

## **Graphene oxide – a new electrode nanomaterial for chemical current sources**

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### **Abstract**

Scientific and technical progress in the field of nanotechnology has contributed to the creation of new functional nanomaterials with unique physical and chemical properties, which gave a rapid impetus to the development of electronic technology. Graphene oxide (GO) is characterized by hydrophilicity, high chemical activity, proton conductivity, and the possibility of changing in stoichiometric composition, the number and type of oxygen-containing functional groups, which allows to vary GO properties and generally consider graphene oxide as an independent nanomaterial with a wide range of applications, this, in turn, indicates the relevance of developing nanostructured functional materials based on GO.

**The relevance of the work** determined by the comprehensive research of the properties of nanostructured functional materials based on graphene oxide. The performed research fills in the gaps about several practically important properties of graphene oxide, as well as determines the conditions for obtaining nanostructured functional materials based on graphene oxide and their application. The dissertation developed physical and chemical principles for creating nanostructured functional materials (films, microspheres, aerogels) based on graphene oxide, conducted comprehensive studies of their structure and properties, and analyzed the possibility of using the obtained materials to modify existing and create new electroactive components of chemical source of electric energy. The possibility of using graphene oxide as the main current-forming component of the cathode of primary lithium chemical source of electric energy is proposed and demonstrated. Presented process of electrochemical reduction of graphene oxide in a lithium electrolyte. A prototype model of a galvanic cell of the Li|GO electrochemical system is presented, the specific (weight) energy intensity of which reaches 749 W·h/kg, which is 25-390% higher than the energy density of modern primary chemical current sources.

**The purpose of the dissertation** is to develop physical and chemical principles for creating functional nanostructured materials based on graphene oxide, to establish the features of their structure and properties, to determine the possibility of their use for modifying existing and creating new electroactive components of chemical source of electric energy. Establishing the process of electrochemical reduction of graphene oxide by studying the reaction products formed in this process.

### **Main tasks were:**

1. Analysis of the possibility of changing the content of oxygen-containing functional groups in graphene oxide by changing the synthesis conditions;
2. Study of the process of chemical and thermal recovery of graphene oxide thin films;
3. Development of a method for obtaining films with specified characteristics from graphene oxide dispersion;

4. Development of a technique for obtaining microspheres from graphene oxide dispersion;
5. Development of a method for obtaining aerogel with specified characteristics from graphene oxide dispersion;
6. Investigation of electrochemical characteristics of the obtained functional nanostructured materials;
7. Investigation of structural and chemical changes in graphene oxide during electrochemical reduction;
8. Investigation of the possible mechanism of the process of electrochemical reduction of graphene oxide in a lithium electrolyte.

**Scientific novelty:**

1. For the first time, a method is presented for producing thin films of reduced graphene oxide on the surface of an aqueous dispersion of graphene oxide, by forming them during directed heat treatment of the surface of an aqueous dispersion of graphene oxide with a stream of hot air, followed by transferring the resulting films to a solid substrate. The technology allows the growth of films "from the bottom up" because the transport of the initial components of the films occurs from a liquid medium (graphene oxide dispersion). A change in the physical and chemical properties of reduced graphene oxide films depending on the duration of the film formation process has been established, which demonstrates the possibility of obtaining films with specified properties by changing the heat treatment conditions, as well as the concentration and composition of the graphene oxide dispersion used.

2. For the first time, the possibility of direct application of graphene oxide as the main current-forming component of the cathode of a primary lithium chemical current source with a high specific discharge capacity reaching practical values of  $720 \text{ mA} \cdot \text{h/g}$ , which exceeds the values of the discharge capacity of known cathode materials used in the production of primary chemical current sources by 1.6-3.2 times. The dependence of changes in the discharge capacity of graphene oxide on the oxygen content, surface area, layer thickness, and discharge currents is established, which demonstrates the possibility of manufacturing primary chemical current sources with specified characteristics by changing the conditions for the chemical synthesis of graphene oxide and the technological conditions for obtaining cathode materials based on it.

3. A process for electrochemical reduction of graphene oxide in a lithium electrolyte is Proposed, based on the results of studies of changes in the bond structure, surface morphology, and composition of graphene oxide during electrochemical reduction. For the first time, a theoretical calculation of the electrochemical reduction capacity of graphene oxide corresponding to  $3292 \text{ C/g}$  or  $914 \text{ mA} \cdot \text{h/g}$  is presented.

4. For the first time, based on practical results, a model of a prototype of a galvanic cell of the Li|GO electrochemical system was calculated, the specific (weight) energy density of which reaches 749

W·h/kg, which exceeds the values of the specific (weight) energy intensity of lithium galvanic cells produced by industry by 25-390%.

### **The practical significance of the work**

This work creates a foundation for technical and technological methods and solutions for the use of graphene oxide and is generally aimed at solving the problems of developing new functional nanomaterials with specified properties. The obtained information about the properties of functional materials based on graphene oxide will be in demand in laboratory and technological processes when designing and predicting the characteristics of chemical current sources.

The developed method for producing thin films based on reduced graphene oxide on the surface of an aqueous dispersion of graphene oxide by forming them during directed heat treatment of the surface of an aqueous dispersion of graphene oxide with a hot air flow opens up new opportunities for obtaining carbon coatings with specified properties.

The developed method for obtaining porous electrodes based on graphene oxide opens up the possibility of creating cathode materials of chemical current sources with a discharge capacity reaching 720 mA·h/g, which exceeds the values of the discharge capacity of known cathode materials by 1.6-3.2 times.

The evaluation of the theoretical relevance of the results, presents a process of electrochemical reduction of graphene oxide in a lithium electrolyte, demonstrating the dependence of the capacitance of the electrochemical reduction of graphene oxide content of oxygen-containing functional groups that can be considered as the method of establishing the degree of oxidation of graphene oxide complementary to known analytical techniques.

The presented practical feasibility of creating a primary battery based on graphene oxide opens up wide opportunities for autonomous electronic devices, since the specific energy density of a lithium primary chemical current source with a graphene oxide cathode can reach 749 W·h/kg, which is 25-390% higher than the energy density of modern primary battery, which will proportionally increase the operating time of autonomous electronic devices and, accordingly, improve their consumer properties.

### **Defense Provisions**

1. Technological features of obtaining functional nanostructured materials (films, microspheres, aerogels) based on graphene oxide;
2. Results of research of physical and chemical properties of nanostructured materials (coatings, films, microspheres, aerogels) based on graphene oxide;
3. Electrochemical characteristics of functional materials based on graphene oxide in lithium chemical current sources;

4. Results of complex experimental studies of the influence of synthesis modes and parameters on the main characteristics of functional nanostructured materials based on graphene oxide, which determine the design criteria for power supplies;
5. The process of electrochemical reduction of graphene oxide in a lithium electrolyte;
6. Primary battery of the Li|GO electrochemical system with parameters exceeding industrial analogues.