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## Carbon dots for food packaging applications

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Nanotechnology in food packaging has emerged as a viable commercial option. In particular, nanoparticles are being included in packaging materials to enhance the storage life and ensure the safety of food. Carbon dots offer several desired physical and chemical qualities, including their nanosize, numerous surface functional groups, nontoxicity, outstanding biocompatibility, excellent antibacterial and antioxidant activities, low cost, and ease of synthesis. Because of these reasons, carbon dots have the potential to be used in a wide variety of applications in the food packaging industry. Herein, the carbon dots for food packaging industries have been reviewed. The fabrication of carbon dots from microbes, plants, animals, and food items, the effect of carbon dots on engineering properties of polymeric films, and carbon dots as active antioxidant and antimicrobial agents are all discussed. In the end, challenges and future recommendations on carbon dots for food packaging are discussed.

### 1. Introduction

Food packaging has seen increased growth in recent years from users with hectic lifestyles who want food to be convenient and easy to consume.<sup>1</sup> Over the past few years, marketing and distribution network involvement in package innovation and design has risen dramatically. Compared to the price of the spoiled products, food waste can cause a much higher economic penalty.<sup>2</sup> In developed economies, consumer demand for pre-packaged food is still rising, and developing countries are also changing their lifestyle. Packaging helps to ensure the long-term viability of environmental resources by reducing

product deterioration and waste and protecting products until they have fulfilled their purpose. An optimum level of packaging is essential to decrease waste across the distribution chain and reduce the overall cost of the product.<sup>3,4</sup>

Biopolymer-based packaging is widely used due to its non-toxicity, good biocompatibility, and environmentally-friendly nature. Due to external factors, nearly a third of the food produced worldwide is wasted annually. Consequently, to resolve this issue, people have actively sought to retain good-quality food, use efficient techniques to increase the shelf-life, safety, and quality of the food, or embraced modern preservation and packaging technology. For food safety, one needs some functional or active materials that can slow the rate of food oxidation and stop microorganisms from growing in the food.<sup>5-7</sup>

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An active packaging film is a thin, shelf-standing film with functional qualities that is used throughout food packaging. Active coatings directly applied over the food surface are generally used to decrease bacterial growth (antimicrobial coating), reduce the rate of food oxidation (antioxidant coating), and limit enzymatic browning, moisture loss, and respiration of food products. These active coatings work as primary packaging while also enhancing the shelf life of fresh vegetables and fruits. Food can be coated by either dipping into the film-forming solutions or spraying with the solution. The functional coating can act as an ultraviolet and gas barrier (gas exchange between the fresh product and surroundings).<sup>8,9</sup>

The application of carbon dots in biochemical sensing, photocatalysis, optical technology, and biological imaging has been widely explored. Because of their advantageous properties, such as low cost, simple synthesis, high water solubility, bioactivity, less hazardous, and light absorption, they are frequently used in medical applications.<sup>10</sup> Nanomaterials based on carbon dots have gained popularity because of their superior electronic, optical, thermal, and mechanical characteristics (Fig. 1). There has been a rising demand for carbon dots (CDs) in food packaging, as carbon dots are considered to be active materials and can be used to form antioxidant or antimicrobial-based food packaging. Carbon dots (CDs) are synthesized from natural materials, and provide additional functions to food packaging, ultimately increasing the quality, freshness, and shelf life of food products.<sup>11</sup> A review by Qu *et al.* (2018) gives an idea for using carbon dots for food safety and preservation. Carbon dots can be easily synthesized from waste materials or eco-friendly substances such as peels, leaves, honey, and fruits. The functionality (strength and barrier) of packaging is improved by the use of these active materials, which are cheap and have great physical and chemical properties.<sup>12</sup> Researchers have recently used a variety of carbon dots from various sources to create active packaging materials that improve the shelf life of fresh produce.<sup>2</sup> The study has shown that enoki-mushroom-based carbon dots incorporated into gelatin/carrageenan-based

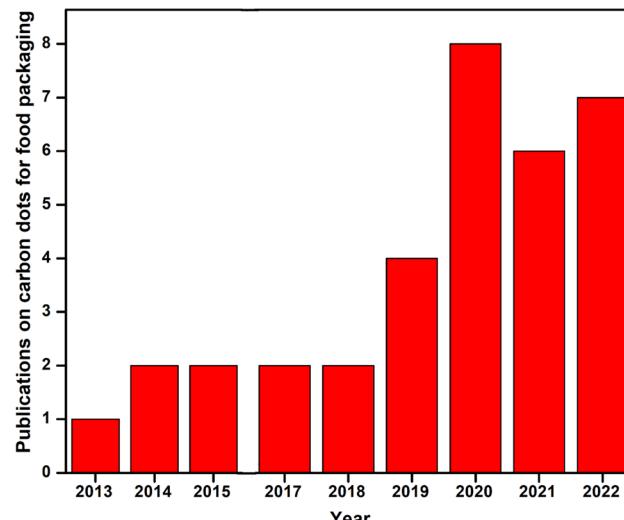


Fig. 1 Publication trends in oxygen scavenging films used in food packaging from 2013 to 2022. Data were collected from Scopus using the keywords "carbon dot" packaging" and "carbon dot food".

composite films enhance mechanical, antioxidant, and UV-blocking capabilities without compromising the film's transparency.<sup>3,8,13</sup>

This review provides information about carbon dots, the application of carbon dots in food packaging technology, and a brief introduction to the synthesis of carbon dots from microorganisms, plant, animal, and food items, followed by their engineering properties and applications as antioxidant, and antimicrobial agents. Furthermore, this review provides an overview of the ongoing studies and their application in food packaging. In the end, the challenges and future recommendations of carbon dots for food packaging applications are briefly discussed.



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## 2. Carbon dots: a new type of carbon-based nanomaterial

In recent days nanotechnology has been a thriving area in every field. Carbon dots are a type of carbon nanomaterial that was discovered through the purification of singular-layer carbon nanotubes.<sup>14</sup> Based on the dimensions, nanostructures can be classified into four categories: zero-dimensional (0-D), one-dimensional (1-D), two-dimensional (2-D), and three-dimensional (3-D) nanomaterials. Carbon dots are unique, discrete small particles and zero-dimensional semiconductors, smaller than 10 nm and completely spherical form of nanoparticles.<sup>15</sup> CDs have high-quality physical and chemical properties, such as low cost of the source, good tensile strength, ultraviolet barrier, good photostability, low toxicity, good biocompatibility, antioxidant, antimicrobial, small size, good quantum yield, abundant availability, and photoluminescence, giving opportunities in lots of applications in various fields. On the surface of carbon dots, there are naturally occurring or active functionalized groups, such as hydroxyl (−OH), carboxyl



(-COOH), and amino (-NH<sub>2</sub>), providing the chain formation reaction site or increasing the cross-linking properties in the chemical reaction.<sup>16,17</sup>

CDs' surface functional groupings help enhance the water solubility, the adsorption effect, chemical reactivity, and derivatives of some polymeric, biological, or organic materials. CDs are commonly classified into carbon quantum dots (CQDs), graphene quantum dots (GQDs), graphene oxide quantum dots (GOQDs), and carbon nanodots (NCDs). CD formation depends on the surface adjustment of carbon nanoparticles with polymeric and organic molecules. The chemical composition or the interior structure of the carbon dots decides which kind of carbon source is the primary material and it depends on the synthesis method.<sup>18</sup> Carbon dots extracted from non-biomass materials, such as polymer waste, liquid fuels, and battery discharge, are cytotoxic in nature and sometimes, after the addition of metal doping, change the properties of the whole matrix. These carbon dots are not suitable for food contact applications. Carbon dot surfaces can be altered by adding functional groups, such as amino, hydroxyl, carboxylic, and aldehyde groups, allowing them to interact with a wide range of molecules.<sup>19</sup> CDs can also be synthesized from eco-friendly substances. Some environmentally-friendly sources such as animal-origin food (honey, meat, butter, kinds of seafood), fruits/vegetables, and plant-origin (waste paper, fruit juice, tea residue waste, plant extract, plant seed) are the most popular carbon dot sources for food grade applications, and are described in Fig. 2. These food grade carbon dots are synthesized without using any harmful, and non-food-grade chemicals.<sup>20,21</sup>

### 3. Synthesis of carbon dots

Based on the source, CDs are synthesized in two ways. The first is the "top-down," and the second is the "bottom-up" approach. The top-down is a physical approach in which bulk nano-material breaks down into carbon dots using chemical or electrochemical and physical techniques. The top-down approaches

refer to chemical oxidation, arc discharge, laser ablation, and electrochemical methods.<sup>22</sup> Another aspect of the bottom-up approach is the synthesis of carbon dots from smaller carbon units into ultra-small units by using some energy, such as hydrothermal treatment, ultrasonic treatment, thermal decomposition, pyrolysis, carbonization, and microwave synthesis.<sup>23</sup> The surface properties can be optimized during the formation or synthesis process.

The bottom-up approach is more compatible because of reagent requirements, low cost, efficiency, and compatibility with the chemistry green world. Fig. 3 explains the various processes for synthesizing carbon dots.<sup>20</sup> For the synthesis of carbon dots, several synthetic (polymer waste, liquid fuels, batteries discharge) and natural compounds (biomass, micro-organism, plants) with carbon have been employed as precursors. In recent years, biomass has been extensively explored as a green source for generating food-grade carbon dots. Food-grade carbon dots can be synthesized from plants, food products, and microorganisms. It includes a wide range of organic substrates, including carbohydrates, proteins, alkaloids, and carotenoids. The hydrothermal and microwave methods are best suited for the synthesis of carbon dots. In the hydrothermal method, the degradation of biomass sources in the presence of water solvent at a certain temperature and pressure, and the range between water/sources is 5:1 to 75:1.<sup>24</sup> The chemical, physical, and also functional carbon dot characteristics are enhanced through the doping process with heteroatoms such as phosphorus (P), nitrogen (N<sub>2</sub>), fluorine (F), sulfur (S), copper (Cu), magnesium (Mg), titanium (Ti) and different combinations of atoms. Based on the type of material, 2-types of doping may be performed in carbon dots, *i.e.*, metal and non-metal doping, for example-nitrogen glucose-based carbon dots (NGCD) or N-functionalized carbon dots.<sup>25</sup> Carbon dots have various properties, such as unique structure, nontoxicity, tiny size, drug delivery, diagnosis, bioimaging, bio-labeling, photodynamic therapy, easy synthesis, high surface charge, environmentally friendly, which ultimately make them suitable for food packaging applications.<sup>26</sup>

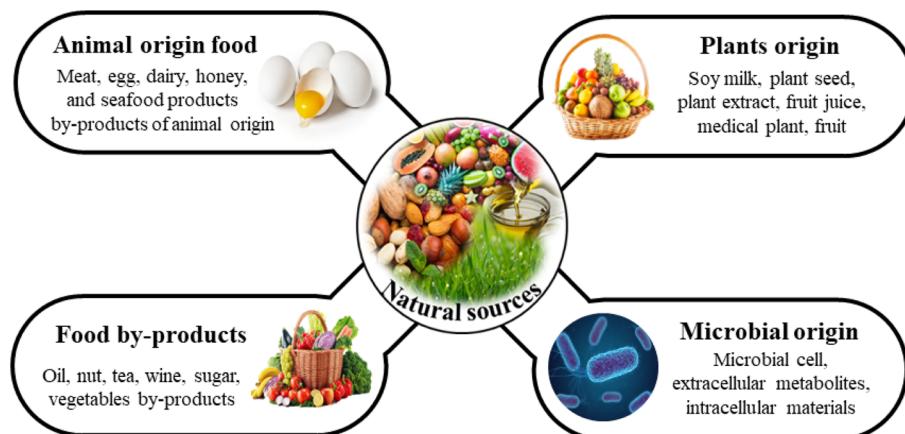


Fig. 2 Various sources, including animal food, human origin, plant and microorganism origin materials used for the fabrication of carbon dots for active food packaging applications.



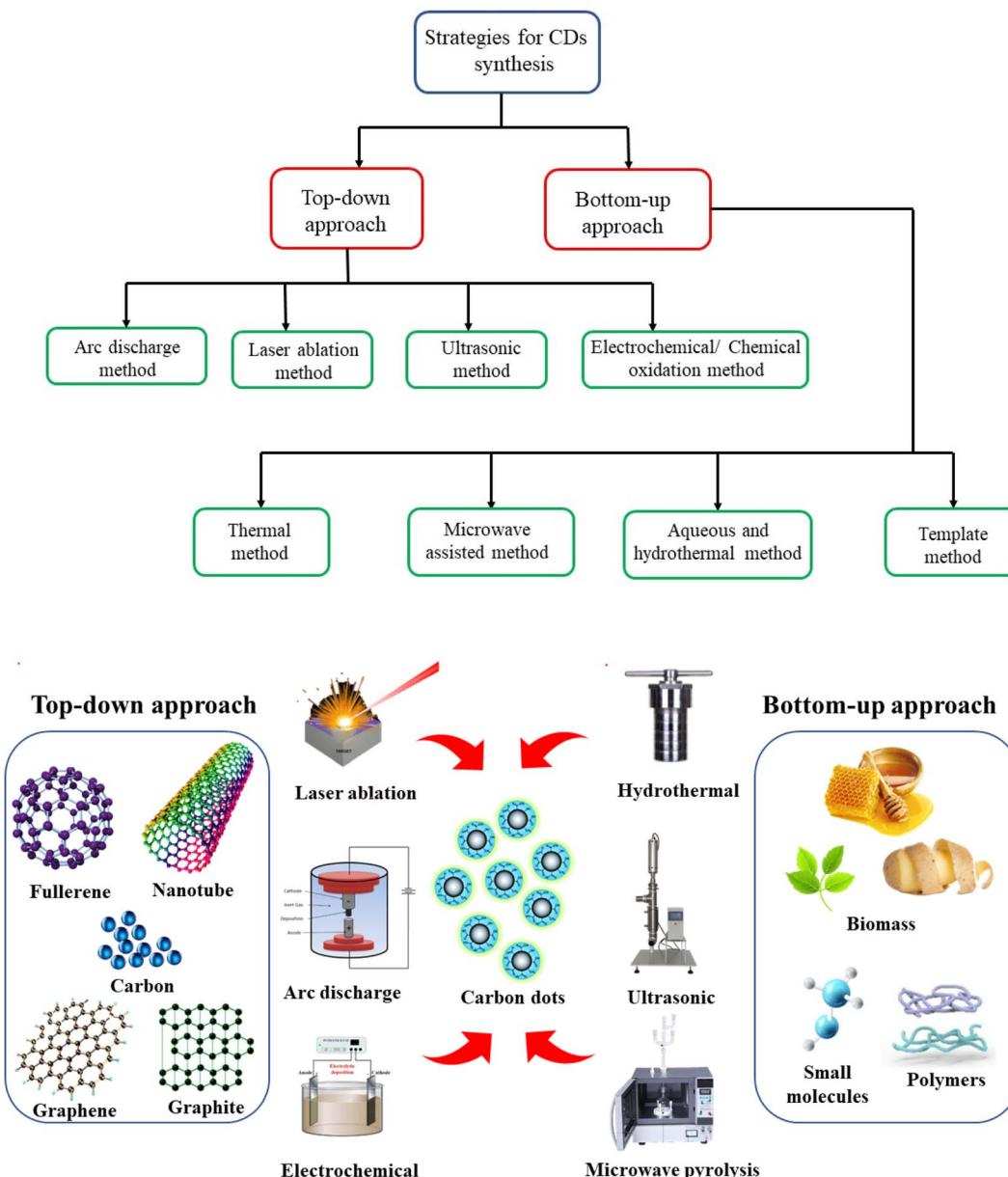


Fig. 3 Potential synthetic techniques top-down and bottom-up approaches for the production of carbon dots from various sources for food packaging applications.

#### 4. Carbon dots for packaging applications

Carbon dots play the most crucial role and have a diverse range of potential applications in packaging. They can help develop new biodegradable, anti-ultraviolet, antibacterial, anti-oxidation, and biocompatible films for food packaging to retain the quality and freshness and increase the shelf-life of the food. The food packaging industry has higher requirements for carbon dots to extend product storage life and safety.<sup>12</sup> The addition of carbon dots as active material enhances the physical, chemical, and mechanical properties of packaging without any effect on the remaining properties of the film and foods. It

is aimed to develop carbon dots as a new biodegradable packaging material with antimicrobial and antioxidant properties, which can be used to preserve and enhance the shelf-life of food.<sup>27</sup> Table 1 presents the application of carbon dots in developing active and intelligent food packaging systems.

Carbon dots have various applications and are used in food packaging that can enhance the properties of food products, such as quality and freshness.<sup>28</sup> Research has shown that adding carbon dots increases the film's light-blocking property without affecting water vapor permeability (WVP), mechanical properties, and water contact angle (WCA). The composite film generates reactive oxygen species (ROS), which have a strong antibacterial effect on bacteria, exhibit high antifungal activity,

Table 1 The application of carbon dots in developing active and intelligent food packaging systems and their effect on the composite matrix

Biopolymer	Carbon source	Effect on polymer matrix	Ref.
Gelatin	Potato peel	The gelatin/carbon dots film exhibits UV-barrier, antioxidant, and antimicrobial properties	29
Gelatin/chitosan	Glucose	The composite film generates reactive oxygen species, exhibits high antibacterial and UV-blocking properties without altering mechanical properties and water vapor permeability	30
Cellulose	Glucose	The film showed antibacterial activities, enhanced water contact angle, and UV-blocking properties without reducing the transparency of the film	25
Pectin/gelatin	Turmeric	The film showed intense antioxidant, antimicrobial activity and improved the UV-blocking function without significantly affecting the transparency of the film	31
Chitosan	Silk sericin	The bio-nanocomposite film has enhanced flexibility, antimicrobial, UV shielding, biocompatibility, and antioxidant properties	32
Carboxymethyl cellulose	Chitosan	The developed film exhibits enhanced antioxidant, antimicrobial, and mechanical properties	33
Polyvinyl alcohol	1,4 dihydroxyanthraquinone	The glyphosate film has carbon dots, and Cu <sup>2+</sup> ions have good innovative sensing properties	34
Polyvinyl alcohol	Banana paste	The developed film protects against UV radiation, and increases the shelf life of fried meatballs	35
Gelatin/carrageenan	Enoki mushroom	The enoki-mushroom-based active antioxidant film prevents food oxidation. The film also exhibits high UV barrier and strength properties	3
Chitosan/polyvinyl alcohol	Waste acorn cups	The developed composite film has enhanced the UV absorption capacity	36
Polypropylene	Cysteine	The film has fluorescent, antioxidant properties, which can extend the shelf life of food products	37
Bacterial nanocellulose	Lactic acid	The developed composite film enhances stretchability and flexibility, and also increases antimicrobial, photoluminescent, and ultraviolet-blocking properties	38
Polyvinyl alcohol	Waste tea residue	The developed films are transparent, flexible, and showed high mechanical strength, and enhanced UV blocking capabilities	13
Chitosan	Banana	The developed film has good microbial stability, and heat treatment for improving the shelf life of soy milk	39
Starch	Soy protein isolate	The developed composite film reduces water sensitivity and enhanced mechanical properties	40
Zein	Zinc acetate dihydrate	The film with quantum dots was effective against <i>S. aureus</i> and <i>E. coli</i> , and showed antimicrobial properties	41
Bacterial cellulose	Postbiotics of <i>L. acidophilus</i>	The developed film has good UV-blocking, antioxidant, and antimicrobial properties against <i>L. monocytogenes</i> and <i>E. coli</i>	38
Polyvinyl alcohol/nano cellulose composite	Cyanobacteria	The developed composite film has good flexibility, UV, and infrared blocking tendency, and anti-counterfeiting properties	42
Polypyrrole-chitosan/polypyrrole - chitosan-poly ethylenediamine	Citric acid and D,L-cysteine	The developed composite film has antimicrobial activity against <i>E. coli</i> and <i>S. aureus</i>	43
Chitosan coating	Kelp ( <i>Laminaria japonica</i> )	The developed coating decreases water mobility and effectively inhibits microorganisms which increases the quality as well as the shelf life of the food	44
Regenerated cellulose	Lactose	Nitrogen-doped films are highly transparent with enhanced mechanical properties	45
Collagen	Cotton cellulose	The composite thin films have high ionic strength and good UV barrier properties	46
Polyvinyl alcohol-cellulose nanofiber	The residue of radiata pine	The composite film has good UV barrier properties, mechanical properties, water resistance properties, and anti-counterfeiting	47
Poly methyl methacrylate	Carbon black pigment	The quantum carbon dots-based film has good UV-blocking properties	48
Nanocellulose- zinc oxide	Nanocellulose and 4,7,10-trioxa-1,13-tridecanediamine	Zinc oxide-based carbon dots composite film has good UV barrier properties and exhibits excellent thermal stability and photoluminescent stability	49
Polymethylmethacrylate	Cow milk	The cow milk-derived nanocomposite films have good antimicrobial and flexible properties	50

and increases tensile strength and the elastic modulus of the film. This review sum-up up all recent research on carbon dots in food packaging as active packaging, intelligent-sensor packaging, anti-counterfeiting, the addition of nanoparticles in

foods to increase shelf-life, and application in food preservation as a coating agent.

Carbon dots can be used in anti-counterfeiting applications for packaging. Anti-counterfeiting packaging is the practice of safe packaging that ensures product safety and prevents



replicas. Carbon dots have photoluminescent properties that can help detect counterfeiting. For instance, carbon dots that emit a specific fluorescence signal could be embedded in the packaging substance and observed by flashing fluorescent lights. Carbon dots may be embedded in the interior of the polymer matrix, or they may be coated explicitly onto the surfaces of the polymer matrices. It is more challenging for counterfeiters to imitate expensive products. These labels are handy for expensive goods that might be produced locally.<sup>12</sup> The application of carbon dots as additives, coating agents, antioxidant agents, and antimicrobial agents in food packaging as shown in Fig. 4 is self-explanatory because of some unique properties, such as good bio-compatibility, photoluminescence properties, and higher stability. As such, carbon dots are successfully applied to different fields such as active packaging, intelligent packaging, coating agent, and nano-level additives to enhance the food's shelf life. CDs have high functional properties, and thus they are more demanding in food packaging sectors.<sup>12</sup>

## 5. Effect of carbon dots on engineering properties of the polymeric film

Mechanical properties (tensile strength, elongation at break, modulus of elasticity), barriers (water barrier, gas barrier, and UV barrier), and thermal stability effectiveness of polymer films are fundamental characteristics of packaging materials.<sup>51</sup> This section highlights the thickness, mechanical properties, thermal properties, gas barrier, and morphological changes in polymer films. Table 2 describes the active and intelligent packaging system, which uses carbon dots made with natural

resources as carbon sources and their effect on the composite matrix.

### 5.1. Mechanical properties

Mechanical properties affect the film's behaviour during processing, preservation, transportation, and distribution until the consumer uses the food product. Packaging protects food products from various external mechanical loads during food supply chain processes.<sup>52</sup> Mechanical properties such as tensile strength (TS), elastic modulus (EM), and tensile strain of the film are increased by adding carbon dots in biodegradable polymers and forming composite films for food packaging.<sup>53</sup>

The use of thinner film packaging is made by improved mechanical performance, which lowers the cost and amount of the material used.<sup>12</sup> The addition of carbon dots in the gelatin/carrageenan polymer matrix creates a suitable composite film with excellent improvement in mechanical properties.<sup>3</sup> The tensile strength depends on the intermolecular interaction between the two biopolymers. The addition of enoki mushroom-based carbon dots in the gelatin/carrageenan film increases the tensile strength of the composite film because the interaction between the -OH group of carrageenan and the -COOH group of gelatin forms an ester bond, and the enhanced tensile strength was 61 MPa. Elastic modulus (EM) and elongation at break (EB) are also affected by the addition of enoki mushroom-based carbon dots (mCDs). The elongation at break and the thickness value without adding mCDs were 4.0% and 55  $\mu\text{m}$ , respectively, and after the addition of mCDs, the values increased by nearly 4.4% and 57  $\mu\text{m}$ , respectively. Similarly, the value of elastic modulus linearly decreases with an increased concentration of enoki mushroom-based carbon dots (mCDs).<sup>3</sup>

The addition of carbon dots derived from waste tea residue (WTR-CD) in the PVA matrix increases the mechanical

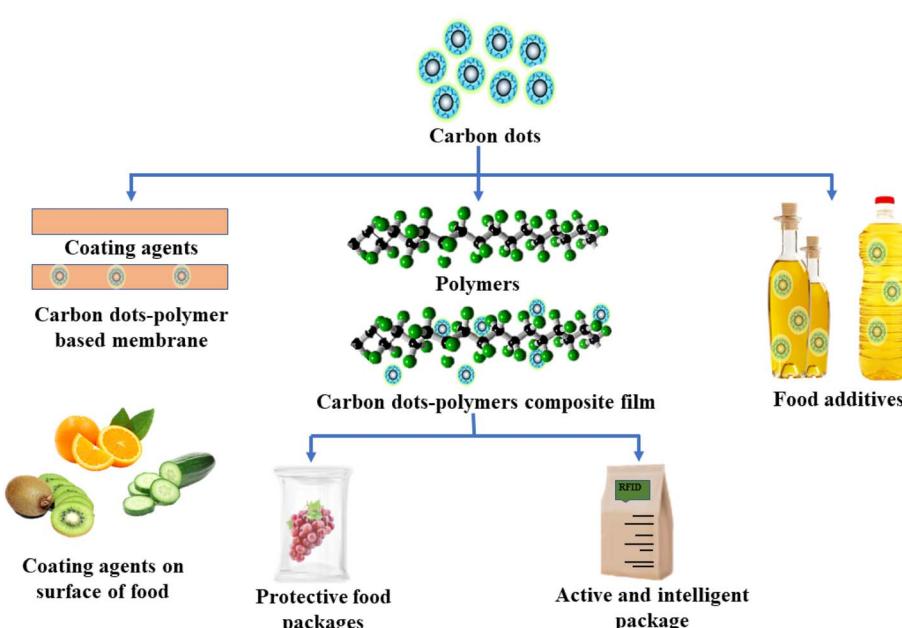


Fig. 4 Potential applications of carbon dots as additives, coating agent, and active and intelligent agents for food packaging applications.



**Table 2** Various kinds of carbon sources, synthesis method, active properties, and variation in thickness, tensile strength, modulus of elasticity, and elongation at break after incorporating carbon dots in different types of composite films

Polymeric film	Carbon source	Synthesis method	Thickness		Tensile strength		Modulus of elasticity		Elongation		Active properties	Ref.
			Control film	Active film	Control film	Active film	Control film	Active film	Control film	Active film		
Carboxymethyl cellulose/carbon quantum dots	Chitosan	Hydrothermal at 180 °C for 12 h	58.3 µm	61.3 µm	30.7 MPa	31.5 MPa	1.3 GPa	2.1 GPa	6.4%	7.9%	Antioxidant, antibacterial and antifungal activities	33
Silk sericin to silk sericin protein/chitosan	Silkworm cocoon	Hydrothermal at 100 °C for 2 h	0.05 mm	0.06 mm	10.39 MPa	22.01 MPa	—	—	18.5%	24.04%	Antibacterial, antioxidant, and UV blocking	32
Silk sericin protein/chitosan to silk sericin protein/chitosan/carbon dots	Silkworm cocoon	Hydrothermal at 100 °C for 2 h	0.06 mm	0.07 mm	8.21 MPa	19.83 MPa	—	—	24.0%	54.7%	Anti-counterfeiting, antioxidant, and antimicrobial	32
Chitosan/gelatin to chitosan/gelatin/carbon dots	—	Hydrothermal	51.8 µm	52.9 µm	79.3 MPa	82.1 MPa	2.8 GPa	3.1 GPa	6.4%	7.3%	Antioxidant, and antimicrobial	54
Cellulose nanofiber-based graphene carbon dots to N-functionalized graphene carbon dots	Glucose	Hydrothermal at 200 °C for 6 h	32.8 µm	36.1 µm	75.2 MPa	76.0 MPa	6.2 GPa	5.9 GPa	3.9%	4.1%	UV-blocking and antimicrobial	25
Polyvinyl alcohol to polyvinyl alcohol/waste tea residue carbon dots	Waste tea residue	Muffle furnace at 200 °C	0.040 mm	0.07 mm	249.8%	209.99%	—	—	4929.12 gf	4926.847 gf	UV-blocking	13
Gelatin/carrageenan to gelatin/carrageenan/enoki mushroom-based carbon dots	Enoki mushroom	Hydrothermal at 200 °C for 6 h	54.5 µm	61.4 µm	52.8 MPa	81.2 MPa	1.3 GPa	0.8 GPa	3.9%	6.4%	Antimicrobial	3
Pectin/gelatin to pectin/gelatin/carbon dots	Potato	Hydrothermal at 200 °C for 6 h	—	—	55.3 MPa	60.6 MPa	4.3 GPa	2.0 GPa	8.9%	6.4%	Antioxidant, and antimicrobial	31
Pectin/gelatin/carbon dots to pectin/gelatin/sulphur-doped carbon dots	Potato	Hydrothermal at 200 °C for 6 h	—	—	52.8 MPa	20.1 MPa	2.3 GPa	1.1 GPa	6.4%	12.5%	Antioxidant, and antimicrobial	31
Cellulose nanofiber to cellulose nanofiber-based graphene carbon dots	Glucose	Hydrothermal at 200 °C	32.8 µm	33.1 µm	73.3 MPa	75.2 MPa	6.3 GPa	6.2 GPa	3.8%	3.9%	Antioxidant and antimicrobial	25

properties of the polymer film. The stress-strain study of PVA and PVA/WTR-CDs composite films was conducted at a high concentration of waste tea residue-carbon dots. The tensile strain, tensile stress, and extension properties of PVA films are 249.78%, 194.06 kg cm<sup>-2</sup>, and 63.44 nm, respectively. After adding WTR-CDs to the PVA film, the tensile strain, tensile stress, and extension were 209.97%, 104.53 kg cm<sup>-2</sup>, and 53.44 nm, respectively. From the above results, Patil *et al.*<sup>13</sup> (2020) observed that the value of extension and stress-strain parameters in the PVA film were high compared to the PVA/WTR-CDs films. The gelatin film's thickness, tensile strength,

and elongation at break are 60.7 MPa, 62.3 µm, and 12.2%, and after adding carbon dots, they increase to 65.3 MPa, 69.0 µm, and 12.0%, respectively. The flexibility of the gelatin/CD film did not change the cruciality, but the stiffness of the composite film decreased significantly. Min *et al.* (2022) reported that a low concentration of carbon dots increased the mechanical strength of the gelatin/CD composite film. This result indicates that hydrogen bonding and interfacial interaction between the gelatin/CD matrix increased at low concentrations.<sup>29</sup>

The addition of carbon dots increased the tensile strength of the biopolymer matrix. The tensile strength of pectin/gelatin-



based films depends on the solid intermolecular interaction between the -OH group of pectin and the -COOH group of gelatin. The interfacial interaction between the pectin/gelatin-based biopolymer and carbon dots is strong compared to the pectin/gelatin-based composite film. Adding carbon dots in pectin/gelatin-based films increased the tensile strength and modulus of elasticity and decreased elongation at break.<sup>52</sup> The addition of sulfur-doped carbon dots (S-CDs) reduced tensile strength and increased elongation at break. The different results of mechanical properties of CDs and S-CDs composite films depend on the surface functional groups on CDs.<sup>31</sup> The addition of carbon dots into cellulose nanofiber (CNF) does not significantly alter the thickness of the film. The tensile parameters of the CNF film are relatively high; for example, tensile strength was nearly 74 MPa, young modulus of elasticity was 6.1 GPa, and elongation at break was 4.1% compared to pectin, gelatin/carrageenan-based polymeric films. The addition of graphene quantum dots and the nitrogen-doped graphene quantum dots in the CNF did not affect the mechanical properties of the composite film.<sup>25</sup>

## 5.2. Barrier properties

Barrier properties are essential for retaining food refreshment, nutritional values, and quality. Ultra-violet (UV) barriers, water vapor permeability (WVP), water contact angle (WCA), and gas barriers help to protect food from the atmosphere. This section provides a brief idea about the significance of barriers in food packaging.<sup>29</sup>

**5.2.1. Ultraviolet light barrier.** Carbon dots exhibit good barrier properties and resist harmful ultraviolet radiation between the food product and the package. Dairy products with vitamin B<sub>2</sub> lose much nutrition when they come in contact with ultraviolet radiation, and UV radiation also promotes oxidation, off-flavor, decomposition, and the creation of some hazardous chemicals in food. UV radiation affects the oil in food, which can lead to oxidation, change in the food's color, and reduce vitamin C and vitamin D in food.<sup>55,56</sup> Carbon dots have good scattering and adsorption properties in that they immediately convert UV photons into heat, which means they can be used to protect food from UV radiation. Fig. 5 represents the UV barrier mechanism when carbon dots are incorporated into biodegradable packaging. Recent research on carbon dots illustrates that carbon dots are active agents as UV barriers and play an essential role in food packaging.

Patil *et al.* (2020) reported that increasing the concentration of tea waste residue-based carbon dot nanoparticles into neat polyvinyl alcohol film (WTR-CDs/PVA) increased the UV-blocking properties of the composite film. The UV-barrier properties of the composite films depend on the various carbon dot concentrations in the polymeric films. The UV-barrier capability changes dramatically when the concentration of waste tea residue-derived carbon dots in PVA increase. Increasing the 0.5 mg concentration of the waste tea residue-derived carbon dots in PVA can prevent the transmission of light of UV-A (310 nm to 400 nm) and UV-B (280 nm to 310 nm) and also reduce UV-C (235 nm to 280 nm). This result indicated

that enhancement of the UV adsorption capability changes dramatically when the concentration of waste tea residue-derived carbon dots in PVA increases in the PVA film. There was no significant change in thickness, or mechanical characteristics, such as tensile strength, along with the transparency of the film after adding WTR/CDs UV blocking agent in the polyvinyl alcohol film.<sup>13</sup>

Further, another study showed that adding carbon dots in chitosan/gelatin composite film increased UV-blocking of the radiation without changing other properties, such as transparency and tensile characteristics. Ultraviolet radiation generally encourages the oxidation and decomposition of nutrients in foods due to various chemical compounds, which form hazardous compounds, and leads to the decolorization of food products. The pectin/gelatin film was excellent and had visible light transparency. Incorporating carbon dots and S-CDs decreased the transparency of the films while increasing the light transmission to visible light. The addition of carbon dots and sulfur-doped carbon dots in the pectin/gelatin film decreased the transparency of the composite film but increased the UV barrier properties. At 280 nm, light transmittance blocks UV-A (320 nm to 400 nm) and UV-B radiation.<sup>31</sup> Glucose carbon dots (GCD) and N-functionalized carbon dots (NGCD) were used to prepare cellulose nanofiber (CNF)-based composite films. GCD and NGCD provided high UV blocking properties to the CNF film without changing the transparency of the composite film. At 280 nm light transmittance, the transparency of the CNF film was considerably reduced from 53.8% to 5.2% and 1.3%, while the addition of the glucose carbon dots and N-functionalized carbon dots in the CNF film at 660 nm light transmittance decreased from 86.7% to 80.2% and 78.4%, respectively.<sup>25</sup>

Carboxymethyl cellulose (CMC) films had good ultraviolet (280 nm) and visible light (660 nm) transmittances of 61.0% and 84.2%, respectively. The ultraviolet-barrier characteristics increased with the addition of carbon quantum dots to CMC. At the carbon quantum dot (CQDs) concentrations of 1.0%, 3.0%, and 5.0%, the ultraviolet-barrier characteristics of the composite film increased by 81%, 98%, and 100%, respectively, because CQDs have good light-adsorbing properties. But at the other hand, the inclusion of carbon dots did not affect the transparency of the composite film. The size of CQDs is much smaller than the 660 nm wavelength of light, which does not interact with the transmitted light. As such, it showed that the addition of carbon quantum dots in the CMC blocks the UV radiation without any impact on the transparency of the composite film.<sup>33</sup> The above results indicated that adding carbon dots to the composite film enhanced the composite film's ultraviolet barrier characteristics, which can be utilized in the food packaging sector.<sup>56</sup>

**5.2.2. Water vapor barrier.** The transmission of water vapor through a film involves several physical phenomena, including the adsorption of water vapor on the film's surface, water molecules passing through the film, and water evaporation on the opposite side of the composite film.<sup>12</sup> The reduced water activity in the food helps inhibit the growth of mold and microorganisms. Water solubility is also the main factor in food



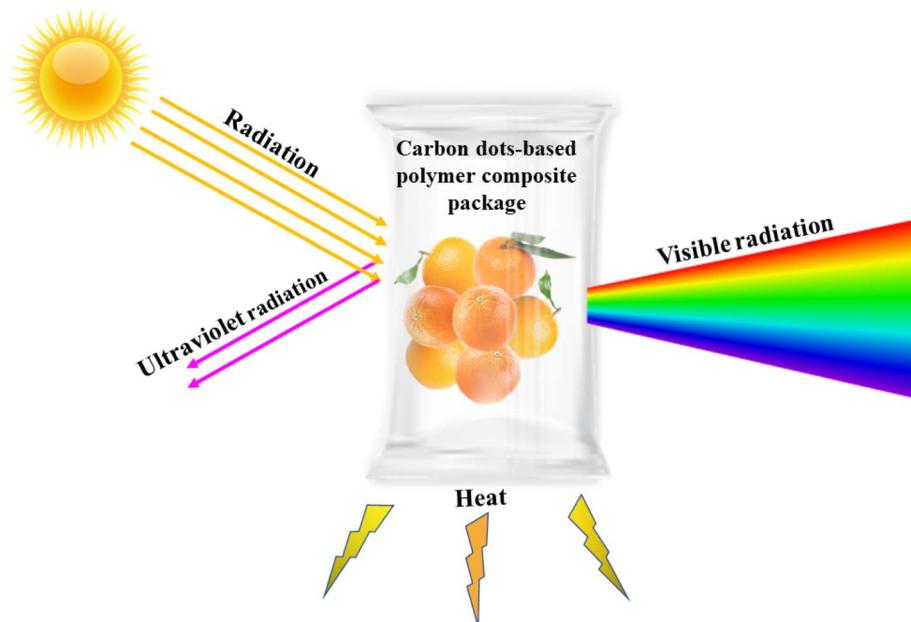


Fig. 5 Ultraviolet light barrier mechanism of carbon dots based on biodegradable packaging illustrating food product's protection from ultraviolet radiation.

packaging to increase the shelf life of the food. Water solubility retains moisture-sensitive foods through packaging. The water solubility in the pure cellulose nanofiber-based film was 18%; however, adding nitrogen-doped carbon dots and graphene carbon dots increased the water solubility by 24%. The graphene carbon dots have good hydrophilic nature, which increases water solubility. The rate at which functional materials discharge and their magnificent qualities, such as anti-bacterial and antioxidants, are anticipated to be influenced by the water-resistance properties, water solubility, and swelling ratio.<sup>57</sup> The low water vapor permeability (WVP) of the packaging films helps maintain the shelf life and quality of food products. Food quality changes when moisture is transferred between the food and the surrounding atmosphere. The water vapor permeability depends on the interaction between the filler and the polymer matrix.

Research suggested that the WVP of the pure CNF films did not alter with the addition of graphene carbon dots in the polymer matrix. But the addition of nitrogen-doped graphene carbon dots to the CNF composite film enhanced the WVP activity of the film. This might be due to the high hydrophilic tendency of CNF films containing nitrogen-based graphene carbon dots compared to the CNG films with graphene carbon dots. This claim can also be justified by the water contact angle values of both films. The water contact angle of pure cellulose nanofiber film was nearly 30°, indicating that the CNF film was highly hydrophilic. The addition of graphene carbon dots in the cellulose nanofiber composite film decreased the water contact angle because carbon dots have hydrophilicity properties. The surface wettability and hydrophilicity of the CNF composite film depend on the mode of the interaction between the polymer and additives.<sup>25</sup>

The addition of carbon dots (2 wt%) in a neat gelatin film increased the water vapor permeability of the composite film, which decreased the water vapor because of the surface hydrophilic functional groups of the carbon dots. At the same time, the water contact angle of the composite film was significantly reduced by adding carbon dots. The water contact angle and water vapor permeability of gelatin/carrageenan film were 58.3° and  $0.95 \times 10^{-9} \text{ g} \times \text{m m}^{-2} \times \text{Pa} \times \text{s}$ , respectively.<sup>29</sup> The addition of enoki mushroom-based carbon dots (mCDs) in the gelatin/carrageenan had no significant change in the water barrier properties. Enoki mushroom-derived carbon dots (mCDs) had surface hydrophilicity properties, which increased water vapor permeability but decreased the water contact angle in the gelatin/carrageenan/mCDs composite film. As such, it is concluded that the hydrophilic nature of the composite film depends on the concentration of carbon dots and nature polymers.<sup>58</sup>

**5.2.3. Oxygen barrier.** The transmission of gases between food products and the surrounding environment also affects the food shelf life, an important aspect of the food packaging domain. Oxygen gas can oxidize several kinds of compounds in food and reduce the shelf life of the food. For maximum storage life, the food should be stored in an oxygen-free atmosphere in the packages. The oxygen transmission rate of the neat cellulose nanofiber-based film is nearly 5 cc per m<sup>2</sup> per day. However, the addition of carbon dots, graphene carbon dots, and nitrogen-doped carbon dots decreases the oxygen transmission rate of the cellulose nanofiber film. The addition of carbon dots prevents oxygen in a biopolymer-based neat film. Decreased gas transfer, resulting from the addition of carbon dots to the clean CNC film,<sup>25</sup> between fresh food and the atmosphere inhibits the growth of microorganisms.



## 6. Carbon dots as an active material

Active packaging interacts with the packaged product effectively, either chemically or biologically. It affects the product in the packaging, for instance, preventing microbial growth, maintaining the quality and freshness of food, extending shelf-life or inhibiting fungal growth, and providing information about the product's condition. Active packaging mainly includes antimicrobial and antioxidant properties.<sup>59</sup>

### 6.1. As an antioxidant agent

Antioxidant-releasing packaging techniques effectively reduce the food oxidation rate, increasing the food shelf life.<sup>60</sup> Currently, a series of CDs, such as GQD, selenium-doped CDs, nitrogen-doped CDs, and chlorine-doping CDs are widely used as antioxidants for reducing reactive oxygen species. The sources utilized to synthesize carbon dots with antioxidant properties are primarily plant-origin compounds, and the formation technique typically involves an autoclave. Das *et al.* (2021) found that the DPPH free-radical blocking properties of nitrogen, phosphorous, and sulfur-doped (NPSC) carbon dots synthesized using a one-step autoclave method were reduced. By coating with nitrogen, phosphorous, and sulfur-doping of carbon dots polypropylene composite film, the antioxidant capacity of the NPSC dots was efficiently employed on plastic packages. Many types of radicals, such as DPPH radicals, hydroxyl radicals,  $\text{KMnO}_4$  radicals, and superoxides anion radicals, elevated the antioxidant properties in the NPSC-dots. DPPH (2,2-diphenyl-1-picryl-hydrazyl-hydrate) is a kind of electron transfer and stable radical used to evaluate the antioxidant properties of the compounds. This method is both economical and practical for the measurement of antioxidant properties. When DPPH reacts with an antioxidant, it absorbs the hydrogen-radical to form the DPPH-hydrogen molecule, which has a pale-yellow appearance. Because it contains nitrogen atoms with lone-pair electrons and is enclosed by 3 benzene rings, DPPH is a stabilized free radical. The antioxidant properties of the NPSC dots were enhanced as the concentration increased nearly to  $417 \mu\text{g mL}^{-1}$ . The nitrogen, phosphorous, and sulfur-doped CDs (NPSC-dots) have the highest scavenging activity of 74% for DPPH radicals.<sup>37</sup>

The gelatin/CDs film shows high antioxidant activity and is intended to inhibit the degradability of packaged foods sensitive to oxidative degradation. Depending on the concentration of carbon dots, considerable antioxidant activity in the ABTS (2,2-azino-bis(3-ethylbenzothiazoline-6-sulfonic acid)) and DPPH techniques were determined. Carbon dots have good shielding ability, which could be attributed to surface structural features that contribute to removing free radical scavenging to produce more developed systems in the ABTS and DPPH methods, and in most of the films, antioxidant properties from the DPPH method are less compared to ABTS methods. Gelatin-based films have some scavenging activity assessed using the DPPH and ABTS methods. Antioxidant activity in gelatin film was 27.0% because of some antioxidant peptides present in gelatin. The addition of carbon dots at 2.0% and 4.0% levels

enhances the scavenging capacity of the film to 99.0% and 99.6% in the ABTS method.<sup>29</sup>

Similarly, incorporating carbon dots in the gelatin film increases the free radical scavenging activities by 72.1% and 93.2%, respectively, for 2.0% and 4.0%, when assessed using the DPPH method. From the above results, it can be concluded that the antioxidant activity of the carbon dot incorporated films was more significant when measured using the ABTS method, as compared to the DPPH method, which could be due to the higher expanding rate of the gelatin film in an aqueous ABTS mixture than in a methanolic DPPH solution. Carbon dots' solid antioxidant activity can be linked to their surface functional groups capable of scavenging free radicals. Carbon dots added in the biopolymer-based composite films have more antioxidant activity, such as pectin/gelatin and gel/carr composite films.<sup>29</sup> Some parameters influencing the antioxidant activity are electron flow, unpaired electrons, hydrogen donating behavior, hetero-atom doping,  $\text{sp}^2$  hybrid carbon domain, and surface functional group type. Research indicates that the presence of carbon dots at 2 or 3  $\text{mg mL}^{-1}$  concentration helps improve the films' antioxidant activity by about 18-fold and prevents oxidative rancidity.<sup>31</sup> The antioxidant properties of the cellulose-based film were determined by DPPH and ABTS methods. The value of antioxidants for the bacterial nanocellulose (BNC) film without adding carbon dots was 14.0% and 5.0%, from the ABTS and DPPH methods, respectively. After adding carbon dots, the antioxidant activity of the BNC composite film was enhanced.

The addition of GCD and NGCD in the CNF film increased the capacity to scavenge free radicals to nearly 98.1% and 98.0%, respectively, in the ABTS method, but for the DPPH method, the values were 79.0% and 84.0%, respectively. The antioxidant activities of graphene carbon dots/BNC composite film were more significant in the ABTS method as compared to the DPPH method. The surface functional groups of the graphene carbon dots that take part in free radical scavenging are responsible for good antioxidant activity in the CNF film. CNF/NGCD and CNF/GCD composite films presented the same antioxidant properties in the ABTS method. But, the NGCD/CNF composite films presented a more excellent antioxidant activity in the DPPH method as compared to NGCD/BNC, and due to the high antioxidant properties, these films can be used in the active packaging domains to reduce the rate of food oxidation.<sup>25</sup> The enoki mushroom-based carbon dot/gelatin/carrageenan composite film significantly increased the free-radical release effect. Similarly, they increased the antioxidant properties compared with pure gelatin/carrageenan films.<sup>3</sup> The increased activity in ABTS is because mCDs are more soluble in an aqueous ABTS solution than in the DPPH methanol solution. The occurrence of surface hydroxyl groups on mCDs may explain their excellent antioxidant property. Enoki mushroom-based carbon dot/gelatin/carrageenan composite film significantly increased the free radical release effect, resulting in a film with higher antioxidant properties as compared to pure gelatin/carrageenan films. Carbon dots work as active agents with heteroatom doping in the films in the above process.<sup>3</sup>



## 6.2. As an antimicrobial agent

Carbon dots from various sources exhibit antimicrobial properties against a broad spectrum of food-borne pathogens and spoilage microorganisms. The antimicrobial properties of carbon dots depend on their shape, size, surface charge, and various functional groups.<sup>52</sup> It was accomplished by incorporating an active ingredient into the product package, depositing a coating on the product, or incorporating it within the packaging matter. Carbon dot's antibacterial properties are affected by numerous mechanisms such as cytoplasmic leakage, cell structure breakdown, and reactive oxygen species (ROS) production of genomic deoxyribonucleic acid (DNA) fragmentation/condensation.<sup>38</sup> The addition of carbon dots to the films demonstrated different antibacterial actions in the gelatine-based film based on the number of carbon dots and bacterium test.<sup>38</sup> The composite films inhibited the development of *E. coli* but disturbed the growth of *L. monocytogenes*, and the action was enhanced when the concentration of the carbon dots was increased. Paper is commonly used as packaging material for food packaging and other fields since it is recyclable, ecological, and affordable. Li *et al.* (2021) developed a paper with antibacterial or waterproofing properties that can be utilized for food packing to increase the shelf-life of food. The antibacterial activity of paper, nano-ZnO, and graphene quantum dots was produced on the surface using a hydrothermal synthesis approach. The BNC paper displayed antimicrobial properties against Gram-negative *E. coli* and *L. monocytogenes*. Some CDs were implanted with nanocellulose using an ex-situ process to produce antimicrobial/ultraviolet-protected nanopaper. The antibacterial potential of carbon dots at different concentrations, such as 100, 250, and 500 mg mL<sup>-1</sup>, was investigated using the agar well diffusion methods.<sup>25,61</sup>

In another research, the viable cell colony count method was used to assess the antibacterial activity of the pectin/gelatin composite films towards food-borne pathogenic bacteria, *L. monocytogenes*, and *E. coli*. The pure pectin/gelatin layer did not show any antibacterial properties, but the addition of carbon dots in pectin/gelatin showed antibacterial properties that prevented the growth of *E. coli* and *L. monocytogenes* by nearly 2 log CFU mL<sup>-1</sup>. Nevertheless, the addition of sulfur-doped carbon dots (S-CDs) in the pectin/gelatin composite film showed antimicrobial properties that increased rapidly because S-CDs have some functional groups in sulfur, such as sulfonic acids and sulfonates. These functional groups help bind the enzymes, produce reactive oxygen species, and degrade lipids or proteins.<sup>31</sup> The carbon dots derived from the turmeric and sulfur-functionalized carbon dot composite film showed good antimicrobial properties through ABTS and DPPH methods. The zone inhibition method was used to determine the antimicrobial activity of carbon dots and sulfur-based carbon dots.<sup>62</sup> Carbon dots are also used in biopolymer solutions to create an antibacterial coating around food. In this way, the food is dipped into the antimicrobial solution. For example, the coating on freshly-cut cucumber dipped in the antimicrobial additive polymer solution. The kelp-derived carbon dots were

dipped into the chitosan solution and coated on the cucumber because the coating on the fresh cucumber under modified atmosphere conditions prohibited the growth of microorganisms, mold, and yeast.<sup>44</sup>

The addition of nitrogen-doped carbon dots in a cellulose nanofiber film demonstrated more excellent antimicrobial properties than graphene carbon dots. Therefore, NGCD/CNF-based nanofiber showed excellent antimicrobial properties for food packaging to prevent food spoilage and enhance the shelf-life of food. The graphene carbon dots inhibit the growth of the microorganism; meanwhile, microbial killing was demonstrated by the NGCD-added film. Various studies have demonstrated that generating carbon dot-generated reactive oxygen species leads to cell death. Therefore, the antimicrobial properties of carbon dots were performed using the oxygen radical absorbance capability method, and the formation of reactive oxygen species solutions by adding carbon dots and sulfur-doped carbon dots increased over time. The aforementioned research studies on carbon dots conclude that a broad spectrum of antibacterial action and antimicrobial properties are greatly influenced by the type of carbon used, particle size, doping atom and shape, and the production process.<sup>38</sup>

## 7. Challenges in carbon dots for food packaging

The advanced improvement of nanotechnology is to the creating a novel sensor for food packaging that includes a platform that enhances food quality, reduces unwanted waste, assesses protection, and enhances shelf-life.<sup>63</sup> Numerous innovative nanoparticles are now being studied for application in food packaging. The center of attention on nanomaterials for their safety in food packaging is their toxicity and higher surface region to volume proportion, leading to hazardous reactions. Introducing any novel products into the market must thoroughly examine the impact of the products on the health of the consumer and the atmosphere across the food chain, after which they can be used in the food packaging industry, sales, and marketing.<sup>64</sup>

The nanomaterial utilized in the current technology has the potential to transfer and penetrate the body *via* breathing, ingestion, and skin absorption; it is going to affect consumers' health. Toxicity is a significant factor in determining the capacity-environmental impact of carbon dots.<sup>65</sup> Current research shows that biological hazards demonstrated that carbon dots are biocompatibility and do not induce noticeable pathological changes in the body after insertion. The release of carbon dots from packaging material is a factor of the possible toxicity of carbon dots and requires specific consideration. Furthermore, a few research articles have investigated the effect of incorporating carbon dots into food-inert substances. Many factors influence the liberation of active components from films or active packages to food simulants, including the type of food additives, the suitability of the active material with the film, the solubility of the film, and the quantity or concentration of the substance. For instance, the concentration of mushroom-



derived CDs and the food additives affected how quickly they were released from a gelatin/carrageenan-based polymer matrix. When the concentration of carbon dots was increased from 1.0% to 6.0%, the release rate of enoki mushroom-derived carbon dots in all simulants improved with the increase in concentration. The release rates of carbon dots derived from enoki mushrooms in 95.1% and 50.2% ethanol were lower than 10.0% ethanol and water. Carbon dots are tiny nanoparticles, which may be a concern for some consumers.<sup>3</sup>

## 8. Future recommendation

The application of carbon dots as nanoscale food additives for extending food's shelf life is still lacking. As such, realistic food samples must be used to prove the practical use of carbon dots as additives in packaging materials.

The heteroatom-doped carbon dots also form a composite film; for example, sulfur-functionalized turmeric-based carbon dots like sulfur-based carbon dots (S-CDs) and nitrogen-doped carbon dots. Thus, it would also be crucial to investigate the possible hazards or toxicity of various types of carbon dots to verify that they may be used securely in food applications.

There are two approaches to synthesizing carbon dots: The top-down and the bottom-up. In the bottom-up approach, the hydrothermal method is an easy, environmentally friendly, and one-step synthesis quickly dispersed in solution, and the reaction operates below 300 °C. Furthermore, hydrothermal synthesis is less expensive than other solution synthesis processes in terms of instruments, power, and time. It will be essential to develop cost-effective techniques for generating carbon dots on a broad scale to be economically feasible for culinary applications.

More research is needed to determine the effect of specific carbon dots' features, such as shape, composition, charge, and functional groups, on the properties of packaging materials synthesized from various polymers.

Consumers' opinions on using carbon dots in meals and manufacturing processes are essential. By thermal or pyrolysis methods, brown-colored carbon dots may be restricted to using carbon dots on a wide scale in various meals. For this issue, large-scale production can be handled using new non-thermal methods for carbon dots.

Carbon dots are tiny nanoparticles, which may be a worrying point for some people. Due to their nanosize and dimensions, they can enter the human body through the skin-inhaling process and affect our health. The release of carbon dots from the packages is the main issue. It is necessary to research and determine the carbon dots' migration into the gastrointestinal food system and their potential toxicity. This paper showed that solid waste and residues from food manufacturing could be converted into significant CDs, potentially boosting the sustainability of the world's food supply and economics.

## 9. Conclusion

Carbon dots are a wide source of raw materials at low cost, good photoluminescence, UV blocking, high stability, easy

interaction with substrates, and photoelectrochemical capabilities. The carbon dots offer numerous advantages for advanced or innovative food packaging, including enhanced mechanical properties, water barrier, thermal activity, barrier properties (radiation blocking, water, and gas), and antimicrobial and antioxidant properties. Carbon dots can be combined with biological-chemical indicators to create rapid and sensitive packages for detecting food quality and originality. C-dots can be synthesized with different compositions and characteristics depending on the primary sources and processing conditions. They have a high potential for usage as active material, nano-level food additives, coating agents, and intelligent agents to improve the shelf-life of food products. Carbon dots provide superior safety to other nanoparticles. At suitable concentrations of carbon dots, nontoxicity is observed for human and animal cells. Carbon dots help protect cells from oxidative stress harm in exceptional cases. Research on carbon dots used to create intelligent packaging that detects food freshness and active packaging with antibacterial and antioxidant capabilities is a prospective research area. Carbon dots have been studied as multipurpose nanoparticles for food packaging films, with significant improvements. It expands the range of possible applications for radically-new quantum materials. The migration of carbon dots in the food is the main reason for toxicity. Additional research should be conducted to investigate the potential migration patterns of carbon dots from packaging films into various food systems and create ways to regulate the release of carbon dots.

## Author contributions

DG: investigation, writing – original draft, writing–review, and editing, LK: writing and editing. KKG: conceptualization, supervision, project administration, funding acquisition, writing–review, and editing.

## Conflicts of interest

The authors declare that they have no conflicts of interest.

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