S.m.15. MERCY MANI, P.–Studies on nonlinear dynamics of certain reaction system–1989–Dr. M. Ramachandra Kaimal

The nonlinear dynamics of certain important reaction systems are discussed and analysed in this thesis. The interest in the theoretical and the experimental studies of chemical reactions showing oscillatory dynamics and associated properties is increasing very rapidly. Many researchers work out mathematical models of such reactions to understand the complex nature of the systems. The oscillatory behaviour and other related characteristics in nonlinear systems far from equilibrium has a highly interesting connection with the organization of the structure of the system.

An attempt is made to study some nonlinear phenomena exhibited by the well known chemical os cillator, the Belousov-Zhabotinskii reaction whose mathematical properties are much in common with the properties of biological oscillators. While extremely complex, this reaction is still much simpler than biological systems atleast from the modeling point of view. A suitable model (Field and Noyes, 1974) for the system is analysed and we have studied the limit cycle behaviour of the system, for different values of the stoichiome tric parameter f by keeping the value of the reaction rate (k_4) fixed at $k_4=1$. The more complicated three-dimensional model is stiff in nature. The values of the parameters in the system other than f viz. s, w and q are fixed as 77.27, 0.161 and 8.375×10^{-6}. The existence of limit cycle behaviour in the three dimensional model is shown analytically and are verified numerically. Among many numerical methods to solve such systems of nonlinear differential equations the Runge-Kutta method with step doubling is found to be the most suitable one to solve our system. Limit cycle behaviour is observed for the complete model in the y–x phase plane for f=1.0 and f=1.1, with periods t=143.5 and t=148.9 respectively.

The two variable (y-z) model, being much easier to handle mathematically is examined more thoroughly. The oscillatory behaviour of this system is examined for several values of f lying in the unstable region $1/2 < f < 1+\sqrt{2}$. Isolated limit cycles are obtained for f = 0.5001, 0.74, 0.9 1.0, 1.3,-1.4 and 1.5 in the y-z plane. The birth of more than one limit cycle is observed for the system at the parametric values f=1.1, 1.2 and 1.595288. This may be considered as s signal for the excitability property of the reaction model. Many workers [N.F. Britton (1982), G.B. Ermentrout and N. Kopell (1986)] predicted and some discussed the excitable behaviour of B-Z reaction system for this range of f.

The solution trajectory starting from the initial concentration $(y_0, z_0) = (1.0, 488.8)$ is attracted to another stationary state (1.297, 4.355) for some parametric values f=1.6, 1.7, 1.8, 1.9, 2.0, 2.1 etc. The multiple steady states are observed in this region of f.

The fact that the autocatalysis is the common feature of almost all oscillators (biological and non-biological) inspired us to study a general cubic autocatalytic system.

We have presented a model analysing the auto catalytic system. The cubic autocatalytic step A+2B → 3B lies at the heart of the simplest models for oscillatory and other complex nonlinear behaviour in closed systems, in the C.S.T.R. and in reaction coupled with diffusion. General reaction systems containing either quadratic auto catalytic reaction step or cubic autocatalytic step, coupled with autocatalyst decay step which are two dimensional were studied so far. In order to explain the complex phenomena observed in oscillators like aperiodic oscillation, period doubling etc. a three dimensional model is required. Sustained oscillatory behaviour is observed in the system. The range of the controlling parameter K at which the stationary states are unstable is found analytically. This range is found to be $K_{1c} < K < K_{2c}$ where $K_{1c} = 0$ and $K_{2c} = 0.0003$. The values of the other parametric values n' and t_f are fixed as $n' = 10^{-4}$, and $t_f = 10^4$. The initial value taken for the numerical integration is $x_0=Y_0=Z_0=0.1$. Limit cycles are obtained for the system in X-Y, Y-Z and Z-X phase planes respectively. The trajectories are attracted to a limit cycle and the unstable critical point inside it acts as an attractor, for the supercritical Hopf bifurcation point K_{2c}. When the integration of the system is carried out numerically for other parametric values less than K = 0.0003, large amplitude waves corresponding to the concentrations of each constituents P,A and B are formed. The trajectories corresponding to the system repel from a fixed point $(3\times10^{-4}, 8\times10^{-4}, 3\times10^{-4})$. Thus the numerical result confirms the analytic prediction that the equilibrium state solution of the system in the region $K_{1c} < K < K_{2c}$ is an unstable focus. At the critical Hopf bifurcation value $K_{2c} = 0.0003$, the unstable focus becomes a stable one. The same behaviour of the solutions described above are obtained for some other values of t_f, viz. 9999.0, 10001.0, 10002.0, 10003, 10004.0, 10005.0, 10007.0, 10008.0 and 10009.0.

Several interesting phenomena, other than sustained oscillations or damped oscillations, such as bursting, excitability, intermittency, CDO, Chaotic structures etc. are to be examined more elaborately for the oscillators, considered here. In the case of the three-variable Oregonator model, the sustained oscillatory behaviour of the system is to be examined for all other values of f. Satisfactory models are yet to be developed for explaining some recent experimental results of the B-Z chemical reaction (eg. certain pattern formation). The reaction system based on the cubic autocatalytic step has to be subjected to more studies.