

A Multifunctional Arithmetical Processor Model Integrated Inside a Single Molecule

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Improving the processing power of molecules remains the challenge for molecular logic and computation. Here we report a 2-phenylimidazo[4,5-*f*][1,10]phenanthroline (PIPH)-based three-state molecular switch by controlling its unique emission and absorption spectra in the acid and base condition. On one hand, PIPH can perform simultaneously the functions of an “AND” gate and an “XOR” gate, capable of operating as a half-adder, and the “off–on–off” function as well as comparison function by monitoring its fluorescent spectral changes. On the other hand, the molecule can also implement in parallel the functions of an “XOR” gate and two “INH” gates by monitoring its absorption spectral changes, which constructs two half-subtractors. The cooperative operation of comparator and half-subtractor makes general subtraction operation become possible, which is discussed conceptually in the report.

Introduction

To construct future molecular computers, the mimicry of some elementary functions of neurons and electronic computers at the molecular scale has been attracting more and more interest from chemist community,¹ since de Silva and co-workers published their pioneering paper about molecular AND logic gates.² Subsequently, other logic functions, such as OR,³ XOR,⁴ XNOR,⁵ NAND,⁶ and INHIBIT⁷ have been explored extensively.⁸ More recently, some complicated molecular logic functions such as addition^{9,10} and subtraction^{10,11} involving the simultaneous operations of some logic gates have also been reported. The devices implementing the above elementary binary addition and subtraction are called half-adder and half-subtractor, respectively. It is noteworthy that the molecular arithmetic processor developed by Shanzer and co-workers¹⁰ is capable of both elementary binary addition and subtraction by one molecular species, which was thought to be a significant improvement in enhancing the computing power of molecules.¹² However, the relative magnitudes of minuend and subtrahend need to be accounted for during performing subtraction algebraic operation, so subtraction operations based on half-subtractor are more complicated than addition operations based on half-adder. To get a correct result of subtraction operation, the relative magnitudes of subtrahend and minuend are first compared by a comparator, then the small one is subtracted from the big one by half-subtractor, and finally sign is added to the difference according to the comparing result.^{13,14} That is to say, a comparator is another important device for subtraction operations in addition to half-subtractor.

Many life processes, such as chemical synapses of neurons, can operate normally within a very narrow ion window such as Ca^{2+} and H^+ . This function or behavior can be described as being “off–on–off” or “on–off–on” behavior as a function of ion concentration. Mimicry to this behavior at the molecular scale has been possible for some time with Ca^{2+} , H^+ , or laser as inputs.¹⁵ These studies are highly important not only for the

understanding of complex mechanisms of important life processes but also for the potential application in molecular computing.

We were interested in exploring a single molecular species to execute distinct algebraic operations, even to mimic some elementary functions of neurons by a variety of chemical inputs. While we were preparing for this manuscript, Shanzer et al.¹⁶ reported a molecular arithmetic system based on a plain fluorescein dye, capable of performing a full scale of elementary addition and subtraction algebraic operations. We were prompted to disclose our result because it is distinctly different in the processing functions. The reported system¹⁶ emphasized the reset capability of performing simultaneously the elementary binary addition and subtraction operations with single fluorescein molecule solely in the absorption mode. However, the outputs of the half-adder and half-subtractor in the same mode could interfere with each other. In the present study, a simple phenanthroline derivative, i.e., 2-phenylimidazo[4,5-*f*][1,10]phenanthroline (PIPH), can not only combine addition and subtraction functions with different output modes inside the single molecule but also implement the “off–on–off” function as well as comparison function by different chemical inputs. Meanwhile, the comparator and half-subtractor can be operated simultaneously for general subtraction operations. We believe that the present investigation could provide an excellent model for a multifunctional arithmetical processor integrated inside a single molecule.

Experimental Section

Analytical-grade *N,N*-dimethylformamide (DMF) was dried over calcium hydride for 2 days and then distilled under a reduced pressure to give the anhydrous solvent prior to use. Tetrabutylammonium fluoride ($[(\text{Bu})_4\text{N}]\text{F}$), tetrabutylammonium hydroxide ($[(\text{Bu})_4\text{N}]\text{OH}$), perchloric acid (HClO_4), trifluoroacetic acid (CF_3COOH), and tributylamine (Bu_3N) were purchased from Aldrich and used as received. 2-Phenylimidazo[4,5-*f*][1,10]phenanthroline (PIPH)²³ was prepared according to the literature procedures. Fluorescence spectra were measured in a conventional quartz cell ($10 \times 10 \times 45$ mm) at 25 °C on a

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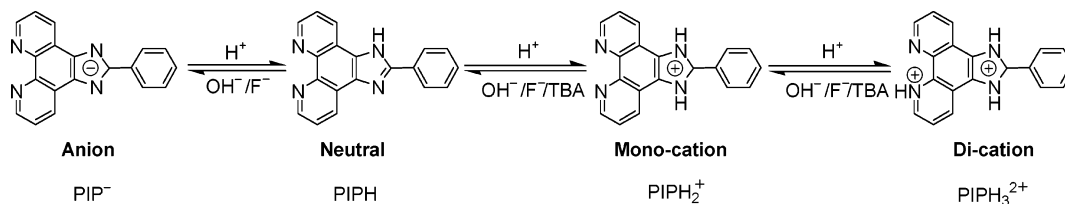


Figure 1. Schematic representation of the interconversion of PIPH among the four ionization states in the presence of acid and base.

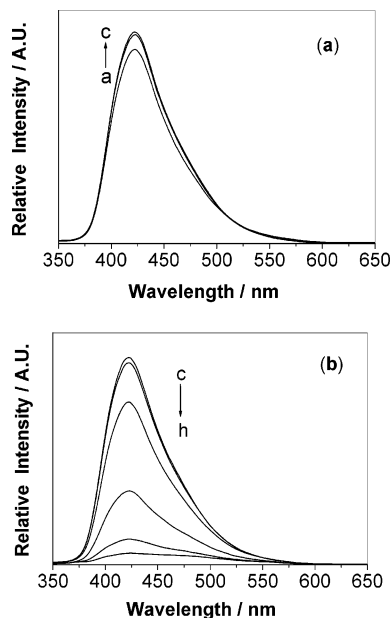


Figure 2. Relative fluorescence intensities of PIPH (0.01 mM) in DMF solution upon addition of (a) 0, 0.5, 1 equivalent HClO_4 (from a to c), and (b) 1, 2, 6, 10, 14, 20 equivalent HClO_4 (from c to h).

JASCO FP-750 spectrometer with the excitation and emission slits of 5 nm width. The sample solutions at a PIPH concentration of $1 \times 10^{-5} \text{ mol} \cdot \text{dm}^{-3}$ were excited at 335 nm.

Results and Discussion

Spectral Properties of PIPH. PIPH is a good transition metal ligand, but its photophysical behavior in the acid and/or base condition is not fully studied. In this paper, we first investigated the spectral properties of PIPH in DMF, indicating that the compound can exist in four ionization forms (dication, cation, neutral, and anion) in the different acid/base conditions, as shown in Figure 1.

In DMF solution, PIPH has the highest emission peak at 423 nm when excited at 335 nm. When 1 equivalent HClO_4 was added to the DMF solution of PIPH, its relative emission intensity increased (Figure 2a), but the addition of a further amount of HClO_4 led to emission quenching (Figure 2b). In light of the protonation abilities of phenanthroline ($\text{p}K_a(\text{PH}^+) = 4.65$, $\text{p}K_a(\text{PH}_2^{2+}) = -6.5$),¹⁷ and imidazole ($\text{p}K_a = 6.993$)¹⁸ in aqueous solution, we could deduce reasonably that there should be the two protonation forms of cation and dication in the different acidity. In the case of 1 or less equivalent acid, the nitrogen atom of imidazole in PIPH is first protonated. When excited by UV light, the poorly emissive $n-\pi^*$ transition is replaced by the strongly emissive $\pi-\pi^*$ state upon the protonation. This may be responsive for the increase of emission intensity.¹⁹ In the presence of more than 1 equivalent acid, one of the nitrogen atoms of phenanthroline is also protonated, and therefore charge and energy transfer between protonated phenanthroline and protonated imidazole could lead to the quenching of fluorescence. The same phenomena were observed when

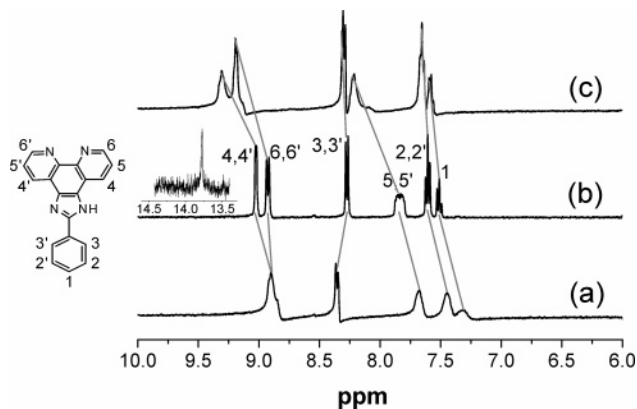


Figure 3. ^1H NMR (400 MHz) spectra of PIPH (0.01 M) in $\text{DMSO}-d_6$ in (b) the absence and the presence of (a) 5.0 equivalents of $[(\text{Bu})_4\text{N}]\text{F}$, and (c) 5.0 equivalents of CF_3COOH .

replacing HClO_4 with CF_3COOH . The NMR experiments show that the resonance signals on its phenanthroline moiety shift more downfield than those on its phenyl rings after the addition of 5 molar equivalents of CF_3COOH (Figure 3), which also confirms the protonation process.

In sharp contrast, the addition of $[(\text{Bu})_4\text{N}]\text{F}$ to the DMF solution of PIPH results in a considerable red shift for both its emission spectra (from 423 to 513 nm) and its absorption spectra. These observations should be attributed to the deprotonation of the NH group of the imidazole ring in PIPH, leading to the formation of anion PIP^- .²⁰ Comparing with the ^1H NMR spectra of PIPH in the absence and presence of F^- (Figure 3), we notice that its N–H signal disappears and the other proton signals shift upfield, except for the two $\text{H}_{3,3'}$ protons. The observation is well consistent with the results of the literature,²⁰ suggesting the formation of a hydrogen-bond complex between the fluoride ion and the NH group at low fluoride concentration, and the formation of the deprotonated PIPH (PIP^-) at excess fluoride. This deprotonation process is also confirmed by the identical fluorescence spectral changes observed in the titration experiment with $[(\text{Bu})_4\text{N}]\text{OH}$ and Bu_3N as that of with fluoride ions. It is known that $[(\text{Bu})_4\text{N}]\text{OH}$ is a stronger organic base than $[(\text{Bu})_4\text{N}]\text{F}$, and Bu_3N is the weakest among them. The addition of a little $[(\text{Bu})_4\text{N}]\text{OH}$ to the DMF solution of PIPH abruptly decreases its emission intensity at 423 nm, but Bu_3N cannot change its emission and absorption spectra.

Half-Adder and Half-Subtractor. Based on these distinct fluorescence properties of PIPH among the three states PIPH , PIP^- , and PIPH_3^{2+} , PIPH can operate within the molecule as not only the adjustable fluorescent “off–on–off”/“off–on” switches, but also a half-adder by acid/base reaction. In the presence of 3 mM CF_3COOH , 0.01 mM PIPH is the dicationic form and is nonfluorescent. Upon the continuous addition of any of $[(\text{Bu})_4\text{N}]\text{OH}$, $[(\text{Bu})_4\text{N}]\text{F}$, or Bu_3N to the solution, the base and acid neutralize first and the fluorescence of PIPH at 423 nm is gradually revived to its maximum until 3 mM base is added (Figure 4). Subsequently, the cases are distinctly different: its emission intensity at 423 nm (a) abruptly reduces

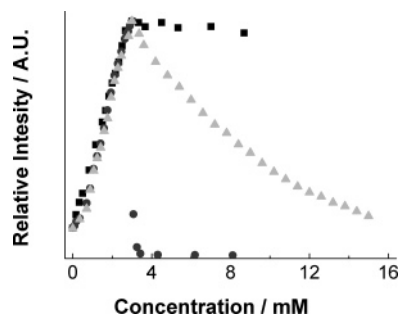
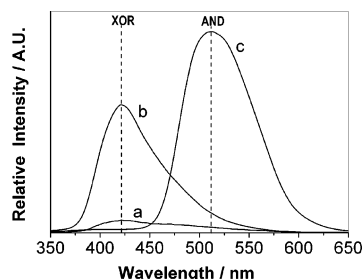


Figure 4. Adjustable “off–on–off” behaviors of PIPH_3^{2+} (0.01 mM) with addition of different bases. $[(\text{Bu})_4\text{N}]\text{OH}$ (●, black), $[(\text{Bu})_4\text{N}]\text{F}$ (▲, gray), and Bu_3N (■, black).

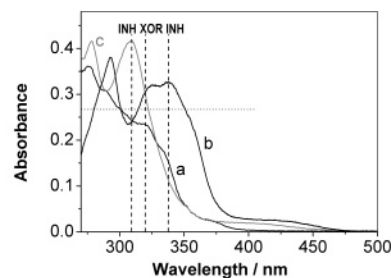


Input		Output		Half-Adder (C/S)
A	B	XOR 423 nm (Sum)	AND 513 nm (Carry)	A + B 513/423 nm
0	0	0 (low, 6)	0 (low, 3)	00
1	0	1 (high, 63)	0 (low, 9)	01
0	1	1 (high, 63)	0 (low, 9)	01
1	1	0 (low, 2)	1 (high, 100)	10

Figure 5. Molecule logic gates for the half-adder. Top: the fluorescence spectra of PIPH (0.01 mM) in DMF solution in the presence of (a) 3 mM CF_3COOH , (b) one of chemical inputs, and (c) two chemical inputs when excited at 335 nm, and the resulting XOR gate (423 nm) and AND gate (513 nm) for the half-adder. Bottom: the truth table for the operation of the molecular half-adder solely in the fluorescence mode. Input: (A) 3 mM $[(\text{Bu})_4\text{N}]\text{OH}$; (B) 3 mM $[(\text{Bu})_4\text{N}]\text{F}$ or $[(\text{Bu})_4\text{N}]\text{OH}$.

with the addition of a little $[(\text{Bu})_4\text{N}]\text{OH}$; (b) gradually decreases with the addition of $[(\text{Bu})_4\text{N}]\text{F}$; (c) does not change with the addition of Bu_3N . The adjustable molecular “off–on–(off)” fluorescent switches by inputting different species are similar to the chemical synapse of neurons, which can operate normally within a very narrow ion window.

Under the same starting conditions as the above “off–on–(off)” switches, this system can operate simultaneously as AND and XOR logic gates if two identical $[(\text{Bu})_4\text{N}]\text{OH}$ (3 mM) or one $[(\text{Bu})_4\text{N}]\text{OH}$ (3 mM) and one $[(\text{Bu})_4\text{N}]\text{F}$ (3 mM) are respectively regarded as inputs. Addition of one of inputs causes nonfluorescent PIPH_3^{2+} convert to neutral PIPH with the blue emission at 423 nm, while simultaneous addition of two inputs results in the formation of PIP^- , with the blue emission at 423 nm disappearing and the green emission at 513 nm occurring. Hence, a characteristic AND logic gate is obtained when the output is read at 513 nm (carry digit), and a XOR logic gate is also achieved when the output is read at 423 nm (sum digit), as shown in Figure 5. This result corresponds to the half-adder function. Herein, the use of identical species as two inputs is not the first example.^{15d} It is worth noting that the replacement of the two inputs by two identical $[(\text{Bu})_4\text{N}]\text{F}$ fails to operate a half-adder logic function, which is attributed to the weaker basicity of $[(\text{Bu})_4\text{N}]\text{F}$ than that of $[(\text{Bu})_4\text{N}]\text{OH}$.



Input		Output			Half-Subtractor (B/D)	
B	C	INH 310 nm (B)	XOR 320 nm (D)	INH 340 nm (B)	B–C 310/320 nm	C–B 340/320 nm
0	0	0 (low, 0.24)	0 (low, 0.23)	0 (low, 0.13)	00	00
0	1	1 (high, 0.42)	1 (high, 0.30)	0 (low, 0.10)	11	01
1	0	0 (low, 0.24)	1 (high, 0.30)	1 (high, 0.32)	01	11
1	1	0 (low, 0.24)	0 (low, 0.23)	0 (low, 0.13)	00	00

Figure 6. Molecule logic gates for the half-subtractor. Top: the absorption spectra of PIPH (0.01 mM) in DMF solution in (a) the absence and (b) the presence of (b) the input (B), and (c) the input (C), and the resulting XOR gate (320 nm) and INH gates (310 and 340 nm) for the half-subtractor. Bottom: the truth table for the operation of the molecular half-subtractor with the function exchanging the order of minuend and subtrahend in the absorption mode. Input: (B) 3 mM $[(\text{Bu})_4\text{N}]\text{F}$ or $[(\text{Bu})_4\text{N}]\text{OH}$; (C) 3 mM HClO_4 or CF_3COOH . B: Borrow; D: Difference.

Starting with neutral form and monitoring changes of absorption as outputs, this molecule can operate simultaneously as INHIBIT (INH) and XOR logic gates when acid (3 mM HClO_4 or 3 mM CF_3COOH) and base (3 mM $[(\text{Bu})_4\text{N}]\text{OH}$ or 3 mM $[(\text{Bu})_4\text{N}]\text{F}$) are respectively used as inputs. With no addition of inputs, all outputs fall below the threshold value (<0.27). Addition of acid results in a dicationic form with high absorption values at 310 and 320 nm, and a low absorption value at 340 nm. When base is added, a monoanionic state is formed with high output values at 340 and 320 nm, and a low output value at 310 nm. When input simultaneously, acid and base neutralize each other and the absorption spectra of the neutral form is observed. Now, a XOR logic gate is obtained for generating the difference digit of half-subtractor when the output is read at 320 nm, and two INH logic gates are achieved for the borrow of half-subtractor when the output is read at 310 and 340 nm, respectively, as shown in Figure 6. The corresponding electronic symbols of molecular half-adder and half-subtractor are illustrated in Figure 7. Because there are two INH gates with different response to the inputs, the order of minuend and subtrahend can be exchanged according to requirement. When the INH gate at 310 nm is combined with the XOR gate, the $B - C$ algebraic operation is executed by the molecular half-subtractor. While using the INH gate at 340 nm, the $C - B$ algebraic operation is performed. This will be very useful in the subtraction operation described below.

Comparator Function. Digital comparators are used to compare the magnitude of two binary quantities and determine the relationship of those quantities. There are two basic types of digital comparators: identity comparators and magnitude comparators.²¹ Identity comparators indicate whether two inputs are equal. The XOR gate is actually an identity comparator.^{4a} Magnitude comparators indicate whether two inputs are equal and, if they are not, which input is larger or smaller. The truth table and the symbol for a magnitude comparator are shown in Figure 8. If $B > C$, i.e., the comparison is “greater than”, then output gt (greater than) is set to 1; if the comparison is “equal”

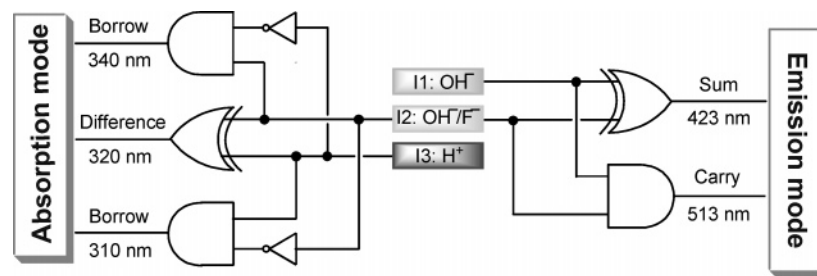


Figure 7. The corresponding electronic symbols of molecular half-adder and half-subtractor.

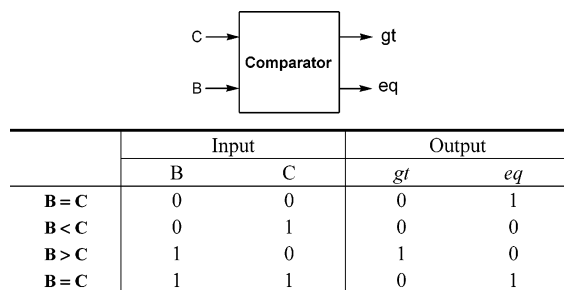


Figure 8. The symbol (top) and truth table (bottom) for single-bit magnitude comparator.

TABLE 1: Truth Table for the Operation of the PIPH-based Digital Comparator in the Emission Mode^a

	input ^b		output	
	B	C	gt 513 nm	eq 423 nm
B = C	0	0	0 (low, 9)	1 (high, 63)
B < C	0	1	0 (low, 3)	0 (low, 6)
B > C	1	0	1 (high, 100)	0 (low, 2)
B = C	1	1	0 (low, 9)	1 (high, 63)

^a Starting conditions: the neutral form of PIPH (0.01 mM) in DMF solution and excited at 335 nm. ^b Input: (B) 3 mM [(Bu)₄N]F or [(Bu)₄N]OH; (C) 3 mM HClO₄ or CF₃COOH.

(B = C), then output *eq* (equal) is set to 1; if B < C, then both outputs are set to 0.²²

Based on the same starting solution and inputs as half-subtractor and monitoring changes of emission as outputs such as half-adder, that is, line b in Figure 5, a molecular digital comparator can now be constructed. With no or both inputs present, B = C, a blue emission is observed and the output *eq* is 1. Addition of base results in monoanionic species, B > C, the solution shows a green emission, and output *gt* = 1. When adding acid, B < C, dicationic form is obtained and the luminescence is quenched completely, both outputs are 0. The corresponding truth table is shown in Table 1.

The molecular comparator and half-subtractor have the same starting conditions and inputs as well as different outputs, so they can be implemented cooperatively without mutual interference. This is favorable for the realization of subtraction operation. Now let us describe the process of subtraction operation based on the simple molecular system for the four input possibilities. Assume expecting to obtain the difference of B - C, we need to compare first the relative magnitudes of B and C by a comparator. If the output *eq* of the comparator is 1, then B = C, and there is no need of subtraction and B - C = 0; else if the output *gt* is 1, then B > C. Here, the half-subtractor integrated from the INH gate at 310 nm and the XOR gate is used to perform B - C algebraic operation, and B - C = 1; else both *gt* and *eq* are 0, then B < C. Here, the half-subtractor integrated from the INH gate at 340 nm and the XOR gate is chosen to execute C - B algebraic operation for

preventing that the bigger number is subtracted from the smaller one, and C - B = 1. Finally, B - C = -1 is obtained by adding the minus sign to the difference according to the output of comparator. It should be emphasized that the comparison and subtraction operation are practically implemented in parallel in this molecular system, and all results are collected simultaneously, which would improve the efficiency of the molecular logic circuit. In addition, the present system can also be reset a few times with acid and base for the operation of both half-adder/half-subtractor and comparator.

Conclusion

The spectral properties of 2-phenylimidazo[4,5-f][1,10]-phenanthroline (PIPH) in the acid and base condition have been investigated by fluorescence and UV-vis spectrometry. The obtained results indicate that PIPH can exist in four ionization forms of dication, cation, neutral, and anion, and three of them have distinct emission and absorption properties. Furthermore, a three-state molecular switch, capable of operating not only as a reversible half-adder and a half-subtractor but also as an adjustable “off-on-off” switch and a comparator, is obtained by controlling the unique emission and absorption spectra of PIPH. The adjustable “off-on-off” behavior maybe mimic some elementary properties of neurons and improve our understanding on the chemical basis of biologic process. The differences of the output modes between the half-adder and the half-subtractor could prevent the interference of output signal, and the combination of comparator and half-subtractor can provide a feasible arithmetic for general subtraction operation. Undoubtedly, these functions integrated inside a single molecule would enhance effectively the processing power of molecules as arithmetical processors.

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