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Hans汉斯

基于碳点的四环素荧光检测

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摘　要

本文以柠檬酸和尿素为原料，通过微波法一步制备出荧光性能良好的碳点(CDs)。基于四环素与CDs之间的相互作用，构建了一种特异性检测四环素的荧光探针。采用紫外－可见吸收光谱和荧光光谱对CDs和四环素进行表征，发现四环素的紫外吸收光谱与CDs的荧光激发光谱之间存在的重叠区域，可能是由于CDs与四环素之间产生了荧光内滤效应(IFE)导致CDs荧光猝灭。讨论荧光体系溶剂组成、pH等单变量因素对CDs荧光猝灭效果的影响，在最佳检测条件下探究CDs荧光强度与四环素浓度之间的线性关系。实验结果表明，四环素的检出限为1.1×10^{-9} mol/L。该方法选择性好、灵敏度高，在特异性检测四环素领域具有较好前景。

关键词

碳点，微波法，荧光检测，四环素

Fluorescence Detection of Tetracycline Based on Carbon Dots

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Abstract

In this paper, carbon dots (CDs) with good fluorescence properties were prepared in one step by microwave method using citric acid and urea as raw materials. Based on the fluorescence quenching of CDs by tetracycline, a fluorescence probe was constructed to detect tetracycline specifically. CDs

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and tetracycline were characterized by UV-VIS absorption spectra and fluorescence spectra. It was found that the UV absorption spectra of tetracycline overlapped with the fluorescence excitation spectra of CDs to a large extent, which may be due to the fluorescence internal filtration effect (IFE) between CDs and tetracycline. The influence of univariate factors such as solvent composition and pH on the fluorescence quenching effect of CDs was discussed, and the linear relationship between fluorescence intensity of CDs and tetracycline concentration was investigated under the optimal detection conditions. The experimental results showed that the detection limit of tetracycline was 1.1×10^{-9} mol/L. The method has good selectivity and high sensitivity, and has a good prospect in the field of specific detection of tetracycline.

Keywords

Carbon Dots, Microwave Method, Fluorescence Detection, Tetracycline

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1. 引言

四环素是一类广谱抗生素，其抗菌范围涵盖了阴性细菌、螺旋体、专性细胞内细菌以及原生动物寄生虫[1]，主要应用于畜牧养殖业中[2]。随着抗生素的兴起，四环素畜禽养殖中使用呈逐年递增趋势。大量的四环素残留造成严重的生态风险和危害人体健康等环境问题[3]。为降低四环素的残留危害，建立设施齐全的检测机制，定期对畜牧养殖场的畜禽及市场内的动物源性食品进行抽查是必不可少的。目前应用于四环素残留检测的分析技术主要有：液相色谱－质谱法(LC-MS) [4] [5]、高效液相色谱法(HPLC) [6] [7]、微生物法[8] [9]、酶联免疫分析法(ELISA) [10] [11]、电化学法[12] [13]等。其中，HPLC、LC-MS 法对操作人员水平要求较高，仪器费用较昂贵；ELISA、微生物法耗时长；电化学法操作繁琐、重复性欠佳。相比之下，荧光分析法因其具有高灵敏度、高选择性、简便的优点，可用于实际样品的检测和可视化分析[14]。

碳点(CDs)是量子点的一种，尺寸通常在 10 nm 以下，具有成本低、合成途径简单多样、水溶性强、环境友好、生物相容性好、表面可功能化、荧光强度高等优良性能[15]-[17]。此外，由于其良好的荧光特性，CDs 已应用于生物体内成像[18] [19]、太阳能电池[20] [21]、光催化[22] [23]、光电器件[24] [25]、化学传感[26] [27]等领域。

由于 CDs 具有较大的比表面积，其表面分布着丰富活性位点和官能团，这些活性位点和官能团可以有选择性地与检测物通过静态猝灭效应(SQE)、动态猝灭效应(DQE)、光诱导电子转移(PET)、分子内电荷转移(ICT)、荧光共振能量转移(FRET)、荧光内滤(IFE)等荧光传感机制发生相互作用，使荧光探针的荧光信号发生变化，实现定性定量检测。截至目前，已有多项基于 CDs 的荧光探针被报道。Sathish 等[28]以三苯基磷为碳源合成了荧光量子产率高达 61%的绿光碳点并应用于亚细胞线粒体成像和四环素特异性检测，检出限为 1.5×10^{-6} mol/L。Gao 等[29]以柠檬酸为原料，采用简单的一步水热法合成了红光碳点，可作为荧光探针用于特殊的 Pt^{2+}、Au^{3+}和 Pd^{2+}的检测。Shan 等[30]用硼掺杂的碳点检测过氧化氢，检出限为 1×10^{-2} mol/L。由此可见，开发荧光性能良好的碳点作为荧光探针进行四环素检测具有极大研究价值。

本文对利用 CDs 作为荧光探针进行荧光检测进行了深入研究，我们采用了节能且环保的微波法[31]快速合成了碳点。四环素可特异性猝灭 CDs，识别过程不受溶液环境限制，可对各类水体进行检验，为四环素污染检测提供了新思路。

2. 实验部分

2.1. 材料与仪器

柠檬酸(纯度 ≥ 99.5%)、尿素(纯度 99.8%)购自北京化工厂。四环素(Tetracyclines, TC)、磺胺甲恶唑(Sulfamethoxazole, SMZ)、酒石酸泰洛星(Tylosin tartrate, TYT)、罗红霉素(Roxithromycin, ROX)、氯霉素(Chloramphenicol, CM)、盐酸左氧氟沙星(Levofloxacin hydrochloride, LEV)、红霉素(Erythromycin, ERY)、泰拉霉素(Tulathromycin, TULA)等购自上海麦克林生化科技有限公司，乙醇、盐酸、氢氧化钠，实验所用试剂均为分析纯，实验用水为去离子水。

UH-5300 紫外可见光谱仪(日本日立公司)；Cary Eclipse 荧光分光光度计(美国瓦里安公司)；H3-18K 台式高速离心机(湖南可成仪器设备有限公司)；M1-L213B 微波炉(广东美的厨房电器制造有限公司上海)；TENSOR II 傅里叶变换红外光谱仪(德国布鲁克公司)；BAS124S 电子天平(德国赛多利斯公司)；Talos F200X G2 场发射透射电子显微镜(美国赛默飞公司)。

2.2. 实验方法

2.2.1. CDs 的制备

CDs 的合成采用微波法。将 1 g 柠檬酸和 2 g 尿素溶解在 20 mL 去离子水中，搅拌均匀，形成无色透明的溶液；然后将混合后的溶液放在微波炉中，中火(700 W)加热 6 分钟，在加热的过程中，溶液由无色透明变成棕色，最后变为深棕色的固体，标志着 CDs 的形成；最后将深棕色的固体重新分散在 30 mL 无水乙醇中，离心 2 次，每次 10 分钟，离心速率为 8000 转每分钟。离心后保留上层清液，去除沉淀的大颗粒固体，得到 CDs 溶液。

2.2.2. 荧光分析法检测四环素

准确称取 0.0444 g 的四环素，将其溶解于 10 mL 无水乙醇中，配置成 1×10^{-2} mol/L 的四环素溶液。使用 1 mol/L 的盐酸和氢氧化钠固体调节 CDs 溶液 pH 至 3.3，用移液枪准确移取 0.30 mL CDs 溶液、0.30 mL 四环素溶液、2.40 mL 无水乙醇，混匀后测其荧光激发光谱，其中四环素溶液浓度为 1×10^{-3} mol/L。在 400 nm 的激发波长下，扫描发射光谱范围为 420~750 nm，测试并记录数据。

3. 结果与讨论

3.1. CDs 的表征

通过紫外可见吸收光谱对 CDs 结构进行表征，如图 1(A)所示，CDs 在紫外吸收区有明显的吸收，说明 CDs 中的 C=C 发生了 π→π*跃迁[32]。合成的 CDs 分散良好，如图 1(B)所示，粒径在 4.5~7 nm 之间。高分辨率 TEM (HRTEM)图像显示，大多数颗粒为无晶格的无定形碳颗粒和石墨；少数颗粒为准球形且具有清晰的晶格条纹，平均晶格间距为 0.21 nm。CDs 荧光量子产率为 14.1%，与已报道的 CDs 相比具有良好的荧光性能(表 1)。

3.2. 探究 CDs 荧光探针最佳制备条件

3.2.1. 溶剂组成对 CDs 荧光强度的影响

溶剂的极性对荧光有较大的影响，在不同的溶剂中有显著改变[43]。本文探究了在不同乙醇－水体积比下 CDs 的荧光强度。按表 2 依次配制水体积分数 fw 为 0%、15%、30%、45%、60%、75%、80%、84%、88%的待测液。在 400 nm 的激发波长下，比较各组样品最大发射波长处 CDs 荧光强度。结果如图 2 所示，纯乙醇体系情况下，CDs 荧光强度最强。随着水体积分数增大，CDs 荧光强度减小。这是由于

CDs 荧光发射由 π→π*和 n→π*跃迁引起，随着溶剂极性的增强，碳点 HOMO-LUMO 能隙逐渐减小，导致荧光发射峰逐渐红移。此外，CDs 表面非成键的孤对电子易与溶剂形成氢键，随着水体积分数增加，氢键作用加强，使 CDs 发生 π→π*跃迁概率降低，量子产率减少，荧光强度降低[44]。

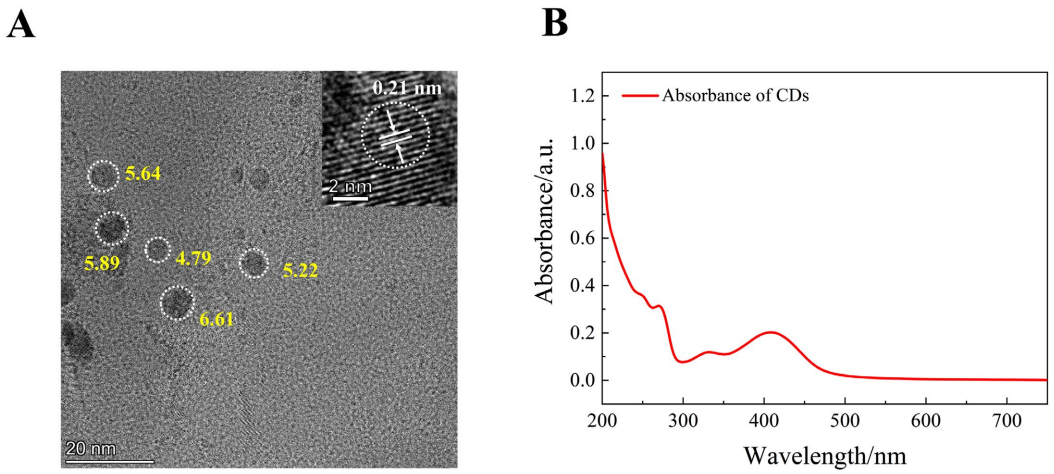

Figure 1. (A) TRTEM images of CDs; (B) UV-vis absorption spectra of CDs
图 1. (A) CDs 的 HRTEM 图像；(B) CDs 的紫外可见吸收光谱图

Table 1. Comparison of quantum yield of CDs
表 1. CDs 量子产率比较

CDs	Quantum Yield	Ref.
CQDs	28. 4 %	[33]
CM-CDs	32.81 %	[34]
Mn, Cl, N-CDs	5.3 %	[35]
CDs	23.7 %	[36]
N-CQDs	22.2 %	[37]
N-CDs	21.15 %	[38]
PTCDA-CDs	53.22 %	[39]
PCDs	10.2 %	[40]
N-CDs	45 %	[41]
PbCDs	4.1 %	[42]
CDs	14.1 %	本文

Table 2. Sample composition table
表 2. 样品组成表

CDs/mL	C_2H_5OH/mL	H_2O/mL	Bulk volume/mL	Volume fraction of H_2O/%
0.3	2.70	0.00	3.00	0
0.3	2.25	0.45	3.00	15
0.3	1.80	0.90	3.00	30

续表

0.3	1.35	1.35	3.00	45
0.3	0.90	1.80	3.00	60
0.3	0.45	2.25	3.00	75
0.3	0.30	2.40	3.00	80
0.3	0.18	2.52	3.00	84
0.3	0.06	2.64	3.00	88

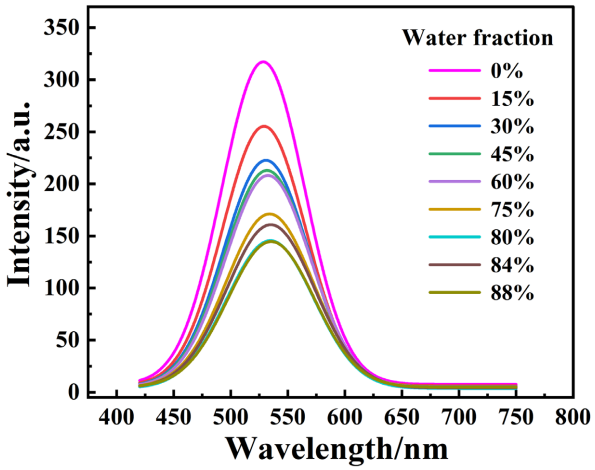

Figure 2. Fluorescence excitation spectra of CDs in different proportions of water-ethanol systems
图 2. 不同比例水–乙醇体系 CDs 的荧光激发光谱

3.2.2. 体系 pH 对 CDs 荧光强度的影响

采用 1 mol/L 的盐酸和氢氧化钠固体调节体系 pH，探究在不同 pH 值下(pH = 1.8~12.3)下 CDs 的荧光强度。结果如图 3 所示，CDs 荧光强度在 pH = 1.8~12.3 范围内保持着较高且稳定的状态，在 pH = 3.3 时，CDs 的荧光强度最大。

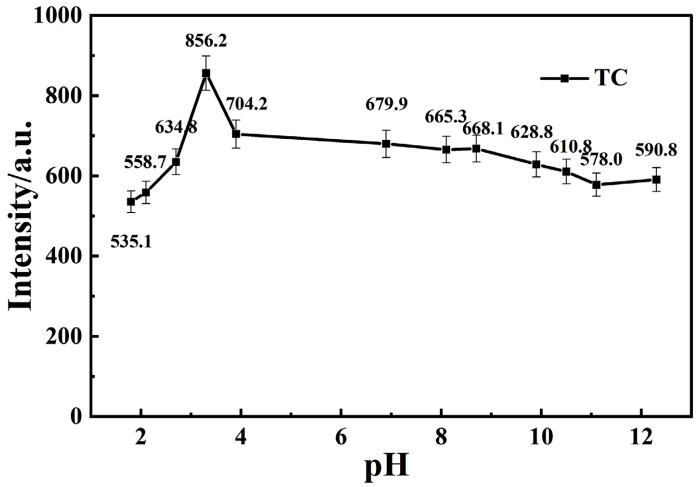

Figure 3. Fluorescence intensity of CDs at different pH
图 3. 不同 pH 下 CDs 的荧光强度

3.3. 四环素对 CDs 的选择性猝灭

以无水乙醇为溶剂，配制浓度为 1×10^{-2} mol/L 的四环素、磺胺甲恶唑、酒石酸泰洛星、罗红霉素、氯霉素、盐酸左氧氟沙星、红霉素、泰拉霉素溶液，所用剂量见表 3。用移液枪准确移取 0.30 mL CDs 溶液、0.30 mL 四环素溶液、2.40 mL 无水乙醇，混匀后分别测定其荧光发射光谱。调节荧光分光光度计激发波长为 400 nm，比较最大发射波长 511 nm 处 CDs 荧光强度的变化。由图 4 可知，仅 CDs 存在下，511 nm 处有明显的荧光发射峰。在四环素存在情况下，CDs 荧光猝灭程度最大，而除四环素外的其它抗生素对体系荧光强度猝灭程度较小，因此实现了在 511 nm 处对四环素的选择性猝灭。

Table 3. Quality of antibiotics
表 3. 抗生素质量

Antibiotic name	The quality weighted/g
四环素(TC)	0.0444
氯霉素(CM)	0.0323
磺胺甲恶唑(SMZ)	0.0253
盐酸左氧氟沙星(LEV)	0.0398
酒石酸泰洛星(TYT)	0.1982
泰拉霉素(TULA)	0.0806
红霉素(EPY)	0.0734
罗红霉素(ROX)	0.0837

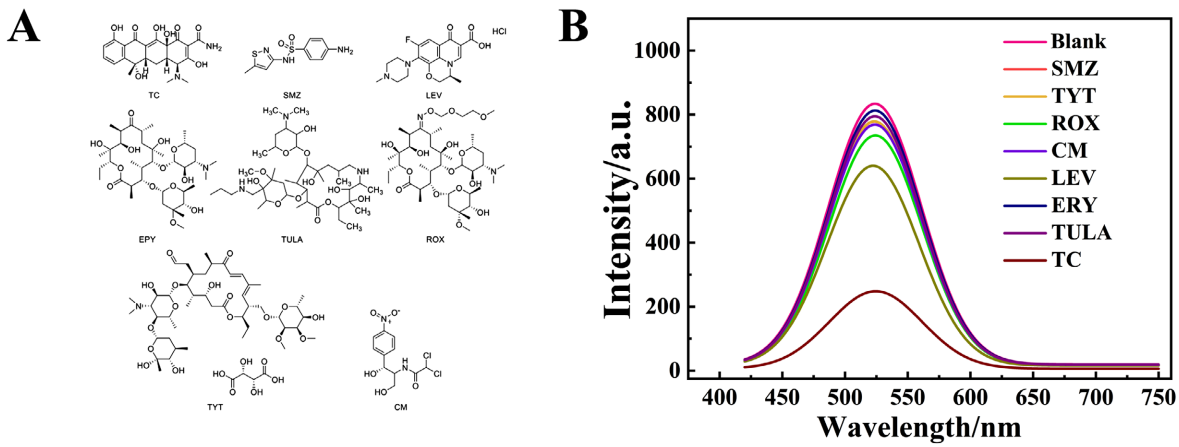

Figure 4. (A) Chemical structure of antibiotic drugs; (B) Fluorescence emission spectra of CDs after antibiotic solution with the same concentration was added
图 4. (A) 抗生素类药物的化学结构；(B) 相同浓度的抗生素溶液加入后 CDs 的荧光发射光谱图

3.4. 四环素对 CDs 的检测条件的优化

采用 1 mol/L 的盐酸和氢氧化钠固体调节体系 pH，以无水乙醇为溶剂，配制浓度为 1×10^{-5} mol/L 的四环素溶液。用移液枪准确移取 0.30 mL CDs 溶液、0.30 mL 四环素溶液、2.40 mL 无水乙醇，混匀后分别测定其荧光发射光谱。由图 5 可知，当体系 pH 为 8.6 时，CDs 荧光猝灭程度最大，因此后续实验均在 pH 为 8.6 的条件下进行。

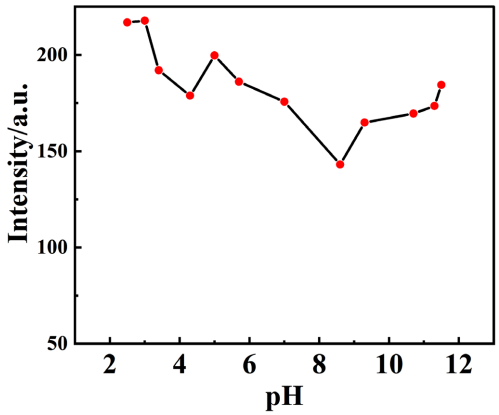

Figure 5. Fluorescence intensity of CDs after adding TC at different pH levels
图 5. 不同 pH 下加入 TC 后 CDs 的荧光强度

3.5. CDs 对四环素的荧光检测性能

以无水乙醇为溶剂，调节体系 pH 为 8.6，探究不同浓度四环素对 CDs 的猝灭程度。结果如图 6(A)所示，当四环素浓度在 1×10^{-8} mol/L~1×10^{-7} mol/L 的线性范围内时，随着四环素浓度的升高，CDs 荧光猝灭程度增大。如图 6(B)所示，线性方程为 $y = -4.85 \times 10^{8} x + 267.65$，线性相关系数为 $R^2 = 0.9944$，根据 $3\delta/k$ (δ 为标准偏差，k 为线性方程的斜率)计算该方法的检出限为 1.1×10^{-9} mol/L。与已报道的 CDs 探针相比(表 4)，本方法检测四环素残留具有可靠性，且具有经济性，为实际样品中四环素残留检测提供新思路。

3.6. CDs 对四环素的荧光检测机理

当吸收体的吸收光谱与荧光体的激发或发射光谱之间存在显著的重叠区域时，吸收体吸光度的变化可以相应地引起荧光体的荧光信号相应改变，这一转换过程即为荧光内滤效应(IFE) [48] [49]。选择红霉素作为典型非四环素类抗生素，测定四环素、红霉素的紫外可见吸收光谱和 CDs 的荧光激发光谱，结果如图 7 所示。可观察到，四环素的紫外可见吸收光谱与 CDs 荧光激发光谱存在部分重叠，四环素将作为吸收体将竞争吸收 CDs 的激发光，导致 CDs 的荧光猝灭，即发生 IFE 过程。相比之下，红霉素的紫外可见吸收光谱与 CDs 荧光激发光谱重叠程度较小，因此对 CDs 几乎无荧光猝灭作用。

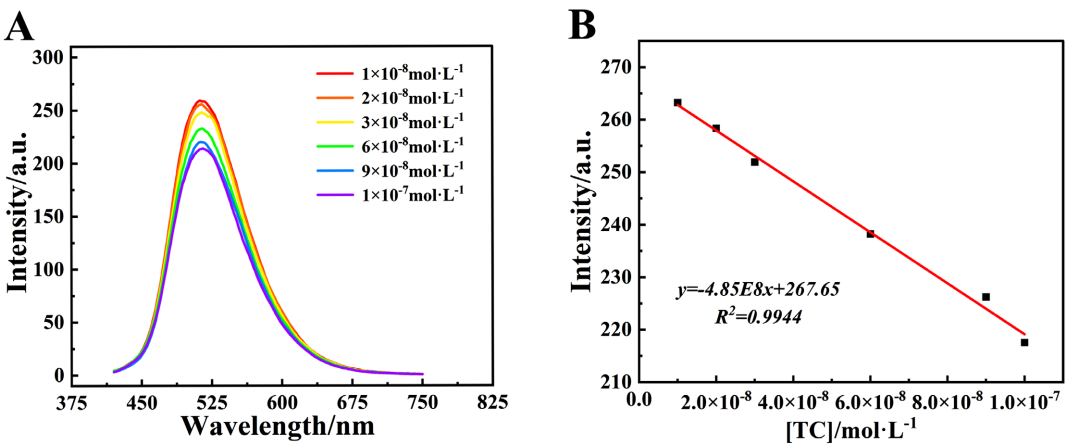

Figure 6. (A) Fluorescence emission spectra of CDs after antibiotic solutions with different concentrations were added; (B) Probe CDs was used to detect the linear relationship curve of tetracycline concentration
图 6. (A) 不同浓度的抗生素溶液加入后 CDs 的荧光发射光谱图；(B) 探针 CDs 检测四环素浓度线性关系曲线

Table 4. Comparison of tetracycline detection methods
表 4. 检测四环素方法比较

Fluorescent probe	Linear range (mol/L)	Limit of detection (mol/L)	Ref.
CDs	1.6×10^{-5}~3.9×10^{-3}	1.5×10^{-6}	[28]
R-CDs	4.0×10^{-5}~3.0×10^{-6}	3.9×10^{-9}	[45]
CDs-AuNCs	5.0×10^{-7}~4.0×10^{-5}	5.6×10^{-8}	[46]
N, Cu-CDs	1.0×10^{-5}~1.0×10^{-4}	3.7×10^{-9}	[47]
CDs	1.0×10^{-7}~1.0×10^{-8}	1.1×10^{-9}	本文

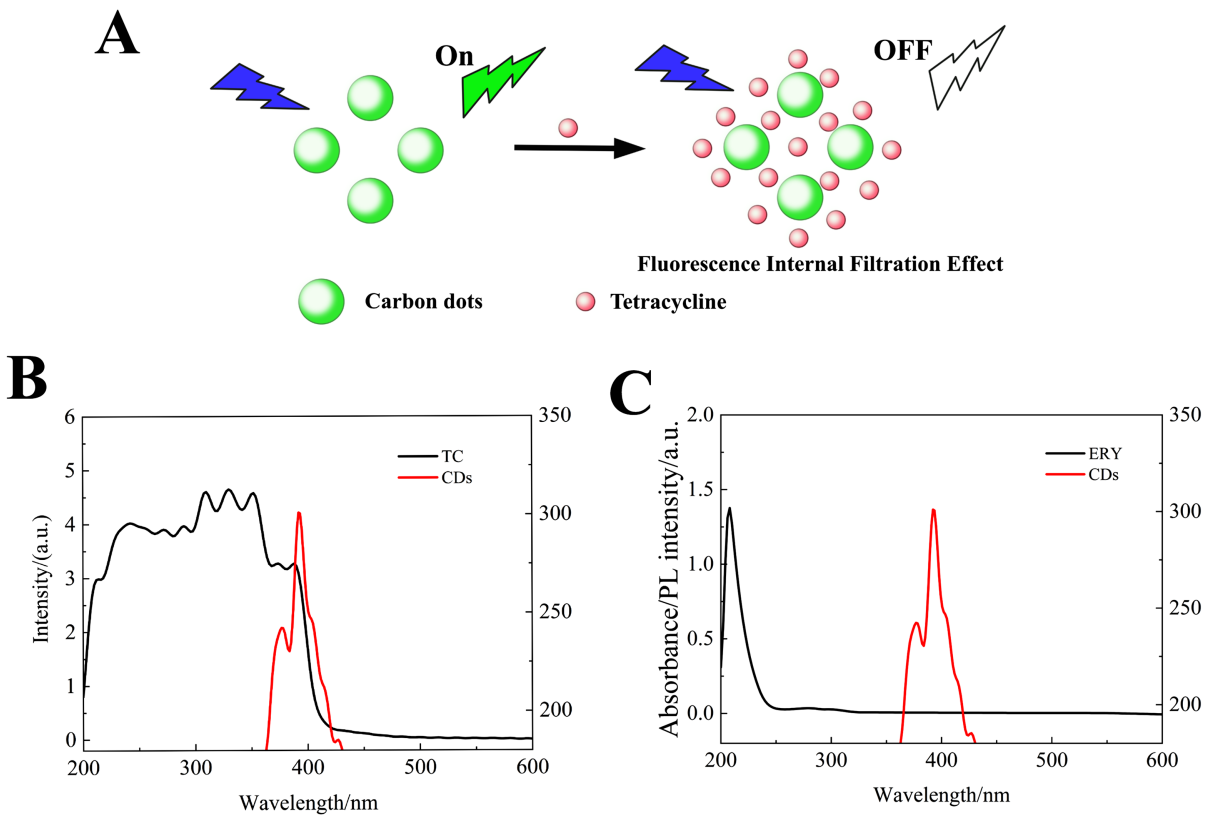

Figure 7. (A) UV-VIS absorption spectra of tetracycline and non-tetracycline antibiotics and fluorescence excitation spectra of CDs; (B), (C) Diagram of the mechanism of fluorescence internal filtration effect
图 7. (A) 荧光内滤效应机理图；(B)，(C)四环素和非四环素类抗生素的紫外可见吸收光谱和 CDs 的荧光激发光谱

3.7. CDs 对四环素的荧光检测的抗干扰能力

为进一步验证 CDs 对四环素的荧光检测的抗干扰能力，本文使用无水乙醇作为溶剂，在 CDs 溶液中加入等量的四环素与非四环素类抗生素如磺酸甲恶唑、酒石酸泰洛星、罗红霉素、氯霉素、盐酸左氧氟沙星、红霉素、泰拉霉素，最终四环素与非四环素类抗生素浓度均为为 1×10^{-3} mol/L。以不添加四环素类抗生素的溶液作为空白对照，在 400 nm 的激发波长下，比较 CDs 的荧光猝灭程度。如图 8 所示，与非四环素类抗生素混合后，四环素仍能对 CDs 产生较强的荧光猝灭效应，说明该 CDs 荧光探针抗干扰能力较强、可应用于实际样品中四环素含量的监测。

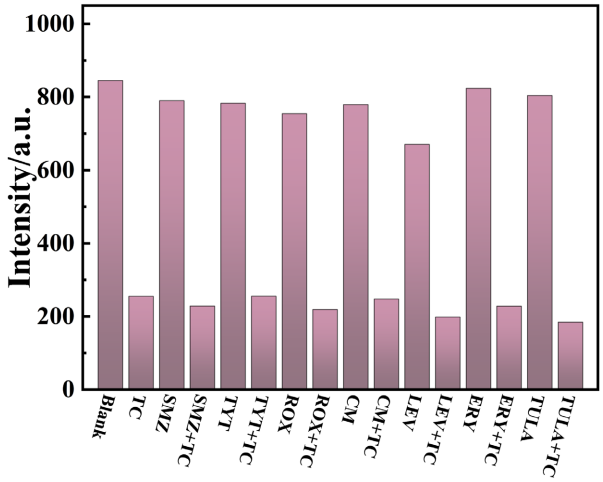

Figure 8. Fluorescence intensity of CDs after mixing tetracycline with other antibiotics
图 8. 四环素与其它抗生素混合后 CDs 的荧光强度

4. 结论

采用微波法一步合成了 CDs。基于四环素与 CDs 之间的荧光内滤效应，以 CDs 作为荧光探针，建立了一种快速、高效的荧光分析法。制备出的 CDs 荧光强度高且稳定，四环素对其可选择性猝灭，进而可将其应用在四环素的特异性检测中。为提高 CDs 检测四环素的灵敏度，探究了 CDs 荧光探针的最佳制备条件。实验结果表明，当以无水乙醇作为溶剂，体系 pH 在 3.3 左右时，CDs 荧光强度最大。检验 CDs 对四环素的荧光检测性能，当四环素浓度在 1×10^{-8} mol/L~1×10^{-7}mol/L 内，CDs 荧光强度与四环素的浓度呈现良好的线性关系，检出限为 1.1×10^{-9} mol/L。通过进一步表征，验证了四环素特异性猝灭 CDs 是 IFE 机理。此外，CDs 荧光探针抗干扰能力强、绿色环保，可在四环素检测中得到推广。

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