



Fine Control and Selection of Travelling Waves in Inorganic Pattern Forming Reactions

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ABSTRACT

Under normal reaction conditions [AlCl_3 0.28-0.34M and NaOH 2.5M A.Volford et al.] spontaneous spiral and circular travelling precipitate waves were observed. We constructed a phase diagram for the reaction and identified a large controllable region at lower aluminium chloride levels. We show that it is possible to selectively initiate travelling circular waves and other self-organised structures within this controllable region. In previous work initiation was undertaken before adding the outer electrolyte resulting in disorganised waves. However, marking the gel one minute after adding outer electrolyte resulted in cardioid waves. Increasing the time interval to two minutes caused a transition to single circular waves. If the gel is marked sequentially nested circular waves (target waves) are formed. These reactions were used to calculate simple and additively weighted Voronoi tessellations. The fine control of self-organisation in precipitation reactions is of interest for the synthesis of novel and functional materials. [Article copies are available for purchase from InfoSci-on-Demand.com]

Keywords: *Chemical Computing; Control of Self-Organised Waves; Pattern Formation; Travelling Chemical Waves*

INTRODUCTION

When considering pattern formation in inorganic systems the majority of work spanning over 100 years has been focussed on Liesegang type reactions (Liesegang, 1896). These are periodic patterns formed due to non-equilibrium crystallisation and precipitation. There have

been many models proposed to explain the phenomenon but still none are able to predict robustly the position, spacing and morphology of the precipitate bands in the full range of chemical systems. For example reactions based on cobalt chloride and ammonium hydroxide exhibit chaotic dynamics due to redissolution of the chemical complex at the stratum coupled

with continued band formation ahead of the reacting front. This phenomenon means the entire pattern migrates within the tube (Ghoul and Sultan, 2001).

More recently there has been renewed interest in this class of chemical reactions and the control thereof in order to synthesise functional materials by utilising the inherent self-assembly mechanisms. Work has been carried out to apply electrical fields (Lagzi, 2002), magnetic fields (Sorensen and Madsen, 2000) and changing the conventional geometry of the reactor (Kuo, Lopez Cabarcos and Bansil, 1997). This work is still at an early stage of development and lacks a fundamental mechanistic understanding which would allow a quantum leap forward. Basically our understanding of crystallisation processes and molecular processes *per se* is very limited and to some extent limits the potential of this class of reaction. If better molecular computation and material synthesis via self-assembly is to be achieved then this knowledge gap must be eradicated. The recent discoveries of a series of very simple inorganic reactions exhibiting travelling wave phenomena serves to further highlight this issue (Hantz, 2000; Adamatzky, De Lacy Costello and Asai, 2005; Volford et al, 2008; De Lacy Costello, Ratcliffe and Hantz, 2004).

In terms of modelling inorganic pattern forming reactions a significant amount of work has been directed towards the modelling of sea shell patterns. Shells consist of calcified material secreted by the mantle, they then increase their size by accretion of new material at the “growing edge” of the shell. They show a diverse range of complex pigmentation patterns which can be modelled via cellular automata models (Wolfram, 1994), neural models (Murray, 1993) and reaction diffusion models (Meinhardt, 1995). Thus there is some driving force to study pattern formations in simple inorganic systems in order to gain a better understanding of pattern formation in natural systems. This is in addition to the need to understand the underlying mechanisms in order to exert high levels of control.

Recently a new class of inorganic reactions were discovered based on gels of copper

chloride reacted with sodium hydroxide (Hantz, 2000). These reactions exhibited a range of self-organised patterns only observed previously in more complex reactions such as the BZ reaction. This is highly significant given the very simple nature of the chemical reactants. Most striking was the formation of travelling cardioid and spiral precipitation waves. This phase of the reaction was further studied in (Adamatzky, De Lacy Costello and Asai, 2005) where the use of a different gel media and outer electrolyte (potassium hydroxide) enabled spiral wave evolution to be observed at much smaller length scales.

More recently another inorganic system has been identified based on aluminium chloride gels reacted with sodium hydroxide (Volford et al, 2007). This system is particularly remarkable as unlike the systems mentioned above (Hantz, 2000; Adamatzky, De Lacy Costello and Asai, 2005) the wave evolution can be easily observed in real-time.

In previous work we were able to exert control over another class of inorganic reaction in order to realise a useful computation (De Lacy Costello, Ratcliffe and Hantz, 2004). The reaction involved the addition of copper chloride to potassium ferricyanide immobilised in an agarose gel. At high concentrations of potassium ferricyanide circular expanding waves are spontaneously generated by heterogeneities. Where these waves collide they form a natural tessellation of the plane similar to those obtained in crystallisation processes (multiplicatively weighted crystal growth Voronoi diagrams). By reducing the concentration of potassium ferricyanide then the point and time at which circular waves are initiated could be controlled by marking the gel with a very fine glass needle. By using this methodology user defined precipitation patterns could be created. These user defined precipitation patterns equate to a calculation of a Voronoi tessellation of the plane.

In previous work (De Lacy Costello, 2008) involving the aluminium chloride sodium hydroxide reaction we adopted the same strategy and were able to show that initiation of waves

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