Adaptation to the Edge of Chaos in a Non-Isothermal Autocatalator

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Numerical simulations of a low-pass filtered feedback from a dynamical variable to the system parameter of a non-isothermal autocatalator are examined. Parameter values for which the limiting dynamics is chaotic are found to evolve to nearby values yielding periodic dynamics while parameter values yielding periodic dynamics are uneffected. The system thus exhibits adaptation to the edge of chaos. This suggests that low-pass filters, believed to be quite common in chemical reactions, may be one reason few chaotic reactions have been observed experimentally.

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Introduction

There has been much interest lately in the wealth of behavior possible in open chemical reactions. This includes such behavior as oscillating pH structures[1], Turing and other patterns[2 3 4 5], symmetry breaking[6] and pattern concatenation[7]. Another possibility in open chemical reactions is the presence of chaotic dynamics. Chaotic oscillations were first observed in the Belousov-Zhabotinskii reaction in the 1970's[8]. Since then chaotic dynamics have been observed in heterogeneous catalysis reactions[9], electrodissolution reactions[10] and biochemical systems[11]. These observations have hence brought great interest to the topic of how to control chaos in a chemical reaction[12 13 14 15 16]. In fact, so much has been written on the topic of chaotic chemical reactions and how to control them that one may be lead to believe that chaotic dynamics is quite common in open chemical reactions. This is in fact not the case. Chaotic dynamics is indeed quite rare.

Due to the non-linear nature of most chemical reactions and the presence of feedback through autocatalysis and/or self-heating one would expect chaotic dynamics to be abundant in chemical reactions. However, as mentioned only a small portion of reactions exhibit chaotic behavior. One is thus left wondering, why the apparent lack of chaos in chemistry? Previous studies have investigated the effect of a low-pass filtered feedback from a dynamical variable to the control parameter on the logistic map[17] and the Chua circuit[18]. It was found that the low-pass filter resulted in the systems adapting to a state at the boundary of chaos and order known as the edge of chaos. We examine numerical simulations of a non-isothermal autocatalator in the presence of a similar low-pass filter.

We find that the presence of a low-pass filtered feedback in a non-isothermal autocatalator results in the system evolving to the edge of chaos. Low-pass filters are believed to be quite common in nature, particularly in dissipative chemical reactions.

These results thus suggest that such naturally occurring low-pass filters may be one reason for the apparent scarcity of chaotic dynamics in open chemical reactions.

Numerical Simulations

The following non-isothermal autocatalator model is taken from *Chemical Chaos*[19],

Step 0:	$P \rightarrow A$
Step 1:	$A \rightarrow B$
Step 2:	$A + 2B \rightarrow 3B$
Step 3:	$B \rightarrow C + \text{Heat}.$

This system can be described by the following set of equations,

$$\begin{aligned} \frac{d\alpha}{d\tau} &= \mu e^{\theta} - \alpha\beta^{2} - \kappa_{u}\alpha \\ \frac{d\beta}{d\tau} &= \alpha\beta^{2} + \kappa_{u}\alpha - \beta \qquad (1) \\ \frac{d\theta}{d\tau} &= \delta\beta - \gamma\theta \end{aligned}$$

Here α and β are dimensionless concentrations of A and B respectively, θ is the dimensionless temperature difference between the reaction temperature and the ambient temperature and τ is dimensionless time. The exothermicity of Step 3 is described by δ, γ is a measure of the surface heat transfer coefficient and κ_u is the dimensionless rate constant for the uncatalysed Step 1. The parameter μ, representing the dimensionless initial concentration of reactant P (taken here to be constant), determines the dynamics of the system. For $0.6 < \mu < 0.637$ both concentrations as well as temperature undergo period-1 dynamics. For values of μ between 0.638 and 0.688 all dynamical variables: α, β and θ, undergo a period doubling sequence. For $0.689 < \mu < 0.696$ the dynamics is mostly chaotic however, there do exist several small periodic windows within this range. For $0.696 < \mu < 0.7$ the dynamics is once again periodic. The edge of chaos refers to values of μ for which a small perturbation would take the system from periodic dynamics to chaotic dynamics or from chaotic dynamics to periodic dynamics. Values of μ that are very near 0.689 or 0.696, or that are within the periodic windows are thus at the edge of chaos.

If the system parameter, μ, instead of being held constant is allowed to vary slowly with time this system becomes an adjustable system. This is accomplished by periodically applying a forcing function, f_n, to μ:

$$\mu(\tau) = \mu_n \qquad \text{if } nT < \tau < (n+1)T$$

$$\mu_{n+1} = \begin{cases} \mu_{max} & \text{if } \mu_n + \varepsilon f_n > \mu_{max} \\ \mu_{min} & \text{if } \mu_n + \varepsilon f_n < \mu_{min} \\ \mu_n + \varepsilon f_n & \text{else.} \end{cases} \qquad (2)$$

Here ε is a small constant and T is chosen to ensure an appropriate separation of time scales between the evolution of the system parameter and that of the dynamical variables. The upper and lower bounds, μ_{max} and μ_{min}, are chosen to be any two values outside the chaotic domain. In the following simulations $\mu_{max} = 0.7$ and $\mu_{min} = 0.686$. If the forcing function, f_n, is chosen to be a function of the dynamical variables only the system becomes self-adjusting. The following simulations examine the use of a low-pass filter as the forcing function

In numerical simulations a low-pass filter is achieved using a fourier analysis of the times series of one of the dynamical variables. Here the temperature difference, θ, is chosen however, low-pass filters using either α or β have also produced similar results. If the number of time steps, N, is taken to be odd the fourier sine and cosine coefficients are given by, $a_{nk} = (2/N)\sum_{t=x}^{N-1}\theta(t)\times\sin(2\pi kt/N)$ and $b_{nk} = (2/N)\sum_{t=x}^{N-1}\theta(t)\times\cos(2\pi kt/N)$ respectively, with b_{n0} given by $(1/N)\sum_{t=x}^{N-1}\theta(t)$. Here x, the lower limit of the summation, is chosen such that any initial transient behavior is not included in the analysis. A low-pass filter with dc cutoff and a low frequency cutoff keeps only the lowest frequency terms. A back transformation using only the second fourier component is chosen for the forcing function, f_n,

$$f_n = a_{n2}\times\sin(4\pi n/N) + b_{n2}\times\cos(4\pi n/N)\,. \qquad (3)$$

If the forcing function is applied only every N time steps this simplifies to,

$$f_n = b_{n2}\,. \qquad (4)$$

Results

Numerical simulations of the self-adjusting chemical system using the forcing function defined by (3) were performed. Figure 1 shows the evolution of four different initial parameter values. For $\mu_0 = 0.68712$ the system exhibits period-4 dynamics and there is essentially no change in μ with time. However, for the initial parameter values

$\mu_0 = 0.68950$ and $\mu_0 = 0.69188$, for which the system exhibits chaotic dynamics, μ evolves with time until a parameter value is reached for which the limiting dynamics is periodic. For $\mu_0 = 0.68642$ the system again exhibits period-4 dynamics however, there is a small drift in the parameter value. This drift is due to the windowing function used in the fourier analysis and could easily be avoided by implementing a minimum threshold for the forcing function,

$$f_n = \begin{cases} 0 & \text{if } \log(|a_{n2}| + |b_{n2}|) < \text{Threshold} \\ \text{Given by (4)} & \text{else} \end{cases} \qquad (5)$$

Figure 2 shows a histogram of 100 parameter values at time n = 150. Initially, at n = 0, the distribution is uniform across the interval [0.686, 0.7]. At n = 150 several peaks are observed at the periodic windows within the chaotic domain and at the boundaries of the chaotic domain. There is little change from the initial parameter distribution in the periodic regions with the exception of a small number of parameter values drifting due to the windowing function. It should be noted that this drift simply shifts parameter values within the periodic region to other nearby bins within the periodic region; it does not cause parameters starting in the periodic region to settle in the chaotic domain. At n = 150 there is thus a very low probability for values of the parameter μ for which the limiting dynamics is chaotic. There is however, a high probability for parameter values for which the limiting dynamics is periodic. The probability is particularly high for parameter values within the periodic windows and at the boundaries of the chaotic domain. The system thus exhibits adaptation to the edge of chaos.

The effect of the low-pass filter can be understood in terms of recurrence time. Periodic dynamics has a finite recurrence and thus a lowest frequency fourier component. Chaotic dynamics however, has an infinite recurrence time and thus no lowest frequency fourier component. A low-pass filter exploits this distinction between periodic and chaotic dynamics by using only the lowest frequency fourier components in the forcing function. This results in a sizeable forcing function for parameter values for which the system behaves chaotically and little to no forcing function for parameter values for which the system exhibits periodic behavior.

Simulations were also performed in which the forcing function was normalized by the amplitude of oscillation of the dynamical variable θ,

$$f_n = b_{n2}/A_n. \qquad (6)$$

Here A_n is the amplitude of oscillation. Adaptation to the edge of chaos was again observed indicating that this is a recurrence time phenomena independent of the oscillation amplitude.

The evolution of parameter values yielding chaotic dynamics under a low-pass filter has been shown to follow a diffusive, random walk motion[17]. The time required for adaptation to occur is then a function of the distance in parameter space between the current parameter and the nearest parameter yielding periodic dynamics, the separation of timescales between the system parameter and the dynamical variables and the forcing constant, ε. It is thus expected that reactions show a period of transient chaos while adaptation is taking place.

Conclusions

We have shown through numerical simulations that the application of a low-pass filtered feedback from a dynamical variable to the system parameter of a non-isothermal autocatalator results in the system evolving to the edge of chaos. This suggests that low-pass filters, quite common to dissipative chemical reactions, may be at least partly responsible for the apparent lack of chaos in chemical reactions. Experimental work is currently underway to examine the effects of an externally imposed low-pass filter on the Belousov-Zhabotinskii reaction.

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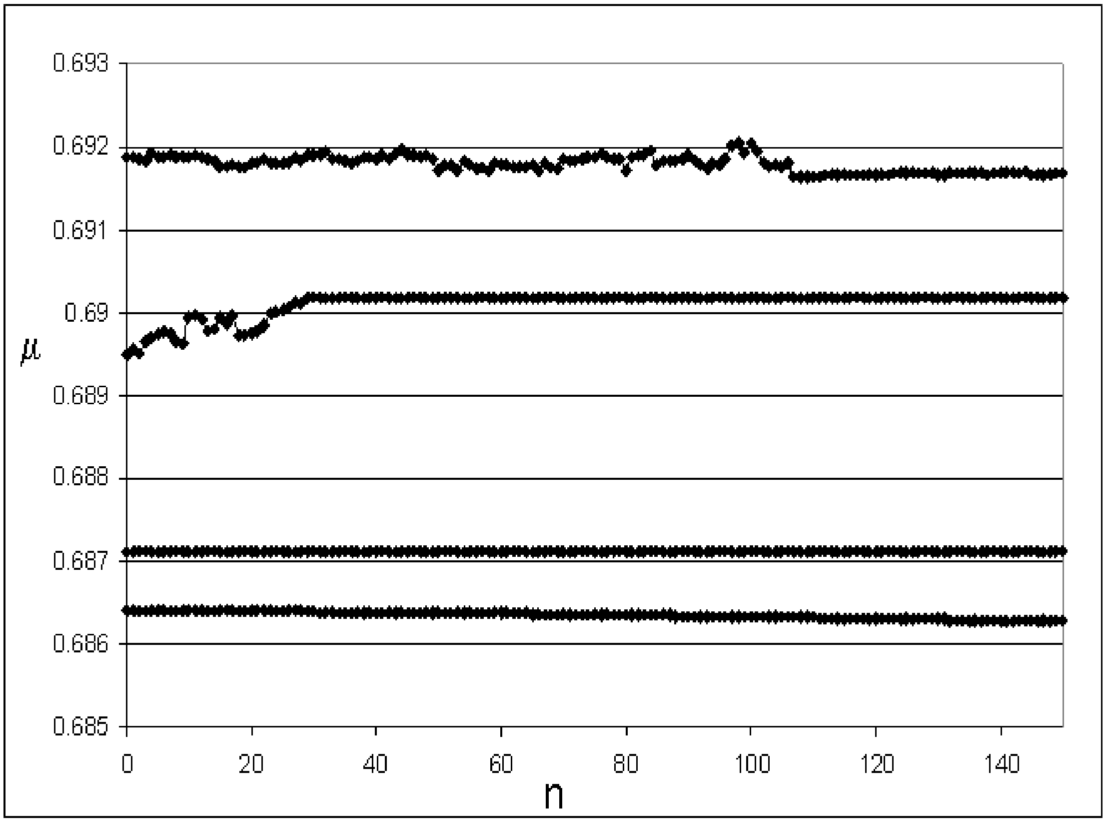
FIG. 1: Time evolution of 4 different initial parameter values. Values μ_0=0.68712 and μ_0=0.68642 correspond to periodic dynamics while μ_0=0.68950 and μ_0=0.69188 correspond to chaotic dynamics. For this simulation κ_u=0.0055, δ=0.1, γ=0.5, N=7500 and ε=0.8.

FIG. 2: Distribution of parameter values between 0.686 and 0.7. At n=0 distribution was flat however, at n=150 several peaks are observed at the boundaries to the chaotic domain as well as within the periodic windows. Periodicity of periodic windows is indicated above each peak. Again κ_u=0.0055, δ=0.1, γ=0.5, N=7500 and ε=0.8 for this simulation.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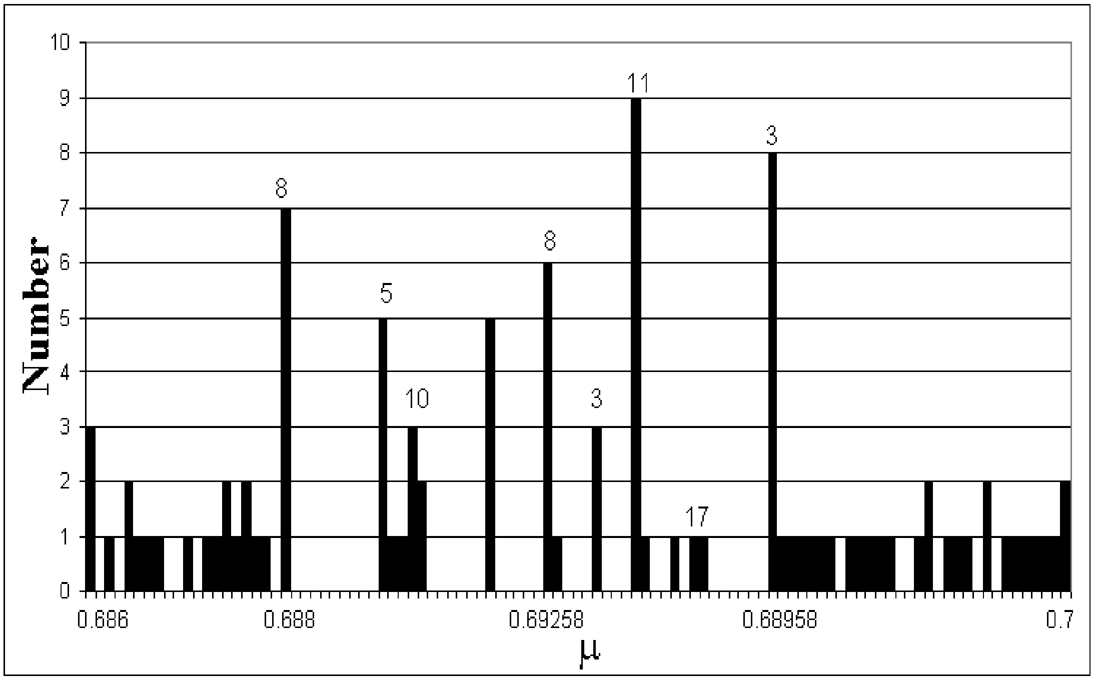

Number
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1
2
3
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6
7
8
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17
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