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Study of composite electrode material formation features based on super C45/RuO_2 and super C45/MnO_2 for asymmetric planar supercapacitors

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Abstract. A method for fabricating an asymmetric planar supercapacitor with dissimilar electrodes based on Super C45/RuO_2 and Super C45/MnO_2 using the electrophoretic deposition method has been developed. The electrode topology was formed using laser engraving. The electrophoretic deposition method was chosen for deposition of the composite onto the surface of nickel-coated sitall plates. The features of sequential deposition of composite materials onto the substrate surface were studied, as well as the influence of electrophoresis modes on the composition and morphology of the formed electrode layers. The research was conducted to identify the dependence of the capacitance characteristics of the formed electrode materials on the process parameters. A technology for producing compact planar supercapacitors with an asymmetric configuration for a wide range of microelectronics applications has been developed.

Keywords: planar supercapacitor, asymmetric supercapacitor, laser engraving, electrode material, electrophoretic deposition, suspension

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Исследование особенностей формирования композитного электродного материала на основе Super C45/RuO_2 и Super C45/MnO_2 для асимметричных планарных суперконденсаторов

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Аннотация. Разработан метод изготовления асимметричного планарного суперконденсатора с разнородными электродами на основе Super C45/RuO_2 и Super C45/MnO_2 методом электрофоретического осаждения. Топология электродов была сформирована с помощью лазерной гравировки. Для осаждения композита на поверхность ситалловых пластин с никелевым покрытием был выбран метод электрофоретического осаждения. Были изучены особенности последовательного осаждения композиционных материалов на поверхность подложки, а также влияние режимов электрофореза на состав и морфологию формируемых электродных слоев. Исследования проводились с целью выявления зависимости емкостных характеристик сформированных электродных материалов от параметров процесса. Разработана подход для получения миниатюрных планарных суперконденсаторов с асимметричной конфигурацией для широкого применения в микроэлектронике.

Ключевые слова: планарный суперконденсатор, асимметричный суперконденсатор, лазерная гравировка, электродный материал, электрофоретическое осаждение, суспензия

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Introduction

Supercapacitors (also known as electrochemical capacitors or ultracapacitors) are particularly attractive for microelectronic devices and renewable energy production because they have excellent power and outstanding service life. The current trend in the development of miniature portable electronic devices has greatly increased the demand for ultra-thin, flexible, and durable micro-supercapacitors on a chip, which have enormous potential to complement or even replace microbatteries and capacitors. Asymmetric planar supercapacitors with dissimilar electrodes based on carbon-containing materials and transition metal oxides are promising electrochemical energy storage devices that can fully utilize the advantages of the unique properties of composite materials through combinations of different charge storage mechanisms or different redox reactions [1–5].

This paper presents the results of research and development of a method to form asymmetric planar counter-pin supercapacitors from representative materials based on carbon material, Super C45, transition metal oxides such as RuO_2, MnO_2, by electrophoretic deposition, and a method to create the topological pattern of a planar supercapacitor by laser engraving. The morphology and composition of Super C45/MnO_2 and Super C45/RuO_2 composite samples, as well as the capacitive characteristics of asymmetric planar supercapacitors based on them, were studied.

Materials and Methods

As electrodes of the planar supercapacitor, a 20×12×0.6 mm cut sitall substrate was used, on which a 200 nm thick layer of nickel was deposited by magnetron sputtering. As an adhesion layer between the substrate and the nickel layer, a 40 nm-thick chromium layer was deposited. Laser engraving was used to create two electrically isolated supercapacitor electrodes, which allows high accuracy in reproducing the topological pattern of the device.

Two suspensions, 50 ml each, were prepared. A dispersant was used to achieve the most uniform dispersion of the suspension and to improve the quality of the final product. A mixture of isopropyl alcohol and acetone in a 1 : 1 ratio was used as a solvent. The components were mixed in a test tube with a mixture of acetone and isopropyl alcohol (Table 1).

The deposition process took place in the electrophoretic cell in two stages: sequential deposition of the composite on one half and deposition of the composite on the second half of the counter-pin electrode. The EDP process was performed in the galvanostatic mode. The electrodes were connected to the power supply by reversing polarity. The main stages of creating the planar supercapacitor are schematically shown in Fig. 1.

Table 1

Suspension composition

Components	Suspension 1	Suspension 2
Super C45, g/l	0.04	0.04
RuO_2*xH_2O, g/l	0.2	–
MnO_2, g/l	–	0.4
Hydroxypropyl cellulose, g/l	0.2	0.2
Iodine, g/l	0.4	0.6
Acetone, l	0.025	0.025
Isopropyl alcohol, l	0.025	0.025

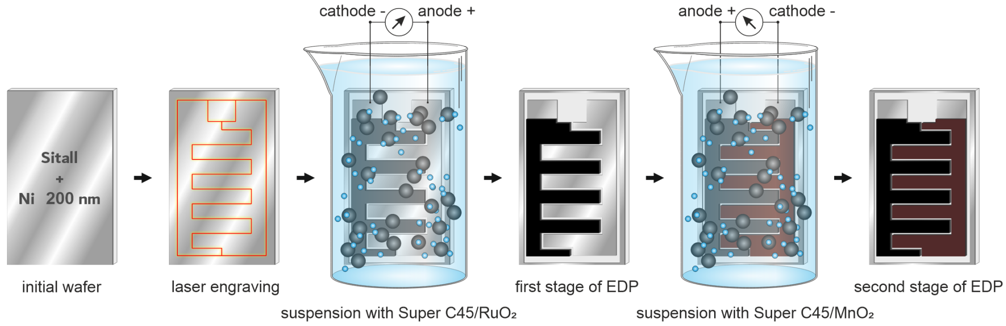

Fig. 1. Schematic representation of the way to create an asymmetric planar supercapacitor with dissimilar composite electrodes

In the power supply, the upper voltage limit was set to 200 V and the current was varied in the range of 5–15 mA. First, deposition was performed for the electrode based on a suspension with RuO_2 in two cycles of 60 seconds each. Then, composite with MnO_2 was deposited in three cycles of 60 seconds for the second electrode.

The morphology and surface composition of the obtained coatings were studied using scanning electron microscopy (SEM) with energy dispersive X-ray spectroscopy, and measurements were taken to determine the capacitive characteristics of the supercapacitors.

The capacity of the cell was calculated by the formula (1):

$$C = \frac{\Delta I}{2\vartheta}, \quad (1)$$

where ΔI is the current swing, ϑ is the scanning speed.

The specific capacitance is calculated by the formula (2):

$$C_{specific} = \frac{c}{m}, \quad (2)$$

where C is the capacity of the device, m is the mass of the sediment.

Results and Discussion

The experimental sample of asymmetric planar supercapacitor with Super C45/RuO_2-based composite on one half and Super C45/MnO_2-based composite on the other half was prepared according to the technique. The results of the morphology and composition study are shown in Fig. 2.

The porous surface structure observed in SEM images is important in increasing the capacitance and power characteristics of supercapacitors. Surface porosity provides a large contact surface between electrode and electrolyte, which promotes efficient ionic diffusion and maximum electrolyte absorption.

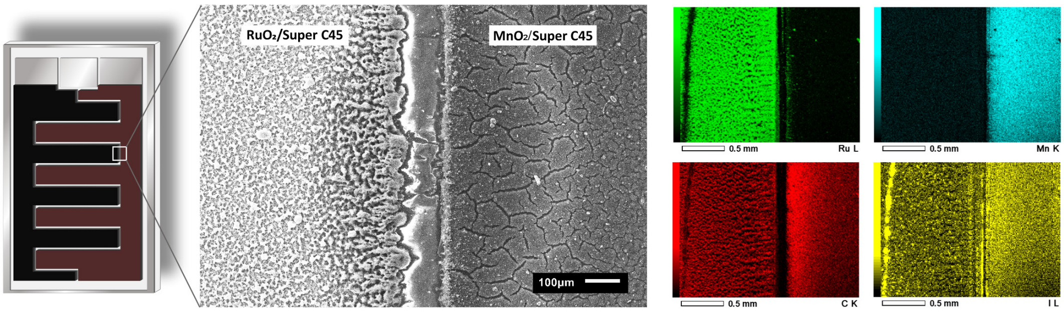

Fig. 2. SEM image and element distribution maps of asymmetric planar supercapacitor

The element distribution maps show a uniform distribution of the components of the electrode material over the surface of the sample, and, importantly, the composites were deposited clearly in their area and did not cross the interface into the opposite electrode. The composite with RuO_2 was deposited only on the left electrode, and the composite with MnO_2 was deposited only on the right electrode. Carbon, which is part of both composites, is present on the two electrodes. Iodine, an auxiliary component that is also present in both suspensions, was equally deposited.

To study the electrophysical characteristics, the supercapacitors were placed in a 1 M KOH electrolyte solution and volt-ampere cyclic sweeps were taken using a potentiostat, after which their capacitive characteristics were measured (Fig. 3).

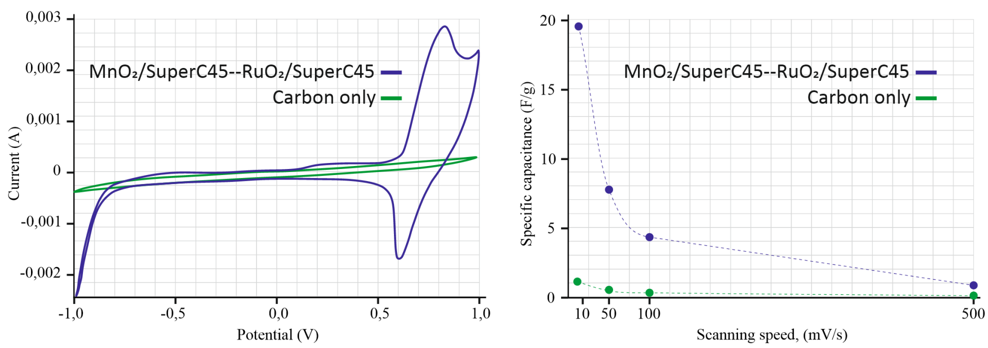

Fig. 3. Graph of the cyclic volt-amperogram (*a*), and the dependence of the scanning speed on the specific capacity of supercapacitors (*b*)

It is well seen, in the asymmetric planar supercapacitor not only a different form of sweep, but also increases the area of the figure, which is directly proportional to the electrical capacitance in contrast to the capacitor with a double electric layer (Fig. 3,*a*). If we compare their shapes, we can see the presence of peaks that correspond to reversible redox reactions.

The specific capacitance of the obtained sample of asymmetric planar supercapacitor was 19.8 F/g, exceeding the specific capacitance of the capacitor with an electric double layer 18 times, which was 1.1 F/g.

Conclusion

In the course of the study, by varying the composition of the suspension and deposition modes, the composition of the composite electrodes was optimized. The peculiarities of the sequential local deposition of composites onto electrodes of planar supercapacitors were studied. These investigations allowed demonstrating the fundamental possibility of creating asymmetric planar supercapacitors with improved capacitance and power characteristics that are compatible with traditional integrated circuit production technology. Further development of new electrode materials, optimization of the synthesis and assembly processes of supercapacitors, and improvement of their electrochemical properties are possible.

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