Physica E 72 (2015) 17–24

Contents lists available at ScienceDirect

Physica E

journal homepage: www.elsevier.com/locate/physe

Ratio-metric sensor to detect riboflavin via fluorescence resonance energy transfer with ultrahigh sensitivity

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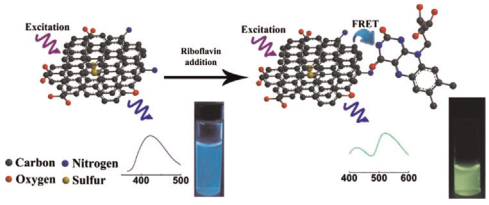
HIGHLIGHTS

- 1N, S doped carbon dots are developed to determine riboflavin in aqueous solutions.
- A novel fluorescence resonance energy transfer ration-metric method is applied.
- The probe obtains high sensitivity with a low limit of detection of 1.9 nM.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 14 March 2015
Received in revised form
5 April 2015
Accepted 7 April 2015
Available online 8 April 2015

ABSTRACT

In this paper, a novel fluorescence resonance energy transfer (FRET) ration-metric fluorescent probe based on heteroatom N, S doped carbon dots (N, S-CDs) was developed to determine riboflavin in aqueous solutions. The ratio of two emission intensities at different wavelengths is applied to determine the concentration of riboflavin (RF). This method is more effective in reducing the background interference and fluctuation of diverse conditions. Therefore, this probe obtains high sensitivity with a low limit of detection (LOD) of 1.9 nM (0.7 ng/ml) which is in the highest level of all riboflavin detection approaches and higher than single wavelength intensity detection (1.9 μM). In addition, this sensor has a high selectivity of detecting riboflavin in deionized water (pH=7) with other biochemical like amino acids. Moreover, riboflavin in aqueous solution is very sensitive to sunlight and can be degraded to lumiflavin, which is toxic. Because the N, S doped carbon dots cannot serve as an energy donor for N, S doped carbon dots and lumiflavin system, this system makes it easy to determine whether the riboflavin is degraded or not, which is first to be reported. This platform may provide possibilities to build a new and facile fluorescence resonance energy transfer based sensor to detect analytes and metamorphous analytes in aqueous solution.

1. Introduction

Fluorescence resonance energy transfer (FRET) is a non-radiative process between a donor and an acceptor. Briefly, the donor absorbs fluorescent energy to an excited state and then transfers energy to the acceptor at the ground state via long-range dipole–dipole coupling. According to Forster's theory, the efficiency of the FRET process is the quantum yield of the energy transfer transition, and is highly sensitive to many other factors like: the overlap between emission spectrum of a donor and absorbance spectrum of an acceptor, the quantum yield of the donor, and the relative orientations of two dipoles. The spatial distance between donor and acceptor is also an essential influential factor, and the FRET process is extremely sensitive to spatial distance on the nanometer scale (1–10 nm) [1]. These unique characteristics of

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http://dx.doi.org/10.1016/j.physe.2015.04.006
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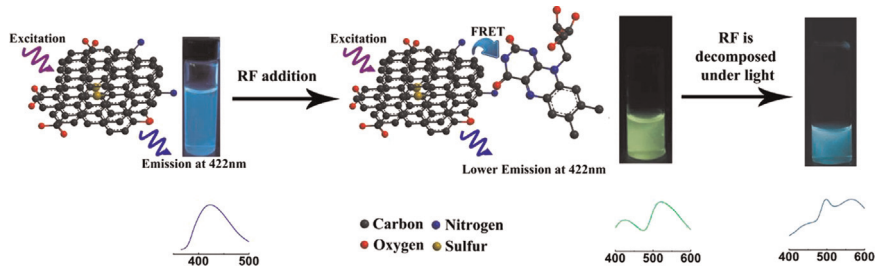

Scheme 1. Scheme of N, S-CDs-based RF probe.

Table 1
Quantum yield of the as-prepared N, S-CDs.

Sample	Integrated emission intensity (I)	Abs. at 360 nm (A)	Refractive index of solvent (η)	Quantum yield (ϕ)
QS	2504394589	0.0418	1.33	54% (known)
N, S-CDs	1842225171	0.0389	1.33	42.7%

FRET allows for enormous applications in diverse fields, such as: dynamics, molecular conformation study in biological molecules, [2–4] and sensing or diagnostic systems in analytical science [5–8]. In the last decade, intensive studies have been published including the detection of bio-molecules, [9,10] ions, [11] toxic gases, [12] and even pH [12] values through FRET processing with versatile donors and acceptors.

Riboflavin (RF) is one of the water-soluble vitamins that play a vital role in functioning healthy humans. RF also plays a crucial role in many physiological activities of cells such as nucleic acid repair process, cell apoptosis and electron transfer processes in the respiratory chain [13,14]. Because RF cannot be synthesized in the human body, inadequate dietary intake may lead to fatigue, slowed growth, digestive problems, angular cheilitis and anemia. In addition, RF is susceptible to be degraded to lumiflavin, which is harmful under sunlight exposure. Therefore, it is urgent to find a method to detect the concentration of RF and to differentiate RF and lumiflavin.

Conventional approaches for the determination of RF, such as microbiological assay, [15] spectrophotometric, [16] high

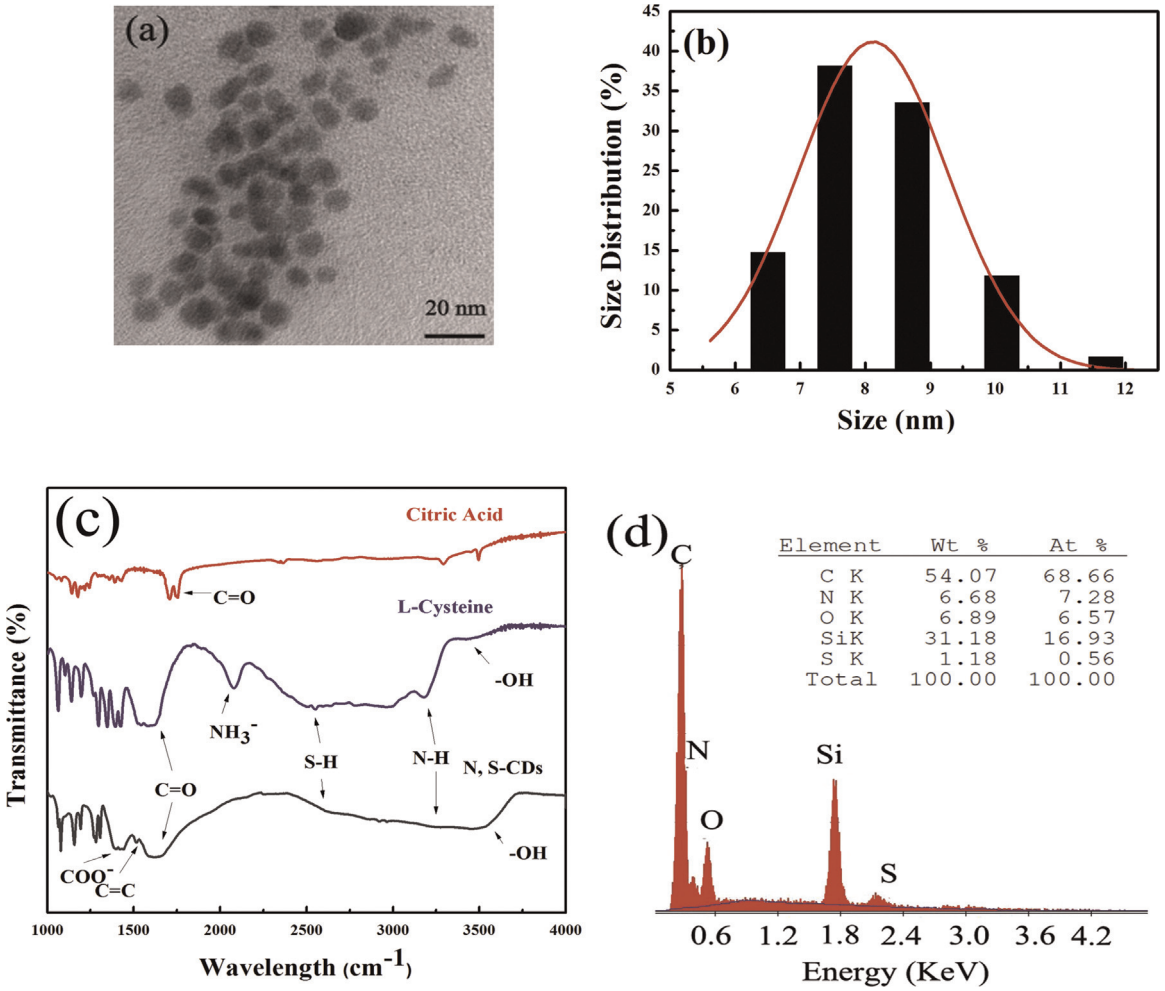

Fig. 1. (a) TEM image of N, S-CDs; (b) size distribution of N, S-CDs via Dynamic Light Scattering (DLS); (c) FT-IR spectra of citric acid, L-cysteine and N, S-CDs; (d) EDX result for N, S-CDs.