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Quantum Dots and its significance in Sustainable Agriculture: A Review

Sandeep Sirohi¹, Nitika Vats², Sanjay Kumar¹, Vivek Srivastava³ and Jaidev Kumar^{4*}

¹Department of Botany, Hariom Saraswati P.G. College, Dhanauri, Uttarakhand 247667, India
(Sri Dev Suman Uttarakhand University, Badshahithol, Tehri Garhwal, Uttarakhand)

² Department of Biotechnology, Meerut Institute of Engineering and Technology, Meerut (UP) 250005, India
(Dr APJ Abdul Kalam Technical University, Lucknow)

³Department of Biotechnology, Rama University, Rama City Mandhana, Kanpur (UP) 209217, India

⁴Department of Chemistry, Hariom Saraswati P.G. College, Dhanauri, Uttarakhand 247667, India
(Sri Dev Suman Uttarakhand University, Badshahithol, Tehri Garhwal, Uttarakhand)

***Corresponding Author – jaidev112233@gmail.com,**

INTRODUCTION

Past few decades have witnessed extensive research over semiconductors that yielded some miraculous results. One of those significant yields includes the – Quantum dots. The quantum dots are practically electrons confined into lines, planes or just dots. Owing to their nanoscale size, they are also an outstanding contribution in the field of nanotechnology. Over the past few years, nanotechnologists have been able to synthesize several kinds of QDs that found utility in varying spheres of innovation and technology. From being used in LCDs to making their way into agriculture, QDs have become worthy of mention. This chapter includes further outlook into the properties, synthesis and application of quantum dots in the field of nanotechnology in general and agriculture in particular.

Nanofabrication techniques are being used in the development of such structures that, in turn, opened endless possibilities in the field of physics and chemistry. Researchers now had the chance to study and observe artificial analogues of atoms [1]. Quantum dots exhibit useful electronic and optical properties thus their usage most widely began as light emitting or absorbing elements. These properties are different from their larger particles owing to quantum mechanics. The electron in the quantum dots gets excited to a higher energy state when illuminated by light. This process corresponds to the transition of the electron from the valence band to the conductance band. After that, the electron returns back into the valence

band, releasing energy in the form of light [2]. Quantum dots are hence used in QD displays or photovoltaic cells. Soon they found significant application in biological sciences too.

The free mean path of a quantum dot is more than its width; it, therefore, shows a quasi-zero-dimensional behaviour. Each of these dots contains 1-200 electrons and are hence called ‘artificial atoms’ where the nucleus potential is served by an artificial potential [3]. Making of a Quantum dot into a real 3D structure traces back to quantum mechanics and Heisenberg’s uncertainty principle. The momentum and the position of electrons cannot be precisely known hence a wide range of momenta equates into higher average energy. Now, if these electrons are confined into an infinitely thin layer, they will exhibit infinite energies. The properties of a semiconductor mainly depend upon temperature or its material properties. If these particles are confined in a thin layer, the effects of the uncertainty principle set aside other considerations. So if the electrons do not have enough energy to be able to get out on the confinement, they become 2-Dimensional. The electrons confined in a dot are not free in any dimension. The length scale of a free conduction electron in a semiconductor is 100 Å where 100 Å equals about the radius of a hydrogen atom. An electron in a semiconducting cube of a 100 Å side is confined to a point [1]. The semiconductor QDs were investigated for many years but were eventually replaced by Carbon QDs due to toxicity caused by heavy metals used in their production [4]. Carbon QDs have lesser toxicity, better biocompatibility, were cheaper and chemical inert, making them an ideal substitute.

PROPERTIES OF QUANTUM DOTS

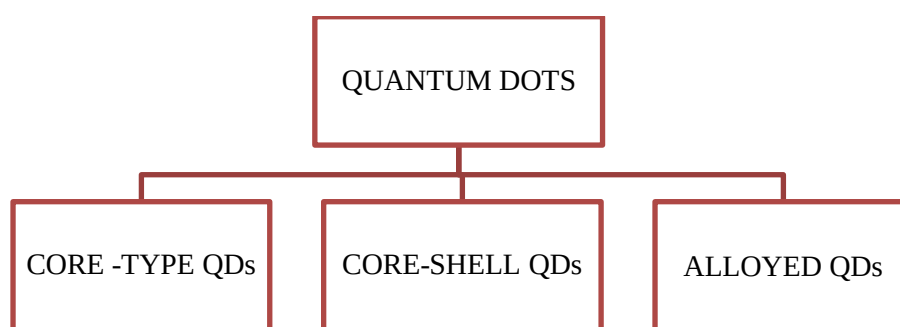
The optical and electronic properties of nanocrystalline semiconductors were first discovered by Ekimov of the Vasilov State Optical Institute in Russia in 1981 who was responsible for synthesizing nanocrystals embedded in a glass matrix[5]. Several other scientists worked on these nanocrystalline semiconductors until Murray, Norris, and Bawendi in 1993 detailed a hot injection synthesis of monodisperse colloidal nanocrystals [5] after which researchers across the globe began considering QDs for commercial usage.

Quantum dots are artificial atoms whose properties can be controlled by using electrostatic gates by applying the magnetic field or by manipulating their geometry [6]. Since we have enumerated several times that quantum dots are 3-dimensional models of atoms, they exhibit similar properties too, such as quantized energy levels and shell structures. These properties are described by the electron wave function that is given by the Schrödinger equation and the Pauli exclusion principle [7]. It wouldn’t be wrong to state that the standard model to describe the electronic structure of QDs is the k·p method [8] which is a semi-empirical method for calculating the band structure and optical properties of crystalline solids(QDs

in this reference). QDs have an intrinsic bandgap through which electrons are bridged by exciting them by incident light. QDs being sparse enough to not create the continuous, valence and conduction bands produce rarefied electronic structures found typically in single atoms. Larger the QD, the smaller the bandgap gets and the closer it comes to that of a bulk semiconductor. Thus, the dependency on the size allows flexibility in the modulation of the bandgap energy by varying the size of QDs [9]. The ‘quantum confinement’ partly contributes to the optical properties of QDs. This means that the energy levels in which the electrons typically stay in become discrete, with a finite separation between them [10]. When a sufficiently energized photon strikes a QD, it might excite the electrons from valence to the conductance band, hence leaving a positive hole. This phenomenon is common in semiconductors, but the average exciton size is smaller than the size of QDs producing confinement energy as the exciton is squeezed into the material [9].

That was the physics behind the properties of the nanoscale quantum dots that we have harnessed and primarily applied in all major commercial spheres. The possibilities are endless, and with the advancement of their utility in the biological sciences, QDs are also making their way into the advanced agriculture industry.

TYPES OF QUANTUM DOTS



CORE-TYPE QUANTUM DOTS

They are made of a single element and thus have a uniform composition. These typically are selenides, sulfides or tellurides of metals like cadmium, lead or zinc. These are the plain QDs with crystalline metal cores and an organic outer layer [11]. Their properties can simply be varied by varying the sizes of the cores.

CORE-SHELL QUANTUM DOTS

When an additional layer is added to the plain metalloid core, many different types of Core-shell QDs can be prepared. This improves their light harvesting properties. Another method to enhance these QDs is to add ions to this configuration.

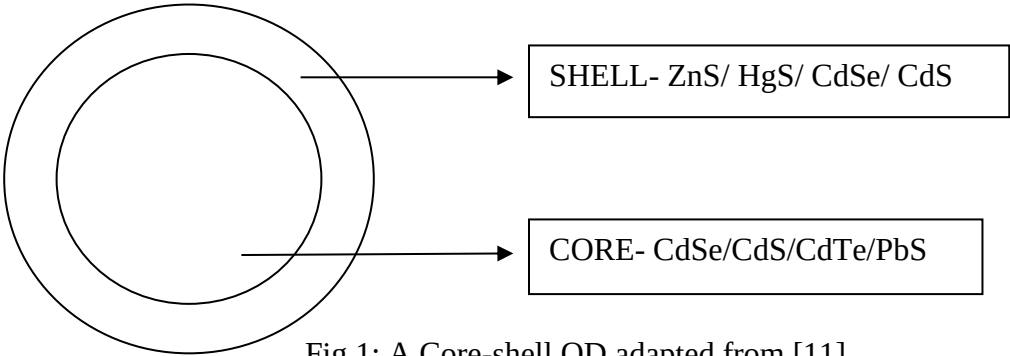


Fig 1: A Core-shell QD adapted from [11]

Depending upon the alignment of the conduction and the valence bands of core and shell, core-shell QDs can be classified into four major classes – Type I, Inverse Type I, Type II, and Inverse Type II. They differ on the basis of their band alignment, the location of carriers, quantum yields, stokes shift, and the average absorption and emission [11]

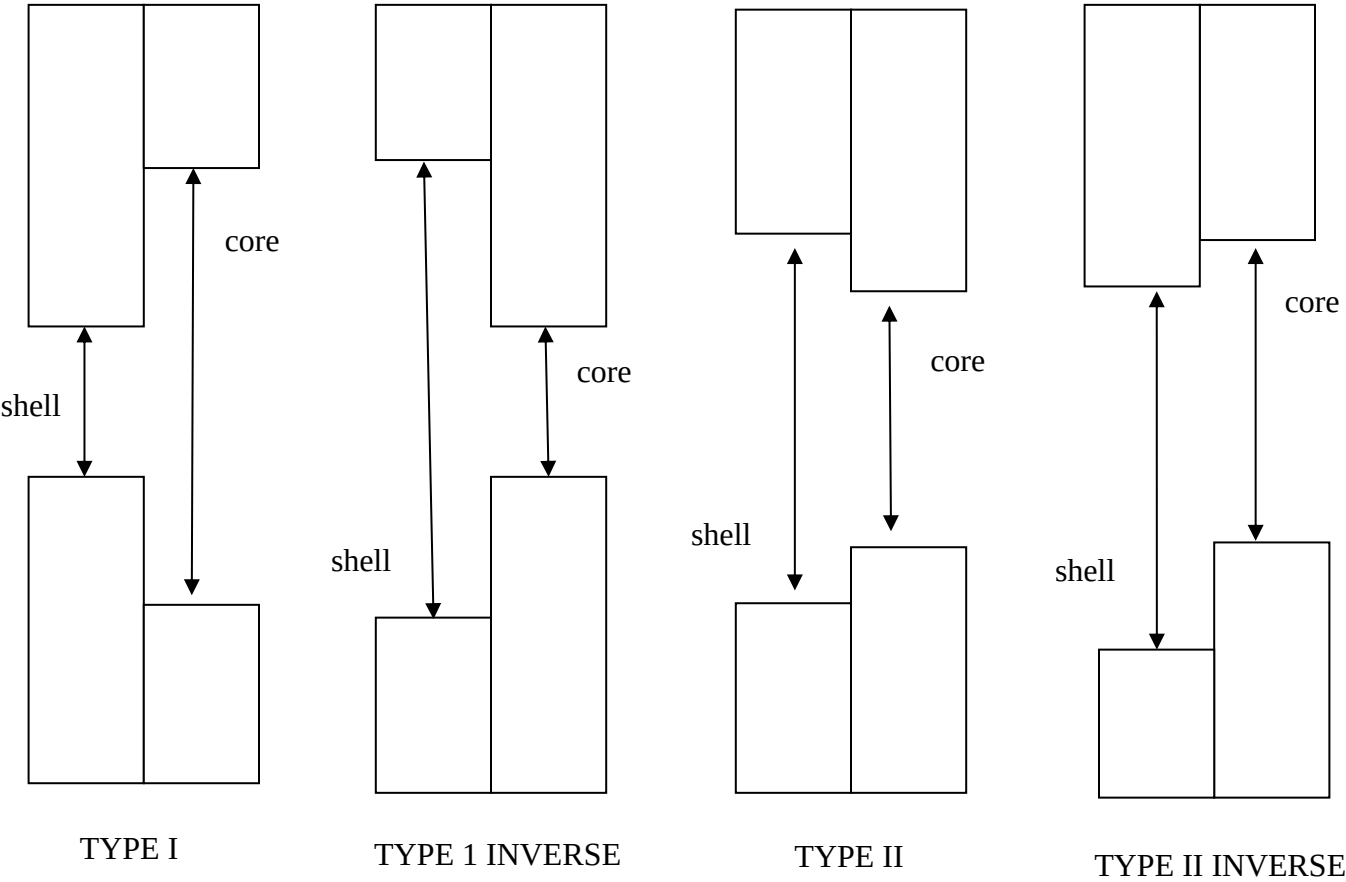


Fig 2: Types of core-shell QDs based on their band alignment [12].

These QDs also differ in the terms of quantum yield where type I and Type II Inverse QDs have a higher yield and better stability as compared to the Type II and Type I Inverse QDs [13,14,15].

The Stokes shift is a term that defines the difference between the peaks of absorption and emission wavelengths [16]. It leads to less possibility of re-absorption and thus, reduction of potential energy loss [11]. The absorption and emission spectra of these dots must match to that of the solar spectrum so that it absorbs photons from the same wavelength [17]. The absorption/emission spectra of these QDs can be manipulated by changing the thickness/size of core or shell [18].

ALLOYED QUANTUM DOTS

The easiest way to manipulate the properties of a QD is by changing its size. That, however, might arise some problems when it comes to application. While, a multi-component QD allows manipulation of properties without actually changing the size of Quantum dots. Alloyed QDs are thus produced by alloying two distinct semiconductors together both having different bandgap energies. These alloyed QDs interestingly exhibit properties that are different from not only their bulk but also from their parent semiconductors.



Fig 3: Alloyed QDs adapted from reference [19]

CURRENT STATUS

- Near-Infrared quantum dots are being used as novel fluorescent labels for their dual advantage as both QDs and NIR light. They have found application into biomedical sciences that includes biosensing, bioimaging and even drug delivery [20]
- Major research on cancer behaviour are using this nanotechnology for biomedical imaging due to the optical and chemical properties of quantum dots. They can even be used in personalized oncology [21,22,23].
- Quantum dots exhibit 'Multiple exciton generation' in which the energy of an absorbed by which the energy of an absorbed photon in excess of the bandgap can be used to create one or more additional excitons instead of being wasted as heat. This effect is being used due to its potential to enhance the performance of solar cells, photocatalysts and lasers [24].
- Organic solvents are generally used to synthesize QDs with high quantum yields and optical stability. However their toxicity, non dissolubility and instability of photo luminescence did not allow much utility of QDs in biology. Polymers are being used to coat QDs to not only make them biocompatible but also to find application [25].
- Due to the optimizable properties of QDs, this nanotechnology is now picking up botanical application. Agriculture industry is looking up to QDs as a solution to many issues these days [26].
- A study has revealed that about 53 companies were involved in the manufacturing and usage of nanomaterials, some including QDs as well by 2005 in the United Kingdom. They were also involved in research and development in the field of nanotechnology [27]. Government of India, on the other hand, has funded nearly 500 research projects throughout the country wherein about 42.5% have already been executed by southern states [28].

SYNTHESIS OF QUANTUM DOTS

QDs can be synthesized by following either the -Top Down Approach or the Bottom Up Approach.

TOP DOWN APPROACH

Top down approach in nanotechnology revolves around a certain goal of miniaturizing technology. In such methods QDs are prepared from larger macroscopic materials like silicon wafers. Some methods following the top down approach of nano-fabrication might include-

1. **Molecular Beam Epitaxy (MBE)**

It is an epitaxial process wherein the growth of material takes place on heated crystalline substrates in a UHV environment. The adsorbed material supplied by a molecular beam interacts with the substrate and grows, having either the same crystalline structure as the substrate, same symmetry as the substrate or has a structure at least 10% similar to the substrate [29]. Quantum dots synthesized by molecular beam epitaxy have great optical properties and have wide application in advanced optoelectronic devices [30][31].

2. **Electron beam lithography**

It is a direct patterning lithography technique that allows highly accurate fabrication of nanostructures. Direct patterning lithography has many advantages over the other methods as it allows an escape from resist deposition, etching and removal hence letting us avoid the issues related to film adhesion, long processing time and generation of roughness in the etching processes [32]. QDs produced by this method have found application in various spheres. Palankar et al. generated high fidelity pre-functionalized water-soluble Quantum dots by using Electron beam lithography [33] while Kaganskiy et al. mentioned to use electron-beam lithography in pre-characterization of QDs before incorporating them into nanophotonic devices. [34]

3. **Ion Implantation**

This process includes implantation of impurity ions in the specific regions of a semiconductor substrate thus forming a semiconductor element. The entire process includes photo-resist development procedure. The ion beam is projected on the entire substrate surface, implanting the ions in specific regions through the resist film opening. It is important for the beam to fall on the substrate at right angles ensuring longer distance between the ion source and substrate [35]. The QDs hence formed often exhibit non linear properties[36].

BOTTOM UP APPROACH

In such an approach a quantum dot is synthesized by using molecular precursors. In other words, bottom up approach includes usage of atoms and molecules that are integrated to form a certain nanostructure.

1. **Wet chemical synthesis**

This process is widely used for QD assembly because it is a versatile and scalable process that requires less labour[37]. It allows the control over kinetic and thermodynamic parameters of QDs thus allowing control over size, shape and composition of QDs[38]. This method has certain limitations as well, one of which includes uncontrolled fluorescent emission[37].

This method might include either colloidal synthesis or spray pyrolysis methods. Typically, in a colloidal synthesis process a solution containing varied ions are mixed together under controlled pressure and temperature thus producing insoluble precipitates. Spray pyrolysis includes the process of atomizing a solution through a nozzle spray. The droplets hence formed are heated to form solid particles.

2. Vapour phase method

This method included self assembly of QDs on the substrate surface by gradually forming layers. The vapour phase precursors are assembled in a hot walled reactor with conditions that promote nucleation of particles rather than their deposition on reactor walls. The vapour phase precursor could be solid, liquid or even gaseous but it has to reach the reactor in a vaporized state only[39].

APPLICATIONS OF QDs IN AGRICULTURE

The essence of Quantum Dots' application in agriculture has proved to enhance horticultural efficiency of sunlight in greenhouses, detection of plant diseases, tracing residual pesticides in agricultural products, the growth rate of plants, vertical farming, plant photosynthetic efficiency in bioregenerative life-support systems (BLSS) and precision farming.

1. Pesticide residue detection

The Food and Drug Administration (FDA) reported about 1045 chemicals as pesticide residues (FDA, 2005). These pesticides have now contaminated our food chain through consistent use over the past 6 decades. Studies have shown that the negative of pesticides have negatively manipulated the growth and reproduction of earthworms, and the harm caused to birds who consumed the insects feeding on pesticide infested plants[43]. Fish and other aquatic biota were seen to be lethally harmed by the pesticide surface run-off infested streams and rivers[45]. Even humans who have been exposed to such compounds faced health issues such as endocrine disruption, genotoxicity, neurotoxicity and carcinogenesis [44].

To reduce such adversity, to help biodegrade or catalyze these hazardous chemical wastes into harmless compounds and to detect its residues in soil and water, the use of QDs is emerging as a promising alternative to the traditional techniques such as traditional gas or liquid chromatography (GC/LC) -mass

spectroscopy (-MS) techniques (Stan and Linkerhägner, 1996; Sicbaldi et al., 1997; Balinova et al., 2007). The latter techniques although reliable and accurate involve a lot of time-consuming steps and skilled manpower.

Considering more than 10,000 samples analyzed by the U.S. Department of Agriculture (USDA), nanosensors can offer a higher sampling rate and overall lower cost for the whole processing[40].

Ung, Thi Dieu Thuy, et al (2012) with the help of Fluorescence biosensors fabricated from CdTe/CdS or CdSe/ZnSe/ZnS QDs and the AChE enzymes, specifically detected PM pesticide at content as low as 0.05 ppm[41].

Methodologies on direct use of QDs [42]

Quantum dots undergo dynamic quenching of their fluorescence upon direct reaction with their analyte. This quenching principle. Doxycycline residues in honey were successfully identified with the aid of the quenching degree investigation. The detection was based on the change of fluorescence intensity.

When docetaxel interacts with CdTe QDs modified with thioglycolic acid, the electrons move from the QDs' to the molecules of doxycycline, causing the QDs' fluorescence to be dynamically quenched.

According to Durán, G. M.; Contento, A. M.; Ríos, Á (2013), the paraquat herbicide was determined in tap water, mineral water, wastewater, and groundwater using the same fluorescence quenching principle.

The capacity of QDs to enhance the luminescence of some chemiluminescence systems further expands their range.

The transfer of an excited electron from energy donor to a receptor through non-radioactive energy transfer process provided, the FRET process is when the wavelength difference between the donor's emission and the receptor's absorption is less than 10 nm.

This process with QDs has proven to detect pesticide residues efficiently. The size of the QDs during synthesis can be changed to maximise the overlap between their emission and absorption wavelengths of the receptor, further improving the system. An expeditious method for the detection of organic phosphorothioate pesticides was developed by Zhang et al. The technique made use of CdTe QDs, which fluoresced green at 520 nm. These were subsequently coupled with dithizone, which in an alkaline media had a maximum absorption wavelength of 510 nm. Dithizone instantly quenches the fluorescence of CdTe QDs because of the FRET system's action.

The hydrolysis products of the organic phosphorothioate insecticide replaced dithizone on the surface of CdTe QDs, causing the QDs to regain their fluorescence. Phosphorothioate pesticides can be detected by this "on-off" very sensitive technique without the requirement for costly antibodies or enzymes.

Chen et al. When CdTe QDs were added as fluorescent background material to the background electrolyte of capillary electrophoresis, the residues of veterinary drugs and pesticides were detected using the inverted peak height of analytes. This type of approach demonstrated the ability to effectively identify three types of nicotine insecticides, three types of strobilurin fungicides, and five types of quinolone antibiotics in milk and honey. Pesticide residue detection has also made extensive use of QD Fluorescence Immunoassay (QD-FLISA), a technique that uses a complex probe conjugating QDs with antibodies or antigens.

Herbicide 2,4-D detection by direct competition Vinayaka et al. developed the QD-FLISA method.

Chen et al. reported using two indirectly competitive QD-FLISA methods in the same year to detect chlorpyrifos residual in drinking water. Among the fluorescent nanoparticles, graphene quantum dots (GQDs), polymer dots, and carbon nanodots are classified as carbon nanoparticles (QDs). Compared to semiconductor QDs, carbon QDs have distinct fluorescence properties and the benefit of low toxicity and good biocompatibility [46].

Nan et al. employed graphene quantum dots (GQDs) in combination with the enzyme AChE to detect the highly lethal organophosphate pesticide paraxon at concentrations as low as 0.2 nM[47].

Various pesticides (glyphosate, methomyl, l-cyhalothrin and parathion-methyl) have also been detected in water by different research groups through the use of fluorescence sensors based on silica (QDSi) nanoparticles[62]-[66].

Quantum dot	Recognition element	Detection technique	Analyte(s)	LOD	Linear range	Matrix	Ref.
CdTe/CdS		Laser Induced Fluorescence	Mevinphos	0.16 mg kg ⁻¹	0.2e28.0 mg kg ⁻¹	Tomatoes	[48]
			Methidathion	0.05 mg kg ⁻¹	0.1e20.0 mg kg ⁻¹		
			Diazinon	0.18 mg kg ⁻¹	0.3e18.0 mg kg ⁻¹		
			Phosalone	0.12 mg kg ⁻¹	0.2e30.0 mg kg ⁻¹		
C [4]/SiO ₂ /CdTe	Calix [4]arene	Fluorescence	Methomyl	80 nM	0.1e50 mM	Water	[49]
C [6]/SiO ₂ /CdTe	Calix [6]arene	Fluorescence	Glyphosate	0.0725 nM	1.0e25.0 nM	Water	[50]
TGA-CdTe	p-sulfonatocalix [4] arene	Luminescence	Fenamithion Acetamiprid	12 nM 34 nM	0e10 ⁴ M 0e10 ³ M	Garlic samples, Aqueous solution	[51]
ZnCdSe films	-	Fluorescence	Carbaryl	<12.4 nM	12.4e12,400 nM	Water	[52]
Graphene-CdS	-	Electrochemiluminescence	Pentachlorophenol	0.0113 nM	37 pMe1.9 mM	Tap water, River water	[53]
GSH-CdS	-	Fluorescence	Dicofol	55 ± 11 ppb	3.32e62.5 mM	Methanol	[54]
RF-CdTe@SiO ₂ -AuNPs	Trypsin/protamine	Fluorescence	Parathion-methyl	0.0684 nM	0.15e1500 nM	Tap water, milk, rice	[55]
MPA-CdTe	-	Fluorescence	Chlorpyrifos	0.1 nM	0.1e10,000 nM	Apples	[56]
TGA-CdTe/CS-AuNPs	-	Fluorescence	Glyphosate	9.8 ng kg ⁻¹	0.02e2.0 mg kg ⁻¹	Apples	[57]
CdTe/CdS	-	Fluorescence	Paraquat	6.39 nM	9.90e1500 nM	Rice, Wheat flour, Lettuce leaves,	[58]

						Waste water	
(3-MPA)- CdSe/ZnS	-	Fluorescence	Paraquat	0.0117 nM	0.039e19 nM	Water	[59]
Double QDs (ZnCdSe and CdSe)	-	Fluorescence	Paraquat Dursban Dipterex	20 nM	2 10 ⁸ e5 mM 5 10 ⁸ e3 mM	Wastewater, Tea	[60]
CDs/AgNPs	-	Fluorescence	Methyl thiophanate Cartap Glyphosate	 12 ng mL ⁻¹	2 10 ⁸ e5 mM 5 10 ⁸ e5 mM 0.025e25 mg mL ⁻¹	Rice, millet, wheat flour, and maize flour	[61]

2. ENHANCEMENT OF PHOTOSYNTHESIS PRODUCTIVITY AND GROWTH RATE

Only 10% of the sunlight is utilized by chloroplast for photosynthesis since the light trapping pigment use restricted wavelengths. Improvement in solar energy utilisation is extremely crucial if one wants to increase plant yield. An artificial photosynthesis hybrid system that uses CdTe QDs by resonance energy transfer between QDs and phytochrome has been reported [67][68][69][70].

A recent research highlighted that a new mexico based nanotech company-UbiQD has brought forth fibre coupled luminescent concentrated technology for optimization of light in controlled environments. These quantum dot-based luminescent fibres open huge potential in case of vertical farming, where harvesting sunlight and optimally transmitting that energy to multiple layers of plants has been made possible providing a better alternative of expensive artificial lighting.

Similar application was used by Jin-Won Song (2016) on quantum dots as phosphor to fabricate LEDs. His experiment confirmed to the possibility that these multi-wavelength QDs can be used as a lightening source as they enhanced the growth average of the flora[73].

An experiment carried out to measure carbohydrate content in mung bean sprouts leaves studied the influence of CDs and photosynthesis. Studies showed an increased carbohydrate content as compared to the control in the mung bean sprouts treated with CDs. This enhanced productivity of carbohydrates in plants indicates the improved efficiency of plant photosynthesis.[71].

A similar study was done to observe the effects of CDs on the growth of rice plants by Hao et al. The yield was found to be improved by 14.8%. Furthermore, CDs degrade to form CO₂ and other plant-hormone analogues which is then converted to carbohydrates (through Calvin cycle of photosynthesis) and promote plant growth respectively[72].

3. DETECTION OF DISEASES IN PLANT

The use of QDs have been applied in the designing of phytopathogen detection devices that are equipped with smart sensing capabilities to avoid the annual loss of 20–30 % in crop production. This technology has proved to be an effective replacement of the traditional time consuming, labor-intensive, costly methods[75].

Qd-labeled McAb and PcAb (immobilized on microspheres) aided fluorescent immunoassay was used by Mingqiang et al. for detection of Lily symptomless virus (LSV). The results of this experiment coupled with flow cytometry were 16 times more sensitive than traditional assays, such as ELISA[74].

Beet necrotic yellow vein virus is transmitted by the plasmodiophorid *Polymyxa betae*. The detection of this vector has been done by conjugating QDs with specific antibodies against GST for the development of a FRET based QD-Ab sensor. This method has been proven to be a rapid, accurate, and cost-effective immunoassay[76]. The witches' broom disease of lime, caused by *Candidatus Phytoplasma aurantifolia* has also been studied for detection by using QDs[77]. A FRET based optical DNA biosensor has been developed for the detection of a specific *Ganoderma boninense* DNA sequence. What makes QDs even more appealing is its capability for on-site detection on excitation by UV light through visible fluorescence[79]. Another FRET based complex biosensor was used to probe Citrus tristeza virus (CTV) in the study conducted by Safarnejad et al[80]. Hao et al. conducted a series of experiment on rice plants with the use of carbon dots. The results suggested that the CD treatment enhanced the disease-resistance ability of the plants by inducing the over-expression of the thionin gene [79].

A paper-based, fluorescent Tb(III)-CD probe fabricated by Huang et al. was used for the detection of 3'-diphosphate-5'-diphosphate- the stringent of plants in response to extreme environmental condition[81].

4. PRECISION FARMING

Precision farming can be explained as, the use of computers, GIS, GPS and remote sensing devices to measure the local and sensitive environmental conditions, identifying the best suitable methods for the growth of crops and its protection to produce the maximum yield. In easier words, maximizing the output (crop yield) through minimum input (i.e. fertilizers, pesticides, herbicides, labor requirement etc.)[78].

The use of QDs through vast platform has proved to be a boon in application of precision farming. The used areas can be stated as follows:

- a) Nano pesticides - the use of QDs for the development of nano pesticides have paved a great alternative to the use of potential toxic and significantly economic burden posing traditional chemical pesticides. Disease Management aided by on-demand agrochemicals can be made possible by Surface-functionalized QDs .
- b) For the growth and improvement of plants (as we have already discussed above).
- c) Utilization of nano-arrays in agriculture – QDs - ZnS, CdSe, CdS can be used as a fluorophores.QDs are also used as immuno-labelsby Muller et al.
- d) Improvement of crop yield - Late et al.2015 reported the use of graphene quantum dots where *Allium sativum* and *Coriandrum Sativum* seeds were treated with 0.2 mg/mL of graphene quantum dots for 3 h before planting. Enhanced growth rate was exhibited in both plants.

Hence precision agriculture can aid in monitoring plant growth, detecting plant diseases, increasing global food production and quality along with promising scopes for sustainable agriculture by reducing waste[83].

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