

TEMPERATURE SENSORS BASED ON GRAPHENE OXIDE AND GRAPHENE OXIDE / 12 TUNGSTOPHOSPHORIC ACID THIN FILMS ON INTERDIGITAL ELECTRODES

Dr. Ž. Mravik¹, M. Pejčić¹, D. Petković¹, P. Stolić²,
M. Stević³, Dr. Z. Stević^{2,4}, Dr. Z. Jovanović¹

¹University of Belgrade, “Vinča” Institute of Nuclear Sciences;

²University of Belgrade, Technical Faculty in Bor;

³Elsys Eastern Europe, Belgrade;

⁴University of Belgrade, School of Electrical Engineering
Serbia

mravik@vin.bg.ac.rs

Easy-to-develop sensing devices are important for modern technology where materials with modifiable properties that are dependent on the environmental conditions play key role. Graphene oxide nanocomposites possess interesting surface chemistry that is closely correlated to their electric properties. In this work, composite of graphene oxide and 12-tungstophosphoric acid was dip coated on interdigital electrode and thermally treated while dependence of its electric properties on environment temperature was examined with impedance spectroscopy. Results outline potential applicability of these materials for temperature sensors.

Keywords: graphene oxide, temperature sensors, nanocomposites, interdigital electrodes, impedance spectroscopy.

Monitoring of environmental parameters is of great importance for modern society. Constant growth of urban areas causes significant local increase of temperature (phenomenon often called the “urban heat island”) which increases the risk of heat-related mortality and lowers substantially the quality of life [1]. Climate parameters like humidity and temperature are essential for agricultural areas and crop development [2]. Because of this, the development of materials that are sensitive and selective on environmental conditions has been permanent subject of scientific research. Graphene oxide (GO) is a 2D material with interesting and easily modifiable surface chemistry that is closely correlated to its electric properties [3]. Additionally, 2D nature of GO allows functionalization with various inorganic compounds that can increase the sensitivity of its electric properties to environmental conditions [4]. In this work, temperature sensors were developed by dip-coating of GO/12-tungstophosphoric acid composite (GO/WPA) on the interdigital electrodes (IDE) where dependence of electric properties on temperature was investigated with impedance spectroscopy.

Experimental

GO/WPA nanocomposite with 6 wt.% of WPA was synthesized following the procedure published elsewhere [5]. GO and GO/WPA aqueous suspensions (3 mg/ml of GO) were used for dip-coating, i.e. deposition of film on IDE. Dip-coating of IDE with GO/WPA composite was performed with a home-made device consisting of Micro step electric drive controlled by microcontroller. The samples were coated by withdrawing the IDE from GO/WPA suspension at an angle of 30° between the line perpendicular to the suspension surface and the IDE (Fig. 1, a). Pause between steps (dwell time) while withdrawing the IDE from the suspension was 60 s. Deposited films were thermally treated at 200°C in argon atmosphere (30 ml/min flow, 10 °C/min heating rate) to reduce the GO and obtain better electric properties [6].

Digital photographs were taken in order to evaluate properties of film before and after the thermal reduction. Impedance was measured at different temperatures in the range from 0.1 Hz to 1 MHz and with

100 mV amplitude and 0 V bias voltage by allowing the sample to equilibrate for a few minutes at every temperature. Additionally, the changes in resistivity of the samples was investigated at 100 mV DC voltage by performing chronoamperometry measurements at every temperature.

Results

Fig. 1, *b*, *c* shows digital photographs of GO/WPA films deposited on the IDE before and after thermal reduction, respectively. It can be observed that color and optical properties of the film change after thermal reduction, while also some degree of film detachment and cracking is evident. However, a large part of the IDE is covered with homogenous film after thermal reduction.

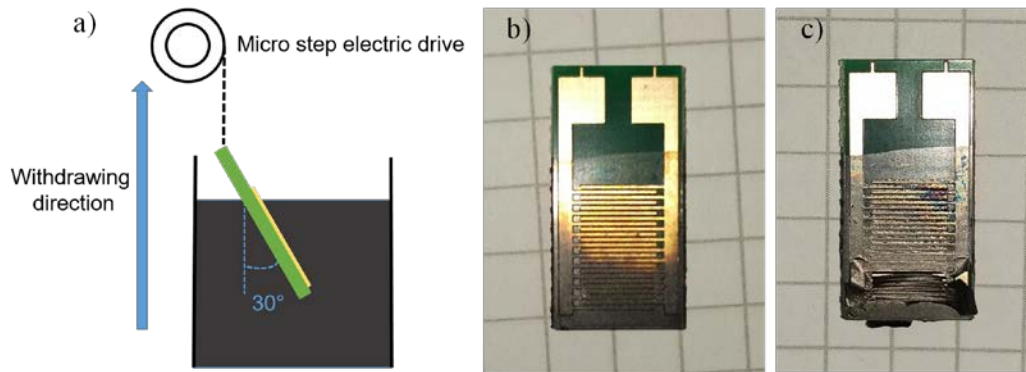


Fig. 1. Digital photographs of GO/WPA films deposited on IDE before and after thermal reduction

Impedance spectroscopy measurements were utilized to investigate electric properties of thermally reduced GO/WPA films at different temperatures. Fig. 2 shows the dependence of impedance on frequency (Bode plot), as well as Nyquist plots obtained after equilibrating the samples at different temperatures. It can be observed that impedance values at lower frequencies are highly dependent on the temperature while at higher frequencies, the slope of logarithmic dependence is similar for all of the examined temperatures. Nyquist plots show close-to semicircle shape where radius of semicircle is highly dependent on the temperature. Additionally, it can be noted that the impedance values vary between 1000 and 2000 Ω which is beneficial for application in temperature sensors and on-site measurements.

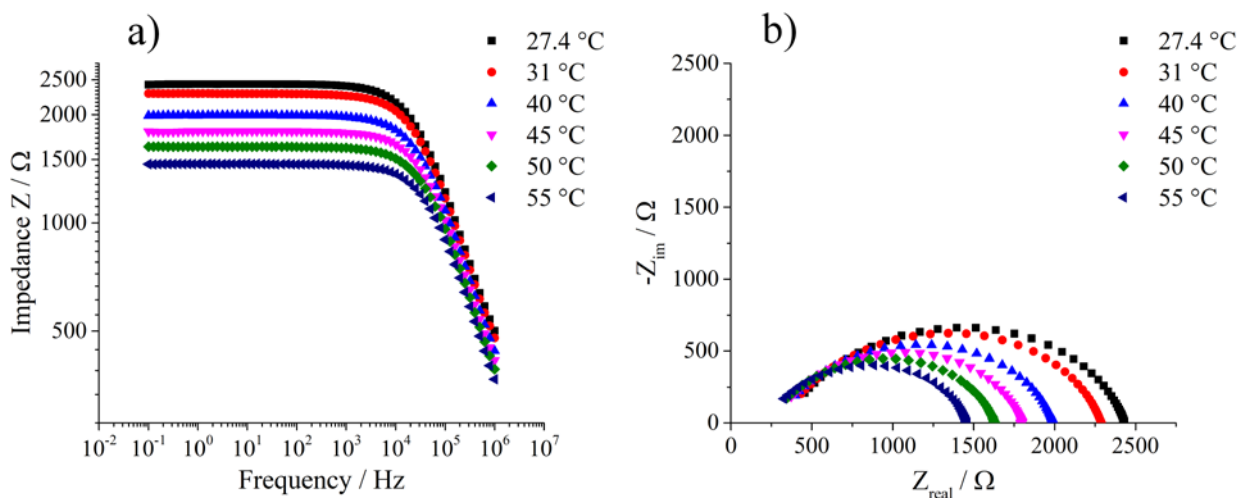


Fig. 2. Logarithmic dependence of impedance from frequency (Bode plots) (*a*) and Nyquist plots of thermally reduced GO/WPA films on IDE (*b*) at different temperatures

The applicability of the material as temperature sensor is also related to the discovery of a simple correlation between electric properties of the material and temperature. Because of this, the temperature dependence of impedance values at the middle of observed plateau (at 10 Hz) from Fig. 2, *a* is plotted against temperature and linearly fitted (Fig. 3, *a*). Very good linear dependence is observed with r^2 value of 0.9994. The results show a decrease in impedance with temperature (negative temperature coefficient — NTC) characteristic to the NTC thermistors. The obtained linear equation shows that for every degree of the temperature increase, the impedance increases for 35.7 Ω , which is very beneficial for temperature sensor application.

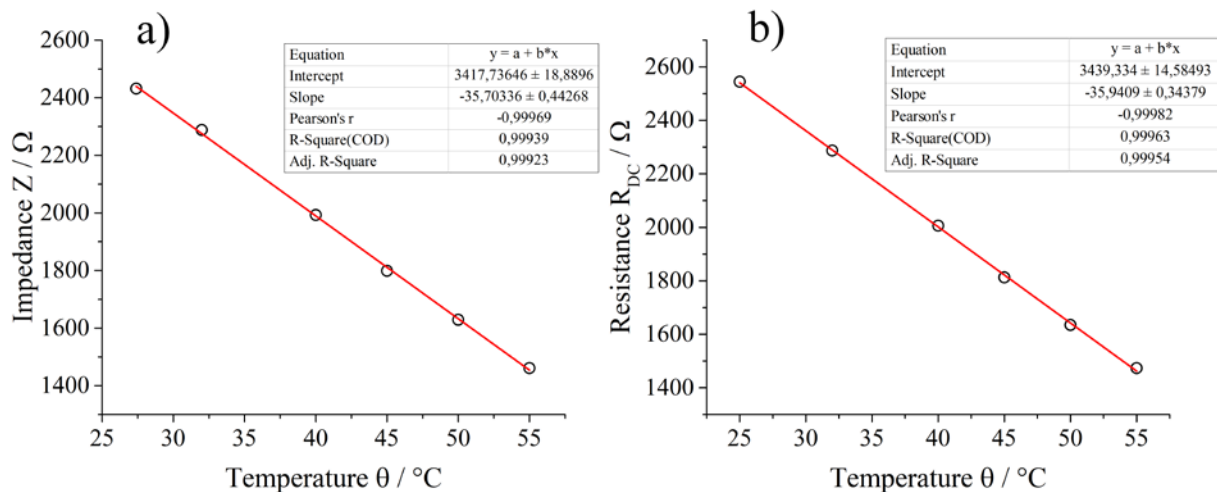


Fig. 3. Temperature dependences of impedance values at 10 Hz (*a*) and of resistance values at 100 mV DC current of thermally reduced GO/WPA films on IDE (*b*)

To evaluate the possibility of using the GO/WPA temperature sensors only with DC current, resistance values at 100 mV were recorded by performing the chronoamperometry measurements (Fig. 3, *b*). The linear plot of resistance values vs. temperature shows almost the exact results as for the impedance at 10 Hz with even better linearity (r^2 of 0.9996) and higher dependence on temperature (35.9 Ω decrease for every degree of temperature increase). This shows that GO/WPA temperature sensors can be used with a DC 100 mV power source.

Conclusions

In this work, a thin film of GO/WPA composites deposited on IDE by the dip-coating method and thermally reduced in argon at 200°C is investigated as temperature sensor. Digital photography before and after reduction shows slight detachment and cracking of the obtained films with a large part of the IDE covered with a homogeneous film. Impedance spectroscopy measurements showed high dependence of impedance values measured at low frequencies from temperature and impedance values of around 1000–2000 Ω , which is beneficial for application. Plots of impedance values at 10 Hz, as well as resistance values at 100 mV DC current, showed the behavior characteristic of NTC thermistors and a good sensitivity to temperature. The results outline good possibility for application of thermally reduced GO/WPA films on IDEs for temperature sensors.

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Ж. Мравик, М. Пејич, Д. Петкович, П. Столич, М. Стевич, З. Стевич, З. Јованович

Датчики температури на основі тонких плівок оксиду графену та композиту оксиду графену та 12-вольфрамфосфорної кислоти на зустрічно-штирьових електродах

Сенсорні пристрої, які легко розробляти, є важливими для сучасних технологій, де ключову роль відіграють матеріали зі змінними властивостями, що залежать від умов навколишнього середовища. Нанокомпозити на основі оксиду графену мають цікаву хімію поверхні, яка тісно корелює з їхніми електричними властивостями. У цій роботі композит з оксиду графену та 12-вольфрамфосфорової кислоти наносили на зустрічно-штирьовий електрод і термічно обробляли. Залежність його електричних властивостей від температури середовища досліджували за допомогою імпедансної спектроскопії. Отримані результати вказують на потенційну можливість застосування цих матеріалів для датчиків температури.

Ключові слова: оксид графену, датчики температури, нанокомпозити, зустрічно-штирьові електроди, імпедансна спектроскопія.