

Recent Progress in Silver-Ion-Selective Fluorescence Chemical Sensors

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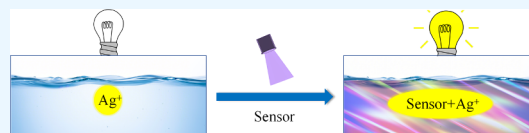
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ABSTRACT: Heavy metal ions easily accumulate in the human body through the food chain, and their binding with sulfur-containing proteins, enzymes, and various metabolites can affect the liver, kidneys, and central nervous system. The development of sensors for detecting Ag and other metal ions provides an economically efficient means of protecting the environment and human health. This review focused on the latest developments in fluorescent and colorimetric organic small-molecule sensors for identifying Ag. We analyzed their recognition principles and mechanisms; discussed their sensitivity, specificity, and stability; and conducted in-depth research on their application performance in practical environments.



1. INTRODUCTION

As a metal ion with unique chemical and physical properties, silver ion plays an important role and is widely used in many fields. Therefore, its detection and research hold substantial significance. In the natural environment, silver ions originate from a wide variety of sources. The mining and smelting process of silver ores can lead to the release of silver ions into the environment. For instance, silver is frequently found in association with some lead–zinc deposits. During the extraction, beneficiation and smelting stages of these ores, silver ions are discharged into the surrounding water and soil along with wastewater, slag and other waste materials.

Moreover, some industrial production processes, such as the silver plating operations employed in the electronics sector and the utilization of silver salts in the photography industry, also serve as significant anthropogenic sources of silver ions. In daily life, silver containing antibacterial products, such as antibacterial fabrics, antibacterial coatings and antibacterial medical devices, will progressively release silver ions as the products are used and age.¹

Ag has been widely used in various aspects of human life, fully leveraging its physical, chemical, and biological advantages in fields such as photography, agriculture, electronic imaging, medical devices, and the pharmaceutical industry.² However, silver may enter soil, water, air, and food with the widespread application of nanomaterials and accumulate in the human body through the food chain, leading to developmental delays, skin damage, liver and kidney toxicity, and central nervous system damage; this is because Ag can coordinate with thiol, imidazole, amino, and carboxyl groups, leading to protein and enzyme inactivation and causing many serious symptoms and diseases.³ Therefore, developing an economically efficient, fast and sensitive silver ion recognition and detection technology has become increasingly important.

Voltammetry, ICP-MS (Inductively Coupled Plasma Mass Spectrometry), AAS (Atomic Absorption Spectroscopy), ion-selective electrodes (ISEs), and electrochemical methods are expensive and time-consuming.^{4–8} Adsorption is an emerging and efficient method that features high adsorption capacity, good selectivity and reusability. It can be used to remove a wide variety of toxic substances in wastewater, such as total nitrogen, total phosphorus and heavy metal ions. Metal ion sensors and fluorescent hybrid materials exhibit high selectivity and sensitivity, as well as good adsorption capacity, which provide new ideas and methods for wastewater treatment.^{9–11}

With the development of materials science, an increasing number of fluorescent colorimetric probes have been developed.^{12–18} Silver ion probes are molecules or materials engineered to specifically recognize silver ions. Its fundamental principle is based on the specific interaction between the probe molecule and silver ions, which can lead to changes in the optical, electrochemical or other physicochemical properties of the probe molecule, thus realizing the detection of silver ions. Among the various types of silver ion probes, fluorescent probes have been the most extensively investigated. It is usually composed of fluorophore and recognition groups. The recognition group is capable of selectively binding to silver ions. When silver ions bind to the recognition group, the fluorescence properties of the fluorophore change, for example, the fluorescence intensity may increase or decrease, or there

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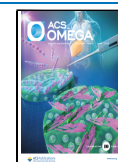


Table 1. Different Analytical Parameters for the Analysis of Ag(I)^a

sensor	binding constants	fluorescence color	solvent media	LOD	sensing mechanism	sensor type	pH range	application
TOV	1:2	colorless to green (435 nm)	ACN/H ₂ O = 9:1	6.138 nM	PET, CHEF	turn-on	neutral pH	water
CHE	1:1	615 nm	EtOH/BR buffer solution = 4:6	250 nM	ICT	turn-off	4–8	water, milk, and pears
DTPAS		green (495 nm)	THF–H ₂ O	860 nM	AIE, ACQ	turn-on	2–10	water
L1	1:1	yellow-green to disappear (546 nm)	DMSO	1.88 nM	PET	turn-off	7.0	drinking water, tap water, and deionized water
KH1	1:1	(350 nm)	DMSO/H ₂ O = 1:4	200 nM	PET	turn-on	7	tap water and wastewater
HL1	1:1	(422 nm)	ACN	7.92 nM	CHEF, PET	turn-on		
DA-NR		no fluorescence to blue fluorescence (481 nm)	Tris–HNO ₃	157.1 nM	catalytic reaction	turn-on	8.0	real water tap water, local lake water, and river water
AQP	1:1	(410 nm)	DMSO/H ₂ O buffer = 1:1	17 nM	ICT	turn-on	7.4	water samples, the soil, rice flour and dried banana flower
AQM	1:1	(405 nm)	DMSO/H ₂ O buffer = 1:1	79 nM	PET, ICT	turn-on	7.4	water samples, soil, rice flour, and dried banana flower
SN-Cl	1:1	light orange fluorescence (385 nm)	DMF/Tris-HCl buffer = 1:1	12 nM	AIE, ESIPT	turn-on	2–7	seawater, lake water, river water, and red wine
TPE-BTA	1:1	blue to dark blue (495 nm)	DMSO/H ₂ O = 1:9	496 nM	AIE	turn-off	5–10	tap water
KL1		orange yellow to blue/bright blue (600 nm)	EtOH/H ₂ O = 1:1	429 nM	catalytic reaction	turn-off		real water
SB	2:1	523 nm	MeOH/H ₂ O = 98:2	36 nM	PET	turn-on	7–11	river water, tap water, pond water, and soil samples
N-FP		661 nm	EtOH/H ₂ O = 9:1	110 nM	ICT	turn-on	1–10	
TMB			H ₂ O	10000 nM	redox			cellulose membrane
ABC	1:1	bright blue to dark blue (465 nm)	DMSO/H ₂ O(HEPES) = 1:1	1.4 nM	PET	turn-off	7.4	borewell, drinking water and real soil sample
TPEPVA	2:1	bright blue to dark blue (426 nm)	ACN/H ₂ O = 1:9	533 nM	AIE	turn-off		test paper
L2	1:1	deep blue to orange (527 nm)	MeOH/HEPES buffer = 7:3	40 nM	PET, CHEF	turn-on	6–9	drinking water, tap water, and river water
AgP	1:1	green to colorless (532 nm)	DMSO/PBS = 1:99		PET	turn-off	4–10	city water, drinking water, lake water, and wild-type (WT) <i>Arabidopsis thaliana</i>
TC-Mal-Et ₃ N		orange yellow to red (590 nm)	BR/ACN = 6:4	60.9 nM	ICT	turn-on	8.5	tap water and Chongde Lake
COH4	1:2	(375 nm, 360 nm)	HEPES buffer/MeOH = 1:1	100 nM	PET, ICT	turn-on	6–8	<i>E. coli</i> cell imaging
DBP	1:1	bright blue to dark blue (425 nm)	EtOH/PBS = 3:7	19 nM	PET	turn-off	5–10	food (shrimp meat, cheese, Barcelona fish fillet), water sample
TQB	1:1	green to no fluorescence (516 nm)	MeOH/buffer = 7:3	650 nM	PET	turn-off	7–11	test strips and actual water sample
I	2:1	blue (499 nm)	MeCN/H ₂ O = 3:7	413 nM	AIE	turn-on	7–10	visualizing <i>Rhizoctonia oryzae</i> mycelia cells

^aAll data except for annotations are silver ions. PET = photoinduced electron transfer. CHEF = chelation-enhanced fluorescence effect. ICT = intramolecular charge transfer. AIE = aggregation-induced luminescence. ACQ = aggregation caused quenching.

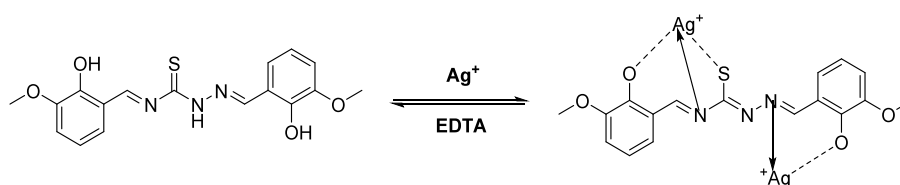


Figure 1. Organic sensor TOV for the detection of Ag(I) anions.

may be a shift in the fluorescence wavelength. Colorimetric probes, in contrast, detect silver ions by causing a color change in the solution after the probe molecule binds to silver ions. This color change can be directly observed with the naked eye or quantitatively measured using instruments such as a

spectrophotometer. Colorimetric probes have the advantages of simple operation and are suitable for rapid detection on site.

In comparison, these methods are simple, fast, sensitive, and efficient. These probes contain N, S, or O atoms in their molecular structure, which can coordinate and form complexes

with silver ions or undergo chemical changes such as oxidation–reduction, resulting in significant color changes or changes in fluorescence intensity. This review focused on the latest developments in fluorescent and colorimetric organic small-molecule sensors for identifying Ag, analyzed their recognition principles and mechanisms, discussed their sensitivity, specificity, and stability, and explored their potential application performance in practical environments. The metal complex value of binding constant, limit of detection (LOD), Sensing mechanism, etc., are shown in Table 1.

2. LIGAND-TYPE SILVER ION PROBE

2.1. Thiourea Derivatives. Hiremath et al. designed and synthesized a probe **TOV** based on the structure of the amino thiourea Schiff base, as shown in Figure 1.¹⁹ The probe **TOV** contained two coordination sites: imine (HC=N) and hydroxyl (–OH). These two sites could form coordination bonds with Ag(I) ions. On the other hand, the isomerization of imine groups and the electron transfer of imine groups to excited benzaldehyde derivatives could lead to fluorescence quenching. After coordinating with TOV molecules at a 2:1 ratio through coordination sites, Ag(I) ions inhibited two fluorescence quenching processes in **TOV**, thereby restoring the fluorescence of benzaldehyde derivatives (green). The probe **TOV** was excited at 315 nm in a solvent of MeCN/H₂O = 9:1 and exhibited emission at 435 nm. After the addition of silver ions, the fluorescence at 435 nm was significantly enhanced and the maximum emission intensity was reached within 50 s; its detection limit for silver ions was 6.138 nM.

In addition, in the UV–vis absorption spectrum, the addition of Ag(I) caused a blue shift in the absorption band of **TOV** at 315 nm, while a new absorption band appeared at 397 nm, and the absorption intensity increased, resulting in a change in the color of the solution from colorless to yellow. By using absorption spectroscopy and fluorescence spectroscopy techniques to study the sensing behavior of **TOV** toward different metal ions, it was found that only Ag(I) could cause significant changes. Therefore, the **TOV** probe could be used for the specific detection of silver ions. Owing to the stable and effective functioning of **TOV** at pH 6–8, this probe could be used for the detection of actual samples and for cell imaging. It also had potential applications in the field of biology.

A novel colorimetric fluorescent probe **L1** capable of specifically detecting Ag(I) and Hg(II) was synthesized using 1,8-naphthalimide as the fluorescent material and hydrazine thioformamide as the reaction site. Ag(I) and Hg(II) combined with the probe to form a 1:1 complex, as shown in Figure 2.²⁰ Ag(I) and Hg(II) had good Lewis acidity and could better accept electrons from the probe, effectively

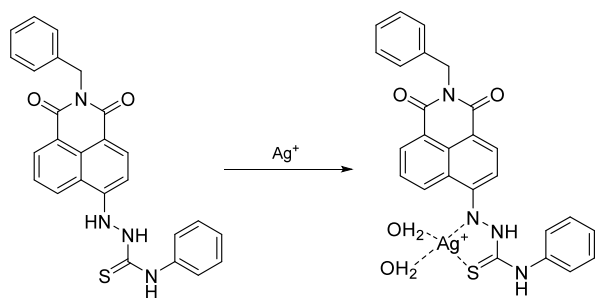


Figure 2. Organic sensor **L1** for the detection of Ag(I) anions.

quenching the probe (PET). Specifically, Hg(II) reduced the fluorescence quantum yield of the probe from 0.035 to 0.00067, while Ag(I) reduced the fluorescence quantum yield to 0.00085, resulting in a decrease in fluorescence intensity to almost 1/50 of its original level. On the other hand, when the probe bound to Hg(II), it caused the absorption peak of the probe to shift red, with a new peak appearing at 494 nm (brown). When it bound to Ag(I), the absorption peak also shift red, with a new peak appearing at 477 nm (brown orange). And the concentration of Ag(I) and Hg(II) showed a good linear relationship with the changes in absorption and fluorescence spectra, which could accurately detect low concentrations of Hg(II) and Ag(I) (LOD: 3.84 nM, 1.88 nM).

Furthermore, by distinguishing Ag(I) and Hg(II) based on the different affinities of ethylenediaminetetraacetic acid disodium salt with Ag(I) and Hg(II), EDTA could capture Hg(II) from the complex between probe **L1** and Hg(II), causing probe **L1** to dissociate, suppressing the PET effect and restoring the intrinsic fluorescence of probe **L1**, thereby achieving reversible detection of Hg(II) and specific detection of silver ions. The test strips could be used for the portable determination of real water samples.

The author synthesized an “open” organic small-molecule fluorescent probe, **KH1**, based on a Schiff base and thiosemicarbazide for the specific detection of Ag(I), as shown in Figure 3.²¹ Due to the PET effect, the fluorescence of

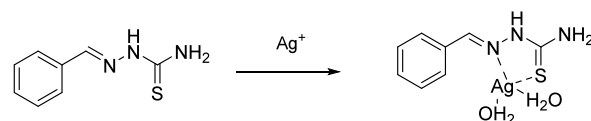


Figure 3. Organic sensor **KH1** for the detection of Ag(I) anions.

the probe itself was weak. When the probe formed a 1:1 complex with Ag(I), the electron cloud density of lone pair electrons of nitrogen atoms on the Schiff base decreased, and the fluorescence quenching caused by the PET effect was suppressed, resulting in an increase in the fluorescence intensity of **KH1** at 350 nm. The fluorescence intensity of the probe stabilized after 30 min of reaction with Ag⁺ at room temperature with a limit of detection (LOD) of 200 nM. This probe was applied for the detection of Ag(I) in actual water samples with good accuracy and precision. In the range of pH 2–7, the fluorescence intensity of the probe increases with the increase of pH value, but when the pH value was higher than 7, the fluorescence intensity began to decrease, which might affect the wide application of the probe.

Naphthalimide was used as the fluorescent material, and hydrazine thioformamide was used as the reaction site. Satyajit Mahata et al. synthesized the fluorescent probe **L2**, as shown in Figure 4.²² In the free state, there was a PET process between the amino group and the carbonyl group of the probe, leading to fluorescence quenching. After the probe formed a complex with Ag(I) and Hg(II), the lone pair electrons of the NH group were unable to participate in the PET process, and the chelation enhanced fluorescence (CHEF) process was activated, resulting in an increase in fluorescence intensity and achieving “off-on” fluorescence detection of Ag(I) and Hg(II). In addition, the decrease of the HOMO–LUMO energy gap led to a red shift in the absorption spectrum. The complex exhibited new absorption peaks at 432 and 528 nm,

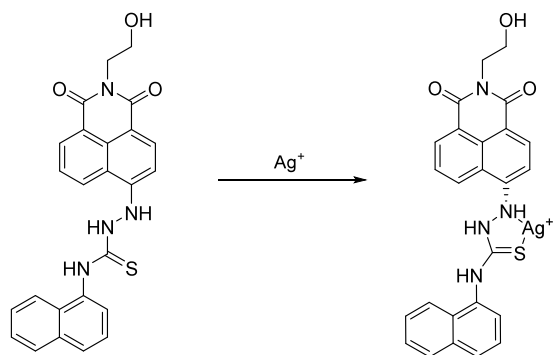


Figure 4. Organic sensor **L2** for the detection of Ag(I) anions.

the color of the solution changed to brown, and the emission peak moved to 527 nm and produced orange fluorescence. This probe had been successfully applied in cell imaging, indicating the potential of the **L2** probe. However, its cytotoxicity drawback needed further improvement.

As shown in Figure 5, Jagadhane et al. designed and synthesized **TPEPVA**, which was based on the AIE effect, as a fluorescent probe for detecting Ag(I) and Hg(II).²³ The probe had a propeller like structure, and in dilute solution, the aromatic ring could rotate freely, leading to non radiative decay and weak fluorescence. With the addition of water, the probe aggregated, limiting intramolecular rotational vibrations and increasing fluorescence emission. When the NH group on the probe formed a stable complex with Ag(I) and Hg(II), the aggregation state of the probe was disrupted, which reduced the fluorescence intensity at 466 nm and changed the fluorescence from bright blue to dark blue. This fluorescence response had been successfully used for detecting Ag(I) and Hg(II) and had great potential in practical applications because of its ability to prepare test strips for onsite detection of Hg(II) and Ag(I).

Among the above thiourea probes, the probes could coordinate with silver ion through the N atom and S atom of thiourea. In contrast, The **TOV** probe also formed stable coordination bonds with Ag(I) ions through the $-OH$ and $-NH$ groups of 2-hydroxy-3-methoxybenzaldehyde, which could simultaneously inhibit the photoinduced electron transfer (PET) and $C=N$ isomerization process, resulting in significantly enhanced fluorescence, improved specificity and sensitivity of the probe, and it showed higher sensitivity and a lower detection limit.

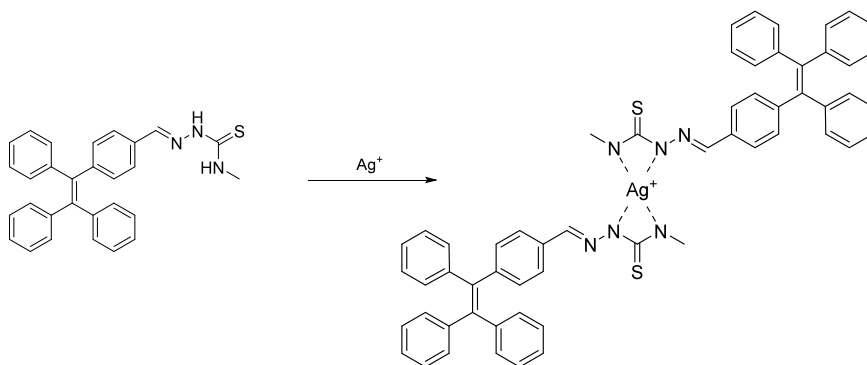


Figure 5. Organic sensor **TPEPVA** for the detection of Ag(I) anions.

2.2. Schiff Base Derivatives. Zhou et al. designed and synthesized an enhanced fluorescent probe, **HL1**, for the specific detection of Ag(I) and Hg(II), as shown in Figure 6.²⁴

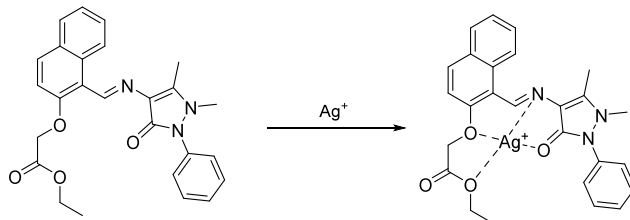


Figure 6. Organic sensor **HL1** for the detection of Ag(I) anions.

After the probe formed a 1:1 complex with Ag(I) and Hg(II), N and O atoms could form coordination bonds with ions, increasing the rigidity of the molecule and limiting its rotation and vibration within the molecule. And the combined molecular structure was more stable, resulting in enhanced fluorescence (PET). The binding with Hg(II) also caused a blue shift in the emission peak (385 nm), and the solution changed from colorless to yellow. The addition of Hg(II) increased the fluorescence intensity of the probe from 220 to 5937, and the addition of Ag(I) also significantly increased the fluorescence intensity. In addition, the detection limits of the probe for Ag(I) and Hg(II) reached 7.92 nM and 5.16 nM, respectively, indicating high sensitivity. The color change and fluorescence enhancement effect of the probe enabled visual detection of Ag(I) and Hg(II). The experiment mainly studied the recognition performance of the probe for metal ions in acetonitrile solution. In practical applications, the sample might exist in various solvent environments, which might limit its wide application.

Zhao et al. constructed a three-response “on” fluorescent probe **SN-CI** based on benzothiazole, salicylaldehyde, and Schiff base frameworks, which exhibited aggregation-induced luminescence (AIE) and intramolecular proton transfer (ESIPT) effects.²⁵ The nitrogen heteroatoms and hydroxyl groups were rich in electrons and had good coordination ability with metal ions. As shown in Figure 7, the binding ratio of the probe to Pb(II) and Ag(I) was 1:1, and the binding ratio to Fe(III) was 2:1. After coordination, the energy gap of the probe decreased, which was beneficial for fluorescence enhancement and also indicated the formation of a more stable structure. The electron distribution of HOMO and LUMO changed before and after coordination, which

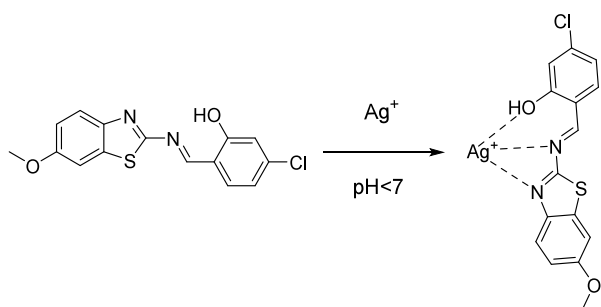


Figure 7. Organic sensor SN-Cl for the detection of Ag(I) anions.

promoted intramolecular electron transfer. On the other hand, after coordination, the molecular structure became more planar and rigid, which was conducive to molecular aggregation and enhanced fluorescence. The probe exhibited fluorescence enhancement at 385 nm with increasing concentrations of Pb(II), Ag(I), or Fe(III) with detection limits of 59 nM, 12 nM, and 8.92 μ M, respectively. This probe had good detection performance for three ions in real water samples and could serve as a good imaging agent for Fe(III) in HeLa cells, providing promising prospects for cell imaging research.

The SB probe synthesized with a Schiff base as the center had relatively weak emission at 523 nm due to the PET effect (electron transfer from the N atom of the imine group to the naphthalene ring portion).²⁶ When the oxygen on the phenolic hydroxyl group and the nitrogen on the imine group of the probe coordinated with Ag(I) and Cu(II), the PET effect was inhibited, and resulted in enhanced fluorescence emission, as shown in Figure 8. After the addition of Ag(I), the color

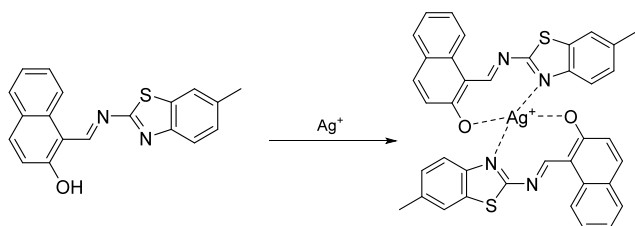


Figure 8. Organic sensor SB for the detection of Ag(I) anions.

changed from yellow to carrot, and after the addition of Cu(II), the color changed from yellow to orange yellow. This red shift was caused by the inhibition of electron transfer from

the nitrogen atom of the probe to the naphthalene ring (ICT) after coordination, as well as the facilitation of charge transfer from the probe to the metal, giving rise to a red shift in the absorption spectrum. The fluorescence intensity of the probe at 523 nm showed a significant enhancement with increasing concentrations of Ag(I) and Cu(II). The detection limits were 25.1 nM for Cu(II) and 36 nM for Ag(I). This fluorescence response had good specificity for Ag(I) and Cu(II) and could be used for the detection of Ag(I) and Cu(II) in water and soil samples. Under acidic conditions, the probe functional groups were protonated, which may prevent complexation with metal ions.

Fluorescent probe 1 was designed and synthesized based on coordination reaction using 1,8-naphthalimide as the fluorescent group and utilizing the sulfur and nitrogen affinity of Ag(I) and Hg(II).²⁷ The fluorescence intensity of the probe itself would increase with the increase of water ratio, and the probe aggregates to form fluorescent groups, giving rise to the AIE phenomenon. However, in the CH₃CN/H₂O (3/7) mixed solution without the addition of Ag(I) and Hg(II), it existed mainly as non fluorescent aggregates, and the fluorescence quenching occurred through a twisted intramolecular charge transfer (TICT) process. Under this condition, when Ag(I) and Hg(II) were added, the metal ions formed a 1:2 complex with the probe (as shown in Figure 9), which led to changes in the morphology and size of the aggregates, limiting intramolecular torsion and vibration, promoting the AIE process, inhibiting the TICT process, and emitting strong blue and green fluorescence at 499 nm (Ag(I)) and 521 nm (Hg(II)). The detection limits for Ag(I) and Hg(II) were 413 nM and 750 nM, respectively, thus, it could be used for the specific detection of Ag(I) and Hg(II). This probe had been successfully used for in vitro imaging of rice *Fusarium* hyphae cells, indicating its potential in examining cell behavior and interactions with various cells.

The structure of Schiff base probes facilitated the formation of stable complexes with metal ions. When amino or hydroxyl groups were introduced, the stability and the fluorescence intensity could be enhanced, which was conducive to improving the sensitivity. For instance, the 1,8 - naphthalimide in probe 1, serving as the fluorescent core, provided the basic fluorescence performance. After forming a V-shaped structure with benzothiazole groups, it promoted the probe to form AIE active aggregates upon binding with ions. This effectively restricted intramolecular torsion and vibration, significantly

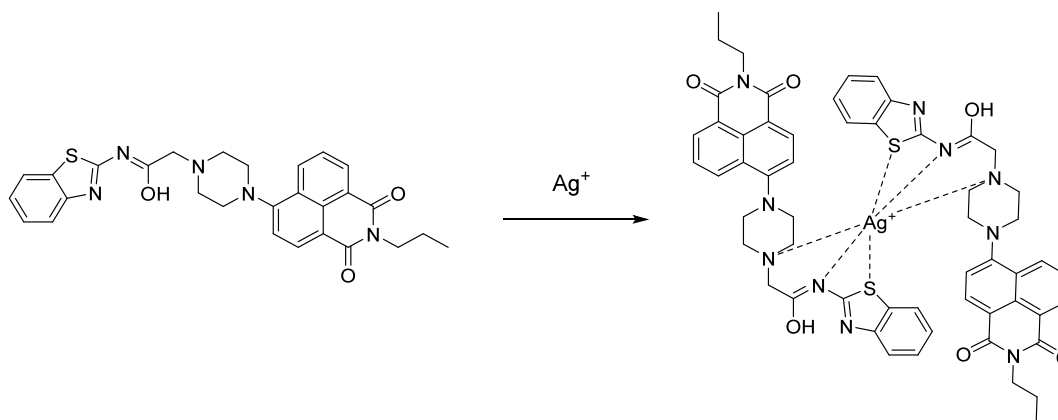


Figure 9. Organic sensor 1 for the detection of Ag(I) anions.

enhancing the fluorescence signal, thereby enabling the effective detection of trace ions.

2.3. Hydrazine Derivatives. Utilizing the strong affinity between N, O, and metal ions, Liu et al. developed a novel ratio of the fluorescent probe **CHE**, which was mainly composed of coumarin and 2-hydrazinobenzothiazole.²⁸ The imine and benzothiazole groups on the probe served as coordination sites, forming stable complexes with Hg(II) and Ag(I) ions in a 1:1 ratio, as shown in Figure 10. DFT

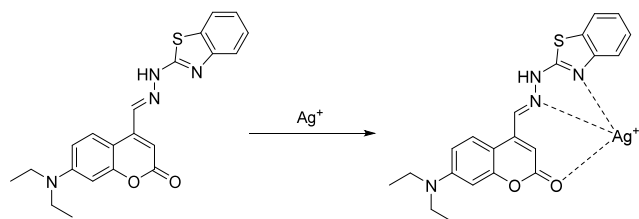


Figure 10. Organic sensor **CHE** for the detection of Ag(I) anions.

calculations showed that the HOMO orbitals of the probe were mainly concentrated in the coumarin portion, while the LUMO orbitals were distributed throughout the entire molecule. After combining with metal ions, LUMO mainly concentrated on the metal ions and extended to the entire molecule, resulting in a significant change in **CHE** color (turning blue) due to charge transfer. Meanwhile, due to the enhanced spin orbit coupling, the fluorescence emission intensity decreased at 615 nm and remained basically unchanged at 475 nm. This ratio change enabled the sensor to more accurately detect Hg(II) and Ag(I).

Furthermore, specific selection of Hg(II) and Ag(I) could be achieved through the different chelating affinities of EDTA for Hg(II) and Ag(I). This probe had been applied to the detection of real samples such as water, milk, and pears, and had been used for the construction of molecular logic gates. Moreover, owing to the reversible interaction between Hg(II) and EDTA, it had broad application prospects in molecular storage.

Song et al. developed a selective fluorescent probe, **AgP**, based on quinoline with a wide measurement range, as shown in Figure 11.²⁹ The probe exhibited bright green fluorescence (emission wavelength of 532 nm) under DMSO/PBS (1:99, v/v, pH = 7.4) excitation at 390 nm. The HOMO–LUMO energy gap decreased after the probe coordinated with Ag(I), which was due to the charge transfer from the ligand to the metal. The structure of the molecule and the distribution of the electron cloud changed, which caused a new absorption band at 383 nm, and the color of the solution changed from transparent to light orange (visible to the naked eye). At the same time, the fluorescence was significantly weakened and

slightly blue-shifted, and the fluorescence color changes from bright yellow green to completely disappeared.

After the addition of Ag(I), the fluorescence response was quenched, and the detection of Ag(I) was stable in the pH range of 4–10. It could be directly used for colorimetric detection of Ag(I) in water samples, which was simple and convenient, and could observe color changes within a few seconds. The recovery rate of the **AgP** detection water sample was between 117.6% and 98.3%, and the relative standard deviation (RSD) was between 0.41% and 2.06%. **AgP** was also suitable for in vivo imaging of Ag(I) in the classic model plant *Arabidopsis thaliana*. The fluorescence of 50 μ M Ag(I) could be completely quenched, providing a new detection tool for studying the distribution of Ag(I) in the environment and living plants. However, when Ag(I) concentrations were high, accurate quantification might be difficult.

Selvaraj Anshmya et al. synthesized a fluorescent probe, **COH4**, with a PET mechanism using coumarin as the parent material.³⁰ The oxygen atoms of coumarin and phenolic hydroxyl groups, as well as the nitrogen atom of the hydrazone group in the probe contained lone pair electrons that could bind to Ag(I) in a 1:2 ratio, as shown in Figure 12. In the free state, the PET process made the probe fluoresce weakly. When Ag(I) was added, it bound with these heteroatoms, inhibiting the PET process while enhancing fluorescence through the ICT mechanism. And the fluorescence emission peaks were blue-shifted (375 to 360 nm) due to the restricted rotation of the carbon–carbon single bond in the molecule. The LOD of the probe was 0.41 μ M and could be reused through EDTA. The probe had been successfully used for imaging Ag(I) in *Escherichia coli* under neutral conditions, indicating the potential of the **COH4** probe in this regard. The probe exhibited optimal detection performance at pH 7.4, but the fluorescence emission was reduced in acidic and alkaline excesses.

Li et al. designed and synthesized a novel fluorescent probe, **TQB**, capable of dual-mode detection of Ag(I).³¹ The probe showed a light yellow color and green fluorescence in a MeOH/H₂O buffer solution (7/3, v/v, pH = 7.23). The probe bound to Ag(I) ions through coordination bonds to form a 1:1 complex, as shown in Figure 13. IR spectroscopy analysis showed that the functional group peaks at –C–N–, –CO–, and –NH– in the probe disappeared, while a new peak appeared at –C=N–. This indicated a change in structure, which in turn caused a change in its optical properties, with the maximum absorption wavelength changing from 500 to 525 nm, and a significant decrease in fluorescence intensity at the maximum emission wavelength of 516 nm (quenching rate of 88.7%). After the coordination reaction with Ag(I), the isomerization of C=N promoted the PET effect, resulting in fluorescence quenching and a pink color under sunlight. It could be used for colorimetric and fluorescence detection of

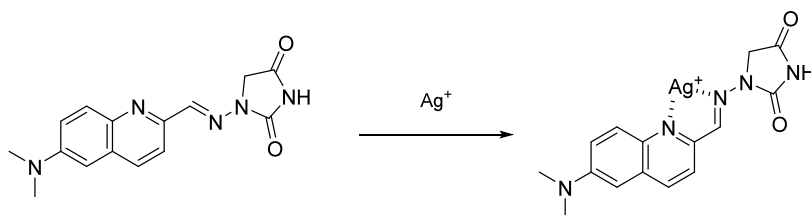


Figure 11. Organic sensor **AgP** for the detection of Ag(I) anions.

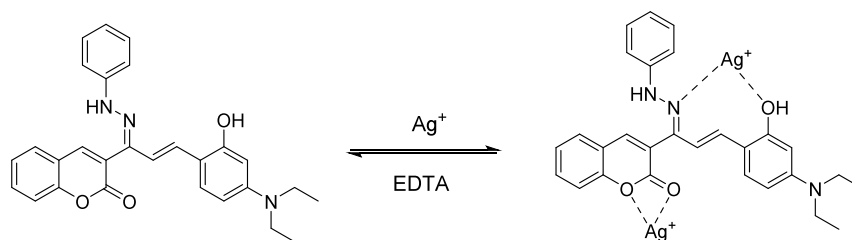


Figure 12. Organic sensor COH4 for the detection of Ag(I) anions.

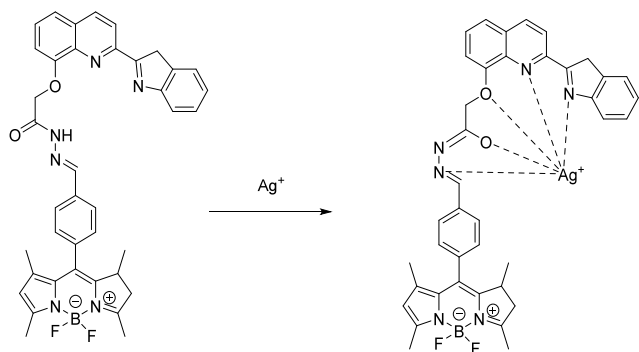


Figure 13. Organic sensor TQB for the detection of Ag(I) anions.

Ag(I) in real water samples, the detection limit was $0.69 \mu\text{M}$ in colorimetric mode and $0.65 \mu\text{M}$ in fluorescence mode. And the test strip prepared with this probe provided the possibility for portable detection of Ag(I).

These probes exhibited strong selectivity for silver ions and formed stable complexes with them through N and O atoms. However, the biocompatibility of the probes posed a limitation to their broader application. It had been discovered that the probe AGP, which had a relatively small molecular weight, demonstrated favorable cytotoxicity and biocompatibility. It could effectively diffuse within living cells, enabling in vivo detection. Moreover, its appropriate excitation and emission wavelengths could also prevent interference from the autofluorescence of biological tissues.

2.4. Amino Derivatives. Suguna et al. designed and synthesized two fluorescent probes, AQP and AQM, as shown in Figure 14.³² In the free state, the lone pair electrons on the nitrogen atoms of the amino quinoline and amino groups in the probe participated in the PET process, resulting in weak fluorescence activity of the probe. When Ag(I) coordinated

with the probe, lone pair electrons on the nitrogen atom were occupied, and the PET process was inhibited. On the other hand, the HOMO and LUMO of the probes were mainly distributed in the quinoline ring and xylene portion. After coordination, the distribution of the electron cloud was changed, which promoted the ICT process and reduced the electron migration energy. This made the complex more polar and chemically reactive, which was conducive to electron transition and fluorescence emission, resulting in a blue shift and increased intensity of the fluorescence emission peak. The synergistic effect of PET inhibition and ICT enhancement significantly changed the fluorescence properties of the probe, thereby achieving the detection of Ag(I). This fluorescent response had good specificity for Ag(I) and high detection sensitivity with LOD values of 17 nM (AQP) and 79 nM (AQM), respectively. It could be used for the detection of actual samples such as water, rice noodles, and banana flowers and for the imaging of Ag(I) in cells. The probe took 5–6 min to saturate with silver ions, which might limit its utility in applications requiring a rapid response.

Bhusanur et al. designed and synthesized a TPE-BTA probe based on the AIE mechanism, as shown in Figure 15.³³ It used

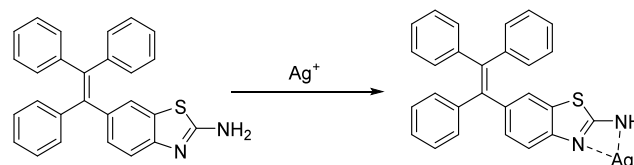


Figure 15. Organic sensor TPE-BTA for the detection of Ag(I) anions.

tetraphenylethylene as the fluorescent group for the detection of Ag(I), Hg(II) and Fe(III). Due to the AIE effect, the fluorescence

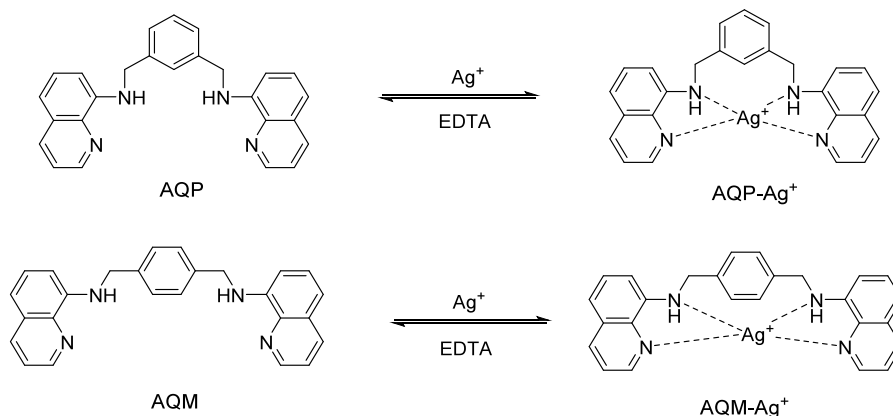


Figure 14. Organic sensors AQP and AQM for the detection of Ag(I) anions.

intensity of the probe in organic solvents was weak, appearing as a deep blue color. As the proportion of water increased, the fluorescence intensity gradually increased, resulting in a bright blue color. Metal ions interacted with nitrogen and amino groups to form 1:1 complexes, causing changes in bond lengths and angles, thus affecting the electronic energy levels of the probes, changing the electronic structure of the probes, and weakening the fluorescence. Among them, Ag(I) mainly affects the LUMO level of the probe, Hg(II) mainly affected the HOMO level of the probe, and Fe(III) simultaneously affected both the HOMO and LUMO levels of the probe. When the probe complexed with Ag(I), Hg(II) and Fe(III), its fluorescence intensity decreased, and the color changed from bright blue to dark blue, thus enabling the detection of Ag(I), Hg(II) and Fe(III). However, for these three ions, the probe did not show significant selectivity.

Suguna et al. synthesized the fluorescent probe **ABC** based on anthracene for the detection of Ag(I), as shown in Figure 16.³⁴ Due to the photoinduced electron transfer effect (PET),

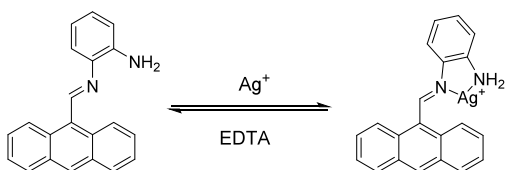


Figure 16. Organic sensor **ABC** for the detection of Ag(I) anions.

the probe itself emitted strong bright blue fluorescence. Selective detection of Ag(I) ions was achieved through quenching of fluorescence intensity when interacting with Ag(I) ions. Ag(I) coordinated with the free amine and imine N atoms on the benzene ring in **ABC**, and electrons were transferred from the benzene ring to the anthracene ring, inhibiting the process of photoinduced electron transfer (PET) and leading to fluorescence quenching. At the same time, the heavy atom effect of Ag(I) increased the probability of non radiative leaps and accelerated the fluorescence quenching of the probe. In addition, after the probe bound to Ag(I) ions, the C=N double bond would be partially isomerized to C–N single bond, which changed the molecular structure and the color dramatically changed to dark blue. The detection limit was as low as 1.4 nM, indicating its strong binding ability and high sensitivity detection ability to Ag(I). The **ABC** could be used for qualitative detection of Ag(I) in water and could be used as a physical developer for LFP development. Although the sensor could be regenerated by adding EDTA, the regeneration efficiency might be reduced.

Sun et al. designed and synthesized a switch-type bifunctional fluorescent probe, **DBP**, based on the structure of the natural product dehydroabiatic acid, which could be used for the detection of formaldehyde (FA) and Ag(I).³⁵ For the detection of Ag(I), the probe coordinated with Ag(I) through the $-NH_2$ group (1:1), as shown in Figure 17. After coordination, the HOMO–LUMO energy gap of the probe decreased to 3.166 eV, with the HOMO mainly located in the triphenylamine group and the LUMO mainly in the coordination site, and the decrease in energy gap stabilized the whole molecular system. Changes in the structure of the molecule and the distribution of the electron cloud led to a new absorption peak in the UV visible absorption spectrum at 440 nm, causing the solution color to change from colorless to

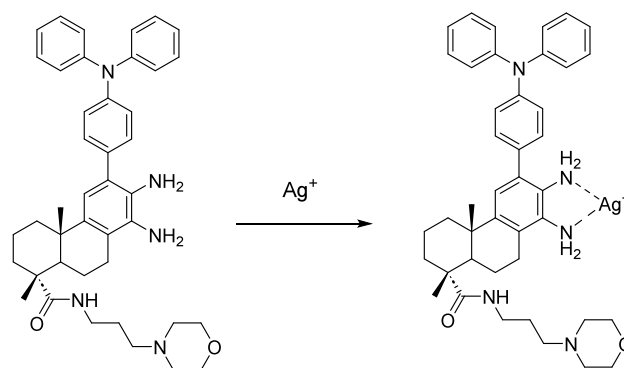


Figure 17. Organic sensor **DBP** for the detection of Ag(I) anions.

brown under sunlight, but the blue fluorescence at 425 nm was significantly quenched. Moreover, the probe could also be used for the detection of FA, with significantly enhanced fluorescence. This probe had low detection limits for FA and Ag(I) over a wide pH range (FA: 38 nM; Ag(I): 19 nM). This probe had been used for the detection of Ag(I) in food and water samples and had been successfully used for MCT-7 cell imaging and zebrafish in vivo imaging.

The response time of the probe also had an impact on its application potential. The conjugated system influenced the electron cloud distribution and energy state of molecules, and electron rich groups offered active sites for binding with metal ions. In the **DBP** probe, active groups such as the amino group reacted with silver ions at a faster rate. Moreover, the amino group in the **DBP** structure had less steric hindrance, allowing silver ions to quickly approach and combine with it. In addition, the conjugated system of **DBP** facilitated the ICT process. The high efficiency of the ICT process enables **DBP** to respond rapidly to target molecules.

3. REACTION-TYPE SILVER ION PROBE

Ruan et al. designed and synthesized a fluorescent probe **DTPAS** with two thioketal groups as the reaction site.³⁶ This probe used thioketal as the reaction site. Its thioketal groups could undergo specific reactions with Hg(II) and Ag(I). Hg(II) could quickly convert the thioketal into a carbonyl group, while Ag(I) could only convert one thioketal. This difference in reaction resulted in the generation of different products, which in turn produced different spectral signals, as shown in Figure 18. Specifically, the difference between the negative and positive regions of the probe was not significant, the dipole moment was small, and the intermolecular interactions were weak, resulting in fluorescence quenching. When reacting with Hg(II), the fluorescence was significantly enhanced (about 14.3 times), and the emission peak changed to 560 nm (red fluorescence). When reacting with Ag(I), the fluorescence intensity also significantly increased (41.2 times), and the emission peak was at 495 nm (green fluorescence). This change in color achieved a two-channel selective detection of Hg(II) and Ag(I). Due to the difference in sulfur affinity, the sensitivity and response speed of the probe for detecting Hg(II) were higher (92 nM, < 1 min). The test strips could be used for the portable determination of real water samples. But the response to Ag(I) was relatively slow, taking about 12 min to reach a stable spectral signal.

Cheng et al. developed a method for detecting Ag(I) and Cu(II) using dopamine (DA) and naphthol (NR) as substrates

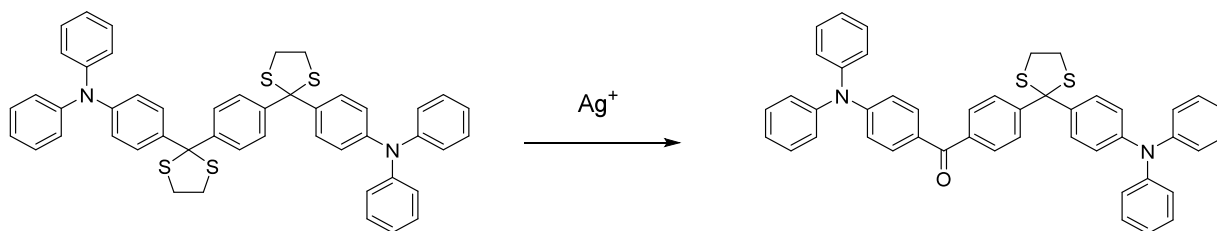


Figure 18. Organic sensor DTPAS for the detection of Ag(I) anions.

by incorporating NaCl or EDTA·2Na.³⁷ Ag(I) acted as an oxidant, and Cu(II) acted as a catalyst (O_2 participated in the reaction) to produced the reaction between DA and NR, as shown in Figure 19. The reaction produced DA-NR, a probe

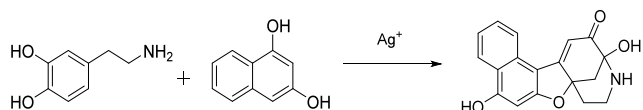


Figure 19. Organic sensor DA-NR for the detection of Ag(I) anions.

with a conjugated structure, whose molecular orbital energy level was changed, facilitating the jump of electrons from the excited state to the ground state and enhancing the fluorescence emission. Its fluorescent signal turns on, changing from nonfluorescent to blue fluorescent, and the color of the solution changed from colorless transparent to yellow. Moreover, the degree of color change was related to the concentrations of Ag(I) and Cu(II), with higher concentrations the more pronounced color changes. This fluorescence response had good specificity for Ag(I) and Cu(II). The detection limits of Ag(I) and Cu(II) were 157.1 nM and 11.9 nM, respectively, which were able to accurately detect low concentrations of Ag(I) and Cu(II). This method did not require synthesis, was simple and easy to operate, and had great potential in practical water source testing. The method needed to be used under specific conditions, which might affect the performance of the probe in practical applications.

Li et al. designed and synthesized a fluorescent probe KL1, with a large Stokes shift using coumarin as the fluorescent material for selective simultaneous detection of Ag(I) and Hg(II), as shown in Figure 20.³⁸ The electronic structure of the probe determined its fluorescence emission characteristics, emitting yellow fluorescence at around 640 nm. Ag(I) and Hg(II), as thiophilic metal ions, had a strong affinity for sulfur atoms in the probe, which could promote hydrolysis reaction of the probe, causing phenyl thiocarbonate to be removed from the probe, resulting in the fluorescence emission band shifting to 600 nm, accompanied by a significant decrease in emission intensity. Its color changed from dark yellow to light yellow/colorless, and its fluorescence changed from orange yellow to blue/bright blue. After adding Ag(I), the fluorescence emission

was immediately quenched and gradually increased, reaching equilibrium in about 140s. This response characteristic made the sensor able to detect the ions in the sample quickly. However, in cases where the detection of trace metal ions was highly demanded, the detection limit of the probe might not be sufficient.

Min et al. synthesized a near-infrared fluorescent probe, N-FP, based on intramolecular charge transfer (ICT) effect, as shown in Figure 21.³⁹ Due to the partial blocking of the ICT effect by thiocarbonate, the probe itself exhibited weak fluorescence. When Ag(I) and Hg(II) were added, due to their strong sulfur affinity, the binding of Ag(I) and Hg(II) with sulfur triggered the cleavage of the C–O bond to generate intermediate N–OH. Its ICT effect was restored, and the fluorescence emission was significantly enhanced. Meanwhile, the red shift phenomenon occurred due to the different absorption and emission properties of the N–OH intermediates generated by the hydrolysis of the probes. Under sunlight, its color changed from orange to magenta/purple. N-FP could quickly detect Ag(I) and Hg(II) over a wide pH range of 1–3 min with the lowest detection limits of 1.1 μ M and 0.72 μ M, respectively. When the water content in the solvent was greater than 50%, the fluorescence intensity of the probe significantly decreased and was almost quenched. This indicated that its application in high water content environments might be limited.

Zhang et al. developed a fluorescent probe, TMB, based on redox reactions, which reacted with Ag(I) and Hg(II) to oxidize amines in aniline, transforming colorless TMB into blue oxTMB.⁴⁰ Based on this phenomenon, TMB was designed and synthesized as a fluorescent probe for detecting Ag(I) and Hg(II), as shown in Figure 22. The main components of the probe included cellulose membranes (CMs) in addition to TMB. CMs had a highly porous structure and abundant active hydroxyl groups. TMB molecules were fixed to CMs through hydrogen bonds. When TMB was oxidized to oxTMB, hydrogen bonds could still be formed to fix oxTMB on the membrane, thereby amplifying the colorimetric signal. The visual detection limit (VLOD) of TMB as a colorimetric probe was 10 μ M, whereas that of TMB@CMs was 1 μ M, which was 1 order of magnitude lower, indicating that the probe fixation platform based on CMs effectively improved detection sensitivity.

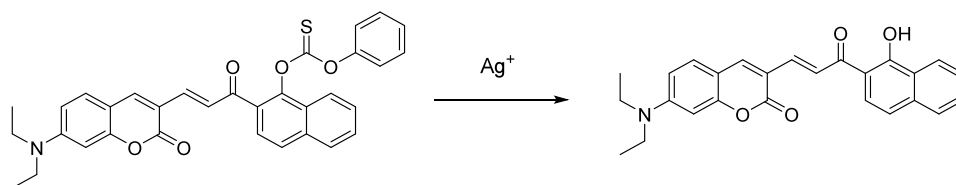


Figure 20. Organic sensor KL1 for the detection of Ag(I) anions.

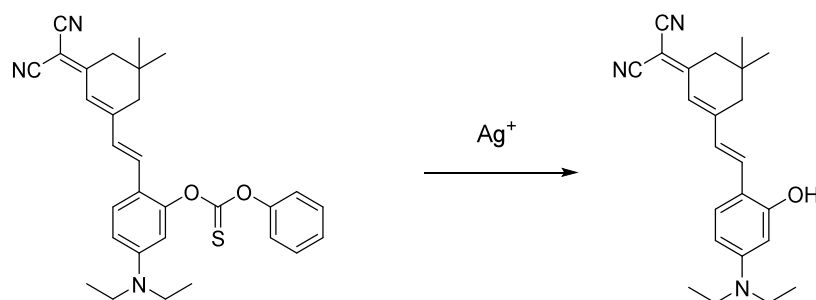


Figure 21. Organic sensor N-FP for the detection of Ag(I) anions.

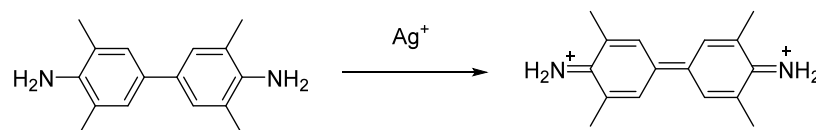


Figure 22. Organic sensor TMB for the detection of Ag(I) anions.

During the cyclic detection of Ag(I), Ag element would gradually accumulate on TMB@CMs, resulting in a gradual graying of the color, which might affect the performance and lifetime of the sensor.

Xu et al. synthesized the coumarin probe **CouMal** for configuring color fluorescent palettes, performing cell imaging, and in vivo imaging.⁴¹ Notably, this small-molecule compound represented a novel low-molecular-weight red-emitting fluorophore with great potential as an efficient and stable red emitter and biocompatible fluorescent imaging chromophore. When combined with triethylamine and malononitrile to form a composite sensing system, it could be used for the precise detection of Ag(I), as shown in Figure 23. The detection

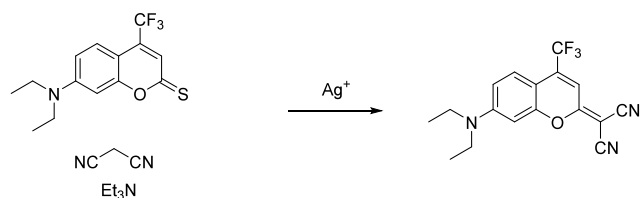


Figure 23. Organic sensor TC-Mal-Et₃N for the detection of Ag(I) anions.

principle was that Ag(I) could interact with coumarin in a non covalent bonding manner, causing the UV visible absorption spectrum of coumarin to be red-shifted. Subsequently, with the assistance of Ag(I), malononitrile rapidly attacked the thiol carbonyl group of coumarin, generating the highly fluorescent probe **TC-Mal-Et₃N**, thus achieving the detection of Ag(I). The probe HOMO was mainly located at the diethylamino group, and coordination led to a narrowing of the HOMO–LUMO energy gap, which was due to the introduction of a strong electron-absorbing group that enhanced the electron-absorbing capacity of the molecule, allowing the probe to be excited by lower energies, resulting in a red shift in the absorption and fluorescence spectra. As the concentration increased, the fluorescence intensity at 590 nm significantly increased.

The detection limit of the composite probe **TC-Mal-Et₃N** was as low as 60.9 nM. The Ag(I)-triggered composite probe had excellent selectivity and a short reaction time and had been used for accurate quantification of Ag(I) and AgNPs in

representative samples. Moreover, it provided a creative approach for the design of fluorescence sensors based on irreversible chemical reactions. However, the probes were based on specific chemical reaction detection and the reaction conditions (e.g., pH 8.5, etc.) were relatively stringent, which might limit their direct application in some extreme or complex environments.

4. CONCLUSIONS

Silver ions, as one of the important sources of pollution, had a great impact on people's lives and might cause various diseases. The development of sensors for detecting Ag and other metal ions provided an economically efficient means of protecting the environment and human health. Organic dyes containing various detection groups achieved specific and sensitive detection of silver ions through changes in apparent color and fluorescence color.

The selection of specific recognition groups was crucial. Sulfur-containing groups and specific heterocyclic structures had high selectivity for Ag(I), which could effectively recognize Ag(I) and eliminate interference from other metal ions. For example, the lone pair of electrons of sulfur atoms could form a strong coordination bond with Ag(I). In complex samples, sulfur-containing groups could specifically recognize Ag(I), which enabled the probe to produce a rapid signal change upon contact with Ag(I), and showed potential in applications such as biological imaging. However, sulfur atoms might be susceptible to oxidation or other chemical reactions under certain environmental conditions, which could affect the stability and lifetime of the probe. Simplifying the molecular structure of the probe and reducing unnecessary steric hindrance enabled faster interaction between the probe and Ag(I), accelerated the production of detection signals and achieved rapid detection of Ag(I). The introduction of more amino or hydroxyl groups could enhance the stability and fluorescence intensity, which was beneficial for improving the specificity and sensitivity of the probe. However, it had been noted that the excessive molecular weight of the probe would also affect the biocompatibility of the probe, thus limiting its wider application. Additionally, the spatial structure of the coordination site was also critical. The coordination site with less steric hindrance enabled silver ions to approach and bind to it rapidly.

In addition, we also discussed the reaction type mechanism probes, which usually had high sensitivity and specificity, because chemical reactions could cause obvious changes in molecular structure and properties, and could be designed as highly specific reactions for Ag(I), which was valuable for the detection of trace amounts of Ag(I) in environmental monitoring. Furthermore, we explored the color changes of organic probes, and these probes with significant color changes will greatly enhanced their practical application value in environmental detection. In the future, the design of probes could be combined with nanotechnology to improve the performance, or could use biotechnology to develop biosensors.

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Notes

The authors declare no competing financial interest.

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