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Forensic Application of Quantum Dots: An Overview

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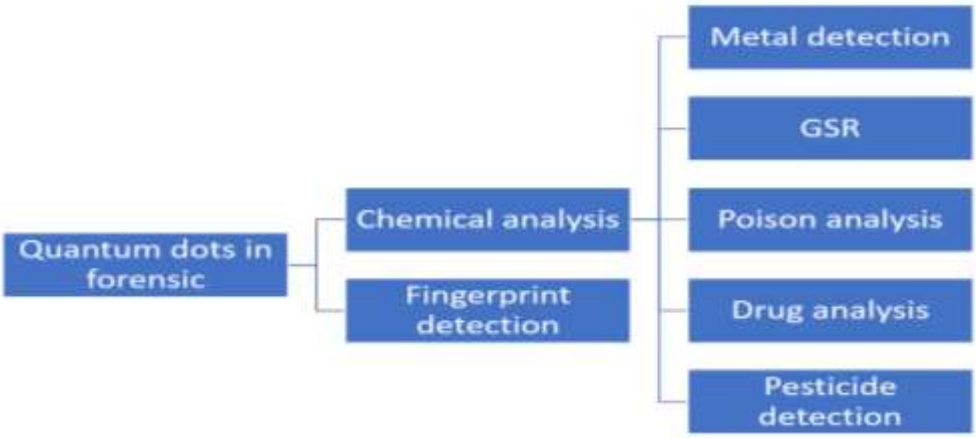
Abstract
Quantum dots (QD) are nanoparticles specially semiconductors with the photoluminescence (PL) property with a variable wavelength. These ranges from the scale of 1-6 nm that can subsist in a liquid or a solution with a constancy state. This photoluminescence or fluorescence is produced due to their surface defects caused on the surface of the QD which results in the absorbance of light from sources of different wavelength. Those tuneable wavelengths of the photoluminescence can be influenced by their shape, size and structure of the QD. The transitional optical properties of QD helps in generating sensors in the field of instrumental analytical chemistry, biosensors to mutual interaction with analytes. These QD can be manufactured through possible colloidal synthesis similar to handed down chemical processes and electrical gating. The high extinction coefficient and incredibly quick optical nonlinearities contributes a high influence in optical properties of QD. More over the QD produced with high efficiency and stability is used for the research purposes which in turn provides a quite narrow emission spectrum, where the size of is directionally proportional to wavelength. The high compatibility, adjustable PL and increased photo stability prioritise the advantage of chemical analysis.

1. Introduction

Quantum dots (QD) are nanoparticles specially semiconductors with the photoluminescence (PL) property with a variable wavelength.[1] These ranges from the scale of 1-6 nm that can subsist in a liquid or a solution with a constancy state.[2] This photoluminescence or fluorescence is produced due to their surface defects caused on the surface of the QD which results in the absorbance of light from sources of different wavelength.[3] Those tuneable wavelengths of the photoluminescence can be influenced by their shape, size and structure of the QD. The transitional optical properties of QD helps in generating sensors in the field of instrumental analytical chemistry, biosensors to mutual interaction with analytes.[4] These QD can be manufactured through possible colloidal synthesis similar to handed down chemical processes and electrical gating. The high extinction coefficient and incredibly quick optical nonlinearities contributes a high influence in optical properties of QD.[4] More over the QD produced with high efficiency and stability is used for the research purposes which in turn provides a quite narrow emission spectrum, where the size of is directionally proportional to wavelength. The high compatibility, adjustable PL and increased photo stability prioritise the advantage of chemical analysis.[5]
As the QD are the unconventional class of inorganic resorcinolphthalein they are briskly put into practise in the arising technologies. When the visible light containing photons hits the semiconductor, the electron gets excited and jump to the higher level. The energy photons are released while returned back to their ground state with a particular emission frequency. The procedure of luminescence interchange of energy

in which radiative charge is carried among the light detecting molecules, an excited donor and the other receiving one. The continuous transfer of energy results in the FL enhancement.[6]

The quantum dots have an extensive window in forensic science as the binding and fluorescence capability of this nano particle can provide much possibilities in the main branches of the subjects like toxicology, biology, fingerprinting etc. And forensic science is the branch of science that welcome the emerging technologies which have higher capacity, accuracy and efficiency in solving crime so that more accuracy provides much deeper hints to the cases.

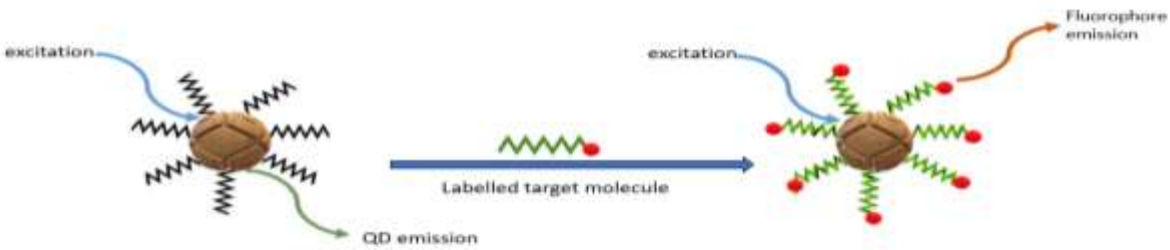


2. Chemical Analysis

The use of quantum dots in the field of chemical analysis can bring a wide range of new perspectives in the detection and analysis procedures.[7] This further indeed can replace the existing traditional technologies in the stage of metal detection, drug analysis, poison analysis, pesticide detection etc. This further provides a newer and faster identification and analysis techniques in the background of forensic toxicology. These days the cases related to forensic toxicology are increasing day by day which seeks for more fast and accurate procedures in examination. This may be due to the uncomplicated availability of chemicals, which also prompts the experts to detect and differentiate the components used in the toxicological cases makes difficult.

In the initial stage quantum dots were put forward as luminescence labels for biological substances, later on new applications on chemistry and forensic toxicology was formulated where the photoluminescence can bring up the reducing use of other chemical binders which can undoubtedly provide much accurate and better results.

The attack of targeted labelled molecule enhancing fluorophore emission.



2.1 Metal Detection

Metal detection is one of the major technical analysis part in the field of forensic science specially in the toxicological division. Heavy metals can be indeed described as the toxic metallic substances or elements that has higher density, specific gravity and atomic weight. The toxic metallic substances like mercury, cadmium, arsenic, lead, thallium etc contribute to the group of heavy metals analysed commonly in the toxicological laboratories. The detection of heavy metals were done by the conformist methods of spectroscopic techniques like AAS, ICP- MS and AFS, followed by electrochemical voltammetry techniques and optical techniques likes XRF and LIBS. The limited ability to distinguish the isotopes along with expensive instrumentation and the requirement of skilled operators and sample vapourization are the few demerits of spectroscopic techniques in the detection of heavy metals. In case of optical techniques there are chances of sufferings from matrix effect which cant be corrected easily. This even seeks for a new method like quantum dots for the detection of heavy metal to make it easy, less time consuming and cost-efficient.[8]

As quantum dots are zero dimensional nano particles, they have distinctive optical properties which enhance the fluorescence properties with broad expansion spectrum which can even provide narrow emission and photo stability. The charge or the energy transfer in the quantum dots helps in detecting the heavy metals which can even be possible with the ligand exchange or transfer. The optical properties like fluorescence intensity, wavelength shift, fluorescence quenching changes while in the interaction with ions. Because of the strong and tunable fluorescence properties CdSe and ZnS quantum dots are used commonly for the detection and analysis. Even PbSe and PbS quantum dots are used for the detection in the conditions where quantum dots are sensitive to infrared regions. When the presence of heavy metals affect the energy transfer efficiency which leads to alter the fluorescence signals quantum dots are used as acceptor or donors in FRET.[9]

The detection of heavy metals in the cases of drowning is common in toxicological analysis. If the drowning occurred in a polluted environment there are more chances for the presence of metallic toxins like Pb, Hg, Cd or As. This can even provide the details regarding the location of the environment where drowning actually occurred.[10]

2.2 GSR

When a gun is triggered out, along with the bullet there will be some other particles like smoke, sooth, unburned or partially burned substances, semi burned ashes and dust. These gradually increases the chances of identification of the weapon, type of injury, the range of fire etc in the forensic ballistic division as to conclude justice in the criminal cases.[11,12] GSR is the gun shot residue that has been protruded out while shooting which can be collected from the barrel, surface of weapon contact, even in the victim body or surfaces. It can give a clear cut shot idea about the types of powder used in the weapon too.

The analysis of gunshot residue is done through the traditional method of detecting the presence of lead, barium, antimony that is considered unique about their combination while formation of GSR.[12,13] But this even cant alone provide details regarding the residue as the detection of both lead- barium combination is not individualised for the presence of GSR. SEM- EDS is the primary instrumental technique used for the detection of GSR as the morphological resemblance of the particles in GSR are detected in EDS. Even this provides good results in GSR detection, the expenditure in the instrumentation and the amount of time consuming in targeting each sample marks a major drawback. The continuous exposure of ammunition other than lead can reduce the characteristics features of SEM- EDS.[12,114] Another instrumentation techniques that are commonly used in detection of GSR are mass spectrometry, Ion mobility spectrometry, liquid chromatography mass spectrometry etc. In some cases the high setup cost and the bulky bench bound machineries also marks the requirement of new innovative techniques for the detection of GSR.

The use of graphene quantum dots remarked the higher sensitivity and selectivity in fluorescence sensor and this can the less particle size reveals the specificity in detecting the GSR particles. [16]

2.3 Drug analysis

The drugs which can in term called as chemical substances are detected from various parts of the body and body fluids, such as over dosed poisoning results in extraction of poison from vomit, stomach contents, urine, liver, blood, hair samples etc and this may vary according to the mode of administration and how metabolism took place. The modern drug detection methods are GC-MS, LCMS based technologies. These detection methods give quick to quick results but its limit of detection is less than 1 ppb.[17]

The 2019 World Drug Report of UNDOC, in the domain of stimulants, amphetamine and cocaine have a wide space, as an effect their identification and detection are becoming significant to restrict their exploitation. TLC, GC, HPLC were the common anesthetics used for the papers of these drugs.[18] Even though they provide exceptional accurateness their time consuming process and procedure remarks them bit problematic. These portrayal apparatuses are very classy and repeatedly necessitate a longer detection cycle to complete an accurate measurement. Sometimes the immediate detection may be also difficult for the large-scale instruments. [19,20]

The detection and identification of local drugs which are abused such as cocaine, codeine, heroine etc are having proved QDs based sensor system such as CQDs in aqueous solution and function papers which can in term known as CQDFPs. But in case of morphine, the detection in visual limit was 0.01mg/L.[14]

The paracetamol oxidation in the incidence of an enzyme like thirosinase for the formation of N- acetyl-p-benzoquine which interact strongly through a biosensor based polypyrrole with GQDs to efficaciously snuff out the fluorescence over electron transfer.[21] Supplementary QD based sensors are N doped GQD, NGQDs, BiOBr etc with the detection maximum of 3.33nM and 1.6 nm. The photoluminescent CdTe quantum dots with a size around 5 nm utilising mercaptosuccinic acids as a stabilising agent. The fluorescence quenching mechanism by these quantum dots are used to detect the presence of Aspirin.[22] The CdTe quantum dots have a fluorescence vigour with aspirin concentrations ranging from 1.7 to 56 μmol/L. In case of ibuprofen detection, an optosensor accompanying CdTe QDs which is overlayed by non-ionic alginate with a detection boundary of 0.004 μmol/L. But the existence of ibuprofen can sometimes leads to the decrease in the fluorescence intensity.[23]

As the number of diseases are increasing the urge to develop more medicines which gives speedy relief was also increasing. The use of antibiotics was worth enough in the medical field to supress much of the diseases. The chromatographic techniques like HPLC, LC-MS, and ELISA were used for the detection of antibiotics. But presently the importance of chemo and bio sensors increased as they are using for the purpose of detecting antibiotics. The most excellent choice for detecting specific enoxacins from biological fluids is the Mn doped ZnS QDs capped with L-cysteine. This QD has a detection limit upto 58.6 nm and their phosphorescence emission apex is 590 nm.[24]

2.4 Poison Analysis

The extraction and purification of poisons from the complex biological samples are the key procedure in the process of poison detection in forensic toxicology laboratories rather than the other chemical analysis.[25,26] The detection and identification of poisons urge is developing day by day as the cases related to toxicology is also increasing. Even though the availability of some specific poisons are restricted to an extend still the people started preparing or exposed to more toxic mixture poisonous substances which will enhance the toxicity in the body on the other hand make it difficult for the experts to identify. The poisons which marks under hydrogen sulphide, carbon monoxide, cyanide are the most important classes in toxicological analysis[26,27] In the normal conditions, diffusion even in the absence of mechanical agitation and chromatogram like GC can conclude the test with an accurate result of the test determination.

The consolidation of screening test and specific chromatograph technique were the most accurate method used in the field of detection of gaseous poison.[18] Lead acetate or silver paper test were used to detect the gaseous toxics like hydrogen sulphide (H_2S). Eventually the QDs were able to replace them as chemical sensors.

In forensic toxicology, the extraction and purification of poisons from complex biological samples are critical procedures, often prioritized over other chemical analyses. As the number of toxicology cases increases, the demand for precise detection and identification of poisons is becoming more urgent. Despite restrictions on certain toxic substances, there has been a concerning trend of individuals creating or using more toxic mixtures. These mixtures not only enhance the level of toxicity in the body but also make it increasingly challenging for experts to identify the specific poisons involved.

2.4 Pesticide Detection

Pesticides are made of different chemical constituents which are also known as economic poisons which are majorly classified into four: organochlorine, organophosphorus, carbamates and pyrethroids.[28] Conventional methods of distillation and TLC are used for extraction and identification of pesticides respectively. Precise molecules can functionalise with quantum dots which include enzymes or antibodies which have the properties to bind selectively to particular molecules of pesticides. The energy transfer or charge transfer helps the pesticide molecules to quench with the fluorescent molecules of quantum dots. The quantitative analysis is done by measuring the degree of quenching and the proportionality of concentration of the pesticides.

Organophosphorus compounds (OPCs) and carbamates are extensively familiar for their part as potent inhibitors of acetylcholinesterase (AChE), an enzyme critical for nerve function. OPCs, which contain esters of phosphoric and phosphonic acids, performance as irretrievable inhibitors of AChE, leading to lengthy and hypothetically lethal inhibition of the enzyme. Carbamates, conversely, are reconstructable inhibitors, permitting for provinsional inhibition of AChE with a certain of continuing toxicity.[29]

One of the noteworthy toxicological procedures linking OPCs is the metabolic conversion into more active inhibitors. Phosphorothionate compounds experience metabolism to produce corresponding oxones, which are highly active AChE inhibitors.[30] A familiar occurance is the pesticide parathion, that has been metabolized in the liver to formulate paraoxon, a vigorous AChE inhibitor, and latterly to p-nitrophenol. The active inhibitory forms of OPCs which is characterise by the recognition of these oxones are performed with the help of N- bromosuccinamide (NBS) spray or bromine vapour, accompanied by cholinesterase (ChE) spray, that enhance the detection of the inhibited enzymes. Even though they are effective methods but have particular confines, specially as of in the cases of sensitivity and specificity.[29]

In the current years, the progress in the discovery of OPCs and carbamates aided the expansion of QD based biosensors. The desirable properties of quantum dots enhances the quality in designing biosensors like the high optical fluorescence, tunable emitting property, varying size etc. Even these increases their specificity to the analytes that to be targeted with high sensitivity due to the presence of more layers of recognition in bio elements, like that of enzymes or antibodies. The QD based fluorescent probe which was used for the detection of OPCs and carbamates can be utilised CdTe for the fluorescence which is enhanced by multiple layers of bienzymes, particularly acetylcholinesterase and choline oxidase. [30]

These bi-enzymes serve as biological receptors that interact with OPCs and carbamates. The detection mechanism is based on the inhibitor-enzyme interaction, where the sequential enzymatic reactions produce hydrogen peroxide (H2O2), a compound with a strong ability to quench the fluorescence of the CdTe QDs.[31] The indication of the OPC, somehow retards the active involvement of AChE, and as a result stops

the creation of H_2O_2. This results, the fluorescence slacking of the quantum dots is compacted, in which the measurement of OPCs concentration in a sample is determined. Paraoxon, dichlorvos, and parathion at picomolar level detection is remarkably sensitive by these biosensors. This high sensitivity remakes its effectiveness in looking for trace quantity of toxic compounds in biological and environmental samples.[31]
In a similar study, a nanostructured thin film was generated by placing layer by layer technique, utalising CdTe quantum dots and AChE as the enzymes. These biosensors were generated to work on the code of competitive inhibition, where the substrate acetylthiocholine strives with OPCs for the active site of AChE. In the presence of OPCs, they hider the activity of enzymes, which retards the conversion of acetylthiocholine to thiocholine, which can sometimes leads to the reduced fluorescence quenching by CdTe QDs.[31] The notable property of these biosensors is the sensitivity with a detection limit, highlighting the potentiality of detecting the substances at their extreme low concentration.
The customisation of biosensors according to the nature of target is another crucial characteristic of quantum dots in a wide range of analytes pasts OPCs and that of carbamates. The alteration of the surface chemistry of QDs and merging various elements of biological recognition, these biosensors can be modified for the detection of various toxins or toxic substances and various pathogens including some harmful substances too.
The naturally occurring insecticidal compound like pyrethrins which contain ester that is evolved from stereoisomers of chrysanthemic and pyrethric acid are comparatively low toxic in nature and photochemically not so stable as compared to their synthetic similarities, which is inturn commonly called pyrethroids.[28] As pyrethroids are the commonly used pesticides which is commercially used can metabolise the body to form phenoxybenzyl alcohol or results in the formation of 3-phenoxybenzaldehyde which enhances the detection and binding properties of QDs to emit larger detection and identification quality.[28]

PESTICIDE	LINEARITY	LOD	Sensitive QD
Malathion	0.908- 303nM	303pM	CdSe
Carbaryl	0-14 µg/ mL	0.12ng/mL	CdTe
Imidacloprid	5-4000nM	0.823nM	GQD
Organophosphorous	0.0001-160µg/mL	96.7 µg/mL	CdSe/ ZnS
Acephate	0-100ng/mL	0.052ppb	CQD
Glyphosate	0-1000nM	2pM	CdTe/ CQD

Pesticides and its sensitive QD along with their linearity.[29]
Simple chromatography techniques along with gas chromatography- flame ionisation detection and gas chromatography – mass spectrometry were the conventional methods of detecting pyrethroids and pyrethrins. The time consuming and requirement of sophisticated equipment gave much draw back which can overwrite its advantage of effectiveness.

3. Fingerprint detection

Fingerprint is a vital part of forensic science. As the palmer surface is covered with a layer of skin with individualised ridge pattern for the purpose of holding stuffs and to provide grips is the fingerprint or ridge pattern. This not only provide firmer grip for holding but also gives the unique identity for an individual. The development of latent fingerprint starts from simple dusting methods. As the significance of developing fingerprint was increasing day by day results in the formation of chemical methods like

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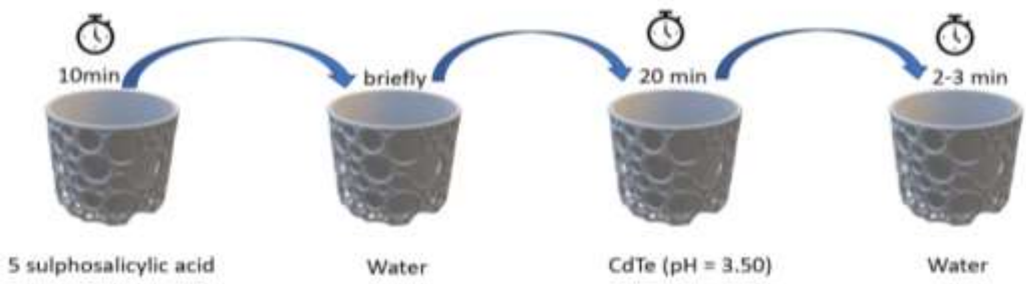
ninhydrin and silver nitrate methods. The gradual technology replaced these with laser technology for the development of fingerprint from different surfaces.[31]

The biological residues like sebum, amino acids, lipids which are the fingerprint components left on the surface can be binded with specific components of quantum dots. The enhanced sensitivity with high fluorescence intensity provides quantum dots make the latent fingerprint even the most faded into a clear visible print.[31] This also helps in identifying finger marks from multi coloured surfaces without considering the nature of the surface. i.e., it is independent of the nature of the surface and background. The specificity of targeting particular chemical compound augment the detection of lipid components.[31]

The effective interaction occurs with the fingerprint residue when the surface is dry and clean. The grave part is the selection of quantum dots for each surface. The cadmium selenide, cadmium sulfide, or indium phosphide, are the major quantum dots with different emission properties. The major benefit is the flexibility of quantum dots with specific emission wavelength varying from ultraviolet to infrared which enables the experts to choose the quantum dots according to contrast to the background and the lighting set up of the examination. As the fingerprint composed of sweat which is combination of water, sodium chloride and some organic compounds like urea and amino acids along with tryglyceride and wax ester sebum and proteins with amino acids provides multiple binding sites for quantum dots. The modification of surface of QDs in functionalization helps to acquaint with specific ligand or molecules which bind with the latent fingerprint. Some chemical groups like thiols and amines can form electrostatic and covalent bonds with the contents of fingerprint residue like proteins and amino acids. Antibodies and peptides can be conjugated with QDs that specifically bond with sweat which contain protein and biomolecule. Even the functional groups of carboxyl and hydroxyl group enhance the binding with sweat contents.

The fingermark detection experimental protocol of Andy Becue et.al., using quantum dots are as follows (1) The specimen is immersed in 5 sulfosalicylic acid in water for 10 min; (2) Brief rinsing in water (3) QD staining solution is used to immerse the specimen for 20 min using a rotating platform for stirring. (4) The specimen is again rinsed in water and observed luminescence probe using mini crimescope under a UV wavelength of 300-400 nm.[33]



Schematic representation of fingermark detection protocol using QD

Electrostatic interactions, covalent bonding, hydrophobic interactions and the Van der Waals force are the major mechanisms which enhance binding. Spraying, brushing and dipping are the major methods of developing fingerprint from different surfaces. Brushing is done normally similar to traditional dusting method. This developing method along with the mechanism of binding makes it quick and selectively stick into the latent ridge marks of fingerprint. The fluorescence imaging helps in the better visualisation of the fingerprint when they got excited when light is emitted with a higher contrast.

TYPE OF QUANTUM DOTS	USES	TARGETED SUBSTANCE
CdSe, ZnS	Heavy metal detection	Mercury, lead, cadmium, copper
PbSe, PbS	Heavy metal detection in IR region	
GQD	GSR	Lead, antimony, barium
GQD, NGQD	Drug detection	paracetamol
CdTe	Drug identification	Aspirin, ibuprofen
ZnS	Antibiotics detection	Enoxacins
CdTe, CdSe, ZnS, CQD, CIS, SiQD	Fingerprint identification	Fingerprint residue

4. Summary

The study revealed the viability of quantum dots in the field of forensic science. This new technique appears to work particularly well for chemical analysis of drugs, pesticides and metals. The luminescence property of this nanoparticle can be brought up to further new examinations which can create much more detection propertied that chemicals luminescence cant be exhibited by every materials. Although the implementation of suitable and appropriate instruments are existing still the technologies like application of quantum dots in forensic science laboratory can further increases the efficiency of the tests and cases results which can promote the acceptance of the innovative, simpler techniques. The betterment use of the quantum dots in forensic science field may result in incorrect interpretation on the performance of this newly explored method.

The varying properties of QDs, which comprises the tunable fluorescence, comparatively higher sensitivity, and the wide emission spectra, which built the best sensor with better sensing applications. The paper analysis the different types of quantum dots in particular on cadmium, Carbon, zinc, silicate, graphene which pinnacle their benefits in detection and analysis in the field of forensic science. Even detailed explanations are discussed on the alteration of QDs for the better performance and increase their ability to target and detect the peculiars which comprises the detection and analysis of drugs, pesticides, heavy metals, poisons and fingerprint identification.

The quantum dots are the eco friendly, low toxic as compared to other chemicals with good bio compatibility and comparatively very low cost. The use of QDs are simple, transportable and for reduced testing period its generated with pre detection tool which can intact reduce the laboratory and instrument cost. This makes it to stand different. Consequently, as a whole this review may give supplementary references to the exploration in the operational visualization method.

This review also enlighten the encounters related to the usage of quantum dots, specially their possibilities toxicities and environmental concerns. The paper deliberate the ongoing developments in generating safe and eco friendly, non hazardous quantum dots which can in deed perform according to the needs and norms. In a nutshell the paper represents the applications and significance of quantum dots that is effective and potential enough to revolutionise the field of forensic science with its rapid sensitive, highlighting properties in detection and identification in various fields.

5. Future Perspective

The further development in the field of research in quantum dot applications in forensic science can prevail the progress of non toxic quantum dots and focussing on the manufacturing of these QDs, eco friendly with bio compactable capacity can be further used for the safer application in monitoring. The use and necessity of field deployed devices and portable devices are increasing day by day as the urge to have an integrated, safe and easy to use devices are increasing in detection, identification and analysis stages respectively. The integrated QD based sensors with above mentioned devices can emerge a revolution in the toxicological analysis like on site pesticide detection etc.

The advanced development and functionalisation techniques can improve the specificity in sensors of QD and this can results in minimising the false positives and can enhance the reliability in the detection. The technical advancement in the fluorophore emission can in turn bring the easy detection of the substrate with high fluorescence capacity and ready to avail identification system specially in toxicological and fingerprint identification.

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