

Review

Input Selection Drives Molecular Logic Gate Design

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Abstract: Optical detection devices have become an analytical tool of interest in diverse fields of science. The search for methods to identify and quantify different compounds has transposed this curiosity into a necessity, since some constituents threaten the safety of life in all its forms. In this context, 30 years ago, Prof. Prasanna de Silva presented the idea of sensors as Molecular Logic Gates (MLGs): a molecule that performs a logical operation based on one or more inputs (analytes) resulting in an output (optical modification such as fluorescence or absorption). In this review, we explore the implementation of MLGs based on the interference of a second input (second analyte) in suppressing or even blocking a first input (first analyte), often resulting in INHIBIT-type gates. This approach is interesting because it is not related to attached detecting groups in the MLG but to the relation between the first and the second input. In this sense, flexible and versatile MLGs can be straightforwardly designed based on input selection. To illustrate these cases, we selected examples seeking to diversify the inputs (first analytes and interfering analytes), outputs (turn on, turn off), optical response (fluorescent/colorimetric), and applicability of these MLGs.

Keywords: molecular logic gates; INHIBIT molecular logic gates; chemosensors; chemodosimeters; optical devices; analyte sensing; supramolecular analytical chemistry



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1. Introduction

Curiosity about what things are made of has led to a long and still ongoing quest to understand the chemical composition of matter. This journey began long before the advent of chemistry as a science. However, curiosity became a necessity when chemical components in diverse matrices threatened life [1,2]. In this sense, analytical chemistry emerges as a fundamental tool in several science fields for identifying and quantifying constituents in different matrices. Therefore, numerous analytical techniques have been perfected over the years, aiming to meet various criteria, such as selectivity, precision, accuracy, robustness, speed, and versatility, among many others [3–6].

In many cases, small concentrations (ppm or ppb) of substances, such as potentially toxic metals and harmful organic substances, may cause severe damage to human health. In this sense, analytical techniques for detecting and quantifying these small concentrations are essential. Optical methods play a key role among the approaches employed for this purpose because they generally have high sensitivity when recognizing an analyte. In this scenario, an optical detection device should preferably be ASSURED (Affordable, Sensitive, Specific, User-friendly, Rapid/Robust, Equipment-free, and Deliverable), such as point-of-care diagnostic devices based on the World Health Organization's conception [7,8].

The applicability of optical detection devices has evolved over the years from single-to multiple-analyte detection and, preferably, presenting responses in more than one channel. In this sense, a Molecular Logic Gate (MLG) is a molecule (device) that performs a logical operation using one or more logic inputs leading to a logic output. Analytes can be considered inputs, whereas a spectroscopic response could be described as an output. In the same way as conventional logic, the logical operations performed by molecules correspond

to both traditional operations, such as AND, OR, NOT ... INHIBIT, and even complex operations. Through this review, some of these logics will be explained and illustrated.

Organic synthesis provides several strategies for designing molecules to act as MLGs. Although different molecular architectures can be devised, this review shows that sensors can be interpreted as MLGs, mainly exploiting the reversible detection aspect. For instance, after a sensor detects a cation through complexation, the sequential addition of a specific anion or a neutral compound can remove this cation from the complex, releasing the optical device in a reversible process. In other words, this logical operation via an input selection may drive an MLG design and not necessarily the structure itself. The solvent is also essential to be evaluated in the analyte detection process because it interferes with the availability (solubility, diffusion, and kinetics) of a species to be detected and to interact with an MLG, leading to changes in optical properties.

2. Analytes

Cations, anions, or neutral molecules that present potential risks to human health have been used as targets for designing optical sensors. In this review, we focus on some of these analytes and decomplexing agents, such as competitive analytes, that can be detected by an MLG.

2.1. Potential Toxic Metals

The accumulation of toxic metals, such as lead, cadmium, mercury, and aluminum, in the human body can damage health [9]. The potential toxicity of these metals is influenced by several factors, including the dosage, route of exposure, and individual characteristics such as genetics, nutrition, and age [10].

Mercury is a toxic element that can cause significant harm to both children and adults. This metal can damage the nervous, renal, cardiorespiratory, immune, and reproductive systems [11–14]. Mercury toxicity is attributed to its affinity for thiol groups in proteins and enzymes [15–17].

Lead contamination commonly occurs through inhaling dust particles containing the metal, ingesting contaminated food and water, or contact with lead-containing paints [18,19]. Prolonged exposure to lead or its salts can result in nerve damage (especially in children), leading to blood and brain disorders [20,21].

The primary routes of cadmium contamination are inhaling cigarette smoke, ingesting contaminated food, and exposure to metallurgical industries. Cd^{2+} have an affinity for metallothioneins, composed of cysteine fragments and found in the liver, kidneys, intestines, and brain [22–24].

Aluminum is a versatile metal that become a practical component in daily human activities, including the production of utensils, electronic and electrical appliances, vaccines, wastewater treatment, and aerospace engineering [25,26]. However, exposure to aluminum has been linked to several neurological disorders, including Alzheimer's disease and other health issues, such as osteomalacia and breast cancer [27–29].

Nickel is important for biosynthesis and metabolism, but excessive amounts of this ion may lead to various health issues such as dermatitis, asthma, pneumonitis, disorders of the central nervous system, and cancer [30,31].

Copper is a vital cation that plays a critical role in redox activity, but an excess of this ion may cause amyloid precipitation and contribute to the development of Wilson's disease, prion diseases, and Parkinson's disease [32–34].

Zinc is an essential structural cofactor in metalloproteins and is associated with regulating gene expression [35,36]. In addition to that, Zn^{2+} also plays a crucial role as a regulator of cellular apoptosis and is present in most DNA or RNA polymerases [37]. The toxicity of this ion stems from its strong affinity for diverse amino acid side chains, notably histidine, cysteines, aspartates, and glutamates, causing several pathological processes, including diabetes and Alzheimer's disease [38,39].

Iron is a fundamental chemical element for maintaining life, as it is involved in several essential biochemical processes in human beings [40], such as redox reactions and syntheses of hemoglobin RNA and DNA [41].

2.2. Alkaline and Alkaline Earth Metals

Sodium and potassium are found in mineral form, mainly in saline water. These elements are involved in critical biological processes, including regulating blood and body fluid homeostasis; contraction of cardiac, skeletal, and smooth muscle; taste and pain sensation; hormone secretion; and animal signal transduction [42–46]. Potassium salts are commonly used in the production of fertilizers. When in excess in the human body, Na^+ can cause various health issues, including water retention and hypertension [47].

Mg^{2+} is an essential ion in several biochemical cellular processes, such as cell proliferation and death, enzymatic processes, and several others [48,49]. The unbalanced concentration of this ion in the body may be associated with several disorders, such as diabetes, osteoporosis, metabolic syndrome, and chronic renal failure [50].

2.3. Anionic Species

Cyanides are a family of chemical compounds with different variations in chemical complexity and which contain a CN^- portion. The extreme toxicity of this ion is due to its ability to form complexes with ferric ions in some enzymes' active sites. Upon binding to an enzyme, cyanide interferes with the electron transport chain, which impairs the tissue's ability to utilize oxygen effectively [51,52]. In this sense, exposure to CN^- can cause acute and systemic toxicity, leading to rapid cardiovascular collapse [51,53–55].

Fluoride (F^-) is related to healthy bones, hair, and nails. Furthermore, this anion is used in toothpaste and drinking water to avoid dental caries. Nevertheless, excess F^- can result in dental or skeletal fluorosis, renal lithiasis, and other disorders [56]. Due to its highest reported charge density, F^- is classified as a hard Lewis base, which inspires the development of several optical sensors bearing acid moieties to detect this anion [57,58].

Acetate (AcO^-) is an important ion of several biological processes. For instance, the rate of AcO^- formation and oxidation could be used as an indicator of organic decomposition in marine sediments [59,60]. Furthermore, sodium acetate can be applied to control microbial growth and added to foods to extend shelf life and improve sensory properties [61,62].

Citrate, employed as a flavor in foods and pharmaceuticals, is an important intermediate in the Krebs cycle [63]. Abnormal citrate levels could be associated with several diseases, such as nephrolithiasis, nephrocalcinosis, and glycogen storage disease [64].

Phosphates are essential for building fundamental molecules in living organisms like DNA and RNA [65]. They also play a crucial role in membrane lipids and biological processes, such as skeletal development, energy metabolism, and protein synthesis. About 85% of the body's phosphate is in bones, with deficiency leading to severe consequences like osteomalacia and central nervous system dysfunction. On the other hand, excessive phosphate (hyperphosphatemia) can also be harmful, causing seizures, kidney disease, and soft tissue calcification [66–69]. Pyrophosphate (PPi) consists of two phosphate groups bonded together and plays a crucial role in biological processes, such as control of the accumulation of calcium salts in tissues, blood vessels, and organs [70,71]. They also serve as biomarkers for medical conditions, essential in energy transfer, enzymatic regulation, and DNA and RNA synthesis [72,73].

Despite the relevance, the detection of anions is more complex than that of cations. Designing anionic recognition sensors requires considering factors such as molecular geometry, stereochemistry, and energy-related properties such as enthalpy, entropy, Gibbs free energy, and solvation [74,75]. In addition, anions such as HCO_3^- , NO_3^- , PO_4^{3-} , and HSO_4^- can exhibit different sizes and charges at different pH values, which can impact their recognition and detection [76].

2.4. Aminoacids

Detecting electrically neutral analytes by chemical and biochemical sensors is a significant challenge, mainly due to the limitation of ligands and indicator molecules capable of accurately recognizing neutral molecules [77]. Many neutral molecules have been the target for sensor design. Among these molecules, we can mention, for instance, biogenic amines [78], chemical warfare agents [79–81], illicit drugs [82,83], and pharmaceutical drugs [84–87]. In addition, recently, considerable attention has also been given to amino acids.

Amino acids are essential for various animal and plant metabolic processes and biological functions. Lysine (Lys) is critical in the Krebs–Henseleit cycle and polyamine synthesis, and it plays a vital role in animal growth and weight gain [88,89]. Histidine (His) is essential for the active sites of many enzymes and functional proteins, and it controls the transmission of metallic elements in biological systems [90,91]. It is present in the muscle and nerve tissues and forms nerve peptides in the brain. Tryptophan (Trp) is crucial for protein biosynthesis, animal growth, and plant development. Deficiency of specific amino acids can lead to various abnormalities [92]. For instance, lack of cysteine (Cys) can result in slow growth, capillary depigmentation, edema, lethargy, liver damage, and muscle and fat mass loss [93–95].

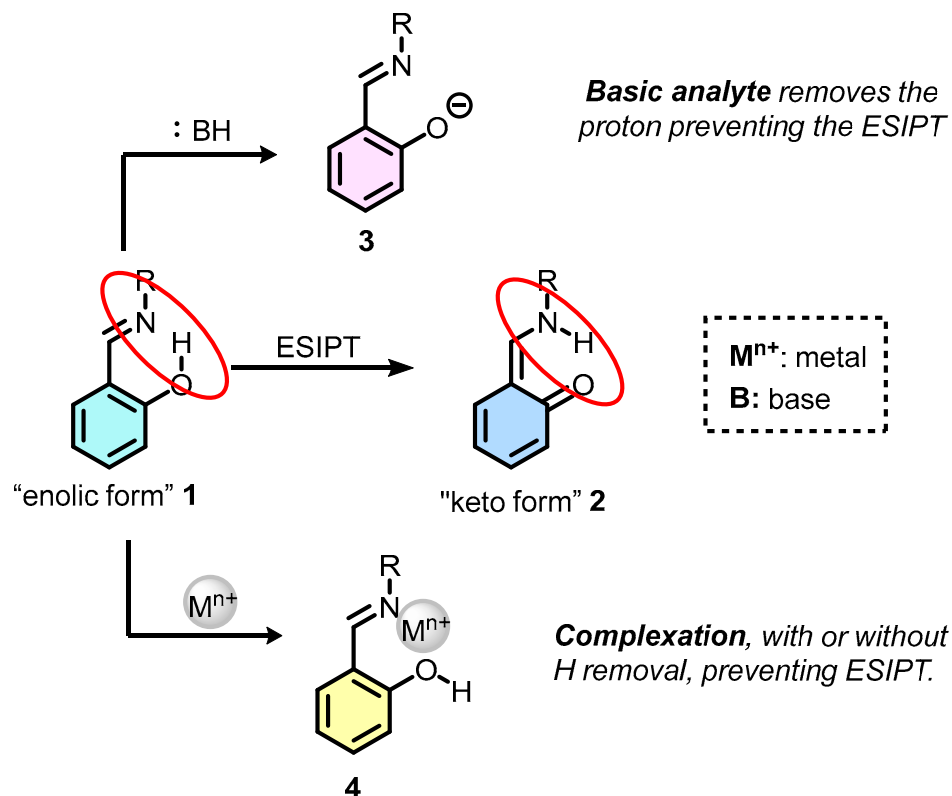
3. Optical Detection Devices

A molecule designed as a detection device usually bears a chromophore/fluorophore unit as the signaling unit for the optical response, as well as a receptor group, which interacts with the analyte. Moreover, according to the molecular architecture, different molecule moieties can act as donors (D) or acceptors (A) of electron density. Therefore, systems such as D-A, A-D-A, and D-A-D can be elaborated. Upon analyte detection, the optical response is usually associated with changes in the electronic density of the sensor interfering in the donor/acceptor system. Different processes can describe this change, but the photoinduced electron transfer (PET) process is the most common. In this phenomenon, light absorption leads to the transfer of an electron from one molecular entity to another, resulting in significant changes in the system's energy levels and optical properties. Some subclasses of PET can be considered:

- a. Internal (or intramolecular) charge transfer (ICT): An electron-rich donor moiety transfers charge to an electron-deficient acceptor moiety; therefore, the entire process occurs within a single molecule [96].
- b. Metal–ligand charge transfer (MLCT): Involves the transfer of electronic charge between a metal ion and a ligand, as the d orbitals of the metal atom and the ligand orbitals (p, d, or f, depending on the type of ligand) can overlap. It predominantly occurs in metal complexes [97].
- c. Twisted internal (or intramolecular) charge transfer (TICT): The occurrence of a direct transfer of charge between nonplanar molecular conformations through the formation of a donor–acceptor complex [98,99].

Another phenomenon usually explored in the elaboration of optical detection devices is the Excited State Internal Proton Transfer (ESIPT, Scheme 1) [100]. As the name implies, a proton from an “enolic form” (1), usually attached to an atom such as nitrogen or oxygen, is transferred to another atom after absorbing energy, resulting in a “keto form” (2). However, an analyte can interfere with this process, preventing this phenomenon from occurring. For instance, a basic analyte can remove this proton (3), or a cation can coordinate to this site (4) [101]. In these cases, detection will occur by disrupting the ESIPT by modifying the optical properties of the sensor, inducing a separate double fluorescence emission in a cycle of four levels that incorporates the ground and excited states of two different tautomers. ESIPT-based molecules have received significant attention due to the large Stokes-shifted fluorescence emission ($6000\text{--}12,000\text{ cm}^{-1}$) without self-reabsorption, which is particularly interesting in the construction of sensors as they prevent interference from other fluorescent species [102,103]. ESIPT-based sensors are extremely sensitive to

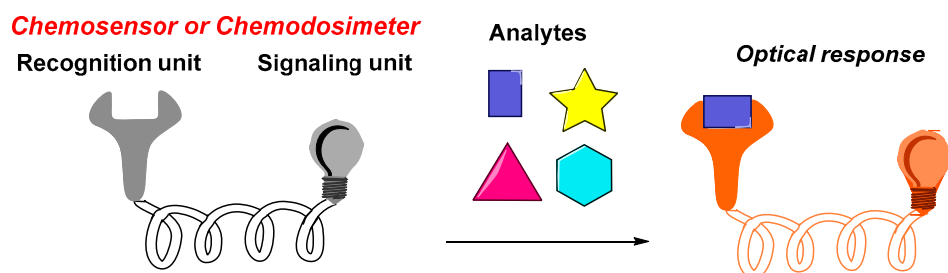
environmental changes. Therefore, their application is often restricted to aprotic solvents, as depending on the solvent, such as water, it can act either as a hydrogen bond donor or as a disturbing acceptor [104]. Many sensors have been designed exploiting this strategy of ESIPT interference by analytes, and some of them are discussed in this review.



Scheme 1. Generic representation of the ESIPT process and its prevention by basic and metallic species in case of analyte detection.

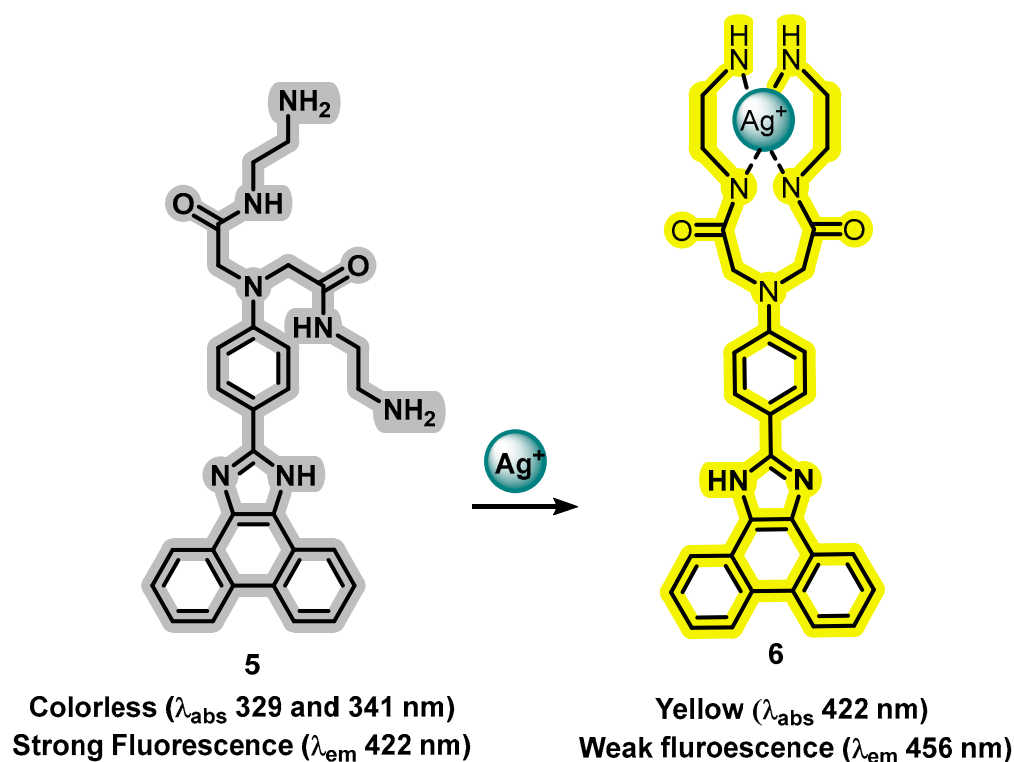
Several other processes, including aggregation phenomena (AIE and AIQ) [105–107], FRET [108,109], and Two-Photon Fluorescence [110], can also be described. A detailed discussion of these phenomena is beyond the scope of this review.

Sensors can be categorized as chemosensors or chemodosimeters, depending on the type of interaction with the analyte. The difference between the two terms is related to bond formation/breaking as well as the reversibility of the process. Chemodosimeters generally involve breaking and forming bonds in an irreversible process. On the other hand, chemosensors are usually associated with reversible processes in a way that does not involve structural transformation of the sensor. Both devices can be simplified schematically by a signaling unit (chromophoric or fluorophoric) coupled to a receiver, according to Scheme 2.



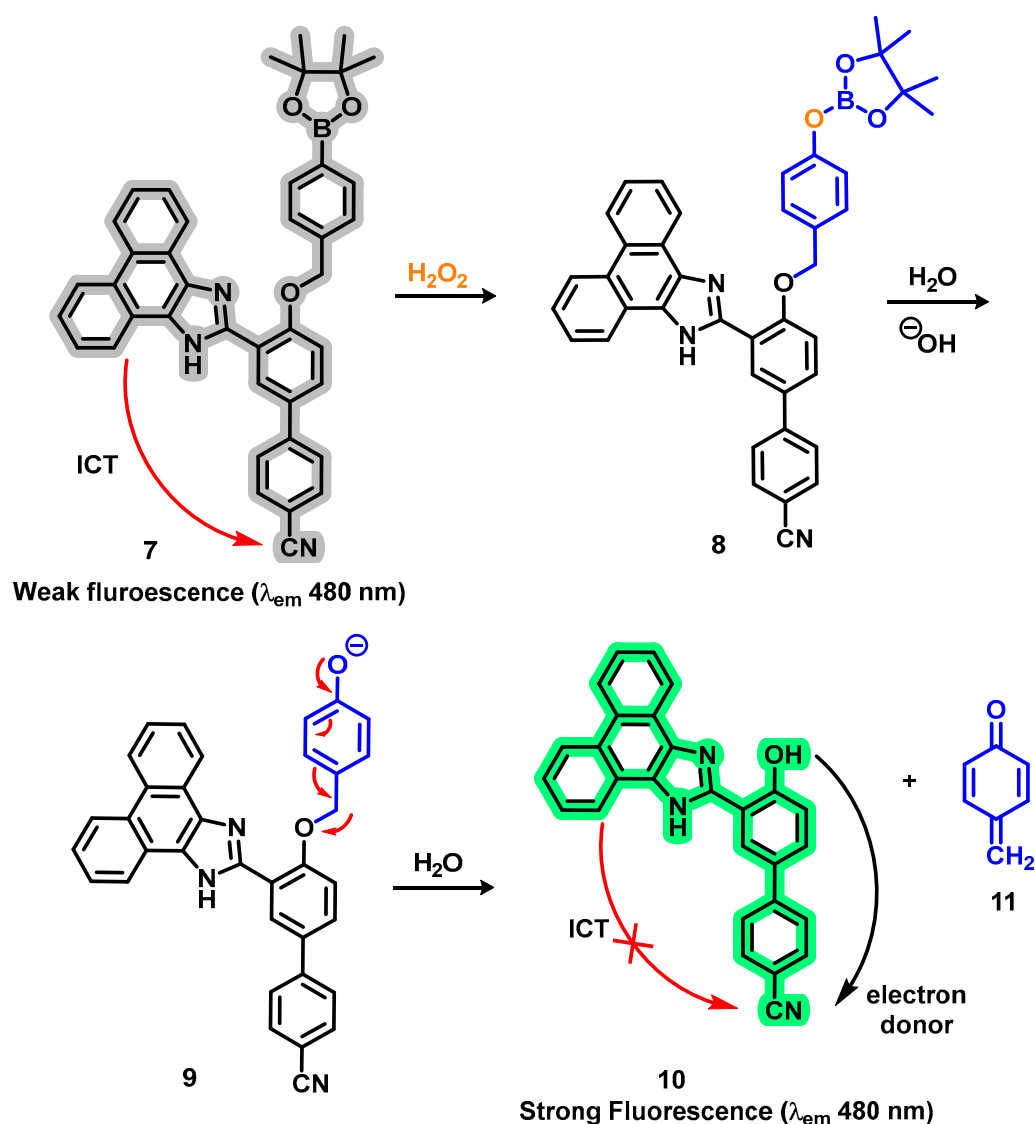
Scheme 2. General representations for an optical chemosensor and a chemodosimeter selective for a specific analyte.

Concerning chemosensors, a basic analyte can interact with a proton donor (NH, OH, SH) via hydrogen bonding, and a cation can interact with basic (coordinating) groups. These chemical processes can change a chemosensor's color or fluorescence, identifying and quantifying the analyte. Zhao et al. described an example of a chemosensor (**5**) based on the fluorescent phenanthro[9,10-*d*]imidazole core with a flexible ethylenediamine binding group (Scheme 3) [111]. The compound **5** solution exhibited absorption bands at 329 and 341 nm, and adding Ag^+ led to a new absorption at 422 nm, with a naked-eye color change, turning the colorless solution into a yellow solution. Furthermore, the fluorescence emission of **5** at 422 nm was reduced with a red-shift of 34 nm due to the addition of Ag^+ . Based on FTIR and NMR investigations, the authors proposed that the optical modification results from the interaction of Ag^+ with **5** affording compound **6** (Scheme 3).



Scheme 3. Chemosensor **5** was described by Zhao et al. based on the fluorescent phenanthro[9,10-*d*]imidazole core [111].

Regarding chemodosimeters, Chen et al. described the weak emissive compound **7**, also bearing a phenanthro[9,10-*d*]imidazole core (Scheme 4) [112]. Adding hydrogen peroxide caused a slight disturbance in absorption intensity at 367 nm, with no wavelength shift. The fluorescence intensity significantly increased at approximately 480 nm due to the formation of fluorescent product **10** induced by hydrogen peroxide (H_2O_2). The change in optical properties was attributed to the inhibition of the ICT process. The limit of detection (LOD) was determined to be $148 \text{ nmol} \cdot \text{L}^{-1}$, and H_2O_2 exhibited a robust linear relationship ($R^2 = 0.999$) within the concentration range of 0 to $150 \text{ } \mu\text{mol} \cdot \text{L}^{-1}$. As seen in this case, contrary to the example described by Zhao et al. (Scheme 3) [111], the sensor undergoes a chemical transformation; so, the reverse reaction is unlikely. Therefore, **7** can be classified as a chemodosimeter.



Scheme 4. Chemodosimeter **7** described by Chen et al. for the detection of H_2O_2 [112].

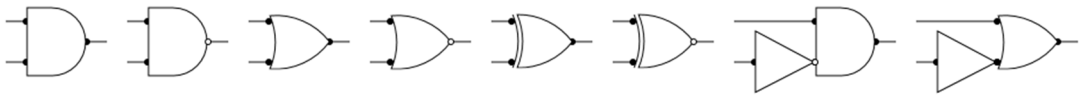
4. Molecular Logic Gates

In a computing device, the flow of the information, as proposed by George Boole, is based on binary code language whose variables can assume one of two possible values: false or true (F, V), high and low (H, L), or simply (0, 1). The binary digits (0, 1) are associated with the two states of the logic circuit. Usually, a threshold value is taken as a reference, and the state of the circuit is defined as “0” if the value observed is lower than this threshold or “1” if the value is higher than this limit [113].

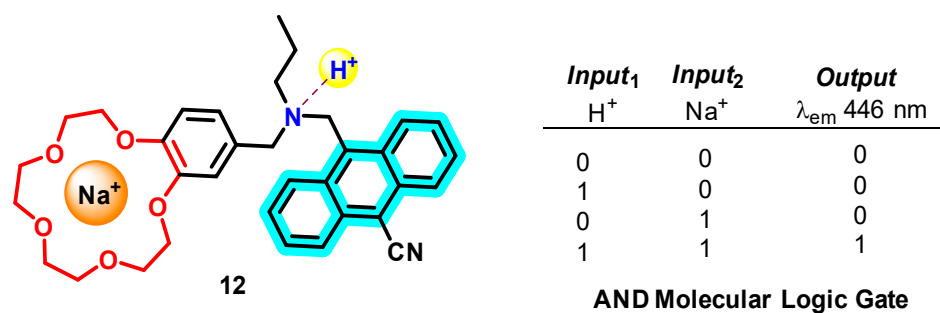
Based on different inputs and outputs, logic gates (LGs) are classified, based on Boolean logic functions, as NOT, OR, AND, and all their possible combination as described through conventional symbols or a truth table (Table 1) [114]. By applying these concepts to detection devices, absorption or emission above a defined value can be considered state “1” and, below this value, state “0”. The presence of an analyte (input) at a given concentration can also be described as a “1” status, while the absence of that same analyte (input) can be assigned a “0” status.

Table 1. Truth tables for generic examples of the most common LGs found in the literature.

Logic Gate Truth Table with Single Input				
Input	Output PASS 0	Output NOT	Output YES	Output PASS 1
0	0	1	0	1
1	0	0	1	1

Logic Gate Truth Table with double input									
									
Input ₁	Input ₂	AND	NAND	OR	XOR	NOR	XNOR	INHIBIT	IMPLICATION
0	0	0	1	0	0	1	1	0	1
0	1	0	1	1	1	0	0	0	1
1	0	0	1	1	1	0	0	1	0
1	1	1	0	1	0	0	1	0	1

Three decades ago, Prasanna de Silva et al. introduced the concept of the *Molecular Logic Gate*, a molecule that performs a logical operation based on one or more stimulus signals (inputs), resulting in a response (output). In their groundbreaking work, the authors elucidated a new compound (**12**) featuring an anthracene derivative connected to a crown ether via a tertiary amine (Scheme 5) [115]. The fluorescence emission of anthracene moiety in this compound was suppressed due to the PET phenomenon, attributed to the methylamino group attached to the anthracenyl moiety. Consequently, in the absence of additives (input “0”), the fluorescence response remains weak (output “0”). The tertiary amine undergoes protonation by adding acid (input H⁺ “1”) to **12**; however, the PET process persists, maintaining a low fluorescence response (output “0”). Likewise, the addition of sodium cations (input Na⁺ “1”), in the absence of acid (input H⁺ “0”), leads to chelation by the crown ether group, but PET still occurs, resulting again in a low fluorescence response (output “0”). However, the simultaneous addition of H⁺ and Na⁺ stimuli (input H⁺ “1” and input Na⁺ “1”) triggers a remarkable increase in fluorescence intensity (output “1”). Therefore, according to the respective truth table, **12** can be classified as an AND-type logic gate, as illustrated in Scheme 5.

**Scheme 5.** MLG **12** of the AND-type described by Prasanna de Silva et al. in 1993 [115].

If we consider multiple analytes (inputs) resulting in different responses (outputs), different MLG combinations can be proposed based on truth tables interpretations, and some examples and rationalization of them are described through this review.

4.1. General Strategies for Building MLGs

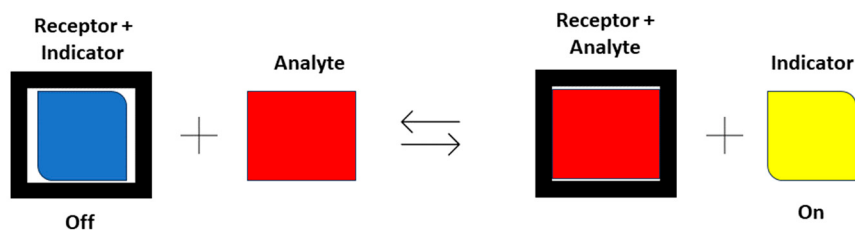
Any optical detection device based on fluorescence emission intensity can be interpreted as an MLG (Table 1). If this device is low or nonemissive (Output “0”) in the absence of chemical species (input “0”), a YES-type MLG can be described if the addition (operation) of a specific chemical species (input “1”) leads to a significant increase in fluorescence

(output “1”). Similarly, a NOT-type MLG, with a single-input operation (input “1”), leads to reduced fluorescence (output “0”). These are the most straightforward cases of MGLs, in which only one stimulus signal (single input, single analyte) leads to a response. On the other hand, when two or more analytes are being recognized, different types of MLGs can be constructed. For instance, in the case of two (or more) simultaneous analytes in a system (input₁ “1” and input₂ “1”) producing an optical response (output “1”), an AND-type MLG will be described [116,117].

The INHIBIT gate is another type of logic gate applied for two inputs, which integrates the NOT and AND gates, although in different connectivity of a NAND gate. In INHIBIT logic, the NOT operation is only applied to input₂. Consequently, input₂ serves as a disabled input, which, when “1 (high, true)”, inhibits the output regardless of the state of input₁ [118].

This review focuses on MLGs where input order does not dictate priority, allowing observation of specific outputs. Although more intricate devices such as those involving addition and subtraction, transfer and keypad lock mechanisms, multiplexers, encoding, and comparators have been addressed in the literature [119–128], and some examples are mentioned in this review, they are not elaborated in detail.

Chemosensors based on displacement assays, capable of detecting two or more analytes, can be described as an INHIBIT-type MLG when a second analyte vetoes the optical response of the first one. Optical devices based on displacement tests are effective for the recognition of multiple species indirectly in a sequential mode. This strategy allows reversibility experiments, where two species are added alternately to a medium. The process follows Scheme 6, where an indicator (off-blue block) initially coordinates with the receptor (black), which can be a host molecule or a metal atom. Subsequently, a competitive analyte (red block) is introduced, causing the indicator to shift into a solution (on-yellow block) [129–131].



Scheme 6. Schematic representation of displacement assay rationalization.

In practice, MLGs based on displacement assays are interesting, particularly in multiple analytes detection, because they provide a simple way to identify a second analyte (second input) according to the target analyte (first input). In this sense, the response generated between the interaction indicator and the first analyte is blocked by a competitive analyte, in other words, inhibiting it. INHIBIT MLGs are notably identified by constructing the two analytes, input₁, and input₂, with an inhibited response, i.e., the response of the first complex corresponding to the output.

4.2. Strategies for Building MLGs Based on Molecular Architecture

An optical detection device is usually designed to contain two groups: a signaling unit (chromophore and or/fluorophore), responsible for the optical response, and a detector unit, responsible for interacting with the analyte. Although the distinction between these groups is not evident in many cases, their arrangement is the basic architecture of molecular sensors. Therefore, integrating these two groups may change the electronic density upon interaction with an analyte, modifying processes such as PET, ICT, and ESIPT and causing the optical response.

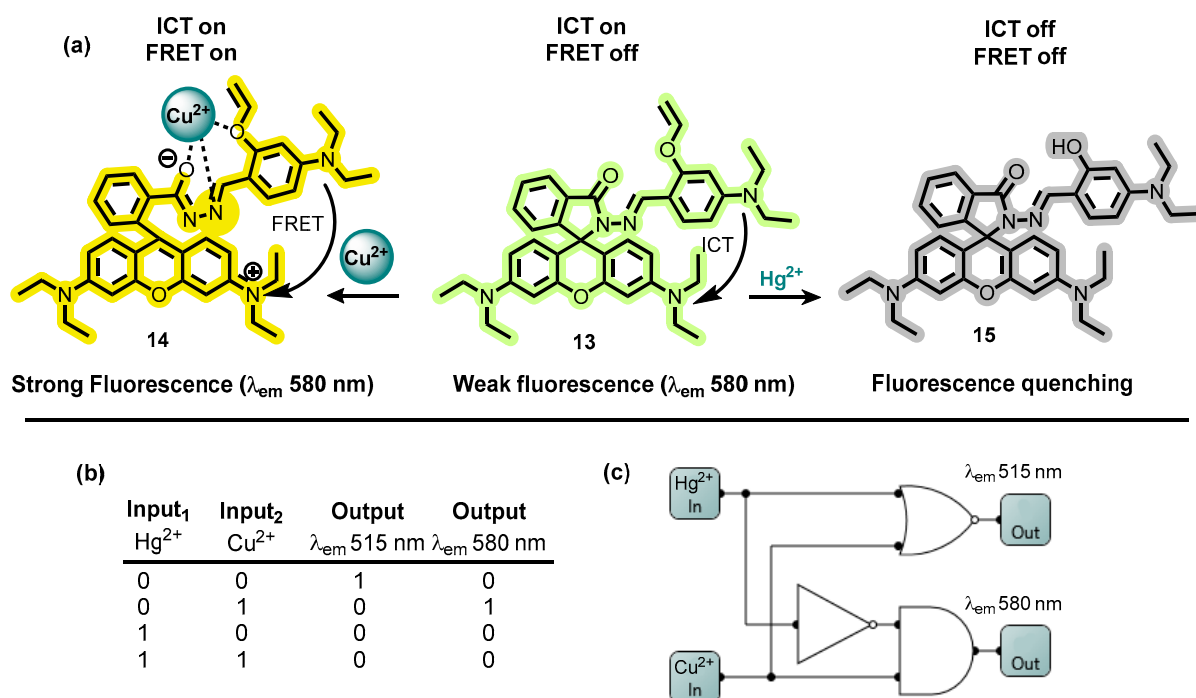
Different heterocycles have been proposed in the literature as signaling units, generally conjugated and planar compounds capable of displaying color and fluorescence. Some

examples include imidazoles [132–135], phenazines [136,137], rhodamines [138–140], and coumarins [141–144], among others [145–162].

Several molecular groups can be attached to the signaling units to recognize chemical species through emission and absorption change. For instance, macrocycles, such as crown ether, have been described in the literature as chelators of different metals according to the size of the macrocyclic and its heteroatoms [163–167]. Acidic groups like phenolic hydroxyls recognize basic species, while basic units like amines recognize acidic substances. Coordinating moieties can selectively interact with metallic species. Another well-known example is the formyl group, which may react with bisulfite, leading to a change in the conjugation of the molecule and, consequently, changes in absorption and emission [168,169]. Furthermore, in distinguishing between reversible chemical processes (chemosensors) and irreversible chemical processes (chemodosimeters), the detector unit holds an essential position within the molecular architecture of MLGs. Detection mechanisms can occur through different processes in this detection unit: chemical reactions involving functional groups, acid–base reactions or interactions through hydrogen bonds, and metal complexation and redox mechanisms.

In this sense, different synthetic strategies can be thought to design organic molecules that can act as MLGs. Molecular groups can also be installed targeting a specific analyte, and signaling units can be designed to present a particular response.

Building structures with two interaction/reaction sites or different detection mechanisms effectively may discriminate cations. For instance, Dong et al. synthesized a dual-function MLG **13** bearing a rhodamine hydrazone and O-vinyl-protected hydroxyl benzaldehyde (Scheme 7a) [170]. This compound displayed a weak emission at 580 nm. However, **13** showed ratiometric detection of Cu^{2+} with a large Stokes shift (150 nm) via an emission enhancement at 580 nm, attributed to the ring-opened rhodamine derivative **14**. Additionally, **13** exhibited high selectivity and sensitivity as a chemodosimeter for Hg^{2+} through removing vinyl ethers, resulting in significant fluorescence quenching due to the formation of **15**.



Scheme 7. (a) Mechanism of the hydrolysis reaction of vinyl ethers in **13**. (b) Truth table and (c) MLG diagram reported by Dong et al. [170].

Combining Cu^{2+} and Hg^{2+} as input signals on MLG **13** led to a logic operation characteristic for a two-input INHIBIT gate on the emission at 580 nm (Scheme 7b,c). In

other words, the presence of Hg^{2+} disallows the presence of Cu^{2+} , since the sensing occurs by different mechanisms.

Beyond molecular architecture, there is an interesting strategy widely employed in elaborating MLGs: a second input (analyte) that interferes with “first analyte/MLG interaction”, causing a perturbation in the system. In this case, no molecular groups are attached at the MLG to detect a second analyte once it interferes with the detection of the first one. In this sense, as discussed in the following examples, usually after detecting an analyte (input_1), a chemical species (input_2 : cation, anion, or neutral molecules) can be added to interfere in forming the previously formed complex. This second chemical species can operate in different ways, such as removal by complexation and removal of the cation (input_1) through the formation of a poorly soluble salt, among others [171].

4.3. MLGs Based on EDTA as Input_2

Complexing agents, such as EDTA, are commonly employed in competitive assays for constructing MGLs based on forming metal–ligand complexes to enable the description of a sensor as MLG. In this approach, a molecule (e.g., a complexing agent) capable of interacting more efficiently with the metal is introduced into the medium. Consequently, the metal–sensor complex is undone, releasing the sensor and recovering its optical properties. Moreover, the added complexing agent inhibits the action of the sensor, which can then be described as an INHIBIT MLG.

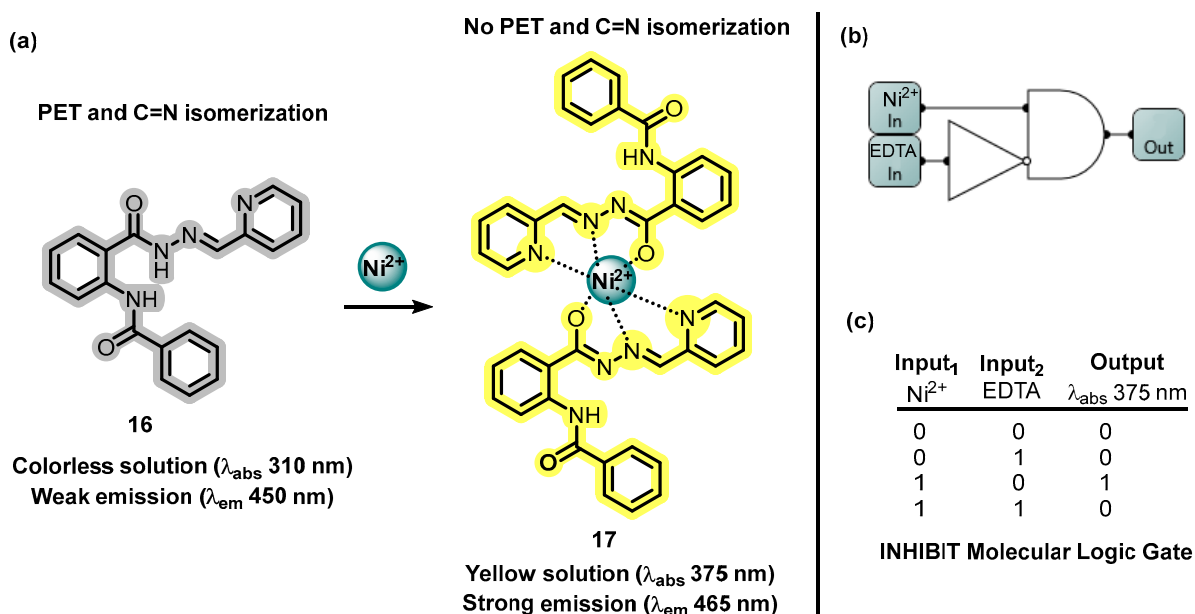
EDTA acts as a remarkable input_2 due to its ability to form chelates with nearly all cations, excluding those of alkali metals. What sets it apart even further is the exceptional stability most of these chelates exhibited, as evidenced by the formation constants in Table 2 [172]. This high stability is attributed to the multiple complexing sites within the EDTA, which give rise to a cage-like structure. As a result, the cation becomes effectively enclosed and isolated from the surrounding solvent molecules.

Table 2. Complex EDTA–Cation formation constants.

Cation	K_a	Log K
Mg^{2+}	4.9×10^8	8.69
Ni^{2+}	4.2×10^{18}	18.62
Cu^{2+}	6.3×10^{18}	18.80
Zn^{2+}	3.2×10^{16}	16.50
Hg^{2+}	6.3×10^{21}	21.80
Pb^{2+}	1.1×10^{18}	18.04
Al^{3+}	1.3×10^{16}	16.13
Fe^{3+}	1.3×10^{25}	25.10

a: The constants are valid at 20 °C and have an ionic strength of 0.1.

An example of a sensor described as MLG based on EDTA input as a complexing agent for analyte removal was described in 2019 by Manna et al. [173]. The authors have proposed a novel hydrazide-based optical MGL **16** capable of detecting Ni^{2+} (Scheme 8a). The rationalization involved designing the chemosensor with a structure sufficiently flexible to complex cations, containing distinct chromophoric groups and -CONH- groups that would enhance solubility in an aqueous media. The studies were conducted in a methanol–Tris-HCl buffer medium solution ($10 \text{ mmol} \cdot \text{L}^{-1}$, pH 7.2, 1:1 *v/v*), where the chemosensor demonstrated stability for two weeks, and no competition with H^+ for metal binding was observed. The free chemosensor in solution exhibited an absorption band at 310 nm, attributed to the $\text{S}_0\text{-S}_1$ transition. Upon interaction with Ni^{2+} , a red shift was observed, with maximum absorption at 375 nm, turning the colorless solution into a yellow solution. According to the authors, the complexation of the metal through the imine moiety delays the isomerization of $\text{C}=\text{N}$ and PET, thereby enhancing the fluorescence emission through the CHEF process.



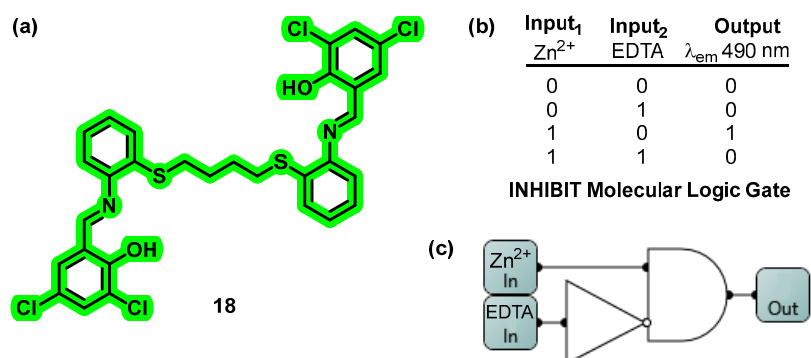
Scheme 8. MLG **16** described by Manna et al. for detecting Ni²⁺ using Na₂EDTA as strategic input: (a) Sensing mechanism. (b) Diagram and (c) truth table corresponding to INHIBIT-type logic [173].

The interpretation of the sensor as a logic gate was based on the reversibility of **17** upon introducing EDTA that coordinates with Ni²⁺, reintroducing the chemosensor in its free form in solution with the recovery of the optical properties of the metal-free system. The absorption intensity at 375 nm was used as the output, with Ni²⁺ and EDTA serving as inputs. The corresponding binary system (Scheme 8b) and truth table (Scheme 8c) represent an INHIBIT-type logic.

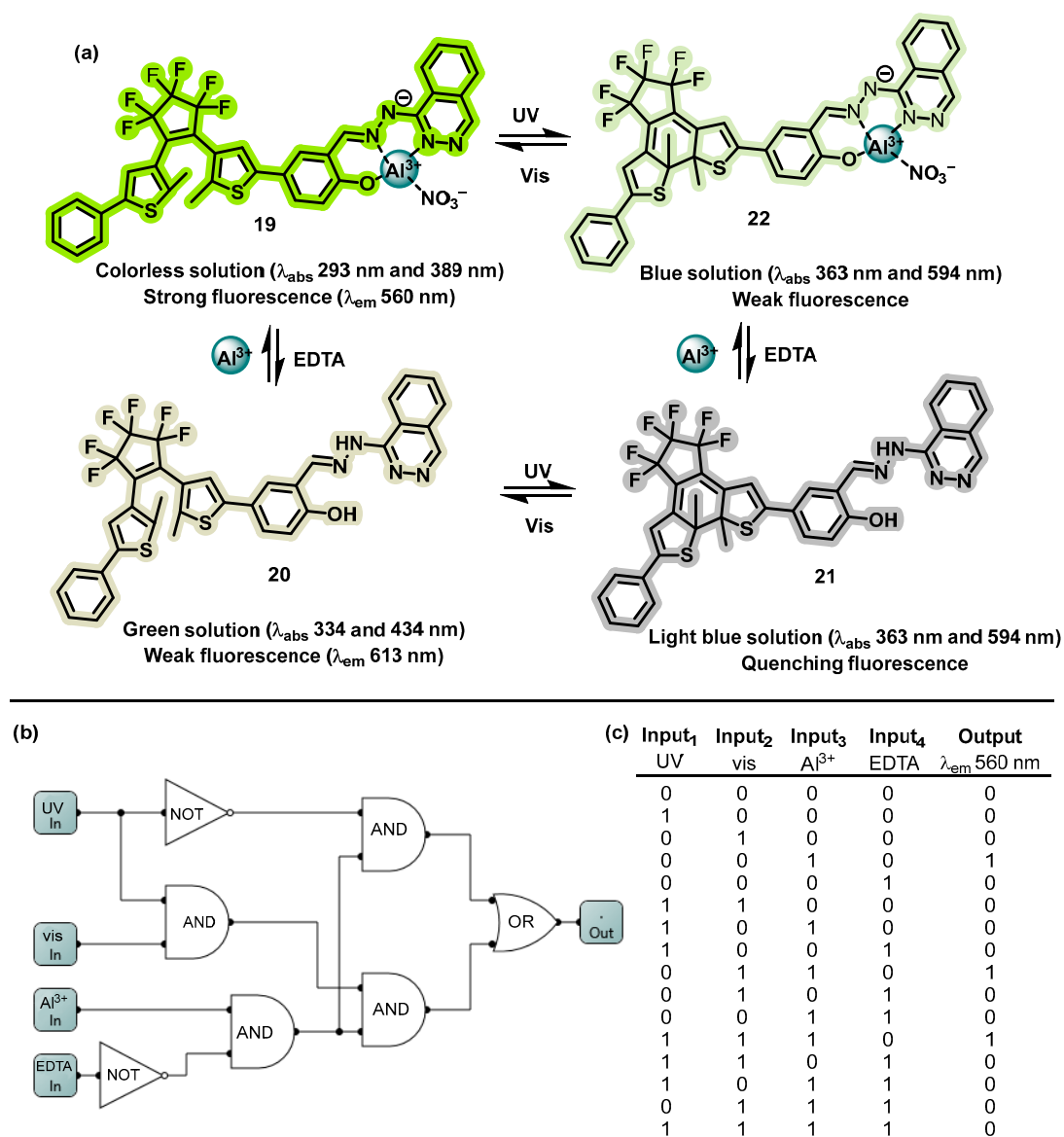
A similar approach was described by Acharyya et al. based on a simple Schiff base fluorescent chemosensor **18** sensitive to Zn²⁺ (Scheme 9a) [174]. ESIPT and PET processes were associated with the fluorescence quenching in the metal-free sensor (which exhibits only a weak emission band at 556 nm). Upon the interaction of thioether groups with Zn²⁺, the new complex inhibits these two processes and establishes the CHEF process significantly, resulting in a new emission band at 490 nm, which is 64 times more intense in the presence of Zn²⁺. The LOD of Zn²⁺ by **18** was calculated to be 1.73 nmol·L^{−1} from fluorescent spectral titration, and the formed complex exhibits an association constant of 6.91 × 10⁴ M^{−1}. Adding EDTA to the chemosensor-Zn²⁺ solution reduces the emission intensity at 490 nm. In other words, EDTA inhibits the response obtained from the complexation of **18** with Zn²⁺. Therefore, the authors received an INHIBIT-type circuit (Scheme 9b,c).

In 2019, Qiu et al. designed the new fluorescent compound **19** containing a phthalazine unit that functions as a multicontrolled fluorescent device under UV/vis radiation and Al³⁺/EDTA (Scheme 10a) [175]. When the solution of **19** is irradiated by UV light (297 nm), the absorbance at 293 nm decreases gradually, and two new bands appear (363 and 594 nm), turning the colorless solution into a blue solution. The influence of the excitation wavelength was analyzed in the fluorescence spectra. Under a 410 nm lamp, a weak emission band at 613 nm is described (formation of **20**). However, when the solution is irradiated with a 297 nm lamp, this emission band is gradually quenched because nonfluorescence isomer **21** was formed. Compound **21** was also used in the detection of Al³⁺, reaching an LOD of 5.5 × 10^{−8} mol·L^{−1}. Upon coordination with Al³⁺, the emission intensity of the solution at 560 nm increases significantly. The formed complex (**22**) inhibits PET and the isomerization of C=N, consequently enhancing the rigidity of the structure. The fluorescence intensity of **19** was recovered by adding EDTA. The authors constructed a combinational logic circuit with those four input signals (UV and Vis lights, Al³⁺, and EDTA) with the

emission intensity at 560 nm as the only output signal. Scheme 10b,c summarizes the total logic in their circuit.

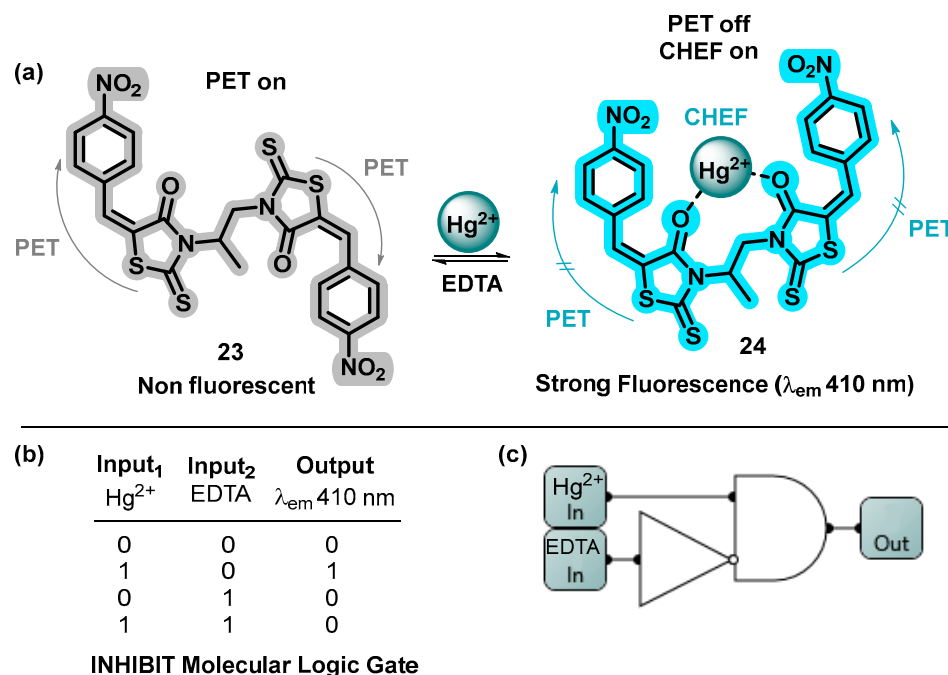


Scheme 9. (a) MLG 18 described by Acharyya et al. for detecting Zn²⁺ using EDTA as strategic input, (b) truth table corresponding to INHIBIT-type logic, and (c) the corresponding diagram [174].



Scheme 10. (a) MLG 19 described by Qiu et al. for detecting Al³⁺ using EDTA as strategic input, (b) the diagram, and (c) the corresponding truth table [175].

In 2021, Kumar et al. described a versatile fluorescent MLG **23** capable of Hg^{2+} recognition (Scheme 11) [176]. The chemosensor comprises a rhodanine and 4-nitrobenzaldehyde derivative that determine Hg^{2+} over other cations through PET and CHEF mechanisms (Scheme 11a). After adding Hg^{2+} , the emission intensity of **23** gradually increased at 410 nm. Complex **24** exhibited a robust linear relationship between fluorescence emission and the increasing addition of the metal, affording an LOD of $7.33 \times 10^{-7} \text{ mol} \cdot \text{L}^{-1}$. The stoichiometry of complex **24**, formed between **23** and Hg^{2+} , was 1:1, and its binding constant (K_a) was estimated to be $8.97 \times 10^4 \text{ M}^{-1}$. Introducing EDTA inhibits the “off–on” response, generating suppression of fluorescence via a reversible process. Therefore, an INHIBIT MLG could be described for **23** (Scheme 11b,c).



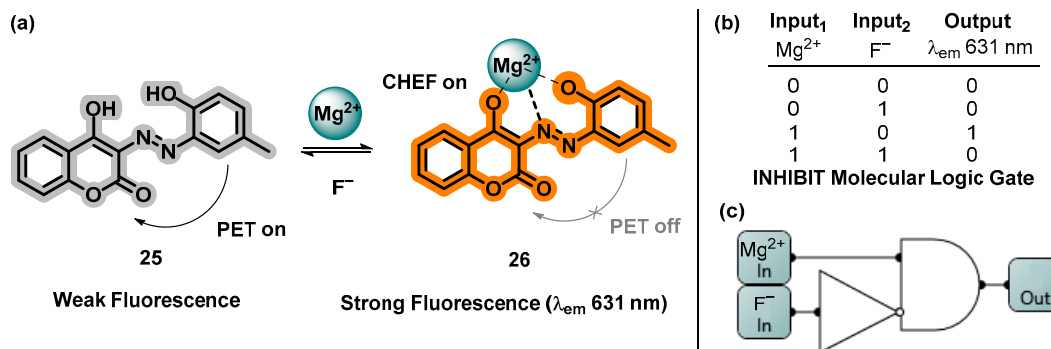
Scheme 11. Rhodanine-based MLG **23** for detecting Hg^{2+} described by Kumar et al.: (a) Sensing mechanism. (b) Diagram and (c) truth table corresponding to INHIBIT-type logic [176].

4.4. MLGs Based on Sulfides, Halides, Carbonate, and Cyanides as Input₂

Although EDTA, as interferent in the analyte–MLG interaction via a decomplexing process, is a strategic approach, other approaches lead to similar displacement assay [177–181]. For instance, some cations form insoluble salts with halides; therefore, ions such as Cl^- , Br^- , and I^- may be used as inputs in cation detection, resulting in insoluble salts and releasing the MLG. Another example involves sulfide ions to form insoluble salts, such as CuS , Ag_2S , and especially HgS . Ions, such as cyanide and carbonate, have also shown this ability to remove analytes from the analyte–MLG system. Some examples are described below.

In 2017, Sarkar et al. synthesized a novel coumarin-based azo-phenol ligand (**25**) applied to Mg^{2+} and F^- detection (Scheme 12a) [182]. Complex **25** exhibited absorbance bands at 408 nm, which decreased upon gradual addition of Mg^{2+} with the appearance of a new band centered at 502 nm. Furthermore, **25** displayed a weak emission with a quantum yield (QY) of 0.008 at 623 nm, and the gradual addition of Mg^{2+} to **25** led to fluorescence enhancement by almost 16-fold (QY: 0.116), with a slight band shifting at 631 nm as a result of **26** formation. Sequentially adding F^- to **26** reduced the fluorescence intensity, leading to the previous emission spectrum of free **25**. The authors have attributed this result to releasing sensor **25**, which operates by an “off–on–off” response, affording MgF_2 . The LODs of Mg^{2+} and F^- were calculated to be 2.4×10^{-8} and $2.3 \times 10^{-8} \text{ mol} \cdot \text{L}^{-1}$, respectively. The association constant of **25** with Mg^{2+} is $1.05 \times 10^5 \text{ M}^{-1}$, which led the authors to conclude that the complex formed was stable. Although the pH has not affected

the fluorescence intensity of **25** in the absence of Mg^{2+} , the authors have described that in the case of **26**, the emission intensity increases according to increasing pH, with maximum intensity at the pH range of 6–8. After this range, the emission was sharply reduced, attributed to the dissociation of **26** complex. In this sense, an MLG with INHIBIT logic gate can be built considering the presence of ions Mg^{2+} and F^- as input “1” and their absence as input “0” and considering the emission at 631 nm as output “1” and their lack as output “0” (Scheme 12b,c).

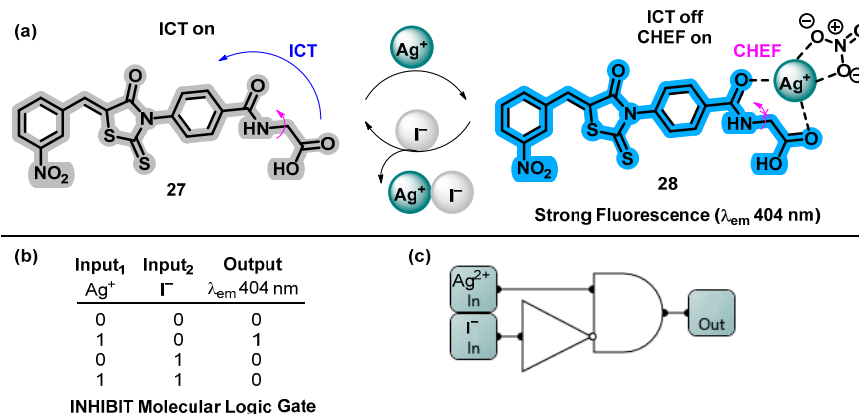


Scheme 12. MLG **25** INHIBIT-type logic described by Sarkar et al.: (a) Sensing mechanism. (b) Diagram and (c) truth table corresponding to INHIBIT-type logic [182].

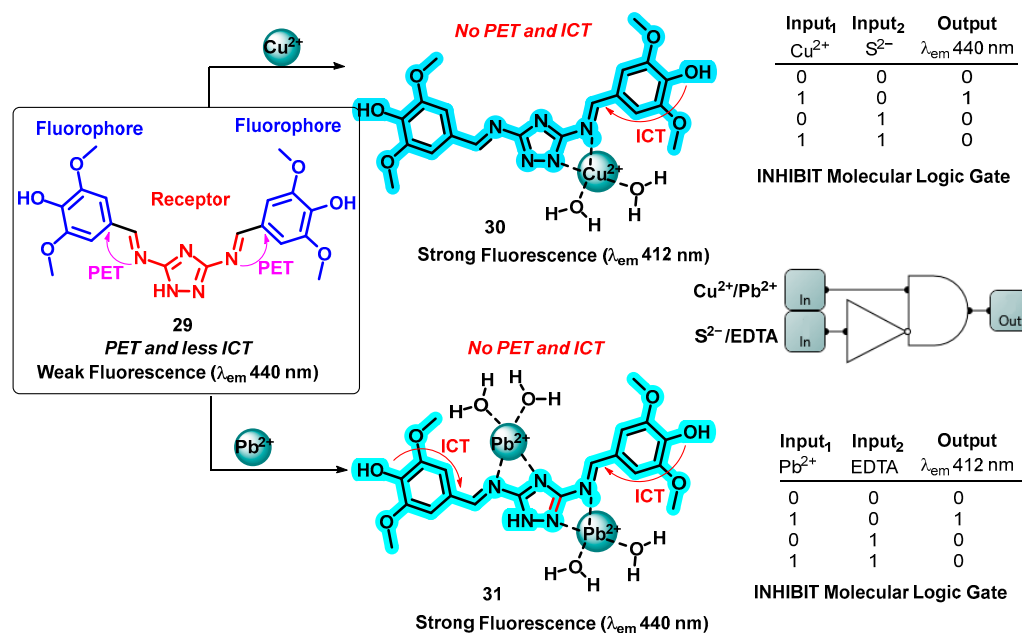
Another example of a halide-promoting decomplexation for the elaboration of an MLG was described by David et al. [183]. The authors designed the rhodanine derivative **27** to detect Ag^+ e I^- (Scheme 13a). The MLG **27** displayed a weak fluorescence at 417 nm, attributed to ICT. However, the addition of Ag^+ ion caused a 100-fold fluorescence emission enhancement at 404 nm, attributed to the increased rigid molecular conjugation via the CHEF effect with restriction of ICT phenomenon. This “turn-on” response is related to the change in fluorescence QY from 0.0013 for **27** to 0.032 for complex **28**. Complex **27** showed an LOD of $24.23 \times 10^{-7} \text{ mol} \cdot \text{L}^{-1}$, and the limit of quantification (LOQ) was calculated to be $80.77 \times 10^{-7} \text{ mol} \cdot \text{L}^{-1}$. The sequential addition of I^- to the **28** led to fluorescence quenching. This process was described as reversible and was repeated about 10 times, keeping the efficiency. For detecting I^- by system **28**, the authors have described the LOD and LOQ of $22.83 \times 10^{-7} \text{ mol} \cdot \text{L}^{-1}$ and $76.11 \times 10^{-7} \text{ mol} \cdot \text{L}^{-1}$, respectively. Job’s plot analysis, FT-IR, ^1H NMR, and theoretical calculations led the authors to propose the formation of the complex **28** and the sequential detection of I^- affording AgI with the release of the weak emissive **27** (Scheme 13). An INHIBIT logic gate was described considering the fluorescence emission of the system at 417 nm as output. Then, in the absence of inputs of Ag^+ and I^- , a weak emissive system was observed, which allowed the attribute of the output “0”. However, upon input of Ag^+ , the system displayed high emission, resulting in an output “1”. When only input of I^- was inserted, no fluorescence enhancement was observed, leading to output “0”. The presence of both inputs, Ag^+ and I^- , did not cause a fluorescence enhancement. Then, an INHIBIT-type MLG was constructed (Scheme 13b,c).

In 2019, Rout et al. described the triazole-bearing Schiff bases **29** as a colorimetric and fluorescent MLG for Cu^{2+} and Pb^{2+} (Scheme 14) [184]. The absorption spectrum of **29** shows bands at 330 and 270 nm, and the addition of Cu^{2+} caused the appearance of a new band centered at 460 nm, turning the colorless solution into a yellow solution. Upon adding Pb^{2+} , a broad band centered at 400 nm was described, turning the colorless solution into light yellow. The weak emissive (QY = 0.002) **29** solution at 440 nm was increased by 15-fold (QY = 0.03) with blue-shift to 412 nm after adding Cu^{2+} . Similarly, adding Pb^{2+} to solution **29** caused a fluorescence intensity enhancement at 440 nm of almost 17 times (QY = 0.035). Based on ESI-mass spectrometry, Job plot, FT-IR, ^1H -NMR spectra, and DFT studies, the authors have ascribed the detection mechanism involving metal coordination, which suppressed the PET, allowing the ICT process (Scheme 14). Adding S^{2-} to the emissive **30** solution led to an almost wholly quenched and sequential addition of more

Cu^{2+} , recovered again de fluorescence emission. For the emissive **31** solution, Na_2EDTA caused fluorescence quenching at 440 nm, and with the sequential addition of Pb^{2+} , the emission was recovered. These experiments indicated that the emissive **30** and **31** solutions could be reversibly restored to the free chemosensor **29** by adding S^{2-} or EDTA, respectively, showing the reversibility application of **29**.



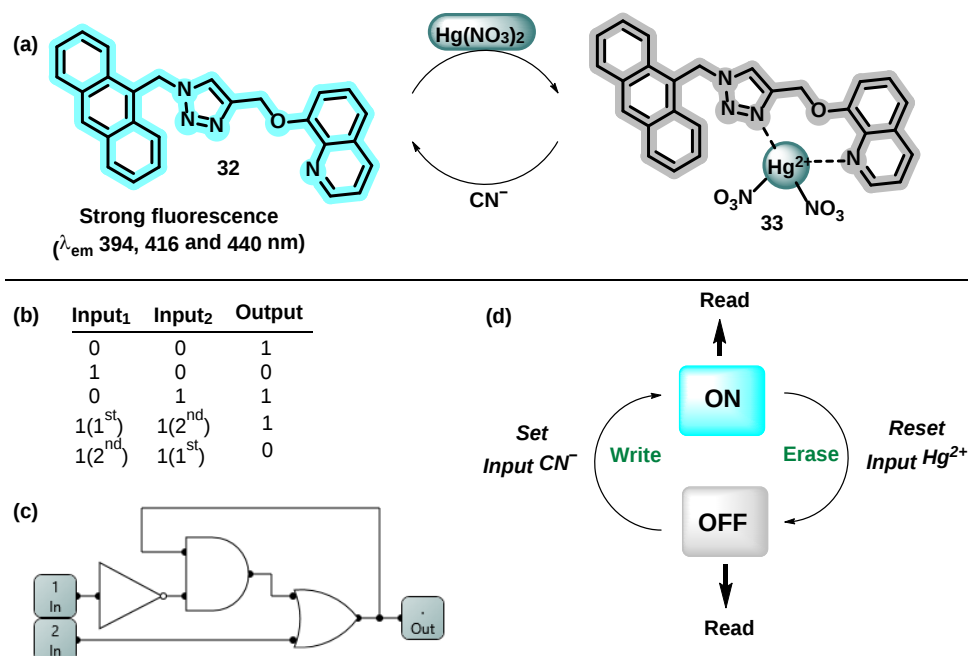
Scheme 13. (a) MLG **27** proposed by David et al. for detecting Ag^+ and, sequentially, I^- via the formation of AgI . (b) Truth table and (c) diagram representing **27** as an INHIBIT-type MLG [183].



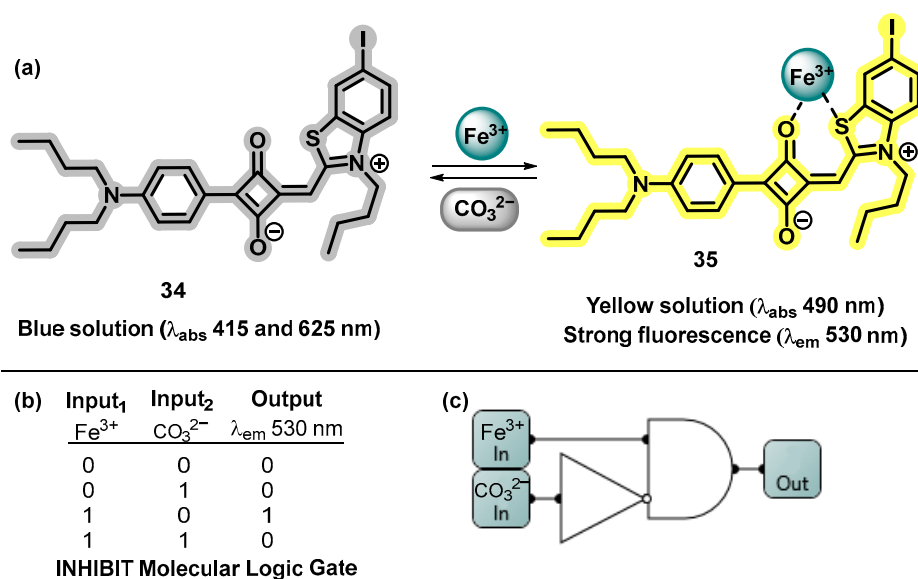
Scheme 14. MLG **29** proposed by Rout et al. for detecting Cu^{2+} , Pb^{2+} using S^{2-} and EDTA to promote reversibility. Truth table and diagram representing **29** as an INHIBIT-type MLG [184].

Considering the emission of **29** at 440 nm as output and four ionic inputs (Cu^{2+} , S^{2-} , Pb^{2+} , and Na_2EDTA), an INHIBIT logic gate was proposed. Adding Cu^{2+} induced emission of **29** at 440 nm ("on" behavior), whereas the sequential addition of S^{2-} recovered the original no-emissive solution ("off" behavior). Similarly, the INHIBIT logic gate was described considering EDTA and Pb^{2+} as chemical inputs and the emission at 412 nm as output. The "off" state of **29** (no fluorescent) was described in the absence of both inputs (Pb^{2+} and EDTA). Adding Pb^{2+} , the "on" state is observed due to the enhancement of emission intensity of **29** at 412 nm, whereas an "off" state was considered in the presence of EDTA as input. In the presence of both inputs, the "off" state of the system was achieved. Therefore, an INHIBIT logic gate could be described based on this operation (Scheme 14).

In 2017, Gupta et al. described the MLG **32**, bearing anthracene and quinoline groups connected by a triazole, for Hg^{2+} and CN^- detection (Scheme 15a) [185]. MLG **32** showed absorption bands at 350, 367, and 388 nm and strong fluorescence emission bands at 394, 416, and 440 nm. Adding Hg^{2+} led to a ~ 16 -fold decrease in the blue–green fluorescence intensity, resulting in a nonfluorescent solution with a QY changing from 0.22 to 0.012. The association constant between **32** and Hg^{2+} was calculated to be $2.99 \times 10^4 \text{ M}^{-1}$ and LOD of $19.2 \text{ nmol} \cdot \text{L}^{-1}$. The sequential addition of CN^- ion to a nonemissive solution of **33** recovered the fluorescence emission with a naked-eye color change from violet to a visible fluorescent blue–green color. The high binding constant of $1.8 \times 10^4 \text{ M}^{-1}$ suggested a demetallation process and displacement of Hg^{2+} from its **33** complex, resulting in $\text{Hg}(\text{CN})_2$ formation. The LOD of CN^- by **33** was calculated to be $1.25 \text{ } \mu\text{mol} \cdot \text{L}^{-1}$. Considering Hg^{2+} and CN^- as inputs and the fluorescence emission as 416 nm as output, **29** was described as an integrated and sequential logic circuit. Therefore, the truth table and its respective diagram in Scheme 15b,c show that the emissive **32** is quenched upon adding Hg^{2+} , whereas the addition of only CN^- caused no significant emission modification. When the **32** solution was first treated with Hg^{2+} and then with CN^- , the fluorescence was not significantly altered either. However, the fluorescence was quenched in reverse sequence: CN^- and then Hg^{2+} . Although the authors did not classify this MLG, it was presented as “write–read–erase–read” in a reversible and reconfigurable system (Scheme 15d). Therefore, considering **32** as a memory device with inputs set (Input CN^-) and reset (Input Hg^{2+}) based on sequential logic operations mimics the function of a memory element. In addition, the reversible and reconfigurable sequences of the set/reset logic operations show a memory similar to “write–read–erase–read” functions. In this sense, the reset input (Input Hg^{2+} “1”) caused a fluorescence quenching and encoded information in the system as “read” and “erased”, and the logic operation is saved as Output “0”. Then, the stored information within the system affords a signal “write” by the set input (In CN^- “1”) with a fluorescence enhancement, and the logic operation is saved as output “1”. This cycle was repeated eight times with no significant change in the relative emission intensity of **32**.



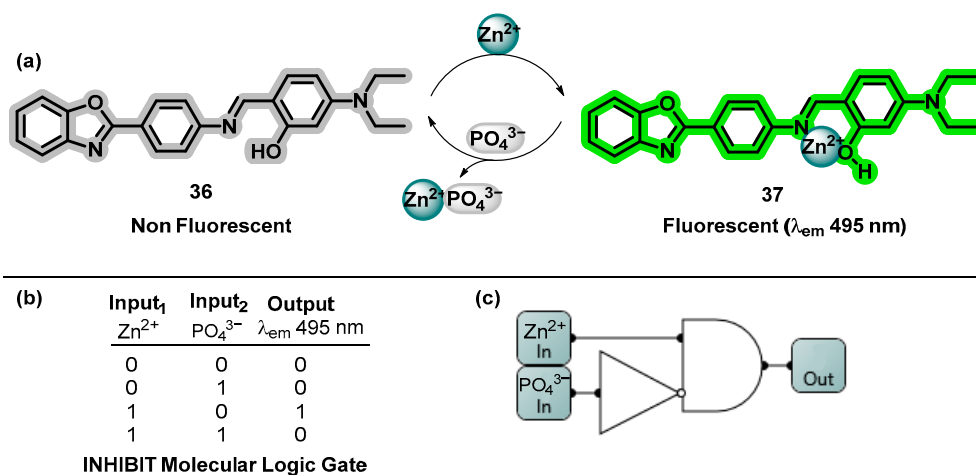
Scheme 15. MLG **32** reported by Gupta et al. for sensing Hg^{2+} and cyanide: (a) Sensing mechanism, (b) the truth table, (c) the corresponding diagram, and (d) the reversible cycle of the “write–erase” system [185].



Scheme 16. (a) MLG **34** proposed by Liu et al. for detecting Fe^{3+} and, sequentially, CO_3^{2-} via removal of the first analyte. (b) Truth table and (c) diagram representing **34** as an INHIBIT-type MLG [186].

4.5. MLGs Based on Phosphate and Derivatives as Input₂

Different analytes containing phosphorus derivatives have been applied as a second input in the description of MLGs. For example, Mahato et al. recently described benzoxazole **36** as an MLG for Zn^{2+} and phosphate (PO_4^{3-}) detection (Scheme 17a) [187]. The weak fluorescence emission of **36** at 495 nm was enhanced upon adding Zn^{2+} . This phenomenon was attributed to the restricted rotation of the N,N-diethylsalicylimine group due to the chelation with Zn^{2+} intensifying the emission 42 times. The reversibility of MLG **36** to detect Zn^{2+} was also investigated upon adding different ions to **37** solution. Adding PO_4^{3-} to the highly emissive **37** solution resulted in fluorescence suppression. This optical response was ascribed to the higher reactivity of PO_4^{3-} with Zn^{2+} , forming a more stable species than **37** and then releasing the nonemissive **36**. The authors have described five cycles of this sequence with minimum changes in the emission process. Finally, considering the presence of Zn^{2+} and PO_4^{3-} as input “1” and their absence as input “0”, the emission at 495 nm as an output “1”, and the no emissive system as output “0”, an INHIBIT logic gate for **14** was described (Scheme 17b,c).



Scheme 17. (a) MLG **36** proposed by Mahato et al. for detecting Zn^{2+} and, sequentially, PO_4^{3-} via removal of the first analyte. (b) Truth table and (c) diagram representing **36** as an INHIBIT-type MLG [187].

In 2019, Jain et al. described the triazole-appended pyrano[2,3-*c*]pyrazolone and triazole-based sensors **38** and **39** for recognition of Fe^{3+} and H_2PO_4^- (Scheme 18a,b) [188]. Sensor **39** exhibits a pincer cavity based on the triazole and pyranopyrazole groups, and **38** is a half-synthetic example similar to the first one. Adding Fe^{3+} to **38** or **39** turns colorless solutions into **40** and **41** pale yellow systems. These optical responses were associated with modifying a broad band in 280–350 nm to 350–450 nm. By analyzing the titration curve, the LOD of Fe^{3+} was found to be 7.0 and 3.0 $\mu\text{mol}\cdot\text{L}^{-1}$ by **38** and **39**, respectively. Binding constants for the formation of **40** and **41** were also estimated to be 5.9×10^2 and $1.27 \times 10^3 \text{ M}^{-1}$, respectively.

Adding different anions to the solution of both sensors did not cause significant changes in their optical properties. However, only adding H_2PO_4^- to **40** and **41** solutions decreased the absorbance intensity at 380 nm, turning the yellow solution into a colorless solution, as observed by the naked eye. The sequential addition of an excess of Fe^{3+} turns the solution yellow again. This process was repeated in 4 cycles, suggesting the reversibility of the sensor. These results are due to the high affinity of Fe^{3+} for H_2PO_4^- , which promoted the release of **39** from the complex **41**. The ^1H NMR, FT-IR, HRMS analysis, and DFT calculation of complexes led the authors to propose the sensing mechanism presented in Scheme 18a,b. In this sense, considering the presence of Fe^{3+} and H_2PO_4^- as input “1” and their absence as input “0”, as well as the absorbance band at 380 nm as output “1” and their lack as output “0”, sensor **39** was described as an INHIBIT-type logic gate (Scheme 18c). The authors also have described this molecular system based on logic operations for the memory element via “writing–reading–erasing–reading” functions (Scheme 18e), similarly to Gupta et al. (Scheme 15a) [185]. In this description, the input of Fe^{3+} is in the absence of H_2PO_4^- (input “0”), and the system writes and memorizes binary state “1”. However, considering both inputs present, state “1” is erased, and the “0” state is written and memorized. Therefore, a reversible and reproducible sequence of logic operations can be described.

Recently, Megha et al. synthesized a julolidine–hydrazone-based chemosensor (**42**) for the sequential detection of Zn^{2+} and PPI (Scheme 19a) [189]. When an aqueous solution of Zn^{2+} was gradually added to solution **42**, the ICT band at 390 nm exhibited a red-shift to 425 nm in UV–Vis spectra. Zn^{2+} resulted in a remarkable enhancement of emission, characterized by the appearance of a bathochromically shifted and intense band at 610 nm, visually observed through fluorescent orange emission. This “turn-on” behavior was attributed to the CHEF process, which restricts the ICT and ESIPT and limits the movement of the molecule. Zn^{2+} complexes are known to be good candidates for constructing sensors sensitive to PPI [73,190–193]; however, interferences from other structurally similar phosphates, such as ADP and ATP, pose a challenge. In this sense, **44** detected PPI selectively through metal displacement, reaching an LOD of 1.7 $\mu\text{mol}\cdot\text{L}^{-1}$. The experiment was used to construct an INHIBIT-type logic gate, using Zn^{2+} and PPI as two inputs and emission at 610 nm as the output (Scheme 19b,c).

4.6. MLGs Based on Other Anions as Input₂

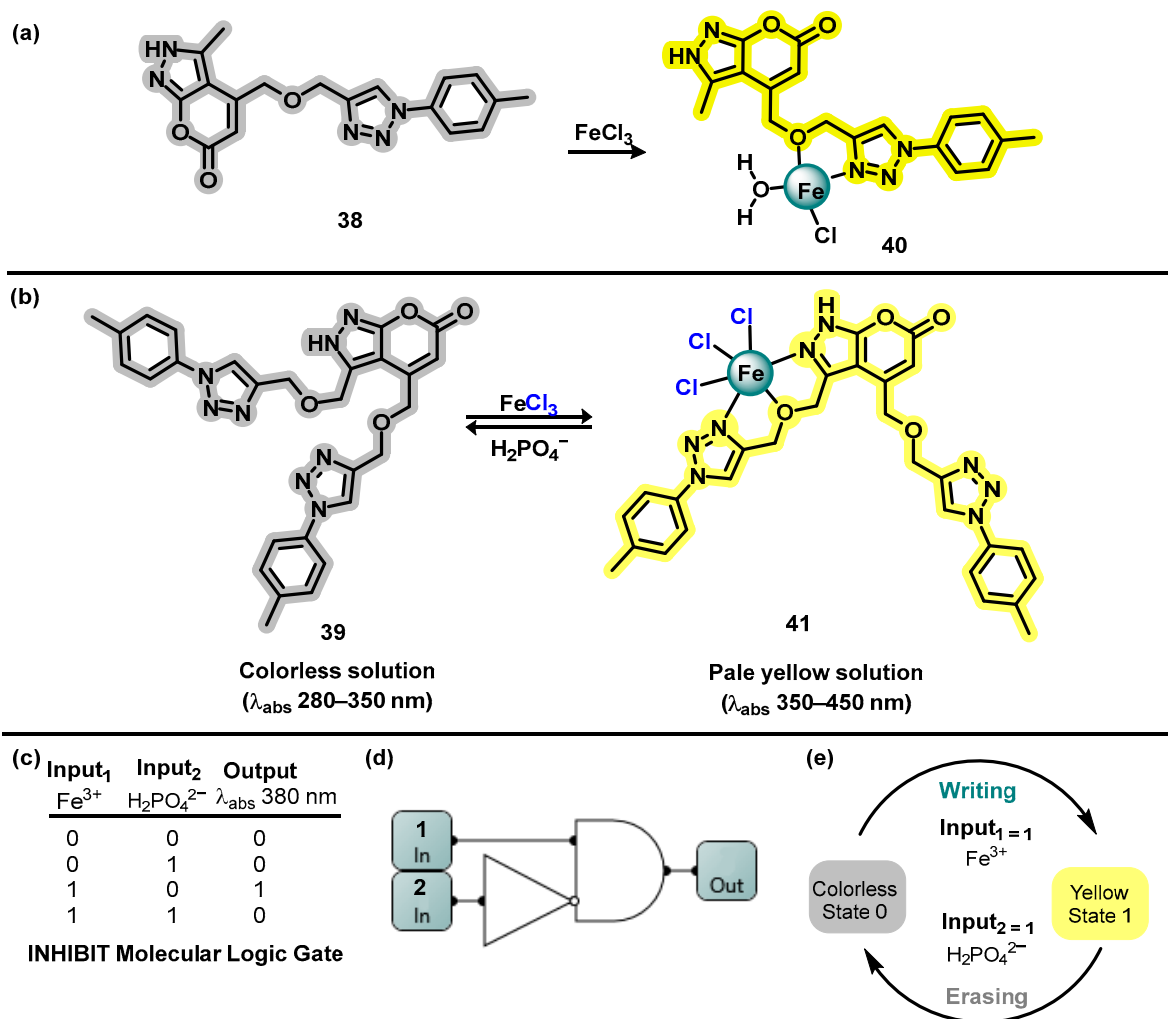
In 2015, Tavallali et al. described the MLG **46** based on rhodamine B for detecting Mo^{6+} using the displacement strategy by citrate anion (cit^{3-}) (Scheme 20) [194]. MLG **46** displayed an absorption band at 558 nm, which was reduced in the presence of Mo^{6+} ions in H_2O . Furthermore, two isosbestic points emerged at 575 and 502 nm, strongly indicating the presence of multiple species in the medium. The molar ratio found 2:1 (**47**) showed an association constant calculated equal to $7.01 \times 10^4 (\text{L}\cdot\text{mol}^{-1})^2$. This optical response was attributed to the formation of the chelated **47** (Scheme 20a). Then, the authors explored the ability of the complex **47** to detect cit^{3-} in aqueous solution via UV–Vis absorption spectroscopy. Adding cit^{3-} to the solution, the absorbance at 558 nm gradually increased. The robust coordination of MoO_4^{2-} with cit^{3-} triggers the alteration in the color of the solution from purple to pink. The stability constants for cit^{3-} and MoO_4^{2-} in the literature are $\log \beta_{(1,1,2)} = 15.00$ and $\log \beta_{(2,2,4)} = 31.02$ [195]. These constants are bigger than the

binding association constant between **46** and MoO_4^{2-} ; consequently, the complex between MoO_4^{2-} and cit^{3-} is formed preferentially (**48**), releasing **46** in its free form and recovering the original absorption band at 558 nm.

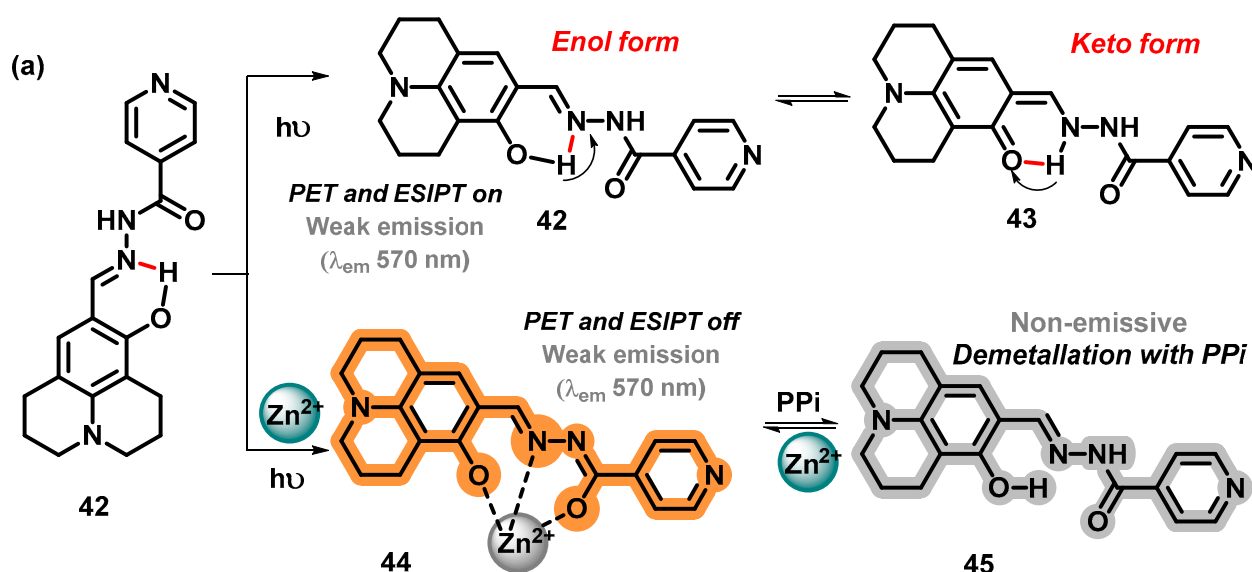
Considering Mo^{6+} and cit^{3-} as inputs (inputs “1”) or their absence (inputs “0”), the intense absorption at 558 nm as output “1” or the weak absorption band at 558 nm as output “0”, the IMPLICATION logic gate **46** was described (Scheme 20b,c).

4.7. MLGs Based on Neutral Molecules as Input₂

In many cases, the identity of the cation (input) that leads to a selective MLG response is fundamental in choosing the second analyte once some cations may have an affinity for specific molecular groups. In 2018, Manna et al. described the chemosensor N-(2-hydroxybenzylidene)-2-(benzamido)benzohydrazide (**49**) for Ni^{2+} and Cu^{2+} detection via colorimetric and fluorometric methods in aqueous medium (Scheme 21) [196]. Adding Ni^{2+} to a colorless **49** solution resulted in a new absorption band at 404 with a color change to yellow. Similarly, adding Cu^{2+} to **49** led to a new band at 394 nm, turning the colorless solution into an intense yellow. In adding Cu^{2+} to **50** solution (**49** + Ni^{2+}), a similar absorbance band of **51** solution (**49** + Cu^{2+}) was described. Therefore, the authors suggested that Cu^{2+} replaces Ni^{2+} .



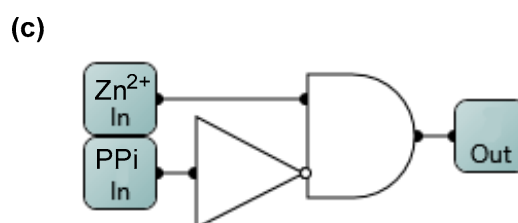
Scheme 18. Compounds **38** and **39** proposed by Jain et al. for detecting Fe^{3+} and, sequentially, $\text{H}_2\text{PO}_4^{2-}$: (a) Sensing mechanism of **38** and Fe^{3+} . (b) Sensing mechanism of **39** and Fe^{3+} . (c) Truth table, (d) the correspondent diagram INHIBIT-type MLG, and (e) writing–erasing cycle **39** representation [188].



(b)

Input ₁	Input ₂	Output
Zn ²⁺	PPI	λ_{em} 570 nm
0	0	0
0	1	0
1	0	1
1	1	0

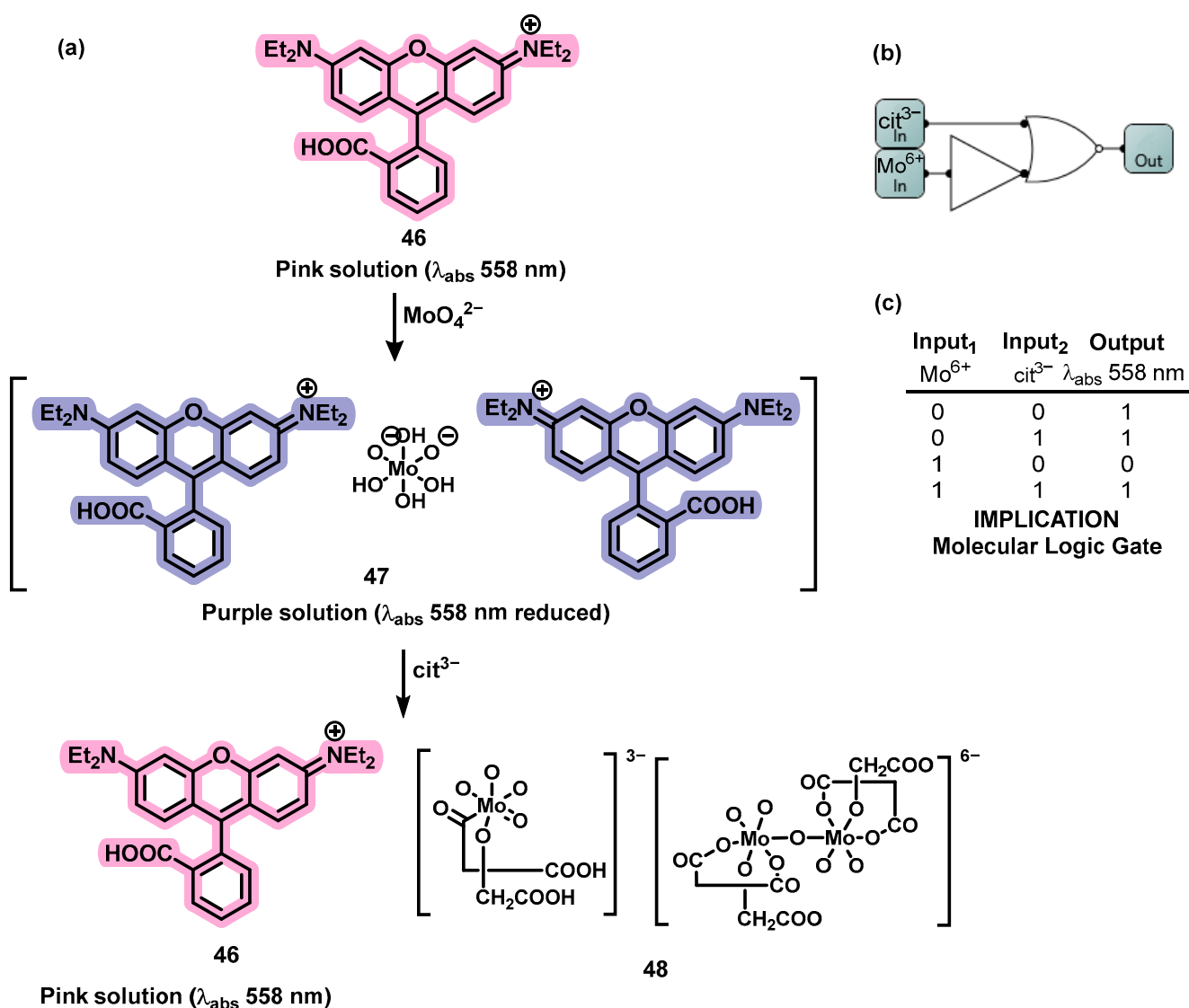
INHIBIT Molecular Logic Gate



Scheme 19. (a) MLG **42** proposed by Megha et al. for detecting Zn²⁺ and, sequentially, PPI. (b) Truth table and (c) diagram representing **42** as an INHIBIT-type MLG [189].

Furthermore, **51** displays a very weak emission at 451 nm when excited at 332 nm, attributed to the PET from the imine “N” to the excited fluorophore salicylazine group and a possible C=N isomerization process (Scheme 21). The authors have also described a small ESIPT process in polar protic solvent, resulting in a small hump at 490 nm in the free **49** and a low cation concentration. Nevertheless, adding Cu²⁺ and Ni²⁺ resulted in a fluorescence enhancement associated with suppressing the PET process after effectively coordinating imine and OH groups. This phenomenon prevents the ESIPT process and produces a more rigid system, promoting a strong CHEF.

Adding Na₂EDTA to solutions **51** or **50** turns the yellow system into a colorless solution with the disappearance of the absorption band at ~400 nm and the appearance of an intense peak around 332 nm. The sequential addition of Cu²⁺ or Ni²⁺ recovered the yellow color of the solutions. Furthermore, the emissive system **51** or **50** was quenched upon adding Na₂EDTA and, once again, became emissive upon the sequential addition of Cu²⁺ or Ni²⁺. The ability of the Cu²⁺ or Ni²⁺ complex with Na₂EDTA associated with these optical responses indicated the reversibility feature of the MLGs observed even after several cycles. In solution **51**, cysteine, which works similarly to Na₂EDTA, can detect Ni²⁺ in the presence of Cu²⁺ once **50** is not affected. The emissive **51** was quenched upon the addition of cysteine.

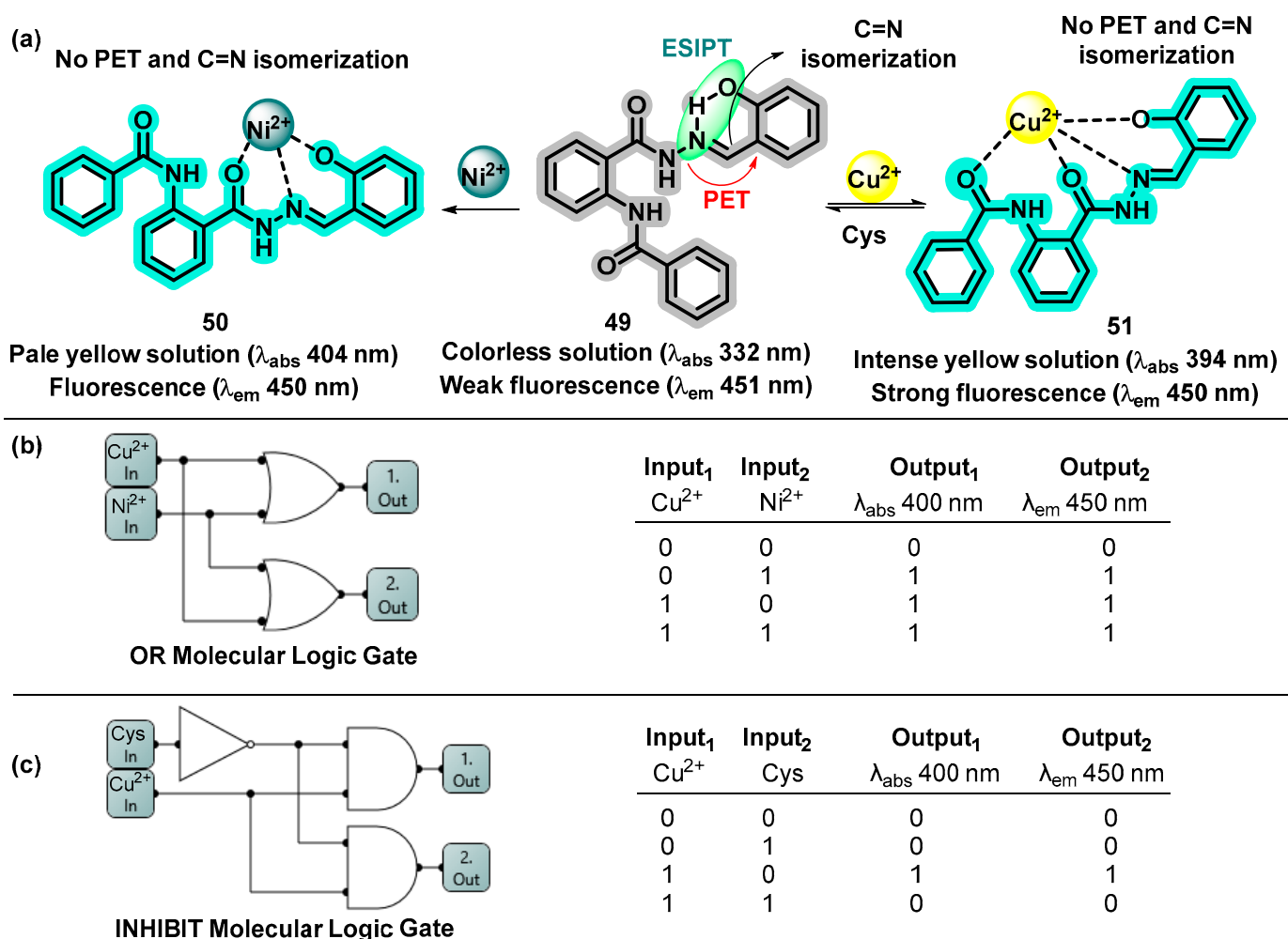


Scheme 20. (a) MLG 46 proposed by Tavallali et al. for detecting Mo^{6+} and, sequentially, citrate anion, (b) diagram, and (c) truth table representing 46 as an IMPLICATION MLG [194].

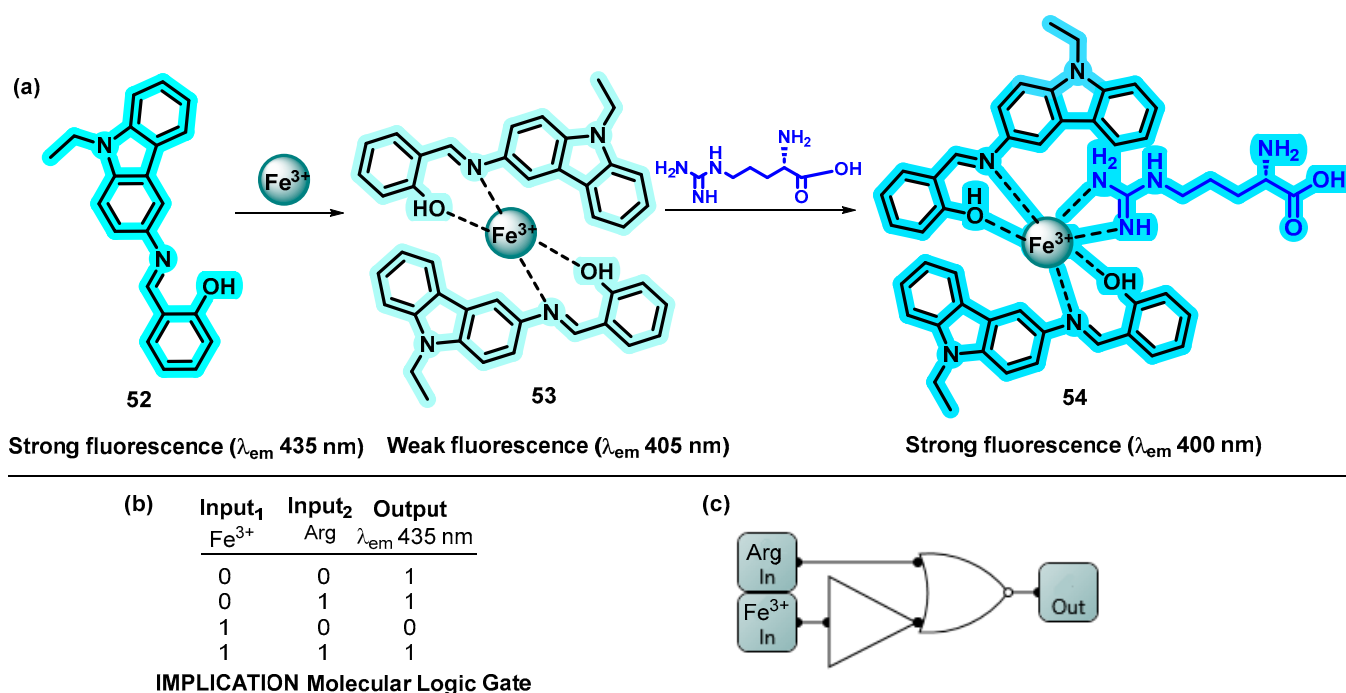
Based on these findings, 49 could be described as an MLG considering the fluorescence enhancement around 450 nm, absorption band around 400 nm as output, and $\text{Cu}^{2+}/\text{Ni}^{2+}$ /cysteine as inputs (Scheme 21b,c). In the absence of $\text{Cu}^{2+}/\text{Ni}^{2+}$ (input "0"), no absorption was observed at 450 nm (output "0"). However, the separated or simultaneous addition of $\text{Cu}^{2+}/\text{Ni}^{2+}$ caused an intense emission at 450 nm, which led the authors to propose the OR-type logic gate. For Cu^{2+} /cysteine association, the INHIBIT logic function was applied for both absorption and emission according to the effect of cysteine in the 51 system previously described.

In 2022, David et al. described the carbazole derivative 52 as a fluorescent MLG to detect Fe^{3+} and the amino acid arginine (Arg) (Scheme 22) [197]. Compound 52 displayed an intense emission at 435 nm, sharply reduced with a blue-shift to 405 nm upon adding Fe^{3+} . Cu^{2+} also caused some suppression in fluorescence but not enough to influence the selectivity of Fe^{3+} . The LOD was determined to be $12.22 \text{ nmol} \cdot \text{L}^{-1}$. The sequential addition of Arg to solution 53 (52 + Fe^{3+}) recovered the fluorescence emission at 400 nm, whereas other aminoacids (Ala, Asp, Asp-A, Cys, Cyst, Glu, Glu-A, His, IsoL, Leu, Lys, Met, Phe, Ala, Pro, Ser, Thr, Try, Tyr, and Val) did not show a similar effect. The binding mechanism proposed by the authors (Scheme 22) involves the formation of complex 53,

resulting in fluorescence suppression due to ICT and LMCT processes. The sequential coordination of Arg to Fe^{3+} in complex **53** led to a stable octahedral complex and induced a substantial fluorescence enhancement. The colorless solution **53** turned brownish after adding Arg (**54**). Considering the strong emission at 435 nm and 400 nm as output “1” and their absence as output “0”, as well as Fe^{3+} and Arg as inputs, an MLG can be drawn. Compound **52** showed a strong fluorescence emission ($\lambda_{\text{em}} = 435 \text{ nm}$ —output “1”) in the absence of Fe^{3+} and Arg (inputs “0”). Adding Fe^{3+} (input “1”), in the absence of Arg (input₂ “0”) caused a fluorescence suppression (output “0”), whereas only the input of Arg caused no significant modification on the emission of **52** ($\lambda_{\text{em}} = 435 \text{ nm}$ output 1). In the case of adding both analytes (input “1”), the fluorescence emission ($\lambda_{\text{em}} = 400 \text{ nm}$ —output “1”) was recorded. Therefore, an IMPLICATION-type MLG could be described for this system (Scheme 22b,c). Interestingly, this example does not involve the replacement of the first analyte detected nor a reversible process, contrary to most of those described here and in the literature in this segment. What happens is that the product formed in the detection of Fe^{3+} (first detection) is, actually, a sensor for detecting Arg through coordination with the metal (analyte) detected.



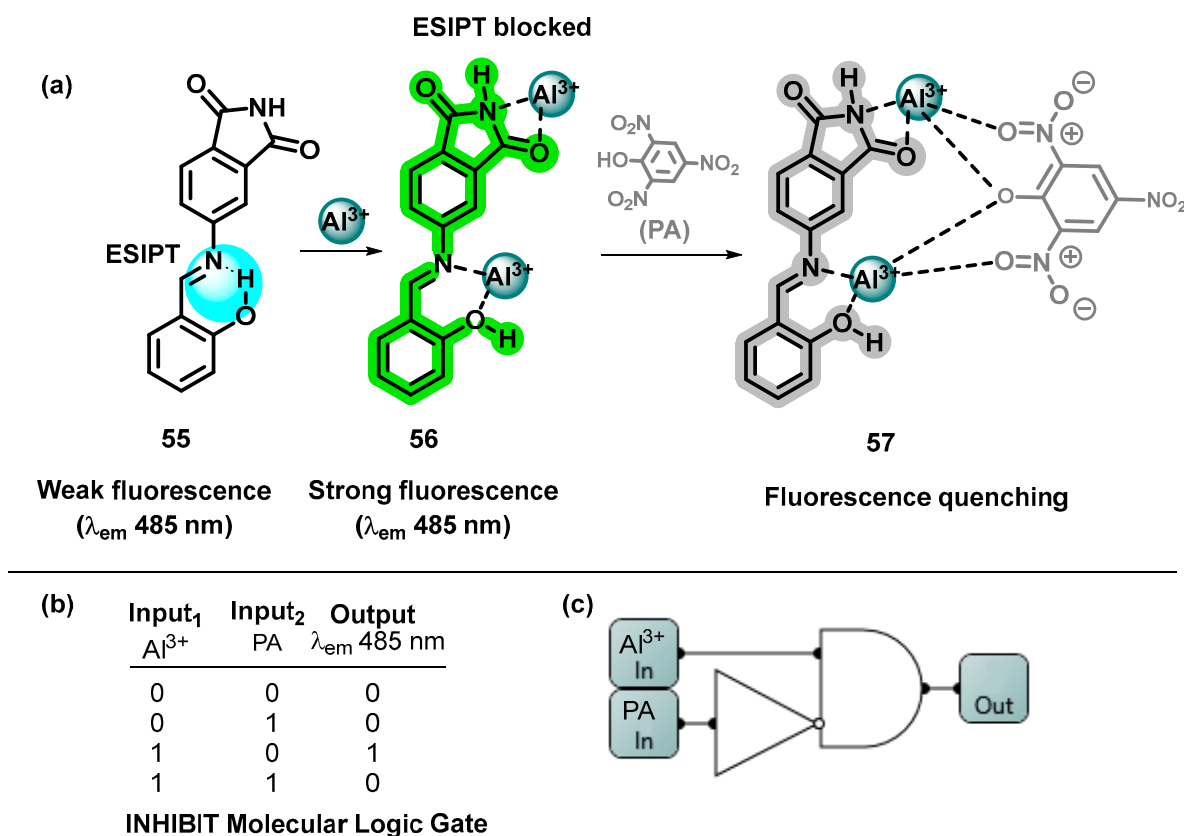
Scheme 21. (a) MLG **49** reported by Manna et al. (b) Interpretation of OR and (c) INHIBIT logic gates for detecting Cu^{2+} and Ni^{2+} , and Cu^{2+} and Cys, respectively, with the correspondent truth tables [196].



Scheme 22. (a) MLG 52 proposed by David et al. for detecting Fe³⁺ and, sequentially, amino acid arginine (Arg). (b) Truth table and (c) diagram representing 52 as an IMPLICATION-type MLG [197].

Recently, Rahman et al. described the 4-aminophthalimide derivative **55** (Scheme 23) for the detection of Al³⁺ and picric acid (PA) [198]. The absorption spectra of **55** displayed two broad bands near about 280 and 350 nm, which were decreased and shifted to 314 and 365 nm, respectively, due to the addition of Al³⁺. The authors have attributed this optical response to the coordination of **55** with this cation through the hydroxyl of the salicylimine group and the nitrogen of imine, increasing the planarity and conjugation of the molecular system (**56**, Scheme 23a). However, no color change was observed. Furthermore, adding Al³⁺ increased fluorescence emission 35 times at 485 nm with an LOD of Al³⁺ calculated to be 0.59 $\mu\text{mol}\cdot\text{L}^{-1}$. The authors attributed the weak fluorescence emission of **55** to the ESIPT process, and the coordination with Al³⁺ blocks as well, increasing the fluorescence emission. This optical response was associated with a color change from a less intense greenish to a highly intense greenish emission in **56**. The sequential addition of PA to the highly emissive **56** solution led to a fluorescence quenching due to the formation of **57**. The interaction affinity was estimated to be $1.3 \times 10^4 \text{ M}^{-1}$, and **56** showed an LOD of PA equal to 0.43 $\mu\text{mol}\cdot\text{L}^{-1}$. Based on these optical results, an INHIBIT MLG was described, considering Al³⁺ and PA as chemical inputs and the fluorescence emission at 485 nm as output (Scheme 23b). The output “0” was considered in the absence of the inputs Al³⁺ and PA. Only an input of Al³⁺ caused the enhancement of the emission, output “1”, whereas only an input of PA did not increase the fluorescence of the system (output “0”). When both inputs were added, no emission was observed (output “0”) either. Therefore, **55** could be described as an INHIBIT logic gate operator (Scheme 23c).

Similarly to the previous case (**52**) [197], the system formed after detecting Al³⁺ can detect PA based on the additional/sequential interaction with the detected cation. These examples show that the second analyte does not always remove the initially identified cation. This strategy of the metal (analyte) to coordinate with the product of the first detection opens up an avenue of possibilities for MLG development by exploiting the cation’s coordination modes. Moreover, this example is a case of INHIBIT-type MLG that did not involve the removal of the cation from the previously formed complex but a sequential coordination on the metal of the complex.



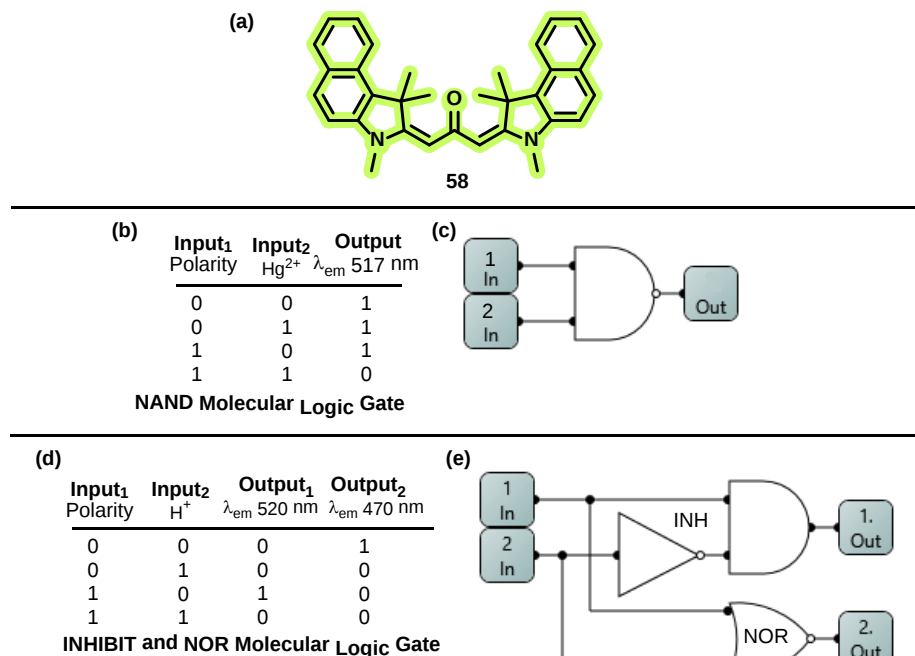
Scheme 23. (a) MLG 55 proposed by Rahman et al. for detecting Al^{3+} and, sequentially, picric acid (PA). (b) Truth table and (c) diagram representing 55 as an INHIBIT-type MLG [198].

5. Strategies for MLGs Based on Solvent Effects

The solvent plays a critical role in the effectiveness of the sensing process, because it can affect the solubility, stability, interaction, and diffusion of the analyte, sensor, and species generated [199–203]. Therefore, the solvent polarity affects the selectivity and sensitivity of the sensor by either facilitating or impeding the recognition of the analyte, making it one of the most important factors to consider in MLG planning [204]. Another important aspect is the solvent's viscosity, which can hinder the diffusion of the target molecule towards the sensor, potentially leading to delayed responses or limitations in real-time detection or even interfering with aggregation phenomena [106,205–208]. Furthermore, the environment's acidity can interfere with the most stable conformation of the optical device and the pathway of a detection mechanism [209,210].

In this sense, Zhang et al. presented an interesting system capable of operating with three different logic functions, namely NAND, NOR, and INHIBIT, in response to solvent polarity, proton, and Hg^{2+} inputs [211]. The authors designed a merocyanine dye, **58** (Scheme 24), which has a carbonyl bearing two benzo[e]indoline fragments at both ends and nitrogen atoms that can either be protonated or act as a quencher agent by PET. Merocyanine dyes have been extensively studied because of their solvatochromic behavior [202,212–217]. Experiments of binary mixtures of THF and water were controlled. Adding a highly polar solvent, such as water, to a strongly nonpolar medium, such as THF, the fluorescence emission is strongly suppressed with a shift from blue (95% THF) to green (5% THF). Furthermore, the isoemissive point at 508 nm can be interpreted as two related emissive species, as one of the proofs for exciplex formation with a polar solvent. To analyze the effect of pH on the optical properties of **58**, a 50% THF solution of the compound was submitted to titration with hydrogen ions (H^+), using $HClO_4$ as the acid. Following the addition of H^+ , the initially yellow solution of **58** changed to a colorless state, with the disappearance of the absorption band at 441 nm and the appearance of a

discrete band at 393 nm. Upon reaching pH 2, the emission is wholly suppressed due to the protonation of nitrogen atoms that cause the dissociation of the entire conjugated system of the molecule. The **58** solution is also efficient in detecting Hg^{2+} by deactivating channels surrounding the metal orbital by coordinating the **58** receptor unit.



Scheme 24. (a) MLG **58** proposed by Zhang et al. (b) Truth table and (c) diagram of NAND-type MLG considering polarity of solvents and Hg^{2+} as inputs. (d) Truth table and (e) diagram of INHIBIT- and NOR-type MLG considering polarity and H^+ as inputs [211].

The authors have used these three possibilities to build logic gates by pairs of stimuli. When analyzing the polarity of the solvent and the addition of Hg^{2+} as inputs, they considered “1” as the input of high polarity (5% THF: 95% H_2O) and “0” as the input of low polarity (50% THF: 50% H_2O), limited due to the low solubility of **22** in THF. They considered the presence of 3 equiv. of Hg^{2+} as “1” and the absence of metal as “0”. Fluorescence emission at 520 nm was identified as output. With these considerations, the authors interpreted the system as a logic gate of the NAND type (Scheme 24b,c).

The second interpretation considers the polarity–protonation pair as input and two emissive bands, 470 and 520 nm, as outputs. In low-polarity solvent (95% THF: 5% H_2O , input₂ “0”) and at pH 8 (input₂ “0”), **58** only shows emission at 470 nm (output₁ “1”; output₂ “0”). When H^+ is added to the solution (pH 2, Input₂ “1”), the protonation of **58** and extinction of fluorescence emission occurs, resulting in both outputs being “0”. However, in highly polar solvent (5% THF: 95% H_2O) (input₁ “1”), **58** exhibits emission at 520 nm (Output₂ “1”), corresponding to a NOR-type gate at 470 nm and INHIBIT-type gate at 520 nm (Scheme 24d,e).

The aggregation of sensor molecules is a frequent phenomenon in the development of optical devices. Aggregation is associated with the ability of solvent molecules to interact preferably with each other rather than with the chemosensor, causing it to aggregate. These interactions are related to the conformation that these molecules assume according to the conditions of the medium. Aggregate formation can result in changes in the optical properties, leading to aggregation-induced quenching (AIQ) or aggregation-induced emission (AIE) [107,208]. Das et al. developed the fluorimetric chemosensor **59**, which operates via aggregation and induces emission (AIE) to Al^{3+} and Zn^{2+} detection (Scheme 25) [201]. The behavior of **59** was investigated in different organic–aqueous media such as MeOH, EtOH, MeCN, DMF, DMSO, and THF with $\geq 70\%$ water fraction. The chemosensor **59** exhibits a

fluorescence sensing behavior capable of discriminating Al^{3+} from Zn^{2+} when the solvent is changed from polar protic solvents (such as MeOH and EtOH) to polar aprotic solvents (such as DMF and DMSO).

In DMSO, **59** is ordered in a six-membered ring structure favorable to the ESIPT mechanism via intramolecular H-bonding (**62**). In a polar protic medium, such as MeOH, there is a disturbance in this H-bond, suppressing the ESIPT. The nonemissive behavior of the solution is attributed to the PET mechanism of electron density donation of the N electron pair of the C=N bond (**60**). When a small-sized and highly charged cations as Al^{3+} is added to a MeOH media, PET is disrupted due to complexation with the metal occurring in the sensor structure with the influence of the hydroxyl electron pair of the solvent (**61**). Consequently, the absorption intensity at 390 nm decreases, while a consistent increase in absorption in the 450 nm region becomes evident. In fluorimetric analysis, **59** in MeOH demonstrates remarkable selectivity towards Al^{3+} with emission at 532 nm.

On the other hand, the chemosensor can complex Zn^{2+} in a polar aprotic medium, inhibiting PET and ESIPT (**63**). Upon introducing Zn^{2+} in DMSO, a notable red-shift ($\Delta\lambda = 63$ nm) of the absorption band at 454 nm was observed, concomitant with a reduction in the band at 391 nm. Fluorescence examination exhibited a relatively weak fluorescence emission band centered around 509 nm in the absence of metal. However, with the Zn^{2+} addition, a significant enhancement in fluorescence was observed at 527 nm.

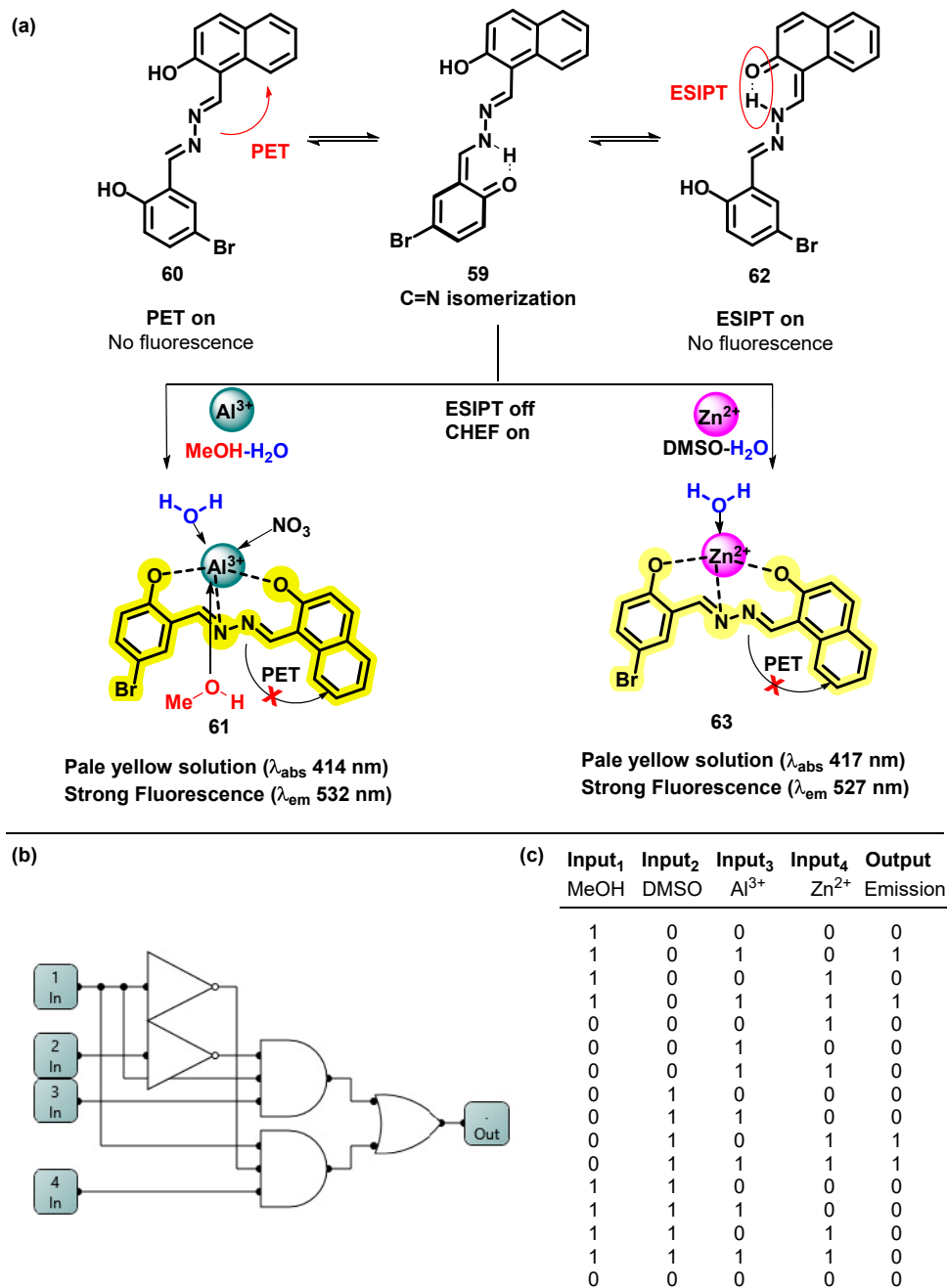
By combining four chemical entities, MeOH, DMSO, Al^{3+} , and Zn^{2+} , for the **59** sensor as inputs and using the emission intensity at 527 nm (for Al^{3+}) or 532 nm (for Zn^{2+}) as outputs, the authors identified diverse logic gates systems, as represented in Scheme 25b,c.

In 2020, Eördögh et al. developed the fluorogenic MLG **64**, capable of visualizing lipid domains (Scheme 26) [218]. MLG **64**, a diazoindanone spirofused to a silicon-containing rhodamine derivative, was planned based on its ability to undergo a photoinduced Wolff rearrangement to afford **65** or **67**, depending on the substrate and reaction conditions. In this sense, the authors investigated the behavior of **64** in various media, particularly nonpolar and polar environments (Scheme 26b). Solutions containing mixtures of **64** with primary alcohols were irradiated at 405 nm. The fluorescence emission intensity is enhanced with an increase in alcohol proportion. Subsequently, experiments were conducted with mixtures of secondary alcohols of similar polarity and water, reducing the fluorescence emission compared with the intensity achieved in experiments with primary alcohols. Then, no fluorescence emission was observed in dioxane, suggesting reduced nucleophilicity might be implicated in the fluorescence mechanism. Therefore, the intermediate **65** was achieved in apolar media, followed by the rapid formation of the fluorescent product **66** through a nucleophilic reaction. The authors have studied the alternative route involving the carbene intermediate **67** via DFT, and product **68** was subsequently confirmed through X-ray crystallography.

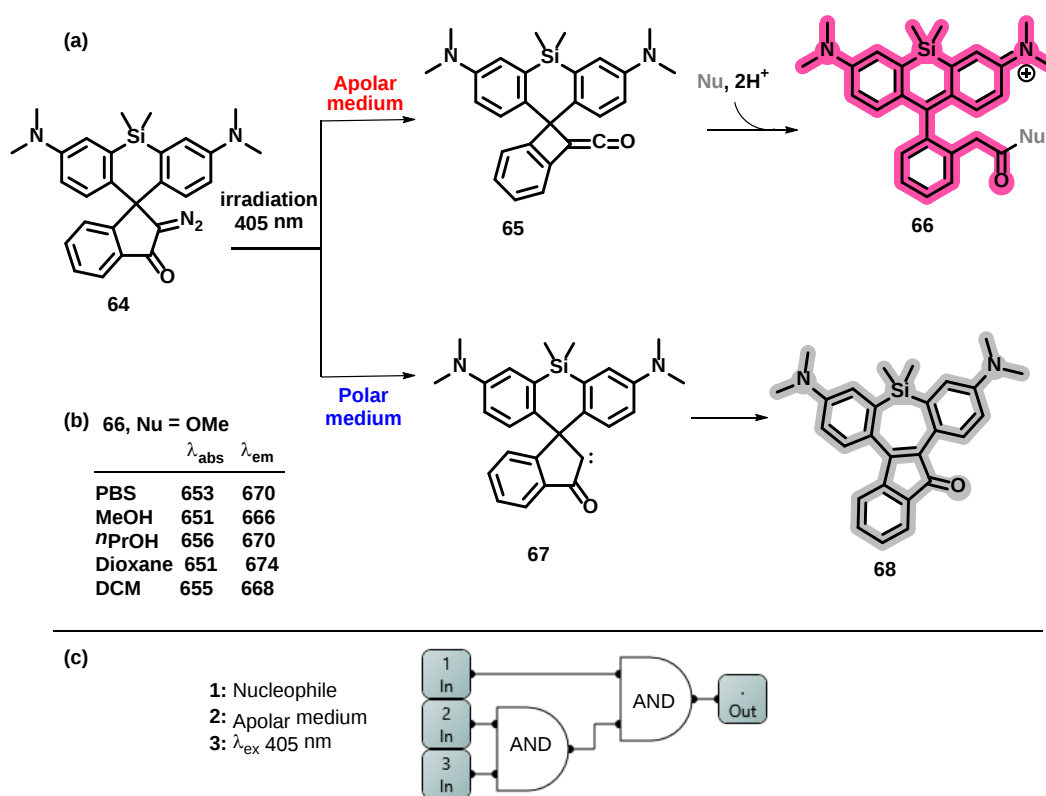
Concerning MLG, irradiation at 405 nm and an apolar media afforded the intermediate **65**, which produced an output fluorescence emission in the presence of a nucleophile, leading to **66**. From a mechanistic perspective, the fluorescent product **66** indicates a preference for the concerted mechanism of the Wolff rearrangement over the stepwise pathway and a double AND-type gate interpretation, as represented in the diagram in Scheme 26c.

In 2019, Nie et al. described the N-alkylated diketopyrrolopyrrole derivatives **69** and **70**, investigated as ratiometric and fluorescent sensors for detecting Cu^{2+} (Scheme 27) [219]. Compound **69**, in acetonitrile, showed absorption bands at 495 and 534 nm, whereas **70** displayed bands at 506 and 543 nm. Adding Cu^{2+} led to a decrease in these bands and the appearance of a new absorbance band at 600 and 596 nm for compounds **69** and **70**, respectively, corresponding to a naked eye color change from yellow to blue. Furthermore, the fluorescence emission of **69** and **70** at 545 and 561 nm, respectively, was suppressed upon adding Cu^{2+} . LODs were calculated to be 65 and 127 $\text{nmol}\cdot\text{L}^{-1}$ by **69** and **70**, respectively. NMR, MS, and EPR measurements and theoretical calculations led the authors to propose that the optical response is associated with forming **69** and **70** radical cations (**71** and **72**). In

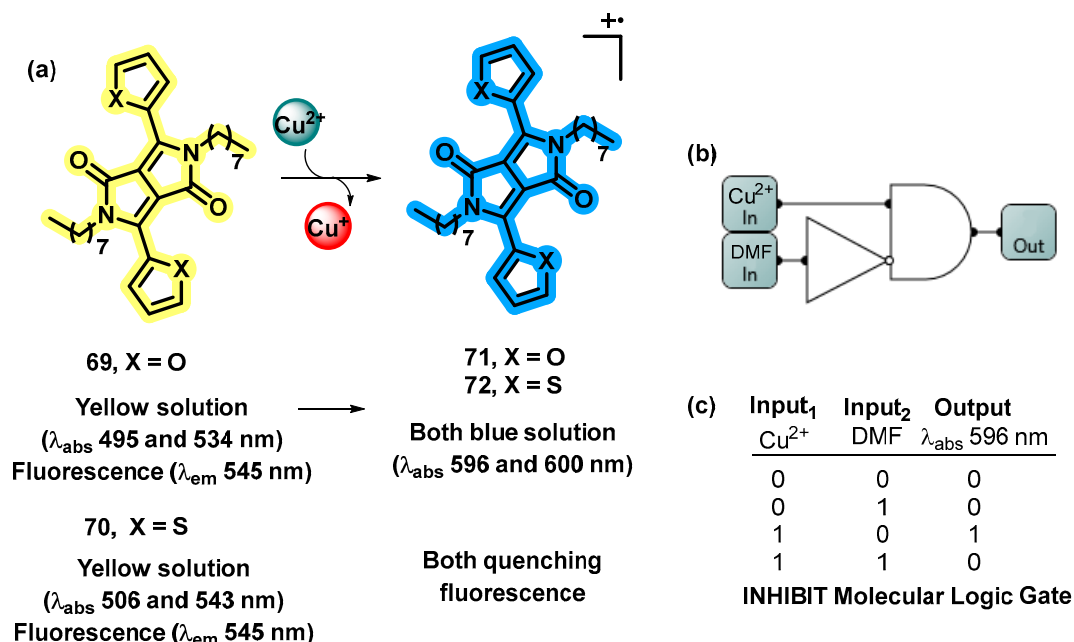
addition, the authors have shown that the optical responses depend on using acetonitrile as a solvent once the redox potential of the $\text{Cu}^{2+}/\text{Cu}^+$ is solvent-dependent. Therefore, solvent used could drastically affect the radical process. For instance, in DMF–acetonitrile (1:100, *v/v*) solution, the reaction between **70** and Cu^{2+} (to form **72**) was reduced, and the new absorbance absorption at 596 nm was reduced. In this sense, an MLG was described considering that the only input of Cu^{2+} in **69** led to an absorption at 596 nm as an output. However, DMF input in **69** kept the original absorption band at 534 nm, regardless of Cu^{2+} presence (Input₁ “1” or Input₁ “0”). Therefore, **69** could be described as an INHIBIT logic gate, based on AND and NOT gates constructed using Cu^{2+} /DMF as inputs (Scheme 27b,c).



Scheme 25. (a) MLG **59** based on solvent effects in detecting metals. (b) Diagram and (c) truth table associated with **59** described by Das et al. [201].



Scheme 26. MLG 64 development by Eördögh et al: (a) sensing mechanism described as function of solvent medium and (b) solvent apolar and polar investigated and (c) the correspondent truth table [218].



Scheme 27. MLG 69 and 70 described by Nie et al.: (a) the sensing mechanism for Cu²⁺ detection, (b) the diagram, and (c) the truth table considering DMF and Cu²⁺ [219].

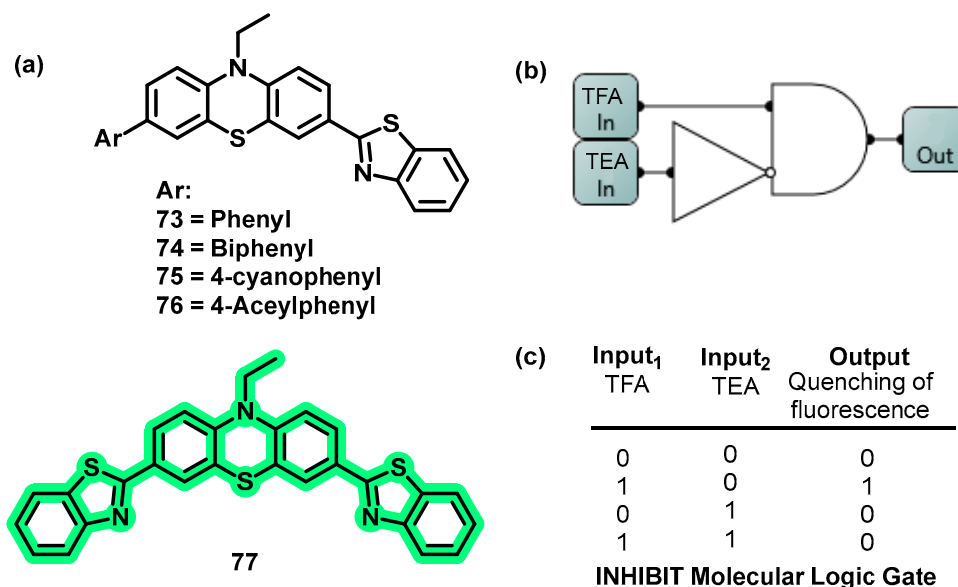
Acid-Based Effect in Reversibility

pH may influence the detection process of sensors that respond to changes in acidity or basicity. This parameter can affect the charge and structure of analyte, consequently influencing its interaction with the sensor. Furthermore, pH can induce some aggregation

effect or even interfere with the excited ground state of a device [220–222]. pH dependence can impair the selectivity and efficiency of the device by preferentially reacting with chemosensor molecules. However, it is interesting to build MLGs based on reversibility tests, considering an acid–base system capable of responding to acidity and basicity stimuli with alternations in the photophysical behavior of the medium [223–227].

For instance, in 2021, Sachdeva and Milton described the phenothiazine–benzothiazole derivatives **73–77** bearing a push–pull system based on the phenothiazine as the electron-donating moiety and benzothiazole as an electron-acceptor group (Scheme 28a) [228]. Fluorosolvatocromic studies indicated that **73–77** displayed green emission in toluene (nonpolar solvent) and yellow emission in DMSO (polar solvent). The authors evaluated the possibility of these compounds, in solution and solid state, detecting acids based on the basicity of the nitrogen of the benzothiazole ring.

The absorption of **73–77** in CHCl_3 around 300 and above 390 nm gradually shifted to around 320 and above 483 nm, respectively, upon adding trifluoroacetic acid (TFA). These optical responses represent a naked-eye color change from green to pink. Solutions **73–77** in CHCl_3 displayed emission around 515–521 nm, gradually quenched upon adding TFA. Compounds **73–77** showed an LOD of TFA of 2.46–1.21 ppm. The acid chromic behavior reversibility of these compounds was confirmed after the sequential addition of triethylamine (TEA) to the weak emissive **73–77**/TFA solutions, leading to the fluorescence recovery. These results disclose that these compounds can be considered acid–base switches for detecting volatile acids in solution.



Scheme 28. (a) MLGs **73–77** INHIBIT-type logic based on acidity media reported by Sachdeva and Milton. (b) The correspondent diagram and (c) truth table [228].

In a solid state, the yellow color of **73–77** was shifted to brown upon exposure to TFA vapors, followed by the original color upon sequential exposure to TEA vapors. Like the solution state, the emissive green/yellow **73–77** under UV light illumination suppressed the fluorescence upon adding TFA fumes. However, the sequential fuming with TEA vapor recovered the emission in the solid state. These results indicate that these compounds could be applied as smart materials as acid–base switches.

The authors have also described the coating of **73–77** chloroform solutions in Whatmann filter paper for a practical application via paper strips. The exposure of TFA vapors to these paper strips resulted in a visual color change from yellow to pink, followed by a reversion due to the subsequent exposure of TEA vapors. The green or greenish-yellow emission of these devices, under UV light ($\lambda = 365$ nm), was suppressed upon fuming with TFA vapors with a reverse effect in fluorescence after exposure to TEA vapors.

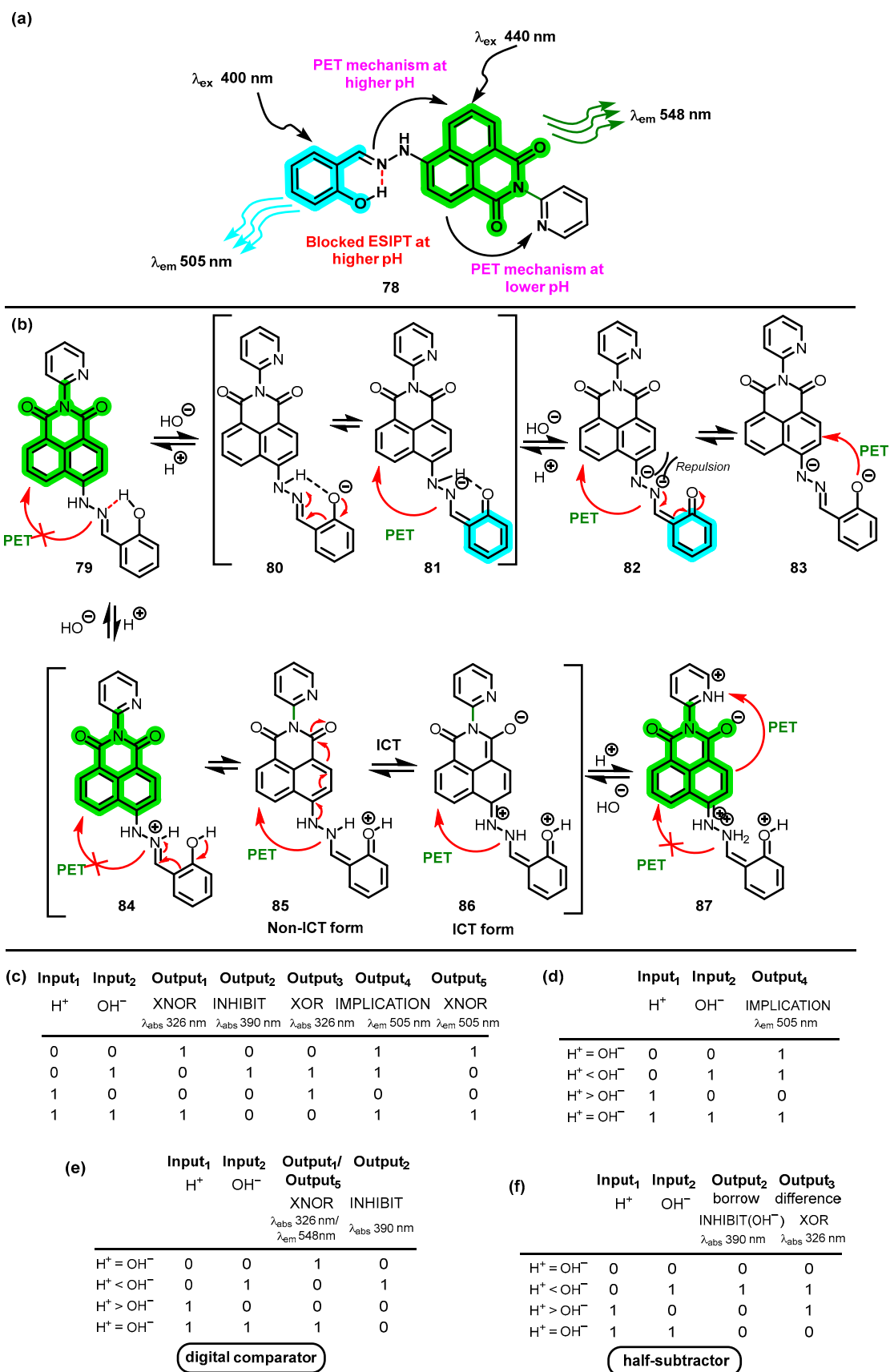
In this system, output "1" was attributed to the fluorescence quenching due to the protonation of **73–77** (Scheme 28b). On the other hand, output "0" was attributed to the strong compound emission in a deprotonated state. Therefore, input (0, 0) corresponds to the original **73–77**, resulting in an output "0". Adding only TFA, thus input (1, 0), afforded an output signal "1", whereas TEA vapors (input (0, 1)) did not change the optical response of the sensors, resulting in an output signal "0". The sequential addition of TFA and TEA, input (1, 1), led to the deprotonated state of benzothiazole derivatives, affording the output "0". In this sense, **73–77**, reversible chromic acid devices, could be described as an INHIBIT MLG (Scheme 28c).

In 2019, Said, Georgiev, and Bojinov described the chemosensor **78**, based on 2-hydroxyphenylhydrazone and 1,8-naphthalimide moieties [229], which operates with an "off-on-off" process as a response to pH alterations (Scheme 29). The structure of **78** was designed to present a modification of electronic molecular distribution via PET, ICT, and ESIPT (Scheme 29a). In a neutral medium, **78** displays absorption at 326 nm, ascribed to the benzenoid form of the salicylaldehyde hydrazone moiety, and at 440 nm, associated with the ICT of the 1,8-naphthalimide group. The absorbance at 440 nm was not affected upon lowering the pH, but the absorption at 326 nm was decreased (87). On the other hand, in an alkaline medium, the absorbance at 326 nm is shifted to 390 nm due to the formation of quinoid form (**82**) from the benzenoid moiety.

The authors also described the effect of pH on the emissive properties of **78**. This MLG exhibited strong fluorescence emission at 548 nm from the 4-amino-1,8-naphthalimides groups at a neutral medium. Upon adding a base, this emission was suppressed, attributed to the deprotonation that unlocked the PET process from the imine nitrogen to the 1,8-naphthalimide group (**81**). A new emission band centered at 505 nm emerged, attributed to the quinoid form of the salicylaldehyde hydrazone group. In an acidic media, **87** showed only the fluorescence from the 1,8-naphthalimide moiety at a lower intensity.

The authors have considered **78** as an MLG using different outputs, resulting in different truth tables (Scheme 29). Then, initially at pH = 7, **78** displayed outputs of absorption at 326 nm (output₁), a low output of absorption at 390 nm (output₂), and fluorescence emissions at 505 nm (output₄) and 548 nm (output₅). Adding only input H⁺ (input) resulted in an output "0". On the other hand, adding only OH[−], the absorption at 326 nm and the fluorescence at 548 nm decreased, resulting in outputs "0", while the absorbance at 390 nm and fluorescence emission at 505 increased, resulting in outputs "1". The simultaneous presence of the two inputs (H⁺ and HO[−]) neutralized each other, affording the initial states of the outputs.

As the transmittance is a reciprocal of the absorbance, the transmittance recorded at 326 nm as output₃ is the inverter of the absorption at 326 nm (output₁). Therefore, the **78** related to the absorption at 326 nm (or emission at 548 nm), absorption at 390 nm, transmittance at 326 nm, and fluorescence emission at 505 nm could be associated with XNOR, INHIBIT, XOR, and IMPLICATION logic gates, respectively (Scheme 29c). Additionally, using the emission output at 505 nm as output₄ and H⁺ and OH[−] as inputs, **78** can be interpreted as an MLG of the IMPLICATION type, respectively (Scheme 29d). More elaborate logic-based operations were carried out. Considering the parallel action of both the XNOR (output₁ or output₅) and INHIBIT (output₂) logic gates, a magnitude digital comparator at the molecular level was described (Scheme 29e). Finally, a half-subtractor could also be identified with the combination of an INHIBIT logic gate (output₂) and XOR gate (output₃; Scheme 29f).

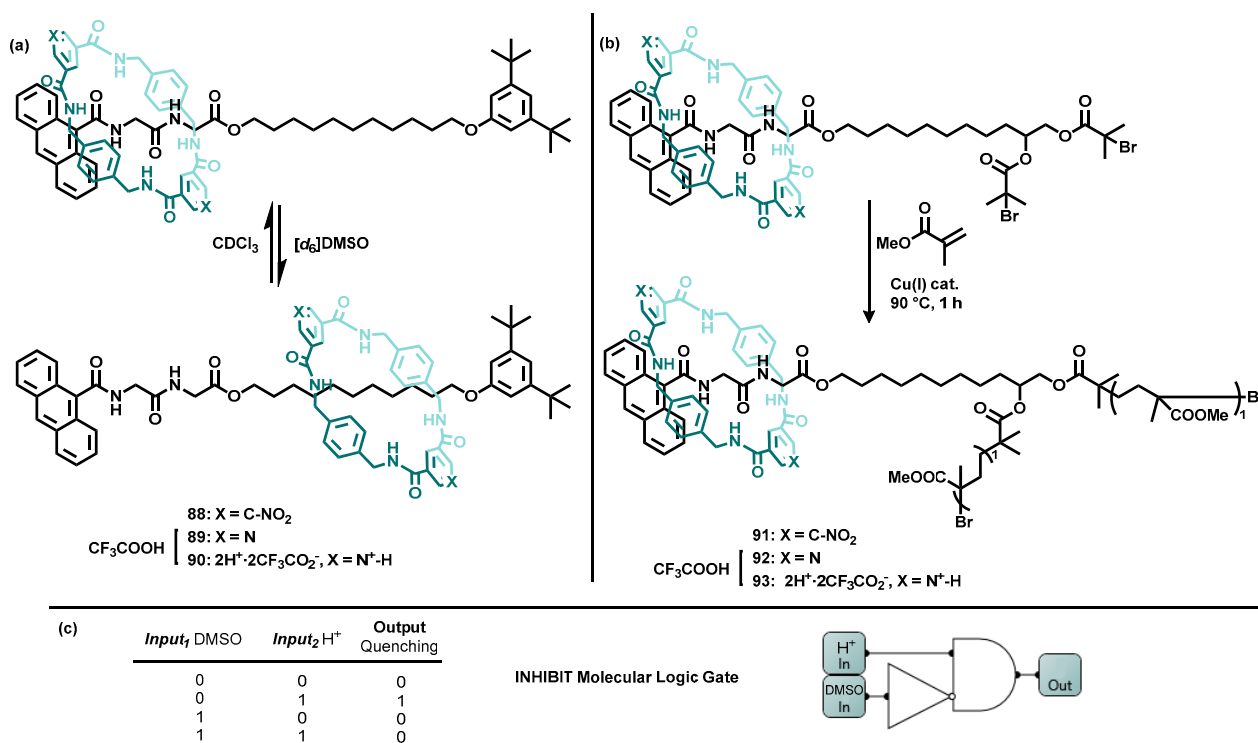


Scheme 29. (a) MLG 78 reported by Said, Georgiev, and Bojinov. (b) Sensing mechanism for H⁺ and HO⁻ and (c–f) different truth tables considering diverse possibilities of outputs [229].

Molecular machines as sensors based on binary logic is a great field to be explored [230]. One notable example is the application of rotaxanes, structurally engineered devices capable of responding to specific stimuli with consequent changes in the photophysical properties, leading to a potential interpretation as a logic gate. These features highlight the potential for utilizing molecular machines to enhance the functionality and versatility of MGLs in various applications.

One example of this approach is described by Leigh et al. [231]. The authors introduced a novel system (88–93) that involved moving a ring along a peptide-based thread to control the fluorescence of a rotaxane by altering the surrounding environment (Scheme 30). Through NMR studies in CDCl_3 , the macrocycle of 89 and 90 predominantly resided over the peptide residue of the rotaxane. However, in $\text{DMSO-}d_6$, the protons of the alkyl chains' signals were shielded, indicating the encapsulation of the alkyl chains by the macrocycle. To investigate whether the same principles could be applied in modified environments, the authors prepared analogous materials to rotaxanes (91–93) through transition-metal-mediated living radical polymerization, as represented in Scheme 30b.

The authors covered quartz slides with 92 films and observed that in the absence of acid, the pyridine units are not protonated and, therefore, the fluorescence emission of anthracene remains. When submitting the films to TFA steam, the films become dark (93). When the dark strips derived from 93 were exposed to DMSO vapor using the same mask rotated at 90° , a cross-pattern was formed. In this pattern, only the segments exposed to TFA vapor (and no DMSO vapor) became dark when illuminated with UV light. According to the authors, this result suggests a displacement of the macrocycle during the process. When interpreting the results according to binary logic, this rotaxane operates according to an INHIBIT logic (Scheme 30c), in which the output “1” is identified as the suppression of fluorescence caused by protonation of the system (input_2), which is vetoed in the presence of DMSO (input_1).



Scheme 30. Molecular machines 88–93 structures proposed by Leigh et al. [231]: (a) Proposed translational isomerism exhibited by being environment-switchable (88–90). (b) Rotaxane initiators and the corresponding PMMA-based polymers (91–93). (c) Truth table and diagram associated with INHIBIT-type MLG 92.

6. Advantages, Limitations, and Challenges of MLGs

The interpretation of optical detection devices as MLGs presents a series of limitations that range from those directly related to the planning of an optical device to those directly related to its application. Ensuring minimum sensitivity and selectivity for detecting a specific group of analytes is a challenge when planning the chemical structure and choosing the solvent and conditions of the medium in which this device will work. The systems discussed in this review present an approach that can be used to facilitate various applications. Furthermore, it is essential to develop systems compatible with the environments in which they are used. Effects of variations in analyte concentration and reaction reversibility must also be analyzed with caution. Integrating Molecular Logic Gates with existing technologies can be challenging, especially regarding communication and control applied to sensitive areas such as medicine and the environment.

One promising direction in MLG research is integration into hybrid systems, mainly polymers, for constructing practical devices. Increasing the surface area, ease of handling and storage, and inertia against different gases and liquids makes it possible to fabricate optical sensors based on sophisticated methodologies that revolutionize supramolecular analytical chemistry.

MLGs may be particularly valuable in medical diagnostics, where the accurate analysis of multiple biomarkers can be critical for identifying a specific health condition. Therefore, applying such devices in real samples is another challenge that must be investigated separately. This challenge lies mainly in the interference of possible constituents of a sample (in addition to the analyte), which may compromise the quality of the results obtained. Thus, the matrix effect plays a crucial role in validating an analytical method that brings the real application of MLGs [232]. For instance, clinical samples (blood, tissue fluid, synovial fluid, and others) are of broad interest for health reasons. However, these are also samples where the matrix effect may be the most important due to numerous constituents such as salts, carbohydrates, lipids, proteins, metabolites, and others [233]. Understanding the matrix effect and its solution is an aspect that needs to be studied on a case-by-case basis, as it depends on the characteristics of the interfering agent and the detection device. However, some approaches in general can be used, such as a standard calibration curve, an internal standard, a matrix-matched standard, and interference removal, among others [234].

Despite these challenges, MLGs represent a fascinating class of components that operate based on chemical interactions or reactions, offering advantages that can revolutionize the analysis and detection of analytes. Unlike many traditional methods that rely on extremely low LODs, MLGs can be designed to selectively recognize chemical targets without requiring extraordinarily high sensitivity, because the focus is on the combination of responses and not necessarily a low quantification of a given analyte. MLGs can be designed to operate in a complex environment containing various chemical species, allowing for simultaneous detection and distinction between them or some of them.

7. Conclusions

Optical detection devices as analytical tools for identifying and quantifying different chemical species have evolved over the years to become increasingly efficient, offering advantages such as lower limits of detection and quantification, greater selectivity and sensitivity, detection of multiple analytes, and different responses to the same analyte, among others. The concept of Molecular Logic Gates represents a significant advance in the area of molecules that perform a logical operation based on one or more stimulus signals (inputs) resulting in a response (output). Therefore, numerous published examples of MLGs involve molecular architectures containing different detection sites for recognizing different analytes. However, in this review, we showed that in many cases, it is not just a question of building various detection sites but exploring strategies such as reversibility in detection based on the removal of the first analyte (input₁) from the complex formed by a second analyte (input₂). Furthermore, there are cases in which the first input triggers the formation of another device, which will then act as a second MLG, generated in situ, for recognizing

another analyte. In this scenario, through a careful analysis of the interaction among analytes, the media (solvent, pH, and viscosity), and the sensor, we explored the state of the art in MLGs based on the selection of a second analyte, making the system more versatile and applicable to a different type of matrix. Therefore, we conclude that the research on classical sensors is being shifted towards describing more elaborate strategies: aspects related to self-assembly and the programming of stimuli that result in controlled movement are variables that can and are being improved. Advances in the construction of smart drug delivery systems, chemical communication technologies applied to nanotechnology, and advances in increasingly urgent environmental monitoring are believed to be the most promising fields of research and development.

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Abbreviations

AcO [−]	Acetate
ADP	Adenosine Diphosphate
Arg	Arginine
ASSURED	Affordable, Sensitive, Specific, User-friendly, Rapid/Robust, Equipment free, and Deliverable
A–D–A	Acceptor–Donor–Acceptor
ATP	Adenosine Triphosphate
CHEF	Chelation Enhanced Fluorescence
cit ^{3−}	Citrate
Cys	Cysteine
D–A	Donor–Acceptor
D–A–D	Donor–Acceptor–Donor
DFT	Density Functional Theory
DMSO	Dimethyl Sulfoxide
DNA	Deoxyribonucleic Acid
EDTA	Ethylenediaminetetraacetic Acid
EPR	Electron Spin Resonance
ESIPT	Excited State Internal Proton Transfer
EtOH	Ethanol
FTIR	Fourier-Transform Infrared Spectroscopy
GSH	Glutathione
HCy	Homocysteine
His	Histidine
ICT	Internal (or Intramolecular) Charge Transfer
INH	INHIBIT Molecular Logic Gates
K _a	Binding Constant
LG	Logic Gate
LOD	Limit of detection
LOQ	Limit of Quantification
Lys	Lysine
MeOH	Methanol
MLCT	Ligand Charge Transfer
MLG	Molecular Logic Gates
MS	Mass Spectra
NMR	Nuclear Magnetic Resonance
nPrOH	Propan-1-ol
PET	Photoinduced Electron Transfer

PA	Picric Acid
Ppb	Part Per Billion
PPi	Pyrophosphate
Ppm	Part Per Million
QY	Quantum Yield
RNA	Ribonucleic acid
TEA	Triethylamine
TFA	Trifluoroacetic Acid
THF	Tetrahydrofuran
Trp	Tryptophan
λ_{abs}	Maximum absorption wavelength
λ_{em}	Maximum emission wavelength
λ_{max}	Maximum wavelength

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