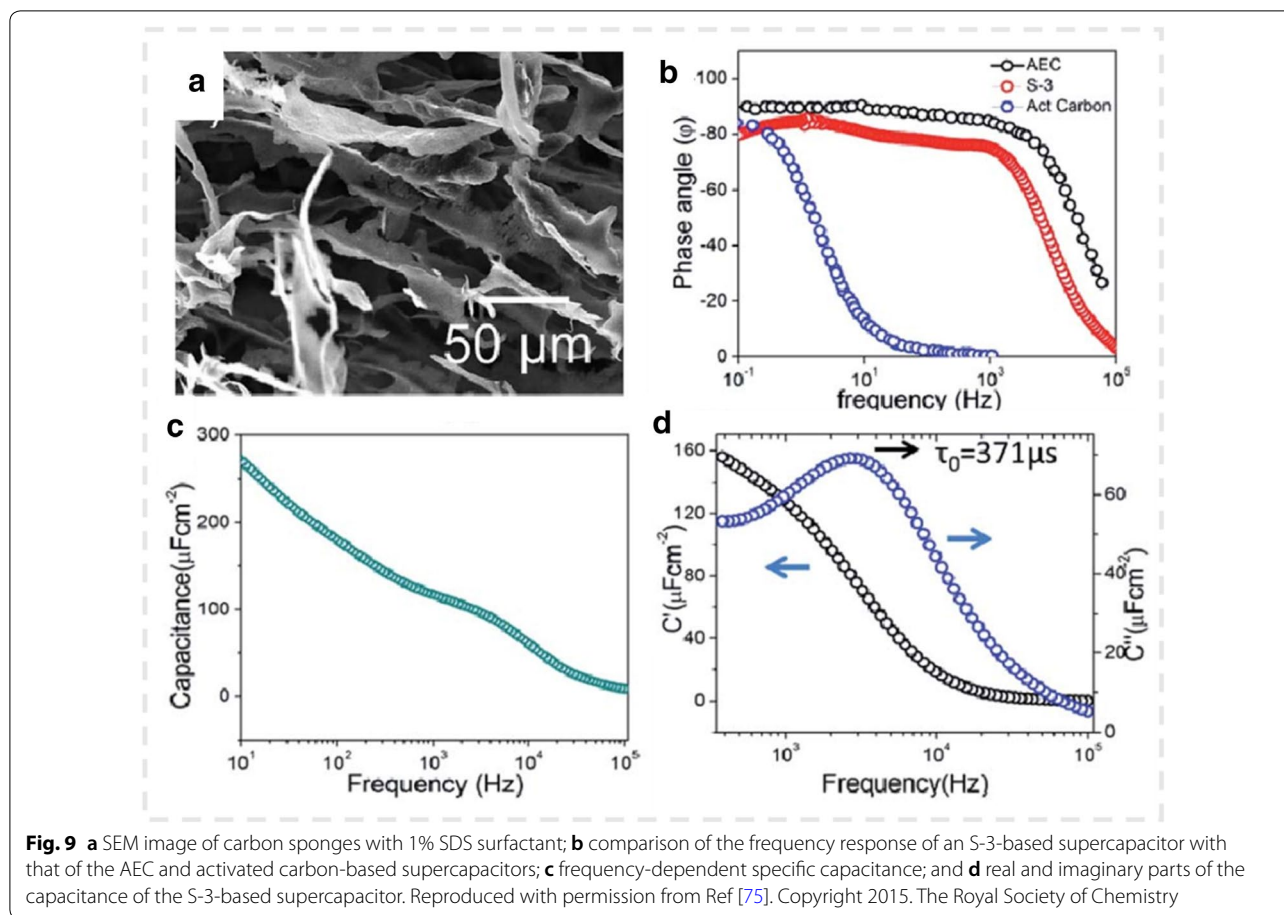
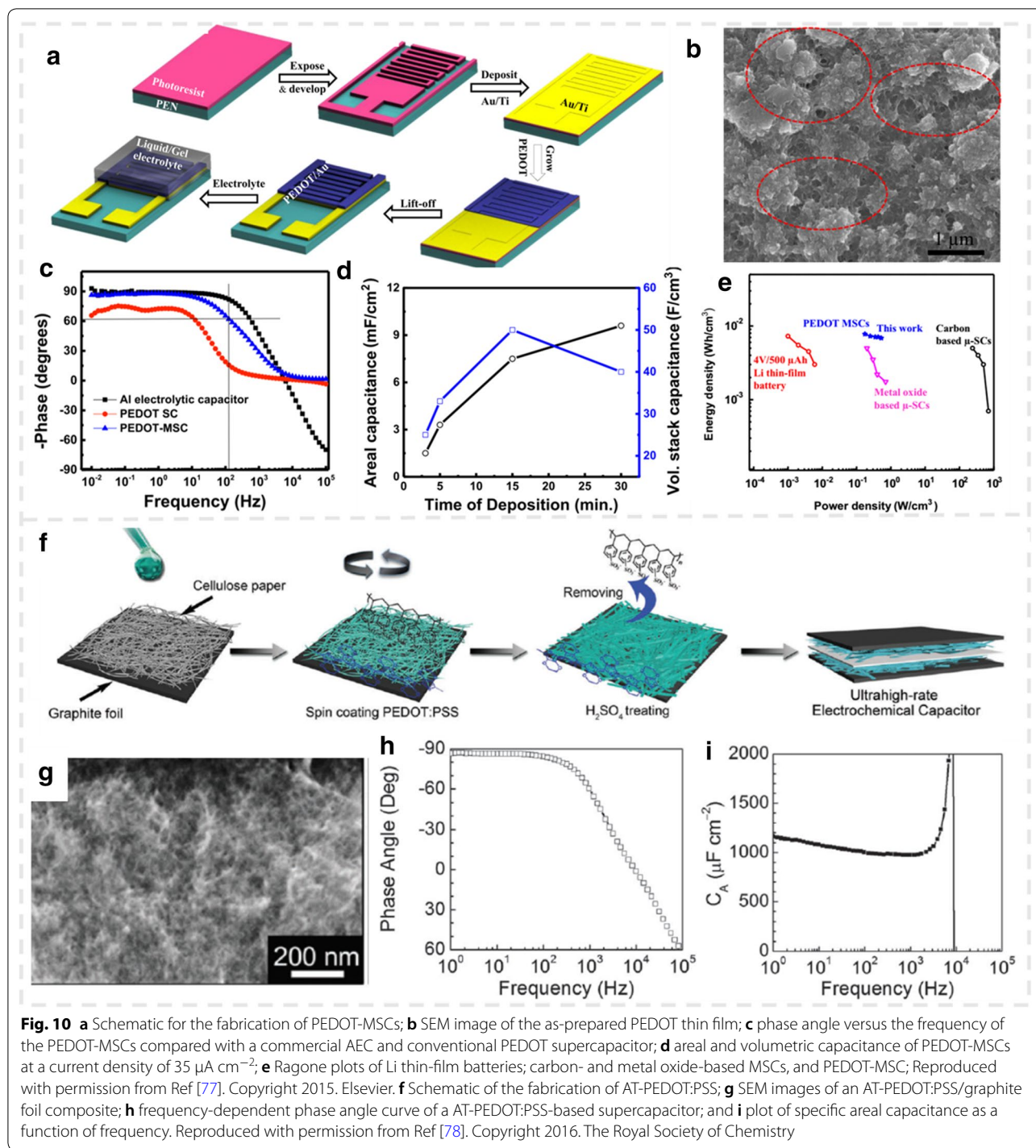


image showed that the micropore wall comprises nanoporous polymer nanoparticles (Fig. 10g). These material-based electrodes exhibited an excellent phase angle of  $83.6^\circ$  at 120 Hz with a short RC time constant of 0.15 ms (Fig. 10h). Concurrently, the ECs exhibited high areal capacitance ( $994 \mu\text{F cm}^{-2}$ ), which was the highest value among the reported ECs with a phase angle of higher than  $80^\circ$  at 120 Hz [12, 60, 74]. It provides the possibility of replacing AECs, thereby reducing the size of electronic devices in future.

This study [19] reported a layer-by-layer (LBL) method for preparing on-chip MSCs with AC line-filtering performance by increasing an azulene-bridged coordination polymer framework (PiCBA) on the inter-digital Au electrode surface. Figure 11a–c show the procedure. The TEM image indicated a uniform macroscopic lateral dimension of the obtained coordination polymer film (Fig. 11d). An impedance phase angle of  $-73^\circ$  was achieved at a frequency of 120 Hz (Fig. 11e).  $\tau_{RC}$  obtained by calculating the Nyquist plot with imaginary capacitance was as short as 0.83 ms (Fig. 11f), thus indicating superior AC line-filtering performance. Highlighting the overall performance of the MSCs, the Ragone diagram for

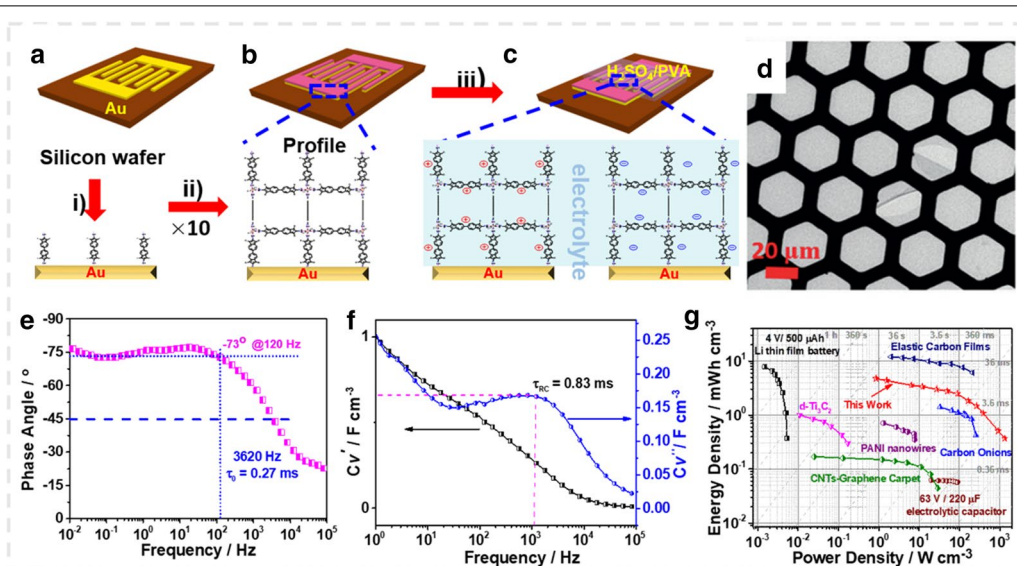
PiCBA-based MSCs exhibited a high energy and power density of  $4.7 \text{ mWh cm}^{-3}$  and  $1323 \text{ W cm}^{-3}$ , respectively (Fig. 11g). This study provided a potential solution for the fabrication of coordination polymer framework-based MSCs with AC line-filtering performance.

Supercapacitors have high capacitive properties; however, complex dynamics limit their frequency response in AC voltage. Park and coworkers [79] demonstrated porous MXene/conducting polymer hybrid-based ECs with AC line-filtering performance. Active materials were deposited through the spray-coating method, including  $\text{Ti}_3\text{C}_2\text{T}_x$  MXene (denoted as  $\text{Ti}_3\text{C}_2$ ) and  $\text{Ti}_3\text{C}_2$ /PEDOT:PSS hybrids, at a mass ratio of 1:1 and 1:2 (denoted as MP11 and MP12), respectively. Figure 12a shows the scalability of the spray-coating method for the ECs prepared with poly(vinyl alcohol)-hydroxyethyl methacrylate (PVA-HEMA) hydrogel. The SEM image of the hybrid thin film shows that the open mesoporous is essential to reduce pore tortuosity and ensure rapid charge transport. Figures 12c and d illustrate the Bode and frequency-dependent capacitance to verify the frequency response of  $\text{Ti}_3\text{C}_2$ , MP11, and MP12 to AC line filtering. The phase angles and RC time constants of  $\text{Ti}_3\text{C}_2$ ,

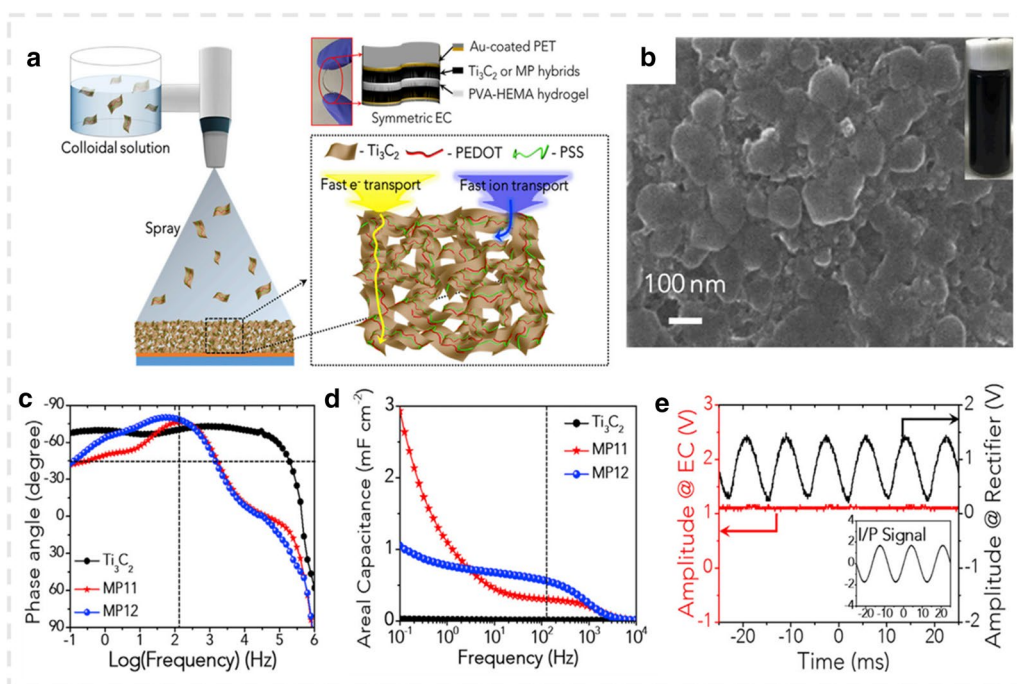


MP11, and MP12 were  $70.0^\circ$ ,  $76.1^\circ$ , and  $79.1^\circ$  at 120 Hz and 0.39, 0.12, and 0.29 ms, respectively (Fig. 12c). The areal/volumetric capacitances of  $\text{Ti}_3\text{C}_2$ , MP11, and MP12 ECs at 120 Hz were  $0.006 \text{ mF cm}^{-2}/0.24 \text{ F cm}^{-3}$ ,  $0.302 \text{ mF cm}^{-2}/8.51 \text{ F cm}^{-3}$ , and  $0.560 \text{ mF cm}^{-2}/24.2 \text{ F cm}^{-3}$ , respectively (Fig. 12d). The input AC signal was rectified

into the pulse signal and was then smoothed through series MP12 ECs, and the output constant DC voltage was 1.27 V (Fig. 12e). The performance gap between high capacitance and the high frequency response was eliminated in this work, and the form-factor-free miniature and scalable devices could be realized. These results



**Fig. 11** a–c Schematic of LBL fabrication of a PICBA film on Au interdigital electrodes; **d** TEM image shows a large area of a PICBA film on a copper grid; **e** impedance phase angle on the frequency for the PICBA-based MSC; **f** plot of capacitance versus the frequency of PICBA-based MSCs; and **g** Ragone plots compared the energy and power density for PICBA and reported supercapacitor and batteries. Reproduced with permission from Ref [19]. Copyright 2017. Wiley–VCH



**Fig. 12** a Schematic of the fabrication of a flexible Ti<sub>3</sub>C<sub>2</sub>/PEDOT:PSS hybrid electrode based on the spray-coating method; **b** SEM image of Ti<sub>3</sub>C<sub>2</sub>/PEDOT:PSS at a mass ratios of 1:2 thin film; **c** frequency-dependent phase angle, and **d** frequency-dependent areal capacitances of Ti<sub>3</sub>C<sub>2</sub>, MP11, and MP12 ECs; and **e** pulsating output signals across the rectifier and constant DC output across MP12 ECs at 60 Hz (Inset indicates AC input signal). Reproduced with permission from Ref [79]. Copyright 2019. Elsevier

emphasize the outstanding filtering performance of series MP12 ECs, which could be next-generation capacitors to replace AEC for AC line filtering.

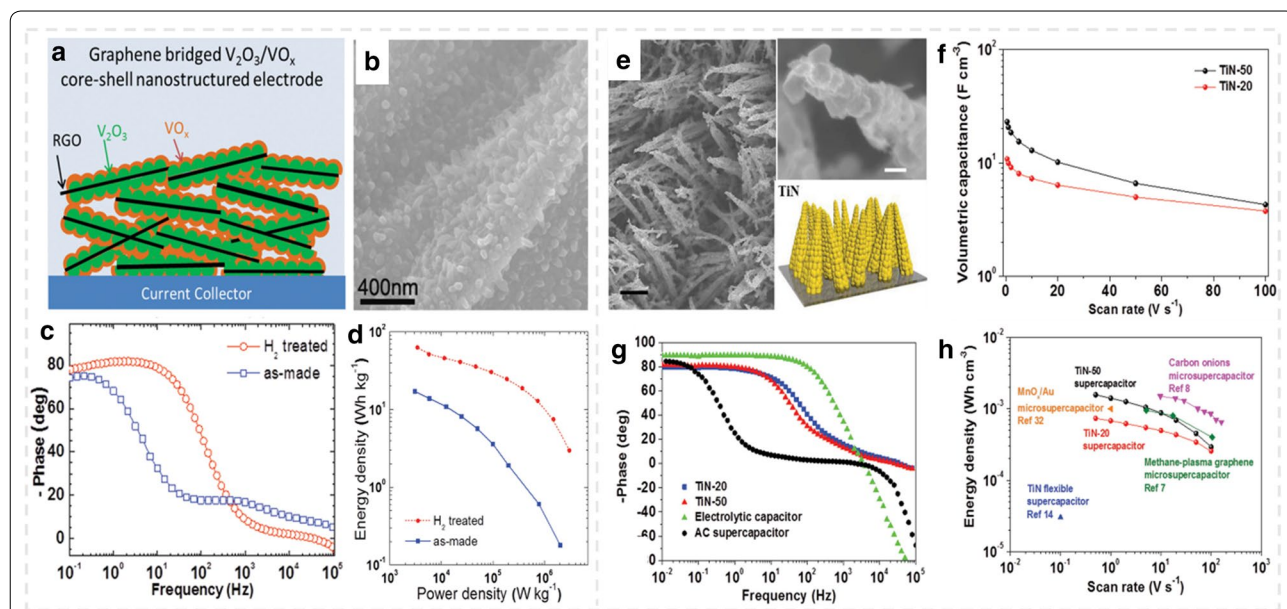
### Transition metal-based electrodes

Some interesting electrode materials combine a double-layer capacitor and pseudocapacitance effects and can be applied to the supercapacitor with AC line-filtering performance. For example, oxides, nitrides, and chalcogenides of transition metals have been widely investigated because of their additional pseudocapacitance contribution. Pseudo-capacitors can provide higher energy density through rapid redox reaction because of their charge transfer, but the limited electronic conductivity of most pseudo-capacitive oxides leads to high electrode resistance. Therefore, compared with electric double layer capacitors and electrolyte capacitors, pseudo-capacitors have lower power density [82]. To compensate for the low conductivity of electrode materials, pseudocapacitance materials are frequently used to bridge highly conductive graphene and similar carbon-based scaffolds.

Transition metal oxides are widely used in the development of high-energy-density supercapacitors because of the inherent large pseudocapacitance effect. However, a typical transition metal with high resistance would affect the charging and discharging rates of supercapacitors, thereby making their application for AC line filtering

difficult. To overcome the challenge of low conductivity, a graphene-bridged  $V_2O_3/VO_x$  core-shell nanostructure electrode was reported [80]. The quasi-metallic  $V_2O_3$  nanocores were dispersed on the conductive graphene sheet, and  $V_2O_3$  was naturally formed with  $VO_2$  and  $V_2O_5$  as active materials. The interconnected graphene and  $V_2O_3$  formed an electron conduction path, whereas the amorphous  $VO_x$  shell promoted ion insertion/extraction to achieve high rate performance (Fig. 13a). SEM images of the synthesized rice-shaped vanadium oxide nanoparticles showed that the average diameter of the nanoparticles was 40 nm, and the covered rGO sheets were uniformly monolayer without clustering (Fig. 13b). Furthermore,  $H_2$  heat treatment of the electrode material significantly improved conductivity. Such an EC with a charge-discharge rate of up to  $50 V s^{-1}$  provided a power density of  $1000 kW kg^{-1}$  at an energy density of  $10 Wh kg^{-1}$ . The characteristic frequency was measured as 114 Hz at a phase angle of  $45^\circ$  (Fig. 13c, d). These observations indicated that graphene-composited transition metal oxides could be favorable for rapid EC development.

In recent years, metal nitrides as electrode materials for high-performance supercapacitors have received considerable attention because of their excellent conductivity [81]. Among them, the conductivity of titanium nitride (TiN) is between  $4 \times 10^3$  and  $5.55 \times 10^4 S cm^{-1}$ , which is



**Fig. 13** **a** A schematic and **b** SEM image of an rGO-bridged  $V_2O_3/VO_x$  core-shell nanocomposite structure as an electrode for fast PCs; **c** Bode plots and **d** Ragone plots of supercapacitors fabricated from hydrogen-treated and as-made nanocomposite; Reproduced with permission from Ref 80. Copyright 2014. WILEY-VCH. **e** cross-sectional SEM image of TiN-50 (scale bar: black 1  $\mu m$  and white 100 nm); **f** volumetric capacitance versus the scan rate at 20 and 50 nm; **g** frequency-dependent phase angle for TiN-20, TiN-50, AECs, and an active carbon supercapacitor; and **h** volumetric energy density as a function of the scan rate. Reproduced with permission from Ref [81]. Copyright 2016. WILEY-VCH

close to that of metal and should improve the rate performance by providing rapid charge transport. Corn-like TiN nanostructures (Fig. 13e) exhibited capacitive performance at scan rates as high as  $100 \text{ V s}^{-1}$  in ultrafast-charging energy storage devices. Figure 13f, h compare the volumetric capacitance and energy density. A volumetric capacitance of  $20.7 \text{ F cm}^{-3}$  could be achieved.  $\tau_0$  values were approximately 4–2 ms (the corresponding characteristic frequencies were 40 and 80 Hz, respectively), which were considerably faster than 400 ms for the AC supercapacitor and was comparable to 0.3 ms for the electrolytic capacitor (Fig. 13g). At a scan rate of  $1 \text{ V s}^{-1}$ , TiN-50 and TiN-20 exhibit a volumetric capacitance of 20.7 and  $10.0 \text{ F cm}^{-3}$ , respectively, higher than that of active carbon (AC;  $9.0 \text{ F cm}^{-3}$ ) and onion-like carbon ( $1.3 \text{ F cm}^{-3}$ ) under the same measurement condition [42, 44]. All these results conspicuously suggest that TiN has great potential for ultrahigh power and energy. Moreover, transition metal sulfides belong to the category of pseudocapacitance electrode materials.  $\text{WS}_2$ -,  $\text{MoS}_2$ -, and  $\text{VS}_2$ -based electrodes have been reported mainly because of their relatively large surface area and pseudocapacitance [82, 83]. However, these materials have no frequency response. Such materials can be further explored for AC line filtering.

### AC line-filtering performance design

The design of supercapacitors with AC line-filtering performance depends on various influencing factors, including preparation of electrode materials, selection of electrolytes, method of preparing electrode materials, and assembly of the device. (1) The first concern is the preparation of electrode materials. Conventional supercapacitors with carbon-based materials exhibit a slow frequency response at 120 Hz because the complex pore structure restricts the transfer of charge [9]. Study and design of electrode materials with an adjustable pore structure or ordered pore structure for supercapacitors with AC line-filtering performance is necessary. Carbon materials have been developed as electrodes for most supercapacitors with AC line filtering, including VOGNs [12, 27, 60, 83], ErGOs [9], and CNTs [30, 48, 74], [9], graphene-PEDOT:PSS [26], and conducting polymers [19, 78–80]. The excellent frequency response of these carbon materials is primarily because of suitable conductivity and the open porous structure for the rapid transportation of electrolyte ions. Therefore, the rational designing and fabrication of highly conductive electrodes with optimized porous nanostructures can improve the performance of supercapacitors with AC line filtering. (2) The second concern is the selection of electrolytes. Aqueous electrolytes, organic electrolytes, and ionic liquids are the three primary types of electrolytes commonly

used in supercapacitors. Among them, the aqueous electrolyte shows a rapid frequency response because of its high ionic conductivity; however, the voltage window is limited to 1 V, thus rendering the low energy density of supercapacitors. Ultrafast organic ECs [38] have organic electrolytes, such as the acetonitrile (AN) solution of tetraethylammonium-tetrafluoroborate. The voltage window can be extended to approximately 3 V, thus providing an energy density of  $472 \mu\text{F V}^2 \text{ cm}^{-2}$ , and the phase angle is  $-80.5^\circ$  at 120 Hz. Kim's group [17] developed a 3-V flexible supercapacitor for AC line filtering by using an ionic-liquid-based polymer as a gel electrolyte. The supercapacitor exhibited 20 times higher areal energy density than the 1-V flexible supercapacitor (0.66 vs  $0.03 \mu\text{Wh/cm}^2$ ). The expansion of the voltage window while maintaining the fast frequency response capability considerably increases the potential for the high-frequency application of supercapacitors in electronic devices. (3) The third concern is the rational preparation of electrode materials. So far, well-established synthetic methods, including electrochemical reduction, CVD, wet chemical synthesis, vacuum filtration, interfacial polymerization, laser scribing, inkjet printing, and screen printing, have been used for the preparation of electrode materials for supercapacitors with AC line-filtering performance [9, 19, 22, 26, 76, 84, 85]. The electrode materials prepared through different synthetic methods could have different structural characteristics, thus resulting in rapid mass transportation and excellent electrochemical properties. However, developing the structure–property relationship between synthetic methods, porous structures, specific capacitance, and AC line-filtering performance is difficult. (4) Finally, the configuration of the device is a concern. The structure and morphology of the electrode material can be controlled to ensure that the electrolyte permeates completely, and thus, the supercapacitor has AC filtering performance [87]. In view of the emergence of different electrode materials, the development of supercapacitors with excellent electrochemical properties provides a new possibility for energy storage. Conventionally, conventional supercapacitors have the sandwich-type structure [11]. In this case, the electrolyte is generally restricted from completely penetrating the electrode material, thus reducing the complete utilization of the electrochemical surface area of electrode materials. It is rarely expected to replace AECs with a large volume in electronic circuits for AC line filtering. An “in-plane” fabrication approach for ultrathin supercapacitors can guarantee each electrode with an effective mass (ions) contact without using a separator. Such a planar structure established a solid foundation for the further development of interdigital electrode MSCs. So far, several well-established synthetic methods have been used



**Table 1 Representative results of supercapacitors with AC line-filtering performance**

Electrode materials	Electrolyte	Phase angle at 120 Hz	f (kHz) at -45°	$\tau_0$ (ms)	$\tau_{RC}$ at 120 Hz	Specific capacitance	References
VOG	KOH	85	20	0.05	-	265 $\mu\text{Fcm}^{-2}$	[60]
VOG	KOH	~90		32.8	-	30 $\text{F g}^{-1}$	[81]
ErGO	KOH	84	4.2	0.17	1.35	283 $\mu\text{Fcm}^{-2}$	[9]
GO film	H <sub>2</sub> SO <sub>4</sub>			13.3		125 $\text{F g}^{-1}$	[85]
EOG	KOH	82	4.03		0.205	0.72 $\text{mFcm}^{-2}$	[86]
EOG	KOH	83	12		0.110	0.6 $\text{mFcm}^{-2}$	[88]
NHG	KOH	81.2	1.2		0.203	0.956 $\text{mFcm}^{-2}$	[89]
GO@CNT	NaSO <sub>4</sub>	81.5		0.82	0.19	662 $\mu\text{Fcm}^{-2}$	[14]
CNT	H <sub>2</sub> SO <sub>4</sub>	81	1.425	0.701	0.199	1.202 $\text{mFcm}^{-2}$	[30]
SWCNT	TEABF <sub>4</sub>	82.2	1.995		0.501	0.564 $\text{mFcm}^{-2}$	[71]
GOMC/CNT	TEABF <sub>4</sub>	80.0	1.0	1.0		0.56 $\text{mFcm}^{-2}$	[70]
Carbon sponges	KOH	78		0.37	0.32	172 $\text{mFcm}^{-2}$	[73]
PENOT:PSS	H <sub>2</sub> SO <sub>4</sub>	84		0.59	0.15	994 $\text{mFcm}^{-2}$	[75]
PiCBA	H <sub>2</sub> SO <sub>4</sub> /PVA	73	3.62	0.27	0.83	34.1 $\text{Fcm}^{-3}$	[19]
Ti3C2/PEDOT:PSS	H <sub>2</sub> SO <sub>4</sub> /PHEMA/PVA	79.1	1.41	0.709	0.29	0.56 $\text{mFcm}^{-2}$	[76]

to prepare in-plane MSCs, such as laser scribing, inkjet printing, and photolithography. Many in-plane MSCs exhibit a superior frequency response, which allows their application in next-generation electronic devices for AC line filtering.

The design of supercapacitors with AC filtering performance could be affected by the above mentioned or other factors, e.g., the electrode material with high conductivity, controllable pore size matched the sizes of electrolyte ions, the distance between the electrodes is short enough to promote the rapid transportation of electrolyte ions. Therefore, micro-sized supercapacitor has sufficient advantages in the synthesis of capacitors with AC filtering properties. To provide an improved overview of this field, Table 1 presents the recent progress in AC line-filtering supercapacitors.

**Conclusion**

In this review, we summarized the research works in the field of supercapacitors with AC filtering performance. The applications of different electrode materials in the preparation of supercapacitors with AC line filtering are discussed. At the same time, some primary factors which could affect the AC filtering performance have been discussed. Compared with the traditional carbon-based electrode materials, the design of electrode materials with high electronic conductivity and using selected electrolytes with suitable aperture, the micro-supercapacitors with short electrode distance are more suitable for the development of capacitors with AC filtering performance.

Because in 2010, Miller first reported the new application of vertically oriented graphene-based supercapacitors in AC filtering, the interesting field has received considerable attention from various researchers and made unexpected remarkable progress. In addition to graphene-based electrode materials, several other electrode materials, including carbon-based materials (CNT, OLC, carbon sponges) and noncarbon-based materials (conducting polymer and transition metal), have been used in the research of AC line filtering and made outstanding achievement. On the basis of the research and discussion of designing supercapacitors with excellent overfiltering performance in the previous study, developing AC line filtering by synthesizing highly conductive electrode materials with an ordered pore structure and selecting an appropriate preparation process and electrolyte for device assembly would be ideal. Although a large number of supercapacitors have been reported and applied to AC line filtering, more efforts are required to apply them to practical electronic circuits, thus encouraging researchers to further develop supercapacitors for AC line filtering and other potential applications.

**Abbreviations**

AC: alternating current; DC: direct current; AECs: aluminum electrolytic capacitors; EDLCs: electrical double-layer capacitors; PCs: pseudocapacitors; EC: electrochemical capacitors; VOGN: oriented graphene nanosheet; CVD: chemical vapor deposition; ErGOs: electrochemically reduced graphene oxides; OMC: ordered mesoporous carbon; DLCs: double-layer capacitors; RC: resistance capacitance; CNT: carbon nanotube; MSCs: micro-supercapacitors; FS: femto-laser; NCNT: nitrogen-doped carbon nanotube; AFM: atomic-force microscopy; MG-MSCs: graphene-based micro-supercapacitors; SDS: sodium dodecyl sulfate; PEDOT: porous conductive polymer; PiCBA: azulene-bridged coordination polymer framework; TiN: titanium nitride.

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**Authors' contributions**

XZ, SH and EK conceived the content of the article and revised the text. DZ collected the literature, DZ, KJ, JL, XZ, CK co-wrote and revised the paper. All authors read and approved the final manuscript.

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**Availability of data and materials**

Not applicable.

**Competing interests**

X. Zhuang and E. Kymakis are the Editorial Board Member and Section Editor of *BMC Materials*, respectively. The authors declare that the draft has gone through official peer review procedure before publication.

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