# OBSERVING THE LIMIT OF OSCILLATION IN AUTOCATALYTIC REACTION

# SYSTEM

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# OBSERVING THE LIMIT OF OSCILLATION IN AUTOCATALYTIC REACTION

SYSTEM

by

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# MENGAWAL BATASAN PENGGANTIAN DALAM SISTEM REAKSI AUTOKATALIK: MODEL BRUSSELATOR

# ABSTRAK

Tindak balas autokatalitik adalah salah satu tindak balas di mana kepekatan perantara reaktan terbentuk sebagai produk dengan berkala. Tesis yang dikemukakan ini menggunakan model brusselator yang merupakan salah satu sistem tindak balas autokatalitik. Autokatalisis telah banyak dieksplorasi dalam bentuk model untuk asal kehidupan sebagai asas untuk organisasi diri. Pengayunan kimia adalah serupa dengan ayunan di mana autokatalisis menggantikan maklum balas positif. Secara amnya, reaksi ini berlaku secara merata dengan kadar yang berbeza-beza hingga pada satu titik di mana ia mencapai keseimbangan. Fenomena dinamik yang kompleks seperti bistabil, ayunan, ketidakstabilan deterministik, dan pembentukan pola spasial atau gelombang secara rawak mungkin berlaku dalam sistem autokatalitik, yang tidak diinginkan dalam pengeluaran kimia. Pembezaan antara autokatalisis orde pertama dan orde tinggi terbukti sangat berguna. Kedua-dua keadaan di mana zarah autokatalitik tunggal aktif di bahagian reaktan diklasifikasikan sebagai autokatalisis peringkat pertama

# OBSERVING THE LIMIT OF OSCILLATIONS IN AUTOCATALYTIC REACTION SYSTEM: THE BRUSSELATOR MODEL

# ABSTRACT

Autocatalytic reaction was one of the reactions where the concentrations of reactants intermediates form as products with periodicity. The presented by this thesis was using the Brusselator model which was one of the autocatalytic reaction systems. Autocatalysis has been widely explored in the form of models for the origin of life as a basis for self-organization. Chemical oscillation was analogous to oscillation where autocatalysis replaces positive feedback. In general, these reactions proceed evenly with varying the rates until at one point where it reaches equilibrium. Complex dynamical phenomena such as bistability, oscillations, deterministic instability, and random forming of spatial patterns or waves may occur in autocatalytic systems, which were undesirable in chemical production. The differentiation between first order and higher-order autocatalysis proved to be extremely useful. Both situations in which a single autocatalytic particle was active on the reactant side were classified as first-order autocatalysis.

# **CHAPTER 1**

## **INTRODUCTION**

Chapter 1 introduces the overview of this research and significance of the autocatalytic reaction system. In general, this chapter summarizes the research background of autocatalytic reaction system, the problem statement, and the objectives of this final year project.

#### 1.1 Research Background

Every living organism has chemical oscillators. Some examples include systems like unit of time clocks and central system swinging activity, as well as numerous organic chemical processes at the cellular level including the glycolytic pathway, peroxidasecatalyzed reaction, and protein synthesis. Scientists were fascinated with oscillatory chemical reactions because they are commonly used as illustrative examples of the behavior that can occur in reactions regulated by non-linear dynamic rules found in chemistry, biology, and engineering. Understanding periodical processes at the molecular level was critical since it holds the key to advanced phenomena such as animal sleep induction or bird migratory behavior, among others.

The phenomenological dynamics of oscillatory chemical reactions reveal that reactant, intermediate, and product concentrations will fluctuate intermittently in area, as during development, or in time, as during unit of time rhythms. Because the reactants were converted to product and then back to reactants, it appears that an oscillatory reaction would need the system's free energy to oscillate, defying the second rule of physics. In fact, this phenomenon was driven by the Gibbs-free-energy decrease of an overall chemical action occurring outside of thermodynamical equilibrium. Such oscillatory systems are related to thermodynamically open systems. In enzyme-catalyzed processes, a biological cell was an open system that may take in nutrients and release waste products. These reactions were complicated and need a number of fundamental stages, the bulk of which involve nonlinear dynamics. If a suitable feedback mechanism was available, long-lasting chemical oscillations will develop. This may be accomplished by continuously supplying reactants and withdrawing product from the reaction vessel.

Another essential aspect of most chemical systems that exhibit oscillations was autocatalysis, which was a way for providing feedback to a periodic reaction in which the progress of the reaction is favored by the product produced. Autocatalysis was the increase of chemical activity of reactions by product created throughout a reaction. The simplest example of autocatalysis is the reaction:

 $A + X \rightarrow 2X$  (Equivalent  $A \rightarrow X$ )

whose rate law given as  $-\frac{d[A]}{dt} = k[A][X]$ 

Chemical oscillations were caused by the existence of one or more contact action stages in a complex process. Autocatalysis, on the other hand, will result in an explosion as the product concentration rises. Therefore, an inhibition step was additionally necessary. The steady state was stabilized when autocatalysis and inhibition appear at the same time, and the net rate of rise of all relevant species was zero. As a result, oscillations will appear to be beneficial only if the inhibition step was delayed in some way (Nicolis & Portnow, 1973).

In 1920, Lotka revised a hypothetical set of chemical reaction (1)-(3)

$A + X \to 2X \tag{(}$	(1)	)
$A + A \neq 2A$	(Ι,	,

 $X + Y \to 2Y \tag{2}$ 

$$Y \to P \tag{3}$$

Longer temporal oscillations in the concentrations of intermediates X and Y occur during the whole process A to P. This can happen if the system is maintained out of range through exchanging reactants and products with the surrounding (Field et al., 1974).

The chemical mechanism of Field, Karas, and Noyes for the oscillatory Belousov reaction has been generalized by a model composed of 5 steps involving 3 independent chemical intermediates, the behavior of the resulting differential equations has been numerically examined, and it had been shown that the system traces a stable closed plane in 3-dimensional space, an equivalent plane was obtained from alternative part points and even from the purpose appreciate steady state resolution of the differential equations. Limit cycle behavior appears to be present in the model. The limit cycle model may be simplified to a system defined by two plane variables by linking the concentrations of two intermediates; this coupled system can be analyzed using theoretical approaches already developed for such systems (Field et al., 1974).

Since a Lotka mechanism comprises two contact action reactions, it shows this unique behavior; nevertheless, catalysis was not required for sustained oscillation. A minimum of one step inside the mechanism must provide feedback, so that the step's product influences its net rate in addition to rule reversibility. The feedback causes nonlinearity in the differential equations that describe the process, the system may display all of the complexity that come with nonlinear dynamic laws. If the oscillations were to be sustained, the system should be far away from equilibrium. Nonlinear kinetic equations and periodic behavior were caused by chemical systems containing a lot of possible types of feedback, according to Ross and colleagues. As its Lotka mechanism causes persistent oscillations in intermediate concentrations, the open system shows a closed plane inside the plane of X and Y concentrations. The frequency and amplitude of such oscillations, on the other hand, were determined by the initial intermediate concentrations as well as the kinetic constants. Furthermore, if the oscillatory system becomes perturbed, the planes inside the X and Y planes will change. A system with limit cycle behavior, on the other hand, can approach a limiting periodic behavior defined purely by the kinetic constants and unaffected by the starting concentrations of the oscillatory intermediates (Field et al., 1974).

A chemical scheme exhibiting limit cycle behavior (1)-(4)

 $A \to X \tag{4}$ 

$$B + X \to Y + D \tag{5}$$

$$2X + Y \to 3X \tag{6}$$

$$X \to E$$
 (7)

has been proposed by Prigogine and their colleague in Brussels. This appears to be a single spectacular chemical theme showing limit cycle behavior with only two intermediate species, as demonstrated by the capital of Belgium cluster's ability to build advanced spatial and temporal patterns similar to those seen in biological systems. Although step (3) was third order within the concentrations of transient intermediates, the subject has been disputed, it clearly illustrates that chemical systems will develop extremely advanced temporal and spatial order (Field et al., 1974).

The experimental behavior of the Belousov reaction strongly implies a limit cycle's stable limiting plane of intermediate concentrations. The current research demonstrates that intended chemical actions were frequently reduced to a simpler generic mechanism that behaves like a limit cycle (Field et al., 1974).

1.2 Problem statement

There were various models that describe this autocatalysis reaction in order to approach it. The concentrations of reactants, intermediates, and product change with periodicity during autocatalysis. Chemical oscillation was similar to electrical oscillation in that feedback is replaced by autocatalysis. Oscillatory responses have been observed in a variety of conditions. In general, the reactions operate smoothly at different rates until they reach equilibrium. These reactions cause concentration oscillations to be observed. The Belousov-Zhabotinskii reaction (B-Z reaction) and the Bray-Liebhafsky reaction were two examples of oscillatory reactions (Kumar, 2020b). There are approximately three models that can simply tell about the autocatalysis model. The Brusselator model, which was one of the most basic explanations for this model, then followed by the oregonator model and the lotka volterra mechanism. Moreover, this model was chosen because it was one of the simplest autocatalytic reactions, the Brusselator model was chosen as a reference to examine the limit of the oscillations.

#### 1.3 Objectives

The objectives of this study are:

- i. To observe the dynamics oscillation occurs in the autocatalysis reaction systems.
- ii. To study effect of the concentration of activator and inhibitor in the reaction systems.
- iii. To determine the limit of oscillation in autocatalysis systems based on the Brusselator model.
- iv. To observe the exact point at which the oscillations will continuously oscillate.

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#### **CHAPTER 2**

## LITERATURE REVIEW

In previous chapter, the introduction about the autocatalytic reaction already been summarize and the observation for the study for the oscillations system also was being introduce using the suitable or the simplest model to describe this autocatalytic reaction systems. Chapter 2 presents the previous discoveries and reviews available from credible scientific data and references that are related to this final year project topic. This chapter covers the overview of the Brusselator model on the stability and the oscillations of this model on where it begins to oscillate. Besides, this chapter also presents important references and information required on studying the limit of the oscillation in autocatalytic reaction using the Brusselator model as a reference.

#### 2.1 Brusselator model description

Once reverse reactions are neglected and the concentrations of A and B are held constant, the Brusselator from reaction (4) - (7) was understood to show oscillating behavior within the species X and Y. Its applicability as a model for any possible oscillating chemical system has been questioned, despite the reality that each real chemical reaction should be reversible to some extent, and it has been claimed that the reversible Brusselator oscillates if the two principles of detailed balance and conservation of matter are violated. The reversible Brusselator was theoretically examined in situations where all the requirements of detailed balance were met, as well as the consumption of A and B; the system described was closed, and matter was carefully maintained. Oscillating behavior was possible for a limited number of reaction parameters (rate constants, initial concentrations, etc.) and should be described consistently. Oscillations have a finite range and last a certain amount of time once they occur. The oscillating behavior within the irreversible system can be obtained as the limiting case of the reversible system when they cease continuously followed by a monotonic decrease to equilibrium (Gray et al., 1988).

The irreversible Brusselator was not the simplest method of its sort for pointing out any of the higher than behavior, and it comes with its own set of problems. In a relatively basic open system continuous stirred tank reactor, it exhibits infinities in intermediate concentrations and oscillating amplitudes, and it loses oscillations. However, it deserves to be included in the history of the issue with the Lotka-Volterra plan because the system was far from equilibrium and the rates of the reverse reactions were adjusted to an appropriate zero. Then, over the duration of interest, the concentrations of the pool-chemical species A and B (the initial reactants) and D and E (the final products) were maintained constant. This leaves just 2 different concentrations, those of the intermediates X and Y, for an isothermal oscillating system's minimal range (Gray et al., 1988).

These chemical oscillations were caused by non-equilibrium conditions, which can be defined as the spontaneous development of symmetry-breaking and therefore the production of chaotic patterns in dissipative systems. If the trajectories approach the limit cycle as time approaches time it was known as a stable or attractive limit cycle. Then if the trajectories approach the limit cycle as time approaches minus-infinity it was an unstable or non-attractive limit cycle. The uncertain Brusselator system was used to establish a theoretical knowledge of non-equilibrium state instability that can be well depicted. The goal of is to research a system that continuously interacts with its surroundings, that is working faraway from thermodynamic equilibrium (Mcdowell, 2008). Consider an activator-inhibitor Brusselator model that represents an autocatalytic oscillatory chemical reaction. The straight-line behavior of the solutions of the Brusselator model numerically was studied varied forms of pattern formation of the Brusselator model arising in chemical reactions with the numerical investigation and observe the periodic traveling wave solutions of the Brusselator model exploitation numerical bifurcation analysis has additionally been studied. The slow stable manifold of the Brusselator model for the primary time and this study advances the field from the previous related work (Nazimuddin & Al, 2020). The reaction–diffusion Brusselator system contains a combine of variables intermediates with reactant and products chemicals whose concentrations were controlled. The system represents a helpful model for study of cooperative processes in chemical dynamics. In recent years, a lot of attention has been paid in literature within the development of numerical schemes for the numerical solutions of reaction–diffusion Brusselator system such as second order finite-difference scheme (Jiwari & Yuan, 2014).

## 2.2 Governing equation of the Brusselator model

As showing the reaction of the Brusselator model from (4) - (7) denote that the concentrations of *A*, *B*, *D*, *E*, *X* and *Y* by [*A*], [*B*], [*D*], [*E*], [*X*] and [*Y*] respectively. Then the development of the concentrations of species as a function of the time *t* using mass action law as given as follows:

$$\frac{d[A]}{dt} = -k_1[A] \tag{8}$$

$$\frac{d[B]}{dt} = -k_2[B][X] \tag{9}$$

$$\frac{d[D]}{dt} = k_2[B][X] \tag{10}$$

$$\frac{d[E]}{dt} = k_4[X] \tag{11}$$

$$\frac{d[X]}{dt} = k_1[A] - k_2[B][X] + k_3[X]^2[Y] - k_4[X]$$
(12)

$$\frac{d[Y]}{dt} = k_2[B][X] - k_3[X]^2[Y]$$
(13)

where  $k_{j}$ , (j= 1,2,3,4) is the reaction rate.

As the species D and E have no effect on others, the equations (10) and (11) can be ignored. To simplify the equation, assume that [A] and [B] are kept constant, much like the reactant, i.e. [A]=a and [B]=b, where a,b > 0. For  $k_j$  (j=1,2,3,4), all reaction rates have been set to unity. Therefore, the ordinary differential equations that describe the Brusselator chemical reaction are as follow:

$$\frac{d[X]}{dt} = a + [X]^{2}[Y] - (b+1)[X]$$

$$\frac{d[Y]}{dt} = b[X] - [X]^{2}[Y]$$
(14)

To simplify the notation, define that x=[X] and y=[Y]

Therefore, the represent Brusselator chemical reaction given by (14) in a compact form as follows:

$$\dot{x} = a + x^2 y - (b+1)x$$

$$\dot{y} = bx - x^2 y$$
(15)

From equations (15) the equilibrium points were obtained by solving the nonlinear equations

$$a + x^2 y - (b+1)x = 0 \tag{15a}$$

$$bx - x^2 y = 0 \tag{15b}$$

where there were some parameters that was inadmissible if a=0 from (15a) as taking x=0. Thus, making equation where b-xy = 0 or xy = b. Using this, it can be simplified that (15a) as

$$a + bx - (b+1)x = 0 \text{ or } x = a$$
 (16)

Therefore, the unique of equilibrium was obtained as:  $(x, y) = (a, \frac{b}{a})$ .

Assume that  $b = a^2 + 1$ , the brusselator chemical model (15) will exhibits Hopf bifurcation and will show a stable limit cycle (Vaidyanathan, 2015).

## **CHAPTER 3**

# METHODOLOGY

This chapter discloses the information on the methods applied in this final year project. It includes the general research flow diagram, MATLAB© software used and the importance parameters for doing the research.

3.1 Overview of the research methodology

Figure (1) shows the activity of the research. First of all, the equation of the model shall be prepared to understand on how the simulation will be run. One of the models had been chosen which was Brusselator model to run the simulation in the MATLAB©. It was used to observe the limit cycle and its oscillation.



Figure 1: The flow diagram of thesis and report writing.

3.2 Design of experiment (DOE)

MATLAB© were used to study the behavior of the Brusselator model on the dynamical systems. For the first part of this study, effect of the activator concentration, A was studied to observe on the dynamical changes in the systems by various it values. Then, the inhibitor concentration, B was studied by observing its dynamical systems . After observing the dynamical systems, the equilibrium points for both in changing of the parameter activator concentration, A and inhibitor, B were observed by looking at the value of the exact point at which the limit of the point before it continuously oscillate or forming the stable limit cycle. The limit point cycle was being observed by those 2 parameters which were activator concentration, A, and inhibitor concentration B, in a MATLAB© software. Table 1 below show the important parameter to study the behavior systems of this Brusselator model.

Variables or parameters	Unit
Α	Concentration (mole/l)
В	Concentration (mole/l)
x	Concentration (mole/l)
у	Concentration (mole/l)
t	time (dependable)

Table 1 : The important of variables and parameters

#### 3.3 Thesis and report writing

The data and results were presented. In the report writing, the dynamical behavior of each variable or parameters was being observed and the limit point cycle for both parameters which were activator concentration, A and inhibitor concentration, B were observed, respectively. Conclusion and suggestions were made based on the results obtained.

## **CHAPTER 4**

# **RESULT AND DISCUSSION**

4.1 Difference mode of spiral or limit cycle and its oscillation by changing the value activator concentration, A and constant the value inhibitor concentration, B.

The parameter A which was acts as activator concentration in the reaction was the first parameter considered in this works where it was referring to active parameter of the solution. The system was analyzed with different values of parameters activator concentration, A to observe on the oscillations and the limit of the cycle to specify the exact of the point at where the equilibrium point was formed in the reaction.

Figure (2), Figure (3), and Figure (4) were numerically solved using MATLAB© software. By applying this software at which the parameters inhibitor concentration, B was then set as constant. Based on the varying of parameters of A activator concentration, it showed that as increasing this parameter activator concentration, A will cause the reaction from having a limit cycle at initial stage until then a spiral was formed when the activator concentration, A was enough to bind with the inhibitor concentration, B.

As change the value of activator concentration A and fix the value of inhibitor concentration B. Therefore, there were some changes from stable spiral or limit cycle to unstable spiral which make the oscillation to occur at certain time then disappeared.



Figure 2: (a)Phase plane model of the activator A = 1 and inhibitor B = 3 (b) Concentration vs time



Figure 3: (a)Phase plane model of the activator A = 1.5 and inhibitor B = 3 (b) Concentration vs time





Figure 4: (a) Phase plane model of the activator A=3 and B=3. Magnified from the Figure 4(a) and (c) Concentration vs time.

4.2 Difference mode of spiral or limit cycle and its oscillation by constant the value activator concentration, A and changing the value inhibitor concentration, B.

The parameter B which was acts as inhibitor concentration in the reaction was the others parameter considered in this works where it was referring to inhibitor parameter of the solution. The system was analyzed with different values of parameters inhibitor concentration, B to observe on the oscillations and the limit of the cycle to specify the exact of the point at where the equilibrium point was formed in the reaction.

Figure (5), Figure (6), and Figure (7) were shown at which the parameter activator concentration, A was set as constant. Based on the varying of parameters of B inhibitor concentration, it showed that as increasing this parameter inhibitor concentration, B will cause the reaction from having a spiral at initial stage until then a limit cycle was formed when the inhibitor concentration, B was excess to bind with the activator concentration, A.

As change the value of inhibitor concentration B and fix the value of activator concentration A. Therefore, there were some changes from stable unstable to stable spiral or limit cycle which make the oscillation to occur from discontinuous oscillation to continuous oscillations.



Figure 5: (a)Phase plane model of the activator A = 1 and inhibitor B = 1.5 (b) Concentration vs time



Figure 6: (a)Phase plane model of the activator A = 1 and inhibitor B = 2 (b) Concentration vs time



Figure 7: (a)Phase plane model of the activator A = 1 and inhibitor B = 3 (b) Concentration vs time

4.3 Equilibrium points for the difference value activator concentration, A and constant the value inhibitor concentration, B.

As reaching the equilibrium point, it must have the exact point at which this point shows the equilibrium point where it was the limit point where the oscillation may occur. This limit point or equilibrium point was determined by using Matcont in MATLAB© software by set the activator concentration, A as the point where it will change until it reached the equilibrium point and the inhibitor concentration B will remain as constant.





Figure 8: Bifurcation diagram for Hopf point or the equilibrium point (a) Activator concentration, A vs concentration x (b) Activator concentration, A vs concentration y

(c) Inhibitor concentration, B vs concentration x (d) Inhibitor concentration, B vs

#### concentration y.

From Figure (8) it shows that the point at which the point was reached the equilibrium point when set the activator concentration, A as the manipulated variable or the variable that change the value and the observed variable were both concentration x and y where it varies as the activator concentration, A is changing. There is only one point of the equilibrium where later as it goes beyond that the stable limit cycle is form which makes the oscillation will continuously oscillate. The H point show the Hopf point where it was the point at which the oscillation will begin as it goes beyond the Hopf point. It shows that the First Lyapunov was (-0.4). This H point indicate that the beginning points for the oscillation to occur.

 Parameters
 Concentration (mole/L)

 x
 1.414215

 y
 2.121318

 A
 1.414215

 B
 3

*Table 2 : The exact point or equilibrium point for constant the inhibitor concentration, B.* 

Table 2 above show that at where the point for the oscillation to begin as set the inhibitor concentration B as constant and other values changes.



Figure 9: Phase plane of model at constant inhibitor concentration, B and various activator concentration, A.

Therefore, as getting this limit point cycle point, it can be seen from the Figure (10) below show that oscillation was occurred as it goes beyond the point of parameter even though in a small changing of values.



Figure 10: Concentration of *x* and *y* vs time

4.4 Equilibrium points for the constant value activator concentration, A and difference the value inhibitor concentration, B.

Same goes to the inhibitor concentration, B as reaching the equilibrium point, it must have the exact point at which this point shows the equilibrium point where it was the limit where the oscillation may occur. These changes until it reached the equilibrium point and the activator concentration A will remain as constant.





Figure 11: Bifurcation diagram for Hopf point or the equilibrium point (a) Activator concentration, A vs concentration x (b) Activator concentration, A vs concentration y

(c) Inhibitor concentration, B vs concentration x (d) Inhibitor concentration, B vs

# concentration y.

From Figure (11) it shows that the point at which the point was reached the equilibrium point when set the inhibitor concentration, B as the manipulated variable or the variable that change the value. The observe variable were both concentration x and y where it varies as the inhibitor concentration, B was changing. There was only one point of the equilibrium where later as it goes beyond that the stable limit cycle is form which makes the oscillation will continuously oscillate. The H point show the Hopf point where it is the point at which the oscillation will begin as it goes beyond the Hopf point. It shows that the First Lyapunov was at (-0.5). This H point indicate that the beginning points for the oscillation to occur.

*Table 3 : The exact point or equilibrium point for constant the activator concentration, A* 

Parameters	Concentration (mole/L)
x	1
у	2
A	1
В	2

Table 3 above show that at where the point for the oscillation to begin as set the activator concentration, A as constant and other values changes.



Figure 12: Phase plane of model at various inhibitor concentration, B and constant activator concentration, A.

Therefore, as getting this limit point cycle point, it can be seen from the Figure (13) below show that oscillation was occurred as it goes beyond the point of parameter even though in a small changing of values.



Figure 13: Concentration of *x* and *y* vs time

Limit cycles and bifurcations are included in this model, which has more intricate dynamics. From the perspective of dynamical systems, this is a wide field to investigate. Using this paradigm, mathematical models based on autocatalytic reactions can be further developed (Kumar, 2020a). The spatial homogeneous solutions of the reaction–diffusion system are the equilibrium and periodic solutions of the ODE system. Diffusion can cause the stability of the solutions to change.(Liao & Wang, 2016). A solution from any arbitrary starting condition rapidly relaxes toward some attractive where the dynamics develop slowly in the Brusselator model (Nazimuddin & Al, 2020).

#### 4.5 Sustainability

The Sustainable Development Goals (SDGs) are a collection of 17 global goals. These goals are designed to be a blueprint to achieve a better and more sustainable future for all. In this blueprint, it recognizes that ending poverty and other deprivations must be handled together with strategies that improve health and education, reduce inequality, and economic growth while at the same time tackling the climate change and working to preserve our natural resources such as oceans and forests. This research is closely related to Goal 4 which is quality education. This goal is to ensure access on understanding the knowledge of the systems which are most important in mathematics, engineering, biology and etc. In this study, it can be said that by perform this understanding on the dynamical systems many of important parameters and solving the problems were being applied to observe on the importance parts. Those things are valuable to understand for the coming future. It also explains about the limit of the cycle which means it can be studied that the reaction in autocatalytic reaction systems. Also based on this knowledge, it can be applied for the future analysis on how these systems can be related to our live simultaneously.