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Couple of Histamine blue fluorescence chemosensor and surface charge selector of FC-modified silica nanoporous for highly specific histamine detection via FRET-process

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ABSTRACT

Histamine poisoning from fish is a worldwide problem. It occurs when bacteria convert high levels of naturally occurring histidine into histamine resulting that when people eat fish containing high levels of histamine, they suffer allergic type reactions. Deeply considering, histamine and histidine consist of the primary amine and imidazole unit in their structures. As a result, the selectively discriminate detection of each species has been a greatly challenge task for determining the freshness of seafood. In the present work, the successful development of a new fluorescence sensors based on the superior Histamine blue (**HB**) doped in nanoporous silica (NPS) and surface modification with **FC** to provide FRET On-Off process and highly discriminate detection of histamine among other biogenic amines through the charge repulsion of **FC** and histidine and the specific reaction of mesoionic acid fluoride based Histamine blue (**HB**) with the amine group of Him via self-catalytic reaction of imidazole group in Him. This simple **HB@NPS@FC** sensory platform allows accurate determination of histamine in the linear response ranges of 29.12-166.67 μM of Him. The detection limit of the sensor was calculated equal to 8.55 μM. This **HB@NPS@FC** platform was investigated with success on real salmon and tuna samples. On the discovery of highly selective Him detection, the couple of two sensors and surface selector offers the potentially specific detection of Him in the field of food chemistry.

1. Introduction

Histamine (Him) is normally found in a postmortem of the muscle in fish, such as tuna and salmon. [1–3] The poisoning compound was easily produced from bacteria by decarboxylation of an essential amino acid, histidine (His). Ingestion food-borne usually contains high levels of Him leading to a variety of symptoms including nausea, vomiting, diarrhea, red rash, oral burning sensation and itching of the skin [1]. Therefore, the determination of Him in food is seriously concerned regarding as one of the biomarker in food quality for the food production, storage and transportation [4,5]. Most fresh fishes normally contain histidine as an essential amino acid and histamine as a poison compound. Specific discrimination of histidine (His) and histamine (Him) is a challenge task for sensing application, in particularly optical detection due to similar structure of both compound. Owning to a high efficient binding between Him and transition metal, most reports demonstrated the ligand exchanged sensing aspect of metal complexes, especially Ni^{2+} and Cu^{2+}. [6–11] There are very few reports demonstrating the covalent reaction between ligand and Him or His. As first exploration, Kielland and coworkers have fantastically reported the promising selective histamine sensing by the specific reaction of mesoionic acid fluoride based Histamine blue (**HB**) with the amine group of Him via self-catalytic reaction of imidazole group in Him (mechanism shown in Fig. S1). [12] Consequently, a large fluorescence change was observed only the case of Him. However, the authors did not report the fluorescence response of **HB** towards His. We have synthesized **HB** and tested the sensing ability with His and found that the fluorescent spectra of **HB** in the presence of His is identical spectrum to **HB**-Him complex. The analogue fluorescent spectra of **HB** towards Him and His indicated a non-specific binding with Him. (Fig. S2) Concerning the discriminated detection of Him without the interference of His, there has been rare in the previous reports because both compounds comprised of similar functional group of amine and imidazole. Due to the efficient **HB** as the superior fluorescence sensor for specific detection of Him and His among

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other amino acids, this inspired us to develop the new sensing system for specific detection of Him by using **HB** as the chemosensor. As a well-defined porous materials for sensing purposes, the nanoporous silica (**NPS**) was an attractive candidate for screening the proper compound passing through the porous. [13–19] Moreover, a few fluorescence chemosensors for biogenic amines sensing employed the fluorescence energy transfer (FRET) as a tool of detection. Therefore, we have designed the sensing system comprising two suitable sensing elements to undertake the FRET process for detection and the use of nanoporous silica as a solid support and gate-selector for specific analyte. This aspect brings us to engineer the NPS materials bearing the donor electron fluorescence sensor as the binding site and the proper electron acceptor fluorophore modified on the surface of NPS. Our motivation is to prepare the two fluorescence chemosensors based on NPS material: (i) **HB** as the binding site and a donor fluorophore, doped in the porous NPS would react with specific amino acid containing an imidazole moiety, namely, Him and His providing the emission band at 436 nm, (ii) the fluorescein dye (**FC**) was fabricated on the surface of NPS materials for taking a function of gate-selector because **FC** containing carboxylate group possibly enables to obstruct the His which has high total negative charge. Herein, this publication demonstrated a highly selective fluorescence system for determination of Him to give a strong FRET-on emission band at 526 nm corresponding to **FC**. We hypothesized that the specific analyte passing to react with **HB**-doped NPS provided the HB-guest complex doped in **HB@NPs@FC** which gave the emission band at 436 nm which was absorbed by **FC** fabricated on the **HB@NPs@FC** material to perform the FRET emission band at 536 nm. The concept design was illustrated in Fig. 1.

2. Experimental sections

2.1. Materials

Histamine blue was synthesized following the literature [9] and Nanoporous silica (NPS) were prepared by Hyehyeon Kim's method. [20] Chemicals and the solvents being standard analytical grade were purchased from Fluka, Aldrich, Carlo erba, Merck, TCI or Lab scan and used without further purification. Dichloromethane were distilled using calcium hydride as drying agent under nitrogen prior to use. Column chromatography was carried out using silica gel (Kieselgel 60, 0.063 0.200 mm, Merck). Thin layer chromatography (TLC) was performed on silica gel plates (Kieselgel 60, F_{254}, 1 mm). Dimethyl sulfoxide as AR grade used in fluorescence measurement was used without drying.

2.2. Apparatus

^{1}H NMR spectra and ^{13}C NMR spectra were recorded on a Varian Mercury 400 NMR spectrometer and a Bruker DRX 400 MHz nuclear magnetic resonance spectrometer. All chemical shifts were reported in part per million (ppm) using the residual proton or carbon signal in deuterated solvent namely $CDCl_3$ and DMSO-d_6. All fluorescence spectra were measured by a Varian Cary Eclipse Probe fluorescence spectrophotometer by personal computer data processing unit. The light source is Cary Eclipse a pulsed xenon lamp and a detector is a photomultiplier tube. All UV–vis spectra were recorded with a Varian Cary 50 Probe UV–vis spectrometer. TEM images were recorded on a transmission electron microscopy (JEOL, JEM-2100 electron microscope). X-ray diffraction pattern (XRD) was determined on D4 X-ray diffractometer using Cu Kα radiation (λ = 0.1541).

2.3. Synthesis of HB@NPS@FC

- **Synthesis of Histamine blue (HB)**

HB was synthesized according to the published procedure [12] Briefly, TFAA (0.393 ml, 2.78 mmol) and isocyanide (0.253 ml, 2.78 mmol) were added to a solution of isoquinoline (360 mg, 2.78 mmol) in anhydrous CH_2Cl_2 (5 ml) at −30 °C. The reaction was stirred for 3 min at this temperature, and then the cooling bath was removed and the reaction mixture was stirred for 14 h at room temperature. A saturated solution of Na_2CO_3 (10 ml) was added and the mixture was extracted with dichloromethane (2 × 5 ml). The combined organic extract was washed with brine (10 ml) and then dried over Na_2SO_4. The silica and Na_2SO_4 were added to the organic extracts with stirring for 1 h and then were filtered through Celite. The precipitate was washed with AcOEt, and the collected

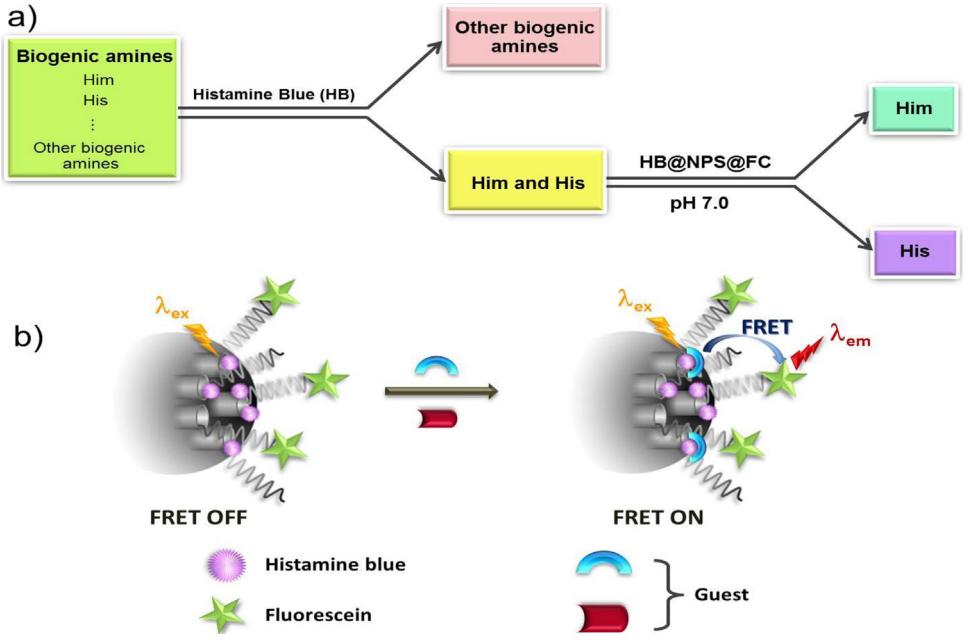

Fig. 1. a) concept for discrimination of biogenic amines, b) the conceptual illustration of fluorescence sensor for specific Him or His detection.