Photoconductivity of functionalized carbon nanotubes

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Investigation of carbon nanotubes is a modern trend due to their combination of unique physical, chemical, electrical, and optical properties. Carboxyl-functionalized carbon nanotubes (fCNTs) for investigation of photoelectrical properties were synthesized. The photo-sensitivity spectra of a carboxyl-functionalized CNT sample for voltage range from 1 to 9 V, and for the spectral range from 400 to 900 nm were investigated. The voltage equal to 1 V generated lower photosensitivity in the broadband wavelength range for visible to near-infrared. The most efficient photocurrents of fCNTs were received for a voltage of 5 V in the wavelength range λ_p~400-800 nm and for voltage U=3V in the broadband spectral range λ_p~400-900 nm. The experimental data analysis helped to determine the widest photosensitivity range, as well as the highest sensitivity value. As result, the voltage U=5V was obtained. Here, the most significant photocurrent peak with I_p~2.67 μA for wavelength λ~720 nm was observed. A comparison between the photosensitivity spectra of fCNTs and pure CNTs shows that the photosensitivity of fCNTs has increased significantly. Thus, the maximum photosensitivity for fCNTs is I_p ~ 2.67 μA, and for pure CNTs, it equals I_p ~ 0.185 μA. A 14-fold enhancement of photosensitivity for fCNT has been registered. The mathematical analysis of spectral dependencies of generated photocurrents under different applied voltages can be described using fourth-order polynomials. The I-V characteristics for wavelengths 760 nm and 780 nm have the same trend with the shift of photocurrent maximum to the lower parameters of voltage. The carboxyl-functionalized nanotubes can be effectively used as light detectors and in optoelectronic applications.

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1. Introduction

A lot of optoelectronic applications are working using carbon low-dimensional nanomaterials (carbon nanotubes, graphene, and its derivatives). Modern investigation of carbon nanocomposite connected with optical, photoelectrical, and structural properties characterization for new applications [1-8]. The most useful synthesis methods for carbon nanotubes are the arc

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discharge method, laser ablation method, and chemical vapor deposition method [9]. Surface functionalization of carbon nanomaterials and nanocomposites can be processed by different strategies [9]: non-covalent binding [10-11] and covalent binding [12-13], nanoparticle functionalization [14-16,30]. A new fullerene-activated carbon nanotube detector with high photosensitivity has been proposed [17]. Electrochemical biosensors based on carbon are perspective medical applications and bioimaging techniques [18]. The electrical properties and photoelectrical phenomena for carbon and composite nanomaterials are explained in [19-23]. In [24-29] physical properties of CNTs and GO were comparatively investigated. In the paper [24], the morphology and structural parameters of carbon nanotubes and gadolinium-doped carbon nanotubes were investigated by using Scanning Electron Microscopy and X-ray diffraction techniques. Optical and structural properties of carbon nanotubes doped with 10% and 15% gadolinium were shown in [24-26]. Doping degree transformed the physical and chemical properties of carbon nanotubes. In our previous research [27], the photosensitivity spectral characteristics for pure CNTs in the wavelengths from 400 up to 900 nm at different applied voltages to the sample were studied and received new information for the next generation of photoelectrical and optoelectronic CNTs devices.

2. Experimental details

Carbon nanotubes were obtained by the arc discharge method and initially functionalized with a carboxyl group. For the measurement of photoelectrical properties, the home-build setup consisting of a white light source, monochromator, and digital registration electronics was used at room temperature [27].

3. Results

Spectral characteristics of carbon nanotube functionalized with the carboxyl group photosensitivity under different wavelengths (400-900 nm) of excitation light at two applied voltage values 1V and 3V are described in Figure 1. The spectral dependencies of photocurrent (Figure 1) indicate that for the spectral range, λ=600-875 nm with maxima at λ_{p1}~600 nm, λ_{p2}~700 nm, λ_{p3}~760 nm, λ_{p4}~840 nm, λ_{p5}~875 nm the photocurrent intensities for U=1 V were evenly distributed.

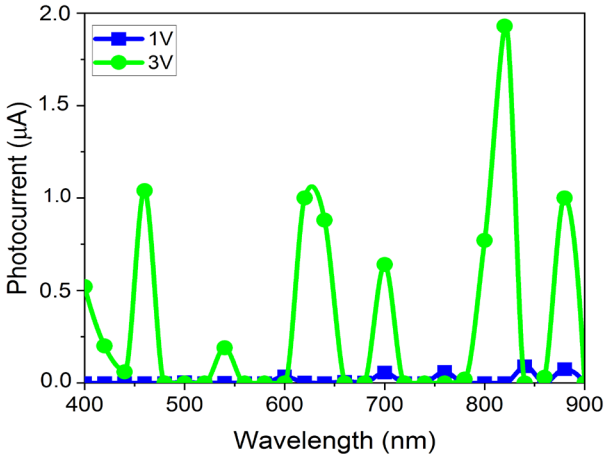

Fig. 1. Photosensitivity spectra of carboxyl functionalized carbon nanotubes for applied voltages U=1V and U=3V.

The highest amplitude of photosensitivity equal to 0.1 μA is registered at λ_{p4}~840 nm. Other peaks have a photocurrent value less than 0.1μA. In the spectral range from 400 to 575 nm, the absence (zero) of photosensitivity is observed. Almost the entire investigated spectral range λ=400-900 nm at U=1V is characterized as small intensity light sensitivity range. Photocurrent intensity peaks with the wavelengths λ_{p1}~400 nm, λ_{p2}~460 nm, λ_{p3}~540 nm, λ_{p4}~620 nm, λ_{p5}~700 nm, λ_{p6}~820 nm, λ_{p7}~875 nm at voltage value U=3 V are observed. In comparison with the photocurrent for λ_{p6}~820 nm at U=1V the photocurrent for U=3V has a maximum peak with a significant increase up to I_{p6}~1.93μA. The spectral peak position λ_{p6}~820 nm is slightly shifted to the low wavelength spectral range. Zero photosensitivity in the spectral range λ=475-525 nm was detected. In this case, the range of zero photosensitivity has narrowed.

Figure 2 shows the spectral characteristics of photosensitivity for fCNT in the spectral range from UV to NIR at three values of the bias voltage U (5 V, 7 V, and 9 V) applied to the investigated sample.

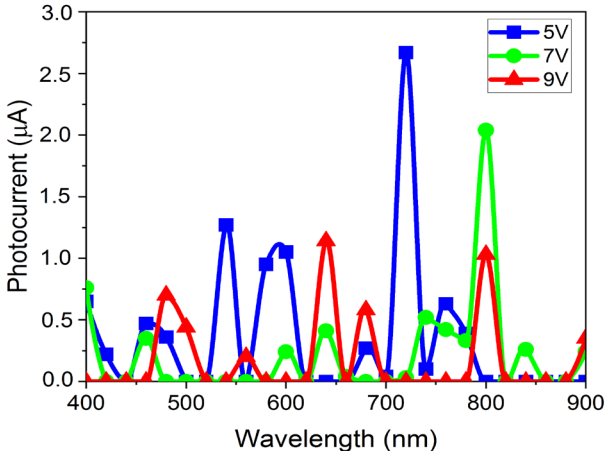

Fig. 2. The photosensitivity spectral dependencies of fCNTs for the bias voltages U (5V, 7V, and 9V) applied to the sample.

At U=5 V, the peaks for wavelengths λ_{p1}~400 nm, λ_{p2}~460 nm, λ_{p3}~540 nm, λ_{p4}~600 nm, λ_{p5}~675 nm, λ_{p6}~720 nm, λ_{p7}~760 nm are registered. The maximum peak at wavelength λ_{p6}~720 nm with photocurrent value I_{p6}~2.67 μA is observed. The third peak at λ_{p3}~540 nm and the fourth peak at λ_{p4}~600 nm have photocurrent values I_{p3}~1.27μA and I_{p4}~1.05μA, respectively. The zero photosensitivity spectral range λ=800-900 nm is $\Delta\lambda$=100 nm. At voltage U=7 V, the maximums intensities of photocurrent for wavelengths λ_{p1}~400 nm, λ_{p2}~460 nm, λ_{p3}~600 nm, λ_{p4}~635 nm, λ_{p5}~740 nm, λ_{p6}~800 nm, and λ_{p7}~840 nm were registered. The maximum peak at wavelength λ_{p6}~800 nm with photocurrent value I_{p6}~2.04 μA was observed. Other peaks have a small photocurrent value of less than 0.5μA. At the spectral range, λ=475-575 nm with zero photosensitivity was registered. At voltage U=9V, for spectral range λ=520-620 nm ($\Delta\lambda$=100 nm) the weak photosensitivity spectra were registered. This spectral range has one weak photocurrent peak at wavelength λ_{p2}~560 nm. The maxima values of photocurrent I_{p3}~1.14 μA and I_{p5}~1.03μA were observed at wavelength λ_{p3}~640 nm and λ_{p5}~800nm, respectively. The peaks at wavelengths λ_{p1}~475nm and λ_{p4}~680nm correspond to photocurrent values I_{p1}~0.7μA and I_{p4}~0.58μA, respectively.

The analysis of measured photocurrent spectra shows that the increase in applied voltage (up to 9V) to the investigated fCNT sample leads to an increase in photosensitivity. The photocurrent data at voltage U=1V for spectral range λ=400-900 nm with small photosensitivity characterized it as ineffective for light detection. The most efficient use of fCNT is on the voltage 5 V in the wavelength range λ_p~400-800 nm and voltage U=3V in the wavelength range λ_p~400-900 nm. For widest spectral range and the highest photosensitivity values can be used at the voltage value U=5 V. Figure 2 shows, the photocurrent at the wavelength λ~720 nm reaches a

value equal to I_p~2.67 μA. These experimental results indicate that fCNTs can be effectively used as light detectors in the tuned wavelength range. A comparison between the photosensitivity spectra of fCNTs and pure CNTs [27] shows that the photosensitivity for fCNTs has increased significantly. Thus, the maximum photosensitivity for carbon nanotubes functionalized with a carboxyl group and for pure carbon nanotubes equals I_p~2.67 μA and I_p~0.185 μA, relatively. A 14-fold enhancement of light sensitivity for fCNTs has been observed. The photosensitivity experimental spectral dependencies at different voltage values can be transformed into I-V characteristics for understanding photoelectrical features of light interaction with fCNTs.

It is of interest to study the observed changes in the photosensitivity ranges for fCNTs as well as their intensities after changing the voltage applied to the samples. It is from this point of view that we draw the graph of the dependence of the voltages on the photosensitivity spectrum at different wavelengths. As can be seen from the figure, the I-V characteristics of the samples used for different wavelengths are described as follows. The I-V characteristics fCNTs (Figure 3(a)) for wavelengths 640 nm and 720 nm can be approximated using the next approach:

$$I = -0.01047 \cdot U^4 + 0.231 \cdot U^3 - 1.692 \cdot U^2 + 4.624 \cdot U - 3.152 \quad (1)$$

$$I = 0.04141 \cdot U^4 - 0.8288 \cdot U^3 + 5.391 \cdot U^2 - 12.45 \cdot U + 7.843 \quad (1)$$

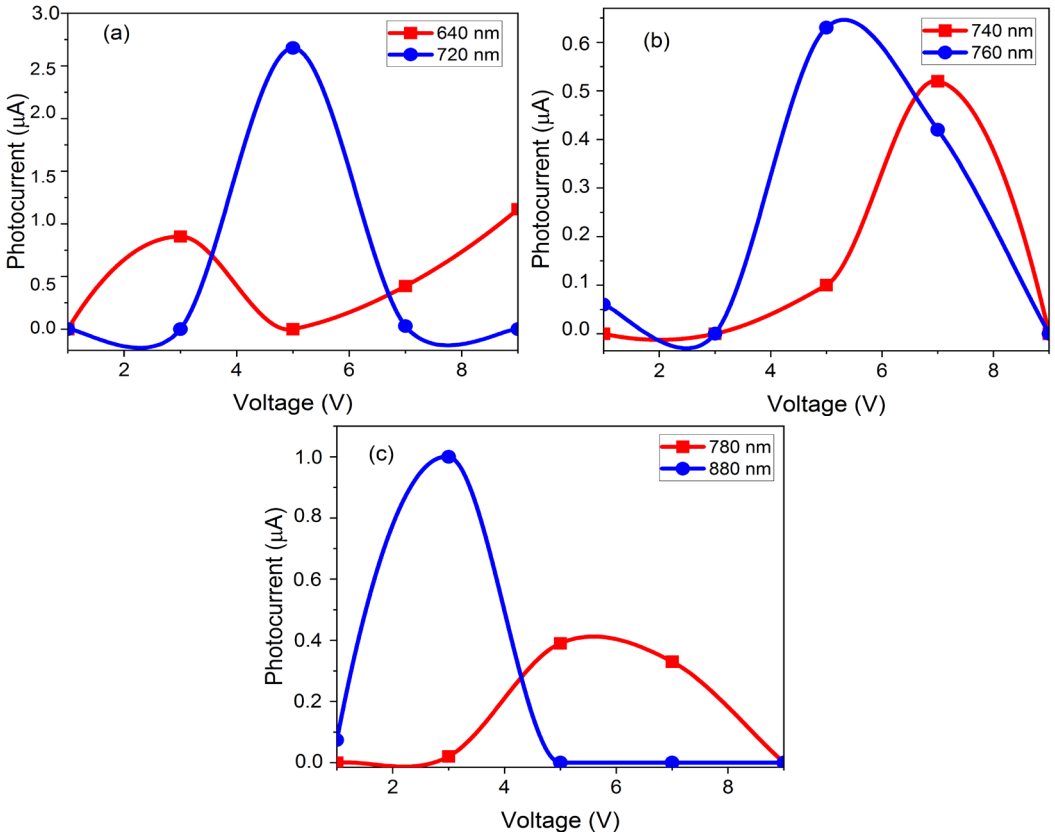

Fig. 3. I-V curves of fCNT for different illumination wavelengths.

I-V characteristics as shown in Figure 3 included different regions that can be associated with specific conductance for carbon materials. The voltage ranges 2.2-5V at wavelength λ ~ 640 nm and in the ranges 1-2.1 V and 6-8.7 V can indicate the presence of negative differential conductance (Figure 3 (a)). Also, another region can be associated with appearing of positive differential conductivity. From the I-V curve at the wavelength 720 nm (Figure 3 (a)), the voltage ranges of 1-3 V and 7-9 V can be associated with zero sensitivity area. In addition, the voltage range 5-7V can be described with an influence of the negative differential photoconductivity

mechanism. The voltage U=5V and illumination wavelength 720 nm lead to the generation of a maximum photocurrent value equal to I_p ~2.7 μA. The absorbed light generated the electron-hole pair's formation. This process (separation of electron-hole pairs) can be controlled by the bias voltage. The carrier concentration will be increased as well as the conductivity and current. I-V characteristics in Figure 3(b) at two wavelengths 740 nm and 760 nm can be approximated by:

$$I = -0.003854 \cdot U^4 + 0.06625 \cdot U^3 - 0.3602 \cdot U^2 + 0.7338 \cdot U - 0.4359 \quad (3)$$

$$I = 0.005625 \cdot U^4 - 0.1219 \cdot U^3 + 0.8569 \cdot U^2 - 2.098 \cdot U + 1.418 \quad (4)$$

I-V curve in Figure 3(b) for wavelength λ ~ 740 nm and the voltage ranges 1-1.5 V and 4.2-7.5 V can be associated with appearing of positive differential photoconductivity. Other voltage ranges 1.7-3 V and 7.5-9 V can be associated with a negative differential photoconductivity mechanism. The voltage 3-4.2V range indicated the presence of an almost zero photosensitivity. The maximum photocurrent value equal to I_p ~0.55 μA is reached at U=7.5V. The weak photosensitivity intensity with a photocurrent value equal to I_p ~0.06 μA takes place at U=1.6V. Thus, the maximum photocurrent value decreased and shifted towards higher voltage values with an increase of wavelength from 740 to 760 nm. From I-V curve in Figure 3(b) at wavelength λ~760 nm also can be associated with two different mechanisms of photoconductivity. The features of the I-V curve for voltage range 5.5-8.5V and 3-5.5V can be connected with negative and positive differential photoconductivity, respectively. In this case, zero sensitivity region was registered in the two voltage ranges 1-3 V and 8.5-9 V. The voltage U=5.5V generated the maximum photocurrent value equal to I_p~0.67 μA, and the peak position shifted towards lower voltage values. Figure 3(c) shows the experimental data of I-V curves of fCNT for two wavelengths, λ~780 nm, and λ~880 nm. For these curves, the following fitting approach has been applied:

$$I = 0.002448 \cdot U^4 - 0.05542 \cdot U^3 + 0.4005 \cdot U^2 - 0.9696 \cdot U + 0.622 \quad (5)$$

$$I = -0.01022 \cdot U^4 + 0.2245 \cdot U^3 - 1.669 \cdot U^2 + 4.627 \cdot U - 3.099 \quad (6)$$

4. Discussion

I-V curves at the wavelengths 780 nm have the same shape as the wavelength 760 nm. The maximum value of photocurrent was maintained at the voltage value U=5.5V but slightly decreased up to the value 0.42 μA. The voltage range with zero light sensitivity takes place from 1 to 3 V under 780 nm illumination light. For illumination with wavelength 880 nm, zero light sensitivity area in the range from 5 to 9V, was detected. The photocurrent peaks with values 1.1 μA and 1.2 μA are registered at U=2.2V and U=9V, respectively. The voltage ranges of 2.3-5V and 8.2-9V at illumination 880 nm on the I-V curve (Figure 3 (c)) can be associated with negative differential photoconductivity. The region of zero light sensitivity from voltage 5 to 7 V was registered. A weak peak at voltage U=8.2V with a photocurrent value of 0.24 μA was detected. The photocurrent peak intensity shifted towards lower voltage values with increasing wavelength above 760 nm- wavelength. For modern optoelectronic applications, the information about photoelectrical properties of fCNTs in the spectral ranges 800–900 nm and 550–900 nm is important. Mostly, these ranges can be overlapped and correspond to laser lines for different types of solid-state lasers. Also, the laser such as argon, krypton, helium-neon, and ruby with emission wavelengths 490 nm, 630 nm, and 690 nm motivated to investigate the photosensitivity of carbon nanomaterials. Comparisons between the collected data from Figure 1-3 are shown the possibility for their future applications but require the chemical content improvement of carbon nanotubes and new experimental conditions.

5. Conclusions

The dependencies of photosensitivity spectra for the fCNT doped with carboxyl group in the wavelength range from 400 nm to 900 nm under voltage values 1V, 3V, 5V, 7V, and 9V have been investigated. The lowest photosensitivity for the fCNTs at a voltage value equal to 1V was registered. This feature takes place almost along the entire above-mentioned wavelength range and characterizes these materials and voltage value (1V) as unsuitable as a light detector. It is important to note that more effective conditions for fCNTs use in the spectral range of 400-800 nm are voltage 3V and 5V. Moreover, the voltage of 5 V leads to the appearance of the widest and the highest photosensitivity at the wavelength 720 nm with an intensity of 2.67 μA. This makes it important to use it as a light detector in different wavelength ranges.

The I-V experimental data for fCNTs have been shown for different wavelengths of excitation light such as ~640 nm, ~720 nm, ~740 nm, ~760 nm, ~780 nm, and ~880 nm, respectively. The approximated approach has been proposed for describing these experimental curves. The fourth-order polynomials described these experimental data. The approximation has been obtained using MATLAB R2021a software. The I-V characteristics for fCNTs at wavelengths 760 nm and 780 nm have the same shape. But, there is some difference related to the tendency of the photosensitivity peak which shifted towards the lower photocurrent value with the increasing wavelength from 760 nm.

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