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Novel metal ion-mediated complex imprinted membrane for selective recognition and direct determination of naproxen in pharmaceuticals by solid surface fluorescence

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ABSTRACT

A novel metal ion-mediated complex imprinted membrane (CIM) was prepared by immobilization of complex imprinted polymer (CIP) onto a polypropylene membrane. CIP was introduced as a novel imprinted material using a "complex template" constructed with Cu (II) ion and naproxen that could improve the selective recognition and enrichment properties of the membrane in water medium based on the coordination interaction rather than hydrogen bonding interactions, which could make CIP a promising material to mimic biological recognition process. A simple, sensitive and selective solid surface fluorescence method was proposed for the determination of naproxen in pharmaceuticals sample, using the CIM as the recognition material. Under the optimum conditions, the CIM exhibited large adsorption capacity and high selectivity to naproxen. A good linearity was obtained in the range of 0.50–20 mg/L with an estimated detection limit of 0.11 mg/L. Finally, the proposed method was applied to the analysis of naproxen in pharmaceuticals without complicated pretreatment. The recoveries were 85.0% and 89.1% and the RSDs were 9.2% and 12.0%. The results were consistent with that obtained by high performance liquid chromatography. CIM integrated extraction, concentration, and detection into one-step, which could make the analytical procedure more efficient.

1. Introduction

The field of modern analytical chemistry requires fast sensitive detection methods to meet the requirements of drug metabolism, disease diagnosis or environmental analysis. In this case, solid surface fluorescence method has received much attention because it enables very fast and sensitive determination for many substances at room temperature without cryogenic conditions because the luminescent molecules adsorbed on a solid substrate are isolate and collision-restricted [1,2]. However, one of the main drawbacks of solid surface fluorescence is its poor selectivity. To solve the problem, molecular imprinted polymer (MIP) was increasingly adopted for recognition process [3–5].

MIP is a kind of special material with high affinity and selective recognition to the template molecule. In general, MIP can be established relying on various interactions, among which hydrogen bond is the most frequently employed. However, its imprinting effect may be weakened or even damaged by strong polar solvents because the hydrogen bond is liable to be destroyed in these environments [6]. To widen the application of MIP especially in aqueous environment, one of the virtual methods is to prepare complex imprinted polymer (CIP) based on metal ion coordination interaction. CIP is a novel material, which has remarkable recognition properties for its "complex template" based on metal coordination interaction. In terms of directionality, specificity, and strength, the metal coordination interaction is stronger and more stable than hydrogen bonding or electrostatic interactions in polar system, resulting in good water-compatibility. Therefore, CIP is widely used in catalysis [7,8], separation [9,10], recognition [11,12], and sensors [13]. Besides, some metal ions can also enhance the fluorescence of analyte, and further increase the sensitivity. Nowadays the combination of CIP and solid surface fluorescence method has been explored by our group [14], which was applied in ethanol polar environment. Therefore, the exploration that CIP can be directly applied in aqueous samples is significant.

Naproxen (6-methoxy-α-methyl-2-naphthalene acetic acid) is a non-steroidal anti-inflammatory drug (NSAID) and was widely used to moderate pain relief in the treatment of many diseases [15]. Its chronic or acute administration shows toxic manifestations generally characteristic of NSAIDs, such as gastrointestinal erosion, bleeding and kidney failure [16–18]. Moreover, it may also increase the risk for cardiovascular events [19,20]. The development of a simple and sensitive method for the determination of naproxen in pharmaceuticals and biological fluids could be very useful for toxicological purposes. Up to now, several methods dealing with naproxen determination have been reported,

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including chemiluminescence [21,22], differential pulse voltammetry [23], capillary electrophoresis [24], gas chromatography mass spectrometry [25,26], and high-performance liquid chromatography (HPLC) with varied detection [27–30]. Although these methods have been successfully applied to analysis of naproxen in various matrices, tedious procedure, high cost and lack of selectivity are the main disadvantages.

In this paper, a simple, sensitive and selective solid surface fluorescence method was proposed for the determination of naproxen in pharmaceuticals based on CIM. The design, synthesis and characterization of the materials and their applicability were investigated. The proposed CIM could enhance the accessibility of naproxen to the imprinted cavities and improve the selective recognition properties in water medium, which makes the determination of naproxen in aqueous samples such as biological fluids possible and convenient.

2. Materials and methods

2.1. Materials and chemicals

Naproxen (NAP) was purchased from Yuancheng Pharmaceutical & chemical Co., Ltd. (Zhuhai, China). Ibuprofen (IBU), aspirin (ASP), 2-phenylpropionic acid (2-PPA), 3-phenylpropionic acid (3-PPA) and 4-acetamidophenol (4-AAP) were purchased from Jingchun Reagent Co., Ltd. (Shanghai, China). Ascorbic acid, uric acid and sodium chloride were obtained from Guangzhou Chemical Reagent Factory (Guangdong, China). Polypropylene membrane was purchased from Zhetao Industrial Co., Ltd. (Shanghai, China). Dimethyl sulphoxide (DMSO), azo(bis)-isobutyronitrile (AIBN) and cupric acetate ($Cu(OAc)_2$) were obtained from Damao Chemical Reagent Factory (Tianjin, China). Acrylamide (AM) was provided by Xilong Chemical Plant (Guangdong, China). Ethylene glycol dimethacrylate (EGDMA) was from Corel Chemical Plant (Shanghai, China). Acetonitrile was HPLC grade (Sigma-Aldrich, USA). Other chemicals were of analytical grade. All aqueous solutions were prepared using ultra-pure water obtained from a Millipore purification system. The pH value of aqueous solutions was adjusted using hydrochloric acid (HCl) or sodium hydroxide (NaOH) from Damao Regent Plant (Tianjin, China).

2.2. Preparation of CIM

NAP (230.0 mg), $Cu(OAc)_2$ (90.6 mg), AM (142.2 mg), EGDMA (3.78 mL) and AIBN (76.8 mg) were dissolved in 40.00 mL DMSO by sonication for 10 min at room temperature. A piece of polypropylene membrane was soaked in the polymerization solution for 5 min. Subsequently, the membrane was taken out and then deposited between two cover glasses as a "sandwich" which was transferred to a weighing bottle. The weighing bottle was deoxygenized with nitrogen for 5 min and then radical polymerization was performed thermally at 65 °C for 6 h in an oven. Finally, the CIM was washed with methanol/acetic acid (9/1, v/v) to remove the template. A molecular imprinted membrane (MIM) was synthesized in the same process of CIM preparation without adding $Cu(OAc)_2$, and also a non-imprinted membrane (NIM) was prepared by adopting the same protocols excluding both $Cu(OAc)_2$ and NAP.

2.3. Characterization of imprinted membranes

A Quanta 600 scanning electron microscope (FEI, Netherlands) was used to investigate the surface morphology of the membranes. A NICOLET AVATAR 330 Fourier transform infrared (FT-IR) spectrometer (Nicolet, Waltham, USA) and a thermal gravity (TG) analyzer (Netzsch-209, Bavaria, Germany) were applied to study the composition and the thermal stability of the coatings, respectively. The HPLC system assembled from Shimadzu LC-20A (Shimadzu, Japan) consists of a model LC 20AB pump, Diamonsil (TM) C_{18} column (250 mm × 4.60 mm I.D, 5 μm) and a model SPD-20A UV detector. The buffer solution was 0.1% acetic acid (v%) in water. The mobile phase was acetonitrile/buffer (80:20, v/v) at the flow rate of 1.0 mL/min and the detector was set at 254 nm.

Fluorescence measurements were performed on a RF-5301PC Spectrofluorophotometer (Shimadzu, Japan). The experimental conditions are as follows: the slit widths of the excitation and emission were both 5 nm, and the excitation wavelength was set at 284 nm with a recording emission range of 300–500 nm while the fluorescence intensity at 352 nm was chosen for quantification. To obtain fluorescence measurements, imprinted membranes were adhered onto the sample disc to record the solid surface fluorescence spectra.

2.4. Fluorescence response of imprinted membranes

NAP-Cu(II) (NAP:Cu(II)=2:1, mol:mol) (3.00 mL) standard aqueous solutions of various concentrations were added to a weighing bottle. Then a piece of membrane was immersed and incubated under shaking for 30 min. Subsequently, the membrane was taken out, washed with ethanol twice and then dried by nitrogen gas. The in situ solid surface fluorescence spectrum was recorded immediately. The fluorescence response of imprinted membrane to NAP (I_{NAP}) or mixture of NAP and its analogs (I_{mix}) for each concentration was calculated as the difference between the emission fluorescence intensity at 352 nm after and before recognizing target molecule. After each assay, the single-use membrane was discarded.

2.5. Selectivity of CIM

The selective recognization ability of CIM was evaluated by using IBU, ASP, 2-PPA, 3-PPA and 4-AAP, which were chosen as competitive agents since their chemical molecular structures are similar to NAP to a certain extent. The concentration of NAP-Cu(II) and each of its analogs-Cu(II) aqueous solution was 25 μmol/L. Besides, the mixture solution was prepared with an equal amount of NAP and IBU, ASP, 2-PPA, 3-PPA and 4-AAP. Selectivity test was also performed on both MIM and NIM for comparison.

2.6. Sample preparation

Two capsules of naproxen were both purchased from the local drugstore. First, a suitable weight of the drugs was transferred into a 50 mL volumetric flask. The volume was adjusted with ultra-pure water and the solution was sonicated and filtered by 0.45 μm nylon membrane. Then, the filtrate was properly diluted in order to make sure that the concentration of naproxen was within the linear range of the method. Finally, analysis was conducted by the procedure described above. The spiked contents were set at the same level according to the labeled amount. A triplicate determination of each concentration was conducted.

3. Results and discussion

3.1. Synthesis of CIM

Metal ions have the ability to bind to a wide range of functional groups through the donation of electrons from the heteroatom of ligand to the unfilled orbit of the outer coordination sphere of the metal [31]. Therefore, for metal ion mediated imprinting, the template first reacted with metal ion to form complex template, and then the complex template was allowed to establish interactions with functional monomers. Consequently, metal coordination was employed as an alternative of association between

template and functional monomer in the construction of imprinted polymers [31]. Subsequently, the resulting substance was copolymerized with cross-linkers in the presence of a free radical initiator. After removal of the template, imprinted cavities were available for rebinding template (Fig. 1A).

In this work, CIP was synthesized through free radical polymerization and immobilized on a polypropylene membrane through one-step reaction to obtain the metal ion-mediated CIM. The preparation procedure was simple in designing and preparation. Moreover, CIM could directly be used to enhance the solid surface fluorescence intensity of analyte (Fig. 1B).

The molar ratio between cupric acetate and naproxen was chosen according to the previous reports [32,33]. To prepare homogeneous polymer, appropriate porogen was very important. DMSO was selected as the porogen, because it can solubilize the reagents (complex template, initiator, monomers and cross-linker) to obtain homogeneous polymerization solution as well as to produce adequate porosity. AM, MAA and 4-Vpy were selected as monomers to investigate the effect on the fluorescence response (Fig. S1). It could be seen that CIM produced the strongest fluorescence intensity when AM was used as monomer. It may be because it meets the requirement of interaction energies between complex template and the monomer [34]. The polymerization time was also studied (Fig. S2). It showed that the fluorescence intensity was accelerated along with the increase of reaction time till 6 h and then decreased. Short reaction time produced few specific sites, whilst long reaction time would cause higher crosslinking degree of polymer which may block the imprinted cavities. The reactivity ratio between the cross-linker and monomers would influence the homogeneity of the polymer [35,36], which may have an effect on the solid surface fluorescence intensity. Therefore, the reactivity ratio between the cross-linker and monomers was studied by varying the amount of EGDMA (Fig. S3). The results revealed that the strongest fluorescence intensity was reached when the reactivity ratio between cross-linker and monomers of 10 mmol/1 mmol was used. It could be explained that small reactivity ratio produced a less dense polymer with few specific sites for binding naproxen. However, large reactivity ratio resulted in reduction of the distance between polymeric chains, which may block the imprinted cavities.

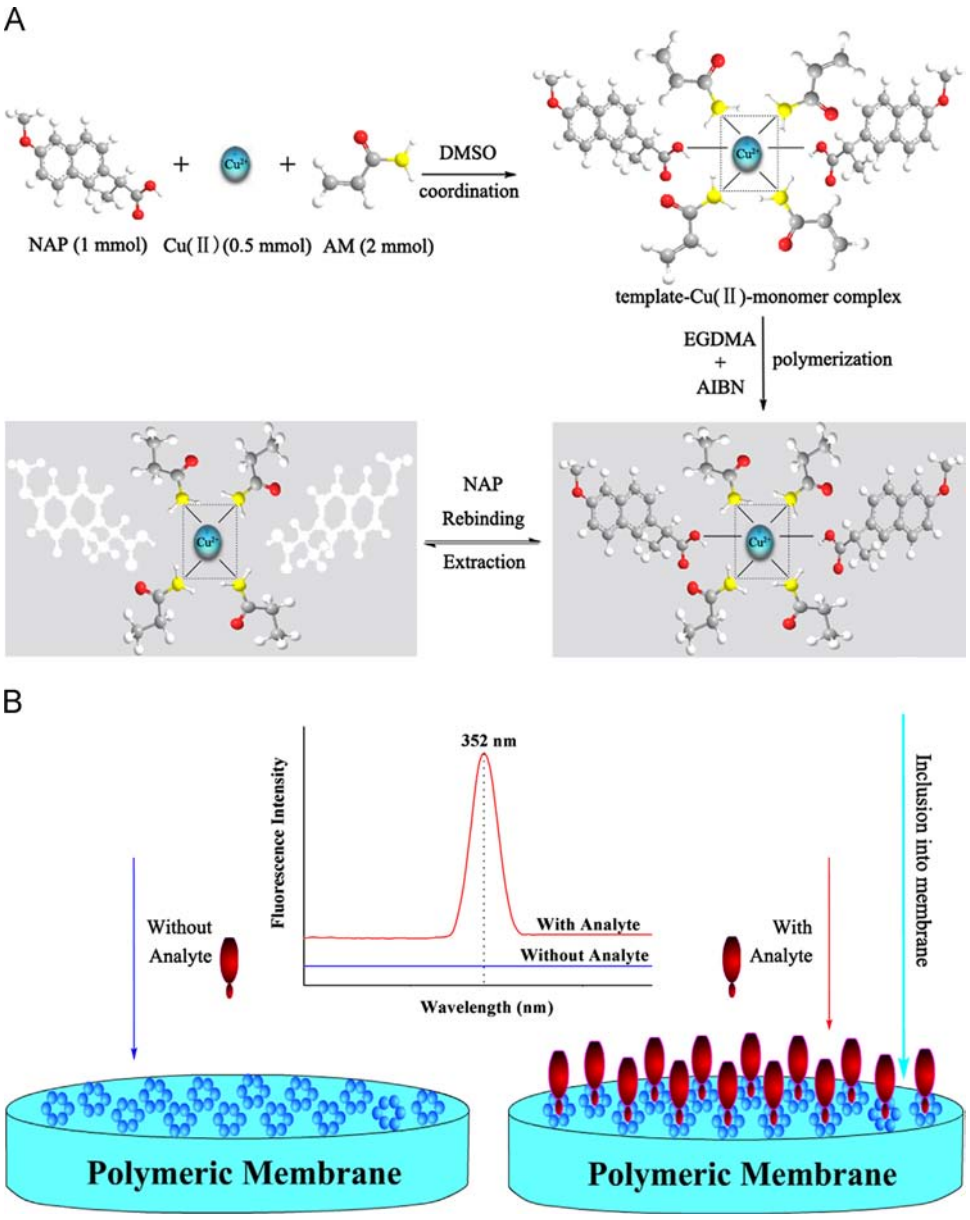

Fig. 1. (A) Schematic illustration of metal ion mediated imprinting and rebinding of naproxen and (B) the fluorescence detection of naproxen.

3.2. Characterization of imprinted membranes

3.2.1. SEM characterization

The SEM images of imprinted membranes and the substrate are provided in Fig. 2. As can be seen, the apparent difference of morphology between polypropylene membrane and CIM clearly showed the successful chemical modification of the substrate. It was revealed that the surface morphology of CIM, MIM and NIM were homogeneous and porous, which was beneficial to mass transfer.

3.2.2. IR characterization

FT-IR spectra of CIM, MIM and NIM are compared in Fig. S4, which indicated that these materials had the same adsorption

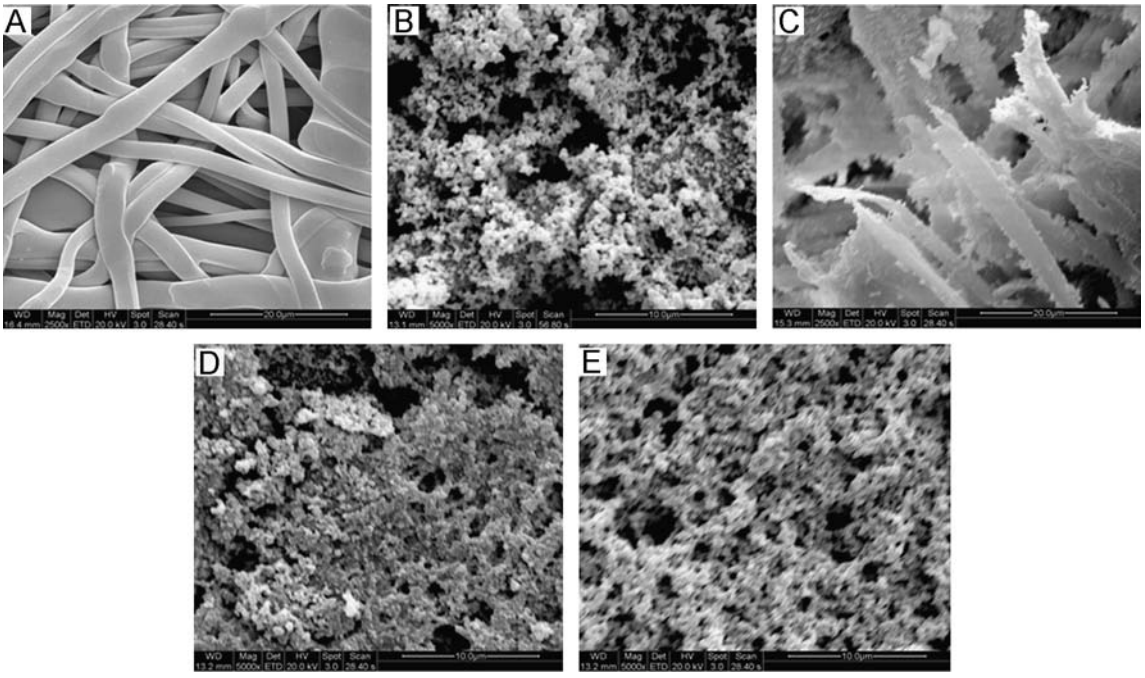

Fig. 2. SEM images of (A) polypropylene membrane, (B) CIM, (C) CIM cross-section, (D) MIM, and (E) NIM. Magnifications: (A, C) 2500 ×, (B, D, and E) 5000 ×.

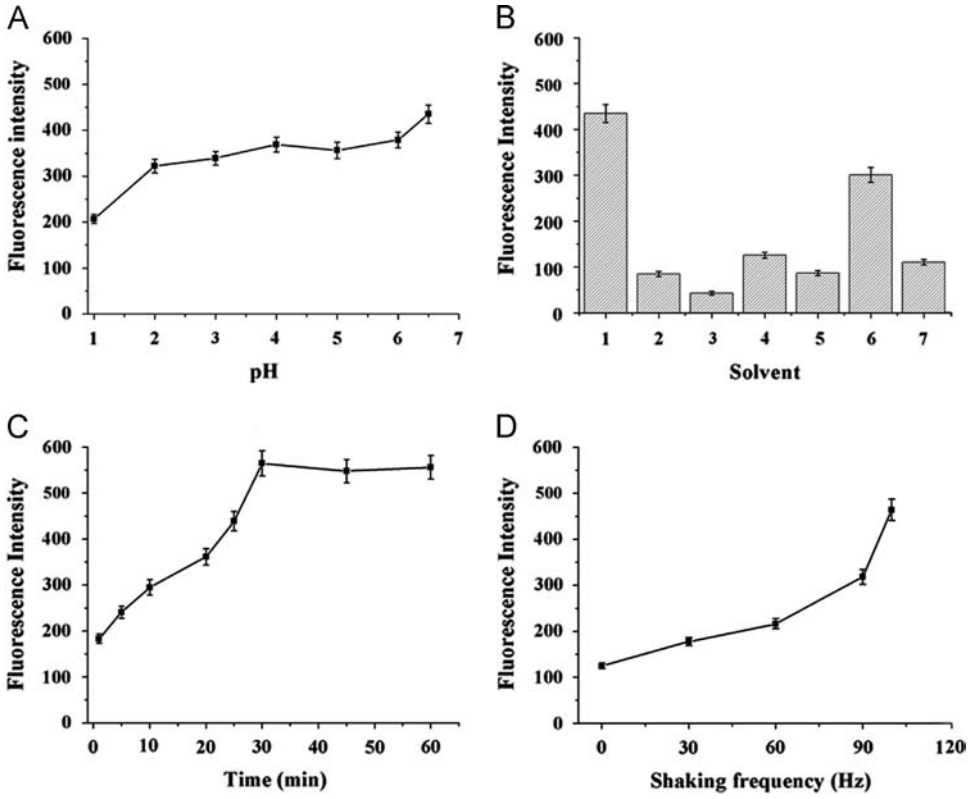

Fig. 3. Influences of CIM fluorescent response to 10.0 mg/L NAP-Cu(II). (A) pH and (B) incubation solvents: (1) ultra-pure water, (2) DMF, (3) DMSO, (4) ethanol, (5) acetonitrile, (6) hexane, and (7) dichloromethane. (C) Incubation time and (D) shaking frequency.

bands. The strong and broad peak around 3435 cm^{-1} indicated the N–H stretch. Other observed bands at 2984 cm^{-1} and 2954 cm^{-1} corresponded to the C–H stretching. The presence of the bands at around 1730 cm^{-1} was the C=O stretching vibration. The bands at 1456 cm^{-1}, 1389 cm^{-1}, 1258 cm^{-1} and 1156 cm^{-1} were the C=C stretching vibration, C–H bending vibration, C–O stretching and C–O–C asymmetric stretching, respectively. The results indicated that CIM, MIM and NIM had the similar chemical constitution.

3.2.3. TG characterization

The thermal stabilities of CIM, MIM and NIM were investigated with the thermogravimetric analysis. The results indicated that the weight loss curve and corresponding 1st derivatives of CIM were similar with both MIM and NIM (Fig. S5). The obvious mass loss occurred at about 220 °C for all membranes, and the apparent mass loss for CIM, MIM and NIM occurred at 343.1, 350.5 and 354.5 °C, respectively.

3.3. Influences of experimental conditions on CIM fluorescence response

3.3.1. Effect of pH

The pH value affects the dissociation of complex template and thus has great effect on the fluorescence intensity of CIM. Fig. 3A indicated the fluorescence intensity of CIM after being immersed in 10.0 mg/L NAP-Cu(II) solution at different pH values. As can be seen, the fluorescence intensity of CIM decreased as the pH decreased because low pH value would lead to the dissociation of the NAP-Cu(II) complex, resulting in low specific recognition. Besides, Cu(II) ion may undergo hydrolysis in alkaline solution. Finally, ultra-pure water (pH 6.5) was selected for further experiments, which means CIM can be directly applied in water medium.

3.3.2. Incubation solvent

Ultra-pure water, dichloromethane, dimethyl formamide (DMF), DMSO, ethanol, acetonitrile, and n-hexane were chosen to investigate the influence of incubation solvent on the fluorescence intensity of CIM to 10.0 mg/L NAP-Cu(II) solution (Fig. 3B). The results illustrated that CIM had the strongest fluorescence when water was used as the incubation solvent. This phenomenon confirmed that aqueous environment did not interfere with the selective recognization ability of CIM due to its metal coordinate interaction, which makes the determination of naproxen possible in aqueous samples such as biological fluids.

3.3.3. Incubation time

The response equilibrium of CIM was investigated in a solution containing 10.0 mg/L NAP-Cu(II) and it was observed (Fig. 3C) that the fluorescence intensity was accelerated along with the increase of incubation time until equilibrium was reached at 30 min. The solid surface fluorescence spectrum was recorded immediately after incubation of 30 min because the fluorescence intensity of CIM decreased slightly as the storage time increased (Fig. S6).

3.3.4. Influence of shaking frequency

The influence of shaking frequency on fluorescence intensity of CIM was studied at 0, 30, 60, 90 and 100 Hz with 10.0 mg/L NAP-Cu(II) solution (Fig. 3D). It was found that the fluorescence intensity of CIM increased along with the increase of shaking frequency due to faster kinetics. Shaking frequency over 100 Hz would cause damage of the membrane. Hence 100 Hz was chosen as the optimized shaking frequency.

3.4. Binding capacities of CIM, MIM and NIM

Under the selected conditions, a series of NAP-Cu(II) standard aqueous solutions with concentration from 0.50 to 50.0 mg/L were prepared to evaluate the fluorescence response of CIM, while MIM and NIM were chosen for comparison (Fig. S7). A notable observation is that fluorescence intensity increased gradually along with the increase of concentration of standard solutions. Generally, the fluorescence intensity depends on the adsorptive affinity of membrane to the complex template. It was found that CIM had better affinity to NAP-Cu(II), which can be concluded from the phenomenon that higher intensity was observed for CIM than both MIM and NIM. In the case of CIM, the fluorescence intensity increasing was mainly achieved by the affinity of the imprinted cavities with template due to the metal coordination interactions between NAP-Cu(II) and monomer. The MIM and NIM had similar affinity to NAP-Cu(II) because hydrogen bond interaction was dominant for MIM recognition. The strong polarity of solvent would disturb the hydrogen bond interaction between naproxen and monomer and result in weak imprinting effect of MIM. When the fluorescence response of imprinted membrane was saturated, the maximum intensity of CIM was 1.9 times as much as both MIM and NIM.

3.5. Selectivity

Molecular recognition selectivity is an important parameter in evaluating imprinted material. In order to better study the selectivity of the CIM, IBU, ASP, 2-PPA, 3-PPA and 4-AAP were chosen as competitive adsorption molecules for comparison due to their similar molecular size and active coordination site to NAP (Fig. 4A), among these analogs IBU, ASP and 4-AAP may coexist with NAP in human urine sample. A comparison of the fluorescence response caused by absorption of NAP, IBU, ASP, 2-PPA, 3-PPA and 4-AAP was performed on the CIM, MIM and NIM, and the results are shown in Fig. 4B. It could be seen that CIM revealed higher selective recognition to NAP compared with MIM and NIM. The fluorescence response of analogs was very weak, because the analogs were less luminescent compound. Moreover, the fluorescence response of CIM to 25.0 μmol/L mixed solutions of NAP and each of its analogs was investigated to find out whether CIM could selectively recognize NAP when analogs existed. The results are shown in Fig. 4C and Table S1. In the presence of 3-PPA, the fluorescence intensity of CIM, MIM and NIM decreased about 10%, which may be caused by the strong coordination capability of 3-PPA compared with other analogs. This result illustrated that the accessibility of analytes to the complex imprinted cavities was not only based on molecular size but also coordination capability. Since 3-PPA will not coexist with NAP in human urine sample, it will not influence the detection of real sample. Other analogs had very little effect on fluorescence intensity of CIM. Therefore, CIM can be used for highly selective analysis of NAP in the presence of other interferents. On the other hand, the change of fluorescence intensity of CIM was less than both MIM and NIM, indicating that the immunity from interference of CIM was stronger than MIM and NIM.

3.6. Application

The calibration curve for analysis of naproxen using the proposed CIM showed a linear relationship over NAP-Cu(II) concentration in the range of 0.50–20 mg/L, and the linear regression equation was $Y=11.23+29.31X$ ($R^2=0.9942$) with a detection limit of 0.11 mg/L ($S/N=3$) (Fig. 5). The precision of the method was tested with six replicate analyses of a NAP-Cu(II) standard solution at a concentration of 1.0 mg/L and was 5.7% (within batch) and 9.8% (batch to batch).

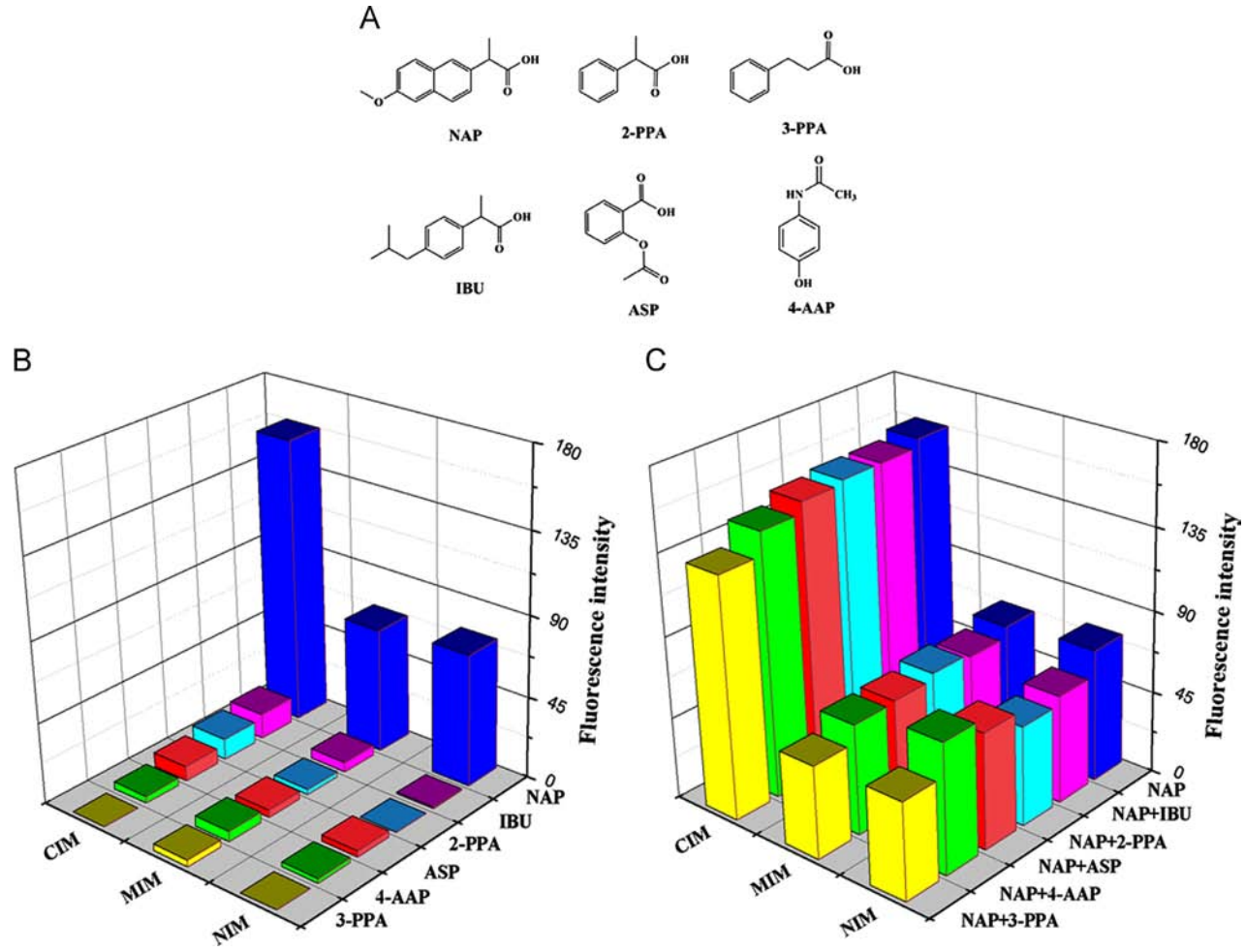

Fig. 4. (A) Molecular structures of NAP and its analogs. Fluorescence intensity of CIM, MIM and NIM to 25.0 μmol/L NAP-Cu(II) and its analogs-Cu(II) (B) and 25.0 μmol/L mixed solutions of NAP and each of its analogs (C).

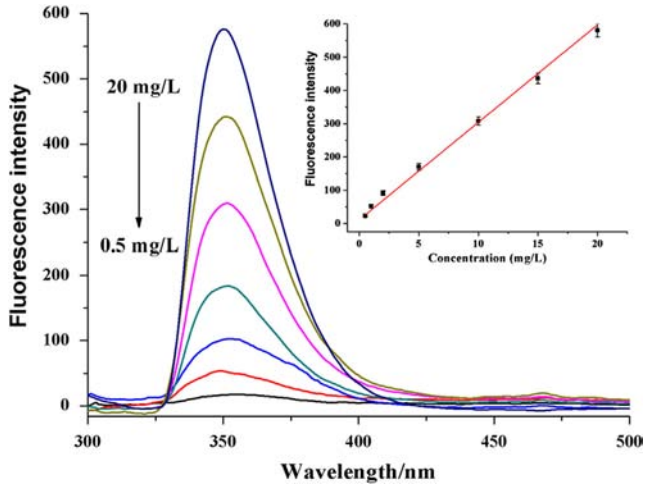

Fig. 5. Fluorescence response calibration curve of CIM over NAP-Cu(II) concentration in the range of 0.50–20.0 mg/L.

To investigate the feasibility of the CIM for real sample application, a solid surface fluorescence method for determination of NAP in pharmaceuticals was established. The results were further compared to high performance liquid chromatography (HPLC) method. Fig. 6 shows the fluorescence emission spectra of determinations of NAP in pharmaceuticals samples by employing CIM, MIM and NIM. The results revealed that CIM had the highest fluorescence response because CIM could both enhance the accessibility of naproxen to the imprinted cavities and improve the selectivity in water medium due to its metal coordination interaction. The recoveries were 85.0% and 89.1% and the RSDs were 9.2% and 12.0% (Table 1). Furthermore, the results obtained by the proposed method and the HPLC-UV method were analyzed by the paired *t*-test. It was shown that there was no statistically significant difference between the results obtained by the proposed method and the traditional HPLC method ($t_1=1.44$, $P=0.95$; $t_2=1.55$, $P=0.95$).

3.7. Comparison of the analytical methods

Several methods for the determination of naproxen have been published. Each method has its advantages and limitations in terms of specificity, sensitivity and interference of matrix compound. The proposed method was then compared with some of the previous reports and the results were illustrated in Table 2. As outlined in Table 2, the proposed method combined the recognition ability and predetermined selectivity of CIP with the high sensitivity of fluorescence analysis. Moreover, the CIM integrated sampling, extraction, concentration, and detection into one-step, which could make the procedure of sample analysis easier and more efficient. Therefore, the novel CIM has great promise for selective and sensitive analysis of naproxen in aqueous samples.

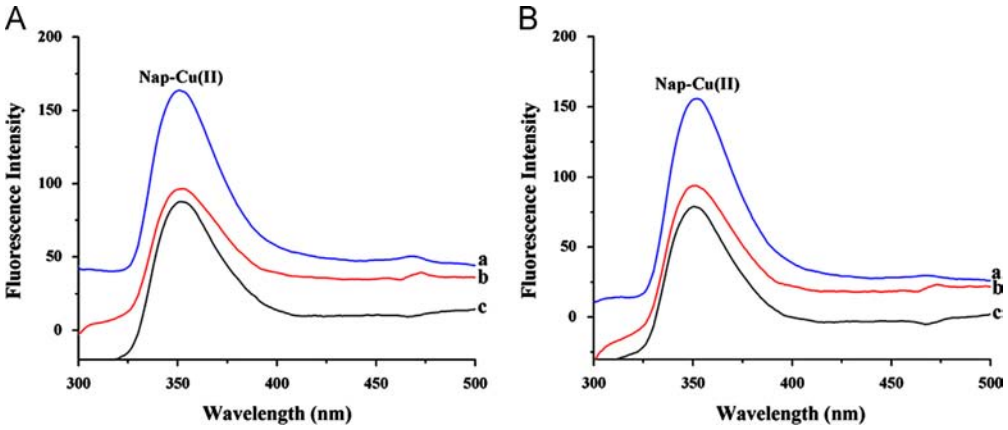

Fig. 6. Fluorescence emission spectra of the detection of NAP in real samples by (a) CIM, (b) MIM, and (c) NIM. (A) Pharmaceutical 1 and (B) pharmaceutical 2.

Table 1
Comparison of naproxen values in pharmaceuticals samples obtained by the proposed method and the HPLC-UV method ($n=3$).

Sample	Solid surface fluorescence method				HPLC-UV measured values	Labeled values
	Measured values	Spiked	Recovery (%)	RSD (%)		
Pharmaceutical 1 (g/g)	0.49 ± 0.03	0.60	85.0	9.2	0.52 ± 0.02	0.60
Pharmaceutical 2 (g/g)	0.45 ± 0.04	0.55	89.1	12.0	0.49 ± 0.02	0.55

Table 2
Comparison of the proposed method with existing reports.

Method	Matrix	Linearity range (mg/L)	Sample preparation technology	Approx. time consuming (min)	Approx. organic solvent consuming (mL per single sample)	Selectivity	Ref.
The proposed method	Pharmaceuticals	0.50–20	CIM	30	0	Yes	Current paper
HPLC-UV	Urine	0.2–20	SPME	60	10	No	[28]
HPLC-UV	Urine	0.009–0.11	MISPE	60	15	Yes	[37]
HPLC-DAD	Suppositories	160–240	FMAE	40	30	No	[38]
LC–MS	Bovine serum	0.05–25.0	Centrifugation	35	10	No	[39]
UV	Plasma	5.0–100.0	LLE	60	20	No	[40]
FL	Serum	0–14	LLE	40	10	No	[41]

4. Conclusions

In this work, a simple, sensitive and selective solid surface fluorescence method based on CIM was proposed for the determination of naproxen in pharmaceuticals samples. Compared with traditional MIP, the proposed CIP could enhance the accessibility of naproxen to the imprinted cavities and improve the selectivity of MIP in water medium due to its metal coordination interaction. The fluorescence performance of CIM was better than that of the controlled materials, which indicated CIM could selectively recognize naproxen in water medium. Finally, the proposed method was applied to the analysis of naproxen in pharmaceuticals with satisfactory results. The main advantages of CIM included simplicity in designing, low-cost, high specificity, and utility. Moreover, CIP is a promising material to mimic biological recognition process. The metal ion mediated recognition can be further expected to be used to fabricate various CIP-based membrane for advanced applications.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.talanta.2013.07.022.

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