

Usage of oxygen-modified CNT for electrode material of an air-hydrogen fuel cell

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Abstract

The problem of efficiency of fuel cells is important recently with the introduction of this type of energy sources in a road transport.

The efficiency problem of energy conversion of fuel cell consists of three main parts: the electrochemical activity of the electrode material in reactions creating potential, efficiency of mass transport and efficiency of charges transport. In order to increase the electrochemical activity and intensification of mass transport of cathode material of the hydrogen-oxygen fuel cell with proton conducting membrane by the authors was developed a material containing a short, pre- oxygen modified CNTs and platinum on carbon black (Pt/C). Having attached oxygen-containing groups of the atoms of the CNT provides Red-Ox activity of material with a high degree of reversibility. As a result, CNTs are an additional depolarizer, lowering the activation energy of the process of electrochemical reduction of molecular oxygen. However, the presence of short CNTs allows to obtain a equal by structure composite with larger porosity that improve mass transport.

1. Material (CNT and Pt/C)

Electrode material was prepared by mechanically mixing and by ultrasonic homogenization of Pt/C and CNT. As Pt/C type was used commercial product E-TEK.

The materials were characterized by scanning and transmission electron microscopy, EDAX, adsorption-structural analysis and helium pycnometry, FTIR-spectroscopy. Electrochemical behavior examined on a disk glassy carbon electrode in a 0.5 M sulfuric acid.

CNTs produced by arc discharge between graphite electrodes in liquid hydrocarbon [1]. Unlike traditional arc technologies of CNT growth in the gas phase, the use of a liquid hydrocarbon phase substantially reduces the temperature of the CNT growth and increases the yield of CNT (close to 100%). Upon receipt of the CNT used technical graphite with low content of catalyst (iron impurities 0.03-0.2 wt.%). CNTs were subjected to plasma-chemical modification in high-frequency plasma discharge in argon. Then activated CNTs were prepared by nitrogen and oxygen treatment.

When modifying CNT in frequency plasma closed hemispherical ends are primarily opened, defects in the surface structure are introduced; morphology, porous structure, surface active properties of CNTs are changed. Fig. 1 shows the spherical open ends, defects of side surfaces.

For purification of metal impurities and partial oxidation with addition of oxygen-containing groups of atoms CNT treated with a solution of nitric acid (1:1) at a temperature of ~ 100C for 5 minutes, followed by repeated washing with water and drying. This treatment resulted in a decrease by approximately one order of content of metal impurities.

2. Electrochemical characteristics

Red-Ox activity was ensured by a quinone-hydroquinone equilibrium. The presence of quinone groups were fixed according to the cyclic voltammograms and the characteristic absorption bands on the IR spectra. The presence of oxygen was also recorded by EDAX.

Fig. 2 shows the cyclic voltammograms of CNT. Red-Ox process is characterized by high reversibility.

Electroreduction kinetics of molecular oxygen was investigated on a rotating disk electrode using known relationships Koutecky – Levich. The results of measurement of densities of the kinetic currents by the rotating disk electrode showed a significant increase of kinetic current density of reduction of molecular oxygen in the case of introduction of the Red-Ox CNT additives (Table).

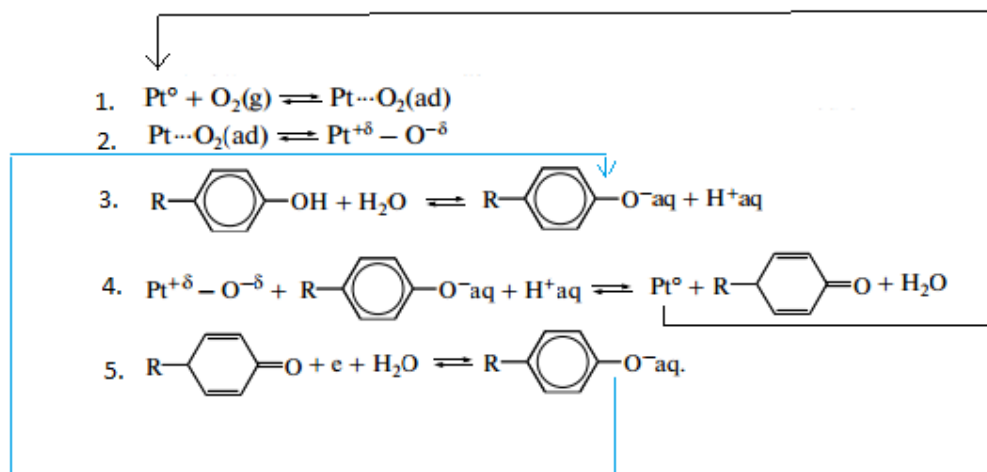
Measurements of components loads and thickness of active layers of samples with different contents of CNTs showed the porosity of the active layer linearly dependent on the content of the CNT.

Degradation of the material under electrochemical influence was investigated. The mechanism of the influence of oxygen modified CNTs on the kinetics of oxygen electroreduction on platinum was proposed. It lies in the inclusion of the quinone–hydroquinone Red-Ox equilibrium of the oxygen reduction.

Table - Density kinetic currents at different electrode materials

Material	Specific current density, mA/cm ² (true surface Pt) at 450 mV
E-TEK	0.23
E-TEK+CNT, 1:1	0.37

The CNTs electrically bonded to platinum CNT reduce (due to the hydroquinone groups) the oxygen adsorbed on it. This is reflected in the experimentally observed decrease in the oxygen coverage of platinum. The mechanism may be represented by the following sequence of equations:



References

[1] G.Z.Paskalov; S.A.Krapivina; A.K.Filippov, US Patent 5,344.462. Sep. 6. 1994

Figures

Fig. 1. TEM image of Plasmas CNTs: a) initial; b) after plasma modification (arrow shows open end)

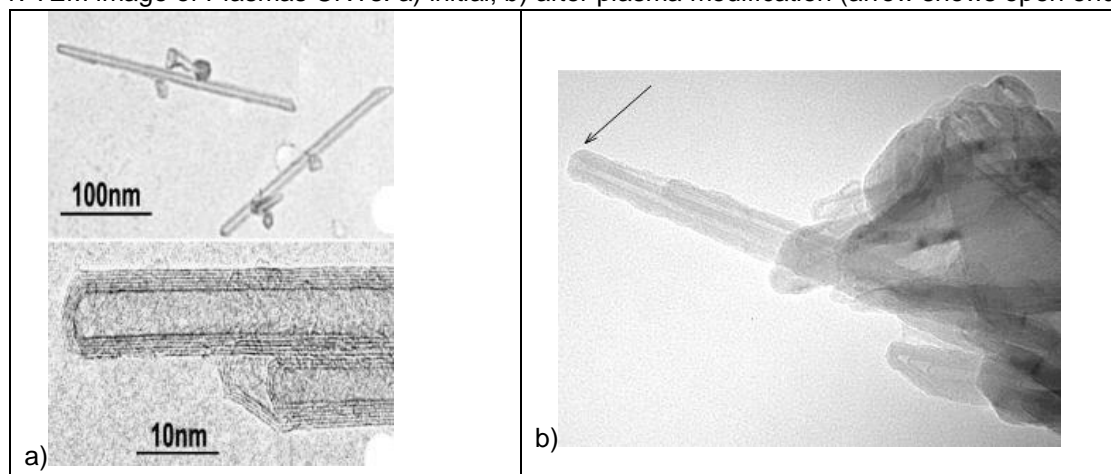
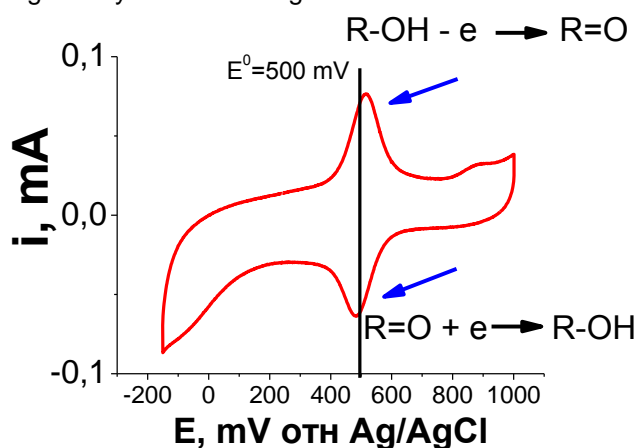


Fig. 2 - Cyclic voltammograms of CNT



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