BioSystems 126 (2014) 12–26

Contents lists available at ScienceDirect

BioSystems

journal homepage: www.elsevier.com/locate/biosystems

Photochromic molecular implementations of universal computation

Jack C. Chaplin[a,b], Natalio Krasnogor[c,*], Noah A. Russell[a,**]

[a] Neurophotonics Lab, Schools of Biology, and Electrical and Electronic Engineering, University of Nottingham, Nottingham NG7 2RD, UK
[b] Institute for Advanced Manufacturing, Faculty of Engineering, University of Nottingham, Nottingham NG7 2RD UK
[c] Interdisciplinary Computing and Complex BioSystems (ICOS) Research Group, School of Computing Science, Newcastle University, Newcastle NE1 7RU, UK

ARTICLE INFO

Article history:
Received 2 June 2014
Received in revised form 11 August 2014
Accepted 3 September 2014
Available online 3 October 2014

Keywords:
Elementary cellular automata
Molecular switches
Photochromic molecules
Turing machines
Unconventional computing

ABSTRACT

Unconventional computing is an area of research in which novel materials and paradigms are utilised to implement computation. Previously we have demonstrated how registers, logic gates and logic circuits can be implemented, unconventionally, with a biocompatible molecular switch, NitroBIPS, embedded in a polymer matrix. NitroBIPS and related molecules have been shown elsewhere to be capable of modifying many biological processes in a manner that is dependent on its molecular form. Thus, one possible application of this type of unconventional computing is to embed computational processes into biological systems. Here we expand on our earlier proof-of-principle work and demonstrate that universal computation can be implemented using NitroBIPS. We have previously shown that spatially localised computational elements, including registers and logic gates, can be produced. We explain how parallel registers can be implemented, then demonstrate an application of parallel registers in the form of Turing machine tapes, and demonstrate both parallel registers and logic circuits in the form of elementary cellular automata. The Turing machines and elementary cellular automata utilise the same samples and same hardware to implement their registers, logic gates and logic circuits; and both represent examples of universal computing paradigms. This shows that homogenous photochromic computational devices can be dynamically repurposed without invasive reconfiguration. The result represents an important, necessary step towards demonstrating the general feasibility of interfacial computation embedded in biological systems or other unconventional materials and environments.

1. Introduction

Conventional computing involves the implementation of algorithmic processes to manipulate data on electronic hardware using binary logic. Unconventional computing on the other hand, uses new logical paradigms and new materials to build computational devices (Calude et al., 1998). Changing the physical materials used to compute opens up the possibility of embedding computers into biological systems at either a physiological or a cellular level. This could be achieved using biologically compatible molecular switches, for example. To demonstrate the feasibility of this possibility we have recently implemented registers and logic gates using photochromic molecular switches (Chaplin et al., 2012). Photochromic molecules (Exelby and Grinter, 1965) are a species of molecule with multiple stable forms. They can be reversibly switched between forms via the absorption of electromagnetic radiation. Spiropyrans are an example family of such photochromic molecules. (Berkovic et al., 2000). They possess a colourless leuco spiropyran form (SP) and a coloured trans-merocyanine form (MC). The transition of a sample of spiropyran molecules predominantly occupying the SP state to the MC state is called colouration, and the reverse is called decolouration.

One such spiropyran molecule is NitroBIPS (1′,3′-dihydro-1′,3′,3′-trimethyl-6-nitrospiro[2H-1-benzopyran-2-2′-2H-indole] or 6-nitro-BIPS or NBIPS). NitroBIPS is a spiropyran with a nitro group on the 6-position of the benzopyran section (Görner et al., 1996; Lenoble and Becker, 1986). The SP form absorption spectrum of NitroBIPS has its peak at ultraviolet wavelengths while the MC form absorption spectrum is in the visible range with a peak at green (Wohl and Kuciauskas, 2005), causing a solution predominantly in the MC state to appear purple or pink.

Absorption of green light by the MC form molecule generates the excited MC* fluorescent form, some molecules of which may immediately decay back to the MC form with the emission of a red photon (Görner, 1997), or undergo isomerization back to the SP

---

[*] Corresponding author: for matters of computer science. Tel.: +44 19 1208 5035.
[**] Corresponding author: for matters of engineering and photophysics. Tel.: +44 11 5846 8847.

E-mail addresses: jack.chaplin@nottingham.ac.uk (J.C. Chaplin), natalio.krasnogor@newcastle.ac.uk (N. Krasnogor), noah.russell@nottingham.ac.uk (N.A. Russell).

http://dx.doi.org/10.1016/j.biosystems.2014.09.004
0303-2647/

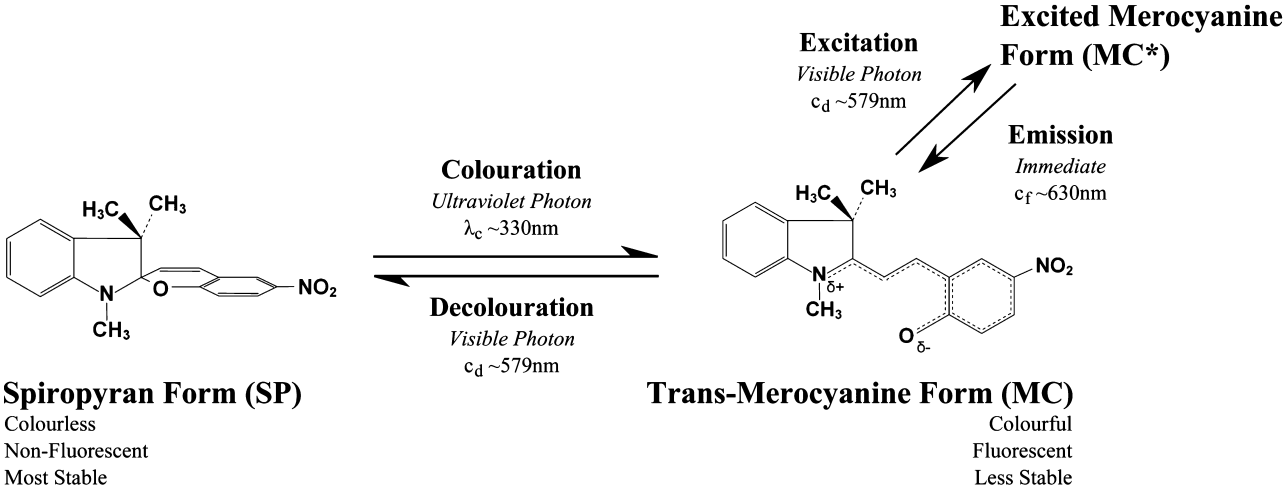

Fig. 1. The two stable forms of NitroBIPS and the transitions between them. As the spiropyran form is the most thermodynamically stable form, a population of NitroBIPS molecules will tend towards a majority-spiropyran equilibrium in the dark. A more detailed transition diagram can be found in Chaplin et al. (2012).

form. A small number of the excited MC* molecules will undergo irreversible bleaching. As fluorescence emission only occurs when the molecule is in the MC* form, the population of MC-form molecules can be estimated by the relative intensity of the emitted fluorescence signal when the sample is exposed to green light. These transitions are illustrated in Fig. 1. NitroBIPS is solvatochromic so the peak wavelength of absorption and the quantum yields (defined as the proportion of absorbed photons that cause a change in molecular form) for each transition are solvent dependent. Note that colouration and decolouration transitions also occur thermally. The SP to MC transition requires more energy than the reverse, so the majority of molecules are in the SP form at equilibrium at room temperature. The advantages of NitroBIPS (and other spiropyrans) compared to other species of photochromic molecules are that they possess higher quantum yields (Petchprayoon and Marriott, 2010), can be used with a wide variety of solvents (Görner and Matter, 2001) and are biologically compatible (Aizawa et al., 1977; Sakata et al., 2005b; Koçer et al., 2005; Ohya et al., 1998). However the MC form of NitroBIPS is subject to thermal relaxation (defined as the spontaneous, thermally driven reversion of a sample of molecules to an equilibrium state. For NitroBIPS the majority of molecules are in the SP form at equilibrium) at a moderate rate relative to other spiropyrans (Wojtyk et al., 2000).

We have previously shown that NitroBIPS can be used to implement registers and logic gates (Chaplin et al., 2012). Here we extend on this preliminary work and demonstrate an unconventional implementation of both parallel registers in the form of a Turing machine tape, as well as elementary cellular automata. Turing machine tapes and elementary cellular automata have been implemented as they both represent universal computing paradigms.

Alan Turing provided the first detailed theoretical description of a simple computational device that was capable of running any algorithm (Turing, 1936). Another simple universal computational device, which has been proven to be Turing complete (Cook, 2004), is the elementary cellular automaton. Elementary cellular automata consist of a one-dimensional array of ‘cells’ where the states of all cells are updated in parallel during each generation. Cells are updated according to their current state, the state of its two immediate neighbours and a predefined set of rules (Wolfram, 1983, 2002). Although Turing machines were introduced as mechanical models of mental processes, attempts have been made to mimic their structure via unconventional molecular processes (Qian et al., 2011; Rothemund, 1996; Shapiro and Karunaratne, 2001). Cellular automata, on the other hand, are typically used for modelling purposes (Nagel and Schreckenberg, 1992) but purpose specific hardware has been engineered to implement several versions of cellular automata (Shackleford et al., 2002; Gers et al., 1997).

The remainder of this paper is divided into five sections. Firstly we state the *Objectives* of this paper. Secondly, a *Methods* section which explains the production of NitroBIPS samples and details the illumination/detection hardware. The *Implementation* section, which builds on the theoretical work in (Chaplin et al., 2012), contains the theoretical basis for parallel registers, Turing machine tapes and cellular automata. Next, an *Experiments* section discusses the results of experiments carried out involving Turing machine tapes and elementary cellular automata. Lastly, a *Discussion* section discussing the strengths and weaknesses of this approach, and possible future directions.

2. Objectives

Our previous paper details the implementation of singular computational elements with photochromic molecules (Chaplin et al., 2012). The molecules were embedded in a polymer matrix and light pulses from LEDs were used to colourise and decolourise them. Emitted fluorescence was recorded with a photodiode. This allowed for data to be stored as the relative proportion of fluorescent molecules, and for logic gates and logic circuits to be executed by exploiting the floor and ceiling restrictions of the colourisation and decolourisation processes. The objective of the research in this paper was to expand upon this by designing and executing parallel computational elements, and implementing universal computation.

This paper builds multiple computational elements in parallel, by allowing a movable illumination area to address multiple regions in the polymer matrix. Parallel registers are implemented, and then expanded upon to implement the tape in a photochromic Turing machine. A combination of parallel registers and logic circuits are then used to implement elementary cellular automata, and hence a form of universal computation. Fig. 2 shows the relationship between these sections and previous work. The theoretical basis for each element is discussed first, followed by the experiments demonstrating these computational processes running on our experimental hardware.