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Research on the Application of a MnO_2-Based Flexible Supercapacitor in AC Filtering

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Abstract: Aluminum electrolyte capacitors (AEC) are widely used in AC filtering as traditional filter capacitors. However, their strong rigidity, high hardness, and high-risk factor have hindered the flexible development of filter capacitors. Flexible supercapacitors, due to their high energy density and outstanding cycle stability, are becoming one of the main directions for the development of filter capacitors in the future. This paper self-assembles a MnO_2-based flexible supercapacitor with high area capacitance and fast frequency response, and preliminarily verifies the feasibility of using the capacitor for AC filtering through simulation tests in Multisim. Finally, the AC filtering test of the flexible supercapacitor using an oscilloscope demonstrates that the ripple factor in the range of 2.7% to 8%, which confirms that the MnO_2-based flexible supercapacitor has a great AC filtering function and can replace traditional aluminum electrolyte capacitors in AC filtering, promoting the flexible development of electronic products.

Keywords: Flexible supercapacitor; AC filtering; MnO_2; Frequency response

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1. Introduction

In the ever-developing modern technology, to ensure the stability of electronic equipment during signal transmission and reduce the interference of AC signals, it is necessary to filter out AC signals and output constant current signals [1, 2]. In the current electronic circuit AC filter system, aluminum electrolytic capacitors (AEC) are widely used as filter capacitors [3–5]. However, with the rapid development of highly integrated circuits in supercomputers, electric vehicles, aircraft, and other fields, filter capacitors are forced to develop towards small size, large capacitance, flexibility, and miniaturization. Supercapacitors (SC), also known as electrochemical capacitors (EC), are a new type of energy storage device derived from the double-layer theory of electrode plates and electrolytes, belonging to the device between traditional capacitors and batteries [6–8]. Due to the unique advantages of SC such as high energy density, low cost, and long cycle life, it has been widely studied and widely used in electronics, data storage systems, industrial power supplies, and energy management. Therefore, supercapacitors are very likely to replace AEC and become the next generation of new AC filter capacitors.

Up to now, research on the application of supercapacitors in AC filtering has also broadened its application to a certain extent and verified that supercapacitors can be used as filter capacitors. For instance, Wu *et al.* designed an asymmetric supercapacitor (PEDOT//ErGO) and applied it to AC filtering [9]. Fan *et al.* prepared carbon samples from various precursors (ZIF 67, Prussian blue, and cellulose). The high-frequency response of the electrode material verified its use for AC line filtering [10–12]. Zhang *et al.* reported a hybrid carbon nano-onion/graphene symmetric supercapacitor (SSC), which can be used as a compact AC filter [13]. Recently, Gogotsi *et al.* explored the AC filtering properties of 2D MXene by changing the electrode thickness, and selected reduced graphene oxide as the electrode to assemble the device [14]. Its 1.35 ms confirmed that it can replace AEC to achieve AC signal filtering [15]. Soomin *et al.* made a flexible supercapacitor based on PEDOT: PSS material that can maintain an ultrafast response speed at 60 Hz [16]. Marinppan *et al.* designed an electrode material composed of SP/SP2 hybrid carbon, and the supercapacitor made of this electrode material showed excellent filtering performance in a wide frequency range [17]. Xue *et al.* used a three-dimensional graphene film with a porous structure to make a capacitor for AC filter with a resistance-capacitance time constant of less than 1 ms [18].

In summary, various supercapacitors have been designed using different electrode materials for AC filtering to replace AEC and promote the development of device flexibility. However, the current research on supercapacitor electrode materials that can replace AEC is mainly oriented to the research of various carbon structure materials. The material types studied are relatively single, and there is less research on other materials. Therefore, it is very necessary to develop new materials with excellent AC filtering performance to assemble supercapacitors to replace AEC, which also promotes the development of flexible filter capacitors.

In this work, MnO_2-based flexible supercapacitors (FASCs) were self-assembled using MnO_2 as the electrode material. The stability, flexibility, and filtering performance of the FASCs were analyzed. In addition, the filtering effects of the FASCs with different waveforms at different frequencies were explored. The results show that the constructed FASCs exhibit excellent rate performance (77%, 50 times), outstanding cycle stability (capacity retention rate of 95% after 10,000 cycles), and fast frequency response (approximately 3.2 ms, equivalent series resistance of about 1.5 Ω). At the same time, the flexible supercapacitors have no obvious performance loss in the bending and folding state, showing excellent flexibility. In addition, the self-assembled FASCs can successfully filter AC signals with different waveforms in the 1 Hz–100 kHz frequency band and output stable signals. Not only has the filtering frequency band of traditional filter capacitors been broadened, but the flexible development of filter capacitors has been promoted to a certain extent.

2. Results and discussion

In this paper, the MnO_2-based flexible supercapacitor was assembled with δ-MnO_2 as the positive electrode of the supercapacitor, activated carbon (AC) as the negative electrode, PVA-Na_2SO_4 electrolyte gel as the electrolyte part, and carbon cloth as the current collector. The energy density, flexibility and stability of the device were tested.

To evaluate the actual performance of the δ-MnO_2 electrode, the device assembly schematic is shown in **Figure 1(a)**, and FASC was assembled with δ-MnO_2 and AC as the positive and negative electrodes, respectively. Next, a bending test was conducted on the flexible asymmetric supercapacitor to verify its flexibility. As shown in **Figure 1(b)**, the photos of the flexible asymmetric supercapacitor with bending angles of 45°, 90°, and 180° vividly demonstrate the excellent flexibility of the device. Excitingly, the CV curves at different bending angles were observed, and the curve shapes basically overlapped with each other as seen in **Figure 1(c)**, which shows that the FASC device has excellent flexibility and stability. Subsequently, a fixed current density (15 mA·cm^{-2}) was set to analyze the stability of the device under long-term charging and discharging, as seen in **Figure 1(d)**. After 4000 cycles, the capacitance of δ-MnO_2//AC FASC can still be maintained at around 87%, demonstrating excellent cycling stability.

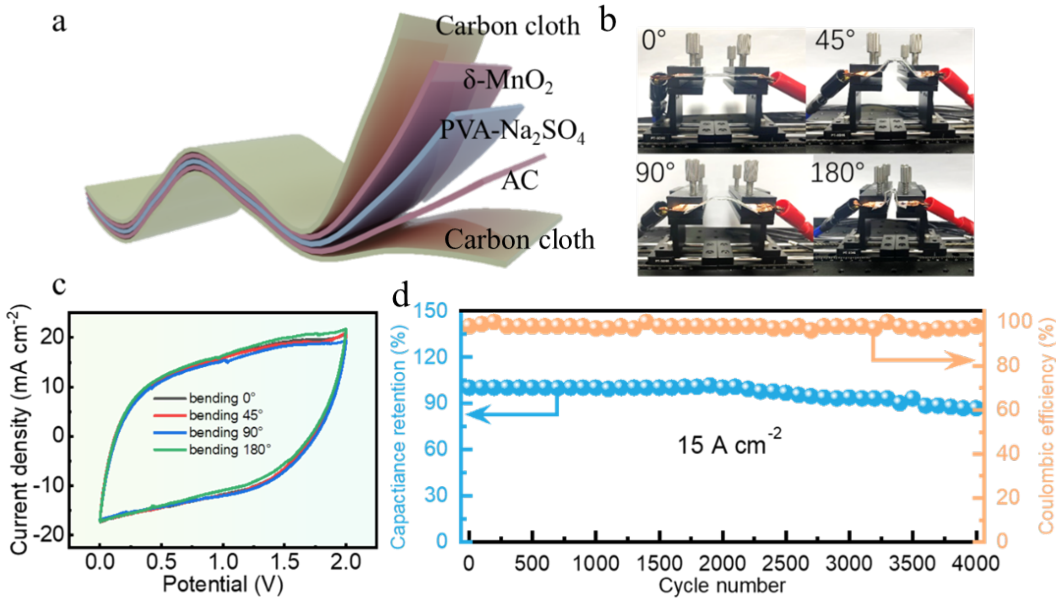

Figure 1. (a)Schematic illustration of the preparation of δ-MnO_2@AC FASC; (b-c) digital photos and capacitance retention performances of δ-MnO_2//AC FASC devices bent at different angles of 0°, 45°, 90°, 180° at 50 mV s^{-1}; (d) Cycling performance of the δ-MnO_2//AC device measured at 15 A cm^{-2} for 4000 cycles.

The area specific capacitance of the device is calculated as follows:

$$C = \frac{I \cdot \Delta t}{\Delta v \cdot S} \quad (1)$$

From Equation 1, I is the current (A); Δt is the discharge time interval (s); S is the effective area (cm^2); ΔV is the potential difference (V).

The calculation equations for energy density and power density are as follows:

$$E = \frac{C \cdot \Delta V^2}{2 \times 3.6} \quad (2)$$

$$P = \frac{3600E}{\Delta t} \quad (3)$$

Here, C is the capacitance, ΔV is the potential window, and Δt is the time it takes to discharge once.

Subsequently, a fixed current density (15 mA·cm^{-2}) was set to analyze the stability of the device under long-term charge and discharge(Figure 1d). After 4000 cycles, the capacitance of the FASC can still be maintained at about 87%, showing excellent cycle stability. Energy density and power density can be obtained from equations (1) to (3). At a power density of 800.5 µW·cm^{-2}, the FASC device shows a high energy density of 53.2 µWh·cm^{-2}, and when the power density rises to 4930 μW·cm^{-2}, the energy density can be maintained at 43 μWh·cm^{-2}.

The assembled FASC exhibits excellent flexibility and outstanding capacitance. However, further analysis is needed to determine whether it can be applied in the field of AC filtering. Frequency response is one of the main methods to study the AC filtering performance of capacitors, where low-frequency response represents the capacitance characteristics. To further verify whether the prepared MnO_2-based flexible asymmetric supercapacitor meets the basic requirements of basic AC filtering capacitor filtering.

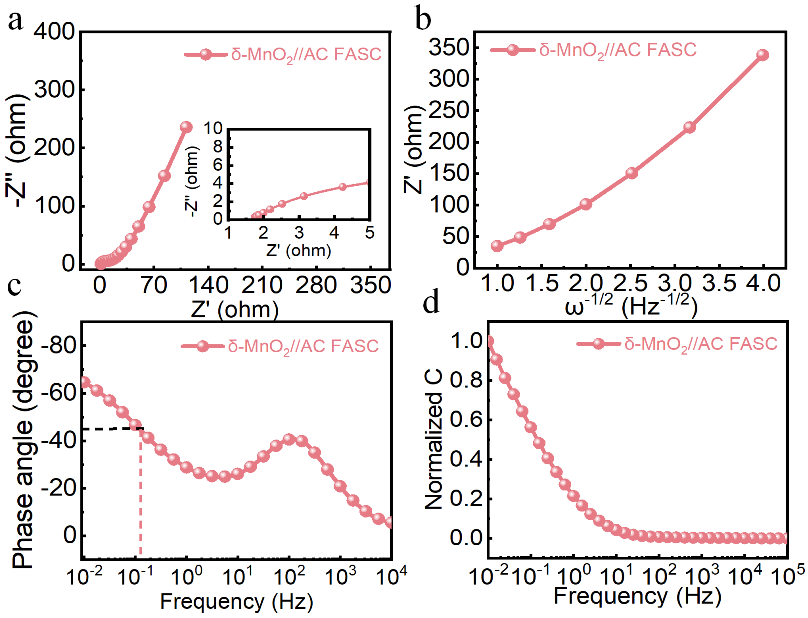

Figure 2. (a) Nyquist plots of δ-MnO_2@AC FASC; (b) The correlation between Z' and $\omega^{-1/2}$ in the low-frequency area; (c) EIS result of δ-MnO_2@AC FASC; (d) Normalized capacitance of δ-MnO_2@AC FASC

Figure 2 shows the frequency response result of the device. As shown in **Figure 2(a)**, the complex plane diagram of δ-MnO_2//AC shows a larger closed slope in the low-frequency range, indicating that it is due to its own rapid electron/ion diffusion ability. In addition, the vertical line characteristics of the capacitance behavior are also obtained by the impedance complex plane diagram shown in **Figure 2(a)** (enlarged diagram of the high-frequency region). The assembled flexible supercapacitors do not have the charge transport semicircle of traditional supercapacitors at high frequencies. More importantly, the enlarged results of the high-frequency region of the impedance complex plane show that the equivalent series resistance (ESR) of δ-MnO_2//AC FASC is 1.83 Ω and 1.5 Ω, respectively. It is worth noting that the equivalent series resistance plays a decisive role in the performance of the capacitor. The smaller the value of ESR, the better the performance of the capacitor, and it is more suitable for AC filtering. In addition to looking at the ESR, the RC time constant (τ_{RC}) is also a very important parameter to determine whether a capacitor is suitable for filtering. It can be observed from **Figure 2(b)** that the RC time constant values of the two supercapacitors are both less than 8.3 ms, proving that δ-MnO_2//AC FASC can be used as an AC filter to achieve the filtering function of the AC signal. This result shows that our flexible asymmetric supercapacitor has great potential to replace AEC ($\tau_{RC} \leq 8.3$ ms) in AC filtering applications. As depicted in **Figure 2(c)**, it can be observed that the inflection point frequency value of δ-MnO_2//AC FASC is within an excellent range, indicating its excellent magnification performance. Additionally, as shown in **Figure 2(d)**, the normalized value of the assembled flexible supercapacitor can be maintained at around 1, which means that FASC has good capacitance behavior and excellent frequency response characteristics. After EIS testing of the flexible supercapacitor, whether it is capacitance value, equivalent series resistance, or RC time constant, it has met the requirements of the filter capacitor.

Next, to evaluate the feasibility of the assembled capacitor in actual AC filtering applications, a filtering circuit was built based on the Multisim platform and carried out simulation tests to confirm that the increase in capacitance promotes the optimization of filtering performance. Based on the oscilloscope platform, the actual filtering performance of the MnO_2-based flexible supercapacitor was tested.

The capacitor value used in the filter circuit is adjusted to 145 mF and the simulated circuit diagram and simulation results are shown in **Figure 3**.

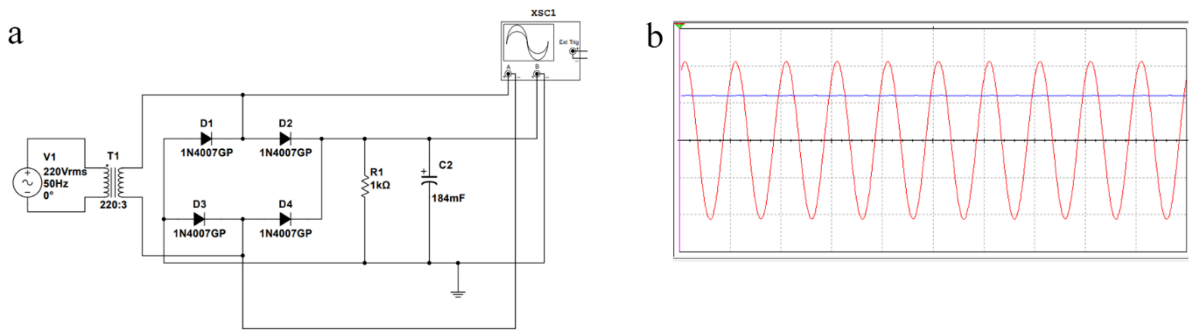

Figure 3. (a) Multisim filter circuit diagram; (b) AC filter simulation test results

From the simulation results, it can be seen that the assembled flexible asymmetric supercapacitor device can perform the filtering function of the AC signal very well, which preliminarily confirms the filtering performance of the MnO_2-based flexible asymmetric supercapacitor. To comprehensively evaluate the actual filtering of FASC, a filtering circuit was built using a bridge circuit, load resistors, and filter capacitors, as shown in **Figure 4**.

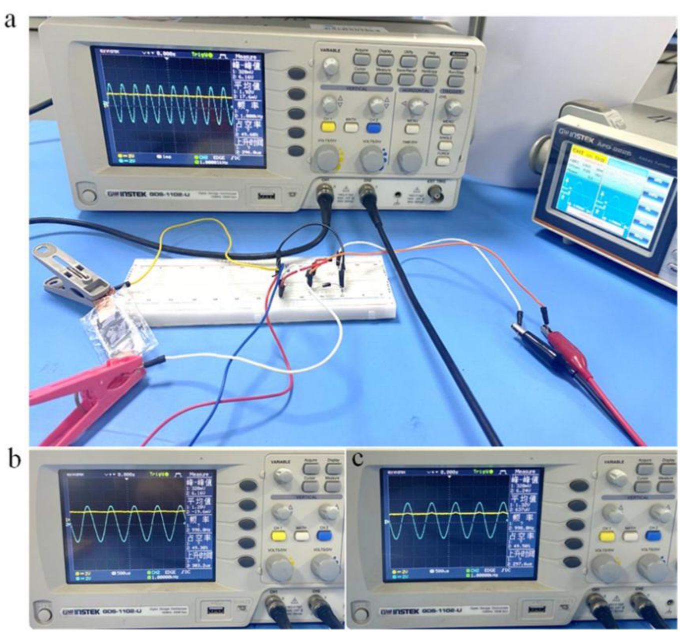

Figure 4. (a) Actual filtering display; (b) MnO_2-based FASC filtering result; (c) Ordinary aluminum electrolyte filtering result

The corresponding signal was input using a function generator (GWINSTEK AFG-2225), and the filtering output was displayed using a GWINSTEK GDS-11-2-U oscilloscope. Using a breadboard as a platform, a simple filtering circuit, as shown in **Figure 4(a)**, was built. The entire filtering circuit consists of four rectifier diodes (model: 1N4148) to form a bridge circuit, and a resistor with a resistance of 1 kΩ is used as a load. A 1000 Hz AC signal is filtered using an ordinary 220 μF aluminum electrolyte capacitor and a MnO_2-based flexible supercapacitor.

As shown in **Figure 4(b)**, the MnO_2 FASC can achieve AC filtering under sinusoidal AC signals. Under the action of the flexible supercapacitor, the input sinusoidal AC signal can successfully filter the 1000 Hz AC signal into a smooth DC signal through the prepared supercapacitor. In addition, a filtering test was conducted using an ordinary aluminum electrolyte capacitor, and the filtering effect was the same as that of the water-based δ-MnO_2//AC flexible supercapacitor, as shown in **Figure 4(c)**. Based on the oscilloscope results, the ripple factor was calculated. The ripple voltage (Vrpp) was determined from the peak-to-peak value (Vpp) using the formula:

$$Vrpp = 1/2 \times Vpp$$

The Vrpp calculated was 0.16V. Given that the input signal voltage is 3 V, the ripple factor, which is the ratio of the ripple voltage to the input signal voltage, was found to be 5.3%. This satisfies the requirement for a filtering ripple factor of less than 10%, confirming that the MnO2-based FASC can effectively serve as an AC filter.

After verifying the feasibility of the assembled δ-MnO_2//AC flexible asymmetric supercapacitor in AC filtering, the filtering capabilities of the δ-MnO_2//AC flexible asymmetric supercapacitor and the traditional aluminum electrolyte capacitor at the same frequency was compared. The rectifier filter circuit built on the breadboard was also replaced with the δ-MnO_2//AC flexible asymmetric supercapacitor and the traditional aluminum electrolyte capacitor prepared by us. The specific data results are shown in **Table 1**.

Table 1. Comparison of ripple coefficients of AEC and δ-MnO_2//AC FASC at different waveform signals at 1 Hz.

Capacitor Type	Frequency (Hz)	Input signal	Ripple Factor (%)	Is it satisfied? (≤ 10%)
Aluminum electrolytic capacitors	1	Sine wave	10.6	no
		Square wave	6.7	yes
		Triangle wave	9.3	yes
δ-MnO_2//AC FASC	1	Sine wave	8	yes
		Square wave	4	yes
		Triangle wave	5.3	yes

From the comparison results, it can be seen that compared with aluminum electrolyte, the filtering effect of δ-MnO_2//AC FASC at low frequency is better than that of aluminum electrolyte capacitors. Even at 1 Hz, when the input signal is a sine wave, the ripple factor of δ-MnO_2//AC FASC is less than 10%, while the ripple factor of traditional aluminum electrolyte capacitors is 10.6% (> 10%), indicating that δ-MnO_2//AC FASC can replace aluminum electrolytic capacitors in AC filtering, and δ-MnO_2//AC FASC is more suitable for AC filtering.

Conclusions

In summary, δ-MnO_2//AC FASC can achieve AC filtering under a variety of AC signals. Under the action of the aqueous δ-MnO_2//AC flexible supercapacitor, all sinusoidal AC signals can successfully output smooth signals through the prepared supercapacitor. In addition, even when the input signal is a square wave or a triangle wave, it still maintains basic filtering performance and the filtering coefficient is maintained at 2.7%–8%. The results verify the wide applicability of δ-MnO_2//AC FASC. At the same time, the device has advantages in AC filtering due to the smallest possible RC time constant, excellent capacitance value, and low equivalent series resistance, which also echoes the frequency response results of δ-MnO_2//AC FASC.

Disclosure statement

The authors declare no conflict of interest.

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