Research on the rapid identification of light green SF using a red-emitting CdTe quantum dot fluorescent sensor

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The study presents a groundbreaking approach to address the pressing issue of ecological contamination caused by Light Green SF (LGSF), a widely used dye that poses significant hazards to aquatic ecosystems. In recognition of the pressing need for a rapid and sensitive method to detect this contaminant, researchers have innovatively developed a highly specialized red-emitting CdTe quantum dot (QDs) fluorescent sensor, specifically designed for the rapid identification of light green SF (LGSF) in aquatic environments. By leveraging fluorescence resonance energy transfer (FRET), a phenomenon where energy is non-radiatively transferred from an excited donor molecule to an acceptor molecule, the researchers found that LGSF acts as an efficient quencher of the CdTe QDs' fluorescence. This quenching effect occurs rapidly and specifically upon the presence of LGSF, enabling the swift and selective detection of the dye in aquatic environments. The study delves deeper into the intricate quenching mechanism, providing valuable insights into the molecular interactions between LGSF and the CdTe QDs. Notably, this is the first investigation to employ QDs for the quantitative assessment of LGSF in lake water, demonstrating the exceptional selectivity and sensitivity of CdTe QDs in this application. Our findings underscore the sensor's potential as a formidable tool for safeguarding aquatic ecosystems from LGSF contamination.

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Keywords: CdTe quantum dots, Fluorescent sensor, Light green SF, Detection mechanism, Environmental monitoring

1. Introduction

The rapid development of science and technology has driven significant progress in light industry, especially the increased demand for dyes. This change is particularly evident in industries such as textiles, papermaking, plastics, and food [1]. Light green SF (LGSF), also known as Acid Green, is a commonly used green triarylmethane dye that is widely applied in industry due to its high coloring strength [2]. Studies have shown that LGSF has the potential for bioaccumulation and penetration, which may stimulate metabolic systems within organisms and has carcinogenic properties if ingested or inhaled [3]. Thus, there is an urgent need to develop an efficient, convenient, and accurate detection method for LGSF to ensure human life safety and health. Currently, various detection technologies have achieved effective detection of dyes, such as liquid chromatography (LC) [4-6], liquid chromatography-mass spectrometry (LC-MS) [7, 8], Surface-Enhanced Raman Scattering (SERS) [9, 10], and electrochemical detection [11, 12]. These technologies demonstrate high accuracy and selectivity but are often accompanied by complex sample pretreatment, prolonged detection processes, high equipment costs, and the necessity for experienced operators and expensive reagents. These factors can impact the accuracy and reliability of detection to some extent and limit the widespread practical application of these methods. Consequently, it is essential to develop a rapid, simple, sensitive, and low-cost novel detection technology to meet the demands of environmental and food testing.

In recent years, due to their unique fluorescent properties and broad application prospects in various fields, quantum dot fluorescence sensors have attracted considerable attention [13, 14]. Among them, CdTe quantum dots (QDs) are a typical fluorescent nanomaterial that exhibits excellent optical performance and stability, showing enormous application potential in biosensing and environmental monitoring [15, 16]. Fluorescent analytical methods are highly favored for their simplicity, high accuracy, and strong sensitivity [17, 18]. Combining the advantages of quantum dots with fluorescence detection technology will undoubtedly provide more efficient and advanced detection methods. Although CdTe QDs have been widely used in fluorescence sensors due to their broad emission spectrum and narrow emission peaks, and they have achieved specific detection of various targets based on Fluorescence Resonance Energy Transfer (FRET), there is currently a gap in the detection of LGSF dyes. In previous studies, Jiménez-López et al. adjusted the surface chemistry of CdTe QDs and gold nanoparticles to achieve the FRET process between donor CdTe QDs and acceptor AuNPs for determining several biologically active thiols [19]. Another researcher, Liang et al. used CdTe QDs with excellent luminescent properties as a fluorescent donor, combined with a two-dimensional metal-organic framework (Cu-TCPP) as an acceptor, to develop a universal fluorescent biosensor for detecting the insertion sequence IS6110 gene fragment specific to Mycobacterium tuberculosis based on

the FRET strategy [20]. These studies fully demonstrate the potential of FRET as an efficient and powerful strategy. Given that the characteristics of quantum dots are consistent with the basic features of FRET applicable to LGSF, we believe that quantum dots can achieve specific recognition of LGSF and provide new ideas and methods for its detection.

This article successfully prepared a CdTe QDs fluorescent sensor, which effectively identifies LGSF in aquatic environments through the phenomenon of fluorescence quenching induced by FRET. To our knowledge, this is the first time that a fluorescent method has been used to detect LGSF. This sensor exhibits specific recognition capability, ultra-high sensitivity, and a short response time for LGSF, indicating that this fluorescent sensor has great potential for application in the field of environmental monitoring.

2. Experimental

2.1. Reagents and instruments

All chemical reagents are commercially purchased analytical grades without further processing. Fluorescence spectra were measured using a fluorescence spectrophotometer (Perkin Elmer LS-55, USA). The morphology of the sensor was characterized by transmission electron microscope (TEM, Tecnai G^2 F30, USA).

2.2. Preparation of CdTe quantum dots

Drawing inspiration from methods detailed in the literature, thioglycolic acid-modified CdTe quantum dots (QDs) were successfully synthesized with minor refinements to the original protocol [21]. In the experimental setup, tellurium powder and sodium borohydride were introduced into a reaction flask. Subsequently, the addition of deionized water facilitated stirring until the solution attained a white hue, followed by allowing the mixture to stand for phase separation to isolate the colorless sodium telluride (NaHTe) upper layer. Under nitrogen-saturated conditions, 0.14 mL of thioglycolic acid was injected into a 40 mL solution of 0.02 mol/L $CdCl_2$, accompanied by NaHTe, with the pH being adjusted to 11.6 using 2 mol/L sodium hydroxide (NaOH). Throughout these steps, the molar ratio of Cd^{2+}: TGA: HTe^- was meticulously maintained at 1: 0.5: 2.5, with the entire process occurring under nitrogen and within an ice bath. The CdTe precursor was then shifted to a reaction kettle and heated to 140°C in a hot air-drying oven for a duration of 230 min to facilitate the synthesis of stable CdTe QDs. Subsequent purification involved the use of methanol, with the resultant precipitate being isolated via centrifugation and vacuum-dried until a constant weight was attained. Ultimately, the CdTe QDs were preserved in powder form at 4°C in darkness for potential future applications.

2.3. Fluorescent detection of LGSF

First, 400 μL of purified CdTe QD solution is accurately pipetted and added to a 10 mL stoppered plastic tube. Next, the CdTe QD solution is diluted with a 0.02 mol/L HEPES buffer solution (pH 8.1) at a dilution factor of 20. Subsequently, 200 μL of the diluted CdTe QD solution is added to each of 11 stoppered plastic tubes, each with a capacity of 4 mL. Then, different concentrations of LGSF solution are added to these tubes, with concentrations of 0, 3, 20, 50, 100, 150, 200, 250, 300, 350, and 400 μmol/L, with each tube receiving 300 μL of LGSF solution. After that, an appropriate amount of 0.02 mol/L HEPES buffer solution (pH 8.1) is added to each tube to ensure that the total volume of the solution in each tube is 3 mL.

Under room temperature conditions, all solutions are thoroughly mixed and allowed to react for 5 min. Then, the fluorescence intensity (F) of each group of solutions is measured using a fluorescence spectrophotometer. The excitation wavelength of the spectrophotometer is set to 365 nm, and the emission spectrum is recorded at a wavelength of 616 nm, ensuring that both the excitation and emission slit widths of the instrument are set to 10.0 nm. To investigate the potential interference of substances in lake water on the experimental results, interference tests are conducted following the same procedure as above, except that the interferent is replaced with a standard LGSF solution, and the concentration of the interferent is set to 20 times that of the LGSF standard solution.

Finally, for practical application, water samples from the Xiuhu Reservoir in Qipan Mountain, Shenyang, are collected for testing. Before testing, the water samples are filtered through a 0.22 μm water filter membrane to remove solid impurities. The fluorescence intensity of the water samples is then measured following the same steps, and the results are analyzed.

3.Results and discussion

3.1. Spectral characterization of CdTe QDs

Fig. 1 presents high-resolution transmission electron microscope (HRTEM) images of the prepared CdTe QDs. The observations reveal that these CdTe nanoparticles exhibit nearly perfect spherical morphology and demonstrate good dispersibility, although slight agglomeration is present. Their particle size distribution is relatively narrow, with an average diameter precisely controlled between 3 to 4 nm, a characteristic that provides a solid foundation for subsequent applications in fluorescence sensors for rapid identification of LGSF. The fine characterization of CdTe QDs not only aids students in gaining a deeper understanding of the preparation and characterization techniques of nanomaterials but also inspires their interest in exploring the practical applications of nanotechnology.

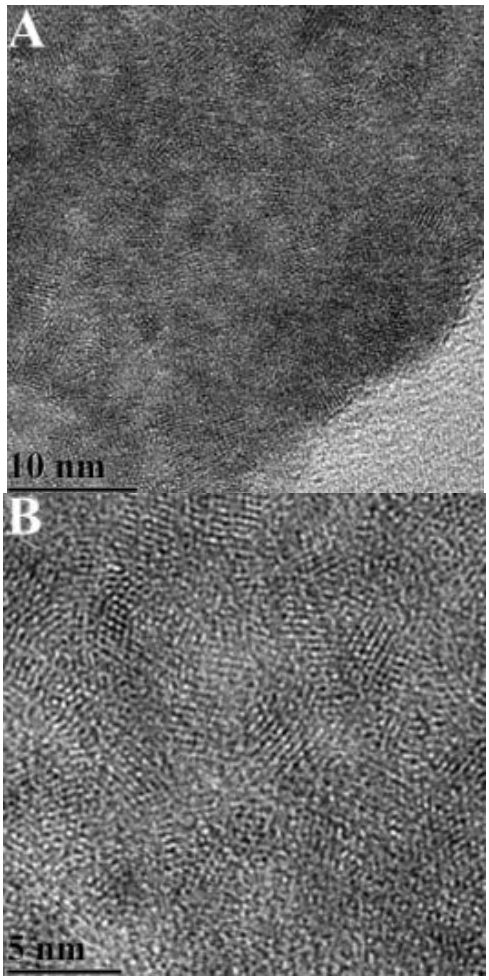

Fig. 1. HRTEM images of CdTe quantum dots

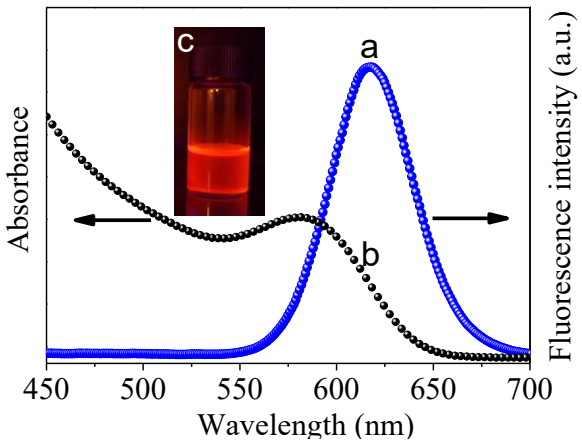

Fig. 2. Absorption and fluorescence emission spectra of CdTe QDs, a and b represent the fluorescence emission spectra and ultraviolet absorption spectra of CdTe quantum dots, respectively (colour online)

After in-depth research on the optical properties of CdTe quantum dots, we obtained detailed ultraviolet-visible absorption spectra and fluorescence emission spectra data, as shown in Fig. 2 these spectral data provide important information about the optical characteristics of the quantum dots. First, by observing the fluorescence emission spectrum, we can clearly see that the emission peak of the CdTe quantum dots is located at 616 nm. This emission peak is not only clearly defined but also exhibits symmetry, with a narrow full width at half maximum (FWHM). This characteristic fully demonstrates the high monochromaticity of the light emitted by the quantum dots, as well as their advantageous low energy dispersion, indicating their potential in optical applications. Furthermore, we used a 365 nm UV lamp to excite the CdTe quantum dots in a dark field and took corresponding photographs. From the photographs, it can be observed that the quantum dots emit bright red light under UV excitation. This observation is consistent with the fluorescence spectrum data, further confirming the excellent luminescent performance of the quantum dots. On the other hand, the ultraviolet-visible absorption spectrum reveals that the CdTe quantum dots have a broad absorption peak, which is continuously distributed. The maximum absorption wavelength appears at 580 nm, indicating the good absorption characteristics of the quantum dots in the ultraviolet region.

3.2. Optimization of detection conditions

In order to obtain the best experimental results, the key factors affecting the detection of LGSF were explored, primarily focusing on fluorescence stability, pH value and reaction time. Detailed investigations were conducted for these three variables. The fluorescence stability of CdTe Quantum Dots (QDs) was investigated (Fig. 3). As illustrated in Fig. 3, the fluorescence emission intensity of this fluorescent sensor remains essentially unchanged over a period of 72 h, indicating its robust and stable fluorescent performance. As shown in Fig. 4, the fluorescence quenching degree of CdTe QDs was maximal at a pH value of 8.1 (LGSF, 20 μmol/L). Given this significant phenomenon, a pH value of 8.1 was determined to be the optimal acidity for this detection system. Furthermore, Fig. 5 provides an analysis of the effect of reaction time on the fluorescence intensity of LGSF. The results indicated that when the reaction time for the addition of LGSF was 5 min, the fluorescence intensity had already reached a stable state, which remained consistent for at least the next 60 min. Therefore, to ensure the efficiency and accuracy of the experiments, a reaction time of 5 min was selected as the optimal duration for the determination of LGSF.

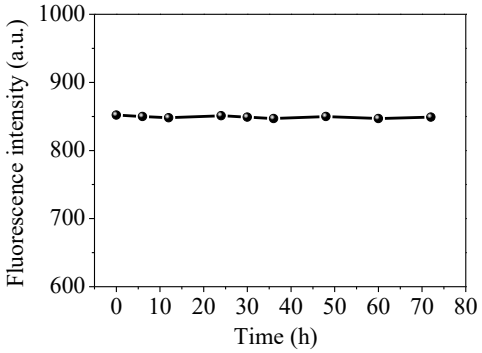

Fig. 3. The fluorescence stability of CdTe QDs in aqueous solution

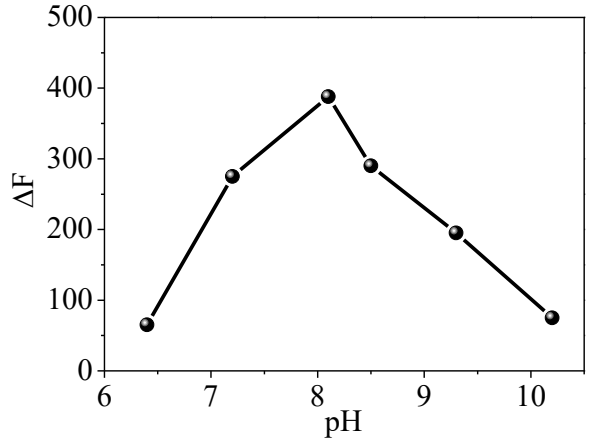

Fig. 4. Effect of pH on fluorescence quenching intensity of CdTe - LGSF system, the concentration of LGSF was 20 μmol/L

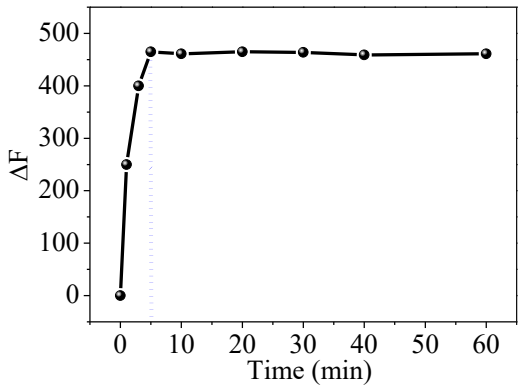

Fig. 5. Effect of reaction time on fluorescence quenching intensity of CdTe-LGSF system, the concentration of LGSF was 25 μmol/L (colour online)

3.3. Detection of LGSF by CdTe QDs

Research has shown that the effect of different concentrations of LGSF on the fluorescence spectrum of CdTe QDs is significant, with results detailed in Fig. 6 From observing Fig. 6, it can be seen that the fluorescence peak of CdTe QDs is located at 616 nm. However, after the addition of LGSF, a noticeable fluorescence quenching phenomenon occurs. Notably, as the concentration of LGSF gradually increases, the fluorescence intensity of CdTe QDs also shows a trend of gradual quenching. Under suboptimal experimental conditions, the degree of fluorescence quenching observed in CdTe quantum dots (ΔF) demonstrates a strong linear correlation with the LGSF concentration, spanning the range of 0.2 to 40 μmol/L. The linear equation is given by $\Delta F = 17.5181C + 26.4339$, with an exceptionally high squared linear correlation coefficient (R^2) of 0.9980 (as depicted in Fig. 7). The limit of detection (LOD) was calculated using the equation $LOD = 3S_0/K$, where S_0 is the standard deviation of blank CdTe QDs measurements (n=5), and K is the slope of the calibration curve. After calculations, the LOD was found to be 0.040 μmol/L. Furthermore, parallel measurements of 15 μmol/L LGSF yielded a relative standard deviation of only 2.7% (n=11), which fully demonstrates that this method possesses high sensitivity and precision.

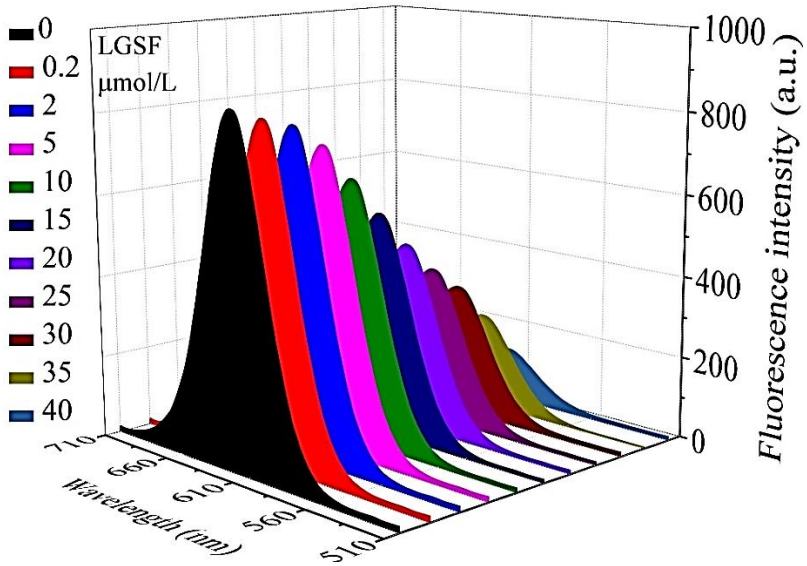

Fig. 6. Effect of LGSF concentration on fluorescence intensity of CdTe QDs (colour online)

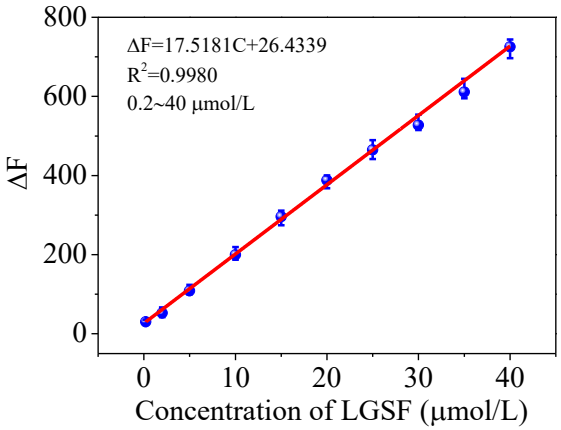

Fig. 7. Relationship between the relative fluorescence intensity of CdTe QDs and LGSF (colour online)

3.4. Selectivity of CdTe QDs fluorescent sensors

Further research explored the effects of various potential interfering substances on the fluorescence of the developed QDs sensor when detecting LGSF. Under conditions where the concentration of LGSF was 25 μmol/L, experimental results showed that even when the concentrations of ions such as K^+, Ca^{2+}, Na^+, Mg^{2+}, Zn^{2+}, Al^{3+}, S^{2-}, Cl^-, Br^-, I^-, NO_3^-, CO_3^{2-}, NO_2^-, and SO_3^{2-} reached 500 μmol/L, they had no quenching effect on the fluorescence intensity of CdTe QDs (as shown in Fig. 8). This finding strongly demonstrates that the proposed method has high feasibility and reliability for analysis of real samples.

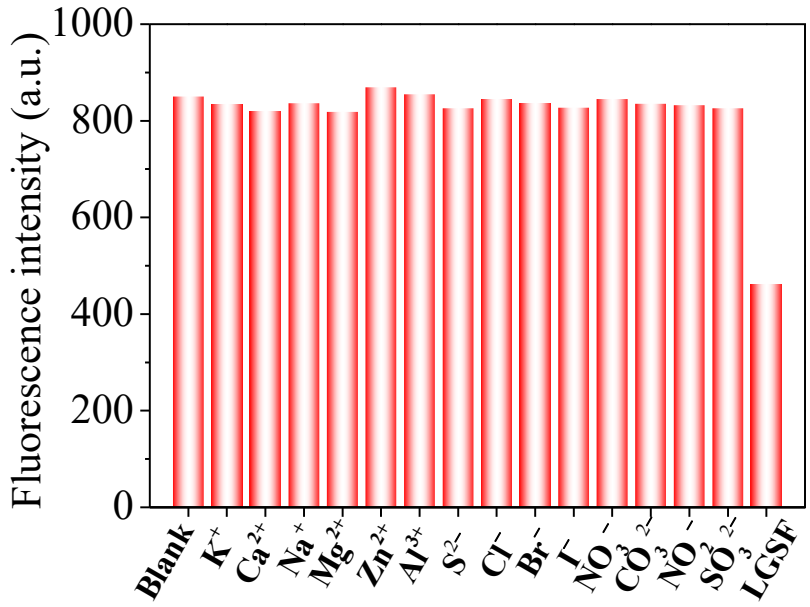

Fig. 8. Fluorescence responses of CdTe QDs to LGSF and various analytes, the concentrations of LGSF and analytes were 25 μmol/L and 500 μmol/L, respectively (colour online)

3.5. Detection mechanism

Herein, we elucidate the potential mechanism underlying the fluorescence detection of LGSF using CdTe QDs. Firstly, CdTe QDs, as semiconductor nanomaterials, possess unique optical and electrical properties, and their surfaces are specifically modified with thiol-capped mercaptoacetic acid groups. These characteristics facilitate interactions between CdTe QDs and a variety of materials or molecules. In the interaction with LGSF, we hypothesize the existence of hydrogen bonding, where hydrogen bond donors such as carboxyl and hydroxyl groups on the surface of CdTe QDs may form hydrogen bonds with hydrogen bond acceptors in LGSF molecules, thereby enhancing the interaction between them.

Furthermore, the fluorescence emission peak of CdTe QDs is situated at 616 nm. Concurrently, the ultraviolet-visible absorption spectrum of LGSF exhibits a prominent absorption peak at 585 nm in the ultraviolet region. Notably, there is a significant overlap between these two spectral peaks (as depicted in Fig. 9), which precisely fulfills the requirements for fluorescence resonance energy transfer (FRET). Therefore, CdTe QDs can strongly associate with LGSF, facilitating efficient FRET. This concept is visually demonstrated in Fig. 10, which clearly portrays the FRET process occurring between CdTe QDs and LGSF through a schematic representation.

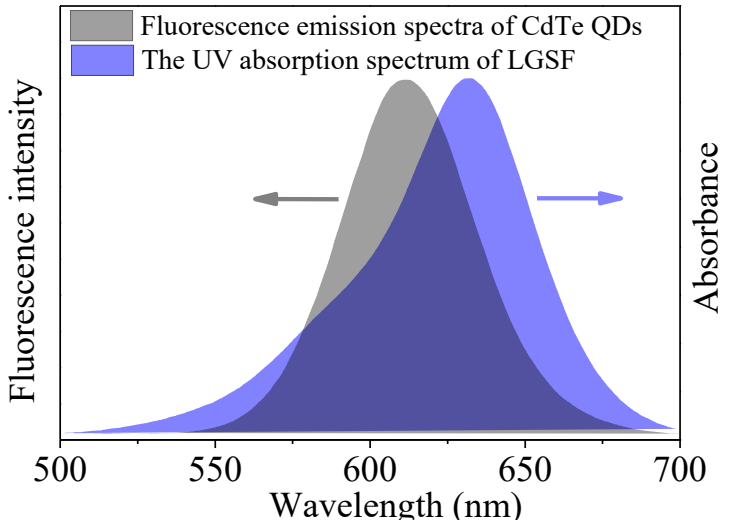

Fig. 9. Overlap of fluorescence emission spectra of CdTe QDs and absorbance spectra of LGSF (colour online)

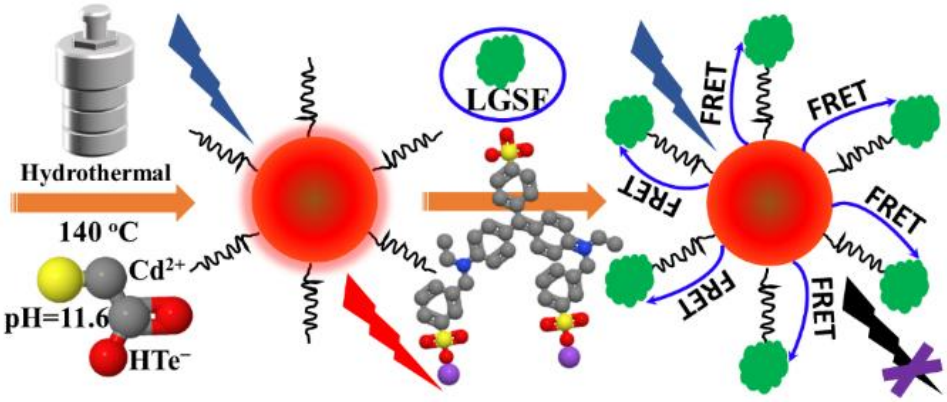

Fig. 10. Synthesis route of CdTe QDs and fluorescence detection strategy for LGSF (colour online)

3.6. Detection of LGSF in lake water

Under optimal experimental conditions, a standard addition method was used to test for LGSF by adding a standard concentration, and the results are shown in Table 1. By observing the data in Table 1, it can be found that LGSF was not detected in the lake water samples. This result suggests that the concentration of LGSF in the lake water sample may be below the established detection limit, making it undetectable. The recovery rate for the spiked samples ranged from 98.0% to 106.0%, and the relative standard deviation (RSD) was between 2.2% and 3.4%, which demonstrates the feasibility and reliability of the standard addition method for testing LGSF.

Table 1. Determination of LGSF in lake water

Samples	Spiked (μmol/L)	Found (μmol/L)	Recovery (%)	RSD (%)
	0	–	–	–
Lake water	0.5	0.49	98.0	2.6
	1	1.06	106.0	2.2
	5	5.16	103.2	3.4
	10	9.97	99.7	2.8
	20	21.08	105.4	3.1

4. Conclusion

This study synthesized CdTe QDs fluorescent sensor for the specific recognition of LGSF in water environments. This sensor induces fluorescence quenching using the FRET mechanism, enabling rapid response to the presence of LGSF in water, along with good sensitivity and selectivity. Additionally, a detailed analysis and summary of the FRET fluorescence quenching mechanism were conducted. The developed detection method features high specificity, high sensitivity, and ease of use, providing a powerful and reliable platform for detecting LGSF in water environments, which holds significant importance for environmental analysis and biological monitoring.

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