International Journal for Housing Science and Its Applications
Publish August 6, 2025. Volume 46, Issue 3 Pages 8882-8890

https://doi.org/10.70517/ijhsa463760

Application and Performance of Cellulose Based Electrode Materials in Flexible Supercapacitors

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Abstract Flexible supercapacitor has the advantages of short charging and discharging time, high power and durability. As a portable power energy storage device, it shows great development potential. So far, there are still some technical problems in flexible supercapacitors with excellent electrochemical and structural mechanical properties. For this reason, this paper would use cellulose based electrode materials to study the performance of flexible supercapacitor. It is found that the curves of galvanostatic charge discharge under different current densities are similar to symmetrical triangles when the flexible supercapacitor is made of it. It shows that the capacitor has excellent electrochemical reversibility. At the same time, the charge discharge cycle test is carried out using the flexible supercapacitor made of the capacitor and the flexible supercapacitor made of biomass carbon materials. It is found that the materials studied in this paper can make the charge discharge cycle of flexible supercapacitor more stable. The final stability is 94.75% after 12000 cycles. In this paper, 20 experts were selected to evaluate the flexible supercapacitor made of two materials. The score based on the materials studied in this paper was above 4.3, while the score based on biomass carbon materials was below 4. Cellulose based electrode materials can be used to produce flexible supercapacitor with shorter production time than biomass carbon materials, stronger conductivity and strong practicability. Therefore, it is meaningful to study the performance of flexible supercapacitor based on cellulose electrode materials.

Index Terms Flexible Supercapacitor, Fiber Electrode Material, Biomass Carbon Material, Galvanostatic Charge Discharge

I. Introduction

With the continuous expansion of market demand for flexible and intelligent wearable electronic components, the market demand for flexible energy storage machines has also expanded greatly. The super capacitor with small size, excellent performance and safety has become one of the hot topics in the scientific research of energy storage machinery and equipment [1], [2]. For the defects of the new lithium metal battery, some scientific research has been completed recently to explore the flexible kinetic energy storage, so as to apply it to such flexible electronic devices as electronic displays, physical and mental health belts, etc. Flexible electronic devices have a variety of innovative applications, such as flexible circuits, compatible RFID tags, electronic paper and touch screens. As a new type of lightweight and energy-saving energy storage equipment, flexible supercapacitor has many advantages. In recent years, there are some micromachining technologies related to the design and preparation of supercapacitors, which have also made great progress. However, the whole preparation process is still complicated, the prepared flexible supercapacitor substrate is unstable, or it is relatively lower than the capacitor, which can not better meet the application conditions under actual conditions [3]. As an important electrode material of supercapacitors, graphene has also been widely concerned, but there are some shortcomings. In this environment, this paper would study another material to make up for the defects of flexible supercapacitor made of graphene materials. Based on cellulose based electrode materials to study flexible supercapacitor, this paper applies this material to flexible supercapacitor to study its influence on flexible supercapacitor performance. It provides a new idea for the development of flexible supercapacitor, and describes the installation method of chemical fiber based electrode materials in flexible power capacitors.

With the increasing demand for flexible displays, many experts and scholars have deeply discussed how to combine them with supercapacitors and how to design flexible supercapacitors, and achieved a series of research results. Cherusseri Jayesh believed that supercapacitor has superior energy storage characteristics, which made it a safer alternative for wearable applications. He focused on the new development in the field of metal organic skeleton composite electrodes for flexible supercapacitors [4]. Lu Chao proposed a full temperature flexible supercapacitor, which enhanced the thermal stability of the hydrogel. He used antifreeze and heat stable materials to form a directional conductive path to promote ion conduction, so that the full temperature flexible supercapacitor has excellent cycle stability [5]. Lu Nan thought hydrogel electrolyte has received great attention in the field of flexible supercapacitors due to its inherent safety. Flexible supercapacitors containing the best antifreeze organic hydrogen gel electrolyte exhibit excellent mechanical and electrochemical stability at sub freezing temperatures [6]. Kim Dong Won developed superheat, flexible and high-performance supercapacitors that operate under high temperatures and mechanical stresses. The multifunctional supercapacitor is prepared by using composite polymer electrolyte. In the flexible test under dynamic mode, the device also shows high long-term stability and mechanical durability after bending cycle [7]. The vast majority of scholars have carried out various researches on flexible supercapacitors, but few scholars have considered the use of cellulose based electrode materials in the containers studied in this paper.

Cellulose based electrode material is a very good material with a wide range of applications. Many scholars have applied it to flexible supercapacitors for research, and the research literature is endless. Sun Zhe believed that supercapacitor, as an advanced energy storage device, has the characteristics of portability and stable performance. He believed that natural polysaccharide cellulose and its derivatives, based on high aspect ratio, specific surface area, porosity, mechanical properties and excellent flexibility, have become promising building polymers for flexible electrode materials [8]. Chang Libo synthesized a pearl layered composite membrane prepared by hydrothermal process of cellulose nanofibers and sodium lignosulfonate. The electrodes of the flexible solid state supercapacitor assembled by it have excellent volume specific capacitance, excellent energy density and excellent electrochemical cycle stability [9]. Tanguy Nicolas R prepared a kind of mechanically strong flexible film electrode with high energy storage performance by using the Faraday reaction of the covalent combination of quinone group in natural lignin and high-strength cellulose nanofibers. It endows the composite electrode with mechanical strength and flexibility, enhances the overall energy storage performance, reduces the cost and environmental footprint, and enhances the mechanical and energy storage performance [10]. Ding Chenfeng synthesized porous carbon and skeleton to form a solid all solid cellulose based supercapacitor with ultra-high area energy density, volume energy density and excellent cycle stability. This achieves significant specific capacitance, including ultra-high weight and volume capacitance even at high current densities [11].

In the current environment of urgent shortage of electric power and energy, supercapacitor, an energy storage device, has received great attention. In addition, it should focus on applying it to wearable electronic components. Because of the rapid development of portable and wearable electronic products, people's demand for flexible and even folding high-performance energy storage systems is also increasing. The flexible supercapacitor has become one of the best choices for energy storage devices of flexible electronic products with its own super advantages [12]. Therefore, how to improve the energy density, power, conductivity and cycle reliability of flexible supercapacitors is an urgent problem to be solved. Therefore, this paper would study the performance of flexible supercapacitor based on cellulose electrode materials, and find that it can actually reduce the cost of flexible supercapacitor production, expand the production scale, and maintain the stability of charge discharge cycle operation.

II. Flexible Supercapacitor and Cellulose Related Theories

II. A. Overview of Capacitor

II. A. 1) Introduction to Super Capacitor

Supercapacitor is a kind of photoelectrochemical equipment based on the process of ion adsorption and slippage generated by positive ions of electrolyte solution on the surface or near the surface of electrode materials to complete energy storage and release. Therefore, supercapacitors are also called photoelectrochemical capacitors [13], [14]. According to the difference of energy storage principle, supercapacitors can be divided into two types. At the same time, according to the above two energy storage systems, the corresponding double layer energy storage raw materials represented by carbon materials have been cultivated. In addition, according to the needs of the use scenario, it needs to provide building countermeasures for different component structures. The energy storage of electric double layer supercapacitor is a physical phenomenon, so it has relatively fast charging and discharging speed and relatively high power. According to the energy storage principle of electric double-layer supercapacitor, the contact surface area and interface characteristics between electrode materials and electrolyte solution are important factors to determine the characteristics of the capacitor. Because it is subject to the principle of surface positive charge storage, the double-layer supercapacitor has fewer positive charges, resulting in low specific capacity and energy [15]. Therefore, the electrode materials of general electric double-layer supercapacitors need very high surface area to enhance the specific capacity and specific energy. The schematic diagram of supercapacitor is shown in Figure 1.

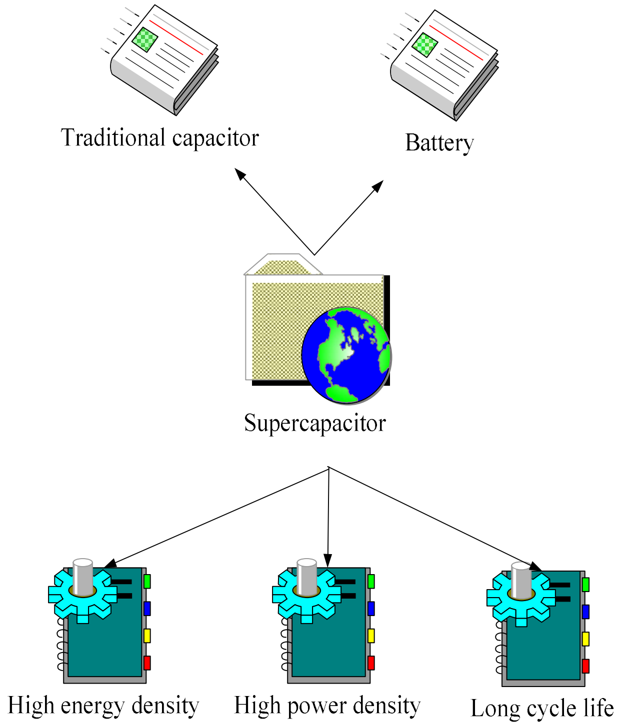

Figure 1: Schematic diagram of supercapacitor

II. A. 2) Introduction of Flexible Supercapacitor

As a kind of portable energy storage machine, flexible supercapacitor has attracted and stimulated the interest of many scientific researchers, especially the lightweight and environment-friendly flexible supercapacitor [16]. Flexible supercapacitor is mainly composed of flexible substrate, electrode materials and solid electrolyte. Compared with rigid capacitors, flexible capacitors have obvious advantages. The application diagram of flexible supercapacitor is shown in Figure 2.

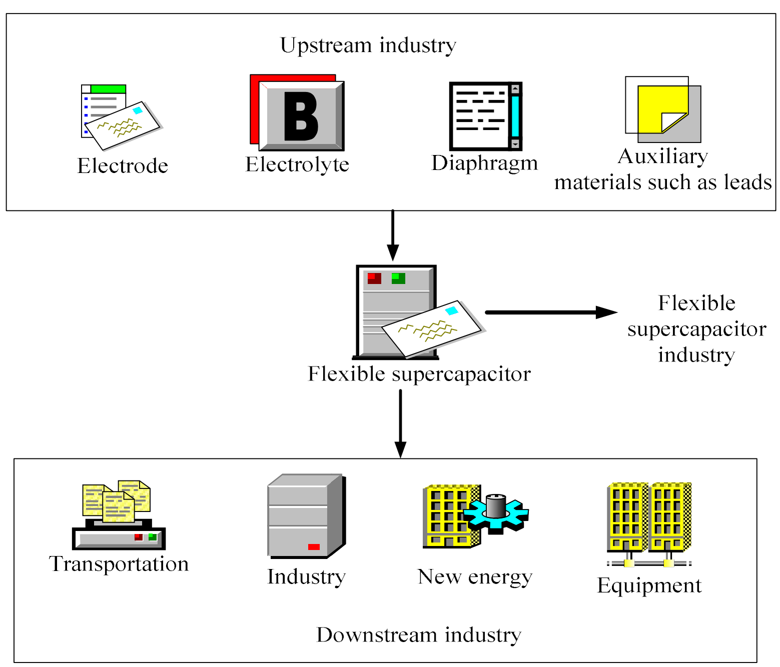

Figure 2: Application diagram of flexible supercapacitor

The exploration and development of flexible energy storage devices and flexible conductors is one of the main factors determining the sustainable development of flexible wearable electronic devices. Flexible supercapacitor has multiple advantages and is a flexible energy storage device with great development potential [17]. The bending characteristics and conductivity of elastic conductor can meet the requirements of the performance test scheme of flexible wearable electronic devices, and has important academic value in its field. In recent years, the exploration of flexible supercapacitors and flexible conductors has made great progress, but there are still many problems in their application in flexible electronic equipment.

As one of the energy storage technology devices with development potential, flexible supercapacitor's excellent device performance is the key to promote its application [18]. Flexible electrical level is the key factor to determine the performance of flexible supercapacitors. The selection and design of electrode materials and flexible substrates would determine the performance of flexible supercapacitors. Cellulose based electrode materials have many characteristics, and it is one of the most ideal electrode materials for supercapacitors. At the same time, designing a power capacitor with cellulose based electrode material array structure can greatly improve the utilization rate, thereby improving the performance of flexible supercapacitors. The structural diagram of flexible supercapacitor is shown in Figure 3.

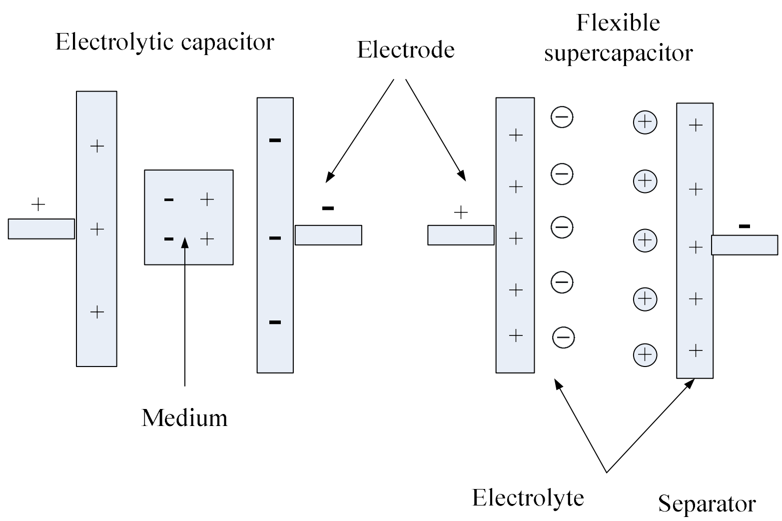

Figure 3: Structural diagram of flexible supercapacitor

At present, there are three main types of flexible energy storage devices. Among them, the flexible solar cell technology is not mature enough and can only be used under the sun. At this stage, it cannot be widely used. The development of flexible electrochemical energy storage device is relatively perfect, and would not be subject to time limit, so it is one of the most popular flexible energy storage components. The performance comparison between flexible rechargeable battery and flexible supercapacitor in flexible electrochemical energy storage device is shown in Table 1. It can be seen from the table that flexible rechargeable batteries have many advantages compared with flexible supercapacitors.

Table 1: Performance comparison between flexible battery and flexible supercapacitor

Type	Flexible rechargeable battery	Flexible supercapacitor
Power density/$W\ Kg^{-1}$	Under 500	100-10000
Energy density/$Wh\ Kg^{-1}$	25-100	05-25
Charging time	2-8h	0.2-20s
discharge time	1-5h	0.2-20s
Charge and discharge efficiency/%	Below 80	Over 90
Cycle life/time	Less than 1000	More than 1000

II. B.Cellulose based electrode materials

There are many cellulose based electrode materials. Carbon fiber materials, nano cellulose and bacterial cellulose can be used as battery materials for flexible supercapacitors.

(1) Bacterial cellulose

Bacterial cellulose (BC), also known as microbial strain cellulose, is a polymer biomaterial with nanotechnology specifications obtained by biological fermentation [19]. Bacterial cellulose is similar to natural cellulose in physical properties, chemical composition and molecular formula. It has become a new trend of branding and intelligence to closely combine low-carbon and environment-friendly biological materials with smart wearable electronic smart home products. Bacterial cellulose has many high quality characteristics, which attracts great attention of researchers all over the world. It has entered the production link in food, pharmaceutical, high toughness paper and other industries. In recent years, bacterial cellulose can be used not only as a composite material, but also as a template or framework with polymers and metal compounds to generate active materials with photocatalytic characteristics. It has a very broad application potential in energy fields such as supercapacitors and lithium batteries.

(2) Nano cellulose

Nanocellulose has good solubility, high density and high strength, and is mainly used in food, microorganism, medicine and other industries [20]. However, in the development process of flexible electrode materials industry, nano cellulose has attracted much attention because of its own advantages, and it is usually used as a framework or substrate to make flexible electrode materials. The nano cellulose membrane flexible electrode has good cycle reliability. Nanocellulose is easy to disperse in the water phase, so it can be evenly dispersed and blended with other dielectrics in the water phase to make flexible electrodes. Nanocellulose is a composite electrode material, which has good physical properties and multiple performance advantages.

II. C. Characterization of electrochemical and mechanical properties of electrode materials

II. C. 1) Cyclic Volt Ampere Test

Cyclic voltammetry is one of the most critical and intuitive test standards for energy storage technology of electrode materials [21]. Before testing, it is necessary to set a relatively stable voltage dialog box and scanning speed for electrode materials. When detecting, it should start from the original voltage, slowly reach the highest point along a certain direction, and then return to the lowest value from the highest point in the opposite direction to conduct a complete cycle time cycle system. The scanning speed would not change in this link. Because the changing scanning voltage would have a corresponding changing induced current, which would show a curve on the screen. The curve can directly reflect the electrochemical performance of electrode materials, and the calculation method is as follows:

$$C_V = \frac{\int I d_v}{2S\Delta V_v} \quad (1)$$

Among them, C_V represents the specific capacitance, I represents the response current, $\int$ represents the surface integral area. S represents the area of the capacitive electrode, and Δ represents the window voltage.

The calculation formulas of the area energy density and power density of the capacitor are as follows:

$$E = \frac{C * \Delta V^2}{2} * \frac{1}{3600} \quad (2)$$

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

II. C. 2) Constant Current Charge and Discharge Test

Constant current charge discharge, also called chronopotentiometric difference method, is one of the key ways to test the electrochemical properties of electronic materials [22]. When galvanostatic charge discharge is used to detect flexible supercapacitor, generally speaking, electrode polarization gives current, so as to accurately measure the change of voltage. The basic principle of constant current charging and discharging is to replace and increase the size on both sides of the flexible supercapacitor, reverse the current in the same direction, and then record the change of voltage on both sides of the power capacitor. When the current direction is positive, the electric level would reflect the battery charging, and the voltage would expand with time. On the contrary, when the whole charge discharge process of the electrode occurs after the current is negative, its voltage would increase and decrease at any time. One charge discharge represents a cycle. The main calculation method is as follows:

$$C_V = \frac{I d_x}{S\Delta V} \quad (4)$$

Among them, C_V represents the area specific capacitance, I represents the added current. Δ represents the discharge time of the capacitor, and S represents the surface of the electrode.

As shown in Figure 4, the specific charging and discharging conditions of the capacitor can be observed intuitively in the curve, which can also be used to further measure the specific capacitance and other properties of the capacitor. In the ideal case, the galvanostatic charge discharge curve of the flexible supercapacitor would show a symmetrical triangle. In the actual working link of the flexible supercapacitor, the galvanostatic charge discharge curve would be very close to the symmetrical triangle due to the sudden change of the internal resistance and its working voltage.

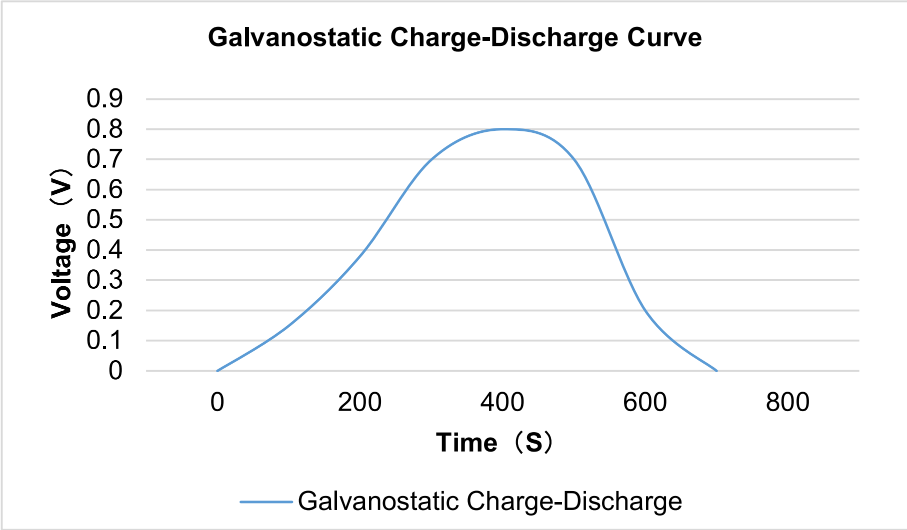

Figure 4: Constant current charge discharge curve

II. C. 3) Cyclic Stability Test

The cycle stability of electrode materials is one of the important factors to show its photocatalytic performance, which can directly reflect the good performance of electrode materials. Under the relatively stable current amount, the electrode material is charged and discharged for more than 1000 times continuously, and the curve of the process is further recorded and made. Finally, the stability of the electrode material is evaluated by comparing and calculating the curve. The calculation method is as follows:

$$R = \frac{C_N}{C_I} * 100\% \tag{5}$$

III. Experimental Analysis of Cellulose Based Electrode Materials for Flexible Supercapacitor

Scientific research is based on the electrochemical characteristics of fiber based electrode materials on flexible supercapacitor, and 6 moles of sulfuric acid electrolyte is used to test it. Before the electrochemical detection of the equipment, the potential range of the flexible supercapacitor should be determined first to prevent the occurrence of such defects as electric polarization or incomplete charge discharge due to too large or too small dialog box. At the same time, the constant current charge discharge (galvanostatic charge discharge) curve did not show obvious deformation within the selected potential range, as shown in Figure 5, further proving this conclusion.

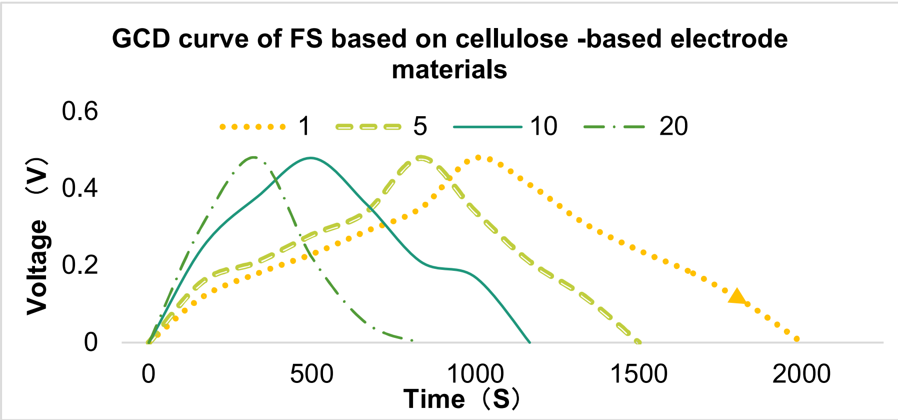

Figure 5: galvanostatic charge discharge curve of flexible supercapacitor based on cellulose based electrode material

By observing the galvanostatic charge discharge curves of flexible supercapacitor based on cellulose electrode material in Figure 5 under different current densities of 6 mole sulfuric acid electrolyte, it is found that all curves are similar to symmetrical triangle shape, indicating that the container electrode has good electrochemical reversibility. The galvanostatic charge discharge curve is not exactly a parallel line, but contains a small amount of skew, which indicates that the capacitor explored in this paper has better performance and fluctuates. In addition, the galvanostatic charge discharge curve is still stable even at high current density, indicating that the overall electrode material has a relatively small characteristic impedance.

In order to study the influence of cellulose based electrode materials on the performance of flexible supercapacitor, cellulose based electrode materials were used to make a flexible supercapacitor, and the charge discharge cycle stability of the flexible electrode of the flexible supercapacitor was studied. This is mainly through repeated charging and discharging to test the experiment. The flexible supercapacitor made of the materials studied in this paper is compared with the flexible supercapacitor made of biomass carbon materials. The flexible supercapacitor made of two different materials is used for 12000 charge discharge experiments. The comparison results of the charge discharge cycle stability of the two flexible supercapacitor electrodes are shown in Figure 6.

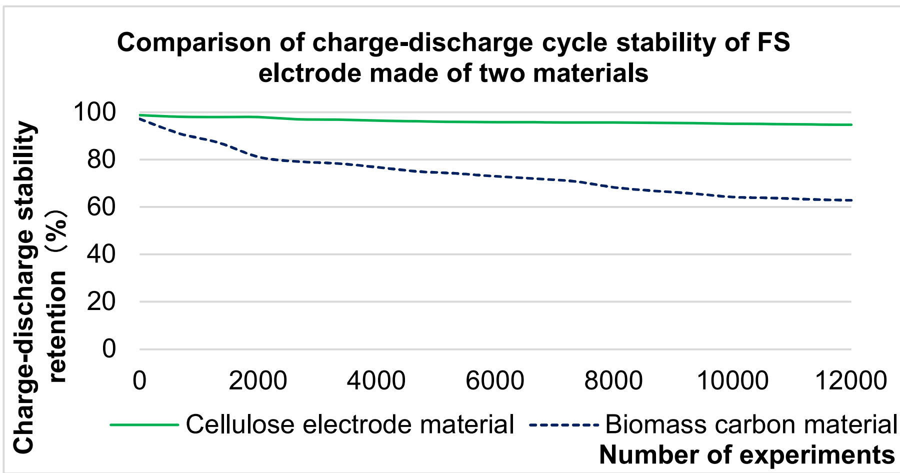

Figure 6: Comparison of charge discharge cycle stability of flexible supercapacitor electrodes made of two materials

As shown in Figure 6, this paper tests the electrode charge discharge cycle stability of two flexible supercapacitor made of different materials. It can be found that after 12000 charge discharge experiments, the final electrode charge discharge cycle stability test of flexible supercapacitor using the materials studied in this paper is kept at 94.75%, while the final value of flexible supercapacitor based on biomass carbon materials is 62.89%. Although the stability of the flexible supercapacitor made of the material studied in this paper has declined, the decline is very small. After 12,000 charge discharge experiments, it only decreased from 98.8% at the beginning to 94.75% at the end, only 4.05%. Once again, the flexible supercapacitor based on the wiki electrode material has an absolute advantage in charge discharge cycle stability. flexible supercapacitor based on biomass carbon materials has decreased greatly after many experiments, and its stability is very poor compared with the materials studied in this paper.

Because the demand for flexible supercapacitor is very large nowadays, how to produce it faster is very important for both producers and consumers. In this paper, the time taken to make flexible supercapacitor based on cellulose based electrode materials and biomass carbon materials is compared, and the specific comparison results are shown in Figure 7.

According to the analysis of Figure 7, it can be found that it takes less time to make flexible supercapacitor with cellulose based electrode materials than with biomass carbon materials, which also shows that the flexible supercapacitor made with the materials studied in this paper has advantages not only in charge discharge cycle stability, but also in production speed. Among them, when there is only one flexible supercapacitor, it only takes 1.5 days to make it based on cellulose based electrode materials, and it takes 2 days for biomass carbon materials. There is little difference between the two. When the flexible supercapacitor is 10, the material studied in this paper only takes 8 days, while the biomass carbon material takes 15 days, which is 7 days more than the material studied in this paper. The production speed is gradually increased. When 20 sets of flexible supercapacitor need to be produced, it takes 12 days to use cellulose based electrode materials, which is 8.5 days less than biomass carbon materials. This shows that the speed of both materials has increased after the expansion of the production scale, but the speed of the materials studied in this paper has increased faster.

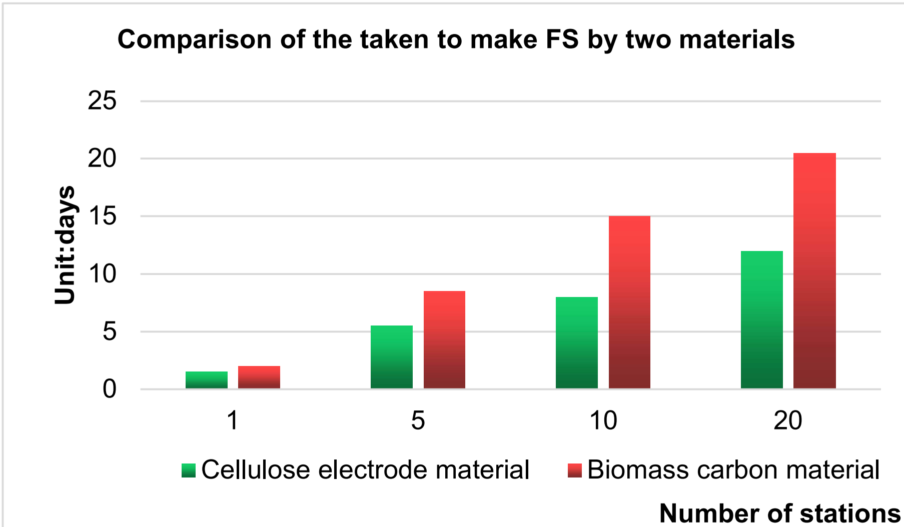

Figure 7: Comparison of the time spent in making flexible supercapacitor with two materials

To further better reflect the research impact of cellulose based electrode materials on the performance of flexible supercapacitor, this paper invited 20 experts to score and evaluate the flexible supercapacitor made of this material and the flexible supercapacitor made of biomass carbon materials in many ways. The evaluation and scoring indicators in many aspects mainly include scale, energy density, production cost, conductivity and practicability, with a score of 1-5 points. The higher the value is, the higher the evaluation is. The final result is the average of 20 experts' scores. The specific scoring of flexible supercapacitor made of two materials is shown in Figure 8.

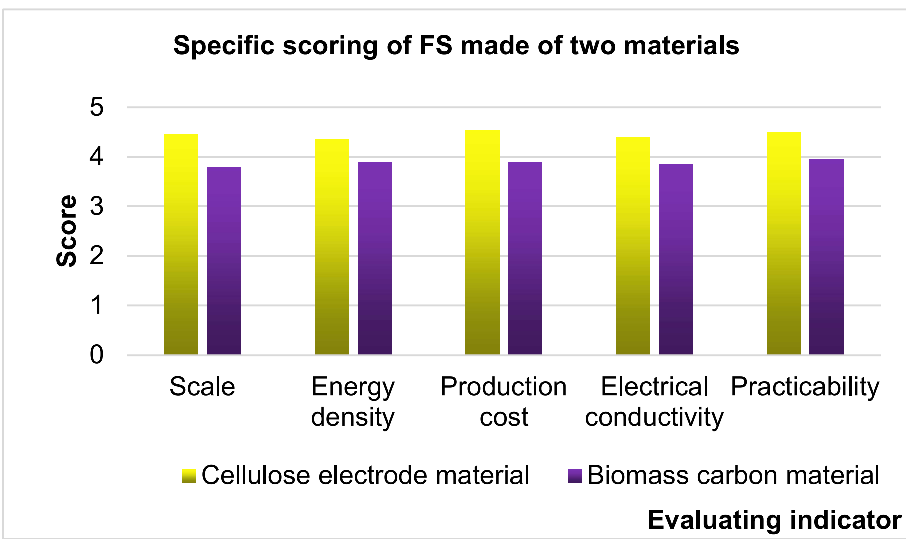

Figure 8: Specific scoring of flexible supercapacitor made of two materials

As shown in Figure 8, experts who make flexible supercapacitor based on cellulose based electrode materials score higher than those based on biomass carbon materials in all aspects. Among them, the score based on the research materials in this paper is above 4.3, while the score based on biomass carbon materials is below 4. Among them, the production cost based on cellulose based electrode materials scored the highest, 4.55 points, 0.65 points higher than that based on biomass carbon materials. The lowest score is for energy density, which is only 4.35 points, but 0.45 points higher than that based on biomass carbon materials. At the same time, the highest score based on biomass carbon materials is in terms of practicality, which is 3.95 points, but 0.55 points lower than the research materials in this paper, while the lowest score is in terms of scale, which is only 3.8 points. Through the research on expert scoring, it can be found that the cellulose based electrode material can expand the production of flexible supercapacitor, make it scale production, and also improve the energy density of flexible supercapacitor. It ensures superior performance, reduces production cost, improves usability, and makes the conductivity function of flexible supercapacitor more powerful.

IV. Conclusions

Cellulose based materials have been widely used in all walks of life because of their good toughness, low carbon, environmental protection, light weight, high quality and low price, and their recyclability. Cellulose based materials also play an important role in the efficient development of flexible supercapacitors. Cellulose based electrode material is a kind of renewable material with stable structure and good compatibility. The importance of flexible supercapacitor in electrical grade active materials is not only as substrate or frame support point material, but also as thickener. In this paper, the performance of flexible supercapacitor is studied based on cellulose based electrode materials, and it is found that its charge discharge cycle stability is better, and the stability is still very high after many experiments. It is conducive to expanding the production scale of flexible supercapacitor, improving the production speed, and also making its energy density better. It is more practical. It not only shows its excellent flexibility, but also shows its excellent photocatalytic performance. The diversity of cellulose based materials and the stability of their structures bring more possibilities for the structural design of electrode active materials.

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