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Модель для оцінки площі поверхні вуглецевих наночастинок

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Model for carbon nanoparticles surface area estimation

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Ми описуємо модель для оцінки площі поверхні вуглецевих наночастнок, а саме одностінних вуглецевих нанотрубок, багатостінних вуглецевих нанотрубок, фулеренів, астраленів, нанопластин. Оцінка площі поверхні ґрунтується на припущенні, що наночастина описується геометричною фігурою з аналогічною наночастинці симетрією та геометричними розмірами. Далі розраховується площа поверхні геометричної фігури. У порівнянні з описаною у літературі моделлю [1], запропонована модель має меншу кількість «розрахункових» кроків для оцінки площі поверхні наночастинок, що зменшить час розрахунків при моделюванні властивостей вуглецевих наночастинок.

Ключові слова: вуглецеві нанотрубки, фулерени, астралени, вуглецеві нанопластини, питома площа поверхні.

We report a simple approach to surface area estimation of carbon nanoparticles, such as single-walled nanotubes, multi-walled nanotubes, fullerenes, astralens and nanosheets. Evaluation of surface area is based on a model in which nanoparticle is presented by geometric figure with the same symmetry and same parameters, and followed by calculation surface area of figure. The results are in good agreement with the microscopic properties and the surface area of carbon nanoparticles which are previously reported in the literature. In comparison with existing models [1], proposed model can reduce the number of steps for evaluation of carbon nanoparticle surface area; this is reducing calculation time in software for modeling properties of nanoparticles. The calculated data are reported in diagrams useful to correlate the specific surface area of samples, which can be helpful to adjust the synthesis conditions of carbon nanoparticles as well as applications where surface area plays an important role, e.g., in development of devices for energy storage, or creation of media for gas storage. At the same time, this approach would open up a new way to the quick estimation of surface properties of nonmaterial, such as nanoparticles and their aggregates.

Key words: carbon nanotubes, fullerenes, astralens, carbon nanosheet, specific surface area.

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Introduction

In contrast to the bulk materials based on carbon, carbon nanoparticles have a developed surface area, which plays a key role in their application. It is explained by the fact that the transition from microparticles to nanoparticles causes not only increasing of surface area, but also cause increasing of the number of atoms that can participate in intermolecular interactions with the environment [2]. For example, carbon nanotubes with a developed surface area can be used for producing a new generation of electrodes in batteries; it will reduce charging time and increase battery capacity. [3]. Fullerenes or carbon nanotubes can be used for creation of more efficient catalysts; it will reduce the temperature and the energy required for beginning of the reaction [4]. Also, the specific surface area plays a significant role in the developing electromechanical actuators [5], devices for storage of energy in electro-chemical capacitors [6] and as media for H_2 storage [7] Examples of other applications of carbon nanoparticles in engineering and medicine are described in works [8, 9]. Note that in liquid medium around the carbon nanoparticles confined water layer is formed [10]. As shown by Korolovych et al. [10, 11, 12] the presence of confined water in the system changes its thermodynamic properties and requires consideration

when applying liquid system as coolant. However, at the moment there is no systematic data on the surface area of carbon nanoparticles. For example, in work [13] specific surface area of carbon nanosheets is equal to ~2500 m^2/g and in work [13] it is 466 m^2/g. A similar situation for single-walled carbon nanotubes, in work [1] specific surface area of nanotubes approximately is 1300 m^2/g, and in work [14] it is 400 m^2/g. In addition, currently there are no simple model for estimation carbon nanoparticle surface area that is depending on their shape and size.

Therefore the aim of our work is to make a report on geometrical calculations establishing the relations between the surface area of main types of carbon nanoparticles (carbon nanotubes, fullerenes, carbon nanosheets, astralenes) and their geometrical characteristics. The results are reported in the form of diagrams which may be useful to other investigators. We also have shown that the experimental data of carbon nanoparticles surface area is in a good agreement with the theoretical calculations of surface area, which was estimated or calculated on the base of the microscopic characteristics, as reported by the respective authors.

Results and discussion

In a modern literature there are carbon nanoparticles of four main forms, namely: a cylindrical (carbon nanotubes), spherical (fullerenes), planar (carbon nanosheet) and toroidal (astralen). Schematic representation of the carbon nanotube, fullerene, carbon nanosheet and astralen are presented on Fig. 2.

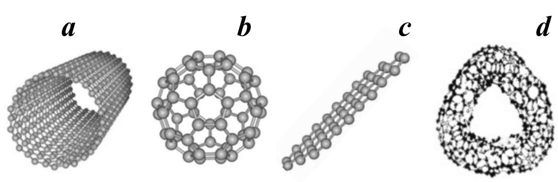

Fig. 2. Schematic representation of the open-ended carbon nanotubes (*a*), fullerene C_{60} (*b*), carbon nanosheet (*c*) and asrtalen (*d*)

Depending on the number of layers, carbon nanotubes (Fig. 2a) are divided into single-walled and multi-walled. The diameter of single-walled carbon nanotubes is in the range from 0.5 nm to a few nm and length from several hundred nanometers to hundreds of microns [15, 16]. Multi-walled carbon nanotubes are also characterized by a diameter and length. Diameter of multi-walled nanotube ranges from a few to hundreds of nanometers, and in length – from several tens of nanometers to a several micrometers [15, 16, 17]. In contrast to nanotubes, fullerene C_{60} fullerene has spherically symmetrical shape and it is characterized by in diameter, meaning of the diameter is about 7 Å [18]. Fullerene C_{70} has not symmetrical shape, so for its characterization two axles are introduced, length of shorter is about 0.712 nm and length of longer - 0.796 nm [19, 20]. It should be noted that in several other works another values are given: 0.72 nm and 0.85 nm [21], or 0.691 nm and 0.792 nm [22]. Astralen is a carbon nanoparticle, which geometric shape is toroid. Astralen is characterized by an outer diameter and pore size. The outer diameter of astralen nanoparticle is in the range from 10 to 150 nm, and pore size from 20 to 60 nm [23, 24]. Carbon nanosheets are characterized by the length of edge, the edge is in the range from 40 nm to several micrometers [25]. Note that in works [26] following data is presented: 1.75 microns and 13 microns, respectively. Thickness of nanosheets typically is more that 1 nm. But we know that nanosheet thickness can vary from a few nanometers to hundreds of nanometers [26].

To evaluate the surface area of nanoparticles with different shapes we proposed a model. The model consists of the description of nanoparticle by geometrical figure that has similar geometric characteristics, and following calculation of the surface area of geometric figure. Note that application of our model does not require consideration of the hexagonal structure of the graphen surface. Also, the model takes into account only the outer surface of nanoparticles. The proposed model can simplify the calculation of the surface area of nanoparticles in comparison to the model described in work [1]. The model proposed in work [1] requires information not only about the nanoparticle weight and number of atom, but also about the molecular structure of nanoparticle hexagonal surface element. Moreover, our model can reduce the number of steps for evaluation carbon nanoparticle surface area; this is reducing calculation time while modeling properties of nanoparticles.

a) Surface area of carbon nanotubes

When calculating the surface area of single-walled carbon nanotube, the nanotube is described by cylinder,

the cylinder height is equal to the nanotube length, and cylinder diameter is equal to the nanotubes diameter. We assume that nanotube "closed", this means that we do not take into account its inner surface. So, the formula for calculation of the nanotube surface area is:

$$S=\pi dl \quad (1)$$

where S - surface area of nanotube, d - its diameter, l - the length of the nanotube.

To calculate the S, we need to know the weight of the carbon nanotube. To calculate the number of carbon atoms that form nanotube, we used the software Nanotube Modeler. Then, the formula for calculation of specific surface area of nanotubes takes the form:

$$SSA = \frac{S}{Nm_C}, \quad (2)$$

where SSA - specific surface area of nanotube, S - its surface area, N - number of C atoms that form nanotube; m_C=1,993*10^{-26} kg – weight of one C atom. When calculating the surface area of multi-walled nanotubes, we applied a similar approach. Specifically, the nanotube is described by cylinder, cylinder height is the length of the nanotube, the diameter of the cylinder - is the outer diameter of the nanotube. The formula for calculation of surface area is similar to the formula (1). To check our model, we calculated specific surface area of single-walled and multi-walled (five layers) nanotubes, taking as a basis the average experimental data on geometrical parameters of carbon nanotubes [15, 17, 10, 17]. The average length of single-walled carbon nanotubes is equal to 30 mm and a diameter is 1.15 nm. For multi-walled nanotubes: the average length is equal to 320 nm and a diameter is 25.4 nm. Calculations using formulas (1) and (2) shows that the surface area of a SWSNT is equal to 4.3332*10^{-13} m^2 or SSA=1303 m^2/g. The obtained data is in agreement with the result (1315 m^2/g) of calculations based on the model described in work [1]. However, the values are too high in comparison with experimental data (285 m^2/g [16]; 400 m^2/g [27]), we explain it by the presence of defects in the nanotubes after synthesis and purification. For MWSNT with parameters, which are equal to averaged value, surface area is equal to S=5,1*10-14 m^2, or MWSNT with 5 layers, SSA = 649 m^2/g. For comparison, the experimental data is 50 m^2/g [16], 10-500 m^2/g [28], 40-300 m^2/g [29]. Note that the value of specific surface area of MWCNT depends on the number of layers which are forming nanoparticle. Increasing the number of layers results in increased weight of the nanoparticle, weight is inversely proportional to the specific surface area, see formula (2), while a value of surface area depends only on parameters of outer layer. In other words, the data calculated by our model is in agreement with the literature data.

Using the proposed approach, we calculated the total surface area of the material formed by single-walled and multi-walled carbon nanotubes (Fig 1).

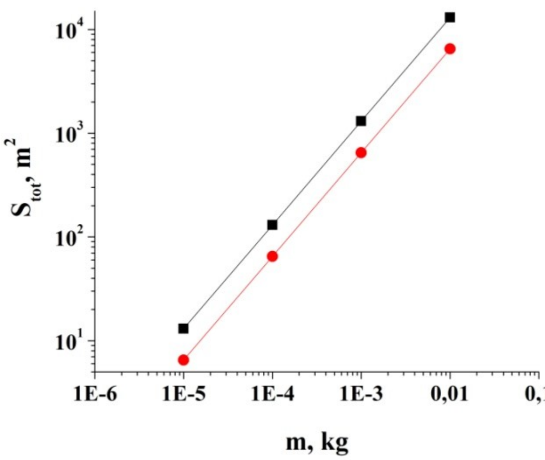

Fig.1. Total surface area of single-walled (■) and multi-walled (●) carbon nanotubes, based on proposed model.

From Fig. 1 can be seen that the surface area of single-walled carbon nanotube is greater than surface area a multiwall nanotubes, with averaged parameters. By increasing the weight of material based on carbon nanotubes, the effective surface area of the material increases linearly.

b) Surface area of fullerenes

To calculate the surface area of fullerene C_{60}, we describe this carbon nanoparticles by a sphere. The diameter of the sphere coincides with the diameter of fullerene. The formula for the calculation of surface area of fullerene is:

$$S=\pi d^2, \quad (3)$$

where S – fullerene surface area, d – fullerene diameter. To calculate the specific surface area of fullerene-based material we used formula (2), where the value of N is defined as the number of C atoms in fullerene (for C_{60} - 60 atoms, and for C_{70} - 70 atoms). The average diameter of C_{60} is 6.83 Å, and C_{70} is 7.59 Å. The surface area of a fullerene C_{60} is equal to 1.46*10^{-22} m^2 or same, SSA=1224 m^2/g. The resulting

value of SSA is greater than the experimental data (5.66-6.44 m^2/g [23]), we explain it by the presence of impurities during the synthesis of fullerene nanoparticles. Surface area of C_{70} is equal to 1.80*10^{-22} m^2 or SSA=1296 m^2/g. Using this model, also was calculated SSA of material formed by nanoparticles C_{60} and C_{70} (Fig. 2).

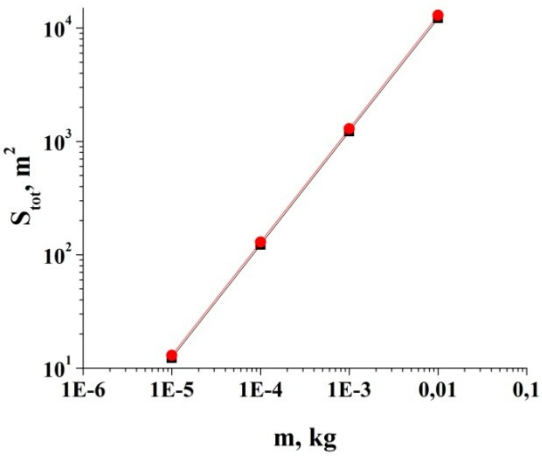

Fig. 2. Total surface area of fullerene C_{60} (■) and C_{70} (●)based on proposed model.

The total surface area of fullerene C_{70} is larger than for a fullerene C_{60}, we explain it by the different size of fullerenes. By increasing weight of material, surface area of material on the base of fullerenes increases linearly.

c) Surface area of astralene

For astralen nanoparticles is used model in which astralen described by geometrical figure - torus. We know that astralen characterized by two parameters: D - outer diameter, and d – pore size. Torus also is described by two parameters, R - radius of large circle of the torus, r - radius of the small circle of the torus. The relationship between these parameters of torus and astralen is: , $D=\frac{d}{2}+r$. Then, surface area of astralen is:

$$S_{astr}=4\pi^2 rR=4\pi^2\frac{D-d}{4}\left(\frac{d}{2}+\frac{D-d}{4}\right)=\frac{\pi^2}{4}(D^2-d^2) \qquad (4)$$

Weight of nanoparticles was determined using software Nanotube Modeler. Due to the geometry of the astralen nanoparticle, we assumed that the number of atoms in astralen is equal to the number of atoms in a single-walled nanotube with 2πR length and radius r. Calculation of the specific surface area is held by the formula (2).

The average outer diameter is 82 nm, diameter of pore – 40 nm. Then, a surface area of astralen nanoparticle is 1.3*10^{-14} m^2 or SSA=1307 m^2/g. For another type of astralen nanoparticles with greater weight, described in work [31], the value of SSA=40 m^2/g, wich is in good agreement with experimental data– 20.55-38.3 m^2/g [23]. Using the proposed approach, the total surface area of the material formed by astralen nanoparticles also was calculated (Fig. 3).

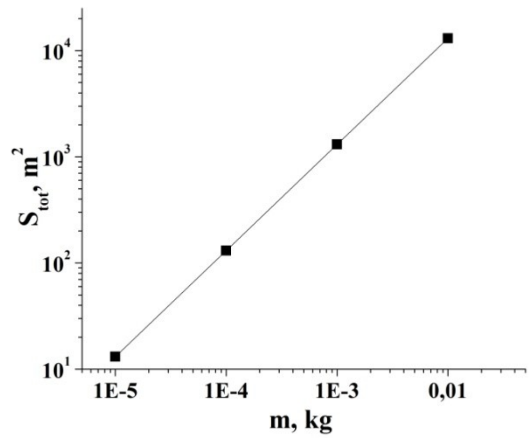

Fig.3. Total surface area of astralens based on proposed model.

By increasing the weight of of the material its effective area increases linearly.

d) Nanosheets

When calculating the surface area of nanosheet, we describe it by a rectangle. Nanosheet is characterized by a length and a width; these values in the proposed model are equal to length and width of the rectangle. The surface area of a rectangle is "S = a*b", where, a - the length of nanosheet, b - width of nanosheet. Number of C atoms in nanosheet was determined using software Nanotube Modeler. Average length and width of nanosheet is 3.6 microns. Applying the formulas (1) and (2) we got value surface area 2.6 *10^{-11} m^2, and the value of the specific surface area 2557 m^2/g for nanosheet. For comparison, the experimental data is 466 m^2/g [13], other theoretical calculations is 2620 m^2/g [13]. We see that the results obtained by the proposed model are in agreement with other theoretical calculations, and they are larger than the experimental data, it is because theoretical calculations reflects the maximum possible value of

surface area of the nanoparticles. In addition, using this model, we calculated the total surface area of the material formed by nanosheets (Figure 4).

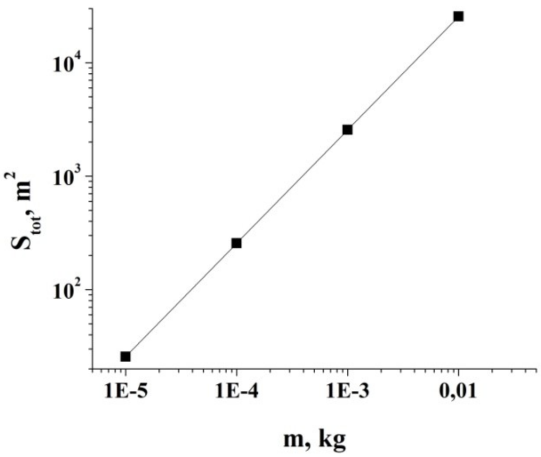

Fig.4. Total surface area of nanosheets based on proposed model.

From Fig. 4 it can be seen, that by the increasing weight of material formed by nanosheets, its surface area increases linearly.

To summarize the results, we compared the values of the specific surface area of different types of carbon nanoparticles (Fig. 5).

From Fig. 5, you can see that the greatest specific surface area have nanosheets. Fullerenes and single-walled carbon nanotubes have similar values of specific surface area. The value of the specific surface area for multi-walled nanotubes may vary depending on the number of layers, and it is less than the value of the specific surface area of single-walled nanotubes with similar characteristics. Usually most interesting for scientists are nanoparticles with the greatest specific surface area, but depending on the application, other parameters of nanoparticles must be taken into account; after determining the importance of all parameters, it can be determined what type of particles should be used.

For example, small particles (<100 nm) would offer a larger external surface, but they cannot be used in most of the biomedical applications due to difficulties of filtering them from the biofluids. Authors of work [29] investigated cytotoxity of carbon nanoparticles and concluded that cytotoxicity follows a sequence order on a mass basis: SWNTs>MWNT10>C_{60}. The measured room temperature thermal conductivity for an individual MWNT (>3000 W/mK) is greater than that of basal plane of graphite (2000 W/mK) [30].

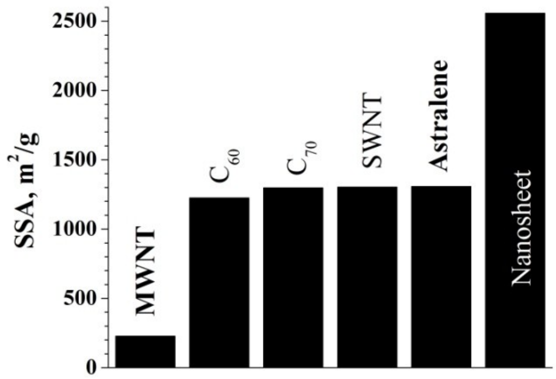

Fig.5. Specific surface area of different types of nanoparticles based on proposed model.

Conclusions

The model for calculating the specific surface area of carbon nanoparticles of different shapes and sizes was proposed. The model consists of comparison of a nanoparticle with geometric figure, and following calculation of the surface area of the figure. The model is based on several assumptions: 1. Nanoparticles can be described by geometric figure with symmetry and geometric parameters that are similar to the corresponding parameters of nanoparticles; 2. Only the outer surface of nanoparticles is taken into account; 3. The surface of the nanoparticles do not contain defects. Unlike the model proposed by the authors in work [1], our model does not require information about the surface structure of nanoparticles. Using the proposed model, was calculated specific surface area of carbon nanotubes, fullerenes C_{60} and C_{70}, astralens and a nanosheets. The results are in a good agreement with theoretical specific surface area calculated by other authors. However, the value of the specific surface area of nanoparticles obtained with the proposed model and the model described in [32] have inflated value compared with experimental data, we explain it by the presence of defects in the carbon nanoparticles as a result of treatment, and the presence of impurities. Thus, the proposed model allows us to calculate the maximum specific surface area of the carbon nanoparticles of different symmetry.

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