

Delving Deeper into Homeostatic Dynamics of Reaction Diffusion Systems with a General Fluid Dynamics and Artificial Chemistry Model

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Abstract

In this paper I present a general modelling framework for coupled fluid dynamics and chemistry problems, and apply it to the simulation of a series of complex, homeostatic reaction diffusion systems. The model can incorporate any number of chemical species and reactions. Those chemical species diffuse, react and are advected by fluid flows. I illustrate some characteristic results from the modelling of the Gray Scott reaction diffusion system with thermally resolved reactions. Extending my previous work on ecological dynamics of non-living structures, I demonstrate that thermal homeostasis of reaction diffusion spots can occur in systems without the use of the porous wall boundary condition that has traditionally been used for the Gray Scott system. I present an initial analysis of the parameter space of this system as well as detailing the mechanism behind the thermal homeostasis.

Introduction

Alongside the dissipation of free energy and autocatalytic growth, a fundamental property of the living state is the ability to regulate a subset of local variables to within windows of specific set points. This ability to self-regulate, encapsulated by the concept of homeostasis, distinguishes life from many non-living dissipative structures such as fire. Fire dissipates free energy and grows exponentially given ample supply of fuel and oxidant. What it is not able to do is perform any kind of feedback with its external environment such that its behaviour does not remove the conditions for its own existence. Once a forest is burned, the fire stops. An adaptive fire would sense any external decline in fuel supply and eventually tune its burning rate to match the rate of tree regrowth. That would allow it to persist far longer than its equivalent, ravenous cousin that consumes with disregard.

One of the key objectives of Artificial Life is to establish general conditions under which the living state emerges. Hence we continually seek simple model systems that exemplify properties associated with life, despite the given system being non-biological. Once all such necessary self-organisation processes are charted and understood in the abstract, we should then be able to map them to more realistic and geologically plausible conditions, and thus attempt to

re-construct the key chapters in the history of abiogenesis on this planet and perhaps others.

Much of my past work has ventured in this direction, and in previous papers I have illustrated the emergence of competition (Bartlett, 2014; Bartlett and Bullock, 2015) and homeostasis (Bartlett and Bullock, 2016) within systems of reaction diffusion (RD) structures. These patterns are spatial instabilities, akin to solitons, manifested as distinct shapes in the concentration profiles of chemical species in a 2-dimensional domain. Their existence serves to channel free energy between two imaginary reservoirs at fixed chemical potential. The original model RD systems were introduced by Gray and Scott (1985, 1994), though we can of course trace the RD concept back to Turing (1952).

They represented a seminal new class of non-linear chemical system, which inspired a wave of theoretical and experimental work (see e.g., Awazu and Kaneko, 2004; Lee et al., 1993, 1994; Lee and Swinney, 1995; Mahara et al., 2008; Pearson, 1993; Virgo, 2011). One of the most striking features of these systems, under certain parameter regimes, is the emergence of self-replicating spot patterns, strongly reminiscent of the multiplication of single-celled organisms (often when I play animations of RD systems to the uninitiated, their first question is whether they are looking at a video of bacteria). It was these systems that I experimented with using simulations in previous work. I found that multiple sets of spot species could spontaneously regulate their local temperature, using a primitive form of reign control (Bartlett and Bullock, 2016), similar to the manner in which populations of Daisies regulate temperature in the Daisy-world model (Watson and Lovelock, 2011; Wood et al., 2008).

This and similar works allow us to consider general, primitive means by which physical and chemical dynamics can lead to self-regulation. In order to explore such phenomena further, it is necessary to have modelling techniques that are physically realistic but also permit a large number of degrees of freedom. This was my primary motivation for developing a reactive, thermal lattice boltzmann model (RTLBM) capable of handling a vast range of chemistries, while also retain-

ing a faithful coupling to the hydrodynamics of the system.

In this work I will present this very general fluid dynamics and artificial chemistry model, and use it to extend my previous work on homeostasis in the Gray Scott system. Regulating a single variable using two populations of individuated structures was a significant result. However the thermodynamic gradients placed across those systems were somewhat constrained. They used the standard, homogeneous porous wall (PW) boundary condition (BC). In order to have full control over the geometry and spatial orientation of the system's gradients, it was necessary to find an alternative set of BCs, such that supply and removal of chemical species could be arbitrarily adjusted. Thus I will present a method for achieving this wherein two extra sets of chemical species are introduced. The inflow and outflow of these two species defines the chemical potential dissipated by the system, and it can be adjusted in any desirable way. I will demonstrate the reproduction of my previous results on thermal homeostasis and go on to present a first parameter space mapping of this system's behaviour.

In the following section I will outline the equations of motion that are solved by my numerical method. Precise modelling details can be found elsewhere (Bartlett, 2014; Bartlett and Bullock, 2015, 2016). I then present results of my simulations including steady state configurations, population dynamics and how resilience varies with two key parameters of the system. I discuss the results and suggest directions for future work in the final two sections.

Model System

The simulations in this work make use of the RTLBM (for full details, please see Bartlett, 2014; Bartlett and Bullock, 2015, 2016), that was developed as an extension of similar models (Ayodele et al., 2011, 2013; Frouzakis, 2011; He et al., 1998; Peng et al., 2003; Shan, 1997). In this section, I will simply outline the governing equations for all the variables simulated.

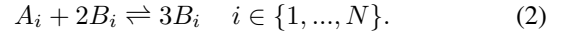
In all cases, the system in question is a rectangular two dimensional fluid with a 4:1 aspect ratio bounded by solid walls enforcing the no-slip velocity BC. The flow of the fluid obeys the standard, incompressible Navier-Stokes equation:

$$\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla) \mathbf{u} - \nu \nabla^2 \mathbf{u} = \beta g (T - T_0) \hat{j}, \quad (1)$$

where \mathbf{u} is the fluid velocity, ν is the kinematic viscosity, β is the thermal expansion coefficient, g is the acceleration of gravity, and T and T_0 are the local and mean temperature, respectively. The term on the right hand side is the gravitational body force due to buoyancy effects (causing convective motion under sufficiently strong temperature gradients). Incompressibility eliminates divergence of the velocity field: $\nabla \cdot \mathbf{u} = 0$.

Let us now define a scheme for representing chemical reactions. For Gray Scott RD systems, we assume that

the non-linear reaction responsible for pattern formation is present:



Note that there is the possibility to include not just one or two, but N such 'spot species' and that the reaction is reversible. I retain the reversibility for the sake of generality. For the above reaction, the reverse direction is forced to be negligibly slow by imposing a very low frequency factor. Allowing a stronger reverse reaction would warrant its own dedicated investigation, which can be taken up in the future.

Instead of using the standard PWBCs, in this work I will use an extra two sets of chemical species (two extra chemical species per spot species), which will produce an equivalent supply and extraction effect. There are several motivations for making this change. Firstly, the PWBCs introduce their own negative feedback into the system since they act to maintain the concentration of the A_i species close to 1. It would be useful to be able to isolate and remove this negative feedback effect to assess whether other negative feedbacks are sufficient to stabilise a given set of patterns. Secondly, it allows all the dynamics of the system to fall within the classes of reaction, diffusion, and fluxes at outer boundaries. Inward/outward fluxes at every point in the system of different chemical species would be difficult to implement experimentally, and don't extend naturally to 3D systems (if we were envisaging that system being re-created in the lab). Thirdly, it allows us to clearly visualise flows of material across the system in an easily recognisable way.

It will also allow us to experiment with, for example, systems where there are no external material fluxes but only energy fluxes (e.g. heat flow). The material within the system would be allowed to pass through cycles of reactions (reversibility of the reactions would be crucial), probably passing through exothermal transitions in low temperature regions and endothermic transitions in higher temperature regions. We can then experiment with the effect that complex pattern formation has on the heat transport abilities of a system (interactions with convective flow structures).

The more general BCs presented in this work allow an arbitrary range of spatially directed gradients. With PWBCs the gradient in chemical potential that drives the system is homogeneous and ubiquitous. But with the system outlined in this work, we can implement gradients of arbitrary structure and direction including finite 2D regions, lines, points, partial boundaries, vertically-oriented, aligned or anti-aligned with temperature gradients, whatever we wish. The PW conditions represent a constraint on the system. I will now illustrate how it can be relaxed.

Let us first address the supply of species A_i , that is conventionally handled by the supply BC. We can use the following simple source reaction:



These two reactions (forward and reverse) must mimic the effect of the $F(1 - \psi_{A_i})$ supply term of the standard Gray-Scott system (where ψ_{A_i} is the concentration of species A_i and $F = 0.03$ is the ‘feed rate’). This is achieved by setting the boundary and initial concentrations such that:

$$\langle \psi_{S_i} \rangle \gg \langle \psi_{A_i} \rangle \quad (4)$$

In the present work, the value $\langle \psi_{S_i} \rangle \approx 100 \langle \psi_{A_i} \rangle$ was used. We must also set the rate constants of the reaction such that:

$$k_{f_s} = A_{f_s} \psi_{S_i} e^{-E_{f_s}/T} \approx F \quad (5)$$

where A_{f_s} is the frequency factor and E_{f_s} is the activation energy (see below).

The reverse rate is set to:

$$k_{r_s} = A_{r_s} \psi_{A_i} e^{-E_{r_s}/T} \approx F \psi_{A_i}. \quad (6)$$

Thus the reaction of S to A proceeds at a rate F , and the backward reaction, A to S , proceeds at a rate $F \psi_{A_i}$, as required.

To recreate the effects of the extraction boundary condition, we simply use a decay reaction:



where the rate constants in this case are set to:

$$k_{f_w} = A_{f_w} \psi_{B_i} e^{-E_{f_w}/T} \approx (F + R) \psi_{B_i} \quad (8)$$

where $R = 0.061$ is the ‘removal rate’. In the reverse direction, we have:

$$k_{r_w} = A_{r_w} \psi_{W_i} e^{-E_{r_w}/T} \ll k_{f_w} \quad (9)$$

The most convenient way to achieve the above condition is simply to set A_{r_w} to a very low value.

Note that it is most straightforward to limit the temperature dependence of the supply and extraction reactions by setting their activation energies to very small fractions of the average system temperature. It would be an interesting experiment to explore the effects of different activation energies for these reactions but that will not be addressed in the present work.

It is straightforward to introduce an arbitrary number of extra chemical species and reactions to the model. In the general case, the concentration field of chemical species C_j will obey the following reaction-diffusion-advection equation:

$$\frac{\partial \psi_{C_j}}{\partial t} = D_{C_j} \nabla^2 \psi_{C_j} - \nabla \cdot (\mathbf{u} \psi_{C_j}) - \sum_{k=1}^R A_k e^{-E_k/T} M_j [\alpha'_{kj} - \alpha^*_{kj}] \prod_{l=1}^{N_s} \psi_{C_l}^{\alpha'_{kl}} \quad (10)$$

where R is the total number of reactions (note that reverse reactions are considered as extra, separate reactions from

their forward counterparts), M_j is the relative molecular mass of species j (for the sake of simplicity, these can be set to unity as long as the stoichiometry of the reactions conserves mass overall), N_s is the total number of chemical species, α'_{kj} is the left hand side stoichiometric coefficient for reaction k and chemical species j (the number of molecules of species j entering as reactants into reaction k), and α^*_{kj} is the right hand side stoichiometric coefficient for reaction k and chemical species j (the number of molecules of species j leaving as products from reaction k). Note that the concentrations of any of the species can be fixed at the boundaries or indeed any point in the domain (we may of course choose to vary those controlled concentrations as a function of time or horizontal coordinate). Alternatively we could fix the flux of any chemical species at any point in the system as well.

If we wish to recreate the self-replicating spot system, it will be necessary to keep the boundary concentration of the source species at a high value $\psi_{S_{i_b}} = 100$. The formation of chemical structures consumes the source species and it can thus become limited by the rate of inward diffusion of that source species. Therefore in this work I reduce the rates of the reactions in Equation 2 by a factor of 10, and increase the length of the simulation runs by the same factor (while also reducing the diffusivities of species A and B). As an alternative, one could also use higher ratios of source species to reactant (e.g. $\langle \psi_{S_i} \rangle \approx 1000 \langle \psi_{A_i} \rangle$).

Finally, we can turn to the advection, diffusion, production and consumption of internal energy:

$$\frac{\partial T}{\partial t} = \chi \nabla^2 T - \nabla \cdot (\mathbf{u} T) - \sum_{k=1}^R A_k e^{-E_k/T} \Delta H_k \prod_{l=1}^{N_s} \psi_{C_l}^{\alpha'_{kl}} \quad (11)$$

where χ is the thermal diffusivity and ΔH_k is the enthalpy of reaction for reaction k (following sign convention, $\Delta H_k < 0$ implies an exothermic reaction).

The numerical framework outlined above allows us to explore an arbitrary range of complex chemical systems. We could even introduce extra interactions such as influences upon diffusivity by chemical species (e.g. the presence of species C_i could reduce the diffusivity of species C_j , or species C_k could cause a reduction in the viscosity of the fluid). Given the generality of the model described above, it will be impossible to systematically and comprehensively explore the parameter space of even fairly simple systems. I will attempt a first mapping of one plane of the parameter space here.

Results

I will first present and discuss some thermal stability simulations to illustrate the similarities and subtle differences between the new system and the standard PW version that

was used previously. With the new BCs, the supply of the source species S_i is relatively slow, since it relies on diffusion from the boundaries. The diffusivity of a given chemical species in the RTLBM cannot be set arbitrarily high. One solution to this issue would be to force the concentration of S_i to its required value at every point in the domain at every time step. However that would defeat the objective of avoiding PWBCs. Thus the diffusivity of the S_i species was set to a high value and the rate of all reactions were reduced by a factor of 10 (as described in the previous section). This caused a general reduction in the pace of the dynamics, but allowed species S_i to diffuse from the boundaries such that the familiar pattern-formation of the conventional system was retained (longer simulation times were another inevitable consequence).

Single Species, Stable Temperature

In this first configuration, there is a single, thermally neutral ($\Delta H = 0$) spot species. The boundary temperature of the system is kept at the hospitable value of $T_B = 1.5$. The steady state configuration of this system is shown in Figure 1.

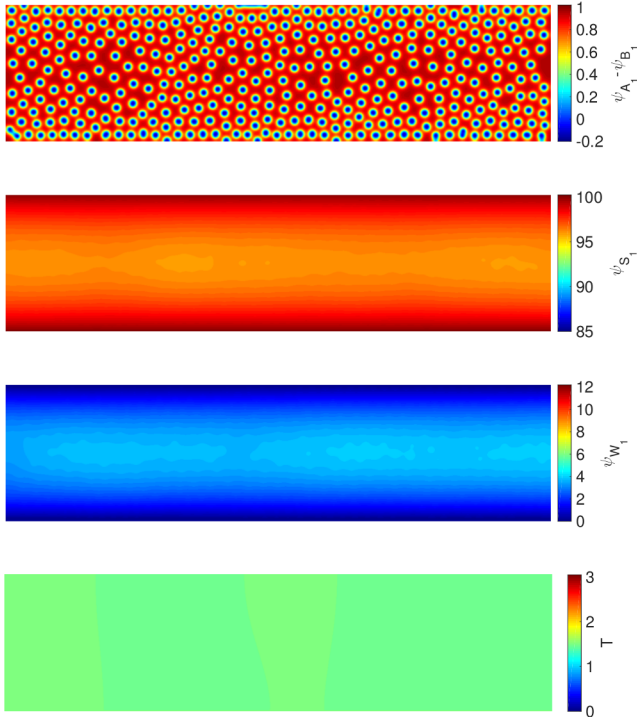


Figure 1: Steady state of a thermal reaction diffusion simulation with a single spot species. The non-linear reaction does not emit or absorb heat. The top figure shows the chemical order parameter, the second figure shows the concentration of the supply species S_1 , the third shows the concentration of the waste species W_1 , and the bottom figure shows the temperature.

We see that with these BCs, the structures can replicate and fill the space without hindrance. However, without the homogenous, PWBCs, the spots rely upon the supply of S_1 to supply the reaction that is key to their structure formation. This means the concentration of S_1 is lowest in the centre of the domain, furthest from the boundaries. In steady state, the concentration remains at $\sim 95\%$ of its optimal value, and hence the spot patterns can persist, albeit at a slightly lower density than those regions closer to the boundary. If it were to drop significantly lower than this, then it would potentially prevent the stable formation of the spots. The waste species shows a concentration excess in the centre of the domain, as we would expect.

Single Species, Variable Temperature

Let us now consider the influence of temperature on such a system. In this first experiment, the temperature was varied horizontally from the low value of $T_B = 0$ on the leftmost edge to the high value of $T_B = 3$ on the right. The steady state of this system is displayed in Figure 2.

As was the case with PWBCs, the spot patterns are only stable within a narrow range of temperature. However, there is an additional feature with the current BCs, caused by the combination of the horizontal temperature gradient and the vertical profile of ψ_{S_1} . As we move right from the centre of the domain, the temperature becomes higher, and hence you would expect the state to transition rapidly through worm-like phases to a lamellar phase and then to the trivial case of the reaction occurring everywhere homogeneously. These changes of phase happen over a longer spatial extent than in the PWBC case, because the supply of S_1 becomes a limiting factor, and as we see, S_1 is heavily depleted on the right side of the figure, where the reaction is proceeding at its maximum rate. If S_1 could diffuse instantaneously, the phase profile would be compressed horizontally.

Instead of keeping the temperature constant in time and varying in space, we can keep it uniform along the two boundaries, but vary it in time. If the non-linear reaction (between A and B) remains thermally neutral ($\Delta H = 0$), introducing a sinusoidal variation in boundary temperature quickly becomes fatal for the chemical structures. The reaction rate is strongly dependent upon temperature and once all patterns disappear due to the reaction grinding to a halt at an excessively low temperature for example, they do not reform once the temperature is raised again.

Homeostasis

Let us now establish that the new model (no PWBCs) exhibits the same homeostatic dynamics as seen in previous work (Bartlett and Bullock, 2016). We now take a system with two spot species and their associated supply and waste substances ($N_S = 8$ individual chemical species in total). The non-linear reaction for the first spot species is set to be exothermic with $\Delta H_1 = -0.2$, while the reaction of the

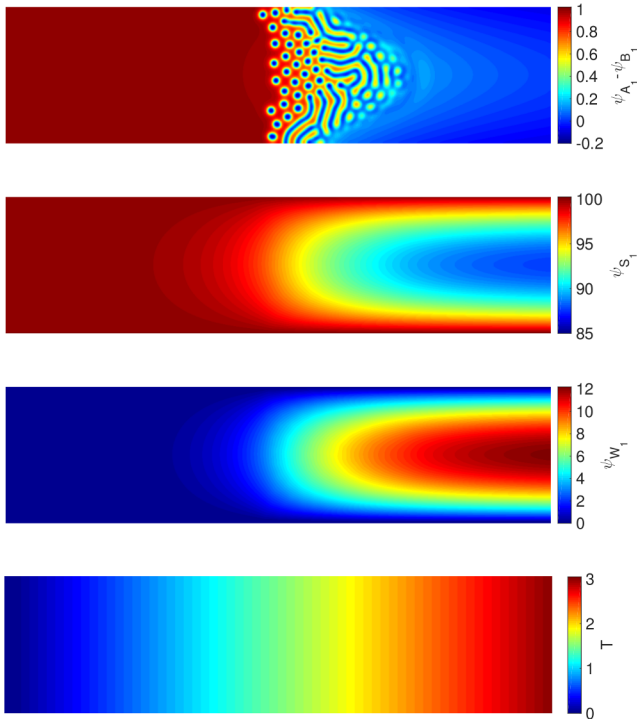


Figure 2: Steady state of a thermal reaction diffusion simulation with a single spot species. The driving reaction does not emit or absorb heat. The top figure shows the chemical order parameter, the second figure shows the concentration of the supply species S_1 , the third shows the concentration of the waste species W_1 , and the bottom figure shows the temperature.

second spot species is endothermic with $\Delta H_2 = 0.25$. In a system with a horizontally varying temperature that remains constant with time, the combined spot system actively increases its viable range compared with the thermally neutral equivalent. Figure 3 displays the steady state of this system.

The spot patterns are clearly a strong attractor for this system, along with the temperature of $T_m \approx 1.5$. These structures have been able to colonise areas that would normally be inhospitable to them.

Let us now view this homeostasis at work when the temperature is kept spatially homogeneous but varied in time. The boundary temperature is varied sinusoidally and produced the population dynamic shown in Figure 4(a).

We see that despite the change in BC, the characteristic homeostatic balance still plays out, wherein the exothermic spots outnumber those that are endothermic when the temperature is lower than optimal, and vice versa when the temperature is higher than optimal (though it seems in this case, in the high temperature regime, the two spot populations are quite close to one another). The variation of the internal temperature is dramatically reduced and it is always feasible

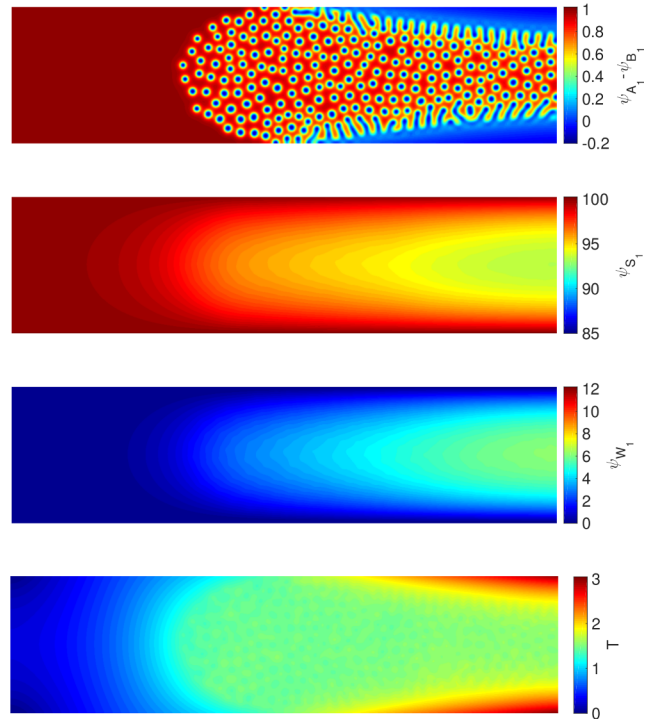


Figure 3: Steady state of a thermal reaction diffusion simulation with two spot species. The first spot species has an exothermic reaction, while the second is endothermic (only the exothermic spot species is shown in this figure, since both species exhibited very similar spatial distribution). The first figure shows the chemical order parameter, the second the concentration of the supply species S_1 , the third shows the concentration of the waste species W_1 , and the bottom figure shows temperature.

for both sets of spots to persist.

Let us now consider how the robustness of this homeostasis can vary. The strongest determinants of this characteristic are the magnitudes of the reaction enthalpies ΔH_i . Of course when this value tends to 0 we revert to the thermally neutral case in which the spots are at the mercy of imposed temperature changes. There is clearly a threshold enthalpy strength required for thermal homeostasis to set in. In what follows, I will assume that the ratio of the two enthalpies is fixed such that $\Delta H_1 = -\frac{4}{5}\Delta H_2$. The additional magnitude of the endothermic enthalpy is necessary to prevent runaway positive feedback from the exothermic reaction.

With that in mind, let us examine a reduction of enthalpy magnitude to $\Delta H_1 = -0.14$. The population dynamics of this system are shown in Figure 4(b). We see that this change does indeed reduce the robustness of the thermal homeostasis, since the internal temperature variations are larger with this smaller magnitude of reaction enthalpy. It is also clear how close this system comes to oblivion. The population of

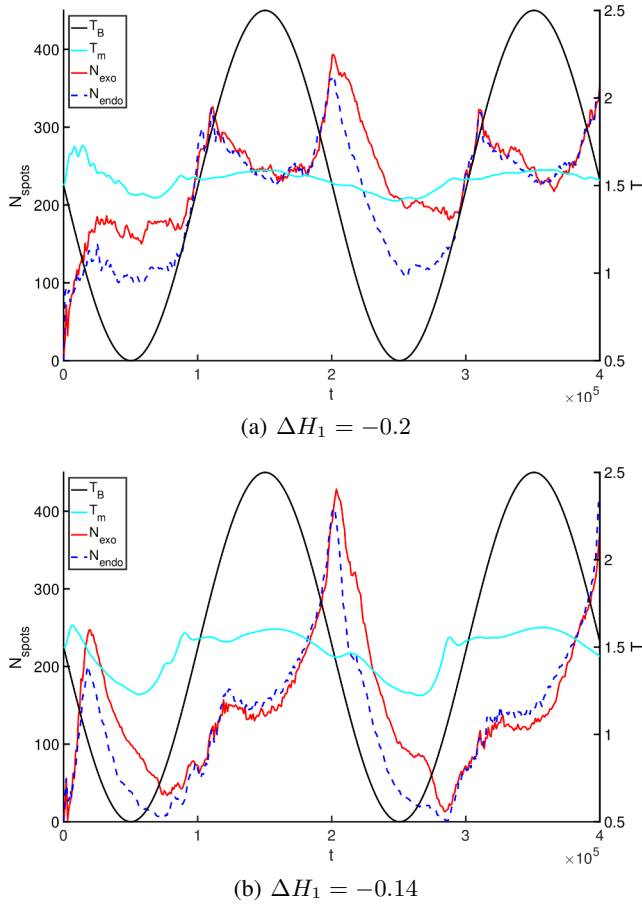


Figure 4: Population dynamics of two spot species RD systems. The two sets are mutually homeostatic with respect to temperature, spontaneously adjusting their population sizes in response to the imposed boundary temperature. The first system had a reaction enthalpy value of $\Delta H_1 = -0.2$, whereas the system corresponding to the lower figure used a value of $\Delta H_1 = -0.14$.

the endothermic spot species in particular comes extremely close to 0. However when the temperature recovers, so do the structures.

The Line Between Survival and Peril

We have been assessing the ability of the chemical patterns to regulate periodic temperature variations. It is plausible that the frequency of these variations also factor into the survivability of the system. Heat travels in a wave-like manner and thus takes some time to propagate and has a degree of inertia when it travels. Therefore high frequency changes in boundary temperature might be less difficult for the survival of the spot patterns, since the destructive temperature changes might not have time to propagate into the core of the system.

In order to assess how robustness of chemical structures

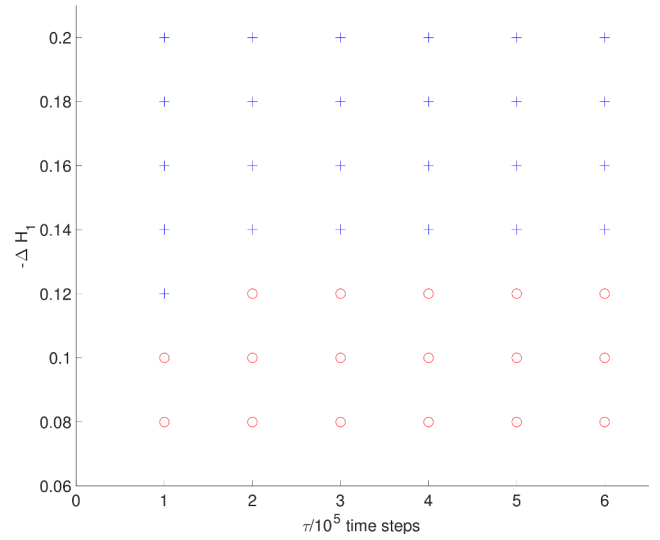


Figure 5: Survival phase diagram for chemical structures in reaction diffusion simulations. The boundary temperature is varied sinusoidally with a period τ .

varies with temperature variation frequency and reaction enthalpy magnitude, I conducted a series of simulations where both of these parameters were varied. The resulting phase portrait is shown in [Figure 5](#). This figure shows that there seems to be a relatively weak dependence of survival on temperature oscillation frequency. When $\Delta H_1 = -0.12$, the spots survive at a period of $\tau = 10^5$ but die when the temperature variations are slower (giving more time for boundary heat fluxes to make their way into the system).

However, there is a phase transition from persistence to destruction at an enthalpy value of $\Delta H_1 \approx -0.13$. This represents the threshold at which the reaction heat fluxes can compensate for boundary heat fluxes that will eventually overwhelm the system. Note that I used a temperature variation of $0.5 \leq T_B \leq 2.5$. It would be instructive to map a similar phase portrait for larger and smaller magnitude temperature oscillations.

The Defence Mechanism

In this final section, I wish to illustrate the means by which the homeostatic two-spot system can prevent excessive heating or cooling. I carried out a simulation in which two small circular regions were held at a high $T_H = 2.5$ and low $T_L = 0.5$ temperature, respectively. The steady state configuration of this system is shown in [Figure 6](#).

The action of the spots keeps the temperature throughout most of the domain at the optimal value of ~ 1.5 . In the region of very high temperature, we see the reaction proceeding at a high rate and an interface region of short worm-like structures at the edge. The total interface contour of the two systems is approximately equal. Thus the absorption of

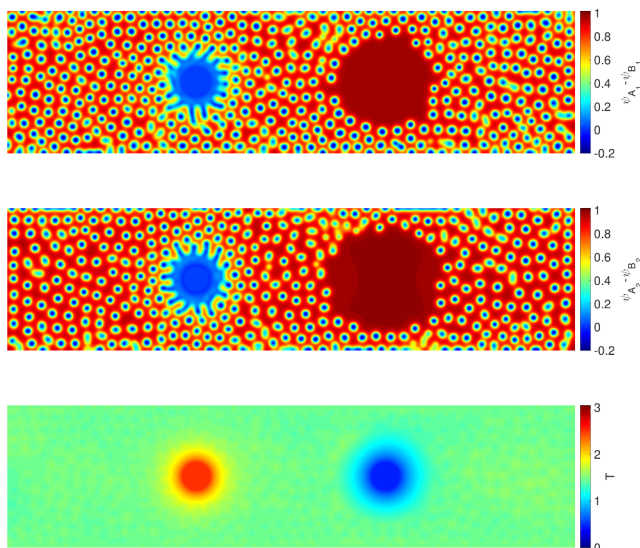


Figure 6: Steady state configuration of a two-spot species reaction diffusion system in which two patches are held at a very high and very low temperature, respectively.

the excess heat from the hot patch is due to the larger enthalpy magnitude of the endothermic spot species. Neither spot species outnumbers the other here, which aligns with the results of Figure 4, which shows the spot populations equal at times of high temperature.

In the case of the cold region, we see that the exothermic spots can survive deeper into the cold region those that are endothermic. This allows them to outnumber their endothermic cousins, who must reside slightly further out from the low temperature region. In both the hot and cold cases, the arrangement of the two sets of spots acts to absorb the excess heat (or cold) fluxes. It is this reconfiguration that confers the thermal homeostasis ability shown in previous sections and previous work.

Conclusions

In this work I presented a versatile modelling framework based on the Lattice Boltzmann Method, able to simulate a vast range of coupled fluid dynamical and chemical systems. It is possible to input any chemical reaction network into this system and observe interactions between fluid flow, heat transfer and chemistry. The model was used to simulate the thermal Gray Scott RD system and it was shown to reproduce previous results of thermal homeostasis, with some subtle changes in phenomenology, despite the fact that it no longer used the standard PWBC. This will open up new possibilities for investigating the pattern-formation, self-organisation and thermodynamic properties of these complex, sometimes life-like, systems. I introduced a parameter space exploration in which I analysed the survival of spot structures as a function of temperature oscillation frequency

and reaction enthalpy magnitude. I illustrated the threshold enthalpy value at which thermal homeostasis sets in, and found that the survival of the spot structures has only a weak dependence on temperature oscillation frequency. In the final section I used a system containing a very hot and a very cold region to demonstrate the actual mechanism behind the thermal homeostasis of these chemical patterns.

Future Work

The modelling platform that I have presented in this work could address a range of questions in the realms of pattern-formation, heat flow and the coupled dynamics of chemistry and fluid flow. We can explore further the possibilities of self-replicating spot patterns with a range of interactions. Looking forward, it's conceivable that we may even be able to construct arbitrary interaction networks amongst sets of RD spots. If we can achieve sufficient separations of time scales, it might be possible to construct networks that are capable of primitive information processing and memory. Such behaviour would give us fascinating new insights into how information concentration can arise spontaneously in physico-chemical systems and may hint at primitive mechanisms that could have played a role in the origin of life.

The next phase in such an investigation would be to ask whether multiple variables, distributed over space and time, could be simultaneously regulated by the action of multiple sets of organised chemical structures. It will be of great insight to explore this possibility of multi-variate homeostasis, and to assess whether separations of length scales and/or time scales introduces new forms of spatial organisation, modularity, or division of labour. Perhaps the complicated task of multi-species regulation might force the system to organise itself in novel ways, illustrating effects of economies of scale, despite the fact that the system is pure physics and chemistry.

Acknowledgements

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