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Review

A supramolecular chemistry basis for molecular logic and computation[☆]

A. Prasanna de Silva[a,*], Seiichi Uchiyama[b], Thomas P. Vance[a], Boontana Wannalerse[a,c]

[a] *School of Chemistry and Chemical Engineering, Queen's University, Belfast BT9 5AG, Northern Ireland*
[b] *Graduate School of Pharmaceutical Sciences, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-0033, Japan*
[c] *Department of Chemistry, Faculty of Science, Chulalongkorn University, Bangkok 10330, Thailand*

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Abstract

The ideas of supramolecular chemistry can be a useful way of advancing molecular logic and computation. Some of these cases occur at the molecular-scale while others require the intervention of bulk materials of various kinds. Even irreversible chemical reactions can provide food for thought. Logic operations from the simplest to small-scale integrated cases are now available, including those with arithmetic capability.

Keywords: Molecular logic; Molecular computation; Molecular arithmetic; Fluorescent switches; Photoinduced electron transfer

1. Introduction

The introduction of supramolecular [1] and host–guest [2,3] concepts has energized coordination chemistry [4]. Further impetus can be provided if ideas of molecular logic and computation [5–12] can also be connected. Conversely, the ideas of supramolecular chemistry can suggest alternative approaches to current issues in molecular computation and molecular electronics [13]. This is especially important for two reasons. These fields have seen much speculation, and even hype. On the other hand, the experimental results have been dogged, at least occasionally, by artifacts.

2. Molecular-scale approaches to logic

The supramolecular chemistry approach to molecular logic and computation has slowly grown without anything like the controversies alluded to above. This review is an opportunity to display and celebrate this growth. Aspects of supramolecular and coordination chemistry can be discerned in many cases of molecular logic operations reported so far, whether they are molecular-scale or not (Tables 1 and 2, respectively). Though by no means comprehensive, these tables are testament to the wide variety of schemes that have been developed by inventive chemists. Many of these schemes incorporate supramolecular chemistry ideas. We hope that these tables will serve the community as stand-alone sources of data, but a few examples are discussed in some detail. The molecular-scale cases are considered first.

For instance, **1** [14] (Table 1) is concerned with how Na^+-binding to a benzo-15-crown-5-ether and H^+-binding to an

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* Corresponding author.
E-mail address: a.desilva@qub.ac.uk (A.P. de Silva).

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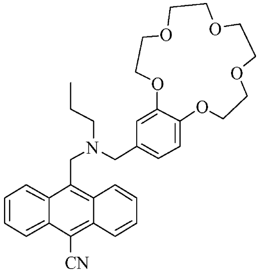
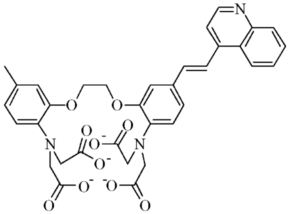
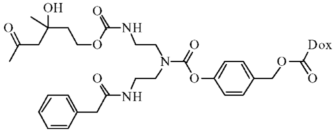
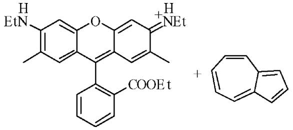
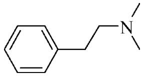
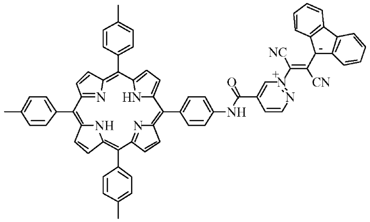
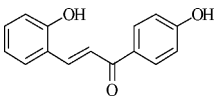
Table 1
Molecular-scale logic operations

No.	Active molecular entity	Input(s)	Output	Logic type	Comments	Ref.
1		H^+ and Na^+	Fluorescence	AND	PET mechanism	[14]
2		H^+ and Ca^{2+}	Transmittance at λ	XOR	ICT mechanism. Other wavelengths provide other logic types	[15]
3		Penicillin G amidase and catalytic antibody 38C2	Drug (doxorubicin = Dox-H) (inhibiting leukemia cell growth)	OR	Enzyme- or antibody-catalyzed hydrolysis of primary amide in prodrug is followed by intramolecular cyclization	[16]
4		Light intensity at λ_1, λ_2 and λ_3	Fluorescence	Nearly full-adder	Two-photon fluorescence and electronic energy transfer (EET) are involved	[17]
5		Light intensity at λ_1, λ_2 and λ_3	Fluorescence and ionization (detected by mass spectrometry)	Full-adder	Photoionization	[18]
6		Light dose at λ_1 and λ_2	Fluorescence	XOR	Photoisomerization-controlled PET is involved. Half-adder is constructed with another porphyrin gate	[19]
7		H^+ and light dose at λ_1	Absorbance at λ_2 or fluorescence	AND	E-Z photoisomerization is followed by acid-catalyzed cyclization	[20]

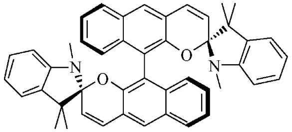
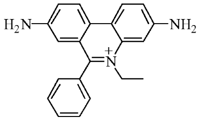
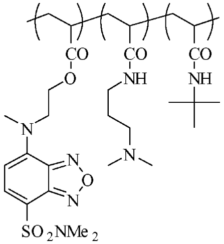
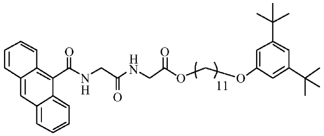
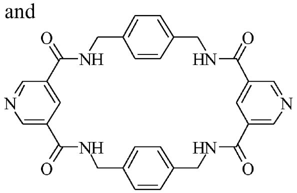
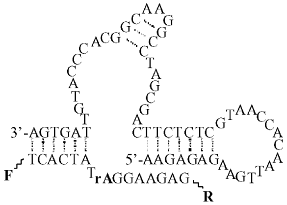
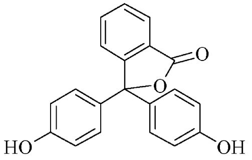
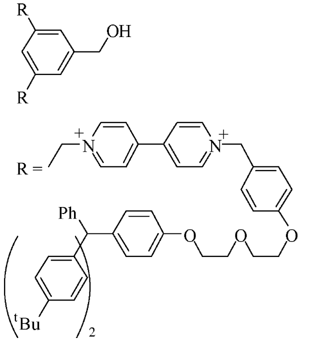
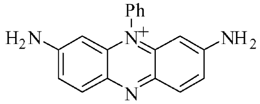
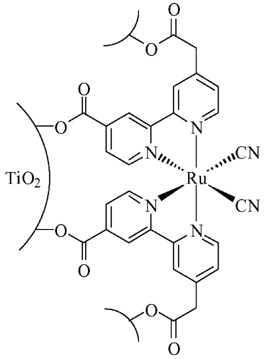
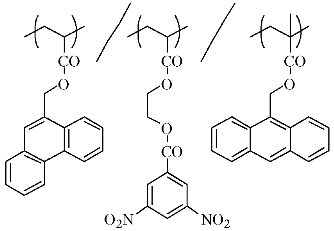
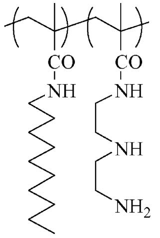
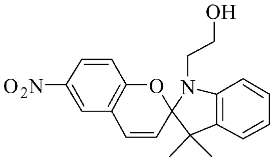
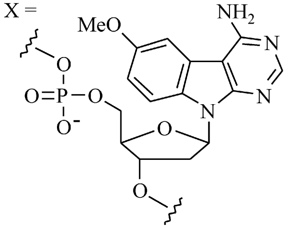
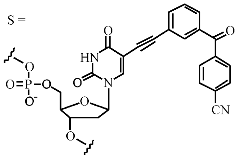
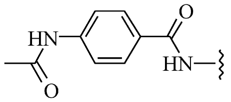
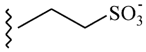
8		H^+ and light dose at λ	Circular dichroism	AND	Photochromism of chiral material is regulated by H^+	[21]
9	Glucose dehydrogenase and horseradish peroxidase	Glucose and H_2O_2	NADH and gluconic acid (measured absorptiometrically)	INH, AND	Cofactor NADH is required. Many other enzymes can be run in tandem in this way	[22]
10	5′-GCCAGAACCCAGTA GT-3′-fluorescein	3′-CGGTCT TGGGTCAT CA-5′ and 	Fluorescence	NAND	Oligonucleotide hybridization is followed by ethidium intercalation which quenches fluorescein emission by EET	[23]
11		H^+ and heat	Fluorescence	INH	Polymer chain folds up in hot water, if the medium is not acidic, so that the water-sensitive fluorophore is shielded	[24]
12	 and 	H^+ and DMSO	Fluorescence	INH	Macrocycle resides on polymethylene chain in hydrogen-bond disruptor solvent DMSO. Then fluorophore is too far from protonated pyridine PET acceptors to cause fluorescence quenching	[25]
13	 F, fluorescein; R, tetramethylrhodamine	5′-CATTGG TGTTAACTT-3′	Fluorescence at $λ_1$	YES	Input strand hybridizes with right-hand loop, opening the inner stem-loop and raising its ribozyme activity. This accelerates cleavage at rA and prevents EET from fluorescein	[26]

Table 2
Other molecule-based logic operations

No.	Active molecular entity	Input(s)	Output	Logic type	Comments	Ref.
14		OH^-	Absorbance at λ	AND, OR	Compound is indicator during acid–base titration in stirred flow reactions. Positive and negative logic can be performed with absorbance and transmittance	[27]
15		Voltage	Current	AND, OR	Logic is not intrinsically molecular, but produced by metallic wiring of the switches. Irreversible reconfiguring was done by oxidatively damaging compound	[28]
16		H^+ and voltage	Fluorescence	INH	Compound is in Nafion® film on indium-tin oxide electrode in electrochemical cell	[29]
17	$[Fe^{II}(CN)_6]^{4-}$	Light intensity at λ_1 and λ_2	Current	OR, XOR	Compound is adsorbed on nanocrystalline TiO_2 electrode in electrochemical cell. Voltage-regulated logic reconfiguring is observed	[30]
18		Cu^{2+} and voltage	Luminescence	NOR	Compound is bound covalently to TiO_2 electrode in electrochemical cell	[31]
19		Light intensity at λ_1 and λ_2	Current	XOR	Compounds are in polymer Langmuir–Blodgett film between electrodes	[32]

20		AMP and Na$_3$PO$_4$	Dimension change	AND	Compound-based gel is observed microscopically	[33]
21		Light dose at λ_1 (at different spatial addresses)	Transmittance at λ_2	NOR	Array of cuvette containing compound is required for integrated logic of several gates, even though the elementary logic operation is molecular-scale. Input/output is not quantitatively homogeneous. The smallest dimension in the cuvette array (cuvette separation distance) should be reducible	[34]
22	$[Fe^{II}(CN)_5NO]^{2-}$	OH$^-$ and K$^+$	Absorbance at λ	Integration of 20 AND and OR gates is achieved for the case of a 2 × 2 × 2 cell array arranged at cubic corners	Inputs regulate reaction of compound with thiol or subsequent photochemical reaction. Comments for the case **21** are mostly relevant here	[35]
23	5′-GGAGATCAAGCTCATACACTCAACCTGATAGACAAGGACA-3′, etc.	3′-GAGTATGTGA-5′, etc.	Hybridization (read by fluorescence)	Full-adder	Compound is assembled on plate. Operation requires ligation, denaturing, and washing or gel electrophoresis	[36]
24	5′-ATTTATAGTGTGGGTTXTTXTTGGGTTATTAT-3′, etc. X =  S = 	3′-TAAATASCACACCCAATAATAACCCAATAATA-5′, etc.	Cleavage (following photoinduced hole transport in duplex)	Full-adder	Compound is hybridized with input(s). Cleavage is visualized after gel electrophoresis	[37]
25	Ar-RVSALEAAVSELEKKVA-COSR′ and H$_2$N-CLEXEVARLKKLVGE-CONH$_2$ Ar =  X = Ar-K R' = 	Ar-RVARLERZ$_1$VSELZ$_2$RKVACLEXEVARLKKLVGE-CONH$_2$ (Z$_1$, Z$_2$) = (K, A) and (K, K)	Ar-RVARLERAVSELERKVACLEXEVARLKKLVGE-CONH$_2$ (detected by chromatography)	OR	Peptide network arises from template-directed ligation	[38]

amine can produce a molecular AND gate when fluorescence is viewed as output. AND gates are common in modern semiconductor computers [39] and are well known to mathematicians [40,41]. Each of these receptors launches a photoinduced electron transfer (PET) process [42,43] to the 9-cyanoanthracene fluorophore across the methylene spacers in order to quench fluorescence until H^+- and Na^+-binding occurs. Then, both PET processes are arrested and fluorescence is revived. This is two-input AND logic since both H^+ and Na^+ are required as inputs before the fluorescence output becomes switched 'on'. This was the first instance where molecules were endowed with computational capabilities in the primary literature [14]. It is still remarkable that a small molecule manages to marshal two inputs from free-swimming chemical species, an ultraviolet light power supply and a blue fluorescence output while providing the internal data processing required.

The case of **11** [24] (Table 1) employs polymer chain folding phenomena to create molecular logic operations. Aqueous solutions of linear polymers or co-polymers with balanced hydrophobicity and hydrophilicity are quite sensitive to temperature. High temperatures cause the chains to contract into a globular form. In fact, this contraction can occur quite sharply. The current example possesses the hydrophobic *t*-butyl and the hydrophilic dimethylaminopropyl units to achieve this delicate balance. The third co-monomer (which is only present at 0.1%) contains the fluorophore which is a benzofurazan system with electron-donor amino and electron-acceptor sulfonamide units. This fluorophore's emission is quenched when exposed to water, which is the case when the co-polymer chain is in the extended form in cold water. The situation changes in hot water as the chain curls up to hide the fluorophore within its folds. Then fluorescence is revived. However this switching 'on' of fluorescence can be prevented by supplying a H^+ input. Protonation of the dimethylaminopropyl unit hugely enhances its hydrophilicity to leave the co-polymer chain extended at all temperatures examined. The mutual repulsion of protonated dimethylaminopropyl units must also contribute to this chain extension. Thus the fluorophore is never hidden from water and the emission is never switched 'on' in acid medium. This is the INH logic where H^+ is the disabling input.

3. Other molecular-based approaches to logic

Though molecular cases requiring bulk materials at some stage of the experiment are gathered separately for ease of classification in Table 2, their value is as high as those in Table 1.

For instance, the case **17** in Table 2 considers nanocrystalline TiO_2 electrodes with adsorbed $[Fe^{II}(CN_6)]^{4-}$ in suitable photoelectrochemical cells which produce photocurrents whose sign depends on the redox state of the complexed species and whether the TiO_2 substrate or the complexed species is being photo-selected [30]. It is then possible to arrange XOR logic by mutual cancellation of photocurrent arising from two light beams with different excitation wavelengths. However, interfacial electron transfer kinetics cause spikes upon switching the light beams 'on' and 'off'. The output has to be chosen as the modulus of the photocurrent for this scheme to work. XOR logic in molecular-scale systems can be arranged by similar mutual annihilation of acid and base [44], but schemes with straightforward ion binding which are more robust for operation in gate arrays are now available [15,45]. The latter were needed for the first demonstration of molecular numeracy [45]. Since the applied potential can be varied at the same time as the excitation wavelength in the case of **17**, the former can be employed as a means of reversibly reconfiguring the logic pattern. However, there are several previous examples of reconfiguring molecular logic. These use the adjustment of the ion input [46] or the excitation/observation wavelength [15,47,48]. In fact, the latter can be used to attain superposed logic where multiple logic configurations are observed simultaneously [15].

4. Chemically irreversible reactions as molecular logic operations

Even chemically irreversible reactions are worth examining in a logic context, especially when the cases concern detection of difficult targets such as diethylchlorophosphate (DCP). There appear to be no supramolecular schemes at present for binding this species. Two representative cases displaying YES logic are **26** [49] and **27** [50]. The electron donor aniline unit of **26** is twisted out of the xanthene fluorophore plane owing to the steric requirements of the carboxylate group. Thus it becomes a PET system with a virtual spacer. Older work [51] from the same laboratory had shown 'off-on' fluorescence switching upon proton binding to the aniline so that PET is suppressed. This time PET suppression is achieved by nucleophilic substitution of DCP with the aniline's amine group so that a phosphoramide is produced.

The PET components of pyrene fluorophore, methylene spacer and amine electron-donor can be easily discerned in **27** [50]. The reaction of the OH group of **27** with DCP first produces a phosphate ester which is intramolecularly displaced by the amine to depart as the corresponding phosphate anion. The resulting ion pair is the fluorescent form. A complication in this case is the generation of HCl from the DCP reaction, which will protonate another mole of **27** causing additional fluorescence switching 'on'. This artifact can be removed by pH-buffering the solution, as was done with **26** [49].

5. More complex molecular logic

Raymo and Giordani cleverly enlarged the scale of integration by running reactions in arrays of cells [34,47,52–54]. Thus, a new way of distinguishing inputs is introduced. Even the same light beam, when aimed at a different cell becomes a different input. Their cases concerned H^+ and light dose as inputs and absorbance as output. Szaciłowski's [35,55] newer example **22** (Table 2) involves a reaction of this type. The one-cell or zero-dimensional case shows colour development (the product **28** absorbing at 520 nm) only if the pH is high (allowing nucleophilic thiolate to be available) and the salt (KCl) concentration is high (so that K^+ stabilizes the trianionic **28** by ion-pairing). So this is two-input AND gate behaviour. Some of the variants can be illustrated by irradiation of **28** at 520 nm to produce **29**

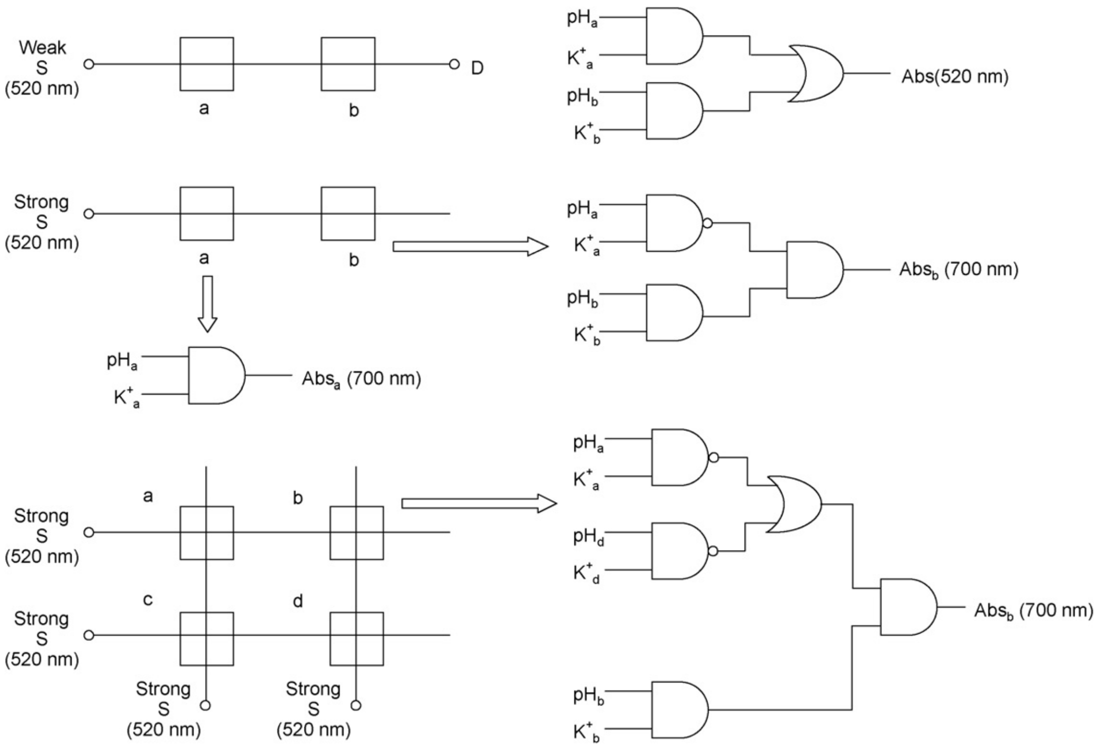


Fig. 1. Szaciłowski's multi-cell array experiment [35]. S, Source; D, Detector.

with an absorption signature at 700 nm. **29** is produced only if **28** is present in the first place, i.e. if both pH and [K^+] are high. This is another two-input AND gate but with a new output. Of course the light dose at 520 nm could have been counted as the third input making this photoreaction a three-input AND gate. But let's stick to the two-input version and watch it evolve to larger gate arrays.

Fig. 1 (top) focuses on the one-dimensional case of a line of two cells, with a weak light source emitting at 520 nm and a photo-detector as the termini. It is clear that a high absorbance is detected if either cell a or cell b develops the coloured **28**, i.e. if either cell has high pH and high K^+. Thus the equivalent logic gate array is composed of two 2-input AND gates feeding an OR gate. Fig. 1 (middle) considers the same cell array but in photochemical reaction mode with a stronger light source. **29**'s absorption at 700 nm is the output we are after. If we examine the front cell a, it needs a high level of **28** (set up by high pH and high K^+) to photochemically proceed to **29**. Thus we have a simple two-input AND gate only. Shifting our focus to cell b at the back gives a different result. A high level of **28** (to be photoconverted to **29**) will arise in cell b if it has high pH and high K^+ within, and if 520 nm light gets through cell a, i.e. if the latter does not have the condition of high pH and high K^+. The equivalent logic array becomes a two-input AND (from cell b) and a two-input NAND (from cell a) feeding an AND gate. Fig. 1 (bottom) takes on the two-dimensional case of a square array of four cells with four light sources. We only consider one scenario of photochemistry occurring in cell b having no sources directly impinging on it. Besides the chemical requirements discussed above, a high level of **29** will accumulate in cell b only if 520 nm light gets through either cell a or cell d. So the equivalent logic array becomes two 2-input NAND gates (from cells a and d) feeding an OR gate whose output combines with that from a two-input AND (from cell b) as inputs into another AND gate.

(a)

IN_1	IN_2	Borrow OUT_1	Difference OUT_2
H^+	OH^-	A_{474}	A_{447}
0	0	0	0
0	1	1	1
1	0	0	1
1	1	0	0
		INH	XOR

(b)

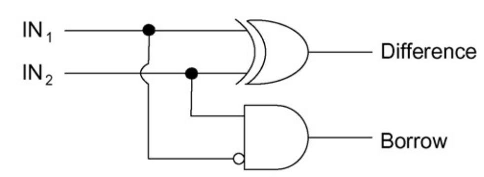


Fig. 2. Half-subtractor due to Margulies et al. [58].

(a)

IN_1	IN_2	Borrow$_{in}$ IN_3	Borrow$_{out}$ OUT_1	Difference OUT_2
H^+	OH^-	OH^-	A_{474}	A_{447}
0	0	0	0	0
0	1	0	1	1
1	0	0	0	1
1	1	0	0	0
0	0	1	1	1
0	1	1	1	0
1	0	1	0	0
1	1	1	1	1

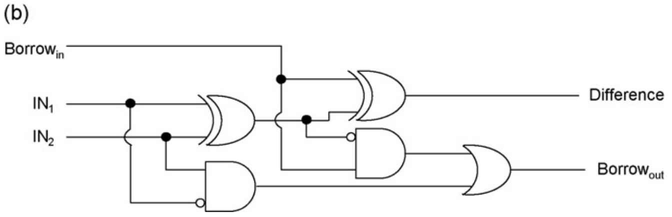


Fig. 3. Full-subtractor due to Margulies et al. [58].

Three-dimensional cases with cubes of cells can be tackled similarly to achieve some of the most complex molecular logic gate integrations seen to date. However, molecular-scale activity is lost the moment separate cells are used. Nevertheless the smallest dimension in the cell array, e.g. cell–cell separation distance, should be reducible without disturbing the principle.

6. Molecular numeracy

After its introduction in 2000 [45] molecular numeracy has been generalized and expanded [17,18,37,56,57], to encompass addition and subtraction within a simple organic molecule [58]. To add to the attraction, this latter work only uses acid and alkali as inputs.

The half-subtractor [59,60], which requires XOR and INH gates (to produce difference and borrow digits, respectively), is constructed from the neutral state **30** [58]. Protonation of **30** to give **31** and deprotonation to yield **32** produce very different absorption spectra. The absorbance at 474 and 447 nm can be interpreted as INH and XOR behaviour (Fig. 2). The simultaneous application of acid and alkali inputs induces mutual annihilation, as if inputs were not applied at all [44].

(a)

IN_1	IN_2	Carry$_{in}$ IN_3	Carry$_{out}$ OUT_1	Sum OUT_2
OH^-	OH^-	OH^-	A_{474}	T_{447}
0	0	0	0	0
0	1	0	0	1
1	0	0	0	1
1	1	0	1	0
0	0	1	0	1
0	1	1	1	0
1	0	1	1	0
1	1	1	1	1

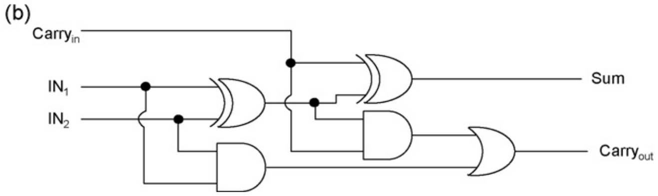


Fig. 5. Full-adder due to Margulies et al. [58].

The full-subtractor is now tackled by using another mole of OH^- as the third input or the borrow-in bit. Note that input$_2$ and input$_3$ are only distinguishable to the operator of the moleculator and not to the molecule itself. However at the end of the day, the operator is the one requiring the correct answer to the calculation. The top four rows of the truth table of the full-subtractor (Fig. 3) are the same as those for the half-subtractor (Fig. 2). Due to the mutual annihilation of equimolar H^+ and OH^-, the fifth and last rows are the same as the second. The seventh row is the same as the first. So only the sixth row remains to be justified. Indeed, treatment of **30** with 2 mol of OH^- (IN_2 and IN_3) produces dianionic **33** which has strong absorbance at 474 nm and little at 447 nm—exactly what's needed.

Now, let's see how the cationic state **31** produces a half-adder (Fig. 4). Note the change of the starting state that is being examined. One mole of OH^- (IN_1) converts it to neutral **30** and another mole of OH^- (IN_2) produced anionic **32**. So we are working with the same three states as we did for the half-subtractor! The old set of absorption spectra are all we need, but we must carefully take the transmittance at 447 nm to attain XOR logic behaviour. Absorbance data at 474 nm still serves to produce AND logic characteristics. Such opportunistic choice

(a)

IN_1	IN_2	Carry OUT_1	Sum OUT_2
OH^-	OH^-	A_{474}	T_{447}
0	0	0	0
0	1	0	1
1	0	0	1
1	1	1	0
		AND	XOR

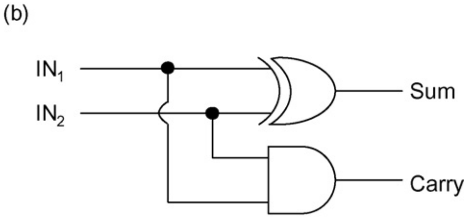
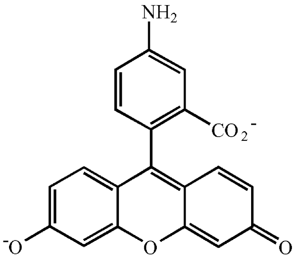


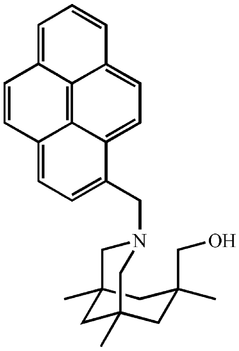
Fig. 4. Half-adder due to Margulies et al. [58].

of coding – switching between positive and negative logic – is legal.

As with the full-subtractor, the full-adder truth table (Fig. 5) has much that is familiar from Fig. 4 for the half-adder, such as the first four rows. Also noteworthy is the three-fold degeneracy of the inputs. The fifth row is algebraically equivalent to the second. The sixth and seventh rows are equivalent to the fourth. So only the last row remains to be justified. Treatment of **31** with three moles of OH^- produces **33** (again!) and hence has strong absorbance at 474 nm and strong transmittance at 447 nm (as noted in different language regarding the full-subtractor). That's the last row down as well.



26



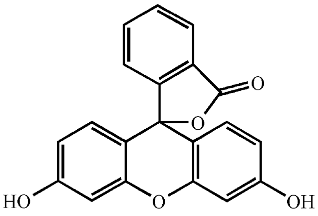
27

$[Fe^{II}(CN)_5N(O)SR]^{3-}$

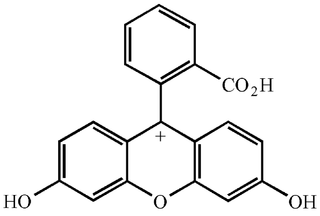
28

$[Fe^{III}(CN)_5SR]^{3-}$

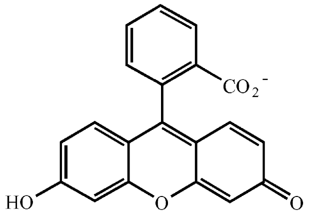
29



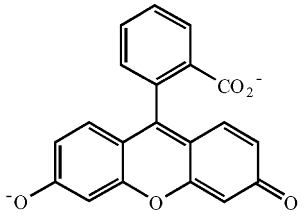
30



31



32



33

This example is particularly remarkable since it achieves such a humanly relevant computation with such a small molecule and such a simple experimental protocol.

7. Conclusion

Recent literature is testament to the general significance of logic interpretations of supramolecular phenomena, especially those of a photochemical nature. Chemical approaches to molecular computation seem to be bearing fruit.

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