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RESEARCH ARTICLE

Unimolecular half-adders and half-subtractors based on acid-base reaction

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According to the structural analysis of reported molecular processors with acids and bases as inputs, we proposed a general method for constructing molecular half-adders and/or half-subtractors based on acid-base reaction. The method is preliminarily supported by four molecular processors (8-hydroxyquinoline, 4-hydroxypyridine, 4-aminophenol and 5-amino-1-naphthol) capable of the elementary addition and/or subtraction algebraic operations. Noticeably, 8-hydroxyquinoline can mimic the functions of three logic devices, i.e. half-adder, half-subtractor and digital comparator, by the use of superposition and reconfiguration. The method described in this paper may be useful not only for designing new unimolecular arithmetical processors with the same inputs and outputs as standard devices for the construction of future molecular computers, but it can also help us disclose the simplest molecules and biomolecules with computational properties concealed around us.

Keywords logic gate, molecular processor, half-adder, half-subtractor, molecular device

1 Introduction

Since de Silva and coworkers published their pioneering work of molecular AND gate [1], molecules capable of logic functions have been attracting more and more interest in the community of chemists [2–4]. Most of the simple logic gates are now accessible at molecular scale [4]. They were even combined to provide advanced logic devices, such as molecular adder [5–16], subtractor [12–20], comparator [15,19], multiplexer [21,22], and keypad lock [23,24]. However, the interconnection between different logic devices for creating more complex logic circuits still remains as one of the biggest challenges in this field since the reported logic molecules are distinct with respect to their structures, inputs, outputs and operation environments. One approach in circumventing this problem is to integrate multiple logic functions inside single molecules by the use of superposition and reconfiguration [25]. In this respect, structurally simple compounds with several logic functions are especially desirable for extendable functions through structural modification and interconnection. On the other hand, the standardization of logic devices, i.e. using the same inputs and outputs, is also beneficial for parallel operation and even interconnection. Therefore, a series of single molecules capable of complex logic functions with the same inputs, outputs, and operation environments will be good candidates as standard devices for achieving more complex logic circuits.

Among the available unimolecular half-adders and half-subtractors, those based on acid-base inputs and optical outputs have attracted special attention for their ease in manipulating, monitoring and resetting. These logic molecules can change reversibly between three or more spectral states as the pH value of their solution changes. Such a property is important for achieving molecular logic functions. Herein we describe four simple molecules, i.e. 8-hydroxyquinoline (8-HQ), 4-hydroxypyridine (4-HP), 4-aminophenol (4-AP) and 5-amino-1-naphthol (5-AN), capable of performing elementary addition and/or subtraction algebraic operations based on acid-base reactions. It is of particular interest to us to unravel the underlying rules for constructing molecular half-adders and/or half-subtractors based on acid-base reactions. Such rules may be highly useful not only for designing new unimolecular arithmetical processors with the same inputs and outputs as standard devices for the construction of more complex logic circuits, but may also help us to disclose the simplest molecules and biomolecules with computational properties concealed around us [13].

The truth table for the logic circuits of half-adders and half-subtractors is shown in Table 1.

Table 1 Truth table for the logic circuits of half-adder and half-subtractor

input		output			
A	B	carry	sum	borrow	difference
0	0	0	0	0	0
0	1	0	1	1	1
1	0	0	1	0	1
1	1	1	0	0	0

2 Experimental section

Analytical-grade CH_3CN in the presence of K_2CO_3 and $KMnO_4$ was refluxed for 10 hours and then distilled. The distilled CH_3CN was then dried over P_2O_5 for one day and then distilled to provide pure solvent for spectral measurement. 8-HydroxyQuinoline, 4-hydroxyPyridine,

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4-aminoPhenol, 5-amino-1-naphthol, $[(Bu)_4N]OH$ and CH_3SO_3H were purchased from Aldrich and used as received. Fluorescence spectra were measured in a conventional quartz cell (10 mm × 10 mm × 45 mm) at 25°C on a JASCO FP-750 spectrometer with excitation and emission slits of 5 nm width.

3 Results and discussion

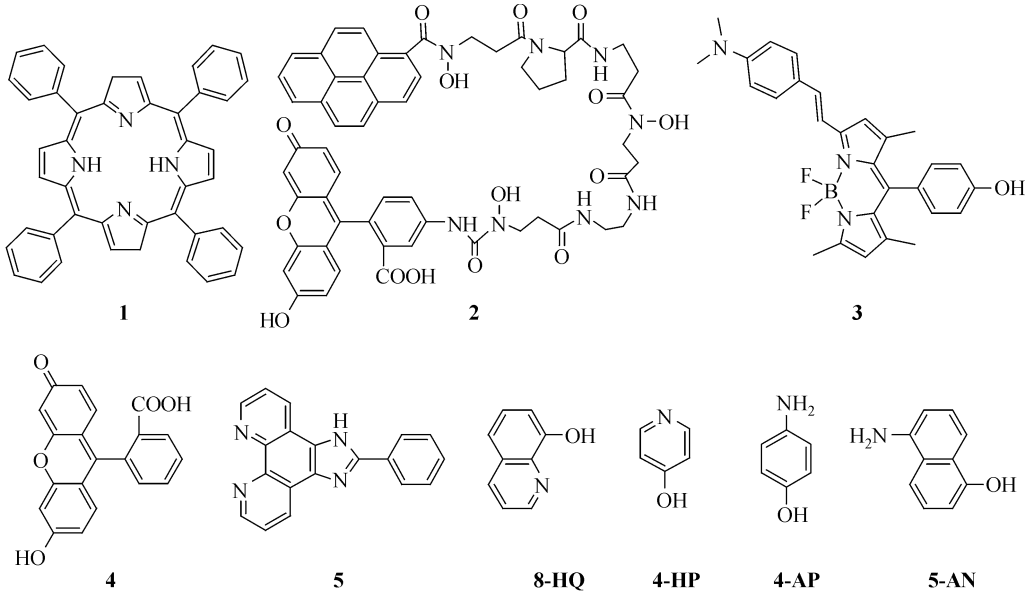
3.1 Design concept

Compounds **1-5**, as shown in Scheme 1, have been demonstrated to operate effectively as half-adders and/or half-subtractors based on acid-base reactions [12–15,17,18]. The acidic groups in **1-5** include –OH, –NH– (sp^3 hybrid, endo-nitrogen) and –COOH, and the basic groups –N = (sp^2 hybrid, endo-nitrogen) and $–NR_2$. These acidic and basic groups are responsible for the spectral state change of compounds **1-5** when the pH values of their solutions are changed. Therefore, these acidic and basic groups may also be the characteristic groups of other logic molecules based on the acid-base reaction. The theoretical analysis about the effectiveness of these groups is described as follows.

The deprotonated forms of –OH and –NH– ($–O^-$ and $–N^–$) are stronger electron donors than –OH and –NH–. Compared with neutral species, the deprotonated groups increase the HOMO energy level, and thus diminish the energy gap between HOMO and LUMO. From the photochemical viewpoint, this will enhance the internal charge transfer (ICT) process and likely result in the bathochromic shift of electronic absorption and emission spectra. This theory is supported by the spectra of compounds **1**, **2**, **4** and **5** under the basic condition. The spectral change caused by the deprotonation of the –COOH group in fluorescein is mainly due to the existence of the lactone form. This phenomenon is not observed for common aromatic compounds with carboxyl groups. Therefore, the –COOH group could be excluded as a candidate for an effective acidic group to achieve obvious spectral state change.

When –N = is protonated, the LUMO and HOMO energy levels of the molecule lowers, which could change the energy gap between HOMO and LUMO and then lead to the shift of the electronic absorption spectra and changes in the emission spectra. This is confirmed by the spectral change of compound **1** and **5** when adjusting the pH to an acidic condition. Considering the $–NR_2$ group, the protonation changes its electronic property from the original electron donor to the electron acceptor, resulting in the blue shift of electronic absorption spectra and emission spectra as clearly seen in the case of compound **3**.

On the other hand, the emission spectra could also be influenced by the protonation or deprotonation of the four groups, –OH, –NH–, –N =, and $–NR_2$ through photo-induced electron transfer (PET) and the ICT process [18]. Consequently, it is reasonable to infer that the molecules conjugated with two or more of –OH, –NH–, –N =, and $–NR_2$ can change among several spectral states when the pH values change and they can thus be considered as candidates for molecular processors with acid and base as inputs. Four simple molecules, 8-HQ, 4-HP, 4-AP and 5-AN (see Scheme 1) are selected to test this hypothesis. These molecules all contain two of –OH, –NH–, –N =, and the $–NR_2$ (R = H) groups and can change reversibly among three distinct spectral states

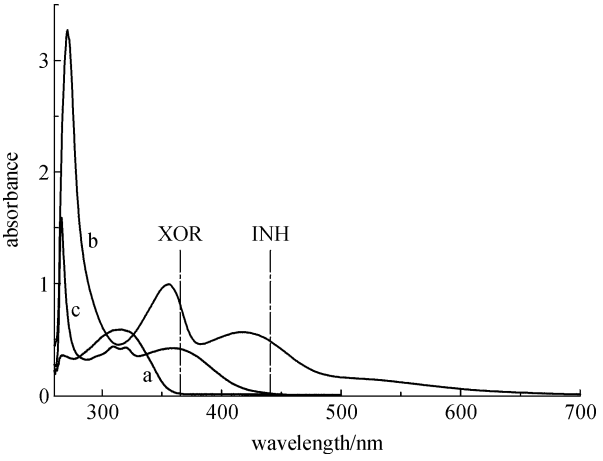


Scheme 1 The molecular structures of molecular processors.

upon the addition of acid and base, which are then applied to demonstrate their logic operations as a half-adder and/or half-subtractor.

3.2 Logic functions of 8-HQ

8-HQ, which contains –OH and –N = groups, is famous for its application in organic light-emitting diode (OLED) devices [26]. However, the logic functions concealed in 8-HQ have not been well explored [22]. With the addition of acid or base, 8-HQ can convert among three ionization forms accompanying three distinct electronic absorption spectra. Compared with 8-HQ under neutral conditions, the absorption spectra of 8-HQ under the acidic and basic conditions are both shifted to long wavelengths due to the enhanced ICT process in the charged forms as discussed above. The bathochromic shift under the basic condition is more obvious than that under the acidic condition, probably because of the stronger ICT process in the deprotonated species of 8-HQ (see the spectra in Figure 1). While we monitor the absorbance changes of 8-HQ at 375 nm and 425 nm as the outputs, with acid and base as two inputs, a molecular processor could be constructed starting with the solution of neutral 8-HQ. When observed at 375 nm, the absorbance is high (coded for binary 1) with the input of either acid or base. The simultaneous inputs of acid and base annihilate each other and generate a low absorbance (coded for binary 0) at 375 nm. These behaviors correlate very well with the XOR logic function. Additionally, the absorbance at 425 nm is high (coded for binary 1) with the input of base only. For the other three combinations of the two inputs, the absorbance is low (coded for binary 0). Therefore, an INHIBIT (INH) logic gate is established when observing the absorbance changes at 425 nm. The parallel operation of these XOR and INH gates thus gives rise to a half-subtractor (see the table in Figure 1). Noteworthily, the highest absorbance coded for binary 0 is only 0.04 while the lowest absorbance coded for binary 1 is 0.37. The difference is remarkable among molecular half-adders or half-subtractors reported solely in the absorption mode, and significantly improved signal/noise ratio of the resulting half-subtractor.

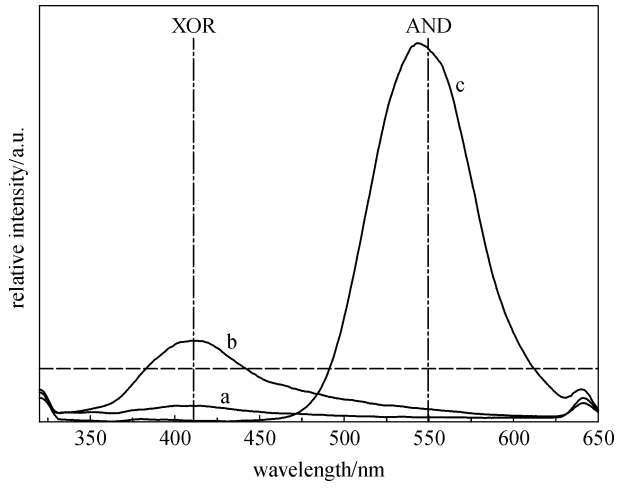


input		output		half-subtractor (B/D)
B	C	XOR 375 nm (Difference)	INH 425 nm (Borrow)	C–B 425/375 nm
0	0	0 (low, 0.01)	0 (low, 0.01)	00
1	0	1 (high, 0.51)	1 (high, 0.55)	11
0	1	1 (high, 0.37)	0 (low, 0.04)	01
1	1	0 (low, 0.01)	0 (low, 0.01)	00

Figure 1 Molecular logic gates of the half-subtractor. Top: the absorption spectra of 8-HQ (0.2 mmol/L) in CH_3CN solution in (a) the absence and (b) the presence of the input (B) or (c) the input (C), and the resulting XOR gate (375 nm) and INH gate (425 nm) for the half-subtractor. The output threshold value is 0.25. Bottom: the truth table for the operation of the molecular half-subtractor solely in the absorption mode. Input: (B) 16 equiv. $[(Bu)_4N]OH$; (C) 16 equiv. CH_3SO_3H.

It is well known that the fluorescence of 8-HQ in solution will be quenched due to the excited-state intramolecular proton transfer [27], but it still can show weak emission in dry CH_3CN [28]. As mentioned above, the addition of acid and base will lead to the bathochromic shift in the absorption spectra due to the enhanced ICT process. The improved ICT process usually leads to the enhancement and the bathochromic shift of the emission spectra, too. Surprisingly, the addition of acid just quenches the emission of the fluorescence with no obvious shift (Figure 2 line a). This phenomenon is presumably also due to the excited-state proton transfer. According to the results of neutral 8-HQ [27], the intramolecular hydrogen bond may still exist between NH^+ and the oxygen. Excitation could change the acidity of neutral 8-HQ and thus transfer protons to surrounding water molecules [27]. Protonated 8-HQ could be more acidic than 8-HQ in the excited states and likely result in proton transfer to residual water in CH_3CN. This process may consume the excited-state energy and quench the fluorescence. However, with the addition of bases, the relative emission intensity was enhanced, accompanying the bathochromic shift from 410 nm to 550 nm at the maximum. Starting with the protonated 8-HQ with two identical bases as two inputs, a molecular half-adder can also be achieved by monitoring the fluorescence changes at 410 nm and 550 nm. As shown in the table in Figure 2, with the changes of the combinations of these two inputs, the output at 410 nm is high only in the presence of one input, and the behavior of the absorption changes corresponds to a XOR logic function. Similarly, an AND logic gate is achieved when observing at 550 nm. In binary addition, the XOR gate output

is the sum digit and the AND gate output is the carry digit. The combination of the above XOR and AND gates can mimic the function of a half-adder (Figure 2).



input		output		half-adder (C/S)
A	B	XOR 410 nm (Sum)	AND 550 nm (Carry)	A + B 550/410 nm
0	0	0 (low, 3)	0 (low, 0)	00
1	0	1 (high, 21)	0 (low, 2)	01
0	1	1 (high, 21)	0 (low, 2)	01
1	1	0 (low, 0)	1 (high, 98)	10

Figure 2 Molecular logic gates for the half-adder. Top: the fluorescence spectra of 8-HQ (0.2 mmol/L) in CH_3CN solution excited at 320 nm in the presence of (a) 16 equiv. CH_3SO_3H, (b) one of chemical inputs (A or B), and (c) two chemical inputs (A and B), and the resulting XOR gate (410 nm) and AND gate (550 nm) for the half-adder. The output threshold value is 15. Bottom: the truth table for the operation of the molecular half-adder solely in the fluorescence mode. Input: (A) 16 equiv. $[(Bu)_4N]OH$; (B) 16 equiv. $[(Bu)_4N]OH$.

Additionally, 8-HQ can also mimic the function of digital comparators, which can help the subtractor implement general subtraction. The first molecular comparator model and the general subtraction at the molecule-scale have been reported by our group [15]. Starting from the neutral species with base (B) and acid (C) as two inputs, the fluorescent output at 410 nm is *eq* (equal) digit and the output at 550 nm is *gt* (greater than) digit. With no or both inputs present, i.e. when B = C, the output *eq* is 1. When adding the base, i.e. B > C, the output *gt* is 1. The addition of an acid, i.e. B < C, leads to the quenching of the complete emission of 8-HQ, and both outputs are 0. The corresponding truth table for the molecular digital comparator is shown in Table 2. Because the half-subtractor and the digital comparator inside 8-HQ have the same starting condition and inputs but different output modes, general subtraction can also be achieved by the combination of the half-subtractor and the digital comparator through the method reported in our previous paper [15]. In summary, the addition, subtraction and comparison functions have been integrated inside 8-HQ, a simple molecule with –OH and –N = groups.

Table 2 The truth table for the operation of the 8-HQ-based digital comparator in the emission mode.[a] The output threshold value is the same as that of the 8-HQ-based half-adder

	input [b]		output	
	B	C	*eq* 410 nm	*gt* 550 nm
B = C	0	0	1 (high, 21)	0 (low, 2)
B < C	0	1	0 (low, 3)	0 (low, 0)
B > C	1	0	0 (low, 0)	1 (high, 98)
B = C	1	1	1 (high, 21)	0 (low, 2)

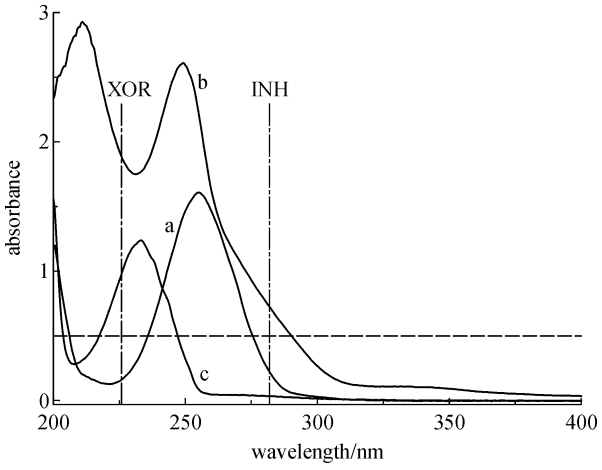
a Starting conditions: the neutral form of 8-HQ (0.2 mmol/L) in CH_3CN solution excited at 320 nm.

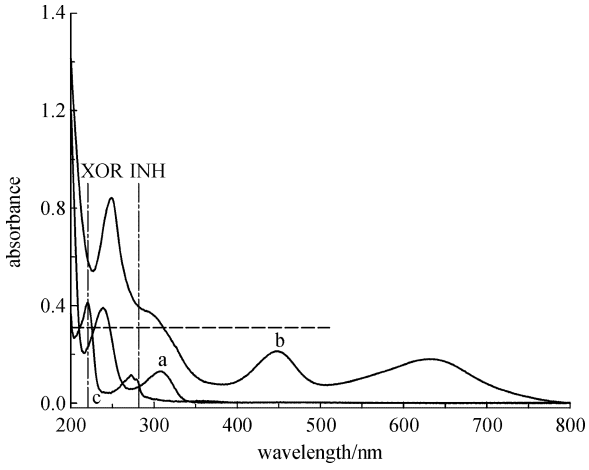
b Input: (B) 16 equiv. $[(Bu)_4N]OH$; (C) 16 equiv. CH_3SO_3H.

3.3 Logic functions of 4-HP, 4-AP and 5-AN

The success with 8-HQ motivated us to further explore along this line. To further miniaturize the volume of molecular logic devices, smaller aromatic systems could be coupled to the functional groups mentioned above. For example, 4-HP contains the same functional groups as 8-HQ but smaller aromatic systems. Monitoring the absorbency change at 225 nm and 280 nm as outputs with neutral 4-HP as starting species and acid and base as inputs, a half-subtractor combining XOR and INH gate is generated (Figure 3). Unfortunately, 4-HP has no fluorescence. Consequently, its output is only in absorption mode. It is worth emphasizing that the molecular weight of 4-HP is the lowest one among all molecular half-adders and half-subtractors that have been reported until now. Additionally, besides 8-HQ and 4-HP, the [2,2′-Bipyridyl]-3,3′-diol reported recently by Das et al. [20] also confirmed the logic characteristics of the molecules with –OH and –N = groups.

4-AP, which also has no fluorescence, is another kind of very simple molecular processor. The –OH and $–NH_2$ groups are incorporated inside 4-AP for executing logic functions with acid and base as inputs. When monitoring the changes of its electronic absorption spectra at the appropriate wavelength, a molecular half-subtractor can be achieved (Figure 4). In contrast, 5-AN with an extended aromatic system shows an emission property. With the neutral form as starting species and two identical bases as inputs, an XOR gate is generated by monitoring the fluorescence at 380 nm as output. Under identical starting conditions and with the identical inputs, one can obtain an AND gate by monitoring the absorbance at





input		output		half-subtractor (*B*/*D*)
A	B	XOR 225 nm (Difference)	INH 280 nm (Borrow)	A–B 280/225 nm
0	0	0 (low, 0.15)	0 (low, 0.29)	00
1	0	1 (high, 0.93)	0 (low, 0.04)	01
0	1	1 (high, 1.93)	1 (high, 0.78)	11
1	1	0 (low, 0.15)	0 (low, 0.29)	00

Figure 3 Molecular logic gates for the half-subtractor. Top: the absorption spectra of 4-HP (0.1 mmol/L) in CH_3CN solution in (a) the absence and the presence of (b) the input (B), and (c) the input (A), and the resulting XOR gate (225 nm) and INH gate (280 nm) for the half-subtractor. The output threshold value is 0.5. Bottom: the truth table for the operation of the molecular half-subtractor solely in the absorption mode. Input: (A) 50 equiv. CH_3SO_3H; (B) 50 equiv. $[(Bu)_4N]OH$.

input		output		half-subtractor (*B*/*D*)
A	B	XOR 220 nm (Difference)	INH 280 nm (Borrow)	A–B 280/220 nm
0	0	0 (low, 0.23)	0 (low, 0.06)	00
1	0	1 (high, 0.41)	0 (low, 0.09)	01
0	1	1 (high, 0.59)	1 (high, 0.40)	11
1	1	0 (low, 0.23)	0 (low, 0.06)	00

Figure 4 Molecular logic gates for the half-subtractor. Top: the absorption spectra of 4-AP (0.44 mmol/L) in CH_3CN solution in (a) the absence and the presence of (b) the input (B), and (c) the input (A), and the resulting XOR gate (220 nm) and INH gate (280 nm) for the half-subtractor. The output threshold value is 0.3. Bottom: the truth table for the operation of the molecular half-subtractor solely in the absorption mode. Input: (A) 18 equiv. CH_3SO_3H; (B) 18 equiv. $[(Bu)_4N]OH$.

255 nm. The combination of the two logic gates constitutes a molecular half-adder (Figure 5).

4 Conclusions

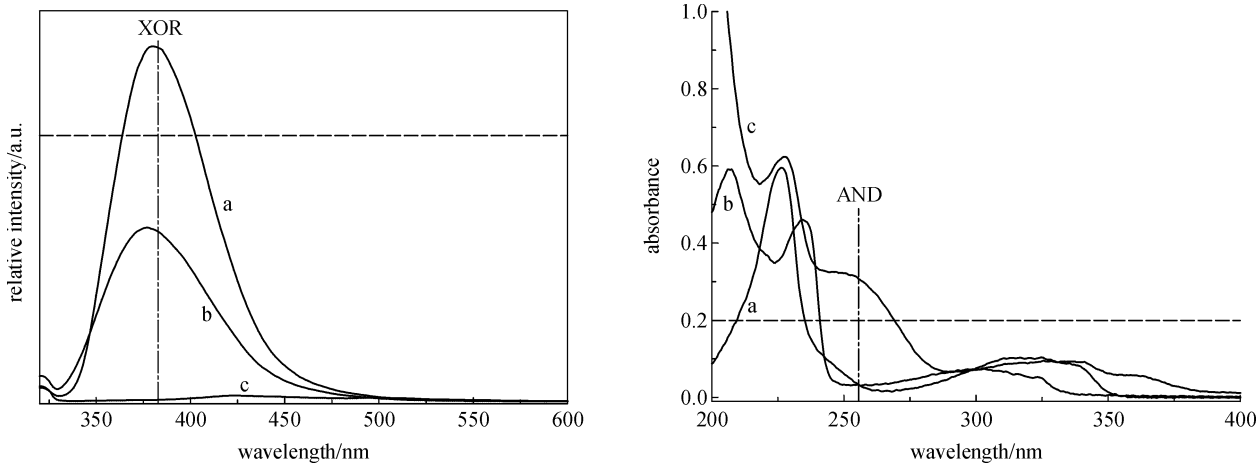
The logic functions and output modes of all single molecular adders and subtractors based on acid-base reaction are summarized in Table 3. The hypothesis that the molecules, conjugated with two or more of –OH, –NH–, –N =, and $–NR_2$, could be considered as candidates for molecular logic devices is supported by these ten logic molecules. Noticeably, inside a single 8-hydroxyquinoline molecule, three advanced logic functions, including a half-adder, half-subtractor and digital comparator, are integrated. Different combinations between the arene matrixes and the four acidic or basic groups could be helpful in designing and disclosing a great diversity of unimolecular arithmetical processors with acid and base as inputs. These processors could be used for integration in future molecular computers. However, more examples are definitely required to evaluate the effectiveness of these four groups as functional groups for constructing molecular processors with acids and bases as inputs.

Table 3 The logic functions and output modes of unimolecular processors based on acid-base reaction

molecules	logic functions	output mode
1	HS	A & E
2	HA & HS	E
3	HS	E
4	HA & HS/FA & FS	A
5	HA & HS	A & E
[2,2′-Bipyridyl]-3,3′-diol	HS	A & E
8-HQ	HA & HS	A & E
4-HP	HS	A
4-AP	HS	A
5-AN	HA	A & E

HA: half-adder; HS: half-subtractor; FA: full-adder; FS: full-subtractor; A: absorption mode; E: emission mode.

In addition, some features and/or perspectives of the unimolecular processors of this genre could be summarized:



input		output		half-adder (*C*/*S*)
A	B	XOR (emission) 380 nm (Sum)	AND (absorption) 255 nm (Carry)	A + B 255 nm/380 nm
0	0	0 (low, 0)	0 (low, 0.03)	00
1	0	1 (high, 100)	0 (low, 0.03)	01
0	1	1 (high, 100)	0 (low, 0.03)	01
1	1	0 (low, 49)	1 (high, 0.31)	10

Figure 5 Molecular logic gates for the half-adder. Top: the emission (right) and absorption (left) spectra of 5-AN (0.01 mmol/L) in CH_3CN solution in the presence of (b) 100 equiv. CH_3SO_3H, (a) one of chemical inputs, and (c) two chemical inputs, and the resulting XOR gate in the emission mode (380 nm) with the output threshold value of 75 and AND gate in the absorption mode (255 nm) with 0.2 as the threshold value, for the half-adder. Bottom: the truth table for the operation of the molecular half-adder. Input: (A) 100 equiv. $[(Bu)_4N]OH$; (B) 100 equiv. $[(Bu)_4N]OH$.

(a) the simplest molecule with –NH– and –N = groups, i. e. imidazole, cannot be operated as a molecular processor because the absorption spectra of the protonated form is the same as that of the neutral one; (b) the differences between the arene matrixes may significantly influence the logic functions, for example, 8-HQ and 4-HP, 4-AP and 5-AN; (c) based on the performance and structure of fluorescein, one may infer that the logic molecules containing only acidic or basic groups could also perform addition and/or subtraction operations as long as they have spectroscopically unique states and their $pK_{a/b}$ values between different states are distinct; (d) it is possible that the four- or more-state molecules with three or more acidic and/or basic groups could operate as full-adders and/or full-subtractors. One example is fluorescein, as demonstrated by Shanzer et al. [13]; (e) all of the molecular processors could be reset efficiently by acid-base neutralization after each operation [13].

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References

1. de Silva, A. P.; Gunaratne, H. Q. N.; McCoy, C. P., *Nature* **1993**, *364*, 42

2. Callan, J. F.; de Silva, A. P.; Magri, D. C., *Tetrahedron* **2005**, *61*, 8551
3. Pischel, U., *Angew. Chem. Int. Ed*. **2007**, *46*, 4026
4. Szaciłowski, K., *Chem. Rev*. **2008**, *108*, 3481
5. de Silva, A. P.; McClenaghan, N. D., *J. Am. Chem. Soc*. **2000**, *122*, 3965
6. Remacle, F.; Speiser, S.; Levine, R. D., *J. Phys. Chem. B* **2001**, *105*, 5589
7. Andréasson, J.; Kodis, G.; Terazono, Y.; Liddell, P. A.; Bandyopadhyay, S.; Mitchell, R. H.; Moore, T. A.; Moore, A. L.; Gust, D., *J. Am. Chem. Soc*. **2004**, *126*, 15926
8. Guo, X. F.; Zhang, D. Q.; Zhang, G. X.; Zhu, D. B., *J. Phys. Chem. B* **2004**, *108*, 11942
9. Qu, D. H.; Wang, Q. C.; Tian, H., *Angew. Chem. Int. Ed*. **2005**, *44*, 5296
10. Zhou, Y. C.; Wu, H.; Qu, L.; Zhang, D. Q.; Zhu, D. B., *J. Phys. Chem. B* **2006**, *110*, 15676
11. Andréasson, J.; Straight, S. D.; Kodis, G.; Park, C. D.; Hambourger, M.; Gervaldo, M.; Albinsson, B.; Moore, T. A.; Moore, A. L.; Gust, D., *J. Am. Chem. Soc*. **2006**, *128*, 16259
12. Margulies, D.; Melman, G.; Felder, C. E.; Arad-Yellin, R.; Shanzer, A., *J. Am. Chem. Soc*. **2004**, *126*, 15400
13. Margulies, D.; Melman, G.; Shanzer, A., *Nature Mater*. **2005**, *4*, 768
14. Margulies, D.; Melman, G.; Shanzer, A., *J. Am. Chem. Soc*. **2006**, *128*, 4865
15. Liu, Y.; Jiang, W.; Zhang, H. Y.; Li, C. J., *J. Phys. Chem. B* **2006**, *110*, 14231
16. Sun, W.; Xu, C. H.; Zhu, Z.; Fang, C. J.; Yan, C. H., *J. Phys. Chem. C* **2008**, *112*, 16973
17. Langford, S. J.; Yann, T., *J. Am. Chem. Soc*. **2003**, *125*, 11198
18. Coskun, A.; Deniz, E.; Akkaya, E., *Org. Lett*. **2005**, *7*, 5187
19. Guo, Z. Q.; Zhao, P.; Zhu, W. H.; Huang, X. M.; Xie, Y. S.; Tian, H., *J. Phys. Chem. C* **2008**, *112*, 7047
20. Suresh, M.; Jose, D. A.; Das, A., *Org. Lett*. **2007**, *9*, 441
21. Andréasson, J.; Straight, S. D.; Bandyopadhyay, S.; Mitchell, R. H.; Moore, T. A.; Moore, A. L.; Gust, D., *Angew. Chem. Int. Ed*. **2007**, *46*, 958
22. Amelia, M.; Baroncini, M.; Credi, A., *Angew. Chem. Int. Ed*. **2008**, *47*, 6240
23. Margulies, D.; Felder, C. E.; Melman, G.; Shanzer, A., *J. Am. Chem. Soc*. **2007**, *129*, 347
24. Guo, Z. Q.; Zhu, W. H.; Shen, L. J.; Tian, H., *Angew. Chem. Int. Ed*. **2007**, *46*, 5549
25. de Silva, A. P.; McClenaghan, N. D., *Chem. Eur. J*. **2004**, *10*, 574
26. Tang, C. W.; VanSlyke, S. A., *Appl. Phys. Lett*. **1987**, *51*, 913
27. Bardez, E.; Devol, I.; Larrey, B.; Valeur, B., *J. Phys. Chem. B* **1997**, *101*, 7786
28. Goldman, M.; Wehry, E. L., *Anal. Chem*. **1970**, *42*, 1178