

A Molecularly Imprinted Fluorescence Sensor Based on Upconversion-Nanoparticle-Grafted Covalent Organic Frameworks for Specific Detection of Methimazole

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1. Validation by HPLC

For HPLC analysis, the pretreatment process and sample analysis procedures of pork and milk were executed in conformity to the Chinese National Standard (GB/T 22990–2008) [1].

Table S1. Optimization of additional amount of COFs.

Polymer	Dosage(mg)	F ₀ /F(MIP)	F ₀ /F(NIP)	IF
MIP ₁	1	1.331	1.137	2.416
MIP ₂	1.5	1.164	1.062	2.645
MIP ₃	2	1.181	1.123	1.472
MIP ₄	2.5	1.193	1.115	1.678

Table S2. Optimization of the molar ratio of MMZ to AM to MAA to EGDMA.

Polymer	Molar ratio	F ₀ /F(MIP)	F ₀ /F(NIP)	IF
MIP ₁	1:4:4:6	1.185	1.101	1.832
MIP ₂	1:4:4:8	1.225	1.091	2.473
MIP ₃	1:4:4:10	1.104	1.053	1.962

Table S3. Optimization of the matrix of tests.

Polymer	Matrix	$F_0/F(\text{MIP})$	$F_0/F(\text{NIP})$	IF
MIP ₁	Methanol	1.297	1.132	2.25
MIP ₂	Ethanol	1.217	1.125	1.736
MIP ₃	Acetonitrile	1.232	1.123	1.886

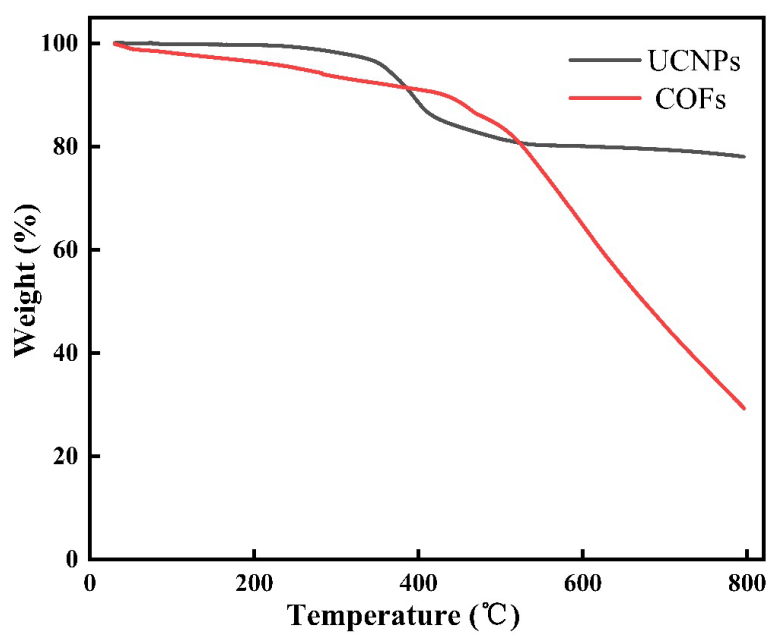


Figure S1. TGA curves of UCNPs, COFs.

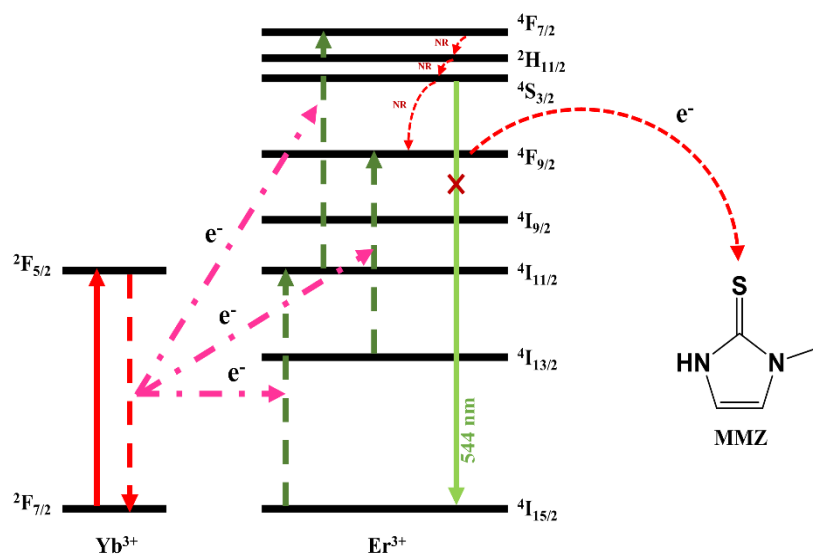


Figure S2. The energy transfer principle.

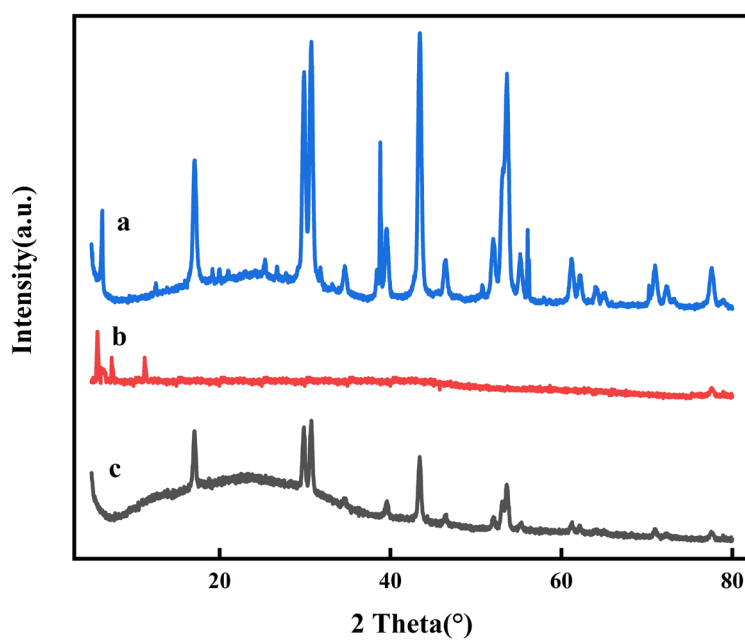


Figure S3. XRD curves of UCNPs (a), COFs (b), UCNPs-grafted COFs@MIPs (c).

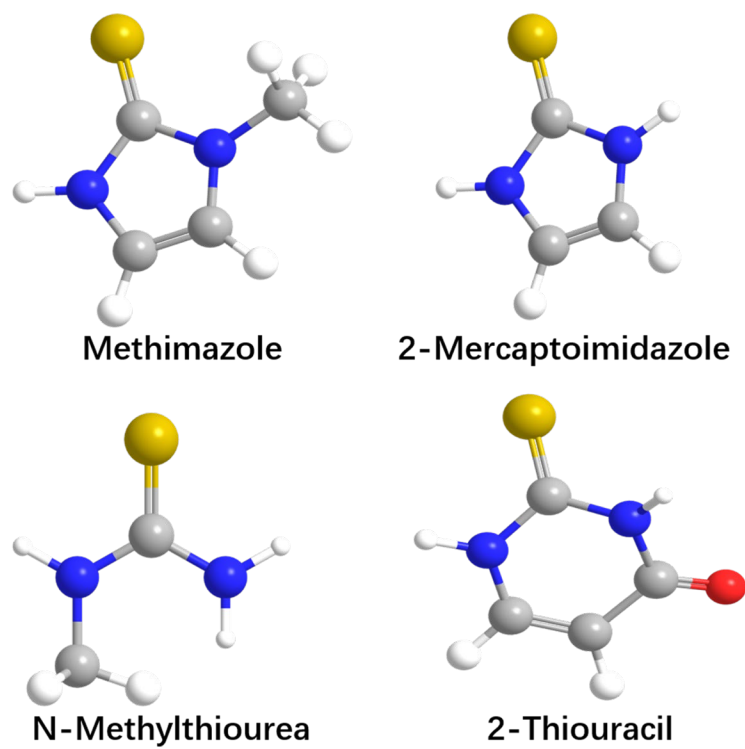


Figure S4. The chemical structures of methimazole (MMZ), 2-mercaptoimidazole (MZY), N-methylthiourea (Metu) and 2-thiouracil (2-TU).

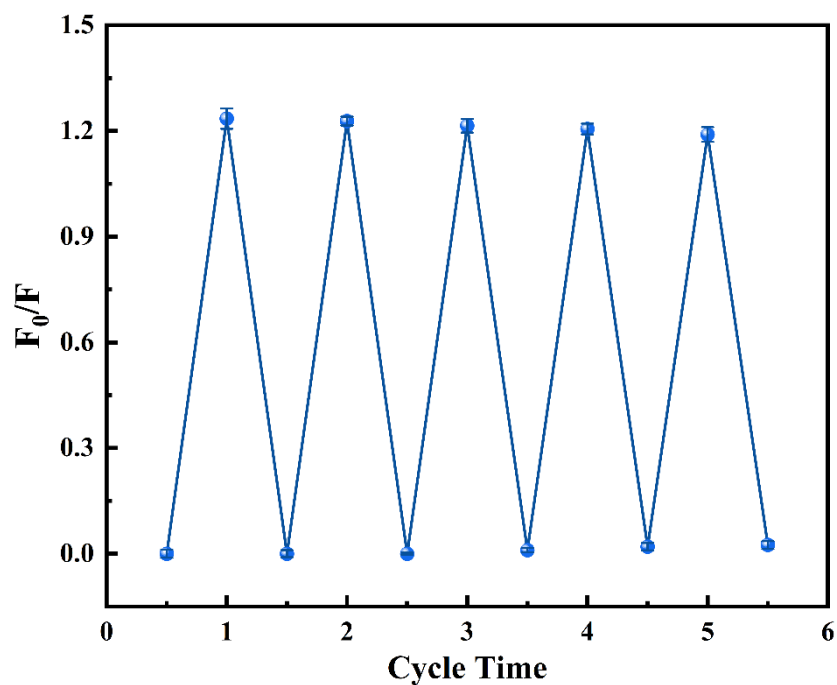


Figure S5. Reusability of UCNPs-grafted COFs@MIPs for the determination of MMZ.

1 **Table S4.** The comparison of the analytical methods with other reported methods.

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Analytical method	Linear range ($\mu\text{g L}^{-1}$)	LOD ($\mu\text{g L}^{-1}$)	Recovery (%)	Reference
MIP QCM sensor	$0.5\text{-}20\times 10^3$	109	84.4-91.7	[2]
ECL	$0.5\text{-}6\times 10^3$	48	87.8-101.8	[3]
HPLC	$0.23\text{-}1.14\times 10^3$	110	97-102	[4]
N-CQDs	$0.34\text{-}6.85\times 10^3$	9.13×10^{-2}	99-102	[5]
Aptamer	1.71-857.14	2.13	81.54-100.96	[6]
ELISA	2.5-40	1.56-2.72	81.25-103.19	[7]
UCNPs grafted COFs@MIPs	$0.05\text{-}3\times 10^3$	3	88.24-91.54	This work

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