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Research paper

Metal-organic framework as luminescence turn-on sensor for selective detection of metal ions: Absorbance caused enhancement mechanism

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ABSTRACT

The luminescence turn-on sensors are more suitable for the practical applications, because they can effectively avoid the wrong response and have a higher signal-to-noise in dark background, compared to the luminescent quenching probes. In this work, we use IRMOF-3 as a luminescent probe to systematically investigate its sensing behavior for detecting metal ions. By examining the relationship of absorbance and luminescent intensity of IRMOF-3 before and after adding metal ions, we propose a novel luminescence turn-on mechanism, i.e., absorbance caused enhancement (ACE) mechanism, to reveal the luminescence turn-on phenomena of IRMOF-3 for selectively sensing M^{3+} (Cr^{3+} , Al^{3+} , Ga^{3+} , In^{3+}) metal ions. By exploring the effects of M^{3+} ion concentrations on absorbance and luminescent intensity, we further confirm the ACE mechanism responsible for luminescence turn-on phenomena. The subsequent experimental results indicate that IRMOF-3 exhibits not only high sensitivity and anti-interference ability against M^{2+} and M^+ metal ions, but also have excellent reusability and stability. In short, the ACE mechanism provides a possible strategy for designing luminescence turn-on probes.

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1. Introduction

With the rapid development of science and technology in recent years, more and more attention has been paid to the environmental pollution problems all over the world, especially for the pollutions induced by heavy metal ions. Therefore, it is very significant to selectively sense heavy metal ions for human health, and quickly identifying and detecting metal ions has become a current hot issue [1,2]. To date, a number of analytical methods for detecting metal ions have been developed, including capillary electrophoresis (CE), [3,4] electrochemical, [5,6] inductively coupled plasma mass spectrometry (ICP-MS), [7,8] atomic absorption spectroscopy (AAS), [9,10] liquid chromatography [11,12] and luminescence assay method [13–15]. Considering operability and cost-savings of the detection, luminescence based detection method has obvious advantages, like high sensitivity and selectivity, real-time response,

excellent photostability, local observation and easily be identified by naked eyes, which have been regarded as one of the most promising detection methods [16–18].

In the past few years, much effort has been devoted to rapidly detect heavy metal ions by luminescence quenching effect, and therefore the luminescent quenching mechanism had also been extensively studied [19–21]. Xu et al. reported a water-soluble luminescent hybrid composites for detecting Cu^{2+} [22]. Wu et al. reported Ag nanoclusters for sensing Hg^{2+} by luminescent quenching and found that the luminescent quenching mechanism was related to the anti-galvanic reduction process [23]. Zhao et al. used protein-functionalized Mn-doped ZnS quantum dots to selectively detect Cr^{3+} by luminescent quenching owing to the electron transfer from photo-excited Mn-doped ZnS QDs to Cr^{3+} [24]. Yan et al. fabricated a luminescent lanthanide functionalized MOF by encapsulating Eu^{3+} cations in the pores of MIL-53-COOH (Al) nanocrystals, and used it to detect Fe^{3+} via luminescent quenching caused by the cation exchange between Fe^{3+} and Al^{3+} in the framework MIL-53-COOH (Al) [25]. Wang et al. developed a conjugated polymer film sensor to detect Cu^{2+} ions via electron or energy transfer, because Cu^{2+} ion has higher thermodynamic affinity and can fast chelate with N atoms in ligand [26]. Cao et al. also inves-

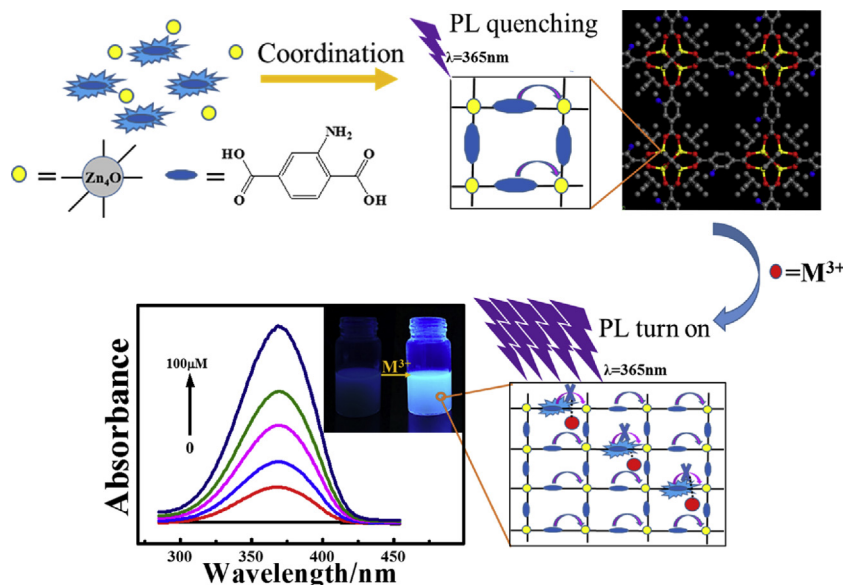
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Scheme 1. A schematic illustration of absorbance caused enhancement (ACE) mechanism of IRMOF-3 for luminescence turn-on detection of M^{3+} metal ions.

tigated the sensing behavior of porous covalent-organic polymer (COP) luminescent probes for detecting Fe^{3+} , and further revealed the absorption competition quenching (ACQ) mechanism of COP luminescent probes [27,28].

Compared to these luminescent quenching probes, luminescence turn-on sensors are more suitable for practical applications, because they can effectively avoid the wrong response and have a higher signal-to-noise in dark background [29,30]. A series of luminescence turn-on probes including nanoparticles, [31] quantum dots, [32] carbon dots, [33] covalent organic polymer, [34] metal organic framework, [35] have been reported to detect metal ions, small organic molecules and explosives. Li et al. reported a series of Ln-based luminescent probe materials exhibiting the luminescence turn-on effect [36,37]. Yuan et al. designed a kind of ruthenium(II) complex-based luminescence probes for detecting cysteine (Cys) and homocysteine (Hcy) by remarkable luminescence enhancement effect, which induced by the degradation of the strong electron withdrawing effects from Ru(II) complex after adding Cys and Hcy [38]. Song et al. synthesized a luminescent lanthanide MOF that can selectively detect DMF vapor by luminescent turn-on effect, which may be caused by DMF-ligand interactions and thus facilitates the LMET (ligand-to-metal energy transfer) process [39]. Cao et al. used MOF material Fe^{III} -MIL-88- NH_2 to sense hydrogen sulfide by luminescent enhancement effect, and elaborated luminescent turn-on mechanism based on collapse of MOF structure [30]. Fang et al. used amphiphilic Tb^{3+} complex as luminescent turn-on probe for sensing guanosine triphosphate (GTP), and revealed that the energy absorbed by GTP is transferred to the Tb^{3+} center and therefore causes luminescent enhancement [40]. Manos et al. presented an luminescent Mg-based MOF for real-time detection of water in tetramethylene oxide (THF) solvent through luminescence enhancement effect, and found that water molecules displaces the THF molecules into the pores of Mg-based MOF to induce the formation of a hydrated MOF [41]. Jiang et al. successfully synthesized a Pd(II)-porphyrinic MOF that can detect Cu^{2+} through luminescence enlargement, owing to the stronger binding affinity of Cu^{2+} over Pd^{2+} to the nitrogen atoms in the MOF [42]. Besides these mechanisms mentioned above, the luminescence enhancement also includes following aspects: (i) oxidation–reduction reactions by adding analyte; [43] (ii) reverse photo-induced electron transfer (reverse PET); [44] (iii) fluorescence resonance energy transfer (FRET) [45]. To our best

knowledge, the mechanism of luminescence enhancement induced by absorbance has not been reported.

In this work, as shown in Scheme 1, we used zinc nitrate hexahydrate ($Zn(NO_3)_2 \cdot 6H_2O$) and 2-amino-1,4-benzenedicarboxylate (BDC- NH_2) to synthesize IRMOF-3, and further explored its sensing applications in detecting metal ions. Based on absorbance changes of IRMOF-3 before and after adding metal ions, we proposed an absorbance caused enhancement (ACE) mechanism to reveal luminescence turn-on behavior of IRMOF-3 as the luminescent probe for selectively detecting M^{3+} (Cr^{3+} , Al^{3+} , Ga^{3+} , In^{3+}) metal ions. Finally, the sensitivity, anti-interference ability, stability and reusability of IRMOF-3 for sensing M^{3+} metal ions were also investigated.

2. Results and discussion

IRMOF-3 was synthesized at room temperature, and systematic characterizations of the as-synthesized sample were also performed. The experimental details were presented in Supporting Information. Fig. 1a shows the powder XRD pattern of the synthesized IRMOF-3, which is in good agreement with the simulated one [46]. The FTIR spectrum of IRMOF-3 in Fig. 1b displays several characteristic bands at 3483 cm^{-1} , 3365 cm^{-1} , 1576 cm^{-1} , 1377 cm^{-1} , 1252 cm^{-1} and 771 cm^{-1} , which are assigned to the asymmetric and symmetric stretching vibrations of $-NH_2$, $C=C$ in the benzene ring, $C-N$, $C-O$ and the out-of-plane bending vibration of Ar-H, respectively [47]. The XRD and FTIR results indicate that IRMOF-3 has been synthesized successfully. Fig. 2a show the SEM image of IRMOF-3, which clearly presents well-shaped cubic structure. TGA curves of IRMOF-3 in Fig. 2b shows two weight loss steps. The first step occurs at $160\text{ }^\circ\text{C}$ with 12.5% weight loss owing to the DMF solvent removed from IRMOF-3. The second weight loss happens at $360\text{ }^\circ\text{C}$ with about 40% weight loss, which may be attributed to the structure destruction of IRMOF-3 framework [48]. The porosity of IRMOF-3 was also characterized by the N_2 adsorption isotherm at 77 K in Fig. 2c. The BET specific surface area of IRMOF-3 is $354.81\text{ m}^2/\text{g}$, and the pore size distributions (see Fig. S1) indicated that IRMOF-3 is a mesoporous material with average pore size of 2.17 nm.

The photoluminescence (PL) emission spectra of solid IRMOF-3 and BDC- NH_2 were shown in Fig. S2. The excitation and emission peaks of IRMOF-3 are 379 nm and 450 nm, respectively; while they are 399 nm and 547 nm for BDC- NH_2 , respectively. Compared

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