

Laser-induced dry electrodes based on carbon nanotubes and graphene for ECG monitoring

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Dry electrodes for long-term ECG monitoring have been developed based on composites of hybrid nanostructures of single-walled carbon nanotubes (SWCNT) and reduced graphene oxide (RGO) flakes in a polydimethylsiloxane (PDMS) matrix, formed by laser processing. A method for forming a liquid dispersion for uniform homogenization of carbon nanomaterials within the composite has been developed. Exposure of the composite to laser radiation with a wavelength in the near-infrared range and an intensity of 15 kW/cm² has been found to result in a 13-fold decrease in resistance to (8 ± 2) k Ω due to the formation of hybrid SWCNT/RGO nanostructures. The formation of conductive networks in the PDMS matrix has been shown to increase the defect density due to the formation of bonds between the carbon nanomaterials. A study of the effect of mechanical pressure on the impedance of the formed electrodes showed that increasing pressure (0–12 kPa) leads to a decrease in impedance across the entire frequency range of 10–500 Hz. Electrodes based on the PDMS/SWCNT/RGO composite demonstrated high impedance-frequency stability in the same frequency range during prolonged contact (7 days) with a suspension simulating human sweat, compared to traditional Ag/AgCl electrodes. Furthermore, the high stability of the PDMS/SWCNT/RGO composite electrodes was confirmed by 7 day recording of impedance changes during their skin placement. ECG measurements revealed that the signal quality recorded by the PDMS/SWCNT/RGO composite electrodes was comparable to the signal from the Ag/AgCl electrodes, with the amplitudes of the main peaks being more pronounced. The biocompatibility of PDMS/SWCNT/RGO composite electrodes was demonstrated in studies of the viability of connective tissue cells on their surface. Therefore, the developed PDMS/SWCNT/RGO composite electrodes are suitable for use in wearable devices, including for continuous ECG monitoring for at least 7 days. The absence of a gel layer in dry PDMS/SWCNT/RGO composite electrodes offers an advantage over commercially available alternatives for long-term ECG monitoring.

Keywords: carbon nanotubes, reduced graphene oxide, hybrid nanostructures, laser processing, electrodes, electrocardiogram, polydimethylsiloxane, cells.

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Introduction

Allotropic modifications of nanocarbon provided broad prospects for the development of electronics. Carbon nanotubes (CNT), which are thread-like structures of carbon atoms with adjustable structural parameters, are used as piezoresistive elements, field-emission cathodes, and energy storage devices [1–9]. The potential for using graphene and its derivatives to create capacitors, resistive sensors, conductive composites and gels is explained by their unique electrophysical properties [10–17].

CNT and graphene, possessing unique properties unattainable by traditional conductive materials, have become the game-changers in creating electrical conductors for biomedical applications. These carbon nanomaterials combine electrical conductivity, biocompatibility,

and mechanical flexibility, making them ideal for next-generation biomedical components. CNT with a given structure demonstrate high electrical conductivity up to 100 MS/m, exceeding the conductivity of copper conductors (59.6 MS/m) [18]. The internal *sp*² hybridized structure results in exceptional mechanical strength and chemical stability of the nanotubes, as well as the ability to function as electrical conductors and semiconductors [19]. Graphene that is featuring a conductivity of as high as 1738 S/m and high specific surface area (~ 2630 m²/g), makes the transfer of electrons between the electrodes and biomolecules quite effective [20].

To register the biopotentials of a human body, including electrocardiogram (ECG), electroencephalogram (EEG) and electromyogram (EMG), metal electrodes are predominantly used, with disposable silver/silver chloride

(Ag/AgCl) gel electrodes being the most widely used in clinical practice [21]. Despite their cost-effectiveness and ease of use, these electrodes have a number of serious limitations. Within development of the biopotentials long-term monitoring system, the key problem is the degradation of the registered signal, mainly because of destruction of the gel layer upon contact with skin secretions during a prolonged mechanical action. An additional factor that degrades the quality of ECG signal is the presence of the stratum corneum of the skin [22]. The need for preliminary skin preparation, including depilation, surface cleansing and partial exfoliation of keratinocytes, can induce inflammatory reactions, which further reduces the quality of the registered signal. There has also been an increase in cases of allergic reactions to the gel component and adhesive components of the electrodes [23]. The combination of these limitations narrows the scope of application of gel electrodes in long-term physiological monitoring systems.

Dry electrodes, including CNT- and graphene-based structures, can overcome the shortcomings of conventional electrodes. Due to their high electrical conductivity, carbon nanomaterials have significant potential for flexible electronics. The integration of carbon nanomaterials into polymer matrices allows the creation of dry electrodes [24–27]. Polydimethylsiloxane (PDMS) is the most common polymer matrix for flexible conductive interfaces, characterized by high chemical stability, low toxicity and biological inertness, which minimizes the risk of inflammatory reactions upon contact with living tissue [26–29]. Due to its high elasticity and flexibility, PDMS effectively adapts to skin irregularities and ensures stable contact during biopotentials registering. However, despite the absence of a gel layer, dry electrodes have insufficient electrical conductivity. During the electrode formation process, the dielectric polymer matrix covers the surfaces of CNT, insulating them and hindering the transfer of electrons during registration of biopotentials, which determines the typical conductivity level of such systems in the range of 0.0001–10 S/m [30].

The electrophysical characteristics of dry electrode composites, including electrical conductivity and contact resistance, critically depend on their composition and fabrication methods. A key challenge in developing dry electrodes is to simultaneously provide low resistance, high mechanical strength, and long-term stability of the conductive carbon network in contact with biological fluids. Research into methods for creating hybrid nanostructures that combine two types of carbon nanomaterials can meet these requirements by complementing their structural and electrical properties while simultaneously increasing conductivity [31,32]. The most accessible and effective method of forming such nanostructures is exposure to external electromagnetic radiation [33]. Laser sources represent the most precisely tunable electromagnetic radiation sources for this purpose, providing contactless, high-energy and precise modification of carbon nanomaterials. In the previous study, the formation of conductive carbon nanomaterial networks during laser processing was investigated [34]. The work

analyzed the mechanisms leading to the formation of C–C bonds between carbon atoms. As a result, covalently bonded CNT atoms and reduced graphene oxide (rGO) atoms provided a highly efficient electron transport in all directions within the formed branched SWCNT/rGO network.

This paper sheds light on the results of fabrication of dry electrodes made of hybrid nanostructures of single-walled carbon nanotubes (SWCNT) and reduced graphene oxide flakes formed in a PDMS matrix by laser irradiation. Structural changes were investigated using scanning electron microscopy (SEM) and Raman spectroscopy (RS). The resistance and impedance of the electrodes were measured before and after 7 days of exposure to an environment simulating a human sweat. The stability of the electrode impedance was studied during their wearing for 7 days. To verify the applicability for biopotentials registration, ECG signals were registered using the fabricated electrodes, followed by comparison with signals registered using traditional Ag/AgCl electrodes. The biocompatibility of the electrodes was assessed by analyzing the growth of human embryonic fibroblast (HEF) cells on the electrode surfaces.

1. Materials and methods

1.1. Formation of electrodes

The following technique was developed to create flexible electrodes based on carbon nanomaterials. The initial step involved preparation of homogeneous dispersions of CNT and reduced graphene oxide in a solvent. SWCNT were used (OCSiAl, Luxembourg). SWCNT were 1–2.5 nm in diameter and 5 μ m long with a specific surface area of 420 m²/g. The reduced graphene oxide powder (Graphenox, Russia) had a specific surface area of 600–700 m²/g and conductivity of > 2 S/cm. PDMS Sylgard 184 (Dow, USA) was used.

The solvent was selected in order to exclude any influence on the curing process of the composite. Ethanol, isopropanol and n-hexane were tested due to their effectiveness in dispersing carbon nanomaterials. The dispersions were processed in a Q700 ultrasonic disperser (Qsonica, USA) for 80 min at a power of 150 W/cm² to achieve homogeneity. In parallel, a 20 mass% dispersion of PDMS in a solvent was prepared and treated in an ultrasonic bath for 30 min. After homogenization, the two dispersions were mixed and ultrasonically treated for 3 h to obtain a homogeneous dispersion. All tested solvents had a low boiling point, which allowed them to be easily removed by evaporation using a heated magnetic stirrer C-MAG HS 7 (IKA, Hungary). The resulting dispersion was then placed in a vacuum chamber for degassing to prevent the formation of macrocavities. A hardener was added to the dispersion in a base-to-hardener ratio of 10:1. The final dispersion was cast into molds with a given geometry (diameter 30 mm, thickness 1 mm) printed on a 3D printer. Wires were integrated into the electrodes during the curing process to

provide electrical connections. The composites included the following: 0.5/0.5 mass% of SWCNT/rGO.

To form hybrid nanostructures with bonds between nanotubes and rGO flakes, the composites were subjected to laser processing with ytterbium laser generating radiation at the fundamental harmonic in the infrared range with a wavelength of 1064 nm. The galvanometric scanner provided precise positioning of the laser beam along the coordinates X and Y . The electrodes were processed using single pulses with a spot diameter of $35\ \mu\text{m}$ and a step of $17\ \mu\text{m}$ at a scanning rate of 240 mm/s, covering the entire area of the of 30 mm diameter electrode. The focusing lens formed a Gaussian beam profile corresponding to the thickness of the electrode. The laser operated in pulsed mode with a pulse repetition rate of up to 30 kHz.

1.2. Study of structural characteristics

The surface morphology of the created electrodes was studied using Helios G4 scanning electron microscope (FEI, USA) at an accelerating voltage of 5 kV and a probe current of 50 pA. Pressure in the chamber was maintained at $3.9 \cdot 10^{-4}$ Pa.

Raman spectra of the electrodes were recorded using a spectrometer LabRam HR Evolution (Horiba, France) fitted with Olympus confocal microscope (objective aperture $\times 100$, NA 0.90) with a diffraction grating of 600 groove/mm and an argon laser with an excitation wavelength of 514 nm. The microscope output power was 0.24 mW. Each sample was measured three times at different points on the surface to obtain statistically reliable data. Before measurements, calibration was performed by recording Raman spectra from the single-crystal silicon.

1.3. Study of electrophysical characteristics

The samples' resistance was measured using a four-probe measuring complex PM5 (Cascade Microtech, USA) connected to a 34401A multimeter (Keysight Technologies, USA). For each sample, at least five measurements were performed, followed by calculation of the average resistance value. The impedance of Ag/AgCl electrodes and dry electrodes fabricated from the aforesaid nanocomposites was measured by the impedance analyzer LCR-8000G (Good Will Instrument, Taiwan) with a test signal of 1 mV. The frequency range of 10–500 Hz was chosen to match the typical ECG bandwidth. The electrodes were placed on the forearm at a distance of 8 cm from each other with registering the dependence of the impedance modulus on frequency. All electrodes were normalized to a contact area of $78.5\ \text{mm}^2$ for comparability of measurement results. Experiments were conducted to study the effect of a medium simulating human sweat on the impedance of electrodes. The simulated human sweat ($\text{pH} = 8.5$) included: lactic acid 2.5 g/l, urea 0.9 g/l, ammonia 1.35 g/l, sodium 0.9 g/l, potassium 0.2 g/l, calcium 0.015 g/l and magnesium 0.0013 g/l. The above components were mixed

in deionized water using a magnetic stirrer. The electrodes were immersed in the prepared solution and kept in it for 7 days. This period was chosen taking into account the fact that most modern ECG monitoring devices register human biopotentials for exactly 1 week. The stability of the developed electrodes over the selected period of time will ensure the collection of information necessary for doctors about the state of the human cardiovascular system.

The tensile test was carried out using a dynamometer. The electrodes were clamped and stretched to 50% of elongation, and the applied force was measured. Additionally, the effect of mechanical pressure on impedance was investigated: a tonometer cuff was used to apply pressure uniformly, and the effect of pressure in the range from 0 to 4 kPa was studied. The ECG signals were registered using CardioVisor-06s electrocardiograph (Medical Computer Systems, Russia) in a three-channel configuration with electrodes fixed with flexible adhesive tape and 30-second recordings. The resulting ECG reflected the averaging of all action potential vectors occurring at a specific moment of cardiac activity.

1.4. Biocompatibility study

To evaluate the biocompatibility of the electrodes, HEF cells were used. Cell suspensions were plated onto the surfaces of electrode samples. The plating density was $3.5 \cdot 10^5$ cell/ml with 1.7 ml of cell suspension added to each Petri dish with electrodes, followed by incubation in a CO_2 incubator for 48 h. After incubation, the cells were stained with Hoechst 33342 dye and examined using a fluorescence microscope BX43 (Olympus, Japan).

2. Experimental results and discussion

At the initial stage, a solvent was selected that ensures homogenization of carbon nanomaterials in dispersion without affecting the PDMS curing process. A study of the composite curing after solvent evaporation showed that several solvents, including ethanol and isopropanol, affected the structure of PDMS matrix, causing partial inhibition of its curing. After the interaction of PDMS base with n-hexane, the curing time of PDMS did not increase. Taking this into account, n-hexane was selected as the most suitable solvent that didn't interfere with curing of PDMS and at the same time ensured uniform distribution of carbon nanomaterials in PDMS matrix. Fig. 1 illustrates the resistances of PDMS/SWCNT/rGO before and after laser exposure with various intensities.

Pulsed laser irradiation with an intensity of $7\ \text{kW/cm}^2$ resulted in a 2.3-fold decrease in resistance down to $(45 \pm 2)\ \text{k}\Omega$. Increasing the irradiation intensity to $15\ \text{kW/cm}^2$ further reduced the resistance by 13 times compared to the original composite to $(8 \pm 2)\ \text{k}\Omega$. However, further rise of intensity resulted in higher resistance of samples: after exposure to $27\ \text{kW/cm}^2$ the composite

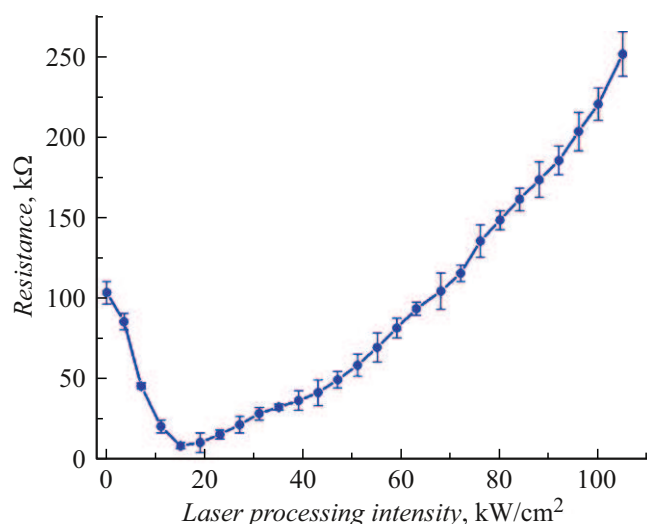


Figure 1. Resistance of PDMS/SWCNT/rGO composite before and after laser processing with various intensity.

resistance was (32 ± 2) kΩ, at 51 kW/cm^2 — (58 ± 7) kΩ, and at 105 kW/cm^2 — it was (251 ± 14) kΩ. According to the authors' previous studies, laser irradiation can induce formation of bonds between carbon nanomaterials to form branched conductive nets [34]. The observed decrease in resistance may be due to the formation of additional covalent bonds both between nanotubes and between SWCNT and rGO flakes. Laser irradiation promoted the generation of defects in CNT and reduced graphene oxide flakes, creating chemically active sites in the walls of SWCNT and graphene flakes, followed by the formation of new C–C bonds between adjacent active centers of carbon nanomaterials, thus, leading to formation of SWCNT/rGO hybrid nanostructures in PDMS matrix with reduced contact resistance (Fig. 1). Further increase in laser intensity caused degradation of the structure of nanomaterials and significant formation of amorphous carbon, leading to an increase in the resistance of the composite.

In order to analyze the structural features of PDMS/SWCNT/rGO composites based on hybrid nanostructures of carbon nanomaterials, their surface morphology was studied using SEM method. Fig. 2 illustrates SEM images of the composite surface.

As can be seen from the images obtained, the surface of the composite was characterized by high uniformity before laser exposure, while SWCNT agglomerates were observed, where all the nanotubes were coated with a polymer layer. Laser exposure with an intensity of 15 kW/cm^2 caused changes in the morphology of the composite surface with the formation of pronounced irregularities, which subsequently provided better conformal contact between the composite and the skin surface containing microcracks and inhomogeneities. The images also illustrate carbon nanomaterials — SWCNT agglomerates and reduced graphene oxide flakes — forming an interconnected conductive network. High-intensity laser irradiation removed the

surface layer of PDMS, thereby improving electron transport between human skin and the electrode during biopotential measurements. Additionally, laser energy was transferred to the carbon nanomaterials, stimulating their interconnection into conductive networks, increasing the conductivity of the composite.

To analyze the process of rearrangement of carbon nanomaterials and the formation of hybrid nanostructures in PDMS matrix after laser exposure, the Raman spectra of the samples were measured (Fig. 3).

Raman spectroscopy provides a rapid, robust, and non-destructive method for quantifying and characterizing defects in SWCNTs formed by laser surface modification, as each carbon nanomaterial has specific modes. The study of CNT functionalization also involves the assessment of the quality of surface modifications through the analysis of changes in specific modes, which allows for accurate structural and electronic characterization [35]. The main contribution to the Raman spectra of the presented experimental samples before and after laser irradiation, respectively, was made by SWCNT, which exhibited all the unique characteristic modes: RBM at 155 and 185 cm^{-1} , *D* at 1348 cm^{-1} , *G*, demonstrating the splitting at *G*[−] (1572 cm^{-1}) and *G*⁺ (1596 cm^{-1} before and 1593 cm^{-1} after laser exposure) which evidences the presence of semiconducting SWCNT in the sample and *G*' around 2670 cm^{-1} [36,37]. Thus, the obtained results are in good agreement with previously obtained data on characteristic modes for carbon nanomaterials.

Laser irradiation of a sample at high energy density can cause amorphization and distortion of the carbon surface due to oxidation of SWCNT, but here, the carbon nanomaterials did not undergo critical structural distortions. Standard evaluation criteria were used, including well-established I_D/I_G and $I_{G'}/I_D$ ratios characterizing purity and crystallinity. Monitoring changes in this ratio before and after processing yields a level of defects that is sufficient to assess the effectiveness and consequences of laser functionalization or ablation processes on nanotubes. In this case, I_D/I_G ratio scales linearly with the defect density, since each defect (*sp*³-center, vacancy, or others) activates additional scattering centers of *D*-band, which also cause a mode shift and an increase in the cross-section. For the initial sample the ratios were $I_D/I_G = 0.092$ and $I_{G'}/I_D = 1.73$, while for the laser processed sample — $I_D/I_G = 0.138$ and $I_{G'}/I_D = 1.65$. An increase in I_D/I_G by 1.5 times and a slight decline in $I_{G'}/I_D$ after laser irradiation indicates the occurrence of distortions in the surface structure (reduction of *sp*² carbon forms) caused by curvatures and local formation of bonds between carbon nanomaterials in defective areas [34]. Presumably, the presence of a binding polymer matrix in a sample based on SWCNT/rGO causes additional mechanical stresses and contributes to the formation of structural defects during laser exposure. Moreover, the broadening of *D*-mode (half-width growth by 1.5 times) and shift of *G*-mode by 3 cm^{-1} was observed. It is important to note that such a change

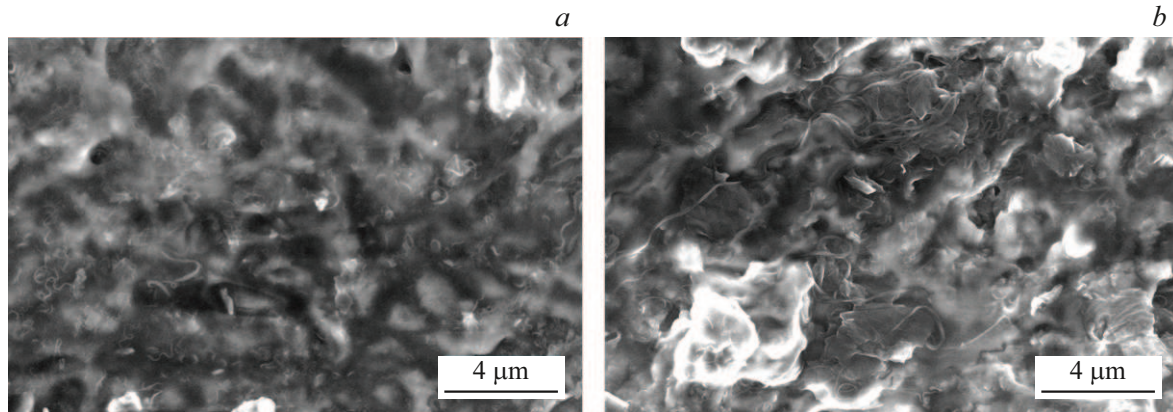


Figure 2. SEM images of PDMS/SWCNT/rGO surfaces before (a) and after (b) laser processing with an intensity of 15 kW/cm^2 .

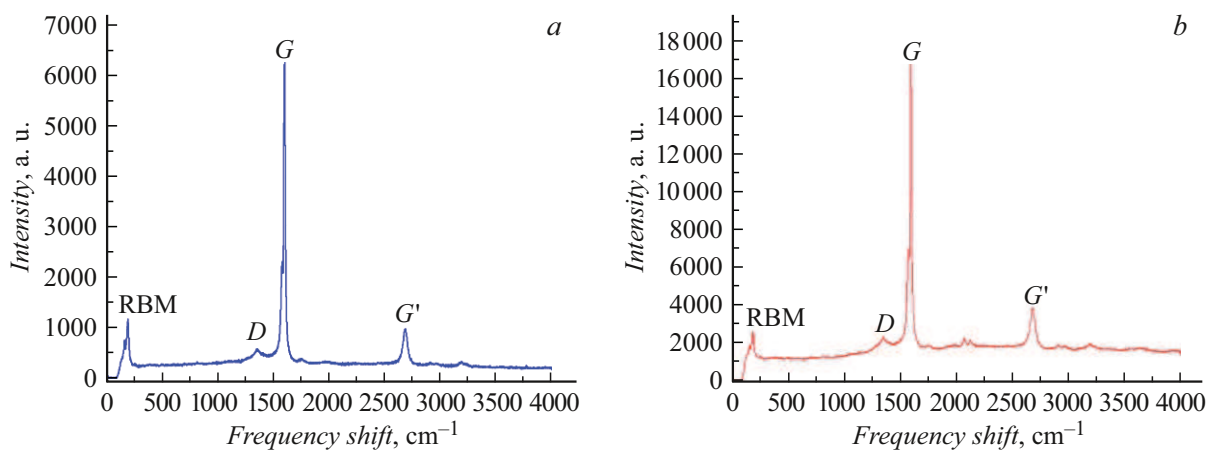


Figure 3. Raman scattering spectra of PDMS/SWCNT/rGO before (a) and after (b) laser exposure with an intensity of 15 kW/cm^2 .

enables to trace the structure defects in SWCNT, but is not critical, which indicates the locality and selectivity of the fusion zones formed after laser irradiation.

After determining the threshold intensity of laser irradiation that ensures the formation of hybrid nanostructures in the polymer matrix, electrodes were manufactured based on PDMS/SWCNT/rGO composites using laser processing (15 kW/cm^2) (Fig. 4, a). To connect to measuring instruments, wires were integrated into the electrodes during manufacture. The elastic moduli of the formed electrodes based on PDMS/SWCNT/rGO composites and commercial Ag/AgCl electrodes were studied. The highest elastic modulus of the commercial electrode was $(386 \pm 12.7) \text{ kPa}$, while the formed electrode was characterized by an elastic modulus of $(180.0 \pm 5.3) \text{ kPa}$. It is known that the average modulus of elasticity of human skin is about 200 kPa . Thus, the obtained results show that the elastic modulus of the fabricated electrodes is close to the elastic modulus of the skin, which in the future will ensure comfortable wearing of electrodes based on PDMS/SWCNT/rGO composites during long-term ECG monitoring.

Since electrodes placed on the skin may be subject to mechanical stress during long-term monitoring, it was needed to study the effect of mechanical pressure on the electrode impedance. A tonometer cuff was used to ensure uniform pressure on the electrodes. The frequency dependences of the impedance of electrodes based on PDMS/SWCNT/rGO composite were recorded without pressure and under applied pressures of 4, 8 and 12 kPa (Fig. 4, b). The frequency varied within $10\text{--}500 \text{ Hz}$.

Applying a pressure of 4 kPa to the electrodes resulted in a lower impedance over the entire frequency range. The greatest impedance decline from 1998 to $426 \text{ k}\Omega$ was characteristic for 10 Hz frequency. At frequencies above 100 Hz , the decrease in impedance was less significant. Further increase in pressure also caused a decrease in impedance amplitude over the entire frequency range. The lowest impedance in the range from 183 to $11 \text{ k}\Omega$ was characteristic for a pressure of 12 kPa .

To evaluate the performance of the electrodes during prolonged contact with the skin, a suspension simulating human sweat was used. One of the key indicators of the effectiveness of skin electrodes for registration of biopotential

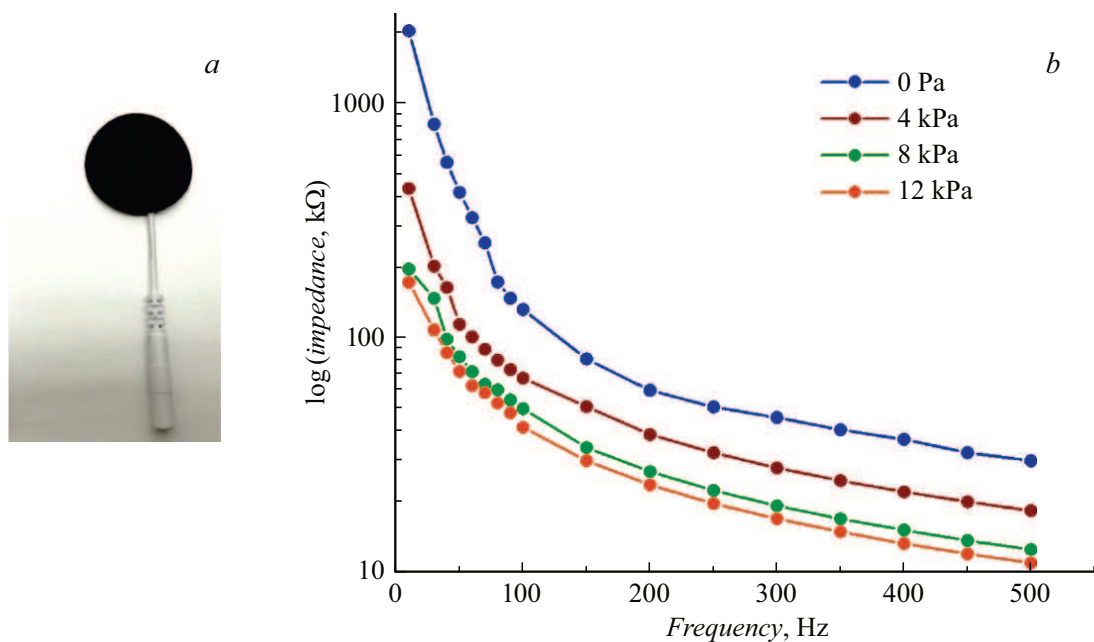


Figure 4. General view of an electrode based on PDMS/SWCNT/rGO composite (a), electrodes impedance versus frequency depending on the applied pressure within 0–12 kPa (b).

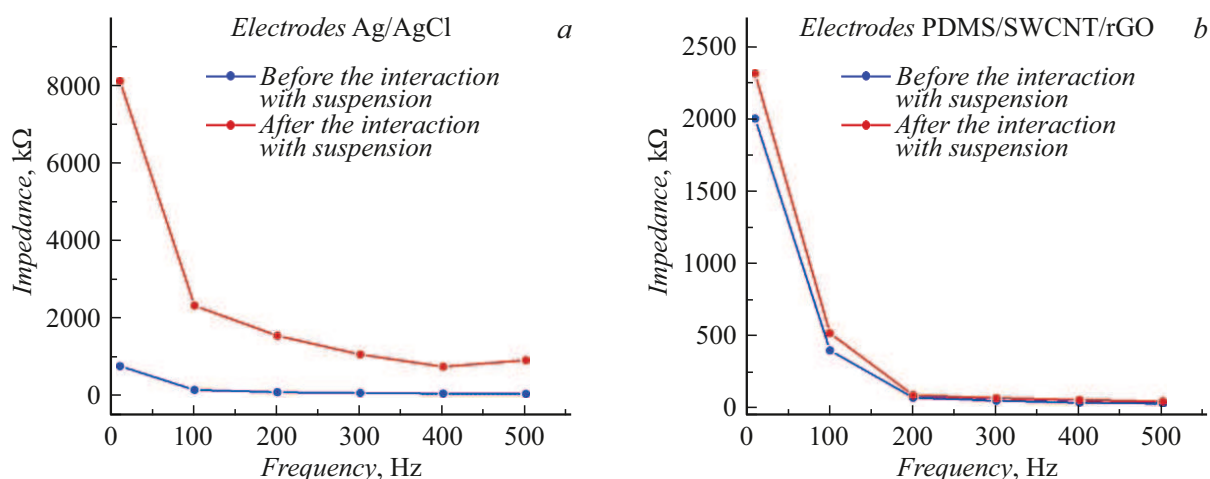


Figure 5. Dependencies of impedance on the frequency for Ag/AgCl electrodes (a), PDMS/SWCNT/rGO electrodes (b) before and after their interaction with a human sweat simulating suspension.

tials is impedance. In connection with this, measurements were made of the dependence of the impedance amplitude of the electrodes on the signal frequency. For comparison, impedance measurements of standard Ag/AgCl electrodes were also performed. Measurements were performed before and after immersion of the electrodes in the suspension. Figure 5 illustrates the obtained curves of impedance amplitude versus frequency.

Initially the amplitude of Ag/AgCl electrodes impedance plummeted from 750 to 35 kΩ with the signal frequency growth from 10 to 500 Hz (Fig. 5, a). After interaction with the suspension the impedance amplitude declined from

8094 to 903 kΩ. An important issue was the rise of amplitude observed at frequencies of 10–100 Hz. These results indicate a high degree of degradation of Ag/AgCl electrodes when interacting with a suspension simulating sweat. Electrodes based on PDMS/SWCNT/rGO composite showed an impedance change from 1998 to 36 kΩ before immersion in the suspension. Exposure to suspension for 7 days did not result in a significant increase in the impedance amplitude, which varied in the range from 2312 to 51 kΩ. Thus, in case of Ag/AgCl electrodes, the interaction with the suspension caused a more significant increase in impedance compared to PDMS/SWCNT/rGO electrodes. This is

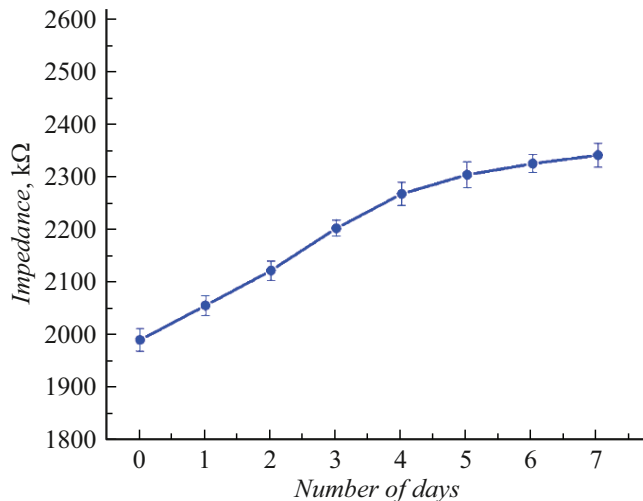


Figure 6. Dependence of impedance on wearing time when an electrode based on a PDMS/SWCNT/rGO composite is applied to the skin for 1–7 days.

expected to be interrelated with degradation of the gel layer of Ag/AgCl electrodes. After modeling the effect of human sweat on the electrode impedance, PDMS/SWCNT/rGO composite electrodes were placed on the skin to record daily changes in impedance for 7 days at a signal frequency of 10 Hz (Fig. 6).

Initially the impedance amplitude was (1992 ± 21) kΩ, and after 7 days it grew up to (2407 ± 25) kΩ. The most significant change in amplitude was registered during 1–5 days of wearing, after which the amplitude changed insignificantly. This effect indicates the stabilization of PDMS/SWCNT/rGO electrode upon contact with human skin. Based on the data obtained, the electrode can potentially provide long-term ECG monitoring for 7 days or more without significant signal degradation. Thus, the results of measurements of the electrode impedance over 7 days confirm the conclusions obtained by modeling the interaction of electrodes with a suspension simulating human sweat. After impedance measurements, ECG signals were recorded using the fabricated PDMS/SWCNT/rGO electrodes, as well as Ag/AgCl electrodes. Figure 7 shows electrocardiograms recorded using dry PDMS/SWCNT/rGO electrodes and commercial Ag/AgCl electrodes.

The quality of the electrocardiogram is assessed primarily by the parameters of PQRST complex peaks. The signal recorded using Ag/AgCl electrodes showed high stability, but compared to the signal from PDMS/SWCNT/rGO electrodes, it was less stable over time. The average amplitude of *R* peak for electrodes based on PDMS/SWCNT/rGO composite was (0.86 ± 0.08) mV. For the commercial Ag/AgCl electrodes the average amplitude of *R*-peak was (0.81 ± 0.07) mV. The findings indicate that, accounting for the measurement errors, the quality of the registered signal is comparable to the signals recorded by the widespread Ag/AgCl electrodes, while the absence of a gel layer in

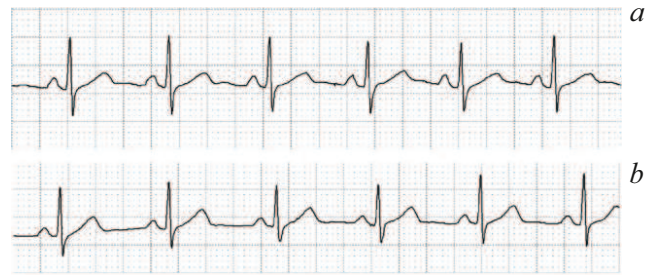


Figure 7. ECG recorded using Ag/AgCl electrodes (a) and electrodes based on PDMS/SWCNT/rGO composites (b).

dry PDMS/SWCNT/rGO electrodes is their advantage over commercial analogues, since no degradation of signal quality will be observed during the long-term use.

To assess the biocompatibility of PDMS/SWCNT/rGO electrodes, studies were conducted on the viability of HEF skin cells on their surface. Figure 8, *a* shows an image obtained by fluorescence microscopy of fibroblast cells after 48 h of incubation on PDMS surface, and Figure 8, *b* — illustrates a similar image of fibroblasts on the surface of the PDMS/SWCNT/rGO electrode.

The resulting images clearly show the oval-shaped cell nuclei, as well as areas of intercellular contacts, indicating numerous attached and grown cells that form structures due to intercellular connections. The high density of cell growth confirms their successful adhesion to the electrode surface and active proliferation.

Based on the obtained experimental data and their comparison with other developments in creating the electrodes for ECG procedures, it can be concluded that the formed electrodes made of PDMS/SWCNT/rGO demonstrate impedance values 8.6 times lower compared to metal electrodes [38]. Compared with PEDOT:PSS-based polymer electrodes and textile electrodes, the developed electrodes showed on average 2.8 times lower resistance and 85 times lower impedance [39,40]. Thus, PDMS/SWCNT/rGO composite electrode developed in this study has a resistance comparable to that of metal-based electrodes and lower resistance and impedance compared to textile or polymer electrodes.

Conclusion

Dry electrodes for long-term ECG monitoring were developed based on composites of hybrid nanostructures consisting of SWCNT and reduced graphene oxide flakes in a PDMS matrix formed by laser processing (PDMS/SWCNT/rGO). A method for uniform homogenization of carbon nanomaterials in the volume of a liquid dispersion composite using a solvent was developed. The study of the composite curing process showed that *n*-hexane is the most suitable solvent that does not inhibit the polymerization PDMS. It was found that PDMS/SWCNT/rGO had resistance of (103 ± 7) kΩ at low concentration of

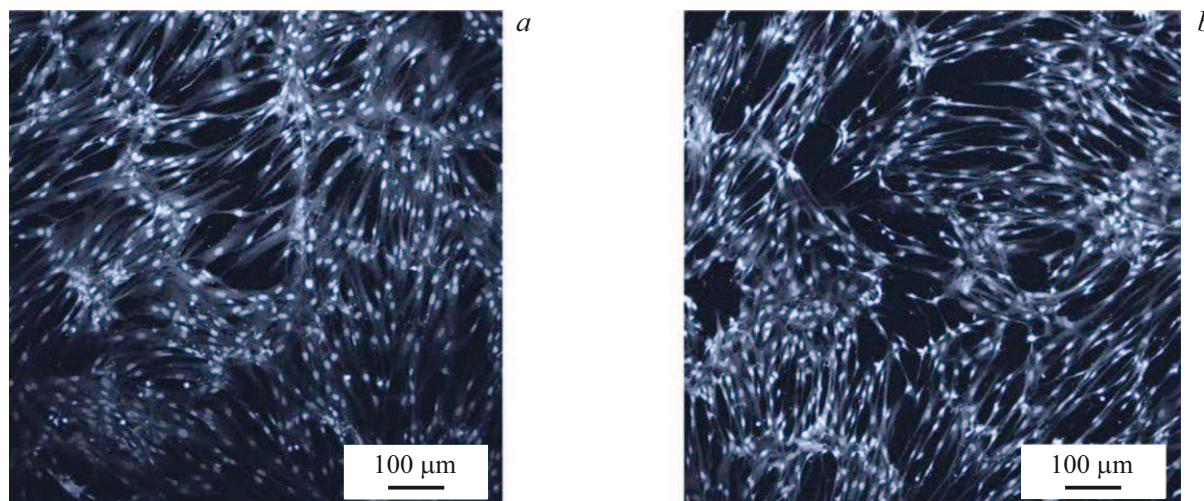


Figure 8. Fluorescence microscopic images of HEF cells grown on surfaces: PDMS (a), PDMS/SWCNT/rGO composite electrode (b) after 48 h incubation.

SWCNT/rGO 0.5/0.5 mass%. Laser treatment in the near infrared wavelength range with an intensity of 15 kW/cm^2 reduced the resistance by 13 times to $(8 \pm 2) \text{ k}\Omega$. It was found that laser irradiation changes the surface morphology of the composite, with carbon nanomaterials forming conductive networks that increase the electrical conductivity of the composite. Studies using Raman spectroscopy revealed an increase in I_D/I_G ratio and, as a consequence, the formation of bonds between carbon nanomaterials at defect sites. Electrodes based on PDMS/SWCNT/rGO composite were manufactured for ECG registering. A study of the influence of mechanical pressure on the impedance of the formed electrodes showed that an increase in pressure of 0–12 kPa leads to a plummeted impedance in the entire frequency range of 10–500 Hz. It was also found that the formed electrodes had a characteristic elastic modulus $(180.0 \pm 5.3) \text{ kPa}$, comparable to the elastic modulus of human skin. After 7 days of exposure to a medium simulating human sweat, electrodes based on PDMS/SWCNT/rGO composites demonstrated high stability, while traditional Ag/AgCl electrodes demonstrated degradation of impedance amplitude. To prove the stability of PDMS/SWCNT/rGO electrodes, a 7-day dynamics of impedance change was recorded at a signal frequency of 10 Hz while the electrodes were placed on the skin. The findings proved high stability of the fabricated electrodes during long-term contact with human skin. ECG measurements showed that the signal quality registered using PDMS/SWCNT/rGO composite electrodes was comparable to that of traditional Ag/AgCl electrodes. However, the absence of a gel layer in dry composite-based electrodes in terms of a long-term use turned out to be a benefit compared to the commercial analogues. The biocompatibility of PDMS/SWCNT/rGO electrodes was demonstrated by studying the viability of HEF cells on the electrodes surface. After 48 h of cultivation, numerous attached and proliferating cells

were observed on the surface of the composite. These results demonstrate that the developed electrodes based on PDMS/SWCNT/rGO composite are suitable for use in wearable devices, including devices for continuous ECG monitoring for at least 7 days.

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Conflict of interest

The authors declare that they have no conflict of interest.

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