



Molecular AND Logic Gates Based on ZnO Nanoparticles Functionalized with Imine Receptors via PET Mechanism

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Abstract: Driven by demand for miniaturized logic systems, development of molecular logic gates based on semiconductor nanoparticles provides a promising platform for next-generation optoelectronic applications. The study showcases the successful realization and application of ZnO nanoparticle-based molecular AND logic gates functionalized with imine-linked receptors. Two systems were designed utilizing photoinduced electron transfer (PET) mechanisms: the first employing 2-aminothiophenol for selective detection towards H^+ and Cu^{2+} ions, while the other employs 2-aminothiols targeting H^+ and Fe^{3+} ions. When both chemical inputs are present simultaneously, distinct strong fluorescence emissions at 440 nm and 490 nm, respectively, were observed, confirming the Boolean AND logic function. The design and functionality of these logic gates were validated via comprehensive fluorescence studies and PET energy transfer mechanisms. The study additionally explores single-input (YES) logic behaviour and clarifies the chemical mechanisms underlying the switching operation. These results pave the way for promising opportunities to integrate, molecular logic operations into nanoscale photonic systems chemical sensors, as well as advanced intelligent biochemical devices.

Index Terms - Molecular logic gates, PET, ZnO nanoparticles, imine receptor, AND gate, fluorescence, chemical sensors.

I. INTRODUCTION AND PET PROCESS MECHANISM

In the information technology era, Boolean logic plays a crucial role in solving algebraic equations, with digital systems employing various logic gates to realize logical operations. Traditional digital systems rely on logic gates fabricated from bulk semiconductors that process input and output signals. Although semiconductor features have been miniaturized down to below 32 nm [1], approaching the scale of a single molecule remains a significant challenge. The concept of molecular logic gates, introduced in the early 1990s [2], opened new avenues for supramolecular constructs capable of mimicking logic functions such as AND, OR, and NOT [3]. Unlike bulk semiconductors which are typically non-fluorescent, semiconductor nanoparticles exhibit fluorescence due to their reduced size and quantum confinement effects, making them ideal for optical sensing and biochemical tagging [4]. The implementation of molecular logic gates using nanoparticles is a novel and actively researched area.

In this study, we demonstrate two molecular AND logic gate systems based on ZnO nanoparticles functionalized via photoinduced electron transfer (PET) processes. The first system employs two PET channels: one binding H^+ ions and the other Cu^{2+} ions to the nanoparticle surface, producing a fluorescent output at 440 nm upon simultaneous presence of both inputs. Similarly, the second system utilizes PET channels for H^+ and Fe^{3+} ions, generating fluorescence at 490 nm when both ions are present, thereby realizing the molecular AND logic operation.

The PET mechanism forms the foundation for these sensing systems due to its modular design and sophistication. Initially reported by Weller in 1998 and later extended by de Silva et al. [5], PET sensors typically comprise three components: a fluorophore (usually an organic dye), a receptor that selectively binds the analyte, and a **spacer** that electronically decouples the two, preserving their individual optoelectronic properties [6]. The fluorophore acts as the signalling unit by emitting fluorescence, the receptor ensures selective and sensitive analyte recognition through chelation or hydrogen bonding, and the spacer—often a methylene or ethylene chain—maintains efficient PET by electronically isolating the fluorophore and receptor [7]. This is illustrated in figure 1.

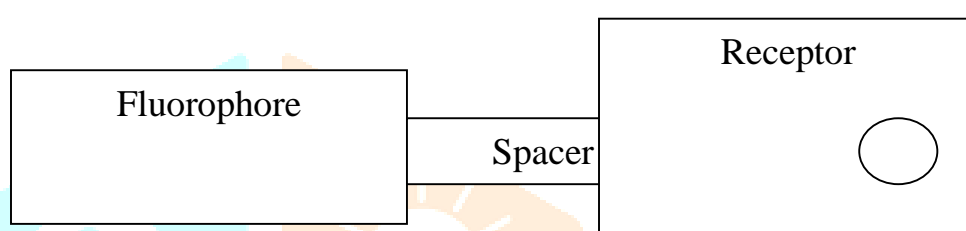


Figure 1: The 'fluorophore-spacer-receptor' structure of fluorescent PET sensors.

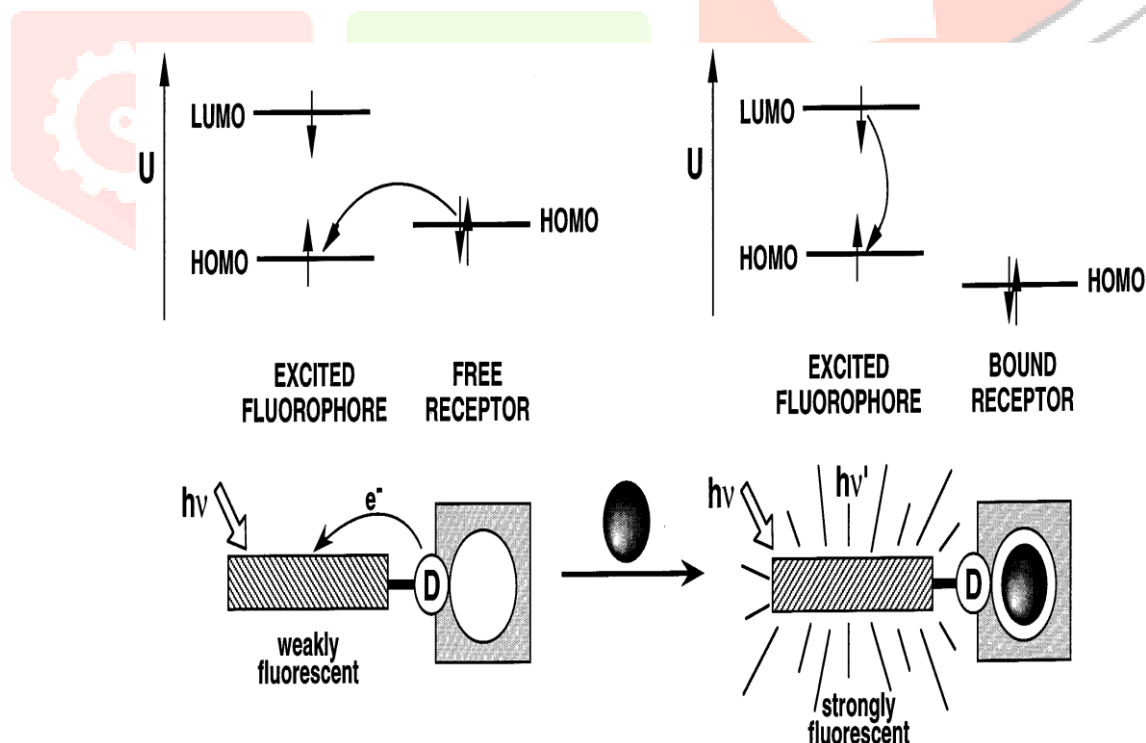


Figure 2: Method of an electron transfer from the analyte-free receptor to the photo-excited fluorophore creates the 'off' state of the sensor. (b) Method of the electron transfer from the analyte-bound receptor is blocked which results in the 'on' state of the sensor.

In the absence of the target analyte, PET from the receptor (electron donor) to the excited fluorophore quenches fluorescence, creating the sensor's "off" state (Figure 2a and 2b). The corresponding frontier molecular orbital energy diagram (Figure 3a) shows that the receptor's HOMO lies higher in energy than the fluorophore's HOMO, facilitating electron transfer and quenching emission. When the analyte binds to the receptor, this raises the receptor's oxidation potential and lowers its HOMO energy, disrupting the thermodynamics of PET. As a result, the electron transfer is blocked, fluorescence is restored, and the sensor switches "on" (Figure 2b). The molecular orbital diagram for the bound state (Figure 3b) illustrates this stabilization of the receptor HOMO below the fluorophore's HOMO, preventing PET quenching.

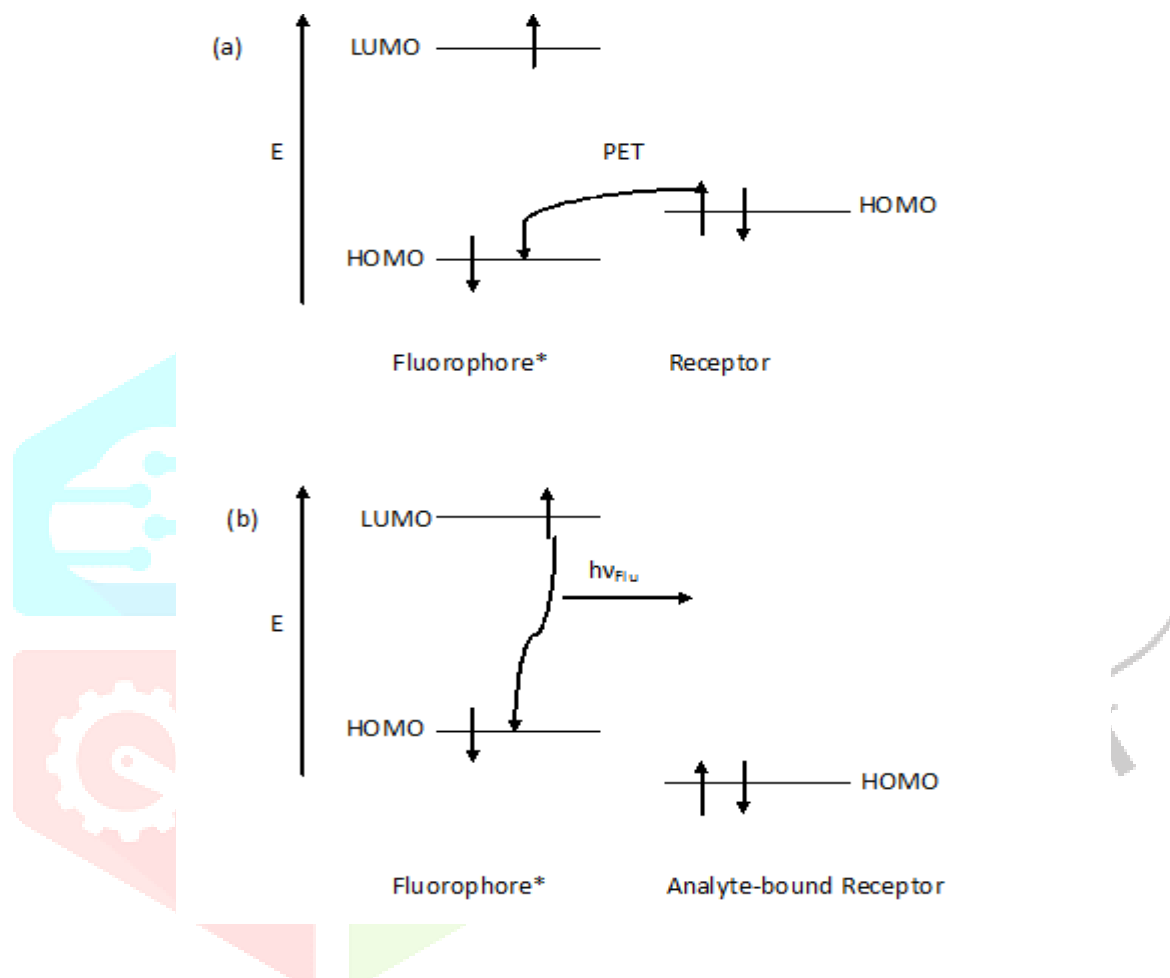


Figure 3: Concept of the molecular orbital energy diagrams which show the relative energetic dispositions of the frontier orbitals of the fluorophore and the receptor in (a) the analyte-free situation and (b) the analyte-bound situation.

Thus, this "on/off" fluorescence switching governed by PET forms the basis of a molecular photonic switch, an essential component for nanoscale photonic devices. The integration of such PET-based molecular logic gates with fluorescence output enables promising applications in molecular computing, chemical sensing, and intelligent biochemical devices.

This work highlights the potential of semiconductor nanoparticles functionalized through PET mechanisms as versatile platforms for constructing molecular-scale logic devices. Such molecular logic gates not only offer a pathway toward extreme miniaturization in computational elements but also pave the way for advanced optoelectronic devices capable of sensitive, selective chemical detection. These systems provide a foundation for future developments in intelligent biochemical devices, molecular diagnostics, and integrated photonic circuits, ultimately bridging the gap between molecular chemistry and nanoscale electronics.

2. Molecular Logic Gates

The capability and efficiency of information storage and processing technology are quickly approaching a limit. Optional material and operating principle for its implementation and communication of data in electronic circuits and optical networks must be recognized fully. Computing at the molecular level is predicted as primary solution to overcome the drawback of the computing device in term of processing speed and storage capacity. Their outstanding feature involves miniaturized dimensions and high degree of control on molecular design by use of the chemical synthesis. In most recent years a range of molecules that respond to different chemical inputs have been synthesized and established as the molecular logic gates. In general, information processing at the molecular level is based on two different approaches [8]. In first approach attempt has been made to imitate the working principle of the solid-state computers which are presently in use, at the nanometer scale with molecular system. This strategy is based on the concept of molecular electronics in which output and input signals are electronics in nature. In second strategy molecules and ions can be used as the input and output signals to process the information by using the molecular substrate. This concept can be implemented in the solution phase and complemented by the photonics, since the chemical and light input and output can be coupled

easily. With each concept of the molecular electronics, photonics and Chemonics [9] information processing can take place in logic gates and data processing relies on the binary nature of the input and output and hence can be manipulated using the concept of the Boolean algebra.

Among the different recording media for the information storage, organic materials have been found to be attractive in the current few years because of their low cost, excellent stimuli responsive property and provide flexibility [10] in the molecular design. In principle, the recording materials must possess two distinct stable states by an external stimulus, where each state can be represented by “0” and “1” values. Generally, any chemical system that exists in two quasi-states of different physical or chemical properties may be considered as a molecular switch or molecular logic gate, provided there must be some chemical or physical stimuli that can alter or change the state of the system.

There are four different possible combinations of the input and output values for the one input and one output logic gates. The pass 0 and pass 1 will provides “0” and “1” respectively, irrespective of the value of input signal. The YES gate will follow the input value to the output. Gates used here will function like a switch, and useful for connecting various devices and for the purpose of the signal amplification. The NOT gate or inverter performs the inversion or complement of the input data. Inverter converts the logic 0 into the logic 1 and vice-versa as shown in table 3.1

Table 3.1 Truth table of single input logic gate

Input	Output			
	Pass 0	YES	NOT	Pass 1
0	0	0	1	1
1	0	1	0	1

Table 3.2 Truth table for the two input logic gates

Input		Output					
		OR	AND	NAND	NOR	XOR	XNOR
0	0	0	0	1	1	0	1
0	1	1	0	1	0	1	0
1	0	1	0	1	0	1	0
1	1	1	1	0	0	0	1

Two input logic gates are used in electronics and made to implement the gate operation like OR, AND, XOR, NOR, NAND and XNOR functions as shown in Table 3.2.

3. Chemical Sensor (Chemically Driven Molecular Logic Gates)

There is great interest in the field of the supramolecular chemistry in developing a variety of smaller molecular devices that can imitate the action of the switches and motors [11]. These devices can show change in the “on” and “off” state by the process of the luminescence which can be tuned by employing with external inputs such as ions, molecules [12] and light etc.

Chemically driven molecular gates or switches generally consists of three most important building blocks named as receptor moiety, spacer or linker and the reporter moiety [13]. Receptor moieties are the specifically designed binding sites for triggering the different molecules and ions. They must provide suitable selectivity and sensitivity for the any value of the chosen triggers. Visualization of the interaction [14] and to report about them can be accomplished by providing a signal that can be analyzed and measured. The structure accountable for creation of this signal will be called as reporter. Spacer will provide the electronics communication between the reporter and receptor moieties. Organic structure held by the covalent bonds and that are capable of binding selectively

ionic or molecular substrate by mean of intermolecular interaction is called as molecular receptor. The sensing function can be integrated at the molecular level. This can be achieved by combining the reporter group and binding site in one molecule. Most optical sensors are based on the phenomena of the fluorescence.

Fluorescence was used as one of the analytical devices to determine the concentration of the different species which may be ionic or neutral. When the analyte is fluorescent, we can find the concentration directly; otherwise, indirect methods such as creation of the various fluorescent complexes or fluorescence quenching has been developed. In fluorescent sensors the fluorophore acts a signal transducer which converts the information into the optical signal. Generally, a fluorescent sensor is an analyte receptive molecular moiety, relating to the fluorophore that indicates the presence or existence of the analyte by change in it fluorescence characteristics. Design of the fluorescent sensors is of key significance because of its greater demand in the field of analytical and clinical biochemistry. Large number of the analytes can be detected by using the fluorescence methods like cations (Na^+ , H^+ , K^+ , Mg^{2+} and Zn^{2+} etc), anions (carboxylates, halides and citrates) and sugar molecules (glucose and sugar)

Generally, there are three methods of providing adequate electronics communication. In first method electronics communications can be provided by use of bridge which can facilitates the overlap of the π -system of the both the moieties. Second method involves the process of the photo-induced transfer, while third method involves arrangement of the reporter and the receptor moieties using the supramolecular interactions to provide the disordering [15] of the electronics structure of the receptor moieties.

Chemically driven molecular gates or switches are based on the optoelectronics phenomenon such as photoinduced electron transfer (PET), Electronic energy transfer (EET), Photoinduced charge transfer (PCT) [16]. The PET based sensor can be triggered by the various physical and chemical based external stimuli such as metal cations, protons, anions and neutral organic molecules. Depending on the desired properties and lifetime of the molecular switch, molecular arrangement may consist of various inorganic or organic fluorophores [17]. Due to the large range of the various organic ligands, it was possible to design the fluorophores that responds to variety of signal inputs appropriate for various logic operations and sensing. In this section we will restrict our discussion for implementation of the molecular AND gate driven by external stimuli such as H^+ and Cu^{2+} and another using the H^+ and Fe^{3+} by use of optoelectronics photoinduced transfer (PET) phenomenon only.

It was de Silva and his associates who have exploited the utilization of Boolean function to describe the relationship between the chemical inputs and fluorescent outputs. First PET based sensor have been designed by using two input molecular AND gates consist of chemical input H^+ and Na^+ respectively. Large number of the organic fluorophores have been reported which can execute different Boolean operations independently with respect to the applied chemical outputs. Boolean operation like AND, OR, XOR, NAND, NOR and XNOR were established in molecules with different inputs including photochemical, chemical, electrochemical and various enzyme etc.

4. Molecular Logic Gate Based on the Single Input (Driven by a Chemical Input)

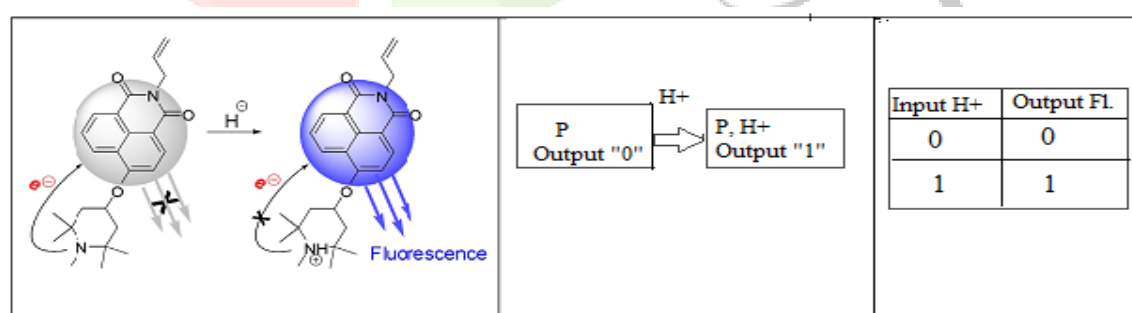


Figure 5: Chemical Sensor performing the YES logic operation under the influence of one chemical input (H^+) and the equivalent truth table

With the help of single input (e.g., binding of analyte) and single output (e.g., fluorescent intensity “switch on”) two simple functions can be implemented. It shows YES [18] when their inputs are 0 and 1 and output will remain the same. It shows NOT when their input 0 and 1 and output will remain opposite i.e. 1 and 0

respectively. It behaves as fluorescent “off-on” sensor when an analyte binds the receptor and hence, we will get strong fluorescence, which correspond to YES logic gates which is illustrated in the figure 5.

5. Molecular Logic Gate Based on Two Inputs and Implementation of Molecular AND Operation by Use of Two Input Chemical External Stimuli

More complicated logic function can be implemented with the help of two inputs in such a way that two different ions occupy two different locations. In general, there are six logic gates which consists of two inputs and one output and named as OR, AND, XOR, NOR, NAND and XNOR [19] gates respectively. Here we will illustrate the AND operation using two inputs. Here AND gate operation will get verified, if we assume logic 0 to be called as the “false” and logic 1 to be called as the “true”, then the logic gate will act in the similar way as a logical “AND” operator. When both inputs are “true” then output will also be “true”. Otherwise, output will be “false”. First molecular AND gate of primary importance was designed by de Silva. Figure 6 displayed that when two PET Channel from receptor was suppressed then we will get strong fluorescence. This can be arranged by providing two guest species on which two receptors were found to be selective. As shown in figure 6 the imine group will require H^+ (shown as input 1) as one of the inputs whereas benzo crown ether moiety will require Na^+ (shown as input 2) as another input.

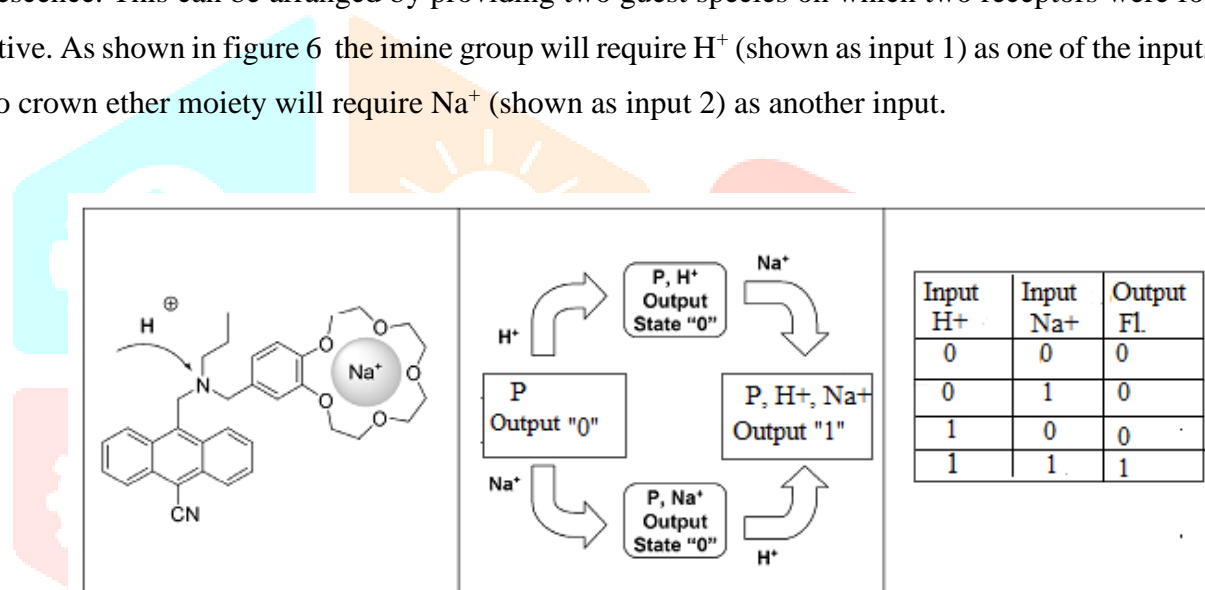


Figure 6: Chemical sensor which performs the molecular AND logic operation under the influence of two chemical inputs (H^+ and Na^+) and the corresponding truth table

6. Imine Link Based Molecular AND Logic Gates

Here in this section, we have considered the examples of the imine link-based receptors capped with ZnO nanoparticles, which can perform Boolean operation like AND function by using two chemical inputs. The AND molecular logic gate realized and constructed with the help of two PET processes. AND gate realization and implementation has been achieved by using two different examples. One example illustrated and shown the AND gate property with the help inputs H^+ and Cu^{2+} , while other exhibit the gate property with the inputs H^+ and Fe^{3+} . So in first example, synthesized imine linked based receptor capped (2-aminothiophenol) with ZnO nanoparticles has been used for realization of the AND gate operation by using PET process with the two gate inputs H^+ and Cu^{2+} . In the second example imine linked based receptor synthesized by using 2-aminothiols as capping agents. Here AND gate property has been realized by using the two inputs H^+ and Fe^{3+} . In second example Fe^{3+} has been used in place of Cu^{2+} . This is because size of Fe^{3+} ion is larger than the Cu^{2+} ion and it could not hold the 2-aminothiophenol molecule due to its straightened structure. The Fe^{3+} ions

could be held in better way by ZnO nanoparticles capped with 2-aminothiols molecule. So ZnO nanoparticle capped with 2-aminothiols has been used to exhibit the AND gate property with inputs H^+ and Fe^{3+} in the second example.

6.1 Synthesis and Characterization of the Imine link Based Receptor by Using ZnO Nanoparticles

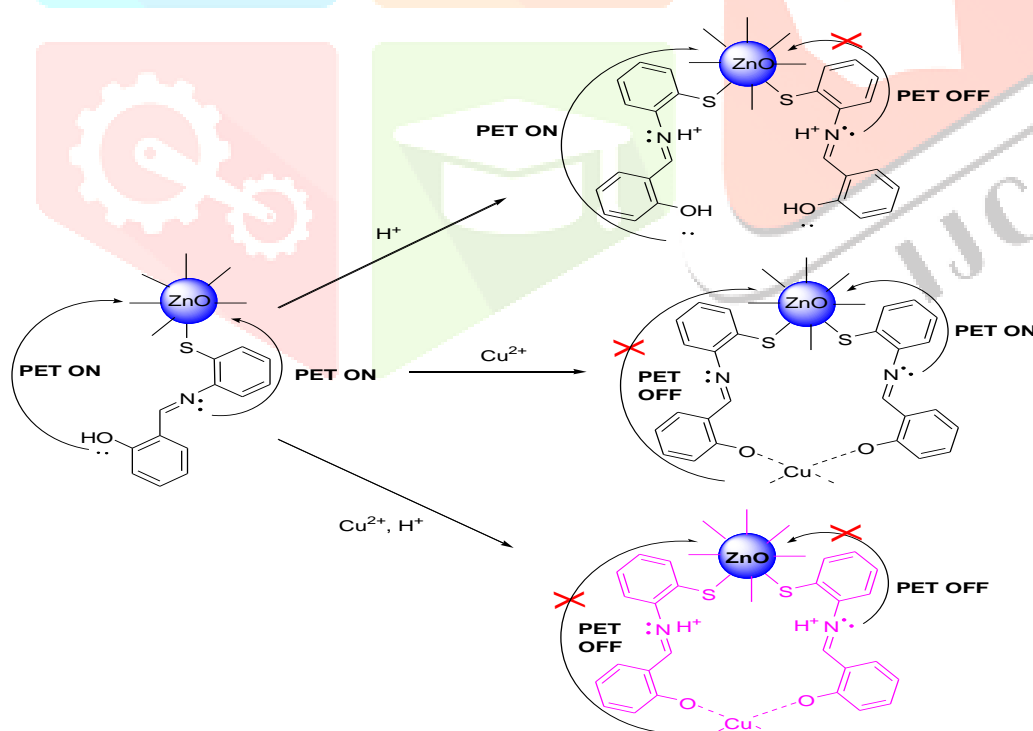
Imine linkage based ZnO receptors have been synthesized and characterized by using ZnO nanoparticles capped with 2-aminothiols and another receptor have been synthesized and characterized successfully.

6.2 Results and Discussions

6.2.1 Realization and Implementation of the Molecular Logic Gate (AND operation) by Using Imine link Based Receptor Capped with ZnO Nanoparticles with External Stimuli H^+ and Cu^{2+}

When input conditions of both low H^+ and Cu^{2+} ions were maintained then the ZnO nanoparticles result in the quenching of fluorescence by using the process of photoinduced electron transfer (PET) from receptor to the excited ZnO nanoparticles. PET operation illustration and realization using (Two different inputs Cu^{2+} and H^+) for 2-aminothiophenyl capped ZnO nanoparticle have been shown in the figure 7

When both the species i.e. H^+ and Cu^{2+} ions were simultaneously present then it results in the restoration of the fluorescence phenomenon or ZnO nanoparticles will emit light. Independent addition of any of these ions has no effect on the process of the fluorescence and hence as a result ZnO nanoparticle will not emit light. Hence, the condition of AND molecular gate has been satisfied with the Cu^{2+} ions and proton (H^+) as inputs and fluorescence emission of ZnO nanoparticles at 440 nm will be observed.



II.

Figure 7: PET operation illustration and realization using (Two different inputs Cu^{2+} and H^+) for 2-aminothiophenyl capped ZnO nanoparticle

Here we will discuss all the four possible different cases by considering Cu^{2+} and H^+ as two inputs. The fluorescence spectra of all the four different inputs signal have been shown in figure 8 (a), while the truth table realization of molecular AND gate property has been given in table 7.1.

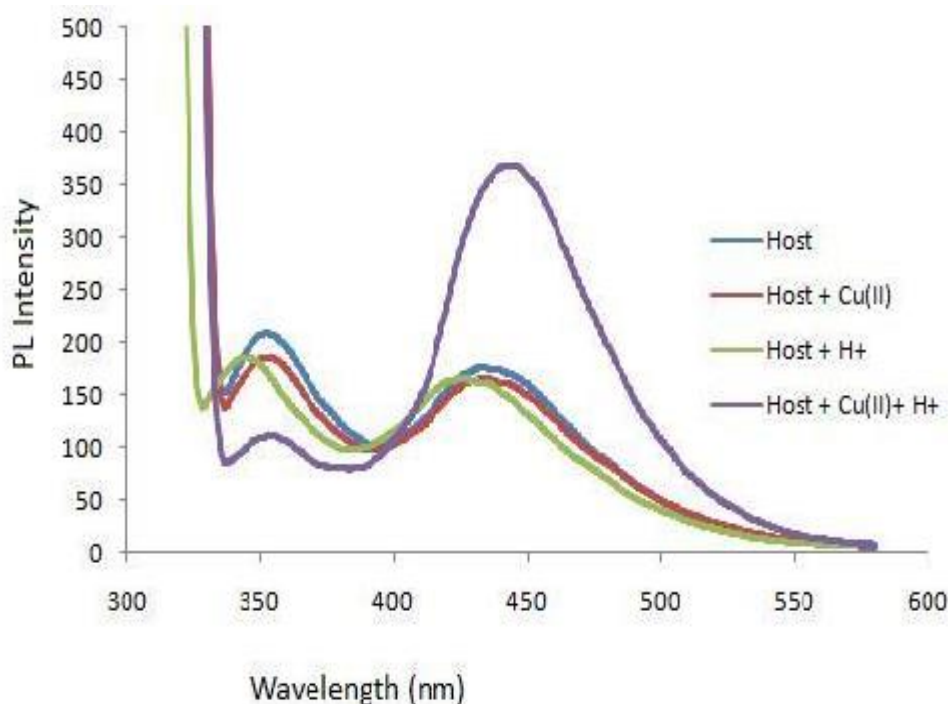


Figure 8 (a) Fluorescence spectra in the presence of: (i) low H^+ , low Cu^{2+} (ii) high H^+ , low Cu^{2+} (iii) low H^+ , high Cu^{2+} and (iv) high H^+ , high Cu^{2+} .

7.1 Truth table for AND logic gate behavior of two chemical input (H^+ and Cu^{2+})

Input 1 (H^+)	Input 2 (Cu^{2+})	Output (Fluorescent Intensity at $\lambda_{\text{max}} = 440 \text{ nm}$)
0	0	0
1	0	0
0	1	0
1	1	1

6.2.2 Truth table Realization of Molecular Logic AND Gate Using Different Inputs

Case-1 (When both Inputs Cu^{2+} and H^+ are Zero or Not Present)

When the two inputs Cu^{2+} and H^+ are not present or both inputs are zero, the lone pair of electrons presents on the nitrogen and oxygen on ZnO nanoparticles capped with 2-aminothiophenols results both PET device to be in the “on” state. The lone pair of electrons present on nitrogen and oxygen groups will get engaged with vacancy of electron created by excited state of ZnO nanoparticles using the PET process. Since both PET devices will be in “on” state hence no fluorescence will be observed resulting no fluorescence or output to in zero state. So, it confirms the Boolean AND gate property corresponding to inputs Cu^{2+} and H^+ at low states (“0”) and output to be at low state (“0”).

Case-2 (When an Input H^+ is Present (High) and Other Input Cu^{2+} is Zero (Low) or Not Present)

In this condition the lone pair of electrons present on oxygen engaged with vacancy of electron created by excited ZnO nanoparticles by using the PET process. So, no fluorescence will be observed in this case. The H^+ ion present as second input will engaged with the lone pairs of electrons of nitrogen group. As a result, no photoinduced transfer (PET), resulting in fluorescence. The combined

effect of both the inputs will be such that no fluorescence will observe at its output. This corresponds to off state or low value ("0") at the output.

Case-3 (When an Input H^+ is Zero (Low) or Not Present and Other Input Cu^{2+} is Present (High))

In this condition the lone pair of electrons present on nitrogen engaged with vacancy of electron created by excited ZnO nanoparticles by using the PET process. So, no fluorescence will be observed in this case. The Cu^{2+} ion present as second input will engaged with the lone pairs of electrons of oxygen group. So, in this case phenomenon of photo induce transfer (PET) will not occur, which results in fluorescence. The combined effect of both the input will be such that no fluorescence will observe at its output. This corresponds to off state or low value ("0") at output.

Case-4 (When Both Inputs Cu^{2+} and H^+ are Present (High))

When the two inputs Cu^{2+} and H^+ are present or both inputs are high ("1"), the lone pair of electrons presents on the nitrogen and oxygen on ZnO nanoparticles capped with 2-aminothiophenols results both PET device to be in the "off" state. The lone pair of electrons present on nitrogen and oxygen groups will get engaged with the H^+ and Cu^{2+} ions respectively, which were present as the high ("1") input signals. Both the PET devices will be in "off" state. Hence at the output fluorescence (high) will be observed. So, it confirms the Boolean AND gate property by employing two inputs Cu^{2+} and H^+ at high states ("1") and output to be at high state ("1").

6.2.3 Realization and Implementation of the Molecular Logic gate (AND operation) by Using Imine Link Based Receptor Capped with ZnO Nanoparticles with External Stimuli H^+ and Fe^{3+}

When input conditions of both low H^+ and Fe^{3+} ions were maintained then the ZnO nanoparticles result in the quenching of fluorescence by using the process of photoinduced electron transfer (PET) from receptor to the excited ZnO nanoparticles. PET operation illustration and realization using (Two different inputs Cu^{2+} and H^+) for 2-aminothiol capped ZnO nanoparticle have been shown in the figure 9.

When both the species i.e. H^+ and Fe^{3+} ions were simultaneously present then it results in the restoration of the fluorescence phenomenon or ZnO nanoparticles will emit light. Independent addition of any of these ions has no effect on the process of the fluorescence and hence as a result ZnO nanoparticle will not emit light. Hence, the condition of AND molecular gate has been satisfied with the Fe^{3+} ions and proton (H^+) as inputs and fluorescence emission of ZnO nanoparticles at 490 nm will be observed. Here we will discuss all the four possible different cases by considering H^+ and Fe^{3+} as two inputs.

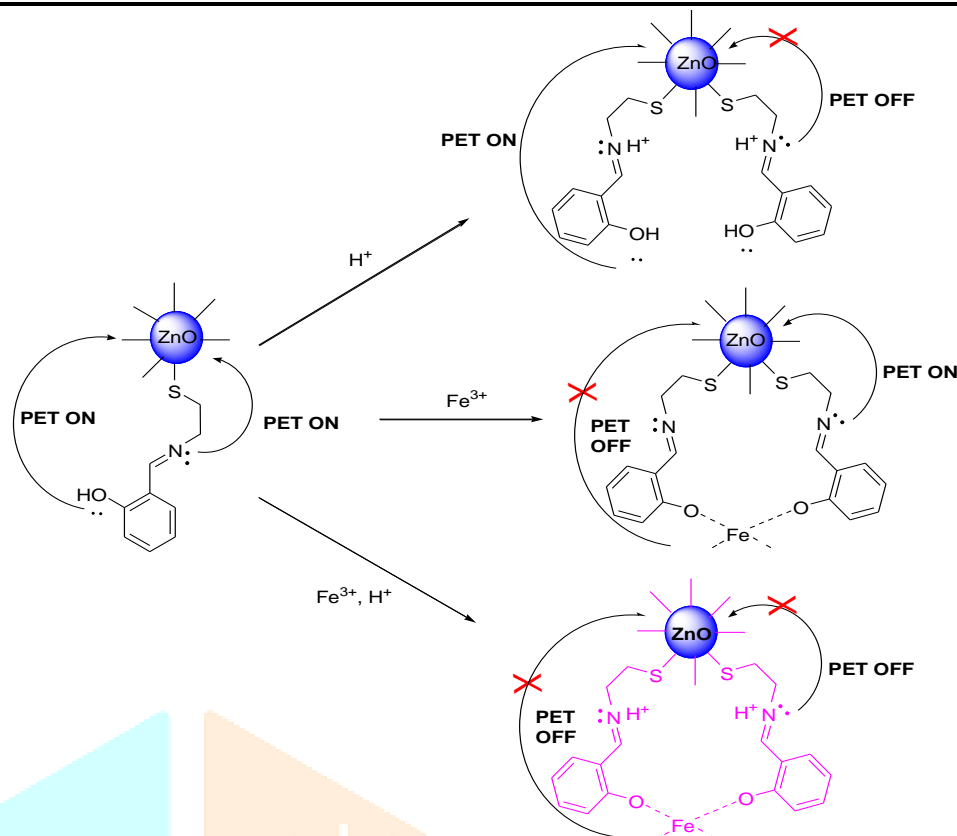


Figure 9: PET operation illustration and realization using (Two different inputs Fe^{3+} and H^+) for 2-aminothiols capped ZnO nanoparticle

The fluorescence spectra of all the four different inputs signal have been shown in figure 9.

6.2.4 Truth Table Realization of Molecular Logic AND Gate Using Different Inputs (H^+ and Fe^{3+})

This is illustrated in figure 10 and table 7.2 respectively. Different cases have been taken and summarized below.

Case-1 (When Inputs H^+ and Fe^{3+} are Zero or Not Present)

When the two inputs and H^+ are not present or both inputs are zero, the lone pair of electrons presents on the nitrogen and oxygen on ZnO nanoparticles capped with 2-aminothiophenols results both PET device to be in the “on” state. The lone pair of electrons present on nitrogen and oxygen groups will get engaged with vacancy of electron created by excited state of ZnO nanoparticles using the PET process. Since both PET devices will be in “on” state hence no fluorescence will be observed. So, in this case output will be zero state. So, it confirms the Boolean AND gate property corresponding to inputs Fe^{3+} and H^+ at low states (“0”) and output to be at low state (“0”).

Case-2 (When an input H^+ is present (high) and other input Fe^{3+} is zero (low) or not present)

In this condition the lone pair of electrons present on oxygen engaged with vacancy of electron created by excited ZnO nanoparticles by using the PET process. So, no fluorescence will be observed in this case. The H^+ ion present as second input will engaged with the lone pairs of electrons of nitrogen group. As a result, no photoinduced transfer (PET) will occur, resulting in fluorescence. The combine effect of both the input will be such that no fluorescence will observe at its output. This corresponds to off state or low value (“0”) at the output.

Case-3 (When an Input H^+ is Zero (Low) or Not Present and Other Input Fe^{3+} is Present (High))

In this condition the lone pair of electrons present on nitrogen engaged with vacancy of electron created by excited ZnO nanoparticles by using the PET process. So, no fluorescence will be observed in this case. The Fe^{3+} ion present as second input will engaged with the lone pairs of electrons of oxygen group. As a result, no phenomenon of photoinduced transfers (PET) will occur, resulting in fluorescence at the output. The combine effect of both the input will be such that no fluorescence will observe at its output. This corresponds to off state or low value (“0”) at output.

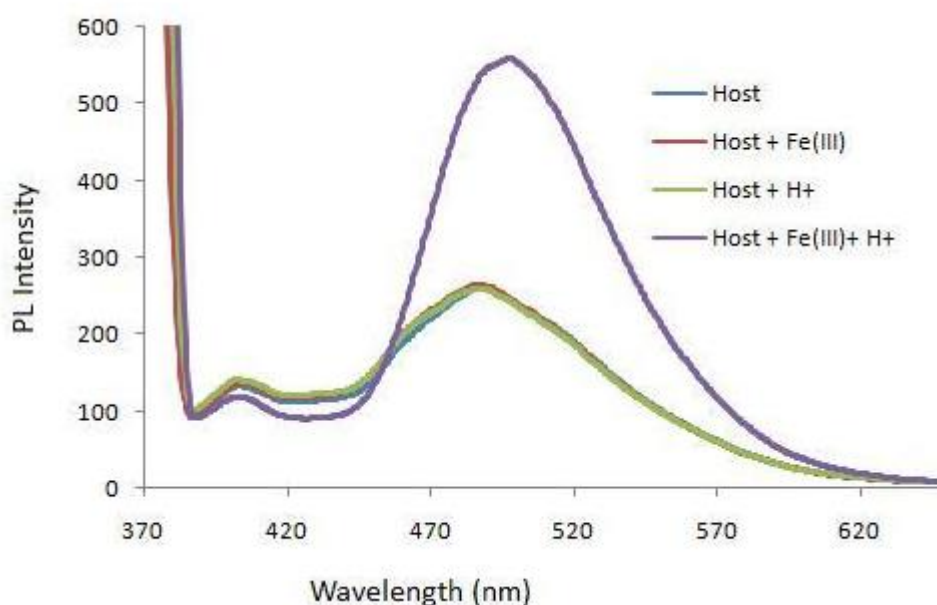


Figure 10: (a) Fluorescence spectra in the presence of: (i) low H^+ , low Fe^{3+} (ii) high H^+ , low Fe^{3+} (iii) low H^+ , high Fe^{3+} and (iv) high H^+ , high Fe^{3+}

7.2 Truth table for AND logic gate behavior of two chemical input (H^+ and Fe^{3+})

Input 1 (H^+)	Input 2 (Fe^{3+})	Output (Fluorescent Intensity at $\lambda_{max}=490\text{ nm}$)
0	0	0
1	0	0
0	1	0
1	1	1

Case-4 (When Both Inputs Cu^{2+} and H^+ are Present (High))

When the two inputs H^+ and Fe^{3+} are present or both inputs are high (“1”), the lone pair of electrons present on the nitrogen and oxygen on ZnO nanoparticles capped with 2-aminothiophenols results both PET device to be in the “off” state. The lone pair of electrons present on nitrogen and oxygen groups will get engaged with the H^+ and Fe^{3+} ions respectively, which were present as the high (“1”) input signals. Both the PET devices will be in “off” state. Hence at the output fluorescence (high) will be observed. So, it confirms the

Boolean AND gate property by employing two inputs Fe^{3+} and H^+ at high states (“1”) and output to be at high state (“1”).

7. Conclusions

The realization and implementation of the computing devices with exceptionally smaller size and extraordinary performance is strong inspiration for the investigations and exploring the molecular based information processing. This field has been extensively studied and developed from the simple molecular switch to produce more complex molecular system that were able to perform logic function and various arithmetic operations also. Although preliminary reports on the molecular logic gates have attracted considerable interests and enthusiasm amongst the researcher and scientists, this research area is still in its early stage in term of the practical applications. Thus, it is significant to investigate the prospective of developing new molecular entities their means and methods to integrate with the various practical devices.

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