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New Materials Based on Carbon-Metal Nanocomposites for Supercapacitor Application

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In this report the approach is developed where the carbon nanotube – metallic nanoparticles composites are used as material of electrodes for electrochemical capacitors based on the double electric layer and pseudo-capacitance effect. To improve the pseudo-capacitance properties the ethanol as the auxiliary reagent is added to the electrolyte to enhance the redox reaction. As active electrode material the two types of carbon-metal composites: carbon nanotube – Ni nanoparticle composites (CNT–Ni) and carbon-palladium hybride films were prepared and studied. The peak on the potentiodynamic curve for CNT–Ni composite electrode in alkaline solutions of ethanol is observed which is ascribed to the ethanol oxidation in alkaline medium. The results obtained are discussed from the point of view both the double layer capacitance and the pseudo capacitance. The carbon-palladium hybride films were prepared by ion evaporation of integrate targets consisting of graphite and metallic palladium. Electron microscopy measurements showed that the nanoparticles of palladium were formed in graphite matrix with average dimension of about 3 nm. The contribution of Pd nanoparticles to the electrochemical processes going on the surface of the films is considered from the point of view of their electrochemical properties .

Keywords: Carbon Nanotubes, Nanoparticles, Pseudo-capacitance, Supercapacitors.

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1. INTRODUCTION

Electrochemical capacitors based on double electric layer [1] and pseudo-capacitance effect [2] (also known as "supercapacitors") attract much attention as sources of electric energy with high power density. The increase of the power capacity of such electrodes can be achieved by using carbon-metallic nanocomposites with high effective surface. The deposition on the surface of carbon nanotubes of catalytically active metallic nanoparticles gives the additional contribution to the capacitance as result of enhancement of Faradaic reactions on carbon-metal nanocomposite media. The pseudocapacitance concept implies that Faradaic redox processes proceed between the electrode and the electrolyte at certain applied potentials. As the active electrode material the transition metal oxides along with nanostructured carbon is mainly used. Organic electrolytes which are used in such systems have serious drawbacks connected with ecological problems and small electrical conductivity that reduces available power. In this connection the development of electrodes where the high effective area will combine with high effective pseudocapacitance properties in aqueous electrolyte is the main point of electrochemical capaci-

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are used as material for the electrodes. To improve the pseudo-capacitance properties the ethanol as the auxiliary reagent is added to the electrolyte to enhance the redox reaction.

2. EXPERIMENT

2.1 The materials preparation and methods

As active electrode material the two types of carbon-metal composites: carbon nanotube — Ni nanoparticle composites (CNT–Ni) and carbon-palladium hybride films were prepared and studied. Carbon nanotubes were grown at 420–450 °C by means of catalytic pyrolysis of granular polyethylene in helium atmosphere using a Ni plate as a substrate. Various methods of promotion of nickel surface substrates were used: abrasive grinding and mechanical and chemical polishing. It allowed the obtainment of nanocomposite materials consisting mainly of carbon nanotubes and Ni nanoparticles. The obtained composites were examined by transmission electron microscopy (TEM) and X-ray diffraction (DRON- 3M, CuKα).

The electrocatalytic properties of CNT-Ni composites were investigated by linear sweep voltammetry. An Ecotest-VA potentiostat was employed for linear sweep voltammetry measurements. Voltammetric experiments were carried out in a three-electrode system at room temperature. Carbon nanotubes-Ni nanoparticles composite electrodes (geometry area $0.2~\mathrm{cm}^2$) were

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used as working electrodes. A platinum foil and saturated Ag–AgCl served as counter and reference electrode, respectively. Three kinds of electrolytes 0.1 M KOH, 0.1 M KOH+0.1 M C2H5OH, 0.1 M KOH+0.2 M C2H5OH were used. These solutions were prepared using high pure chemicals and distilled water. The carbon-palladium hybride films were prepared by means of ion evaporation of integrate targets consisting of graphite and metallic palladium. Films were deposited at substrates from Si, NaCl and glass. The thickness of films obtained was 130-140 nm.

3. RESULTS AND DISCUSSION

The electrochemical activity of CNT–Ni composite electrodes was measured in aqueous solutions of alkali and alkaline solutions of ethanol. The results obtained exhibit high catalytic activity of CNT–Ni composites in the above-mentioned solutions. The linear sweep voltammogrammes of CNT–Ni composite electrode were measured in the aqueous solution of alkali and alkaline solution of ethanol. By comparing with the voltammogramme in the absence of alcohol, an alcohol oxidation peak is clearly observed in the voltammogramme on CNT–Ni electrode in the presence of ethanol. In the oxidation of alcohols at the nickel electrode in alkaline solution, various hypotheses are given in literature. It was suggested that the main reaction proceeds through the following mechanism:

$$Ni OH \rightarrow NiO (OH) + e^{-} + H^{+}$$

Thus the addition of ethanol in aqueous alkaline electrolyte enhance the Faradaic processes between the electrode and electrolyte and can be used to improve the electrochemical capacitance.

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The development of new materials based on carbon nanostructured films is one of the topical tasks of material sciences. The carbon diamond-like films attract particular attention due to the fact that these films contain the nanoscaled graphene planes with dimensions from 0.5 to 5 nm. In this work the results of synthesis and study of new materials - carbonpalladium hybride films are presented. The method based on the ion evaporation of integrate targets was developed. Integrate targets consisted of graphite and metallic palladium. The ion evaporation was carried out by Xe+ ions with energy of 1 keV and current density 200 µA/cm². Films were deposited at substrates from Si, NaCl and glass. The thickness of films obtained was 130-140 nm. Electron microscopy measurements showed that the nanoparticles of palladium were formed in graphite matrix with average dimension of about 3 nm. It was found that the optical gap of composite film is determined by electronic structure of graphite matrix and volume content of palladium nanoparticles. The optical absorption of films is investigated in visible and ultraviolet part of spectrum. The contribution of Pd nanoparticles to the electrochemical processes going on the surface of the films is considered from the point of view of their electrochemical properties.

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