Simulation of Fuzzy *ACSH* on Membranes with Michaelis-Menten Kinetics

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Abstract. Various models have been used to represent natural phenomenon in order to gain insight on what stability is. A computing model called Fuzzy abstract rewriting system on multisets, close to reality is recently designed by introducing fuzziness on computation [1]. As an extension of this model a device named Fuzzy Artificial cell system with proteins on membrane is developed and the corresponding structure is analyzed on its parameters [2]. The aim of the present study is to investigate how the choices made in a simulation affect its accuracy and therefore the reliability of the result.

Keywords: P system · Artificial cell system · Fuzzy ACS · Proteins on membranes · Michaelis-menten behaviour · Simulation

1 Introduction

Fuzzification of membrane systems and their evolution rules which is motivated by some practical applications is a quite recent development. Rigid mathematical models employed in biology are not completely adequate for the interpretation of biological information. This fact has led to the adoption of fuzzy models and methodologies. Also it has been shown that P systems with fuzzy multiset rewriting rules are equivalent to fuzzy Turing machines. Suzuki and Tanaka [3] have introduced the multiset Rewriting system, called Abstract Rewriting System on Multisets (ARMS). Based on this system, they have developed a molecular computing model called Artificial Cell System which consists of a multiset of symbols, a set of rewriting rules and membranes [3,4]. These correspond to a class of P systems which are parallel molecular computing models proposed by Paun [5] and are based on the processing of multisets of objects in cell-like membrane structures [5].

On the other hand, P system with proteins on membranes has been introduced and the power of the system is examined in [6,7]. Following chemical reactions, the kinetics of the sulfoxidation reactions, analogous to biological systems

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were carried out by Jayaseeli and Rajagopal [8]. The computational studies of the work mentioned above, based on membrane computing has been proposed and *Kinetic ARMS* in Artificial Cell System with hierarchically structurable membrane (KACSH) is developed in [9].

Recently we have proposed a computing device that is based on Abstract Rewriting systems on multisets closely related to P system with fuzzy multiset rewriting rules and fuzzy data [1]. As an extension of this model, we have developed a new system called FACSP (Fuzzy ARMS in Artificial Cell System with proteins on membranes) and its behaviour has been studied in [2].

Models of chemically reacting systems have traditionally been simulated by solving a set of ordinary differential equations. Many researches have conducted numerical simulation to establish the simulation conditions and the impact on simulation results. In this paper, the continuous interaction of the system with environment, an operating function from kinetic equilibrium is established. A series of eigenvalues (λ) that satisfy the equation using the corresponding rate of reactions, complexes, oxidant, substrates and the significants according to the real and imaginary parts of the eigen values are obtained.

2 Preliminaries

2.1 Kinetic Studies of the Sulfoxidation Reactions [8]

In many biomimetic approaches, the study of enzymatic reactions are carried out kinetically. Jeyaseeli and Rajagopal [8] followed the spectrophotometric kinetic studies of [Iron(III)-salen] complexes catalysed H_2O_2 oxidation of organic sulfides. When the rate of reaction (k) is plotted against substrate concentration ([S]), a saturation kinetics called Michaelis-Menten behaviour is followed. They have proposed mechanisms based on the results of rate of reactions under various experimental conditions.

2.2 P System with Proteins on Membranes [7]

A system with proteins on membranes is of the form

$$\Gamma = \{O, P, \mu, w_1/z_1, \cdots, w_m/z_m, E, R_1, \cdots, R_m, i_0\}$$

where

- -m is the degree of the system (the number of membranes)
- O is the set of objects
- P is the set of proteins (with $O \cap P = \phi$)
- $-\mu$ is the membrane structure
- $w_i, i = 1$ to m are the (strings representing the) multisets of objects present in the m regions of μ
- $-z_i, i = 1$ to m are the multisets of proteins present on the membranes of μ
- $E \subseteq O$ is the set of objects present in the environment (in an arbitrarily large number of copies each)

- R_i are finite sets of rules associated with the *m* membranes of μ
- $-i_0 \in \{1, 2, \cdots, m\}$ is the label of the output membrane.

Reaction rules are applied in the following manner: In each step, a maximal multiset of rules is used, that is, no other rule is applicable to the objects and the proteins which remain unused by the chosen multiset. At each step we have the condition that each object and each protein can be involved in the application of at most one rule, but the membranes are not considered as involved in the rule applications except the division rules, hence the same membrane can appear in any number of rules of types 1-5 at the same time [7]. By halting computation, we understand a sequence of configurations that ends with a halting configuration (there is no rule that can be applied considering the objects and proteins present at that moment in the system). With a halting computation, we associate a result in the form of the multiplicity of objects present in region i_0 at the moment when the system halts. We denote by $N(\Pi)$ the set of numbers computed in this way by a given system Π . We denote in the usual way by $NOP_m(pro_r; list of types of rules)$ the family of sets of numbers $N(\Pi)$ generated by systems with at most m membranes using rules as specified in the list of types of rules, and with at most r proteins present on a membrane. When parameters m or r are not bounded, we use * as a subscript.

2.3 Fuzzy Artificial Cell System with Proteins on Membranes [2]

Definition. A Fuzzy ACS with Proteins on membranes FACSP is a construct,

$$\Gamma = \{O, P, \mu, w_1/z_1, \cdots, w_m/z_m, E, (R_p, \rho), i_0, J, \omega\}$$

where

- -m is the degree of the system (the number of membranes)
- O is the set of objects
- P is the set of proteins (with $O \cap P = \phi$)
- $-\mu$ is the membrane structure
- $w_i, i = 1$ to m are the (strings representing the) multisets of objects present in the m regions of μ
- $z_i, i = 1$ to m are the multiset of proteins (biological catalysts) present on the membranes of μ
- -E is the set of objects present in the environment (in an arbitarily large number of copies each)
- R_p are finite sets of Fuzzy multiset evolution rules, p = 1 to m of μ
- ρ is the partial order relation over R_p
- $-i_0 \in \{1, 2, \cdots, m\}$ is the elementary membrane (output)
- $-J = \{R_{pi} \in R_p / 1 \le i \le q\}, q = \text{cardinality of } R_p$
- $-\omega: J \to [0,1]$ is the membership function s.t. $\omega(R_{pq}) = i, i \in [0,1]$.

The rules are used in the non-deterministic maximally parallel way:

The same rules are applied to every membrane. There are no rules specific to a membrane. All the rules are applied in parallel. In every step, all the rules are applied to all objects in every membrane that can be applied. If there are more than one applicable rule that can be applied to an object and protein then one rule is selected randomly. If a membrane dissolves, then all the objects in its region are left free in the region immediately above it. All objects and proteins not specified in a rule and which do not evolve are passed unchanged to the next step. At each step we have the condition that each object and each protein can be involved in the application of at most one rule, but the membranes are not considered as involved in the rule applications except the division rules, hence the same membrane can appear in any number of rules at the same time.

By halting computation, we understand a sequence of configurations that ends with a halting configuration (there is no rule that can be applied considering the objects and proteins present at that moment in the system). With a halting computation we associate a result in the form of the multiplicity of objects present in region i_0 at the moment when the system halts.

A Fuzzy ACS with proteins on membranes generates a language L(FACSP)as follows: An object $x \in O^*$ which is present in the region i_0 at the moment when the system halts is said to be in L(FACSP) iff it is derivable from any object $S \in O$ and the grade of membership $\omega_{L(FACSP)}(x)$ is greater than 0, where

$$\omega_{L(FACSP)}(x) = \begin{pmatrix} max \\ 1 \le k \le n \end{pmatrix} \left[\begin{pmatrix} min \\ 1 \le i \le l_k \end{pmatrix} ? \omega(R_i^k) \right],$$

 $x \in O^*$ and n is the number of different derivatives that x has in FACSP, l_k is the length of the k^{th} derivative chain, R_i^k denotes the label of the i^{th} multiset evolution rule used in the k^{th} derivative chain, $i = 1, 2, \ldots, l_k$.

Clearly, $\omega_{L(FACSP)}(x) =$ Strength of the strongest derivative chain for S to x for all $x \in O^*$.

We denote in the usual way by $FACSP_m(pro_r; list of types of rules)$ the family of languages L(FACSP) generated by systems Π with at most m membranes, using rules as specified in the *list of types of rule* and with at most r proteins present on a membrane. When parameters m or r are not bounded, we use * as a subscript.

3 Simulation of FACSP

The mathematical simulation pattern of rate constants (k) with substrate concentrations are analysed.

3.1 FACSP in Oxidation of Sulfides

Process. We describe the formation of intermediate between complex and the oxidant.

(a)
$$Z + X(F3)X \rightarrow X(F4O)X;$$

 $X(F4O)X + Y - RSR' \rightarrow X(F3)X + Y - RSOR'$



R-C6H5, R'-CH3

A simple abstract reaction scheme is followed.

Case I: X = HFollowing convention is used to do the computation. $Y = H = L, Y = OCH_3 = M, Y = CH_3 = N,$ $Y = F = P, Y = Cl = Q, Y = Br = U, Y = NO_2 = V.$ Now (a) will have the following reaction rules

- 1. $Z + H(F3)H \rightarrow H(F4O)H;$ $H(F4O)H + L-RSR' \rightarrow H(F3)H + L-RSOR'$
- 2. $Z + H(F3)H \rightarrow H(F4O)H;$ $H(F4O)H + M-RSR' \rightarrow H(F3)H + M-RSOR'$
- 3. $Z + H(F3)H \rightarrow H(F4O)H;$ $H(F4O)H + N-RSR' \rightarrow H(F3)H + N-RSOR'$
- 4. $Z + H(F3)H \rightarrow H(F4O)H;$ $H(F4O)H + P-RSR' \rightarrow H(F3)H + P-RSOR'$
- 5. $Z + H(F3)H \rightarrow H(F4O)H;$ $H(F4O)H + Q-RSR' \rightarrow H(F3)H + Q-RSOR'$
- 6. $Z + H(F3)H \rightarrow H(F4O)H;$ $H(F4O)H + U-RSR' \rightarrow H(F3)H + U-RSOR'$
- 7. $Z + H(F3)H \rightarrow H(F4O)H;$ $H(F4O)H + V - RSR' \rightarrow H(F3)H + V - RSOR'$

3.2 Behaviour of FACSP

Consider the FACSP

$$\Gamma = (O, P, \mu, w_1/z_1, w_2/z_2, E, (R_p, \rho), i_0, J, \omega)$$

where

- $O = \{Z, A_1, B, S_i, P_i, i = 1, \dots, 7\},\$
- $P = \{A_1, B\},\$
- $\mu = [_1[_2]_2]_1,$
- w_1, w_2 are the multisets of objects present in the regions 1, 2 of μ , $w_1 = \{Z, S_i, i = 1, ..., 7\}, w_2 = \{\phi\},$
- z_1, z_2 are the multisets of proteins present on the membranes 1, 2 of μ , $z_1 = \{A_1\}, z_2 = \{\phi\},\$

$$-E = \{\phi\},$$

- R_p are finite sets of Fuzzy multiset evolution rules, $p = \{1, 2\}$

$$-
ho = \phi,$$

- $-i_0 = 2$ is the output membrane,
- $J = \{R_{pi} \in R_p / q = 1 \le i \le q\}, q =$ cardinality of R_p ,
- $-\omega: J \to [0,1]$ is the membership function s.t. $\omega(R_{pq}) = i, i \in [0,1]$, where

$$\omega_{L(FACSP)}(x) = \binom{max}{1 \le k \le n} \left[\binom{min}{1 \le i \le l_k} ? \omega(R_i^k) \right]$$

and $x \in O^*$

 $R_p = \{R_1, R_2\}$ consists the following evolution rules.

$$R_{11} : \begin{bmatrix} [A_{1}|Z]_{1} & \rightarrow [1B|\phi]_{1}; \\ [1B|S_{1}]_{1} & \rightarrow [1A_{1}] & [2 \ |P_{1}]_{2}]_{1} \\ \text{with } \omega(R_{11}) = 0.0025 \\ R_{12} : \begin{bmatrix} [A_{1}|Z]_{1} & \rightarrow [1B|\phi]_{1}; \\ [1B|S_{2}]_{1} & \rightarrow [1A_{1}] & [2 \ |P_{2}]_{2}]_{1} \\ \text{with } \omega(R_{12}) = 0.01 \\ R_{13} : \begin{bmatrix} [A_{1}|Z]_{1} & \rightarrow [1B|\phi]_{1}; \\ [1B|S_{3}]_{1} & \rightarrow [1A_{1}] & [2 \ |P_{3}]_{2}]_{1} \\ \text{with } \omega(R_{13}) = 0.0059 \\ R_{14} : \begin{bmatrix} [A_{1}|Z]_{1} & \rightarrow [1B|\phi]_{1}; \\ [1B|S_{4}]_{1} & \rightarrow [1A_{1}] & [2 \ |P_{4}]_{2}]_{1} \\ \text{with } \omega(R_{14}) = 0.0016 \\ R_{15} : \begin{bmatrix} [A_{1}|Z]_{1} & \rightarrow [1B|\phi]_{1}; \\ [1B|S_{5}]_{1} & \rightarrow [1A_{1}] & [2 \ |P_{5}]_{2}]_{1} \\ \text{with } \omega(R_{15}) = 0.0011 \\ R_{16} : \begin{bmatrix} [A_{1}|Z]_{1} & \rightarrow [1B|\phi]_{1}; \\ [1B|S_{6}]_{1} & \rightarrow [1A_{1}] & [2 \ |P_{6}]_{2}]_{1} \\ \text{with } \omega(R_{16}) = 0.0009 \\ R_{17} : \begin{bmatrix} [A_{1}|Z]_{1} & \rightarrow [1B|\phi]_{1}; \\ [1B|S_{7}]_{1} & \rightarrow [1A_{1}] & [2 \ |P_{7}]_{2}]_{1} \\ \text{with } \omega(R_{17}) = 0.00027 \\ \end{array} \right)$$

In its initial configuration, the system contatins 2 membranes with 8 objects $\{Z, S_i, i = 1, ..., 7\}$ and a biological protein A_1 on membrane 1. It has two steps. In the first step, any one of the 7 rules is selected randomly. Let the rule R_{11} be applied. Then the protein A_1 is changed into B. In the second step, the protein change back from B to A_1 and the object S_1 evolved into P_1 and move to membrane 2. Since there is no rule that can transform the object in membrane 2 is P_1 .

$$\max_{1 \le k \le n} \begin{bmatrix} \min_{1 \le i \le l_1} (0.0025) \end{bmatrix} = 0.0025;$$

 $\omega_{L(FACSP)}(P_1) = 0.0025$





Similar process will be done when other rules are applied. As a result, the membership values $\omega_{L(FACSP)}(P_i)$ for i = 1 to 7 are obtained. Hence $L(FACSP) = \{P_i/i = 1 \text{ to } 7\}.$

We obtain different languages with corresponding membership values for different complexes $(A_i, i = 1 \text{ to } 7)$. The membership values for different complexes are tabulated as follows (Table 1).

We denote by $FACSP_2(pro_1; 7ffp)$ the family of languages L(FACSP) generated by Γ with atmost 2 membranes using rules as specified in the 7ffp rules and with atmost one protein.

3.3 Mathematical Modeling and Simulation of FACSP

Chemical equations are commonly written in the following way:

$$A + B \longrightarrow C + D$$

indicating that species A and B react together to form species C and D. From the chemical equation we can easily write the rate equation. It is important to note

Complex	$\omega(P_1)$	$\omega(P_2)$	$\omega(P_3)$	$\omega(P_4)$	$\omega(P_5)$	$\omega(P_6)$	$\omega(P_7)$
A_1	0.0025	0.01	0.0059	0.0016	0.0011	0.0009	0.00027
A_2	0.006	0.034	0.023	0.0054	0.0028	0.0029	0.0009
A_3	0.0055	0.023	0.019	0.0062	0.0025	0.0026	0.0008
A_4	0.0017	0.0025	0.0019	0.0009	0.00084	0.00072	0.00023
A_5	0.00089	0.0018	0.00096	0.00062	0.00051	0.0004	0.00017
A_6	0.015	0.066	0.043	0.011	0.008	0.0065	0.0021
A_7	0.00053	0.0011	0.00076	0.00042	0.0004	0.0003	0.00019

Table 1. Membership values $\omega_{L(FACSP)}(P_i) = \omega(P_i)$.

that most chemical systems are assumed to follow mass action kinetics, meaning that the reaction rate is proportional to the concentration of the reactants.

$$-[A] = -r_a = k[A][B]$$

Here [A] represents the concentration of species A, r_a is the reaction rate and k is the rate constant of the reaction. r_a is by convention negative since A is being consumed in the reaction. Now we describe the natural phenomenon of Fuzzy ACS in oxidation of sulfides. The mathematical model [10] is used because of its theoretical simplicity. The mathematical modeling of FACSP is given below.

$$[{}_{1}A_{1}|Z]_{1} \frac{k_{1}}{k_{-1}} [{}_{1}B|\phi]_{1} \tag{1}$$

$$[{}_{1}B|S_{1}]_{1} \xrightarrow{k_{2}} [{}_{1}A_{1}|[{}_{2}|P_{1}]_{2}]_{1}$$

$$\tag{2}$$

In Eqs. (1) and (2), k_i , i = 1, 2 are the reaction rate for each individual reaction, while Z, A_1, B, S_1 and P_1 are species. The molar concentration of A_1 is denoted by $[A_1]$ likewise for the other species. The equations for the evolution of $[A_1]$ and $[S_1]$ are as follows.

$$d[A_1]/dt = k_2[B][S_1] - k_1[Z][A_1]$$
(3)

$$d[S_1]/dt = -k_2[B][S_1]$$
(4)

The above equations are of the form

$$d[A_1]/dt = F_1([A_1], [S_1])$$
$$d[S_1]/dt = F_2([A_1], [S_1])$$

where

$$F_1([A_1], [S_1]) = k_2[B][S_1] - k_1[Z][A_1]$$
$$F_2([A_1], [S_1]) = -k_2[B][S_1]$$

Equilibria. The equilibria of (3) and (4) is given by solving the system

$$k_2[B][S_1] - k_1[Z][A_1] = 0 (5)$$

$$-k_2[B][S_1] = 0 (6)$$

From (5)

$$k_2[B][S_1] = k_1[Z][A_1]$$

$$[B] = (k_1[Z][A_1])/(k_2[S_1])$$
(7)

(5)-(6) gives

$$2k_{2}[B][S_{1}] - k_{1}[Z][A_{1}] = 0$$

$$k_{1}[Z][A_{1}] = 2k_{2}[B][S_{1}]$$

$$[A_{1}] = (2k_{2}[B][S_{1}])/(k_{1}[Z])$$
(8)

$$[S_1] = (k_1[Z][A_1])/(2k_2[B])$$
(9)

From Eq. (5), we obtain $S_1 = \alpha(A_1)$ where $\alpha = k_1[Z]/k_2[B]$

$$([A_1], [S_1]) = ([A_1], \alpha[A_1])$$

From Eq. (6),

$$([A_1], [S_1]) = (0, 0)$$

Hence (0,0) and $([A_1], \alpha[A_1])$ are the equilibrium of the system.

Stability. To evaluate stability, we evaluate the Jacobian at the stationary state.

$$\begin{aligned} \partial(F_1)/\partial[A_1] &= -k_1[Z]; \quad \partial(F_1)/\partial[S_1] = k_2[B]; \\ \partial(F_2)/\partial[A_1] &= 0; \quad \partial(F_2)/\partial[S_1] = -k_2[B] \\ J &= \begin{pmatrix} \partial(F_1)/\partial[A_1] \ \partial(F_1)/\partial[S_1] \\ \partial(F_2)/\partial[A_1] \ \partial(F_2)/\partial[S_1] \end{pmatrix} = \begin{pmatrix} -k_1[Z] \ k_2[B] \\ 0 \ -k_2[B] \end{pmatrix} \\ Trace \ J &= -(k_1[Z] + k_2[B]) \end{aligned}$$

The eigen value equation or characteristic equation is applied in order to evaluate the stationary state.

$$det(J - \lambda I) = 0$$

Arranging these values into matrix form gives

$$\begin{pmatrix} -k_1[Z] - \lambda & k_2[B] \\ 0 & -k_2[B] - \lambda \end{pmatrix} = 0$$

i.e.,

$$\lambda^2 + (k_1[Z] + k_2[B])\lambda + k_1k_2[Z][B] = 0$$

Using Eq. (7)

$$[S_1]\lambda^2 + ([S_1] + [A_1])k_1[Z]\lambda + k_1^2[Z]^2[A_1] = 0$$
(10)

Here we state that,

$$k_1 = 2.5 \times 10^{-3}, [Z] = 5 \times 10^{-3}, [S_1] = i \times 10^{-3}, i = 0, 2, 4, 10, [A_1] = 2 \times 10^{-4}$$

Solving the quadratic Eq. (10) for different values of $[S_1]$ using MATLAB, eigen values of the Jacobian matrix are obtained.

The eigen values for different catalysts for the sulfoxidation reactions are tabulated in Table 2. From the data collected, all Eigen values are real and negative since $\lambda_1 < 0$ and $\lambda_2 < 0$. Thus the system is stable. The changes for the eigen values with substrate concentrations are plotted.

		A_1	A_2	A_3	A_4	A_5	A_6	A_7
s_1	λ_1	-1.25	-3	-2.75	-85	-44.5	-7.5	-26.5
	λ_2	0	0	0	0	0	0	0
s_2	λ_1	-0.05	-0.0017	-0.00115	-0.0125	-0.9	-0.0033	-0.55
	λ_2	-0.005	-0.00017	-0.000115	-0.00125	-0.09	-0.00033	-0.055
s_3	λ_1	-0.0295	-0.00115	-0.095	-0.95	-0.48	-0.00215	-0.38
	λ_2	-0.00148	-0.000058	-0.00475	-0.0475	-0.024	-0.000108	-0.019
s_4	λ_1	-0.8	-0.027	-0.95	-0.0475	-0.31	-0.055	-0.21
	λ_2	-0.0267	-0.0009	-0.48	-0.024	-0.0103	-0.00183	-0.007
s_5	λ_1	-0.55	-0.014	-0.0125	-0.42	-0.255	-0.04	-0.2
	λ_2	-0.0138	-0.00035	-0.00031	-0.0105	-0.0064	-0.001	-0.005
s_6	λ_1	-0.45	-0.0145	-0.013	-0.36	-0.2	-0.0325	-0.15
	λ_2	-0.009	-0.00029	-0.00026	-0.0072	-0.004	-0.00065	-0.003
s_7	λ_1	-0.135	-0.45	-0.4	-0.00115	-8.5	-0.0105	-9.5
	λ_2	-0.0023	-0.0075	-0.0067	-0.000019	-0.142	-0.00017	-0.158

Table 2. Eigen values for different catalysts

When the concentration of the substrate (sulfides) increases there is an increase in rate constant and attains saturation at higher concentration (Fig. 1). When these results are examined mathematically using Fuzzy ACSH on membranes there is a consistancy between the pattern of plots obtained for kinetic results. As the concentration of the substrate increases, the eigen values first decreases and increases. It becomes constant at higher rate constant. This behaviour can be correlated to the saturation kinetics of chemical reactions. The pattern is shown in figure (Figs. 2 and 3).



Fig. 1. k_1 vs. [substrate] for complex 1 catalyzed H_2O_2 oxidation of 1–7



Fig. 2. (S, λ_1)



Fig. 3. (S, λ_2)

4 Conclusion

The new membrane computing model FACSP (Fuzzy ARMS in Artificial Cell System with Proteins on membranes) is analysed in its environment. The stability and equilibrium of the system are determined. The eigen values and the critical points of different catalysts for the sulfoxidation reactions are obtained. A mathematical approach is constructed to show the consistency of Fuzzy ACSH on membranes with the Michaelis-Menten kinetics. It is interesting to note that there is a correlation between the two types of plots.

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