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$CuInS_2$ quantum dots-based fluorescence turn off/on probe for detection of melamine

Siyu Liu, Junjie Hu, Hao Zhang, Xingguang Su *

Department of Analytical Chemistry, College of Chemistry, Jilin University, Qianwei Road 10, Changchun 130012, China

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ABSTRACT

In this paper, a sensitive and simple method for the determination of melamine (MA) was developed based on the fluorescence changes of the water-soluble $CuInS_2$ quantum dots (QDs). The water-soluble $CuInS_2$ QDs capped by mercaptopropionic acid (MPA) was directly synthesized by hydrothermal method based on our previous report. The fluorescence emission of $CuInS_2$ QDs was quenched by the oxidation of the surface of the QDs with H_2O_2, and the quenched fluorescence of $CuInS_2$ QDs could be recovered upon the addition of small amounts of MA, which might be due to the surface passivation of the $CuInS_2$ QDs by MA. The other amino acids such as glycine and lysine had no effect on the quenched fluorescence of $CuInS_2$ QDs. Under optimum conditions, there was a good linear relationship between the fluorescence intensity of $CuInS_2$ QDs and the concentration range of MA from 1.0×10^{-8} to 1.0×10^{-5} mol/L with a detection limit as low as 5 nM. The proposed method was successfully applied to detect trace MA in raw milk with satisfactory results. Compared with previous reports, the proposed method manifested several advantages such as high sensitivity, short analysis time, low cost and ease of operation.

1. Introduction

Melamine (2,4,6-triamino-1,3,5-triazine, MA) is a heterocyclic triazine organic compound with high nitrogen level (about 66% nitrogen by mass), which has been widely used in the chemical industry, including the production of melamine formaldehyde resin (for surface coatings, laminates, and adhesives), plastic and a very durable thermosetting plastic, fertilizer, glue, and flame retardants [1,2]. Undoubtedly, MA is a toxic compound to both animals and human beings [3], and is connected to various diseases, such as, kidney stones and bladder cancer [4,5]. MA molecule shows the similar analytical characteristics of protein molecules in the protein analysis via the traditional Kjeldahl method [6], which is usually employed to quantify the crude protein content in foods, cannot differentiate it from protein molecules. Unfortunately, because of its high nitrogen level of 66% and low cost, MA was added in the infant formula and pet food by unethical manufacturers to enhance the apparent crude protein value [7–9]. It has been reported that the combination of MA with cyanuric acid leads to the formation of insoluble crystals in the kidneys and subsequent tissue injury, which will result in the renal failure and even death of children after a long-term intake of melamine-tainted food [10–13]. Thus it is very necessary to develop a simple and reliable method for the efficient detection of melamine. Up to now, many methods have been developed for MA detection such as gas chromatography (GC) [14], liquid chromatography (LC) [15], capillary electrophoresis (CE) [16,17], mass spectrometry [18], ultraviolet (UV) spectroscopy [19] and near infrared spectroscopy [1]. However, many of these methods are not readily adaptable to routine analysis, because they often need professional technical personnel, time-consuming and complicated operation procedures, and expensive instruments. So, there is an urgent need to develop a simple, rapid, sensitive and low-cost assay for MA determination.

In recent years, fluorescence analysis has attracted increasing attention owing to its operational simplicity, high sensitivity and real-time detection. A series of fluorescence sensors have been designed for the detection of different substances in food and other fields [20–32]. Chen reported a novel fluorescence sensor to probe K^+ based on 15-crown-5-functionalized CdSe/ZnS QDs [27]. Ramanavicius et al. developed an immunosensor to probe target protein based on fluorescence quenching matrix of the conducting polymer polypyrrole [28]. The ability of 2,20-bipyridine-bound copper (II) ions to quench the fluorescence of hydrophobic CdSe QDs was used to create a turn-on fluorescence cyanide sensor [29].

To our knowledge, most of the previous reports on the fluorescence determination of MA were based on gold nanoparticles or

* Corresponding author. Tel.: +86 431 5168 352.
E-mail address: suxg@jlu.edu.cn (X. Su).

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some special organic fluorescence compounds [33–40]. Guo reported a fluorescent analysis for MA based on the high fluorescence quenching ability of gold nanoparticles [33]. Kim and his co-workers represented a selective polydiacetylene (PDA) liposomes sensory system for MA detection based on the multiple hydrogen bondings between cyanuric acid and MA [34]. In this work, we developed a new effective fluorescence quenching analytical method for the detection of MA, the proposed method does not need expensive materials such as Au as fluorescence probe, avoids complex modification of quantum dots, and has the virtue of simplicity of operation, and high sensitivity and selectivity, which was nearly unaffected by some other amino acid molecules.

Based on our previous report [41], we firstly presented the one-pot synthesis of water-soluble $CuInS_2$ QDs, that does not contain any toxic Class A elements (Cd, Pb, and Hg) or B elements (Se and As). Herein, we developed a simple $CuInS_2$ QDs-based fluorescent turn off/on probe to detect MA with high selectivity and sensitivity, which based on the simple chemical redox strategy to quench the fluorescence of MPA capped $CuInS_2$ QD by H_2O_2, and the fluorescence recovery induced by MA.

2. Experiment section

2.1. Apparatus

The fluorescence spectra were obtained by using a Shimadzu RF-5301 PC spectrofluorophotometer equipped with a xenon lamp using right-angle geometry. UV–vis absorption spectra were obtained by a Varian GBC Cintra 10e UV–vis spectrometer. In both experiments, a 1 cm path-length quartz cuvette was used.

2.2. Reagents

All reagents were of at least analytical grade. Copper (II) chloride dihydrate ($CuCl_2 \cdot 2H_2O$), sodium hydroxide (NaOH), sulfourea (CS (NH_2)$_2$), sodium dihydrogen phosphate (NaH_2PO_4), disodium hydrogen phosphate (Na_2HPO_4), MA, acetonitrile(CH_3CN), and trichloroacetic acid(CCl_3COOH) were purchased from Beijing Chemical Works. Mercaptopropionic acid (MPA) and indium (III) chloride tetrahydrate ($InCl_3 \cdot 4H_2O$) were purchased from Sigma-Aldrich Corporation. Raw milk was bought from local War-Mart stores. The 0.1 mol/L PBS (pH 7.4, 0.1 mol/L NaH_2PO_4–Na_2HPO_4) was used as the medium for QDs solution. The water used in all experiments had a resistivity higher than 18 MΩ cm^{-1}.

2.3. Synthesis of $CuInS_2$ QDs

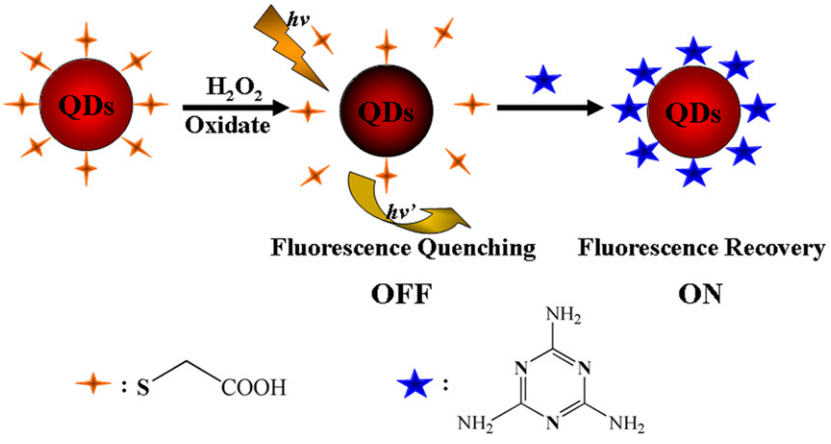
According to our previous report [41] MPA-capped $CuInS_2$ QDs were synthesized in aqueous solution via hydrothermal synthesis method. In a typical experiment, $CuCl_2 \cdot 2H_2O$ (0.15 mmol) and $InCl_3 \cdot 4H_2O$ (0.15 mmol) were dissolved in distilled water (10.5 ml), then MPA (1.8 mmol) was injected into the solution, and then the solution produced yellow granule immediately. After that 2 mol/L NaOH solution was dropwise added into the mixture solution until the pH value was adjusted to 11.3 with stirring. During this process, the solution changed from turbid to clear pink. After stirring for 10 min, CS(NH_2)$_2$(0.30 mmol) was dissolved in the solution. The Cu-to-In-to-S and Cu-to-MPA precursor ratios were 1:1:2 and 1:12, respectively. All the above mentioned experimental procedures were performed at room temperature, and then the mixture solution was transferred into a Taflon-lined stainless steel autoclave with a volume of 15 mL. The autoclave was maintained at 150 for 21 h and then cooled down to room temperature by a natural process. The as-prepared original $CuInS_2$ QDs solution were obtained. Lots of ethanol was added to the solution to obtain $CuInS_2$ QDs precipitate, and the process was repeated three times. The remaining contaminants were removed by the cycled washing. The purified $CuInS_2$ QDs was diluted to the mark, and stored in the darkroom. The final concentration of $CuInS_2$ QDs was about 1.5 mmol/L according to the addition of Cu^{2+} or In^{3+} concentration.

2.4. Fluorescence quenching of $CuInS_2$ QDs by H_2O_2

2.0 mL of 1.5 mmol/L $CuInS_2$ QDs solution, 1.0 mL of 0.1 mol/L PBS (pH 7.4) and different amounts of H_2O_2 were successively added into a 10 ml calibrated test tube, diluted to the mark with deionized water, shaken thoroughly and equilibrated for 20 min until the solution was fully mixed. The fluorescence spectra were recorded from 610 nm to 800 nm, and the fluorescence emission peak intensity was used for quantitative analysis.

2.5. Detection of MA

2.0 mL of 1.5 mmol/L $CuInS_2$ QDs solution, 1.0 mL of 0.1 mol/L PBS (pH 7.4) and a certain amounts of H_2O_2 were added into a 10 ml calibrated test tube, shaken thoroughly and equilibrated for 20 min until the mixture was fully mixed. Different amounts of MA were added into the test tube, diluted to the mark with deionized water, shaken thoroughly and equilibrated for 10 min, and the concentration of H_2O_2 was 0.5 mmol/L. The fluorescence spectra were recorded from 610 nm to 800 nm, and the fluorescence emission



Scheme 1. The schematic illustration of the detection process for MA.

peak intensity was used for quantitative analysis. All the operation was finished at room temperature. The process of the detection of MA could be described as the Scheme 1.

2.6. Milk sample preparation

The raw milk was pretreated as previous reported procedures [38,40]. 1 mL of acetonitrile, 1 mL of trichloroacetic acid, 2.0 mL of raw milk and 7 mL of water were added into a centrifuge tube. Then the mixture was ultrasonically extracted for 20 min and then centrifuged at 5000 rpm for 20 min to eliminate protein. The obtained supernatant was filtered two times. The filter liquor was diluted as a 1:10 ratio with deionized water and the diluents were used for MA detection.

3. Results and discussion

3.1. Spectral characteristic and TEM image of MPA-capped $CuInS_2$ QDs

The PL emission and UV–vis absorption spectra of MPA-capped $CuInS_2$ QDs are shown in Fig. 1A. It could be seen that the fluorescence emission peak of the $CuInS_2$ QDs around 660 nm was narrow and symmetrical, and the UV–vis absorption peak was around 570 nm, which were consistent with our previous report [41]. As shown in Fig. 1B, TEM results showed that the as-prepared $CuInS_2$ QDs were spherical particles with diameters about 2–4 nm.

3.2. $CuInS_2$ QDs fluorescence quenching by H_2O_2

As the optical property of QDs strongly depends on the nature of the surface [42], some analytes that could interact with QDs can cause changes of fluorescence. It has been reported that H_2O_2 as a chemical oxidant could result in fluorescence quenching of QDs [43–46]. Scheme 1 illustrates the process of H_2O_2 induced fluorescence quenching of MPA-capped $CuInS_2$ QDs. In the presence of H_2O_2, the thiol groups of MPA molecule bonding to the surface of $CuInS_2$ QDs were readily oxidized to organic disulfide product (RS-SR). As a result, fewer MPA molecules were bonded to the surface of QDs, leading to the reduced fluorescence intensity [45,46]. In this paper, we investigated the quenching effect of H_2O_2 on the fluorescence emission of $CuInS_2$ QDs. Curve **a** in Fig. 2 described the temporal evolution of the fluorescence quenching of $CuInS_2$ QDs by 0.5mmol/L H_2O_2.It showed that the fluorescence of $CuInS_2$ QDs decreased with the addition of H_2O_2, and remained nearly constant after 10 min. In the further experiments, $CuInS_2$ QDs added with 0.5 mmol/L H_2O_2 was shaken thoroughly for 20 min, and then the fluorescence intensity of the mixture was recorded.

As shown in Fig. 3, the fluorescence intensity (FL) of $CuInS_2$ QDs decreased with the increasing of H_2O_2 concentration upward to 5 mmol/L, and the FL changing trends indicated it was a complex quenching process. It could be also seen from Fig. 3, MA had no effect on fluorescence of $CuInS_2$ QDs even under concentration of 5 mmol/L.

Fig. 4 depicts the fluorescence spectra of $CuInS_2$ QDs with different concentrations of H_2O_2, and the inset showed that there was a good linear relationship between the relative FL intensity $(F_0–F)/F_0$ (F_0 and F were the FL intensity of the $CuInS_2$ QDs in the absence and the presence of H_2O_2, respectively) and the logarithm of H_2O_2 concentration in the range of 0.1–500 μmol/L.

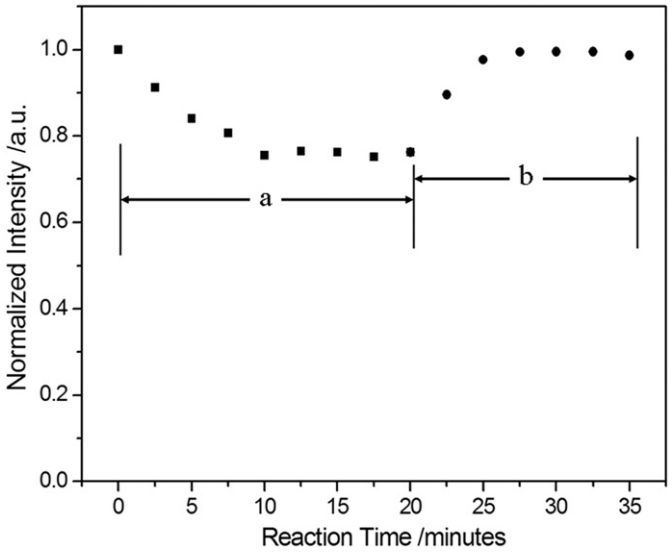


Fig. 2. (a) Fluorescence quenching of $CuInS_2$ QDs by 0.5 mmol/L H_2O_2 in different incubation times. (b) Fluorescence recovery of $CuInS_2$ QDs–H_2O_2 system (incubating 20 min) by addition of 10 μmol/L MA with increasing incubation times. PBS: 10 mmol/L phosphate buffer solution (pH 7.4).

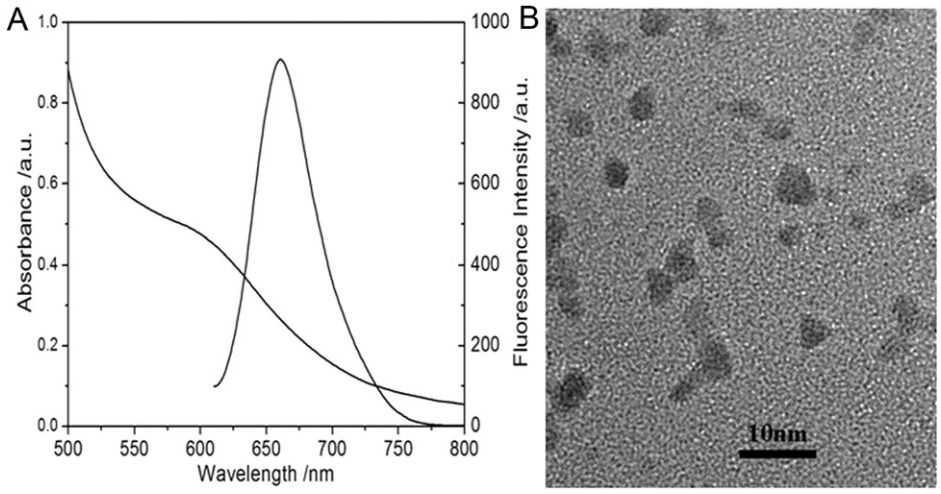


Fig. 1. (A) The fluorescence emission and UV–vis absorption spectra of the $CuInS_2$ QDs solution. (B) TEM image of the as-prepared $CuInS_2$ QDs.

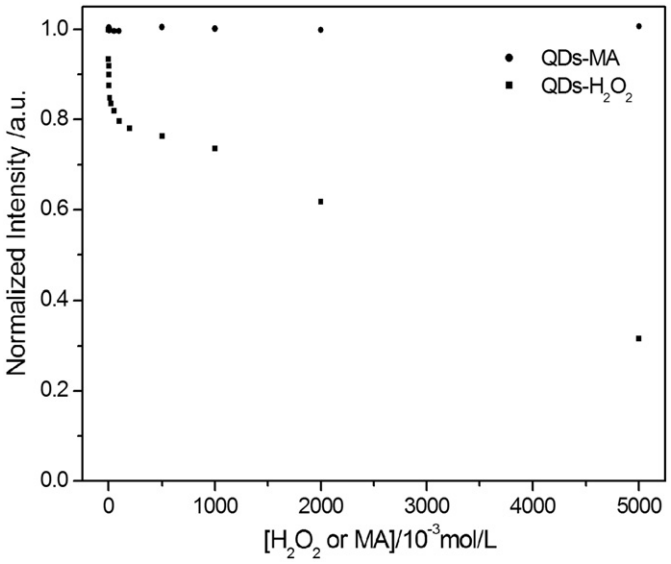


Fig. 3. Fluorescence response of $CuInS_2$ QDs upon the addition of H_2O_2 (0, 0.1, 0.2, 1.0, 2.0, 10, 20, 50, 100, 200, 500, 1000, 2000, 5000 μmol/L) or MA (1, 5, 10, 50, 100, 500, 1000, 2000, 5000 μmol/L).

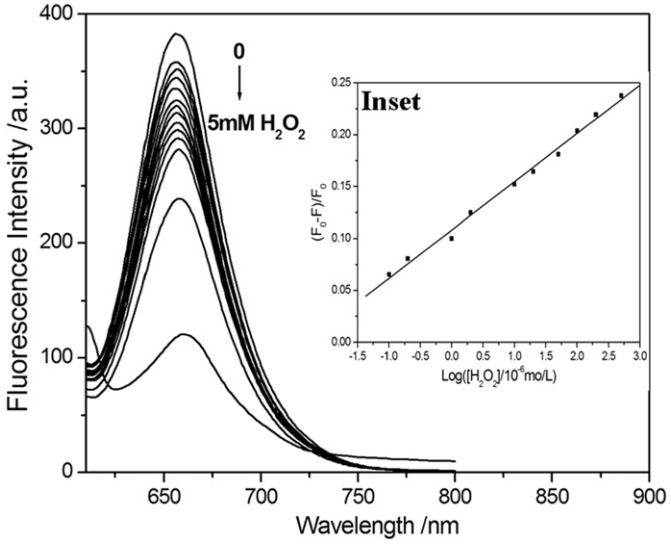


Fig. 4. Fluorescence spectra of $CuInS_2$ QDs upon the addition of different concentrations of H_2O_2 from 0.10 μmol/L to 5 mmol/L. **Inset:** The plot of fluorescence intensity versus logarithm of H_2O_2 concentration in the range of 0.10–500 μmol/L. PBS: 10 mmol/L phosphate buffer solution (pH 7.4).

The regression equation is:

$$(F_0-F)/F_0 = 0.108 + 0.0465\mathrm{Log}\,[H_2O_2],\ \mu\mathrm{mol/L} \quad (1)$$

The corresponding regression coefficient is 0.997, and the detection limit for H_2O_2 was 0.05 μmol/L mol/L, and the standard deviation for nine replicate measurements of 0.5 μmol/L H_2O_2 was 3.6%.

3.3. Fluorescence recovery of $CuInS_2$ QDs–H_2O_2 system by MA

In this study, we found that the fluorescence of $CuInS_2$ QDs quenched by H_2O_2 could be restored upon the addition of MA and it would keep constant after 10 min (Fig. 2b). Fig. 5 shows the fluorescence spectra of $CuInS_2$ QDs quenched by 0.5 mmol/L H_2O_2 upon the addition of different concentrations of MA. It can be seen that the fluorescence intensity of $CuInS_2$ QDs would be enhanced

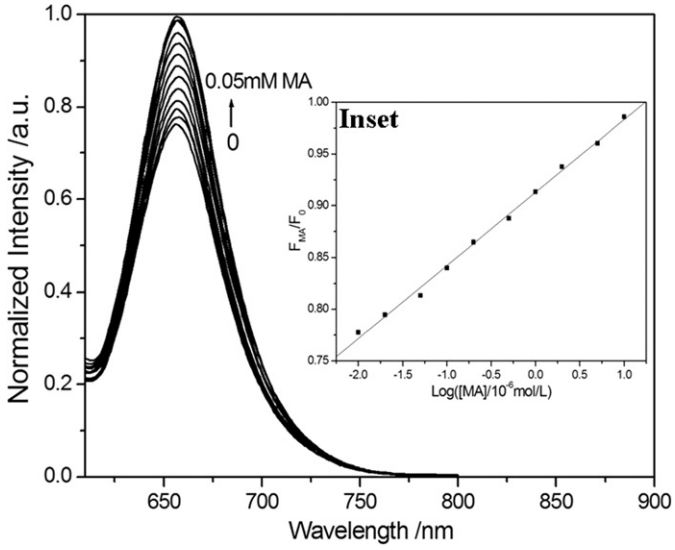


Fig. 5. Fluorescence spectra of $CuInS_2$ QDs-0.5 mmol/L H_2O_2 system upon the addition of different concentrations of MA from 0.01 μmol/L to 50 μmol/L. **Inset:** The plot of F_{MA}/F_0 versus the logarithm of MA concentration in the range of 0.01–10 μmol/L. PBS: 10 mmol/L phosphate buffer solution (pH 7.4).

as the concentration of MA increased from 0.01 to 50 μmol/L with the maximum recovery of nearly 99%. There was a good linear relationship between relative fluorescence intensity F_{MA}/F_0 (F_{MA} and F_0 were the FL intensity of the $CuInS_2$ QDs-0.5 mmol/L H_2O_2 system in the presence or absence of MA, respectively) and the logarithm of MA concentration in the range from 0.01 to 10 μmol/L. The regression equation is:

$$F_{MA}/F_0 = 0.913 + 0.0705\mathrm{Log}\,[MA],\ \mu\mathrm{mol/L} \quad (2)$$

The corresponding regression coefficient is 0.998, and the detection limit for MA was 5 nmol/L, and the standard deviation for nine replicate measurements of 0.02 μmol/L H_2O_2 was 2.3%.

Compared with the previous reports about fluorescence detection for MA, such as MA molecule detection via fluorescence resonance energy transfer (FRET) phenomenon between fluorescein and Au nanoparticles with linear range from 1×10^{-7} mol/L to 4.0×10^{-6} mol/L [33], or based on FRET phenomenon between CdTe QDs embedded in mesoporous SiO_2 and Au nanoparticles with linear range from 7.5×10^{-9} mol/L to 3.5×10^{-7} mol/L [35]. Our method does not need expensive materials such as Au as fluorescence probe, and has the superior detection limit (0.005 μmol/L), and more wide dynamic range (0.01–10 μmol/L).

As in previous reports [43–46], H_2O_2 as an oxidant could oxidize MPA to form disulfide that would fall off the surface of $CuInS_2$ QDs. The loss of the MPA from the surface of QDs induced surface defects, which facilitated the non-radiation recombination, leading to fluorescence decrease. It has been reported that amine group could also bind to the surface of QDs to eliminate the surface defects [47,48], and some molecules containing amine group could enhance the fluorescence of QDs. So the fluorescence recovery of $CuInS_2$ was induced by MA [38,47].

3.4. Optimization for MA detection

As indicated in Fig. 6, the FL intensity of quenched $CuInS_2$ QDs quenched by 1.0 mmol/L H_2O_2 could not be recovered by MA again. Based on our proposed mechanism, when excess H_2O_2 was added into the system, it would not only oxidize MPA molecules, but also destroy the structure of $CuInS_2$ QDs that is why the fluorescence of the $CuInS_2$ QDs quenched by excess H_2O_2 could

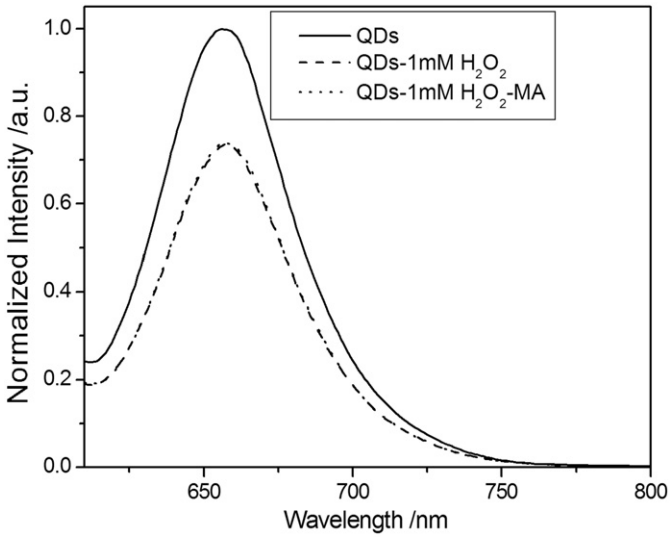


Fig. 6. Fluorescence spectra of $CuInS_2$ QDs quenched by 1.0 mmol/L H_2O_2 upon the addition of 10 μmol/L MA. PBS: 10 mmol/L phosphate buffer solution (pH 7.4).

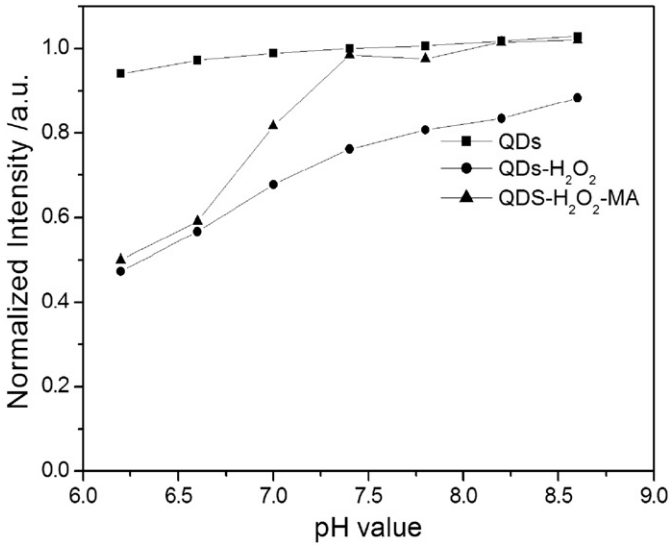


Fig. 7. Fluorescence intensity of $CuInS_2$ QDs with or without 0.5 mmol/L H_2O_2, and $CuInS_2$ QDs-0.5 mmol/L H_2O_2 restored by 10 μmol/L MA in different pH environments.

not be restored by MA. So 0.5 mmol/L H_2O_2 was chosen as the quencher in our experiments.

Fig. 7 shows the FL intensity changes of $CuInS_2$ QDs with or without 0.5 mmol/L H_2O_2 and the quenched QDs restored by 10 μmol/L MA in different pH environments (from 6.2 to 8.6). As shown in Fig. 7, the increase of fluorescence intensity of QDs–H_2O_2 in the pH range of 6.2–8.6 was due to dual attributions: the increased fluorescence intensity of QDs from pH 6.2 to 8.6 and the stronger oxidizability of H_2O_2 in acid condition from the standpoint of electrode potential. It can be seen that when the pH value was higher than 7.0, the fluorescence recovery of $CuInS_2$ QDs-0.5 mmol/L H_2O_2 by 10 μmol/L MA was better than in acid medium, which was because amino group (NH_2-) would exist in the cationic form (NH_3^+-) in acid medium, and the fluorescence recovery reached the maximum at pH 7.4. So physiological condition of pH 7.4 PBS was used in the further experiments.

In this study, we investigated the fluorescence spectra changes of $CuInS_2$ QDs quenched by different concentrations of $KMnO_4$, and its corresponding recovery results by 10 μmol/L MA. As Fig. 8

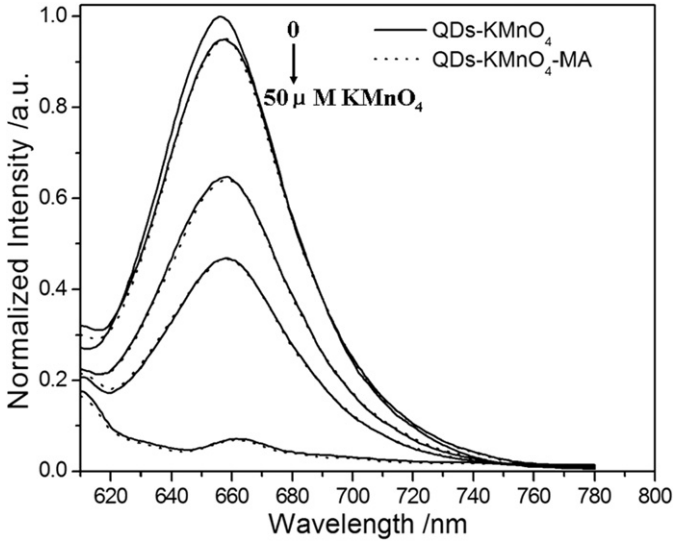


Fig. 8. The fluorescence spectra of $CuInS_2$ QDs quenched by different concentrations of $KMnO_4$ in the absence (Solid line) and the presence (dash line) of 10 μmol/LMA. From top to bottom, the $KMnO_4$ concentrations are 0, 1.0, 5.0, 10.0, 50 μmol/L, respectively. PBS: 10 mmol/L phosphate buffer solution (pH 7.4).

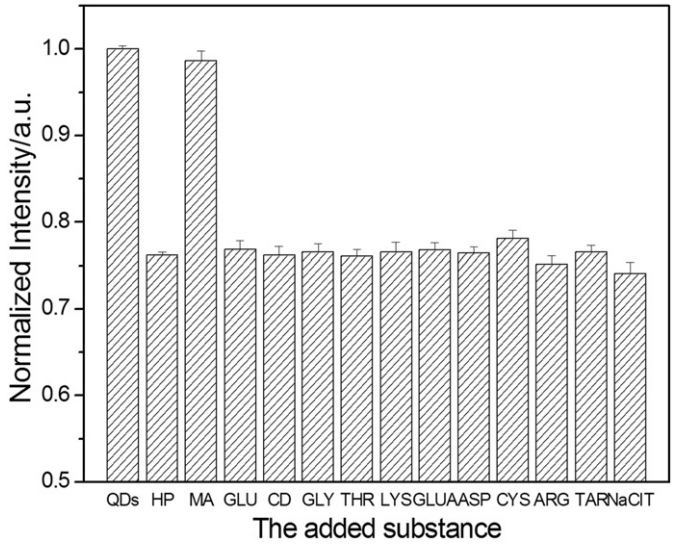


Fig. 9. Effect of a series of 0.01 mmol/L molecules (melamine(MA), glucose (GLU), cyclodextrin (CD), glycine(GLY), threonine(THR), lysine(LYS), glutamic acid(GLUA), aspartate(ASP), cystine(CYS), arginine(ARG), tartaric acid(TAR), sodium citrate(NaCIT)) on the fluorescence of $CuInS_2$ QDs quenched by 0.5 mmol/L hydrogen peroxide(HP). PBS: 10 mmol/L phosphate buffer solution (pH 7.4).

demonstrates, the fluorescence of $CuInS_2$ QDs quenched by $KMnO_4$ even at relatively low concentration could not be restored by 10 μmol/L MA, that is due to that the generated Mn^{2+} from $KMnO_4$ was easily absorbed on the surface of $CuInS_2$ QDs which would inhibit the bonding of MA to the surface of QDs [49,50].

3.5. Interference study

In this study, we investigated the effect of a series of physiological molecules such as melamine(MA), glucose (GLU), cyclodextrin(CD), glycine(GLY), threonine(THR), lysine(LYS), glutamic acid(GLUA), aspartate(ASP), cystine(CYS), arginine(ARG), tartaric acid(TAR), sodium citrate(NaCIT) on the fluorescence of $CuInS_2$ QDs quenched by 0.5 mmol/L H_2O_2. As described in Fig. 9, only MA molecule could restore the fluorescence of $CuInS_2$ QDs

Table 1
Results of the determination of the MA in raw milk.

Samples	Added (μ mol/L)	Found (μ mol/L)	Recovery (%)	RSD ($n=3$, %)
1	0	0	–	–
2	1.0	0.95	95	4.1
3	10.0	9.96	99.6	4.4

quenched by 0.5 mmol/L H_2O_2 and the other molecule nearly had no effect on the fluorescence of the quenched $CuInS_2$ QDs. The result indicated that the selectivity of this method was acceptable.

3.6. Detection of MA in milk samples

In order to test the applicability of the proposed method, it was applied to determinate MA in pretreated real liquid milk which spiked with different concentrations of MA. As Table 1 shows that, MA was not present in original milk samples, the average recovery of the MA was in the range of 95–99%, and the RSD of three replicate determinations was lower than 4.4%. The above results demonstrated the potential applicability of the $CuInS_2$ QD-based fluorescence probe for the detection of MA content in real milk samples.

4. Conclusion

In summary, we have developed a novel fluorescence turn off/on probe for the detection of MA, which based on the fluorescence quenching of MPA capped $CuInS_2$ QD by H_2O_2, and the fluorescence recovery induced by MA. The proposed method avoids complex modification of QDs, and had the virtue of simplicity of operation, and high selectivity, which was nearly unaffected by some other amino acid molecules. The present method was applied to the detection of MA in milk samples with satisfactory results.

Acknowledgments

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