Hindawi
Adsorption Science & Technology
Volume 2022, Article ID 7387409, 9 pages
https://doi.org/10.1155/2022/7387409

SAGE Publishing

Research Article

Surface State Treatment of Carbon Dots Using Sulphur Dioxide Isotherm

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Received 20 January 2022; Revised 13 March 2022; Accepted 28 March 2022; Published 15 April 2022

Academic Editor: R Lakshmipathy

Low toxicity carbon dots are combating the disadvantages of quantum dots. The carbon dots find their applications in many fields due to their versatile nature. Four different types of carbon dots are present, according to the way of manufacturing and application the type is chosen. The water-soluble characteristics of carbon dots help them be involved in biomedicine applications. The optical properties of the carbon dots find applications as drug delivery, biosensors, LED, etc. The properties like fluorescence, photoluminescence, and phosphorescence are found in the carbon dots. The carbon dots occupy the tiny spot that exhibits different optical properties on excitation. The carbon dots excitation is mainly due to surface states. The characterization of surface states is very complex. The surface states contain the core structure of carbon and oxygen functional groups on the surfaces. The anions and cations formed from functional groups on excitation will recombine themselves. The functional groups are usually carboxyl and hydroxyl groups. The Π-collaborative network of the electronic structure contains many quantization levels which help the carbon dots to produce different wavelengths adapting to different applications. Due to the interference of the structure of the carbon dots, the entire property will vary. Doping of heteroatom methods is employed to enhance the fluorescence, and photoluminescence property carried out. They used N, S, P, and B heteroatoms singly and in combination to doping carbon dots. Here, the paper proposes the sulphur dioxide adsorption technique to enhance the optical properties of the carbon dots. The proposed method shows 8.5% efficiency in relative fluorescence intensity and 8% efficiency in terms of photoluminescence intensity.

1. Introduction

In recent years, electronic devices have been miniaturized without compromising their capacity. One such invention is quantum dots also called artificial atoms. These quantum dots are the size of an atom with zero dimensions [1]. The working principle of quantum dots is the same as that of the working principle of the electron in an atom. Quantum dots are a cluster of atoms made up of semiconductor materials of nanosized [2]. They are speckles that fill the single tiny spot. Quantum dots release energy as a light when it is exciting. The frequency of the light depends upon the amount of energy of the bandgap of the quantum dots [3]. The main disadvantage of the quantum dots is toxicity, an exponential decline of fluorescence and blinking happens, degradation inside the living cells, etc. [4]. Xu et al.

conducted the purification process of carbon nanotubes and discovered the carbon dots. Carbon nanotubes are formed by rolling the graphene sheets about the nanosize diameter. Carbon nanotubes replace the metallic wires with their great properties [5]. The main profit of using carbon nanotubes is that they are lightweight, temperature resistant, tensile, etc. The amorphous and carbonaceous impurities present in the carbon nanotubes degrade its performance; therefore, electrophoretic analysis is carried out in the single-walled carbon nanotubes produced by arc discharge soot [6]. During this purification, Xu et al. found two kinds of derivatives from the crude soot. One of the derivatives is carbon nanodots that exhibit fluorescence properties [7]. The carbon nanodots are tiny carbon nanoparticles. When it is excited, it produces photoluminescence. When these fluorescent carbon nanoparticles are separated into several components, they produce various lights with different wavelengths. The carbon dots produced from single-walled carbon nanotubes are only 1/10th of their mass. They produced fluorescent with different wavelengths and combinations of light [8]. A chemical sensor gives precise information about the quality and quantity of chemical species in its environment. Hence, the development of chemical sensors and their applications is an inevitable part of analytical chemistry [9]. The efficiency of a sensor depends on its selectivity and sensitivity of determination towards a particular analyte. Depending on the property to be determined, these chemical sensors are broadly classified into electrochemical, optical, thermal, and mass sensors [10]. The major areas in which the sensors have found application include medical diagnosis, determining the pollutants in the air, water, and soil, getting information about the industrial production process, and determining contaminants in food products [11].

This enhancement in the properties is greatly influenced by their size being in the quantum confinement regime. In the case of bulk semiconductors, the conduction electrons are free to move in the solid and form a continuous energy spectrum [12]. The size confined electrons instead of acing as a continuous energy band show discrete bands with bandgap. This happens when the particle size approaches the exciton Bohr radius [13]. Ya-Ping Sun et al. defined the term carbon dots for the fluorescent nanoparticles. They tried to improve the luminescent photo property of carbon dots by surface passivation instead of oxidation. They found that it produces different wavelengths according to the size of the carbon dots. For the high range of photoluminescence, carbon dots should have a large surface-to-volume ratio for the emissive energy traps for excitation [14]. Xin Wang et al. proposed using inorganic salts like ZnS to overcome the surface defects to increase the fluorescence of the nanotubes [15]. Yun Liu et al. proposed the esterification process on the surface of the carbon nanodots using the carboxylic groups which increased the photoluminescence by rigidity and increased coplanarity of the molecules [16]. Li Cao et al. studied the band gap's involvement in producing various carbon dots of different wavelengths. They also compared quantum dots and carbon dots in different aspects for various applications [17]. Junjun Liu et al. gave a detailed description of various applications of carbon dots currently and discussed its challenges [18]. Hui Ding et al. studied the surface properties of the carbon dots and discussed the surface state derived emissive properties of the carbon dots. The emissive property of the carbon dots is enhanced by doping it with nitrogen, sulphur, boron phosphorous, etc. The various doped carbon atoms are adapted for various applications in different fields [19]. Jayasmita Jana et al. treated the surface of the TiO_2 nanoparticles with sulphur and discussed the properties of the pretreated surface, Coad sorbates, etc. [20]. Jonas Baltrusaitis et al. analyzed the chemical and textural properties of sulphur dioxide adsorbed activated carbon surfaces using a thermogravimetric analysis system. Therefore, the phenomenon of carbon dots is mainly based on surface-emission [21]. The surface of the carbon dots contains some defects and impurities. So far from the literature survey, many authors have used doping to treat surfaces. This paper proposes a new method of introducing the adsorption technique for treating the surface of the carbon dots. The carbon dots are treated according to the surface defects present. The sulphur dioxide adsorption isotherm is used to treat the carbon dots' surface, which uses SO_2 as adsorbents to increase the photoluminescence property. By involving the adsorption technique, the surface density will increase and increase the number of energy trap levels. So that more excitation happens, which in turn increase the photoluminescence property. This paper proposes the novel method of using sulphur dioxide as adsorbate on the carbon dots adsorbent.

2. Carbon Dots

The carbon nanomaterials are partitioned into speckles of small artificial atoms that occupy zero-dimensional space. These kinds of tiny nanoparticles are called carbon dots [22]. These carbon dots have more or less the properties of quantum dots. The carbon dots are overcoming the counterfeit of the quantum dots with their substantial properties. The carbon dots are byproducts obtained during the electrophoretic purification of carbon nanotubes at first, and later many techniques arise for its manufacturing [23]. There are two methods involved in the manufacture of carbon dots: top-down process and bottom-up process. The techniques like electrochemical oxidation method, laser ablation method, supported synthesis method, and combustion/thermal MWA heating methods are employed in the preparation of carbon dots in a top-down approach. The above techniques are difficult and costly too. The other techniques like solvothermal, hydrothermal, and MWA heating methods come under a bottom-up approach, which is quite easy and cost-effective. Usually, natural resources are used to manufacture carbon dots which involves easy processes. The different colours can be obtained from natural sources like bananas for green luminescence and watermelon peel for high blue luminescence. The carbon dots are water soluble applied in natural areas [24].

The carbon dots are nearly 10 nm-sized and have a height of 0.5 to 5 nm. The size of the carbon dots varies according to the process involved in manufacturing and preparation type. According to the interlayer spacing, the fringe spacing of the carbon dots is approximately 0.34 nm. The carbon dots contain many functional groups, mainly oxygen-related groups like carboxyl and hydroxyl. These carboxyl and hydroxyl are water-soluble and appropriate for chemical reactions, so carbon dots are mainly used in bio-applications [25].

The carbon nanotubes are obtained from single graphene sheets. The carbon dots are derivative of the carbon nanotubes. But the carbon nanotube does not have the photoluminescence property like carbon dots. This is because of the absence of energy gaps. The phenomenon of photo elimination is that when an electron excites from the valence band to the conduction band, it emits some energy as light. These light energy wavelengths will differ according to the size of the bandgap [26]. So if the bandgap is absent, photoluminescence will not be present. So the carbon nanotubes require multiple energy band gaps which need to be excited. Therefore, by cutting the carbon nanotubes into multiple pieces, there formed the tiny particles that contained band gap and other methods involved in which sp^2 islands are formed in the graphene sheet itself. There are two ways in which the carbon dots are obtained. They are the top-down approach and the bottom-up approach. In the top-down method, the carbon nanotubes are converted into speckles of nanoparticles. The bottom-up approach produces the carbon dots from small carbon units with some energy. Figure 1 shows different methods involved in manufacturing [27]. The top-down approaches are arc discharge, laser ablation, acidic oxidation, etc. The bottom-up approaches involve combustion routes, microwave pyrolysis, hydrothermal synthesis, and electrochemistry. The bottom-up approach is quite easy compared to a top-down approach. In the top-down approach, the carbon nanotubes need to cut into pieces or islands formed in the graphene sheets. The process will result in a quenching effect [28]. The carbon dots exhibited two photoluminescence emissions; they are discussed as follows.

2.1. Bandgap Fluorescence. Here, the photoluminescence takes place due to different energy traps present. Firstly the sp^2 islands are made in the Π-domain in the graphene sheets. Later, these islands are cut into pieces which form carbon dots. This type yields less emissive compared to the defects derived method. It is quite easy to obtain, but it is the main disadvantage of interlayer/interisland quenching effects. The photons at a particular wavelength cannot be produced when it happens.

2.2. Defect Derived Origins. The photoluminescence of defect derived origins is brighter comparatively. Here, photoluminescence happens because the defects occurred mainly due to sp^2 and sp^3. The carboxyl groups will combine with defect sites atoms for its process. Here, the bandgap is absent, and the system is complex. The characterization of the surface state needs to be done. The proper formulation and properties need to be studied [29]. Depending on the properties, surface functional groups, and structure, the carbon dots are derived into four categories:

(i) Graphene quantum dots

In this type, the graphene sheets are divided into well-defined lattices with functional groups on the edges with edge effects and quantum confinement effects. They are elongated horizontally than vertically.

(ii) Carbon quantum dots

In this type, the core is crystal lattices, and the surfaces contain chemical groups. They are quasi-spherical. Here, luminescence depends on intrinsic states and photoluminescence depends on size of the particles. By tuning the particles, different wavelengths are produced.

(iii) Carbon nanodots

The photoluminescence mainly happens due to defects present on the surface. It contains central core carbon structure and functional groups on the surfaces. There are no proper lattices, functional groups, or structures in this type. Figure 2 shows the types of carbon dots.

(iv) Carbonized polymer dots

In this type, the polymer chain on the surface is aggregated or crosslinked. The carbon core is divided into four categories: (i) CND or CQD, (ii) a crystalline carbon structure, (iii) a highly dehydrated crosslinking polymer structure, (iv) a close-knit frame polymer structure.

The main properties of carbon dots are low toxicity, high photostability, good photoluminescence, appreciable fluorescence, high crystallization, substantial dispersibility, large functional groups, good catalytic properties, multicolour emissions, feasible large-scale productions, good biocompatibility, cost-effective, eco-friendly, cytotoxicity, cytocompatibility, etc. The main advantage of carbon dots is focused on their optical properties. Three important factors come under optical properties [30].

2.3. Absorption. The absorption property varies according to the source of carbon used and its manufacturing types. They are mainly affected by surface groups, cores containing oxygen or nitrogen groups, and the Π-domain islands [31].

2.4. Photoluminescence. This behaviour can be promoted by diverse synthetic methods and pure carbon sources. It mainly depends on the carbon sources used, post passivation method, and synthetic approaches. It is measured with the help of the QY value.

2.5. Phosphorescence. The room temperature phosphorescence is the key feature of carbon dots manufactured by two methods: intersystem crossing and radiative transition. This can be enhanced by proper designing, post-treatment, and appropriating the chemical reactions.

The tuning of carbon dots is required to enhance the emissive property of the surface. Therefore, the doping method introduces the heteroatoms into the carbon dots, which stabilize the functional groups formed due to the surface passivation method. There are two ways by which

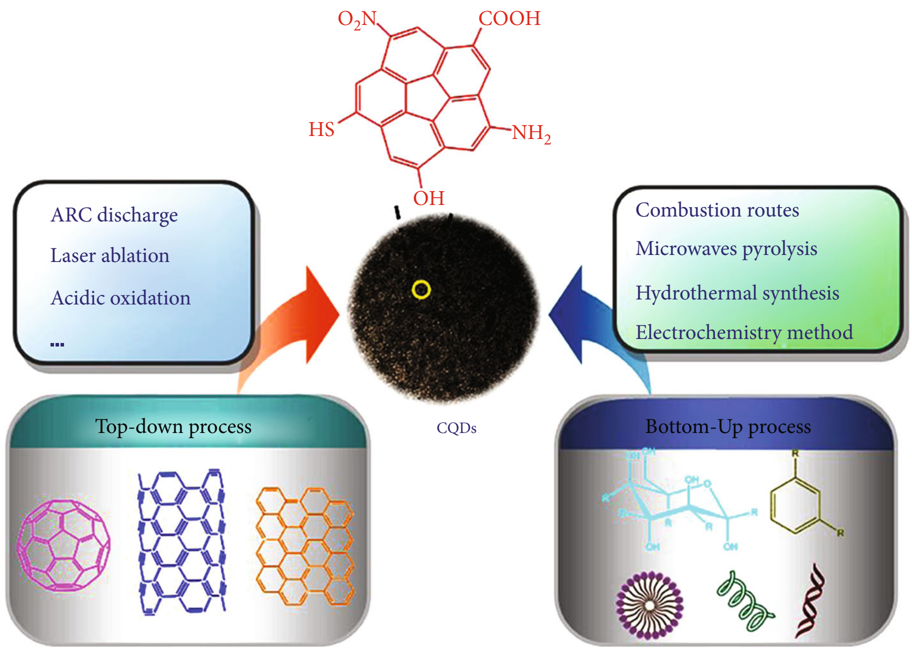

Figure 1: Types of preparation of carbon quantum dots.

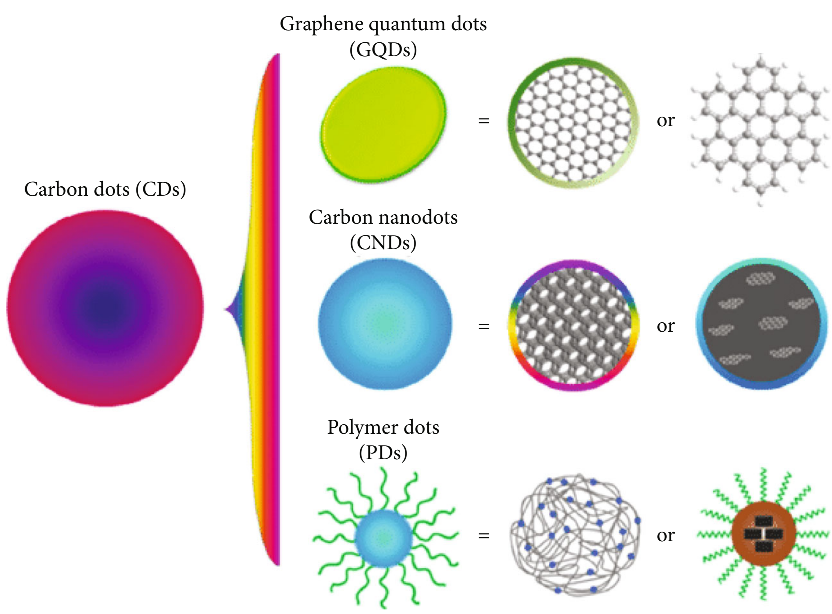

Figure 2: Types of carbon dots.

heteroatoms are introduced: single heteroatom doping and more than two different heteroatoms doped. The heteroatoms used for doping are nitrogen, sulphur, phosphorous, and boron. Among all the dopants, nitrogen-based heteroatoms showed very impressive results [32]. The nitrogen-doped carbon dots are finding their applications in photocatalysis, cell imaging, etc.; the sulphur doped carbon atoms help to increase the high blue colour excitation. It is mainly used for the particular colour tunning process. It finds the main application of ammonia sensing. The P dopant forms surface defects in carbon dots, especially diamond sp^3 surfaces. The B dopant is not widely used, and it has four different categories of processing [33].

The major application of carbon dots is drug delivery, CTV cameras, surveillance, invigilance, supercapacitors, energy storage, catalysis, photovoltaics, light-emitting diodes, sensors, biosensors, biomedicine applications, cell imaging, solar cells, cancer cell imaging, tumour findings, etc.

(i) *Optical applications*: The tiny size, specific surface area, and functional groups make carbon dots more reactive and sensitive, which applies to analyses of the catalytic used. It is used to find cancer cells by imaging and targeting, which is different from normal cells [34].

(ii) *Storage applications are* used in rechargeable batteries by improving the electrochemical process. The cations and anions from functional groups help to store charges in nanoparticles. They are used as solar cells with the help of Π-Π interactions between electronic structures. They are used as supercapacitors with flexible and stable multielectronic storage devices. The large specific area, cross-linked structure, and tiny size help store energy at different levels [35].

(iii) *Bioimaging applications*: Its main application is cell imaging and drug delivery. Various studies are already made on different types of carbon dots and their manufacturing processes. The successful finding outs are carried out at the liver, central nervous system, investigating tumours, etc.; the future study of the carbon dots will help in more biomedical applications [36].

3. Experimental Procedure

The literature shows that the carbon dot's photoluminescence property is mainly due to the surface defects/surface states. But the characterization of the surface state of carbon dots is very difficult because they contain an uneven distribution of functional groups and carbon cores. Many spectroscopic studies like auger electron spectrum, X-ray photoelectron spectroscopy, ultraviolet photoelectron spectrometer, and Raman scattering mechanism are made to characterize the surface states. The luminescence property of the carbon dots is mainly surface layer chemical composition, valence band of the surface state, and surface adsorbent for charge transfer [37]. The key role of surface adsorbent is to transfer the charges excited and illuminate as photons. But due to some uneven distribution and complex characterization of the structures, the adsorbent efficiency is decreased. In turn, it reduces the intensity of the photoluminescence. ASTM E2180 – 18 standards used for the testing for the surface state treatment.

The surface defects on the carbon dots produce photoluminescence of various wavelengths. The wavelengths also need to be in narrow bandwidth to be used in different applications. The intensity of the photons released should be very high for the narrow bandwidth requirement to differentiate it from other narrowband wavelengths. The carriers generated after excitation will be present on the surface of the carbon dots. The ions formed by the functional groups recombine with the carbon cores. But some number of ions will be left without recombination due to the uneven distribution of the surface state. These ions will degrade the performance of the carbon dots. The narrow bandwidth cannot be obtained because of the ions having photoluminescence of various wavelengths. So that wavelengths of a particular colour will not be produced, limiting the application of carbon dots.

Table 1: Simulation parameters.

Simulation parameters	Value
Carbon dots diameter	10 nm
Height	1 nm
Fringe spacing	0.34 nm
Energy of photon	5.7×10^{-19} J
Wavelength	350-400 nm

This paper proposes the novel method of using sulphur dioxide as adsorbate on the carbon dots adsorbent. The adsorbates will distribute on the surface of the carbon dots adsorbents. During the recombination process, the adsorbents have absorbed some of the left uncombined ions when transferring the charges. The adsorbent will themselves produce many energies trap levels when excitation produces high intensities, which is the main requirement for narrow bandwidth. From the literature, it is seen that sulphur acts as a dopant for enhancing the performance of the carbon dots [38]. Here, the paper proposes to use sulphur dioxide as adsorbates that contain oxygen functional groups, which will combat the effects of oxygen functional groups on the adsorbent surface of the carbon dots.

4. Results and Discussions

The simulation of the proposed method is carried out with the help of MATLAB 2021b. A comparison of proposed and existing carbon dots is represented in the graph. The simulation is concentrated mainly at 350-400 nm wavelength. The proposed method is studied only for that particular wavelength [39]. The resultant solution showed reddish-yellow fluorescence. The obtained C. dots were then separated by silica column chromatography, and the fractions showed a gradient of colours from blue to red. All these fractions showed optical uniformity in similar particle size distribution and excitation independent emission spectra. The only factor they differed was the extent of surface oxidation. Hence, they concluded that red-shifted emission was due to incorporating more oxygen species [40]. However, with the deteriorating condition of the environment, the concern for pollution issues has increased nowadays. Metals from various industries contribute extensively towards pollution, which causes adverse effects to the atmosphere and aquatic ecosystem. Besides the fact that many of these metals are important for organisms in small doses, an increase adversely affects human health and the ecosystem.

The simulation is carried out considering the size of carbon dots of 10 nm with a 1 nm height range. The fringe spacing is also considered, which is approximately 0.34 nm. The carbon dots have the surface states, which is considered Poisson distribution for simulation purposes [41]. The

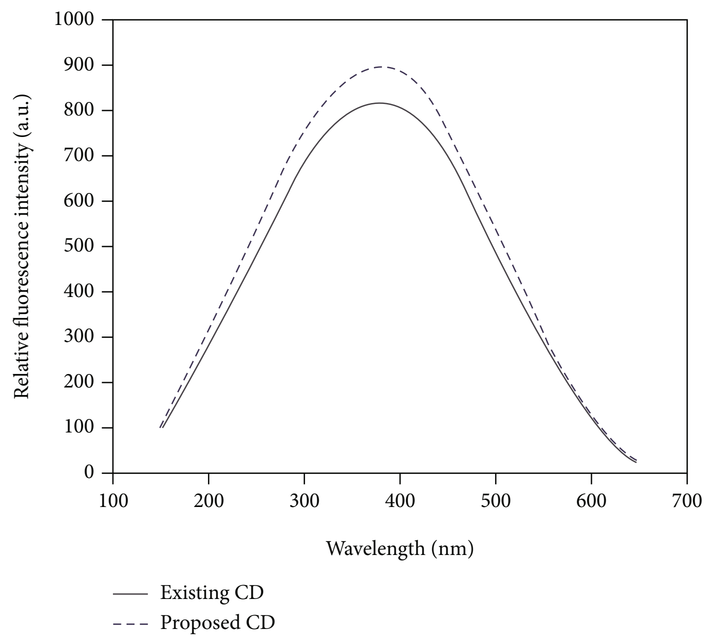

FIGURE 3: Plot of wavelength vs relative fluorescence intensity.

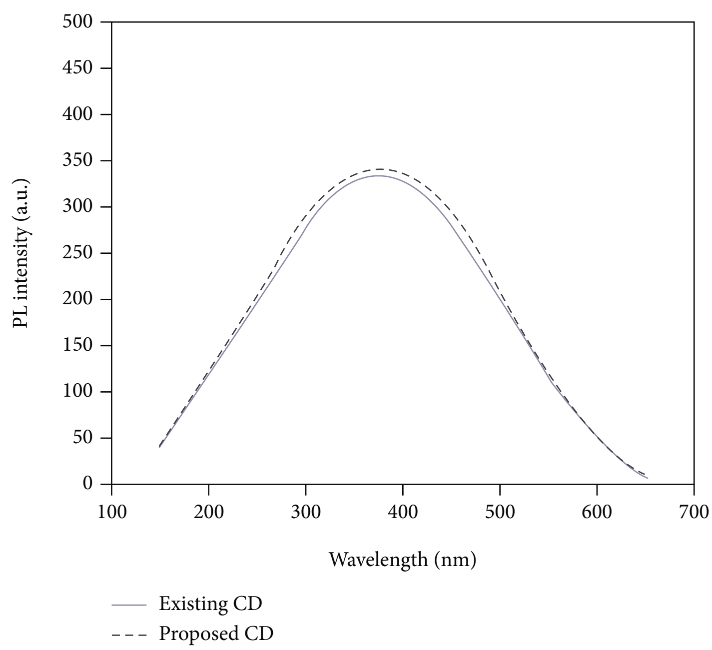

FIGURE 4: Wavelength vs PL intensity.

sulphur dioxide adsorbate is considered to be randomly distributed on the adsorbent surface of the carbon dots. The energy of photons of wavelength is usually $5.7 \times 10\text{-}19$ J. The simulation is carried out using the values present in Table 1.

Figure 3 shows the plot of wavelength in nanometer versus relative fluorescence intensity in the Angstrom Unit. The plot has the highest peak at 375 nm and has a relative fluorescence intensity of 826 a.u. for the existing carbon dots. After applying the sulphur dioxide adsorption technique for the same peak at 375 nm, the relative fluorescence intensity is measured as 903 a.u. The relative fluorescence intensity increased in the proposed method due to the extra energy trap levels created by the adsorbates on the surface of the carbon dots adsorbents [42]. On analyzing the results, the proposed method yields an 8.5% increase in the relative fluorescence compared to the normal carbon dots [43].

It was observed that when the composite materials are fabricated from two chemical species, they exhibit new properties rather than of the individual materials. C. dots have strong electron-donating property. Researchers are intensively applying this property of C. dots to reduce metal salts into metal nanoparticles. Literature shows the formation of C. dot metal nanocomposite for various applications such as optical sensing, electrochemical sensing, and catalysis. Imaging in scientific terms is described as getting a 2-dimensional picture of the specimen under study. Due to the high versatility, selectivity, and sensitivity of fluorescence-based imaging techniques, they are gaining particular interest than the other imaging methods.

In fluorescence imaging, two types of imaging are possible. The first one involves the imaging of chemical species that are intrinsically fluorescent, and the second one involves the imaging of cells and tissues by external fluorescent materials like fluorescent labels or fluorescent nanomaterials. This process of obtaining biological matters images by treating them with fluorescent labels or nanomaterials is known as bioimaging. The internalization of C. dots by cells is possible due to the smaller size of C. dots, thereby finding application as a good bioimaging agent.

This fascinating property allows carbon to form a wide range of structures with different fundamental properties. It can form allotropes where the physical and chemical properties are determined by the structural geometry of atoms and the chemical binding within the molecules. Hard diamond and soft graphite are classic examples of allotropes of carbon. The fluorescence of C. dots is considered its most fascinating property, which includes both excitation dependent and independent emissions depending on intrinsic structural features and surface chemical groups. Another interesting feature of C. dots is its tunable emission. Tunability indicates mainly the shift or change in emission parameters such as emission wavelength, fluorescence Q. Y, and lifetime, with precursors and synthetic conditions.

.Figure 4 shows the wavelength in nm versus the photoluminescence intensity in the Angstrom Unit of the proposed and existing method. The peak wavelength obtained is 357 nm [44]. The peak photoluminescence obtained for existing carbon dots is 338 a.u. For the proposed method, 346 a.u. is obtained. The proposed method has high photoluminescence intensity than the existing method [45]. Size dependency, surface states, surface defects, or surface oxidation can be explained as the possible reason for this. Functionalization refers to the modification of the surface of nanomaterials with various functional groups, which can act as a potential reactive site for the incorporation of specific organic, polymeric, inorganic, or biological functionalities. This may provide favourable attributes like dispersibility, better nanomaterial interaction with polymeric groups, biomolecules, and several organic or inorganic precursors.

Functional modifications of C. dots can be achieved by coating the surface of C. dots with functional groups like carboxyl and amine groups. This functionalization benefits the C. dots to be used in diverse fields like biomedical sciences, catalysis, nanoelectronics, sensors, batteries, and supercapacitors. In the proposed method, the sulphur dioxide adsorbate on the adsorbent surface of the carbon dots is randomly distributed. The distributed adsorbate combats the effects of function group's effects on the surface [46]. The adsorbate in turn will get excited and produces more electron carriers which on recombination emits the photons. These added photons will increase the photoluminescence intensity level. It is analyzed that the proposed method yields 8% increase in the photoluminescence level.

5. Conclusion

The proposal of using adsorption technique in carbon dots is carried out using sulphur dioxide isotherm. The sulphur dioxide acted as adsorbate on the adsorbent surface of the carbon dots. The adsorbate helps increase the energy trap levels and improve the electron carrier's excitation. The proposed method is 8.5% efficient compared to the normal carbon dots in terms of relative fluorescence intensity property. Considering the photoluminescence intensity property, the proposed adsorption technique increases 8% compared to the existing one. The characterization of the surface state is complex in carbon dots. By properly studying the surface states, even more enhanced carbon dots can be manufactured. The paper can be further carried out by considering different wavelengths and their various applications.

Data Availability

The data used to support the findings of this study are included within the article. Should further data or information be required, these are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

Acknowledgments

The authors thank Saveetha School of Engineering, SIMATS, Chennai for the technical assistance. The authors appreciate the supports from Ambo University, Ethiopia.

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