Research Article

Determination of sudan III using nitrogen and sulfur co-doped carbon quantum dots with fluorescence enhancement effect

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Abstract

Sudan III, a synthetic azo dye, is commonly used for coloring oils, waxes, and various plastics. However, its unauthorized presence in food and food products poses serious health hazards, including potential carcinogenic effects. In this study, we investigated the application of nitrogen and sulfur co-doped carbon quantum dots (N,S-CQDs) as a fluorescence sensor to detect trace amounts of sudan III. When exposed to trace amounts of sudan III, the fluorescence signal of N,S-CQDs was significantly enhanced. Under optimal conditions (pH 5.0 and an incubation time of 4 minutes), the fluorescence enhancement of N,S-CQDs showed a linear relationship with sudan III concentration in the range of 7.5×10^{-10} to 1.0×10^{-8} M, with a limit of detection of 2.0×10^{-10} M (equivalent to 0.07 ppb). The method demonstrated high accuracy (recovery from 91.5% to 103.5%), good reproducibility (RSD < 3.86%), and excellent selectivity for sudan III over other azo dyes. We successfully applied this developed method to analyze sudan III in chili sauce and chili powder samples, cross-checking differences between results less than 10% with LC-MS/MS analysis.

Keywords: *carbon quantum dots; sudan III; fluorescence enhancement effect.*

1. INTRODUCTION

Azo-dyes, characterized by their chromophoric azo-group, are a class of synthetic organic colorants extensively utilized in various industrial applications, including solvents, oils, fats, waxes, plastics, printing inks, and floor polishes [1, 2]. Their widespread use is primarily due to their colorfastness and low cost. However, these dyes become biologically

active through their metabolites [3, 4], and have been linked to a higher incidence of bladder cancer among textile and leather workers, painters, and hairdressers [5]. Due to their potential carcinogenic properties, many countries have banned the use of most azo-dyes in products intended for human consumption [5].

Sudans I, II, III, and IV are non-ionic fat-soluble dyes commonly used as additives in gasoline, grease, oils, and plastics. The International Agency for Research on Cancer (IARC) classifies these dyes as category 3 carcinogens due to their ability to induce certain forms of liver and bladder cancer in animals (International Agency for Research on Cancer, 1975). Although the risk level is debated, sudan dyes are prohibited as food additives for humans. The recent contamination of hot chili and derived products from India, which were marketed in the European Union [6], underscores the need for reliable and accurate analytical methods for the rapid identification and quantification of these compounds in food products.

Current detection techniques, including near-infrared (NIR) or Raman spectroscopy [7], electrochemical detection [8], high-performance liquid chromatography (HPLC) [9, 10], and colorimetric detection [11, 12], face several significant challenges. These challenges include expensive equipment, complex sample preparation, lengthy analysis times, and intricate setups, making them impractical for routine use. Detecting sudan III in real-life samples is particularly challenging due to its low concentration and interference from other substances within the sample matrix. This underscores the urgent need for an efficient, highly selective, and environmentally friendly method for identifying sudan III in food samples.

Fluorescent sensors are highly appealing due to their affordability, ease of use, rapid analysis, and high sensitivity. These sensors have garnered significant attention for developing quantitative analytical techniques, leveraging changes in fluorescent intensity or emission peak position through either enhancement or quenching [13-15]. Unlike other semiconductor quantum dots, carbon quantum dots (CQDs) offer straightforward synthesis, efficient water dispersibility, remarkable photostability, non-toxicity, biocompatibility, and superior chemical stability [15-17]. Despite these advantages, traditional CQDs face limitations in fluorescence yield [18, 19]. To address this issue, doping CQDs with heteroatoms such as nitrogen (N) and sulfur has been demonstrated to significantly enhance their optical properties, thereby improving their effectiveness for various applications [20, 21]. Because nitrogen has an atomic radius similar to that of carbon, and sulfur has a similar electronegativity to carbon, sulfur and nitrogen co-doped carbon quantum dots (N,S-CQDs) represent one of the most significant types of doped CQDs [22].

Building on these advancements, this study presents a simple, eco-friendly, selective, and sensitive fluorescence assay for detecting sudan III in foods. Unlike other studies that rely on analyte detection through the quenching effect, such as fluorescence resonance energy transfer [15, 23, 24] or the inner filter effect [25], our approach showcases a significant enhancement in the fluorescence of NCQDs/MIP upon the presence of sudan III. Notably, the sensor exhibits remarkable selectivity in detecting sudan III, even amidst the presence of other structurally similar sudan dyes, underscoring its high specificity.

2. MATERIALS AND METHODS

2.1. Materials

Chemical with analytical grades have been used: citric acid monohydrate ($C_6H_8O_7 \cdot H_2O$; 99.5%; Fisher Chemical^{TM}), thiourea (CH_4N_2O; 99.5%; Fisher Chemical^{TM}), sodium chloride (NaCl; 99.5%; Xilong, China), sodium hydroxide (NaOH; >96%; Xilong, China), sudan II, sudan III, tartrazine, ponceau 4R, sunset yellow FCF (98%, Sigma-Aldrich, USA).

2.2. Apparatus

The fluorescence measurement has been carried out on the fluorescence spectrometer F-4700 (Hitachi, Japan), with excitation wavelength of 360 nm and emission wavelength range from 380 to 600 nm. UV-Vis absorption spectra was recorded on the UV-1601 spectrometer (Shimadzu, Japan). N,S-CQDs were synthesized using MW3-MM25PE(BK) microwave, 25 L, 800 W (Toshiba, Japan).

2.3. Synthesis route of N,S-CQDs

N,S-CQDs were synthesized using a straightforward hydrothermal microwave-assisted method. Initially, 0.5 g of citric acid monohydrate and 0.5 g of thiourea were weighed and dissolved in 25 mL of deionized water. The mixture was then placed in a household microwave and irradiated for 7 minutes at a power of 800W until the solution turned black, indicating carbonization. Following this, an additional 25 mL of deionized water was added, and the solution was subjected to ultrasonication for 10 minutes to dissolve the newly synthesized QDs. The solution was then centrifuged at 6000 rpm for 5 minutes, filtered through a 0.22 µm microfilter, then was undergone dialysis for 24 hours at 25°C in order to eliminate the influence of impurities [26]. Then the dialyzed solution was lyophilized to obtain a brown-blackish solid of N,S-CQDs for storage at 4°C and further use.

The successful synthesis of N,S-CQDs material was proved by XRD, EDX, TEM analyses (provided in more details in another paper of our research group). XRD pattern of of N,S-CQDs indicates broaden peak at 2-theta of 23.3°. Additionally, TEM analysis indicates the amorphous form of CQDs and the spherical morphology of CQDs with the size lower than 10 nm. Hence, we provisionally conclude that the formation of N,S-CQDs was succeeded, even though the boundaries between CQDs and GQDs are not too obvious. As we can witness from EDX data, nitrogen and sulfur were doped in CQDs, therefore the term N,S-CQDs was used throughout our research.

2.4. Detection of sudan III by fluorescence spectroscopy using N,S-CQDs

1.5 mL of the N,S-CQDs solution (100 mg/L) was mixed with sudan III solution and deionized water to a total volume of 4.0 mL and incubated for 4 minutes at room temperature. The absorption spectra of the solution were recorded using a UV-Vis spectrophotometer in the range of 200-700 nm, and the fluorescence spectra were recorded at an excitation wavelength (λ_{ex}) of 360 nm, with the fluorescence intensity (F) measured at an emission wavelength (λ_{em}) of 440 nm. A blank solution of N,S-CQDs was prepared using the same procedure but did not contain sudan III, and its fluorescence intensity was recorded as F_0. The change in fluorescence intensity ($F - F_0$) was calculated for analysis and optimization.

2.5. Chili sample preparation

0.500 g of the sample (chili powder or chili sauce) is accurately weighed. The sample is subjected to hot extraction by being extracted three times with 3 mL of acetone each time, and the mixture is heated in a water bath at 80°C for 5 minutes. The extracts are combined into a 50.0 mL beaker, which is then placed in the freezer to precipitate the excipients from the chili sauce. After two hours, filter paper is moistened with cooled acetone and the cold extract is filtered through it. The filtrate is concentrated in a water bath at 80°C until it begins to boil. The concentrated mixture is then transferred to a 10.0 mL volumetric flask and diluted to the mark with an acetonitrile solvent mixture. Finally, the solution is filtered through a 0.22 μm sample filter.

3. RESULTS AND DISCUSSION

3.1. Fluorescence enhancement effect of N,S-CQDs in the presence of sudan III

The excitation-emission spectrum of N,S-CQDs is shown in Figure 1. The fluorescence spectrum displayed maximum emission intensity at 440 nm with an excitation wavelength of 360 nm. Based on these data, all subsequent fluorescence emission studies were carried out with an excitation wavelength of 360 nm and an emission wavelength of 440 nm for N,S-CQDs.
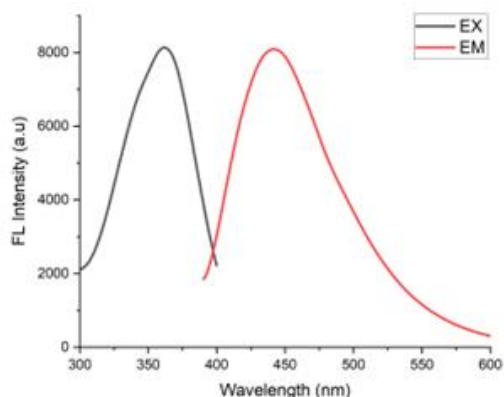
Figure 1. Fluorescence spectra of CQDs and N,S-CQDs

The quenching effect of CQDs in the presence of dyes is very well-known with several proposed mechanisms such as fluorescence resonance energy transfer (FRET) [15, 23, 24] or the inner filter effect (IFE) [25] whereas CQDs plays a role as radiation donor and dye as radiation acceptor. However, this quenching effect was observed by us only at high concentrations, when the color of sudan III was obvious (significant light absorbance). To our surprise, when the color of sudan III was not obvious (i.e, light absorbance is insignificant), instead of quenching, the enhancement effect dominates and provides the basis for quantification of sudan III at very low concentrations, which proved the better sensitivity of our method to traditional UV-Vis method (Figure 2a). Even though the enhancement effect was not frequently mentioned in literature, it still was reported in several studies and was referred to the initial passivation of surface defects at low concentrations

and saturation of surface traps on the N,S-CQDs at high concentrations, which destabilize the N,S-CQDs and lead to quenching [27, 28]. Although several studies have detected sudan III using fluorescence quenching, to our knowledge, there has been no report of using fluorescence enhancement for the detection of sudan III. Therefore, the fluorescence enhancement effect will be utilized for the detection and quantification of sudan III at low concentrations.
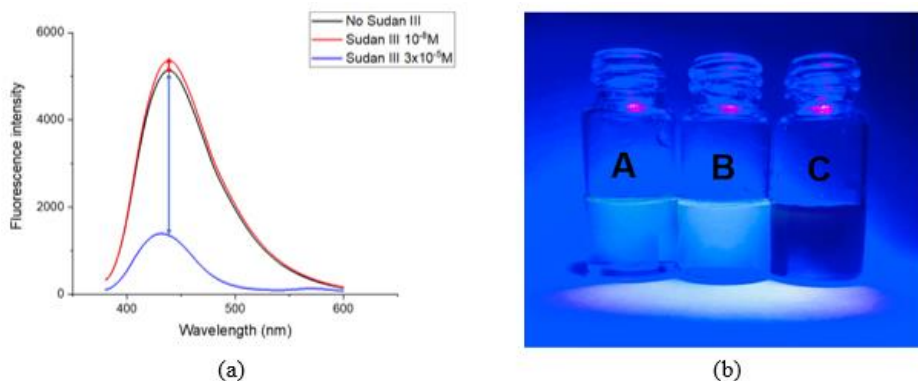
Figure 2. (a) Emission spectra of N,S-CQDs in the absence and presence of sudan III; (b) Solutions of A) N,S-CQDs, B) N,S-CQDs containing 10^{-8} M sudan III and C) N,S-CQDs containing 3 x 10^{-5}M sudan III under UV light

To further verify the fluorescence quenching and enhancement observed in N,S-CQDs solutions after the addition of sudan III, we irradiated three solutions with UV light: a solution containing only N,S-CQDs (solution A), a solution with N,S-CQDs and 10^{-8} M sudan III (solution B), and a solution with N,S-CQDs and 3 x 10^{-5} M sudan III (solution C) (Figure 2b). At high concentrations, the solution appeared black (solution C), indicating minimal fluorescence (fluorescence quenching), while at low concentrations, the solution turned slightly brighter with a green color (fluorescence enhancement, solution B).

3.2. Optimization of analysis conditions

The detection sensitivity of sudan III was optimized by adjusting various parameters, including pH, incubation time, and sudan III concentration. The optimized conditions were selected based on the degree of fluorescence enhancement (F - F_0), where F_0 is the fluorescence intensity of the N,S-CQDs solution without sudan III, and F is the fluorescence intensity of the N,S-CQDs solution containing sudan III.

3.2.1. Effect of pH

The pH value is a crucial factor affecting fluorescence enhancement in the presence of sudan III. As shown in Figure 3a, within the pH range of 3.0 to 10.0 and sudan III concentration of 7.5×10^{-9} M, the degree of fluorescence enhancement (F - F_0) increases with increasing pH, reaching a maximum at pH 5.0 before decreasing. This behavior is attributed to the optimal electrostatic interaction between the negatively charged surface of N,S-CQDs and the small positive charge of sudan III under mildly acidic conditions, which facilitates the adsorption of sudan III molecules onto the surface of N,S-CQDs. Therefore, a pH of 5.0 was chosen for subsequent experiments.
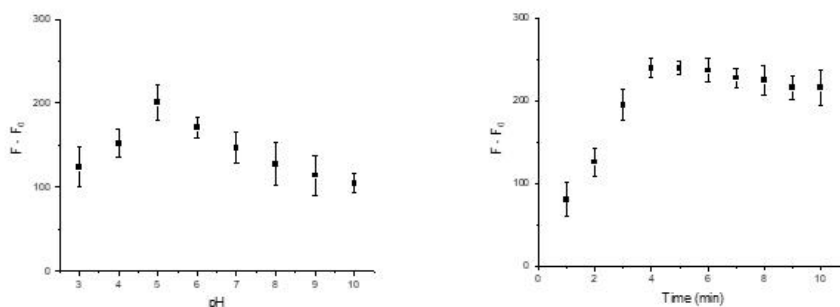
Figure 3. (a) Effect of pH and (b) effect of incubation time on detection of sudan III

3.2.2. Effect of incubation time

Incubation time is a critical factor for allowing sudan III to interact with N,S-CQDs. The incubation time of N,S-CQDs with 7.5×10^{-9} M sudan III was investigated over a range from 0 to 10 minutes. As shown in Figure 3b, the fluorescence intensity increased significantly within the first 4 minutes and then stabilized, indicating complete interaction between N,S-CQDs and sudan III within this period. Therefore, 4 minutes was chosen as the optimized incubation time. Notably, 4 minutes is a relatively short incubation time compared to other studies using CQDs as fluorescence sensors [28, 29].

3.3. Quantification of sudan III by N,S-CQDs

After optimizing the pH and incubation time for the detection of sudan III, the effect of sudan III concentration on the fluorescence enhancement signal was investigated in the range of 5×10^{-10} M to 2×10^{-8} M, at pH 5.0 and an incubation time of 4 minutes. As shown in Figure 4a, there is a linear relationship between the concentration of sudan III and the fluorescence enhancement in the range of 7.5×10^{-10} M to 1.0×10^{-8} M. A calibration curve constructed within this concentration range (Figure 4b) follows the mathematical expression $(F - F_0) = 2.7199$ C (x 10^{-10}) + 0.87, with a high correlation coefficient ($R^2 = 0.9997$).
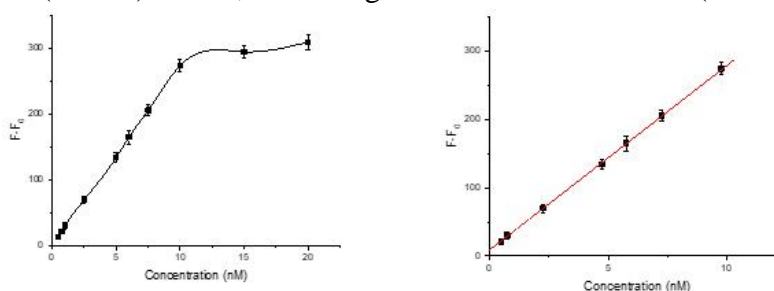
Figure 4. (a) Effect of sudan III concentration; (b) Calibration curve of sudan III

Using the equation LOD = $3S_y/b$, the limit of detection (LOD) calculated from the calibration curve is 2 x 10^{-10} M, corresponding to 0.07 ppb. This achieved LOD is significantly lower than most traditional methods for detecting sudan III, such as HPLC-UV and LC-MS/MS (Table 1). Therefore, our developed method has the potential to detect ultra-trace levels of sudan III in food samples, offering benefits of low cost and environmental friendliness. N,S-CQDs can be synthesized from inexpensive and environmentally friendly precursors using a simple and fast synthesis process that can be performed in any laboratory.

Table 1. *Detection limits of proposed methods for sudan III analysis*

| Method	LOD	Source (year)
Capillary electrophoresis	0.87 μM	[15] (2007)
Fluorescence spectroscopy Waste rubber tires CQDs	0.53 μM	[16] (2020)
HPLC-DAD	0.10 μM	[17] (2018)
PANI-PAN NFMs-HPLC-DAD	42.6 nM	[18] (2022)
HPLC – electrochemical detection	14.0 nM	[19] (2008)
Fluorescence test paper	2.89 nM	[20] (2022)
Electrochemical	2.62 nM	[21] (2019)
ELISA	1.53 nM	[22] (2008)
Fluorescence spectroscopy N,S-CQDs	**0.20 nM**	**This work**

The repeatability and accuracy of the method were evaluated through five replicate measurements at three different concentration levels: 7.5×10^{-10} M, 2.5×10^{-9} M, and 7.5×10^{-9} M. The relative standard deviations (RSD%) were below 5%, indicating good repeatability. The method recovery ranged from 91.5% to 103.5%, satisfying the requirements of the AOAC for trace analysis [30].

3.4. Selectivity

The method's selectivity was evaluated by analyzing sudan III at a concentration of 10^{-8} M in the presence of other synthetic azo dyes, such as Tartrazine (yellow), Ponceau 4R (red), sudan II (red), and Sunset Yellow FCF (yellow), each at the same concentration of 10^{-8} M (Figure 5a). The presence of these other azo dyes simultaneously with sudan III (Figure 5b), all at the above concentration, resulted in insignificant changes in fluorescence enhancement (Tartrazine: -0.67%; sudan II: -5.33%; Ponceau 4R: +8.00%; Sunset Yellow FCF: -6.67%), demonstrating that the detection of sudan III was not affected by the presence of other azo dyes. This indicates that N,S-CQDs exhibit good selectivity towards sudan III.

3.5. Real sample analysis

After successfully synthesizing and optimizing the N,S-CQDs composite for detecting sudan III, we applied this composite to analyze sudan III content in chili samples, including two chili sauces and two chili powders. These samples were obtained from various markets in Hanoi and all came from unknown brands, explicitly selected for their complex compositions containing ingredients like oil, salt, and food colorants. None of the samples contained detectable levels of sudan III. To further evaluate the sensor's capability to detect sudan III in food samples, sudan III standard solutions with varying concentrations were added to samples initially devoid of sudan III. Each spiked sample underwent triplicate analysis, and the average recoveries and relative standard deviations (RSDs) were calculated for each sample (Table 2).
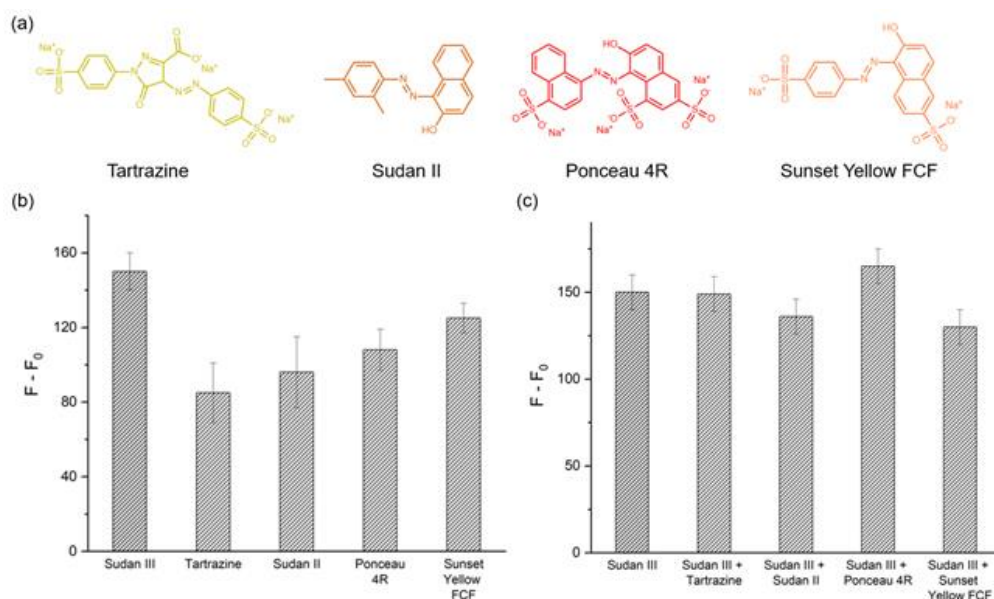
***Figure 5.** (a) Structures of Tartrazine, Ponceau 4R, sudan II and Sunset Yellow FCF; (b) Response of N,S-CQDs to sudan III and other dyes at the concentration of 10^{-8} M; (c) Response of N,S-CQDs to sudan III in the presence of other dyes at the concentration of 10^{-8} M*

The results demonstrate the promising potential of the proposed method in detecting traces of sudan III at the part per billion (ppb) level in food samples with high accuracy. The average recoveries of the analyzed samples ranged from 83% to 107%, with RSDs below 10%.

***Table 2.** Detected results of spiked sudan III*

Sample	Spiked (ppb)	Detected by proposed approach (ppb)	Recovery (%)	RSD (%)	Detected by LC-MS/MS (ppb)	Difference (%)
Chili powder 1	0	ND	-	-	ND	-
	20	17.09	85.45	4.3	-	-
	35	30.92	88.34	6.9	34.14	-9.4
	70	60.98	87.12	3.7	-	-
Chili powder 1	0	ND	-	-	ND	-
	20	16.69	83.45	5.1	-	-
	35	36.46	104.2	4.5	35.43	+2.9
	70	65.04	92.91	6.7	-	-
Chili sauce 1	0	ND	-	-	-	-
	20	18.91	94.55	2.5	-	-
	40	41.89	104.7	3.4	-	-
	100	104.2	104.2	4.1	-	-
Chili sauce 2	0	ND	-	-	ND	-
	20	21.41	107.5	4.7	-	-
	40	37.13	92.82	3.8	38.17	-2.72
	100	102.7	102.7	4.5	-	-

*ND: Not detected; '-': No information; Not analysed

Three food samples were analyzed using liquid chromatography–tandem mass spectrometry (LC-MS/MS), which revealed a detection limit (LOD) of 0.3 ppb. Sudan III was not detected these samples, consistent with our analysis using N,S-CQDs. Spiked samples were also cross-checked with LC-MS/MS and the variation between the results obtained by LC-MS/MS and our method was less than 10%. These findings affirm the reliability and effectiveness of our proposed method in accurately measuring the concentration of sudan III in food samples. Furthermore, the method we developed demonstrates prompt response times, ease of operation, and sensitivity comparable to that of LC-MS/MS.

Cost analysis mostly consists of the expense of chemicals and energy consumption for microwave-assisted hydrothermal synthesis of N,S-CQDs. Chemicals used for material synthesis are citric acid monohydrate, thiourea and deionized water. The prices for these chemicals and energy consumption are shown in Table 3. According to calculations, each run of fluorescence measurement cost only 1.113 VND, much more economical than mentioned analytical methods such as HPLC, LC-MS/MS since these instruments require large amount of costly, quite toxic solvents and intricate equipment. CQDs have also been proven to be environmentally friendly because the applied chemicals are abundant in nature, non-halogenated, non-metallic [15-17].

Table 3. *Estimate cost of analysis*

No.	Category	Original Price	Usage amount (each synthesis)	Final expense (VND)
1	Citric acid monohydrate	1.300.000 VND/1000 g	0.5 g	650
2	Thiourea	4.000.000 VND/1000 g	0.5 g	2.000
3	Deionized water	300.000 VND/1000 mL	50 mL	15.000
4	Electricity	1.728 VND/1000 Wh	95 Wh	164
	Sum (for 1 synthesis route)			17.814

4. CONCLUSION

In this study, we have developed a highly sensitive and selective fluorescence sensor using nitrogen and sulfur co-doped carbon quantum dots (N,S-CQDs) for the detection of sudan III. Our findings demonstrate that N,S-CQDs exhibit significant fluorescence enhancement in the presence of sudan III, achieving a detection limit of 2.0×10^{-10} M (0.07 ppb), which is considerably lower than the limits achieved by traditional methods such as HPLC-UV and LC-MS/MS. The optimized conditions for detection, including a pH of 5.0 and an incubation time of 4 minutes, facilitated efficient interaction between the N,S-CQDs and sudan III, enabling rapid and accurate quantification. The proposed method also exhibited excellent selectivity towards sudan III, even in the presence of other structurally similar azo dyes, and demonstrated high accuracy and reproducibility, with recovery rates ranging from 83% to 107% and RSDs below 10%. Real sample analysis of chili sauce and chili powder samples confirmed the method's applicability for detecting trace amounts of sudan III in complex food matrices. Overall, the N,S-CQDs-based fluorescence sensor offers

a cost-effective, environmentally friendly, and efficient alternative to conventional analytical techniques for monitoring sudan III contamination in food products. This method's simplicity and rapid response time make it suitable for routine food safety assessments and regulatory compliance, contributing to improved public health protection.

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