



Review

An overview of recent advances in the application of quantum dots as luminescent probes to inorganic-trace analysis

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ABSTRACT

Systems based on quantum dots (QDs) have received great attention in recent years in the field of analytical chemistry for the detection of a variety of analytes. In this overview, we review novel methods based on the employment of QDs for inorganic-trace analysis, making use of liquid-phase and solid-phase systems, reversible and even visual and multiplexing analysis. Also, we discuss critical parameters influencing the optical properties of QDs, and the sensitivity and the selectivity of these systems. We present representative examples of QD-based systems and the most promising directions for further investigations.

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Abbreviations: APDC, Ammonium pyrrolidinedithiocarbamate; CL, Chemiluminescence; ECL, Electrochemiluminescence; FRET, Fluorescence resonance-energy transfer; GSH, Glutathione; HDA, Hexadecylamine; IDA, Iminodiacetic acid; ITO, Indium-tin oxide; LbL, Layer-by-layer; MAA, Mercaptoacetic acid; MHA, Mercaptohexadecanoic acid; MPA, Mercaptopropionic acid; MSA, Mercaptosuccinic acid; NIR, Near-infrared; PEG, Polyethylene glycol; QD, Quantum dot; QY, Quantum yield; TOPO, Tri-n-octylphosphine oxide.

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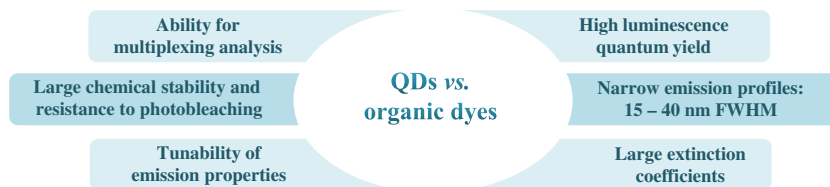


Fig. 1. Summary of the most relevant optical properties of QDs.

1. Introduction

In recent years, semiconductor nanoparticles (NPs), also known as quantum dots (QDs), have emerged as luminescent probes for the detection of a variety of analytes, including many biological, organic and inorganic species [1–4].

QDs exhibit analytical performance superior to conventional organic fluorophores due to their better chemical and photoluminescence stability, and their superior optical properties [5]. Fig. 1 summarizes the most relevant properties of QDs, which make them attractive for optical sensing.

One of the most important problems of organic dyes lies in the difficulty of applying them in multiplexing analysis. By contrast, QDs are suitable for multiplexing analysis, since it is possible to tune the emission of QDs in the ultraviolet (UV) to near-infrared (NIR) spectral region by changing the size, the morphology, or the composition, or even using different preparation procedures [5], and they can be simultaneously excited by a single source when multiple colors are used together. Moreover, QDs provide narrow emission bands with width at half-maximum of ~15–40 nm and sufficiently separated to allow individual detection for multiplexing analysis.

In 1993, Murray et al. [6] developed a relatively simple procedure for synthesizing high-quality CdSe QDs in organic media. From that time, efforts were focused on developing new methods to solubilize QDs in aqueous media, so that they could be used for biosensing and imaging applications. In 1998, this problem was overcome by capping the surface of QDs in order to impart water solubility, as proposed by Alivisatos [7] and Nie [8]. As shown in Fig. 2, most of the QD-based systems focused on bioanalytical applications, but, since 2002, the development of QD-based systems to detect ions has increased markedly.

There has been a significant increase in papers concerning the use of QD-based systems in the past five years. The topic was also addressed in review articles from different perspectives. Recent advances and novel sensing systems were described by Ma et al. [9]. Freeman et al. [2], summarized different methods for functionalizing QDs with different capping ligands in order to use them for sensing low-molecular-weight substrates, metal ions, anions, and

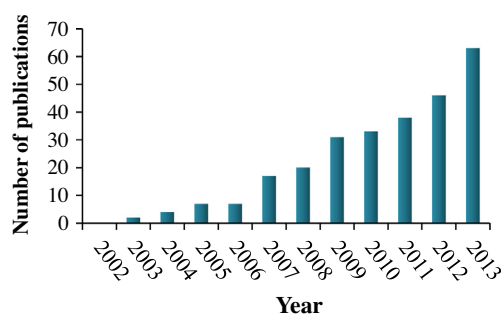


Fig. 2. Evolution of the number of publications concerning the use of QDs for inorganic-trace analysis.

gases. Galian et al. [3] discussed the main mechanisms of interaction between QDs and organic molecules, such as ligand-analyte, bioconjugate-analyte and host-guest, using QDs functionalized with different capping ligands. In 2012, Frigerio et al. [10] summarized the applications of QDs in automated chemical analysis.

Also, different authors published overviews of bioanalytical applications of QDs [11–13]. However, there has been no specific review on the use of QDs for inorganic-trace analysis.

In this article, we present an overview of QD-based systems for inorganic-trace analysis. We discuss most important parameters affecting such systems and outline recent advances in this field.

2. Critical parameters for the development of QD probes for inorganic-trace analysis

Several experimental parameters could affect the stability of QD-based assays for sensing inorganic ions as well as sensitivity and selectivity. Among the most relevant are the functionalization of the QD surface, pH of the dispersant media, or the composition of the QD structure. We briefly discuss these factors below.

2.1. Surface modification and functionalization

One of the main problems encountered in the development of QD-based systems is the selection of a suitable capping ligand in order to achieve adequate solubility of QDs in the dispersion media and to improve the selectivity and the sensitivity of these systems.

Generally, during their high-temperature synthesis, the QD acquires a hydrophobic, chemisorbed surface layer. As a result, QDs are intrinsically soluble in non-polar media and suitable surface modification must be performed if the dispersant solvent needs to be changed.

The polarity of the medium employed to disperse QDs has a strong effect on the luminescent properties, since it determines the stability of capping ligands on the QD surface [14]. We must highlight that most QD-based systems developed so far are applied for sensing in aqueous media, so the transfer of QDs to a polar medium without changing their optical properties and their ability to interact with target analytes is a critical step.

With the aim of transferring QDs to aqueous media, ligand exchange is one of the methods most used to replace the hydrophobic capping ligands from the surface of QDs. To this end, the most widely-used capping ligands are thiol-based species, such as mercaptoacetic acid (MAA) or mercaptopropionic acid (MPA) and L-cysteine or glutathione (GSH). Usually, changing the original hydrophobic capping ligands produces poor-stability QDs and dramatically decreases the luminescence quantum yields (QYs) [15]. This effect is the result of a competition of two processes:

- on the one hand, water molecules adsorbed on the surface of QDs, which passivate surface traps, result in increased luminescence; but,
- on the other hand, these can oxidize the surface of QDs, hence causing the luminescence to decrease [16].

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