

Investigation of the conductive properties of composite polymer materials based on polyvinyl alcohol doped with single- and double-walled carbon nanotubes

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Abstract

The development of nanotechnology requires more advanced computer modeling methods to study the properties of nanoparticles and clusters. These particles are the main “building material” in nanotechnology, determining the unique properties of the created products. Computer modeling makes it possible to predict the behavior of nanoparticles, their interaction with the environment and with each other, as well as optimize the processes of their synthesis and application.

Due to their lightness and flexibility, conductive polymers open up new possibilities for the development of flexible and wearable electronics. Currently, research is quite widespread on the creation of new polymer materials, which are obtained by modifying known polymers with various fillers, including nanomaterials. One of the well-known nanomaterials is carbon nanotubes. The existing applications of nanotubes are almost limitless.

In this work, the well-known polymer polyvinyl alcohol and carbon nanotubes are chosen as the main objects. A theoretical study of the possibility of creating a stable Polymer-CNT complex using the quantum chemical calculation method DFT has been performed. Single- and double-walled carbon nanotubes were used in the study. The mechanism of interaction of nanotubes with fragments of polyvinyl alcohol has been studied. The electron-energy structure of the obtained polymer nanocomposites is analyzed and a conclusion is made about the conductive properties of the resulting complex.

Keywords

density functional theory, electrical and sorption properties, polymer nanocomposites, poly(vinyl alcohol), single- and double-walled carbon nanotubes

1. Introduction

In recent decades, interest in polymer nanocomposites based on CNTs has increased dramatically due to their

unique combination of properties, which is unattainable for traditional polymers [1]. Conventional polymers have a low modulus of elasticity and are dielectrics, which limits their use in electronics. However, the introduction

of carbon nanotubes (CNTs) into the polymer matrix makes it possible to overcome these limitations by creating materials with improved mechanical properties (high modulus of elasticity, strength), electrical conductivity, and versatility (sensory, antistatic, and shielding properties). CNT polymer composites have become a bridge between nanoscience and industry. Their adaptability makes it possible to solve problems from creating flexible electronics to obtaining new materials with predefined properties. However, success depends on overcoming technological barriers, such as scaling up CNT synthesis and developing a method for uniformly dispersing CNTs into a polymer matrix. Great predictions are being made about the possibility of creating so-called “smart” composites, where CNTs will play the role of not only fillers, but also active elements in adaptive systems.

CNTs are ideal fillers due to their unique conductivity and high aspect ratio (even at low concentrations (1–5 wt.%) they form a conductive network through the percolation effect), record-high strength properties, and most importantly, the ability to reinforce a polymer (increase tensile strength by 20–50 % due to load transfer to nanotubes) [2–5].

Composite polymer materials based on CNTs have a number of properties that make them indispensable in modern technologies. For example, due to their flexibility, lightness, conductive properties, and low-temperature processing, these materials can be used as flexible substrates, conductive layers, and light-emitting elements in OLED displays, allowing the creation of energy-efficient lamps and sensor devices. Polymer-based emission layers provide a bright glow with low energy consumption, which makes it possible to use composite polymer materials as organic solar cells. Thus, the active layer of composites (for example, a mixture of donor and acceptor polymers) forms a heterojunction that improves charge separation. This reduces the cost of production and expands applications in flexible and portable solar panels. In addition, some non-toxic polymers are widely used in medicine, and composite materials based on them may become promising materials for creating advanced sensors for soft robots, implantable devices, and drug delivery systems. In addition, the flexibility of the materials ensures integration with biological tissues [6–9].

Polyvinyl alcohol (PVA) is a synthetic polymer used since the early 1930s in a wide range of industrial, commercial, medical, and food applications, including resins, varnishes, surgical filaments, and food contact materials. These hydrophilic biocompatible polymers have a number of unique properties such as excellent flexibility and strong barrier properties, the material is non-toxic and resistant to acid and alkaline conditions. PVA is approved for use in several medical applications, including transdermal patches, the preparation of jellies that dry quickly when applied to the skin, as well as in tablets with immediate and delayed release. Polyvinyl alcohol polymers are also used for the controlled release of oral medications.

Ophthalmic solutions such as synthetic tears may also contain PVA, as it provides good dispersion and coating properties [10–12].

In recent years, flexible wearable electronic devices have received widespread attention in the fields of human health monitoring, bioelectronic interfaces, and human-computer interaction. Conductive hydrogels have gradually become the best candidate materials for flexible wearable electronic devices due to their good conductive properties, modulus of elasticity similar to natural skin tissue, and adjustable mechanical properties. Hydrogel preparation can be performed using both natural and synthetic polymers. Synthetic polymers are usually preferred because natural polymers, although more biocompatible, may not have sufficient mechanical strength and may cause inflammation due to contamination by pathogens. Synthetic polymers, on the other hand, can be adapted according to individual needs in terms of functionality. The use of a synthetic polymer as a drug delivery system usually raises concerns about its biocompatibility. With the discovery of many synthetic polymers, several of them, potentially suitable for use in a drug delivery system, have been investigated for their biocompatibility. Polymers such as polyacrylic acid, polyacrylamide, polysaccharides, poly(N-isopropylacrylamide) and polyvinyl alcohol have potential for hydrogel synthesis. PVA was chosen as a component of the hydrogel because of its favorable water-soluble, physico-chemical properties and its biocompatibility. Chemically crosslinked PVA hydrogel is attracting increasing attention in the field of biomedicine [11].

Polyvinyl alcohol polymers can be used to create mixtures and composites, including various biopolymers. The introduction of CNTs will create composites with a unique combination of flexibility, transparency and conductivity. Thus, PVS composites/CNTs can combine the unique properties of both components, paving the way for the creation of flexible, biocompatible and multifunctional electronics [12, 13].

Thus, the inclusion of conductive fillers in the polymer matrix of polyvinyl alcohol will allow the use of such composite materials for many advanced applications in electronics. The combination of polymers with carbon nanotubes opens the way to innovation, but requires solving technical and economic problems. Already today, such materials are used in experimental smart watches, medical patches, and even flexible displays, and in the coming decades they may become the basis for mass-scale “electronic skin” and autonomous wearable systems.

However, despite the wide possibilities of using PVS-based composites with embedded nanotubes, the mechanism leading to the creation of a stable composite system has not been clarified. Therefore, the theoretical studies presented in this paper on the possibility of creating a stable PVS-CNT complex, performed using the quantum chemical calculation method DFT, are very relevant.

2. Theory/calculation

The density functional theory is used to study the features of the electronic energy structure of a nanocomposite based on PVA doped with carbon nanotubes, and the mechanisms of interaction between CNTs and fragments of polyvinyl alcohol [14]. The essence of this method lies in the use of electron density distributions in the description of atomic-molecular systems. Thus, according to DFT, all the electronic properties of the system, including energy, can be obtained from the electron density $\rho(r)$, without knowledge of the wave functions [15–17]. Therefore, the electron energy in the DFT method can be calculated as:

$$E[\rho] = T[\rho] + V_{\text{en}}[\rho] + V_{\text{ee}}[\rho]. \quad (1)$$

where $T[\rho]$ is kinetic energy, $V_{\text{en}}[\rho]$ is the potential energy of electron-nuclear interactions, $V_{\text{ee}}[\rho]$ is the energy of interelectronic interactions.

The energy of interelectronic interactions can be represented as:

$$V_{\text{ee}}[\rho] = V_{\text{Coul}}[\rho] + V_{\text{xc}}[\rho]. \quad (2)$$

where $V_{\text{Coul}}[\rho]$ is the energy of the Coulomb interaction between electrons, $V_{\text{xc}}[\rho]$ is the exchange-correlation energy.

A hybrid method of approximation, namely the B3LYP method was used to study the system. The main advantage of the B3LYP method is its high accuracy. Calculations were carried out using a valence-split basis set of type 3–21G. This functionality with the selected basic set is well adapted to the selected systems.

3. Results and discussion

3.1. The effect of adsorption of polyvinyl alcohol on the surface of CNT

To predict the possibility of creating a new composite material, the problem of the adsorption activity of the polymer material under study in relation to CNTs is of fundamental importance. The theoretical studies carried out in [18] on the interaction of the structural unit of polyvinyl alcohol (PVA) with the CNT surface using quantum chemical modeling using the Density functional theory (DFT) calculation method confirmed the possibility of creating a stable complex of PVA/CNT. Consideration of the properties of PVS-CNT composites requires a high precision and complex computational method. An ideal candidate for this is the DFT. The method consists of finding the total energy of a system as a unique functional of the electron density, eliminating the need for calculating the many-body wave function. DFT supports the calculation of various structural, chemical, optical, spectroscopic, elastic, vibrational, and thermodynamic properties. Consequently, DFT-based quantum chemical calculations have become a key approach for investigating electronic parameters such as charge distribution, infrared spectra, Raman scattering, ultraviolet-visible (UV-vis) spectra, and nuclear magnetic resonance (NMR) spectra [19]. In the same work, we conducted detailed theoretical studies of the interaction of a fragment of polyvinyl alcohol with CNTs of different layers. Single-walled carbon nanotubes of the (9,9) type and double-walled carbon nanotubes consisting of two nanotubulenes of the (6,6) and (9,9) types are considered (Fig. 1).

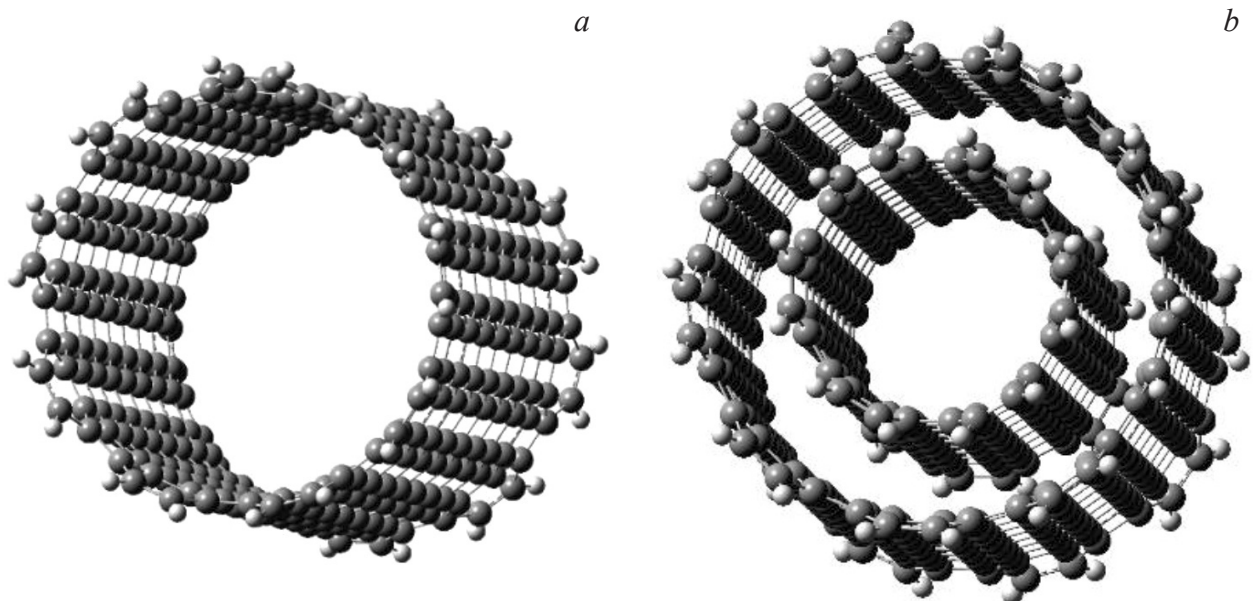


Figure 1. (a) SWCNT type of (9,9); (b) MWCNT type of (9,9) and (6,6)

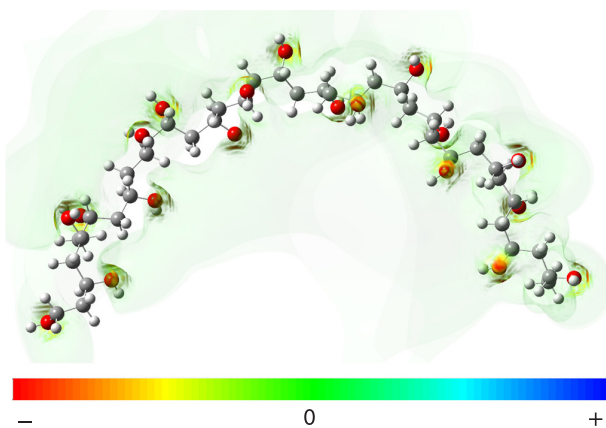


Figure 2. Map of the electrostatic potential of the fragment $[\text{C}_2\text{H}_4\text{O}]_{18}$

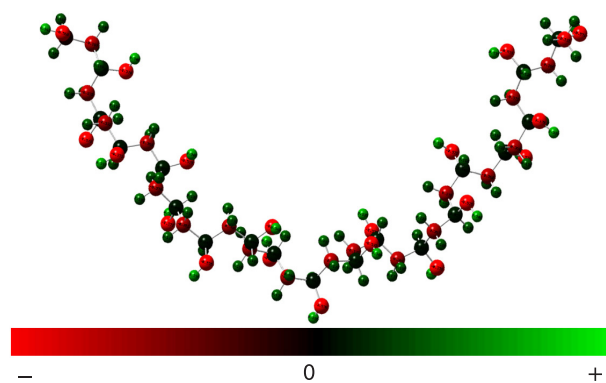


Figure 3. Charge distribution of the fragment $[\text{C}_2\text{H}_4\text{O}]_{18}$

In the structure of a polyvinyl alcohol polymer fragment consisting of 18 structural units of PVA ($[\text{C}_2\text{H}_4\text{O}]_n$, where $n = 18$), the most active center was selected, which is capable of providing stable bonding of the studied fragment to the surface of the nanotubes, namely the oxygen atom. In the course of quantum chemical research, maps of the electrostatic potential (Fig. 2) and the charge distribution in the system (Fig. 3) were also analyzed. In the case of molecular modeling, the electrostatic potential on the surface of a molecule is often calculated based on the electron density distribution obtained from quantum chemical calculations. This makes it possible to visualize the electrostatic properties of a molecule, which is important for predicting interactions with other molecules, solubility, and reactivity. Figure 1 shows a visualization of the electrostatic field in the form of color coding: red – negative potential (high electron density), blue – positive. It can be seen that a high electron density is concentrated near the oxygen atom. Based on the analysis of the obtained results, a high concentration of electron density on the oxygen atom was noted, confirming that this atom can be a reaction center.

The process of adsorption interaction of the PVA fragment with the central part of the CNT cluster was modeled by stepwise approximation of the fragment to the selected carbon atom of the nanotube perpendicular to the CNT

surface. Figure 4 shows an example of an approximation of a PVA to a double-walled CNT (DWCNT).

As a result of the calculations performed, the energy values of the systems were obtained at each step, which made it possible to construct curves of the dependence of the interaction energy on the distance between the components “ $[\text{C}_2\text{H}_4\text{O}]_{18}$ – SWCNT” (see Fig. 3) and “ $[\text{C}_2\text{H}_4\text{O}]_{18}$ – DWCNT” (see Fig. 4). It is established that each curve has a minimum corresponding to the interaction at certain distances.

The adsorption energy ΔE_a was calculated as the difference between the total energies of the adsorption complex and the sum of the energies of noninteracting CNTs and the PVA fragment $[\text{C}_2\text{H}_4\text{O}]_{18}$ under study

$$\Delta E_a = E_{\text{a.d.c.}} - (E_{\text{CNT}} + E_{\text{mol}}). \quad (3)$$

where $E_{\text{a.d.c.}}$ is the energy of the adsorption complex obtained as a result of calculations, E_{CNT} is the energy of the CNT and E_{mol} is the energy of the $[\text{C}_2\text{H}_4\text{O}]_{18}$.

The values of ΔE_a revealed the fact of physical interaction (adsorption) of the $[\text{C}_2\text{H}_4\text{O}]_{18}$ with a CNT cluster (Fig. 5, Table 1):

- 1) The value of the adsorption energy during the interaction of the complex “ $[\text{C}_2\text{H}_4\text{O}]_{18}$ – SWCNT” using the active center of the oxygen atom turned out to be 0.335 eV, the adsorption distance $R_{\text{ad}} = 0.28$ nm;
- 2) The value of the adsorption energy during the interaction of the complex “ $[\text{C}_2\text{H}_4\text{O}]_{18}$ – DWCNT” using the active center of the oxygen atom turned out to be 0.329 eV, the adsorption distance $R_{\text{ad}} = 0.29$ nm.

So, the established fact of the interaction of a fragment of a PVA polymer with the surface of single- and double-walled carbon nanotubes explains the mechanism of creating a composite polymer material based on polyvinyl alcohol reinforced with nanotubes during the adsorption interaction of the polymer with CNTs, leading to the creation of stable complexes.

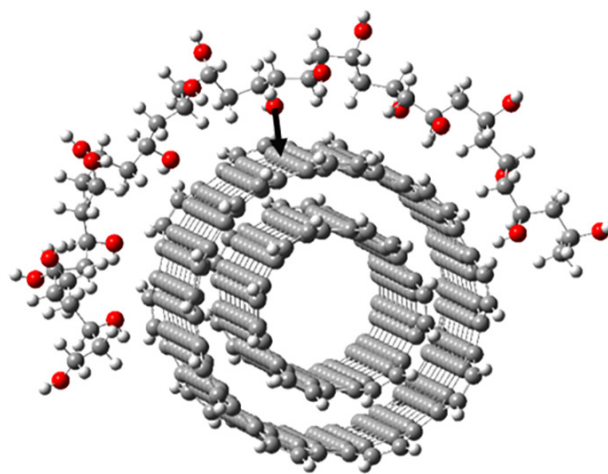


Figure 4. Adsorption interaction of the fragment $[\text{C}_2\text{H}_4\text{O}]_{18}$ and double-walled CNT

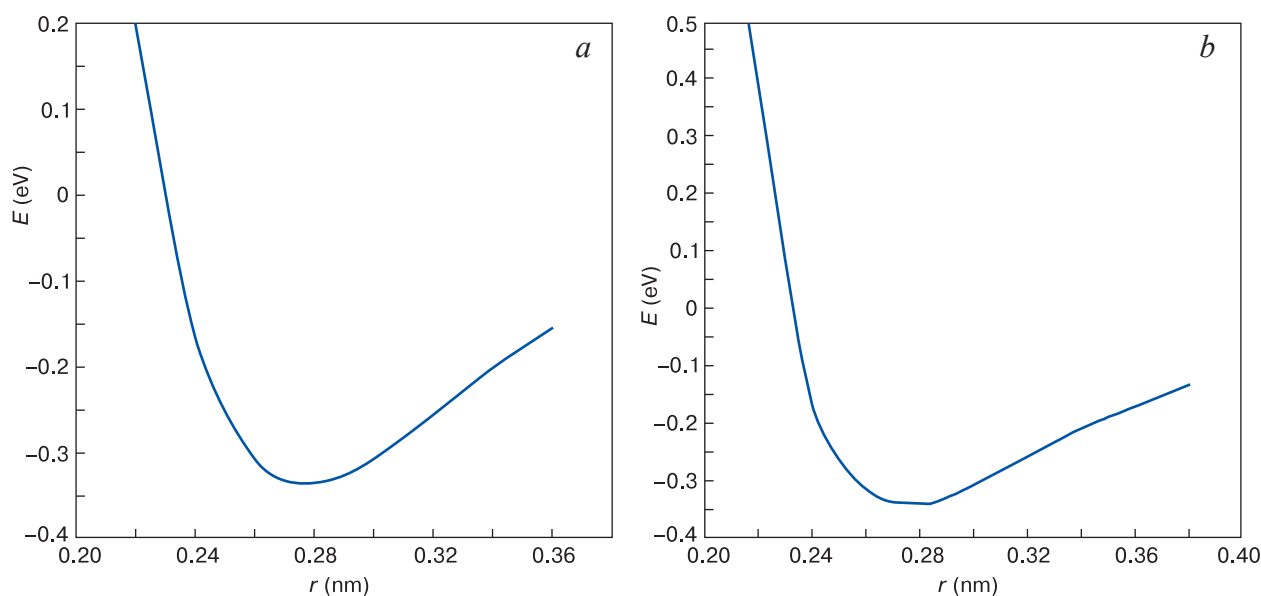


Figure 5. The curve of dependence of the interaction energy on the distance of the system: (a) $[\text{C}_2\text{H}_4\text{O}]_{18}$ – SWCNT; (b) $[\text{C}_2\text{H}_4\text{O}]_{18}$ – MWCNT

Table 1. The results of adsorption interaction of $[\text{C}_2\text{H}_4\text{O}]_{18}$ on the outer surface of CNTs

Types of CNT	Active center	Adsorption distance R_{ad} (nm)	Adsorption energy ΔE_a (eV)
SWCNT	Oxygen atom	0.28	0.335
MWCNT	Oxygen atom	0.29	0.329

3.2. Features of the electron-energy structure of polymer nanocomposites based on polyvinyl alcohol and CNT

The analysis of the electron-energy structure of complexes formed by single/double-walled CNT and $[\text{C}_2\text{H}_4\text{O}]_{18}$. The band gap ΔE_g is calculated, which is defined as the energy difference between HOMO (Highest Occupied Molecular Orbital is the highest-energy orbital with one or two electrons) and LUMO (Lowest Unoccupied Molecular Orbital is the lowest-energy orbital with no electrons) orbitals:

$$\Delta E_g = E_{\text{HOMO}} - E_{\text{LUMO}}. \quad (4)$$

The analysis of the band gap ΔE_g of the adsorption complexes revealed that the system is a semiconductor by type of conductivity (the value of ΔE_g varies from 0.36 eV to 0.43 eV for different complexes) [20–22].

Table 2. The results of the analysis of the band gap width ΔE_g of adsorption complexes

Types of CNT	Connection option (active PVA center)	The forbidden zone width ΔE_g (eV)
SWCNT	Oxygen atom	0.422
MWCNT	Oxygen atom	0.362

Thus, the introduction of CNTs into the polymer matrix of PVS, which is a dielectric by type of conductivity with $\Delta E_g = 7.967$ eV (Fig. 6), leads to the appearance of semi-conducting properties in the resulting polymer nanocomposite. The energy values of the orbitals were calculated using the GaussView 6.0.16 program, which is a set of routines used in combination with the main Gaussian 09 program [23–26]. The values of ΔE_g are shown in Table 2, visualization of single-electron energy levels of polymer clusters, CNTs and the complex “PVS+ CNT” is shown in Fig. 6.

These theoretical results are in good agreement with the results of experimental work related to the study of the conductive properties of composite materials [27–29].

So, the results obtained make it possible to predict the possibility of using nanocomposites based on polyvinyl alcohol doped with carbon nanotubes as a material for micro- and nanoelectronics devices with semiconductor properties and all the advantages of a polymer material.

Conclusions

Based on theoretical calculations performed using the quantum chemical calculation method DFT, the adsorption activity of the polyvinyl alcohol polymer in relation to single- and double-walled carbon nanotubes used to modify the selected polymer has been proven. The results obtained prove that the main mechanism for obtaining stable polymer complexes based on PVA doped with carbon nanotubes is the adsorption interaction of the polymer in question with single- or double-walled carbon nanotubes.

The features of the electron-energy structure of a polymer nanocomposite based on polyvinyl alcohol with embedded carbon nanotubes have been studied and analyzed. It has been established that the band gap of the

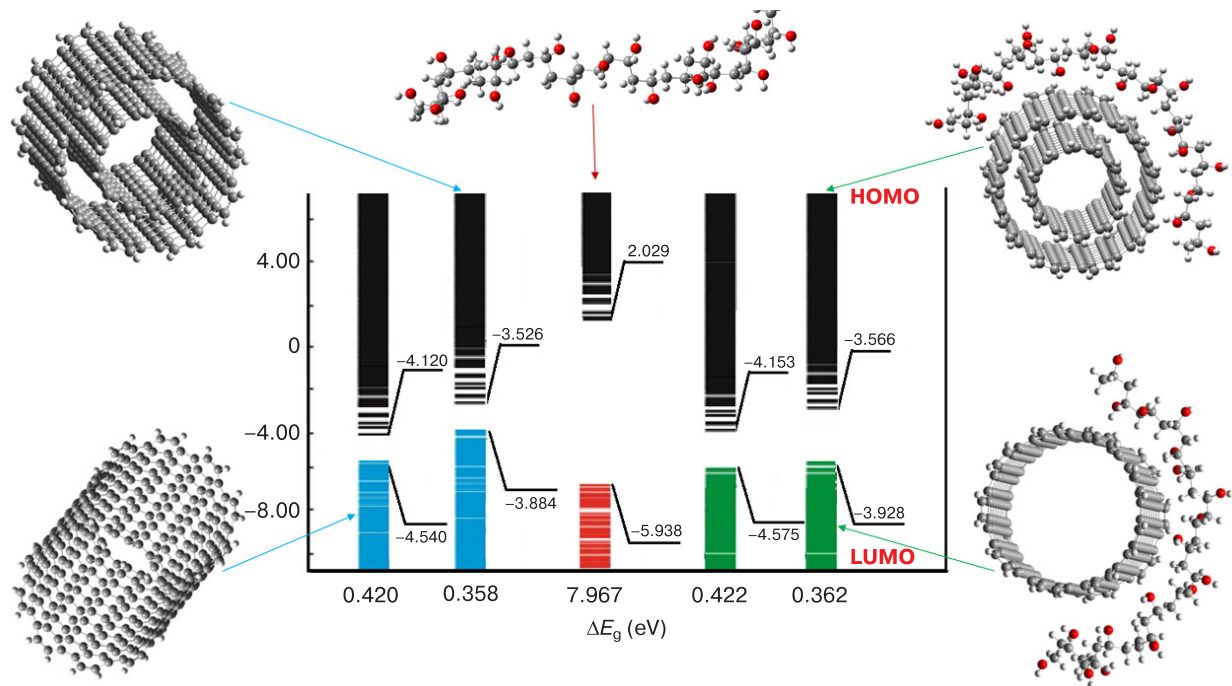


Figure 6. Visualization of single-electron energy levels of CNT/PVA clusters modeling the structure of a composite polymer material. Calculation of MO energies

adsorption complexes, defined as the energy difference between the upper filled and lower vacant orbitals, allows the created nanocomposite to be classified as a semiconductor, which distinguishes it from the original polymer with dielectric conductivity.

Due to the excellent properties of the polymer in question, as well as the results obtained, it is possible to predict the possibility of creating new composite polymer materials with variable conductive properties that can be widely used in electronics as electrode materials, various transistors, sensors, sensor elements, conductive adhesives, etc.

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Conflict of interests

The authors claim that they have no conflict of interest.

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