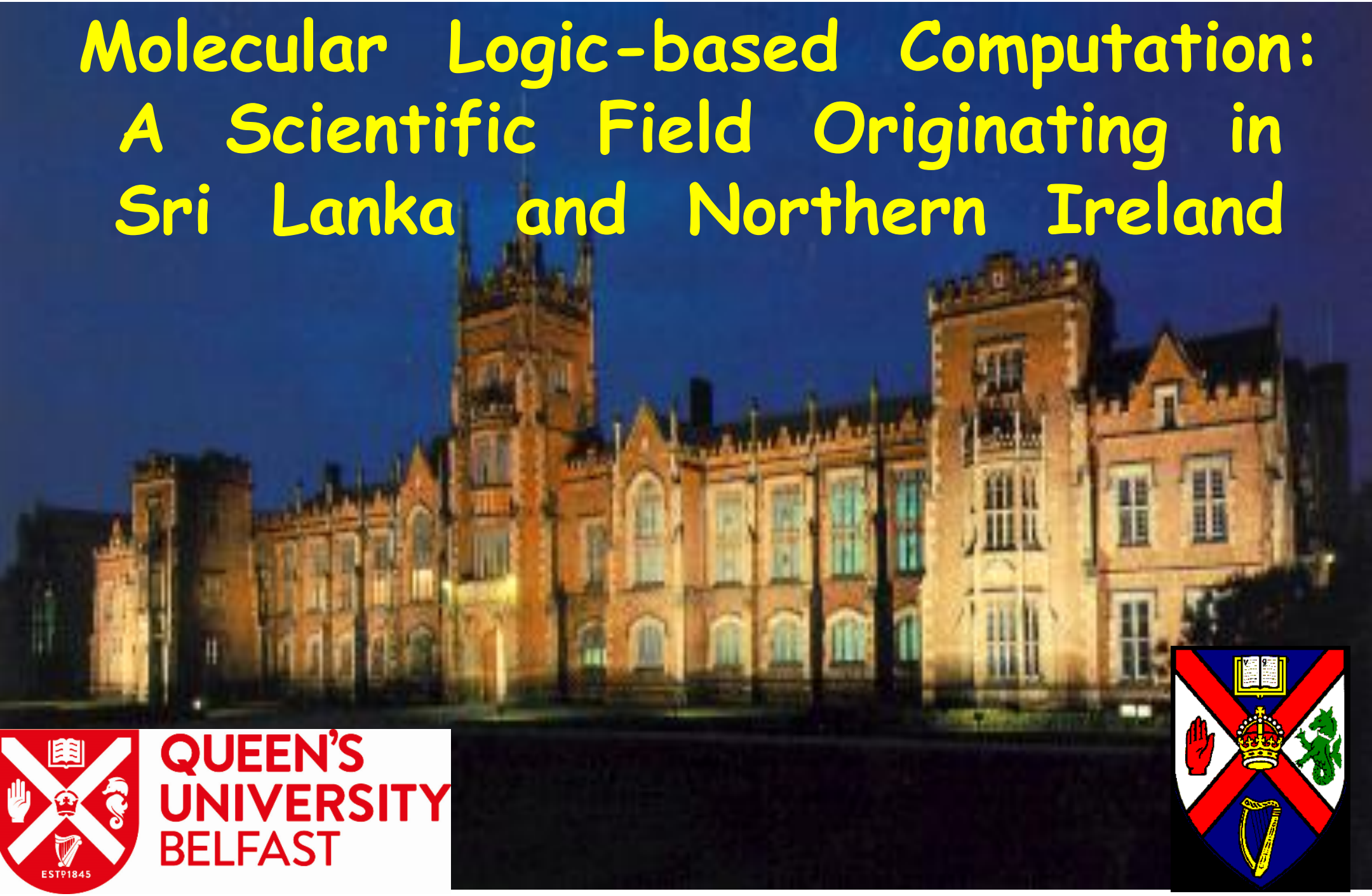
Molecular Logic-based Computation: A Scientific Field Originating in Sri Lanka and Northern Ireland



QUEEN'S UNIVERSITY BELFAST

ESTD 1845

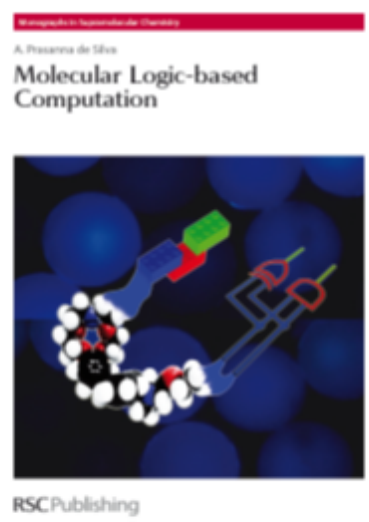
Molecular logic is chemical computation.
It sheds light on chemical and biological information processing

Nimal Gunaratne, Colin McCoy
Nature 1993, *364*, 42

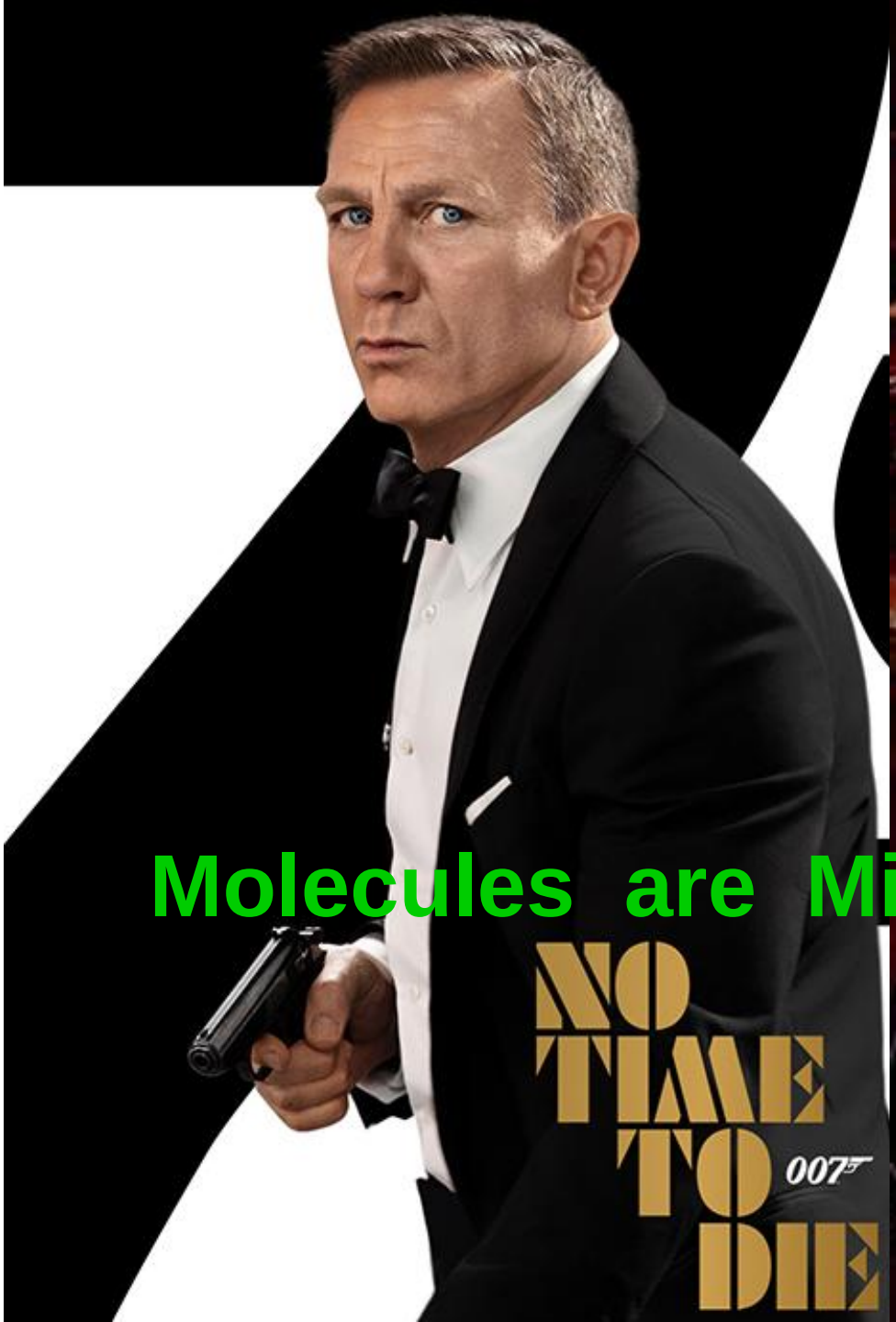
George Boole

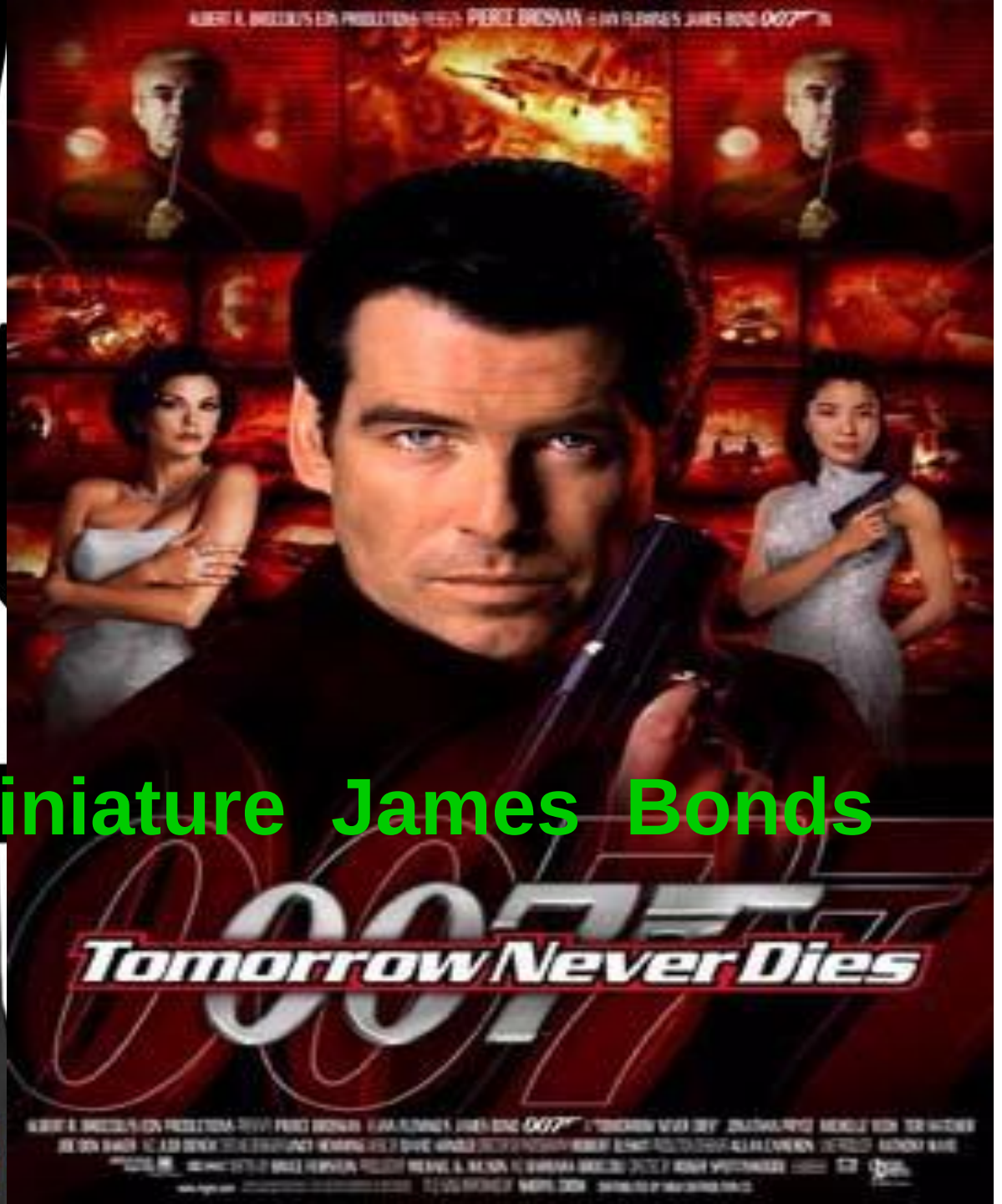
1849-64, Cork

- generalization
- integration
- numeracy & games
- reconfiguring
- improved sensing
- object identification
- Intracellular computing
- Human-level computing



Over 1730 labs have joined.





Molecules are Miniature James Bonds

The PET design (generalized in Colombo/Belfast) is robust

OFF state

Fluorophore

Spacer

Receptor

Chem. Rev.
1997, *97*, 1515

Chem. Soc. Rev.
2015, *44*, 4203

hν

PET

ON state

Fluorophore

Spacer

Receptor

M^+

hν

hν


Over 1200 labs have joined.


Walt, Miller, Blough,
de Silva, Lippard, Cathou,
Weissleder, Langmuir,
Rochford
Vanderkooi,
Yang,
Smith
Koide, Jung,
Kauffman
Czarnik, Pearson,
Zhang, Saeva
Kuhn, Strongin,
Gee, Li
Singaram,
Bertozzi,
Verkman,
Miller,
Chang
Smith,
Aron
Stoddart,
Imperiali,
Marcus,
Cary
Devaraj,
Barbas,
Tsien,
Rebek
Whitten
Inoue,
Machi
D'Souza
Wallace,
Ji,
McCarley
Brown
Jonusauskas, Fages, Burger,
Bouas-Laurent, Desvergne,
Lapouyade, McClenaghan, Michel
Tusa, Fahrni,
Lucchi
Gawley, Leblanc, Zhu,
Schulman, Belfield
Wang, Lindsey,
Finney, Deiters,
Lee
Durantini
Diaz-Fernandez
Lakowicz, Pu,
Chen, Kobayashi,
Geddes
Masilamani
Kulatilleke
Morawetz, Turro, Wu,
Lawrence, Adams,
Pardo, Catalina
de la Escosura, Torres,
Bruckner
Valeur, Miomandre,
Weiss, Leray, Lehn,
Sok
Gunnlaugsson,
O'Shea, Kruger,
Martínez-Calvo,
Evans
Grigg, Moore, Brown, James, Davidson, Beer,
Stoddart, Parker, Williams, Marin, Sutherland,
Nguyen. Beddard, Besley, Mckeown, Corrie,
Gibson, Ward
McCoy,
de Silva
Vendrell, Zysman-Colman,
Plater,
Leigh,
Callan
Rettig, Resch, Wolfbeis,
Moller, Rurack, Schuster
Overkleeft,
Verhoeven,
Jager, Sharma
Soumillion, Elias,
Smet
Petrushenko
Borisov, Müller, Mohr,
Leiner,
Aigner,
Klimant, Wang, Wei,
Knall
Zhang,
Yu, Li2
Reymond
Xu, Zeng, Lu, Zhu,
Liu, Yang2, Feng,
Li2, He2, Finney,
Ruan, Xiao,
Zhao2,
Guang-
Yue,
Dong
Zinic,
Steinberg
Pina,
Pischel2,
Lodeiro,
Prata,
Gois
Lippolis
Freccero,
Fabbrizzi,
Balzani, Prodi,
Barigelletti,
Bianchi,
Pallavicini,
Del Giacco,
Scandola,
Amendola,
Quici
Garcia-
Espana,
Costero,
Miranda,
Hernando,
Escriche,
Gallardo, Luis,
Guirado, Galindo,
Sastre-Santos
Ganguly,
Menon,
Das
Sarkar,
Singh2,
Kumar2,
Kaur,
Mandal,
Milton,
Misra,
Luxami,
Gupta
Shanzer,
Eichen,
Willner,
Margulies
Chang3, Kim9,
Churchill, Lee5, Son,
Yoon3, Park2, Ahn
Qian, Han2,
Peng, Guo,
Song, Liu3,
Duan, Piao,
Zhang2, Li,
Wang, Xiao
Tang, Li
Bharadwaj,
Angamuthu,
Khan
Samanta,
Venkataraman
Chattopadyay, Ghosh,
Dash, Patra, Thakur,
Mahapatra, Sarkar
Bhattacharya,
Maitra, Thiagar
Mugesh, Yelamaggad
Rajasekharan, Pitchumani,
Ajayaghosh,
Ramaiah, Varma,
Arunkumar
Inouye2, Hirai, Akashi, Unno,
Kikuchi, Tachikawa, Majima,
Chatterjee, Kimura, Maeda,
Sakamoto, Ueno, Suzuki, Osa,
Nakatsuji, Fukuzumi, Nishio,
Shizuka, Teramae, Tsukamoto,
Yagi, Iwai, Hamachi, Nakagawa
Tanaka, Nagano,
Imai, Nagamura,
Kagechika, Kubo,
Akasaka, Kondo,
Motoyoshiya, Ito
Shinkai, Kubo,
Hamachi, Ojida,
Imato, Kusano
Qian2, Tian, Lou,
Chen, Hou, Guo,
Ge, Yuan, Yang4,
Shi, Rasheed, Li4,
Zhu2, Wang6
Chen, Chang
Yu, Yuan, Lin2, Ma, Gong,
Zhang2, Zeng2, Chen, Song,
Wang, Wan, Li3, Ou, Lan
Guo, Wang5, Ma2, Hu4,
Hao, Jiao, Yang2, Sun,
Song, Hua, Gao2, Geng,
Zhang4, Liu2, Zheng
Ng, Yam, Chen,
Chan, Zhou, Ma,
Fong, Wang2,
Zhang, Sun, Liu,
Pan, Huo, Luo,
Tan, Wu, Lo,
Ko, Wong, Che,
Meng, Peng
Selinger
Lincoln, Richardson,
English, Wainwright,
Foo

Flame test for sodium

also

switches ‘on’ a light signal

and

has been used to count Na^+

‘Off-On’ sensors selective for Na^+

Jim Tusa, Marc Leiner, Hua-Rui He, Mark Mortellaro

Roche

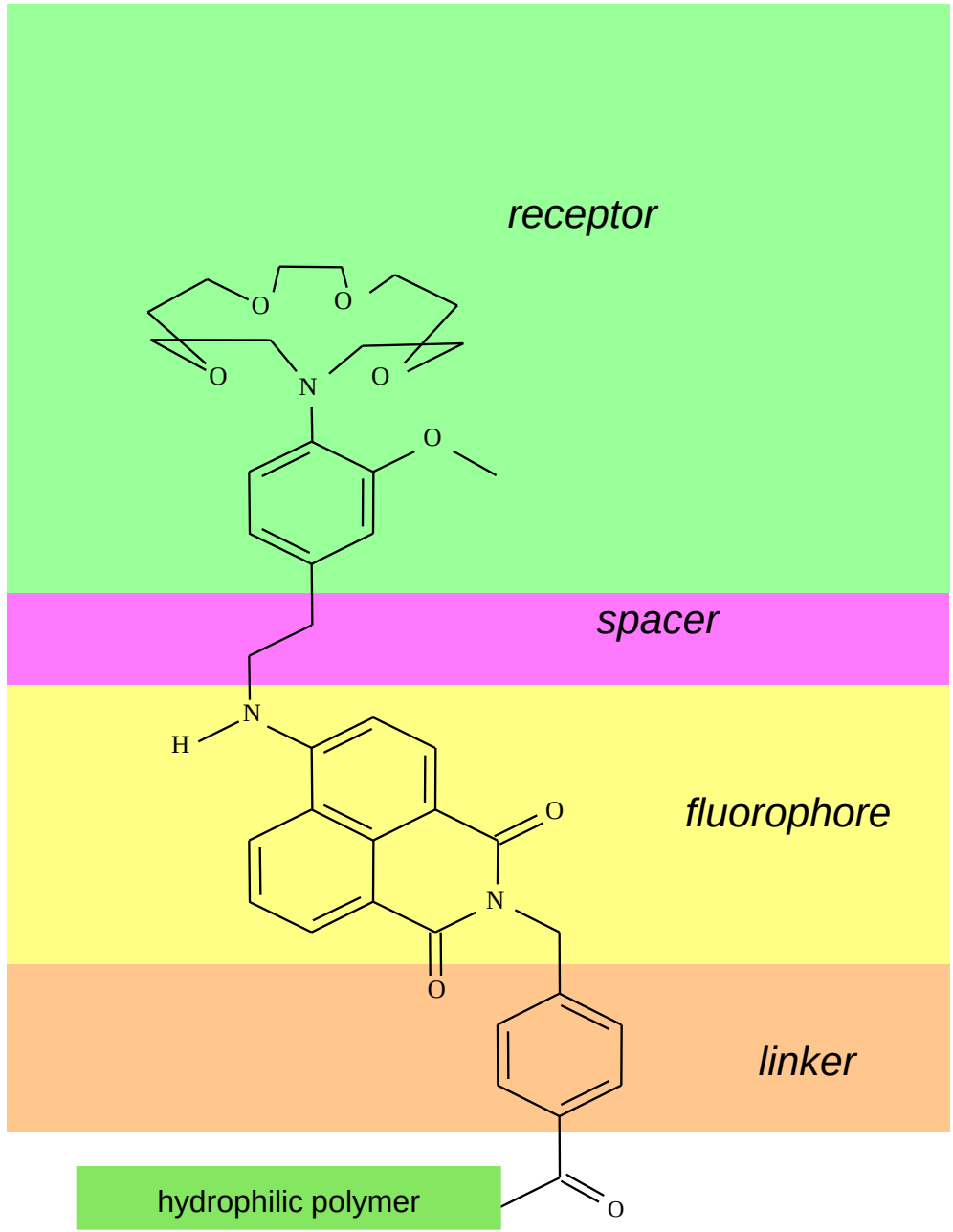
OPTIMedical

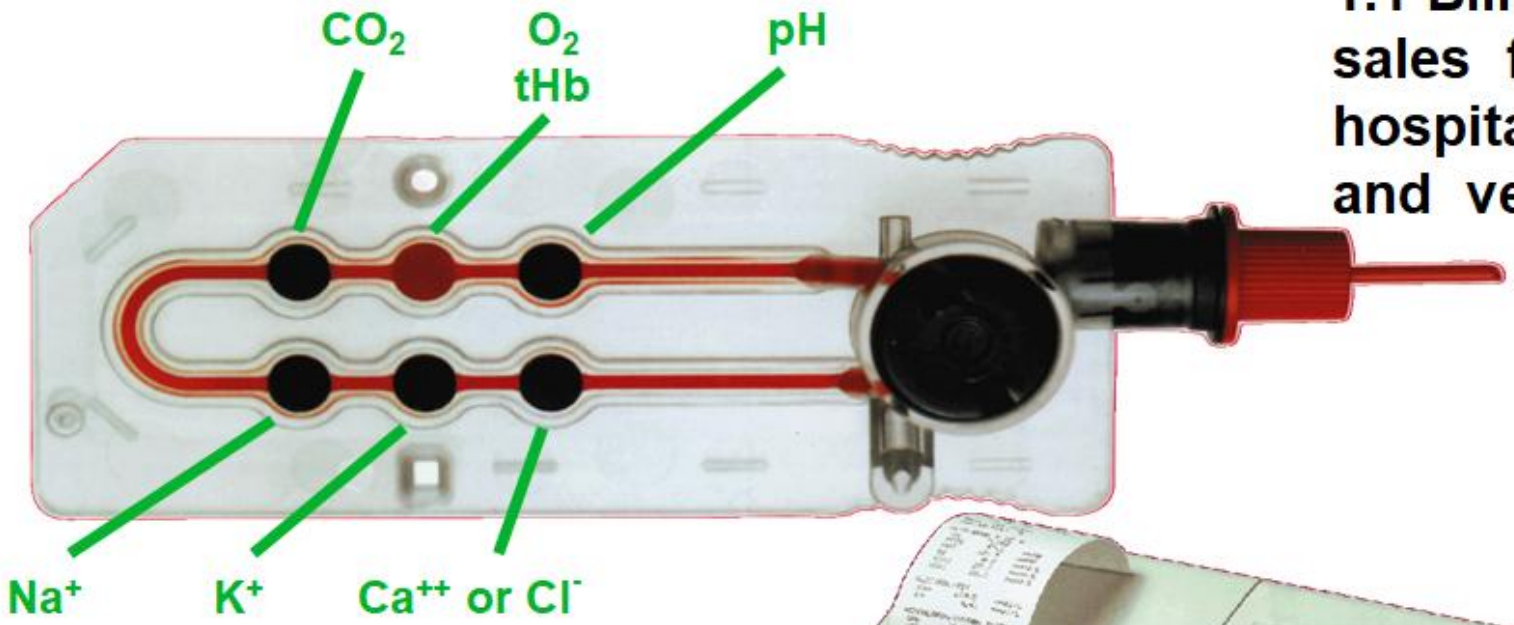
OPTI products

IDEXX LABORATORIES

VETSTAT products

Sodium





1.1 Billion USD Cassette sales for ambulances, hospitals, GP surgeries and veterinary surgeries

Disposable Cassette and OPTI Critical Care Analyzer (CCA). Also IDEXX VetStat Analyzer

Roche

OPTIMedical

IDEXX LABORATORIES

Molecular logic is chemical computation.
It sheds light on chemical and biological information processing

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Nature 1993, *364*, 42

George Boole

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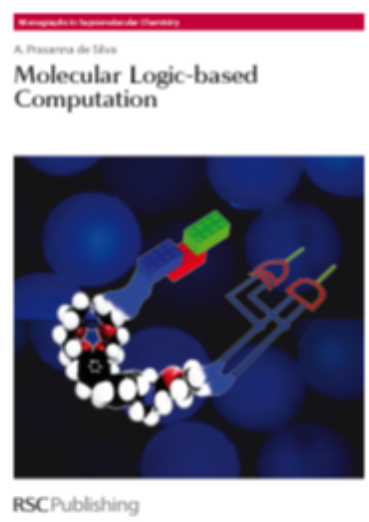

Over 1730 labs have joined.


James, Stoddart, Parker, Beer, Chin, Grigg, Adamatzky, Wang,
Williams, Tucker, Zauner, Fallis, Aldridge, Krasnogor, Dunn,
Davis, Mann
Benenson, Walt, Lin,
Zhang, Collins, Sykes,
Thayumanavan, Lu, Bhatia,
Whitesides, Church, You,
Aprahamian, Konry,
Gagnon, Green, Yin,
Voigt, Sun
Weiss,
de Silva,
Yurke,
Papenfort,
Tavazoie
Konermann,
Puri
Steer, Yeow,
Wettig,
Mondal
Redmond,
Gunnlaugsson,
MacDonaill,
O'Shea,
Bradberry
Wasielewski,
Conrad, Liu,
Smith, Singh,
Blaauw
Bazan
Stoddart,
Heath, Smolke
(Adleman),
Ghadiri,
Almutairi,
Barbas,
Wang,
Lerner,
Hwa
Ji
Deiters,
Reif
Birge, Sen,
Stojanovic,
Seeman,
Avouris,
Katz, Privman,
Sokolov, Wang
Breaker, Minko,
Hamilton,
Medintz, Adhya,
Sadelain, Ray,
Ogihara, Khoo
Gerasimova, Zhu, Tan,
Raymo, Johnson,
Kolpashchikov,
Toma, Araki
Bouas-Laurent, Desvergne, Sanguinet,
Bassani, McClenaghan, Rodriguez
Dickson, Wang, Minko, Nie, Santangelo,
Ke, Zhou
Brown
Fussenegger, Schluter, Wegner,
Diederich, Constable, Marchisio,
Winssinger,
Benenson
Gust, Moore, Li,
Yan, Green, Liu,
Green, Zhang
Stefanovic, Graves,
Doorn
Balzani, Campagna,
Credi, Venturi,
Pallavicini, Fabbrizzi,
Quici, Gentili, Bignozzi
Andreasson, Sun,
Moth-Poulsen, Li,
Landegren
Robertson,
Cockroft,
Callan,
Philp, Leigh,
Goni-moreno
de Silva,
Cao,
Callan
Vasquez-
Lopez
Pina, Pischel
Perez-
Inestrosa,
Pischel,
Ros
Garcia-
Espana,
Miranda,
Covera,
Costero,
Martinez-
manez
Lehn,
Bolze,
Ziessel,
Lutz
Kim(2), Yoon, Choi,
Churchill, Zhang
Schneider, Rurack, Martlbauer,
Voegtle, Schmittel, Schmalz,
Ihmels, Theato(2), Muller,
Schoening, Gupta, Ballauf,
Hellweg, Ramu
Samoc,
Uchacz,
Jurczak,
Pilarczyk,
Szacilowski,
Godlewski
Hoogenboom,
Remacle, Dehaen
Levine,
Speiser,
Shanzer,
Willner,
Shabat,
Ashkenasy(2),
Lotan,
Van der Boom,
Rabani,
Shapiro,
Eichen,
Bachelet,
Klajn, Alon,
Lahav, Fixler
Kikkeri, Sahoo,
Shankarling,
Das(2), Shah,
Thotiyl, Rao
Akkaya, Algi, Ekmekci,
Emrullahoglu,
Erbas-cakmak
Okamoto, Nishimura,
Inoue
Iwai, Sakai, Saito,
Sugimoto, Hirai,
Tomizaki, Hamachi,
Fujimoto, Akashi,
Miyashita, Saito,
Xu
Yuan, Fang,
Zhang, Zhu, Wu
Yan, Li(3), Fu,
Shi(3), Tong, Fan,
Wang(3), Xing,
Wu, Ju, Rao,
Ouyang, Ding,
Jiang, Wen
Wang(2), Ma, Peng,
Yang, Li(2), Lin,
Wei, Tang, Lu,
Shao, Liu
Jin, Gong,
Zhang,
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Fang, Yin,
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Dong(2), Qi
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Upadhyay, Moorthy
Samanta,
Bangal,
Kigga
Yang
Trivedi,
Shaishta
Kumar(2), Iyer, Upadhyay,
Bhalla, Ghosh, Mishra,
Singh(4), Kumari, Luxami, Hasan,
Kaur, Gupta(3), Ghosh, Milton, Basu,
Misra, Awasthi, Pandey, Kar, Mehata,
Ahmed, Maity
Uchiyama, Aida,
Tanaka, Fujita,
Suzuki, Kinbara,
Mihara, Nojima,
Shionoya,
Nishihara
Gao, Wang,
Wen, Gu, Liu(2)
Kimura,
Kubo, Ojida,
Nakakuki
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Huang(2), Zhang(3)
Qian(3), He, Zhu(3)
Xie, Yin(2), Liu, Lu,
Jiang, Wu, Dong,
Qiu(2), Pei(2), Yao,
Shen(2), Yan, Jin,
Zhong, Du, Fan,
Xu(3), Guo, Chen
Yu, Ye, Cheng, Zou
Yang, Ma, Li(2)
Lu, Zou, Liu, Yang,
Zhang(3), Song, Li,
Xiao, Cui, Deng, Shi,
Liang, Ma, Langford,
Wang(2),
Yao, Qiao
Evans,
Hughes

Molecular Logical Edge Detection

Animal vision: survival (security)

Early stage of visual perception. Analysis of an outline to save time in deciding whether it is threat or not

We all do it when awake

Machine vision: automation, security

Rapid screening of image outlines to save time on a production line or on closed-circuit TV.

Present as a subroutine in Photoshop™ or Paintshop™

Jue Ling, Gaowa Naren, Jessica Kelly, Tom Moody

J. Am. Chem. Soc. 2015, 137, 3763

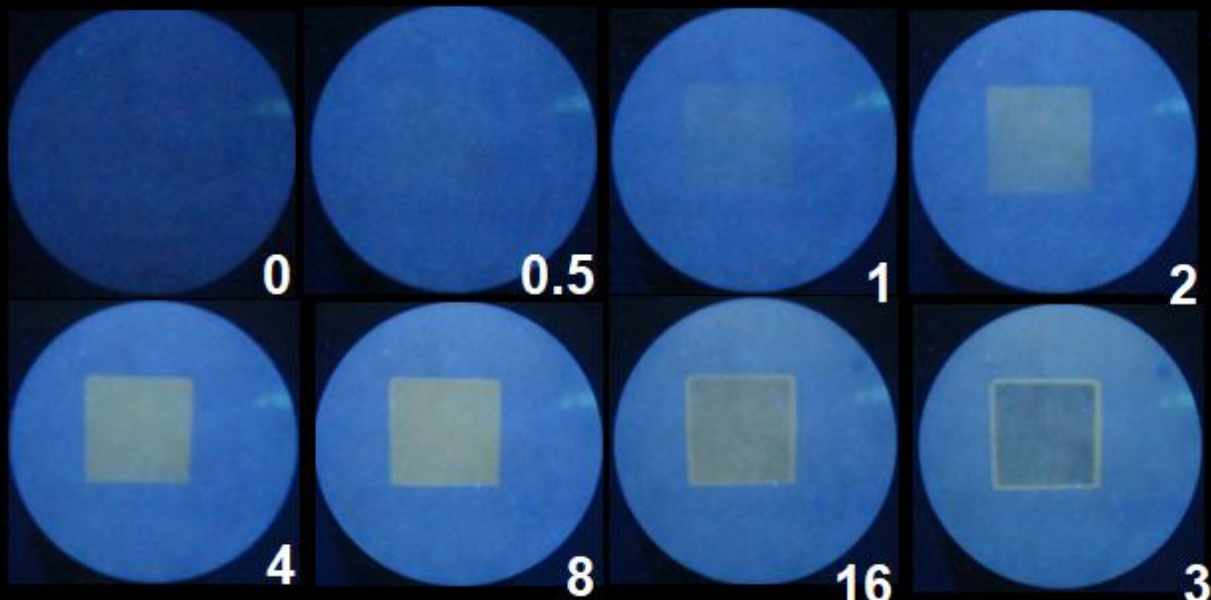
Inspiration:

Voigt, Ellington, et al *Cell* 2009, *137*, 1272

Chen, Ellington, et al *Nature Chem.* 2013, *5*, 1000

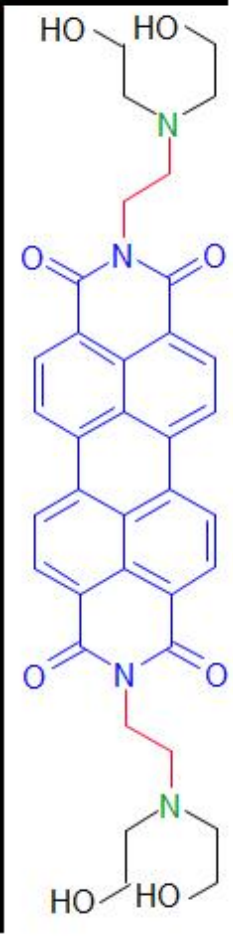
Drying at 50 °C for 4 min
254 nm writing time = t min

← Object (backlit mask)

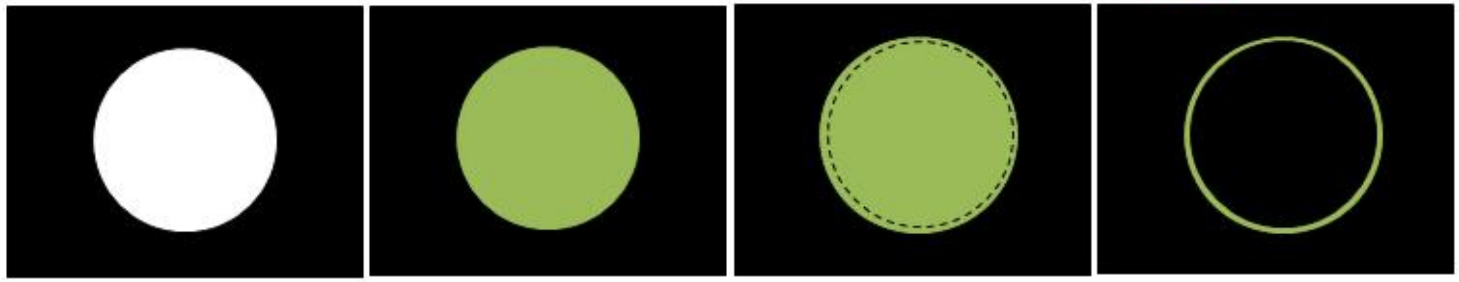


← Fluorescence image
(read with 366 nm)

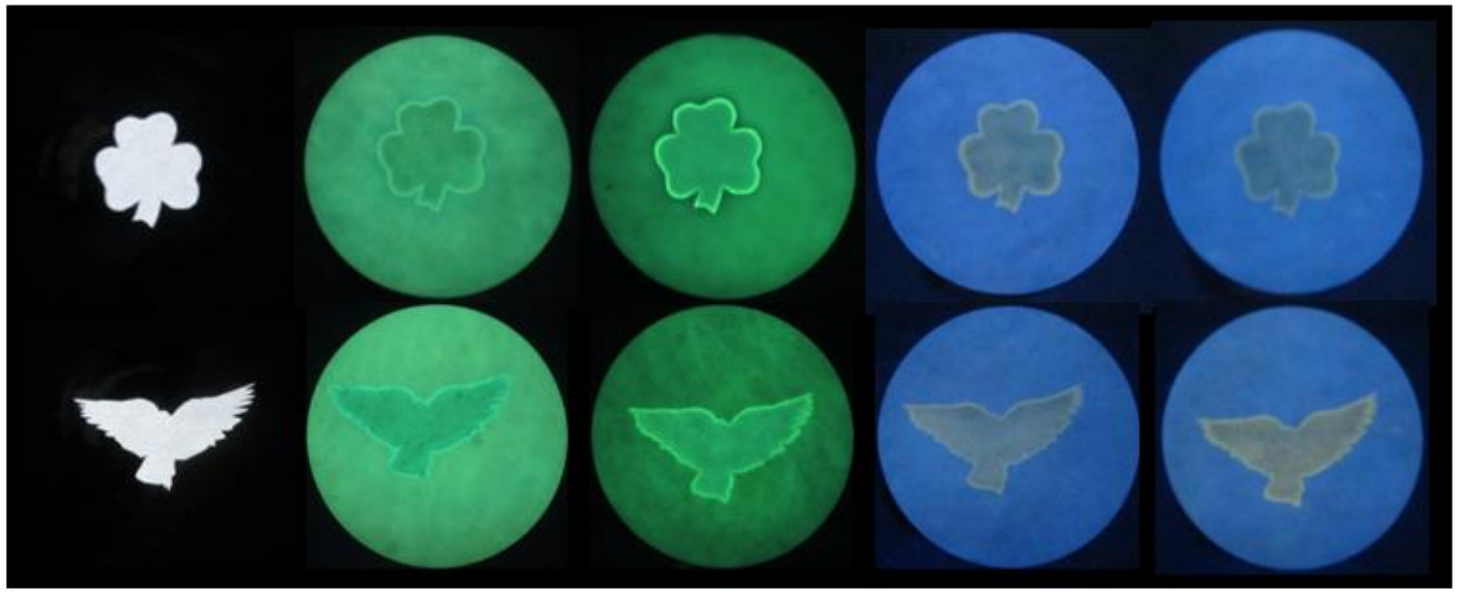
11 cm

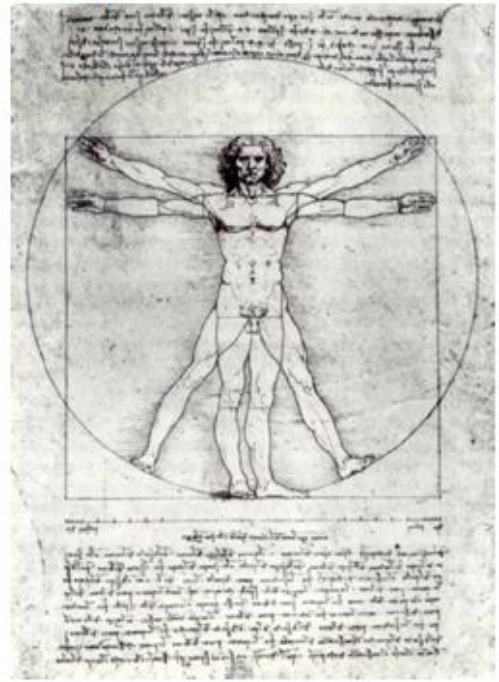


Molecular Outline Drawing



Template | Initial image | Subsequent image | Final image





Jue Ling, Gaowa Naren, Jessica Kelly, David Fox

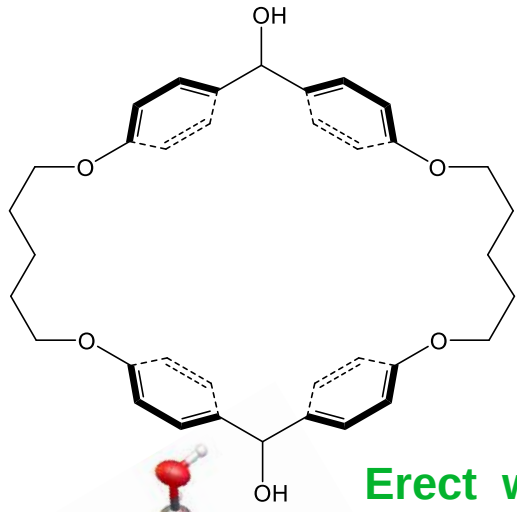
Chem. Sci. 2015, *6*, 4472

Molecular Memory with Downstream Logic processing

The problem of Serial Integration of Sequential and Combinational Logic

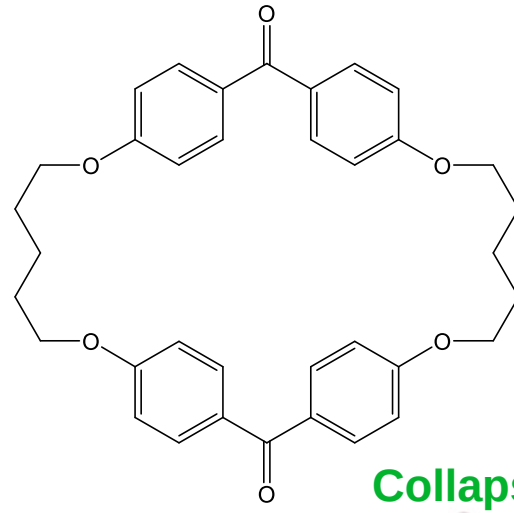
Brian Daly, Nimal Gunaratne, Tom Moody, Allen Huxley, Chao-Yi Yao, Ben Schazmann, Andre Alves-Areias, John Malone, Peter Nockemann

Nature Commun. 2019, *10*, 49



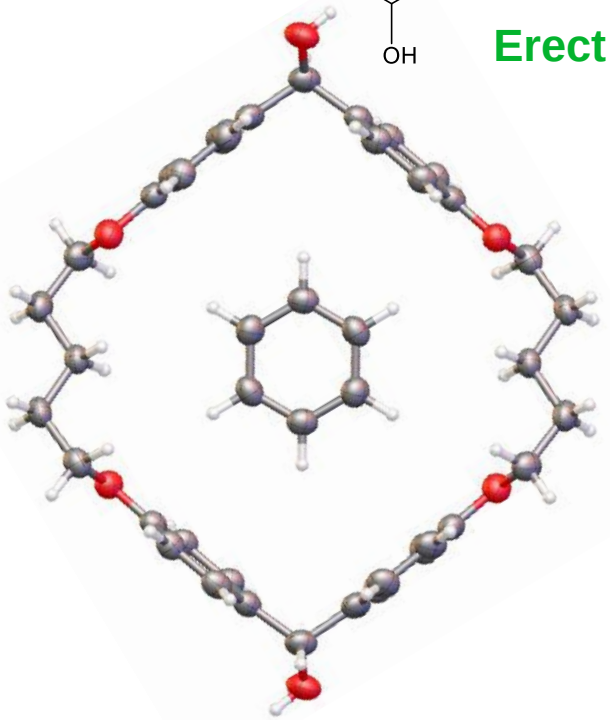
O_2, 110 ^oC, CsOH

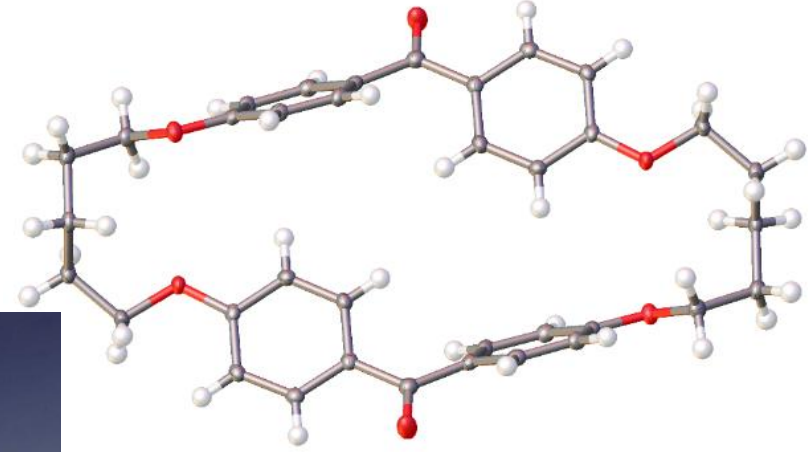
BH_4^-

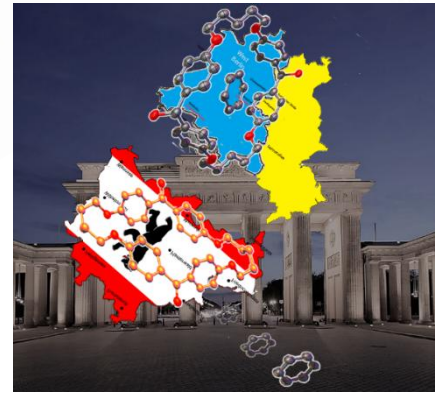


Erect walls

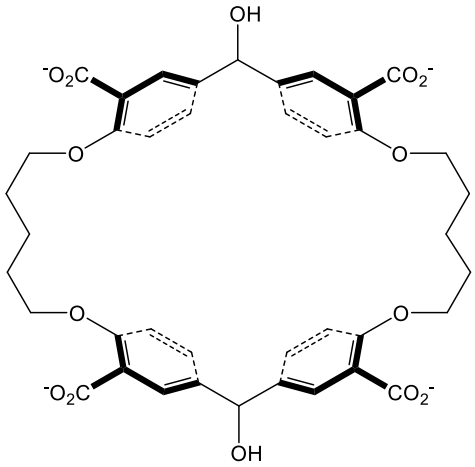
Collapsed walls

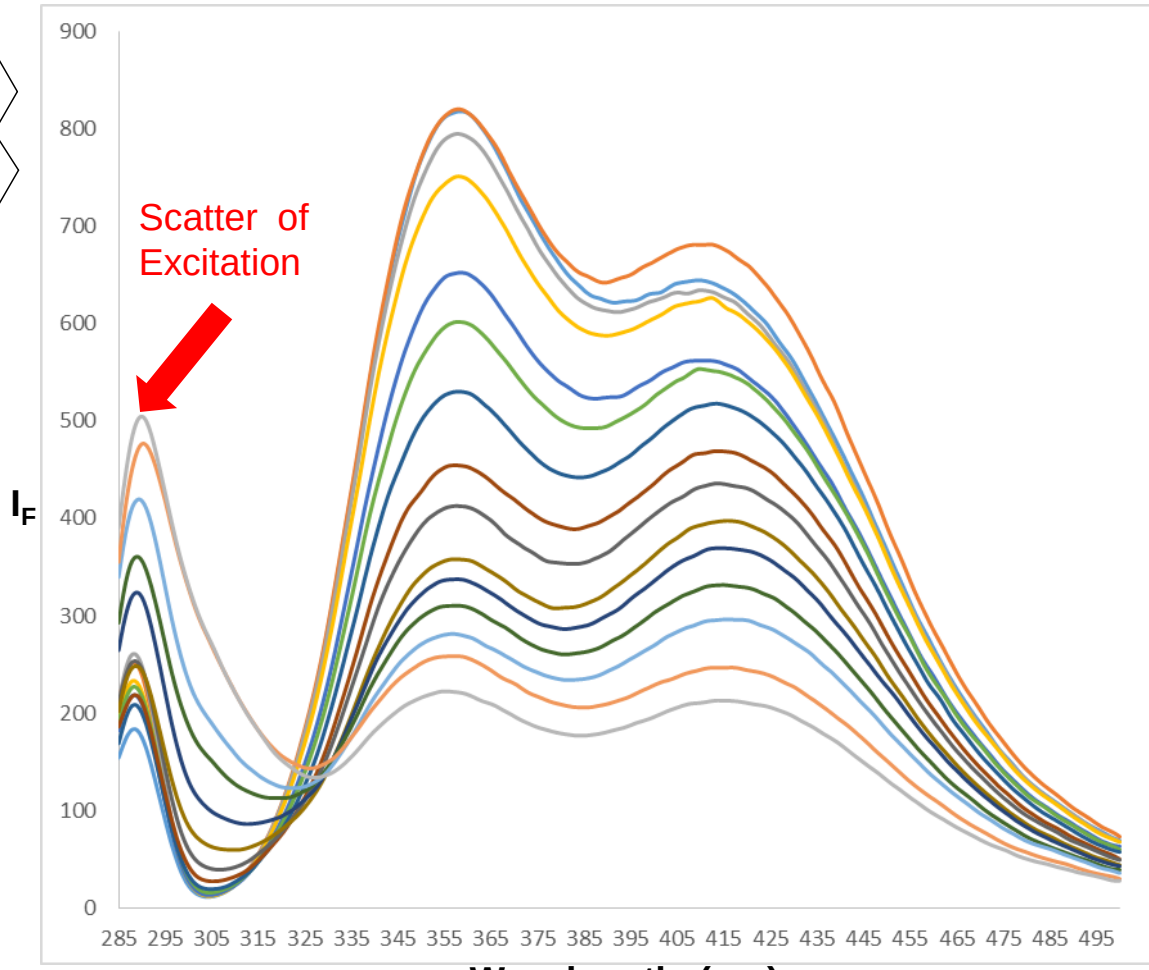


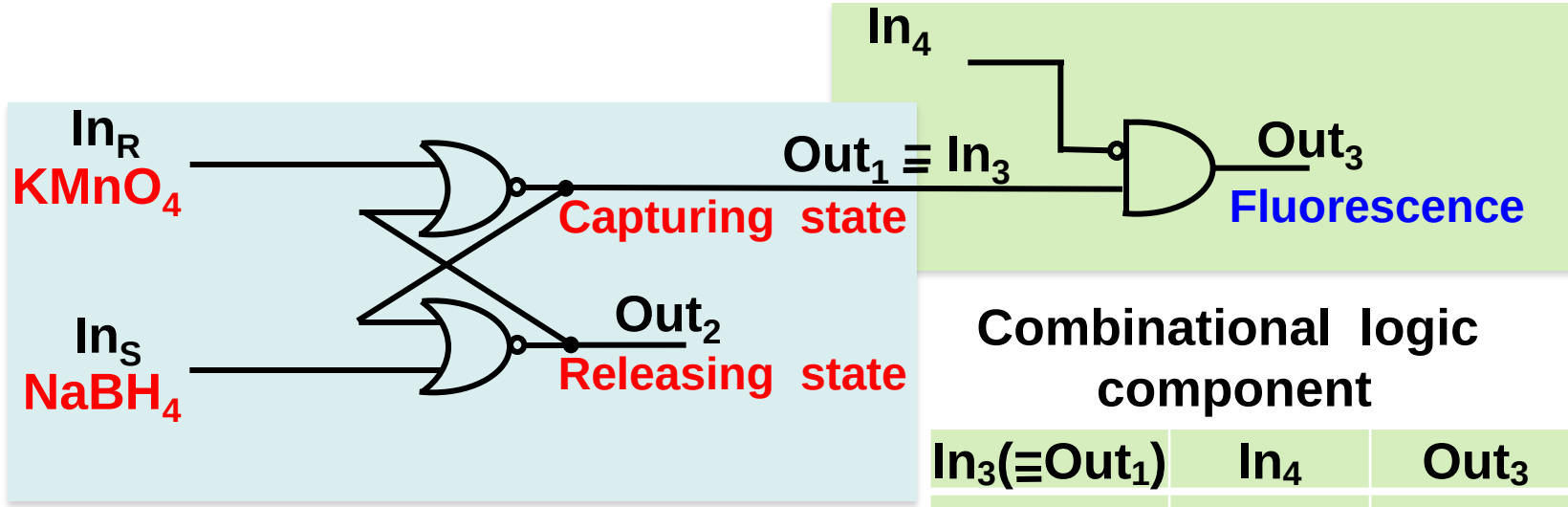




Luminescent self-indication of occupancy







Sequential logic Component (RS Flip-flop)

In_R	In_S	Out_1	Out_2
0	0	holds previous state	holds previous state
0	1	1	0
1	0	0	1

Combinational logic component

$In_3(\equiv Out_1)$	In_4	Out_3
0	0	0
1	0	1
0	1	0
1	1	0

New redox-switchable receptors for sensing and logic

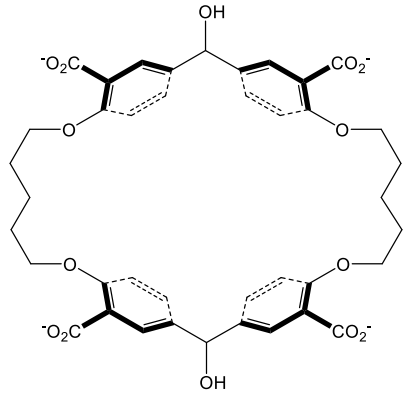
Brian Daly, Nimal Gunaratne, Tom Moody, Allen Huxley, Chao-Yi Yao, Ben Schazmann, Andre Alves-Areias, John Malone, Peter Nockemann *Nature Commun.* 2019, *10*, 49

Chao-Yi Yao, Hong-Yu Lin, Brian Daly, Yikai Xu, Warispreet Singh, Nimal Gunaratne, Wesley Browne (Groningen), Steven Bell, Peter Nockemann, Meilan Huang, Paul Kavanagh *J. Am. Chem. Soc.* 2022, *144*, 4977

Chao-Yi Yao, Hong-Yu Lin, Philip Morgenfurt (Dublin), Tia Keyes (Dublin) *Chem. Sci.* 2022, *13*, 10856

Hong-Yu Lin, Chao-Yi Yao, Jialu Li, Warispreet Singh, Nimal Gunaratne, Meilan Huang, Eric Anslyn *JACS Au* 2023, *3*, 2257

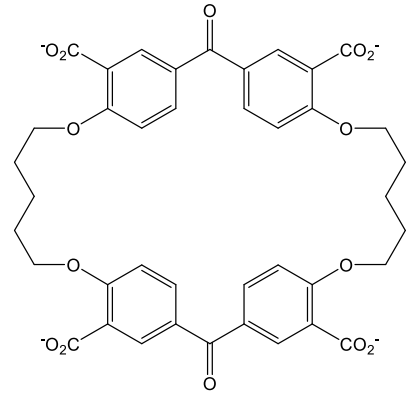
Erect walls



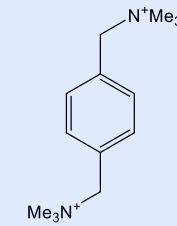
[Ox]

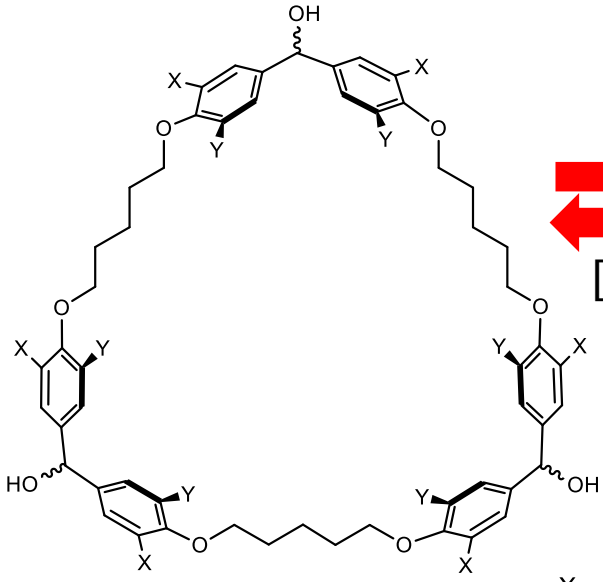
[Red]

Collapsed walls



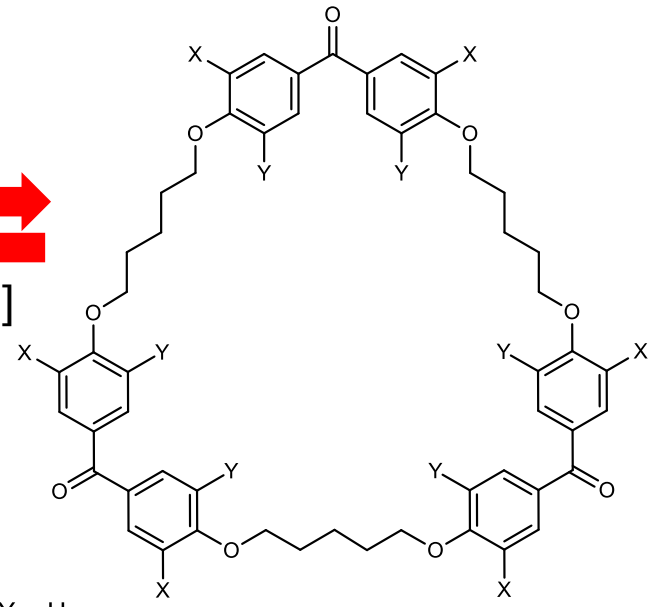
guest





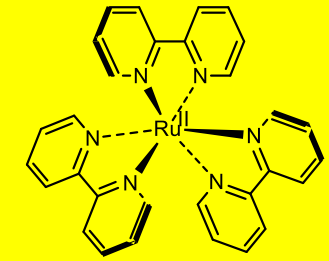
[Ox]

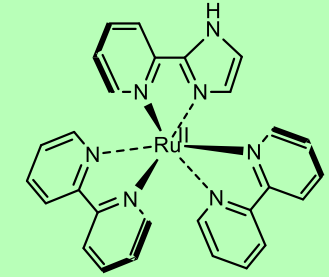
[Red]



X = CO_2^-, Y = H

X = CO_2^-, Y = CO_2^-






McCarroll, Woodruff, O'Halloran, Levine, Liu, Tiwari, Pati, Dyer, Wang, El-Bayoumi
Hall, Watkinson, Spring, Besley, Russell, Bull, Appleton, Resmini, Marken, Kuimova, Vilar, Goldup, Faulkner
Holdt, Plenio, Moro, Schalley, Nau, Langhals, Wurthner
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Houston,
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Ramamurthy, Nandhakumar,
Kolandaivel, Debsharma,
Prasad
Rashatasakhon,
Niamnont,
Wanichacheva
Pfeffer, Bell, Tuck

After 40 years of work…

Established Fluorescent PET Sensing - a general research tool for chemical, cellular and organism research.

***J. Chem. Soc., Chem. Commun*. 1985, 1669.**

Helped a multinational corporation to build the OPTI blood gas and electrolyte analyzer.

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Invented Molecular Logic-based Computation – a new field which bridges chemistry and computer science to shed light on the informational basis of life processes at the level of molecules, cells and organisms.

***Nature* 1993, *364*, 42.**

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Is that all? 40 years wasted?

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Over 1200 labs have joined.

Helped a multinational corporation to build the OPTI blood gas and electrolyte analyzer.

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Chemistry module sales >1.1B $. Helped save 1000s of lives.

Invented Molecular Logic-based Computation – a new field which bridges chemistry and computer science to shed light on the informational basis of life processes at the level of molecules, cells and organisms.

***Nature* 1993, 364, 42.**

Over 1700 labs have joined.

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Alexander, Jones, Steed, Cheong,
Chatterton, Bligh, Marken, Nitschke,
Turberfield, Frost, Phillips, Utley,
Buck, Thanh, Howorka, Mehl, Armes
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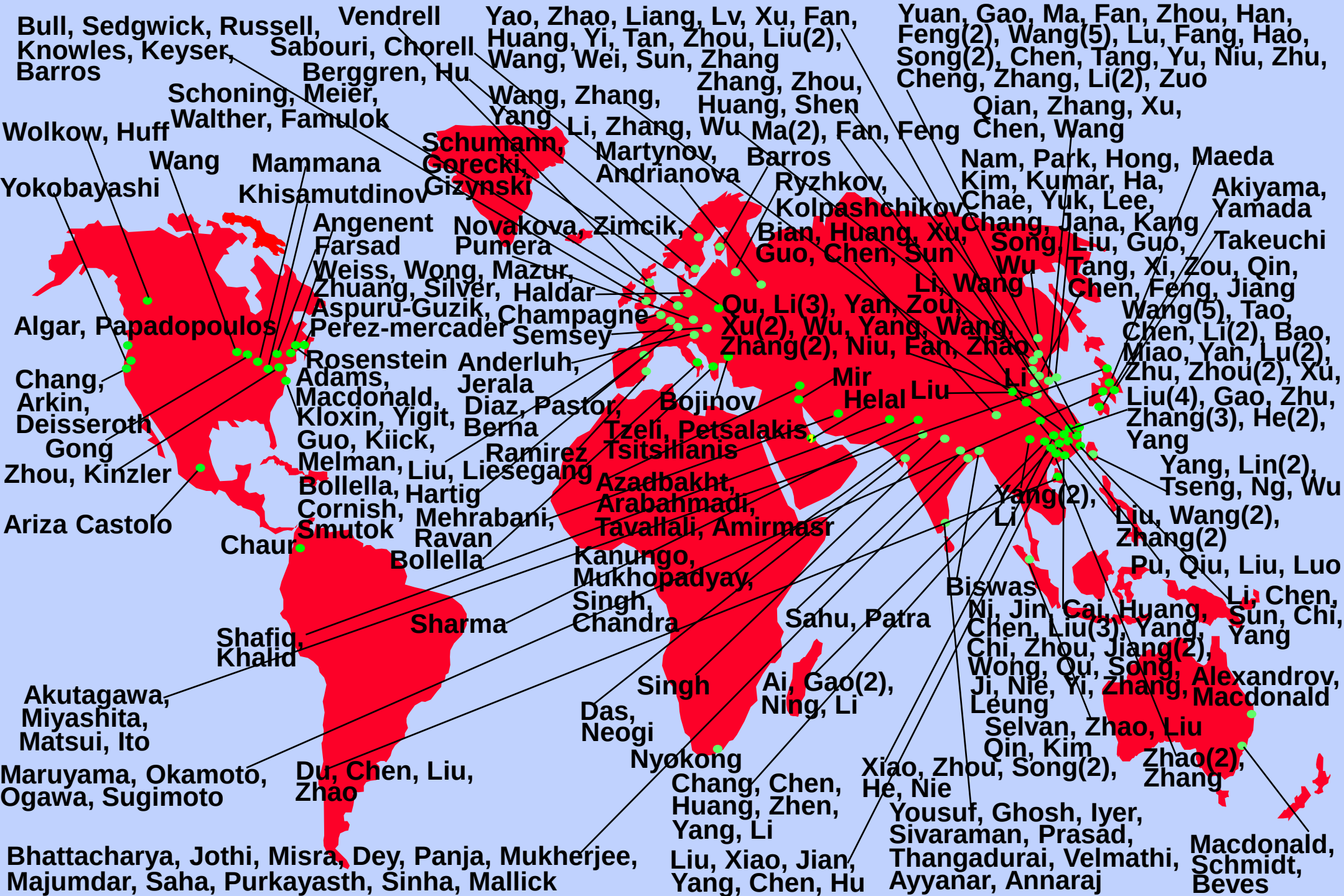
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Mechanism

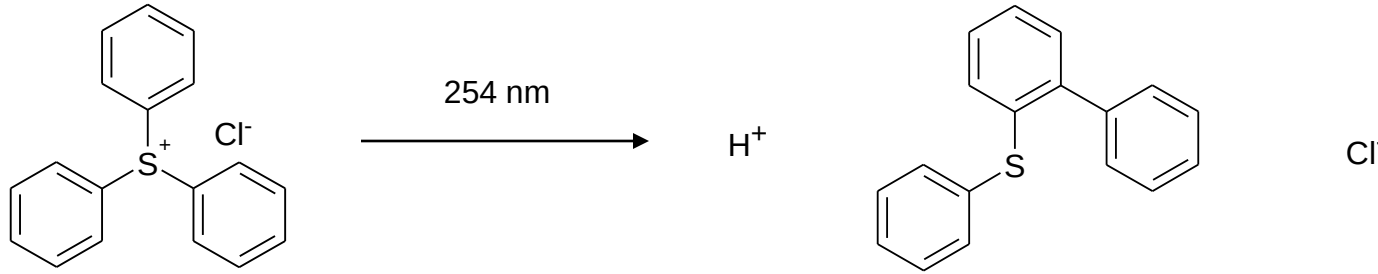
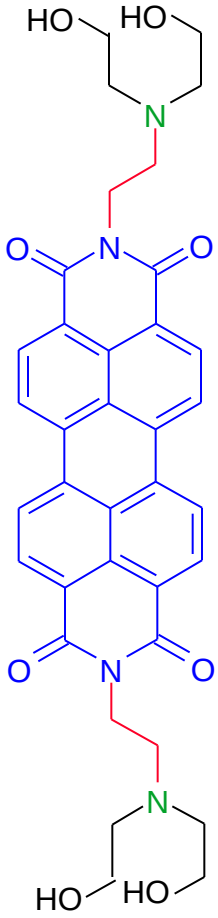


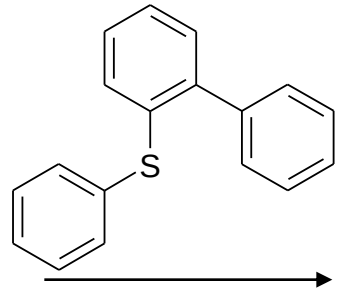
Photo acid generator

Fluorescence quencher via bimolecular PET



H^+

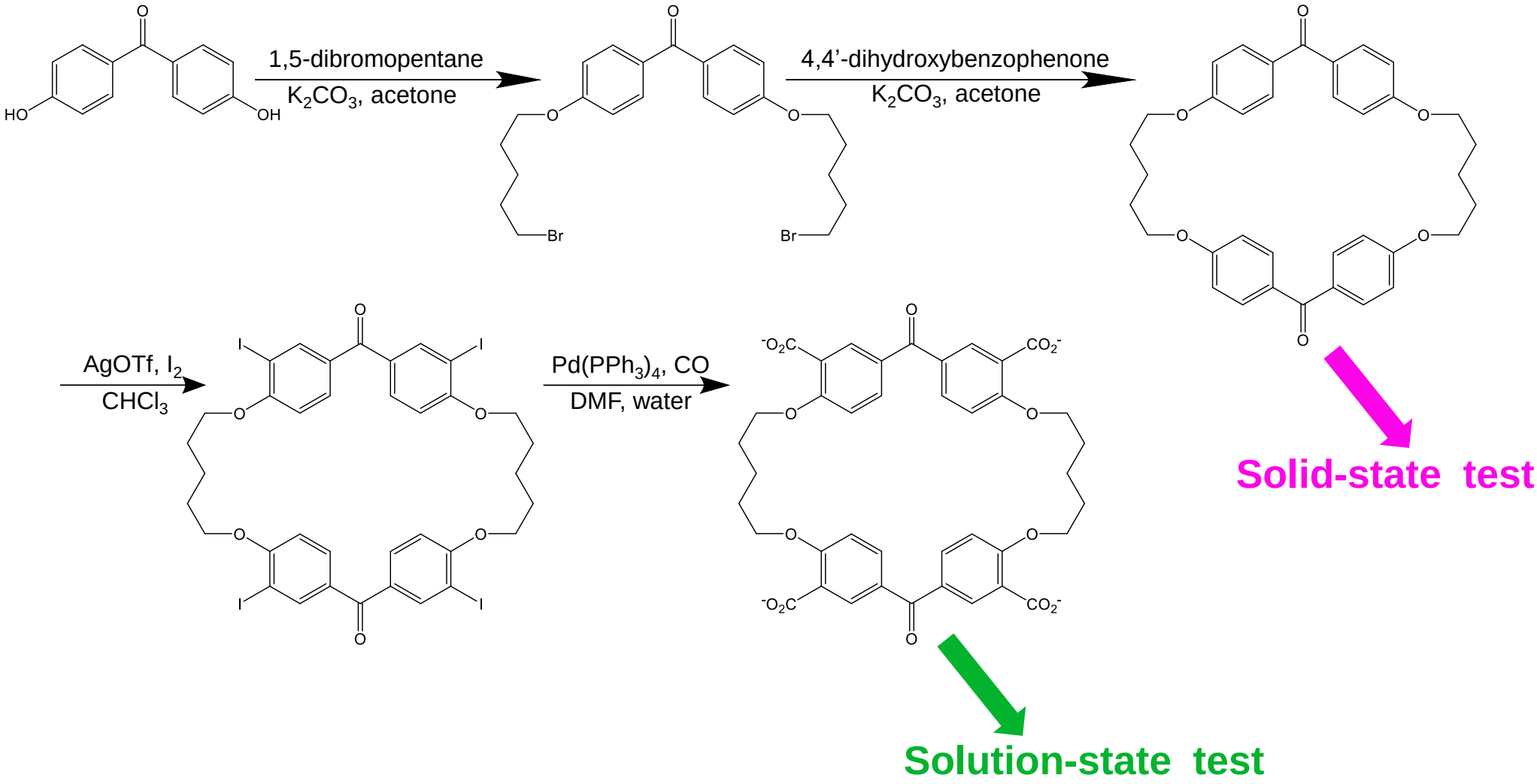
H^+-driven fluorescent PET sensor in ON state in acid solution



H^+-driven fluorescent PET sensor in OFF state in acid solution via bimolecular PET

H^+-driven fluorescent PET sensor in OFF state via unimolecular PET in weakly basic aqueous buffer

‘Off-on-off’ can be XOR logic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4,4'-dihydroxybenzophenone
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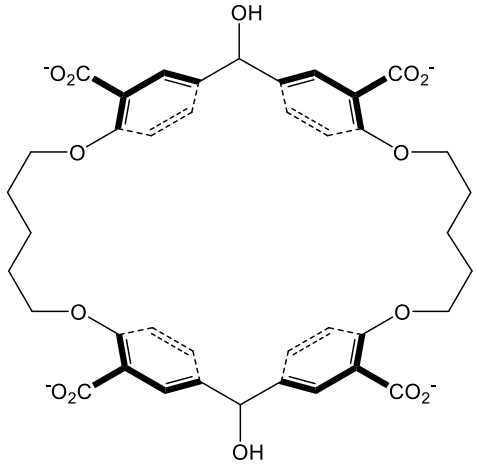
AgOTf, I2
CHCl3
Pd(PPh3)4, CO
DMF, water
Solid-state test
Solution-state test

alone

with



with