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, 2014

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FACULTY OF PHYSICAL CHEMISTRY

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**DSORPTION-DESORPTION  
PROCESSES AT THE SURFACE OF  
PLASMONIC SENSORS**

Doctoral Dissertation

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: 544.7

# **ADSORPTION-DESORPTION PROCESSES ON THE SURFACE OF PLASMONIC SENSORS**

## **Summary**

Chemical and biological plasmonic sensors are devices in which the adsorption-desorption (a-d) processes fundamentally determine their output signal and at the same time limit their ultimate performance by causing stochastic fluctuations of the signal (adsorption-desorption noise). A knowledge about the kinetics and stochastics of adsorption-desorption processes in plasmonic sensors is essential for the optimal design of these sensors and improvement of their performance, especially in the case of contemporary sensors based on metal-dielectric nanocomposites (nanoplasmonic sensors).

The goal of this dissertation is the establishment of tools for the investigation of adsorption-desorption processes in plasmonic sensors which ensures control of sensor figures of merit and thus the optimal design or optimal mode of operation. To this purpose modeling of a-d processes has been performed within the context of the operation of plasmonic sensors as refractometric devices, an analysis of dynamical states and the existing fluctuations of the adsorbed amount of gases on the surface of plasmonic sensors in different modes of operation (transient and stationary modes) as well as the analysis of the influence of real physical parameters of the system (effective surface, temperature, pressure) to the final performance of the sensor (responsivity, response time, selectivity).

In Introduction it is first shown in which manner the operation of plasmonic sensors depends on adsorption-desorption processes. The use of two known and often used models of monolayer adsorption has been considered (the fundamental model of monolayer adsorption and Lagergren model of reaction with first order kinetics) for the definition and the analysis of the signal output of a plasmonic sensor. These two models are both used for the investigation of adsorption-desorption processes throughout this work.

Chapter 2 considers in detail modeling of adsorption-desorption processes which occur in plasmonic sensors at the interface between positive and negative relative dielectric permittivity materials. The mentioned models are also given for the case of multicomponent adsorption, and special attention has been dedicated to the influence of



the adsorbate molecule size to the adsorption kinetics. A method of modeling of adsorption rate constant is proposed that takes into account this influence, as well as a method for the estimation of the maximal surface density of adsorbed particles. It has been shown that the justifiability of the use of Lagergren model depends on the quantitative values of the adsorption and desorption rate constants.

The connection between the number of adsorbed particles and the sensor readout (refractive index change) has been established and described in Chapter 3. The deterministic solutions for the refractive index are given in detail in a convenient analytical form where stationary states are clearly separated from the transient response components and time constants in the transient components are defined. Solutions are given from the general ones to the special cases. Multicomponent and single component adsorption has been considered, with or without taking into account adsorbate molecule size to the adsorption kinetics.

After that, Chapter 4 presents the procedure of stochastic analysis and its application for the investigation of refractive index fluctuations in plasmonic sensors which represent the intrinsic noise of these sensors. The application of this procedure to monolayer adsorption, modeled first by the fundamental model, and then by Lagergren model. The use of Lagergren model is given for the cases of single-component gas adsorption, as well as of a mixture of two or more gases, taking into account the influence of the molecule size to adsorption kinetics. The results of stochastic analysis are summarized in tabular form. It is finally shown how the obtained fluctuations of the number of adsorbed particles of each separate component in the mixture are used to determine refractive index fluctuations. This is first done for the general case of a mixture with an arbitrary number of components, and then the procedure is demonstrated for the examples of 1, 2 and 3 component mixtures.

The application of the obtained theoretical results to numerical experiments is described. First the guidelines are given for avoidance of the propagation of numerical errors in these experiments and for the interpretation of the results. Then a comparative analysis is done of the results obtained by the fundamental model of monolayer adsorption and the results obtained by generalizing Lagergren equation, and within this criteria are defined for the determination of the justifiability of the application of the

simplified linear model. It is shown that the same criteria are valid for the application of the results of deterministic analysis and of stochastic analysis.

According to the obtained results, the level of adsorption-desorption noise is reversely proportional to the area of material with negative dielectric permittivity, which causes higher noise in nanostructured sensors. At the same time, the results of deterministic analysis point to the fact that detection of trace amount of gases using plasmonic sensors is possible and can be improved by cooling and increasing the sensor active area. Noise analysis shows that there is an optimum moment for readout when relative fluctuations are minimal. The results of this dissertation show that there are cases when simultaneous detection of a larger number of analytes is possible using a single measurement on a single element plasmonic sensor. It also turns out that in some cases it is possible to enhance sensing of trace amounts of gases by adding a controlled amount of carrier gas to the mixture, which ensures detection of gas amounts that would elsewhere be below the detection threshold.

The results of this dissertation represent a toolbox applicable for design, optimization and further research of a-d processes in plasmonic devices, but also in other devices based on a similar principle, i.e. those that can be described by the presented mathematical models of adsorption and desorption.

**Key words:** monolayer adsorption, plasmonic sensor, noise, fluctuations, stochastic analysis, gas sensing

**Scientific field:** physical chemistry

**Field of academic expertise:** biophysical chemistry and dynamics of non-equilibrium processes

**UDK:** 544.7

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1	.....	1
1.1	.....	2
1.2	.....	3
1.3 o -	.....	9
1.3.1 o	.....	10
1.3.2	.....	12
2	.....	15
2.1	.....	15
2.2	.....	18
2.2.1	.....	21
2.2.2	.....	24
2.3	.....	26
3	.....	29
3.1	.....	29
3.2	.....	30
3.3	.....	33
3.3.1	.....	37
3.3.2	.....	41
3.4	.....	43
4	.....	48
4.1	.....	48
4.2	.....	50
4.3.	.....	56
4.4	.....	62
4.4.1 -	.....	62
4.4.2 -	.....	67
4.4.3 -	.....	73
4.4.4	.....	76
4.5	.....	84
4.6	.....	87

4.6.1	-	.....	88
4.6.2	-	.....	90
4.6.3	-	.....	91
5		.....	93
5.1		.....	93
5.2		.....	99
5.2.1		.....	99
5.2.2		.....	104
5.3		.....	110
5.4		.....	116
6		.....	122
7		.....	128
8		.....	131
9		.....	139
		.....	139
		.....	143
		.....	145
	(Lapla e)	.....	145
		.....	146
		.....	147
		.....	149
		.....	152
		.....	154
		.....	159

## Списак илустрација

. 1.1	( ), ( ), ( )	5
. 1.2	: 2D	7
. 1.3		8
. 2.1 A		19
. 2.2	: )	19
. 2.3	(8.38-10.87), ) (9.62-16.70), ) (9.08-12.05), ) (10.18-13.36)	25
. 2.4	/	26
. 2.5	( ) ( ) 1 ( ) 1 ( )	27
. 4.1	$\Delta t$	52
. 5.1	MATLAB, 3-	94
. 5.2	, MATLAB	94
. 5.3	1 P	95
. 5.4	0.1 mPa Symbolic Toolbox MATLAB	96
. 5.5	Symbolic Toolbox, MATLAB - 2	96
. 5.6	Symbolic Toolbox, MATLAB2012	97
. 5.7		

	1, 2, 3, 4, 5 1mm <sup>2</sup> , 100mm <sup>2</sup> , 10 <sup>4</sup> mm <sup>2</sup> , 1m <sup>2</sup> 100m <sup>2</sup> . : 50 Pa, 3 dm <sup>3</sup> , 300K.....	100
. 5.8	10 <sup>4</sup> m <sup>2</sup> , 3 dm <sup>3</sup> , : 1, 2 3 : 10 <sup>-3</sup> Pa, 0.1 Pa 10 <sup>5</sup> Pa, .....	101
. 5.9	6.25·10 <sup>14</sup> 3 dm <sup>3</sup> , ( ) ( ) .....	102
. 5.10	= 10 <sup>-6</sup> m <sup>2</sup> , 3·10 <sup>-6</sup> m <sup>3</sup> . 10 <sup>-6</sup> 100 Pa, = 100 , = 300 .....	108
. 5.11		
. 5.12	, V = 3·10 <sup>-6</sup> m <sup>3</sup> .....	108
. 5.13	..... 10 <sup>-3</sup> m <sup>2</sup> (Au), = 300 , V = 0.003 m <sup>3</sup> , p = 0.5·10 <sup>5</sup> Pa, a ( ) ( , ), ( ) ( - ).....	111
. 5.14		
. 5.15	5 : - , - - , -   - .....	111
. 5.16		112
. 5.17		114
. 5.18		114
. 5.19	90 kPa .....	116
. 5.20	, 20% 78% ( 10 <sup>5</sup> Pa) : 2% ) = 400 .....	117
	300 .....	118

. 5.21	(2% CO <sub>2</sub> , 20% O <sub>2</sub> , 78%N <sub>2</sub> , 10 <sup>5</sup> Pa)	119
. 5.22	3 ( , , ) : ( , ) ( CH <sub>4</sub> +CO <sub>2</sub> , - 0.01mm <sup>2</sup> , CH <sub>4</sub> +C <sub>6</sub> H <sub>6</sub> )	120
. 5.23	0.3 mPa (2.1 ppm)	120

### Списак табела

Табела 1	анализе за разне случајеве.....	84
Табела 2	Параметри Рикатијеве једначине у моделу монослојне адсорпције.....	141
Табела 3		146

## Ознаке (по редоследу појављивања у тексту)

$n$

$\sim 11$  а а

$f$  , ,  $i$ -

$A_g$  , ,  $i$ -

$A_a$  , ,  $i$ -

$k_a$  брзинска константа за адсорпцију према егзактном моделу за ,  
 ,  $i$ -

$k_d$  за , ,  $i$ -

$r$  број гасова у смеси

$N_a$  , ,  $i$ -

$N_0$  , ,  $i$ -

$N_{a,\epsilon}$

$k_L$

$\chi_c$  површинска густина адсорпционих центара на ефективној површи сензора

$\chi$  површинска густина адсорбованих честица (максималан број честица којима се површ може прекрити у монослоју, по јединици површине) за ,  
 ,  $i$ -

$S$  -

,  $i$ -

$N_f$  ,

,  $i$ -

$k_l$  константа брзине за адсорпцију за  
 , ,  $i$ -

$v_d$  брзина десорпције

$v_a$  брзина адсорпциј

$v_l$  брзина адсорпције



$\bar{v}$

$n$

$k_B$

(Boltzmann,  $1.38 \cdot 10^{-23}$  J/K =  $8.617385 \cdot 10^{-5}$  eV/K)

$t$

$p_i$

$i-$

$m$

,

$i-$

$i$

$i-$

$N_{av} A$

,

/

( $6.02 \cdot 10^{23}$ )

$V$

/

$\dagger_r$

,

$i-$

$\tau_0$

(

),

$i-$

$R$

$d$

,

$i-$

$\chi$

,

$k_{II}$

$\Delta t$

$F$

$\overline{N_a}$

$\overline{N_a^2}$

$D$

$\dagger$

$u$

$P$

$Q$

$G$

$n_e$

$n_A$

$i-$

$n_{eff}$

$n_{eff}$

$V_{ad}$

$N_a$

$V_{max}$

$V_{lad}$

$w$

$\rho$

$S_f$

### Скраћенице

$SPP$  surface plasmon polariton,

$TM$

( )

[1]–[6].

1998  
 0.5 ng/cm<sup>2</sup> ( 0.003 ) [7],  
 10<sup>-8</sup> [8].  
 [9]–[16]

[17].

## 1.1



## 1.2

1902. " " ( )  
[18]. ([19]– [21]), ([22], [23]), ([Plasmonics Inc](#), [MicroChemLab™](#), [MicroHound™](#), [Sandia National Laboratories](#)).

- [24].  
( / )  
( )  
,  
,  
(surface plasmon polariton, SPP). SPP

( . ).  
,  
je 2006.  
SPP  
VLSI (very large scale  
of integration)

[25].

, a , , -  
[26]–[30].

SPP

( , ),

( )

[9]:

[24].

[31].

(SPP)

$< 0$  (

)

)

$> 0$  (

,

SPP

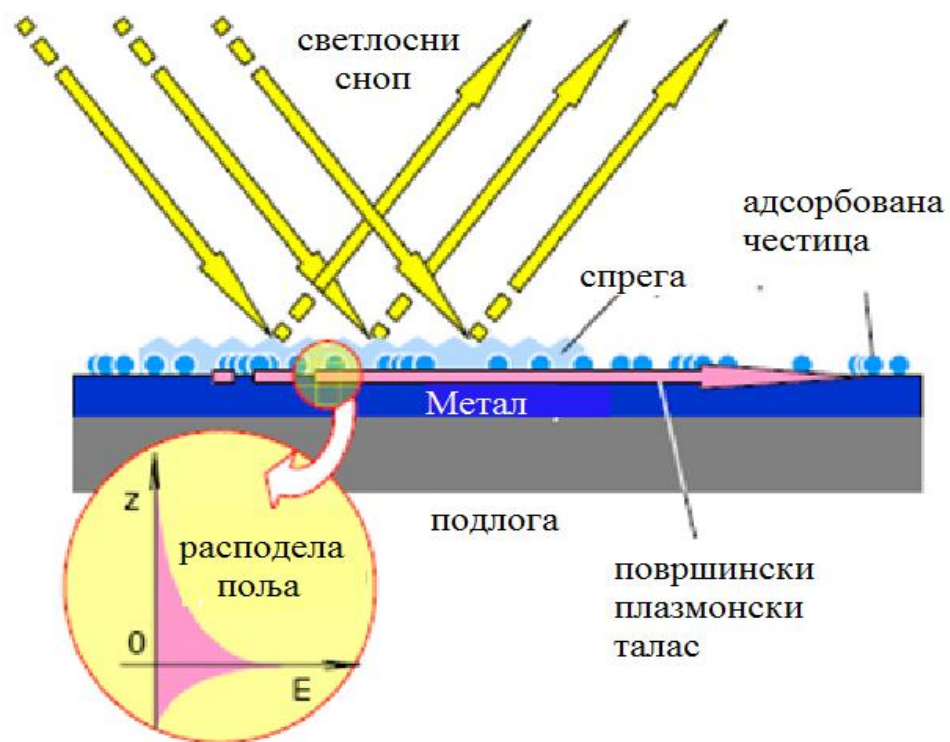
$n$  (

~,

$n = [\sim]^{1/2}$ ).

1.1

[24], [32].



Сл. 1.1 Основни функционални блок у плазмонским направама: упадни сноп зрачења (жуте стрелице), призма за спрегау (светло плаво), слој материјала на коме је могуће успостављање плазмонског таласа (тамно плаво) и плазмонски талас (ружичаста стрелица). У увеличаном кругу је расподела електричног поља.

(Kretschmann),

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 ,  
 (  $p$  TM  
 ), SPP ( )  
 ).

( )  
 " " ( "endfire coupling"),  
 .  
 ( )  
 ).

SPP

SPP- (

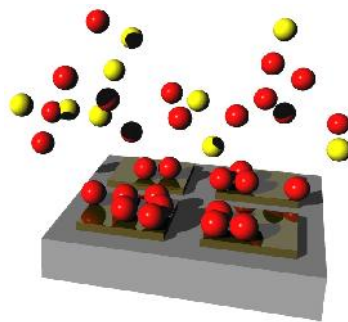


) ,  
SPP ( )  
SPP ,

(transparent conductive oxides, TCO) [33],

SPP

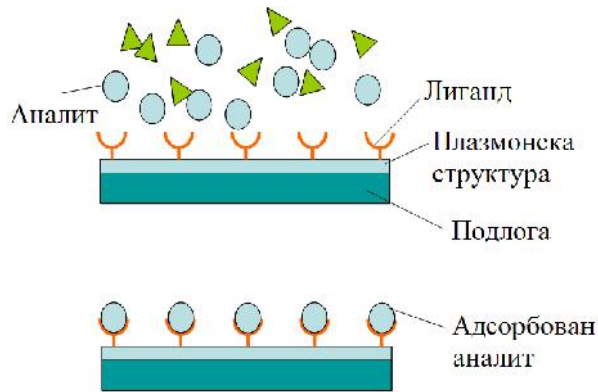
( , ),



Сл. 1.2 Пример метаматеријала на каквом је могуће успостављање плазмонског таласа: 2D периодичан низ квадратних сегмената од метала окружен бинарном смешом гасова.

( )

2D



Сл. 1.3 Основни функционални блок плазмонских сензора са површином функционализованом тако да селективно прихвата циљани аналит

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1.3.

SPP

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### 1.3 Mo

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[6], [34]–[37].

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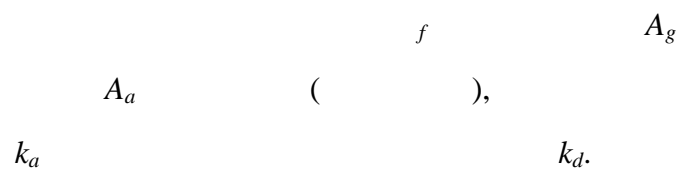
/

· ,

1.3.1

o

[37],



[38]:



$$N_f \quad N_g \quad N_a, \quad :$$

$$\frac{dN_a}{dt} = k_a N_g N_f - k_d N_a \quad (1.2)$$

$$, \quad N_0$$

(Riccati)

,  $N_a$ :

$$\begin{aligned} \frac{dN_a}{dt} &= k_a (N_0 - N_a)(M - N_a) - k_d N_a \\ &= k_a N_a^2 - [k_a (N_0 + M) + k_d] N_a + k_a N_0 M \end{aligned} \quad (1.3)$$

(1.3) [37].

$$N_a(t) = \frac{N_0 M [1 - e^{-k_a(x-s)t}]}{x - s e^{-k_a(x-s)t}} \quad (1.4)$$

(1.3):

$$\begin{aligned} x &= \frac{[k_a (N_0 + M) + k_d] + \sqrt{[k_a (N_0 + M) + k_d]^2 - 4k_a^2 N_0 M}}{2k_a} \\ &= \frac{1}{2} \left[ \frac{k_d}{k_a} + N_0 + M + \sqrt{\left( \frac{k_d}{k_a} + N_0 + M \right)^2 - 4N_0 M} \right], \end{aligned} \quad (1.5)$$

$$\begin{aligned}
S &= \frac{[k_a(N_0 + M) + k_d] - \sqrt{[k_a(N_0 + M) + k_d]^2 - 4k_a^2 N_0 M}}{2k_a} \\
&= \frac{1}{2} \left[ \frac{k_d}{k_a} + N_0 + M - \sqrt{\left( \frac{k_d}{k_a} + N_0 + M \right)^2 - 4N_0 M} \right].
\end{aligned} \tag{1.6}$$

### 1.3.2

(Homola, 2006) [39]

(1.3)

$$k_l \quad k_a N_0,$$

$$\frac{dN_a}{dt} \cong k_a N_0 (M - N_a) - k_d N_a = k_l (M - N_a) - k_d N_a. \tag{1.7}$$

[39],

1898.

[40].

[41]

[42]

( [43]

$N_{a,\infty}$ ,

,  $k_L$ .

$$\frac{dN_a}{dt} = k_L(N_{a,\infty} - N_a). \quad (1.8)$$

(1.7) (1.8)

$k_L$

$$k_L = k_a + k_d,$$

$$N_{a,\infty} = Mk_l/(k_l + k_d).$$

[39],

:

),

( ),

, SPR

(surface plasmon resonance)

[5], [44]–[46]

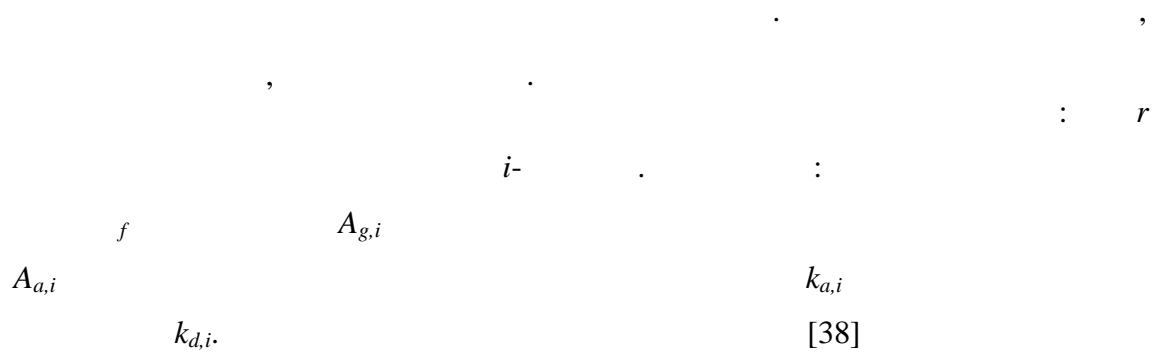
(1.3)



2

2.1

(1.1)-(1.6)



$r$

$$\frac{dN_{a,i}}{dt} = k_{a,i}N_{g,i}N_f - k_{d,i}N_{a,i} \quad i=1,\dots,r. \quad (2.2)$$

$$N_f = M - \sum_{j=1}^r N_{a,j} \quad (2.3)$$

$$\frac{dN_{a,i}}{dt} = k_{a,i} (N_{0,i} - N_{a,i}) \left( M - \sum_{j=1}^r N_{a,j} \right) - k_{d,i} N_{a,i}, \quad i = 1, \dots, r. \quad (2.4)$$

$$\begin{aligned}
\frac{dN_{a,i}}{dt} &\cong k_{a,i}N_{0,i} \left( M - \sum_{j=1}^r N_{a,j} \right) - k_{d,i}N_{a,i} \\
&= k_{l,i} \left( M - \sum_{j=1}^r N_{a,j} \right) - k_{d,i}N_{a,i}
\end{aligned}
, \quad i = 1, \dots, r. \tag{2.5}$$

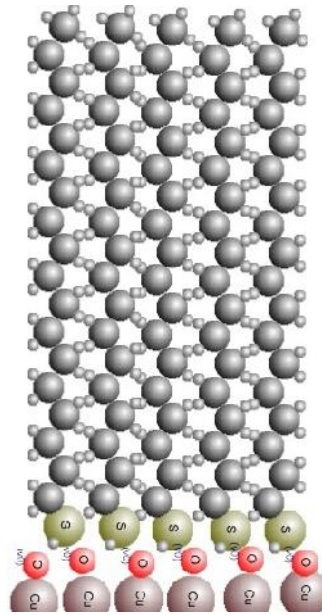
## 2.2

2.1 Marvin Sketch [47]

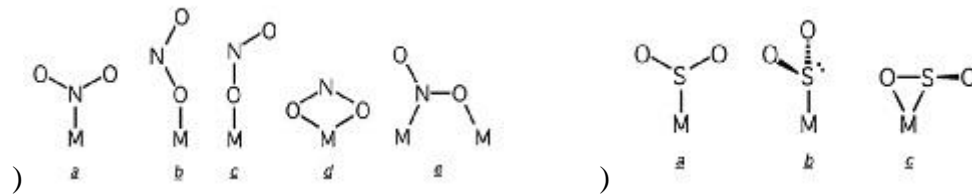
( )

[48]–[50].

( 2.2)

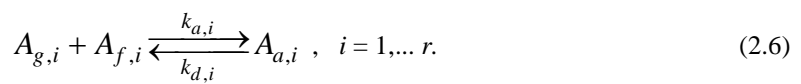


Сл. 2.1 Адсорпција меркаптана на бакар оксиду



Сл. 2.2 Адсорпција зависи од структуре и оријентације молекула: а) азот диоксид б) сумпор диоксид,  $M$  представља адсорпционе центре на површи метала. Слика је преузета из [51]

$$\begin{aligned}
 & \text{),} \\
 & \text{),} \\
 & \text{);} \quad (2.1)
 \end{aligned}$$



(2.2),  
 $N_{f,i}$

$$\frac{dN_{a,i}}{dt} = k_{a,i}N_{g,i}N_{f,i} - k_{d,i}N_{a,i}, \quad i = 1, \dots, r. \quad (2.7)$$

,  $\chi_i$ ,  
 $M_{,i}$ ,  $\chi_i$   
 $i$ ,  $M_{a,i}$ ,  
 $S$ ,  $S\chi_i$

$$N_{f,i} = \left( S - \sum_{j=1}^r \frac{N_{a,j}}{j} \right). \quad (2.8)$$

$N_{0,i}$

(2.7),

$$\begin{aligned} \frac{dN_{a,i}}{dt} &= k_{a,i} \left( N_{0,i} - N_{a,i} \right) \left( S_i - \sum_{j=1}^r \frac{N_{a,j}}{j} \right) - k_{d,i} N_{a,i} \\ &= k_{a,i} \left( N_{0,i} - N_{a,i} \right) \left( M_{a,i} - \sum_{j=1}^r \frac{1}{j} N_{a,j} \right) - k_{d,i} N_{a,i} \end{aligned} \quad , \quad i = 1, \dots, r, \quad (2.9)$$

$$\begin{aligned} \frac{dN_{a,i}}{dt} &\cong k_{a,i} N_{0,i} \left( M_{a,i} - \sum_{j=1}^r \frac{1}{j} N_{a,j} \right) - k_{d,i} N_{a,i} \\ &\cong k_{l,i} \left( M_{a,i} - \sum_{j=1}^r \frac{1}{j} N_{a,j} \right) - k_{d,i} N_{a,i} \end{aligned} \quad \begin{array}{l} i = 1, \dots, r \\ k_{l,i} = k_{a,i} N_{0,i} \end{array} \quad (2.10)$$

### 2.2.1

Multiphysics),

(COMSOL

...).

,  $n$ ,  $\bar{v}$  :

$$\Phi = \frac{n\bar{v}}{4} \left[ \frac{1}{m^2 s} \right] \quad (2.11)$$

$p_i$

$$n_i = \frac{p_i}{k_B T} \quad (2.12)$$

$k_B$  je

(Boltzmann)  $T$  .

$$\sqrt{8k_B T / f m_i} \quad [52], \quad i-$$

$$\Phi_i = \frac{p_i}{4k_B T} \sqrt{\frac{8k_B T}{f m_i}} = \frac{p_i}{\sqrt{2f m_i k_B T}} \left[ \frac{1}{m^2 s} \right] \quad (2.13)$$

$m_i$

$i$

$$: r_i p_i N_{f,i} / \sqrt{2f m_i k_B T} .$$

$i-$

,  $N_{a,i}$ , a

$\dagger_{r,i}$  .

$i-$

(2.8).



$$\frac{dN_{a,i}}{dt} = \frac{r_i p_i}{\sqrt{2f m_i k_B T}} \left( S - \sum_{j=1}^r \frac{N_{a,j}}{j} \right) - \frac{N_{a,i}}{\tau_{r,i}} \quad (2.14)$$

$i$

$$\frac{dN_{a,i}}{dt} = \frac{r_i}{\sqrt{2f m_i k_B T}} \frac{N_{g,i} k_B T}{V} \left( S - \sum_{j=1}^r \frac{N_{a,j}}{j} \right) - \frac{N_{a,i}}{\tau_{r,i}} \quad (2.15)$$

(2.9),

$i$ -

$$\frac{dN_{a,i}}{dt} = \frac{r_i}{iV} \sqrt{\frac{k_B T}{2f m_i}} (N_{0,i} - N_{a,i}) \left( iS - \sum_{j=1}^r \frac{i}{j} N_{a,j} \right) - \frac{N_{a,i}}{\tau_{r,i}} \quad (2.16)$$

(2.9) (2.16)

$$k_{d,i} = \frac{1}{\tau_{r,i}} \quad (2.17)$$

,  $\tau_{r,i}$ ,

[52]:

$$\tau_{r,i} = \tau_{0,i} \exp(E_{d,i} / RT) \quad (2.18)$$

$T$ ,  $R$

$\tau_{0,i}$

(

10 Hz, [5].

(2.9) (2.16)

$i$ -

:

$$k_{a,i} = \frac{r_i}{V} \sqrt{\frac{k_B T}{2f m_i}} \quad (2.19)$$

$$\left( \chi_i, \chi_c \right) \quad (2.19)$$

$\chi_i$      $\chi_c$ ,

$$k_{l,i} = k_{a,i} N_{0,i} = \frac{r_i}{V} \sqrt{\frac{k_B T}{2f m_i}} \frac{p_i V}{k_B T} = \frac{r_i p_i}{i \sqrt{2f m_i k_B T}} \quad (2.20)$$

### 2.2.2

( )

[PubChem](http://pubchem.ncbi.nlm.nih.gov/)

<http://pubchem.ncbi.nlm.nih.gov/>.

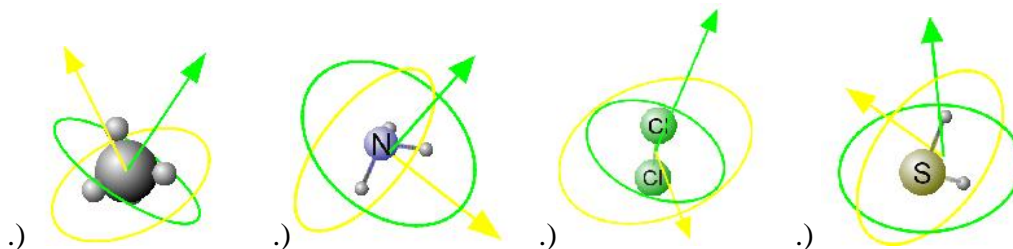
CID (unique chemical structure identifier)

Marvin 5.9.3 2012.

ChemAxon

<http://www.chemaxon.com>.

2.3.



Сл. 2.3 Ефикасни пресек молекула у  $\text{\AA}^2$  за: а.) метан (9.08-12.05), б.) амонијак (8.38-10.87), в.) хлор (9.62-16.70), г.) и водоник-сулфид (10.18-13.36)

( 2.4 )

4/

25

( 2.4 ).



Сл. 2.4 Густина паковања кругова на квадратну/правоугаону површ

0.40786 nm, <100>  
 0.40786 nm      0.2884 nm,  
 4 molecules/nm<sup>2</sup>.  
 [53]      "      10<sup>-5</sup> mol/m<sup>2</sup>      6·10<sup>18</sup>  
 ."      "

[34], [54],

MATLAB

[55], [56].

## 2.3

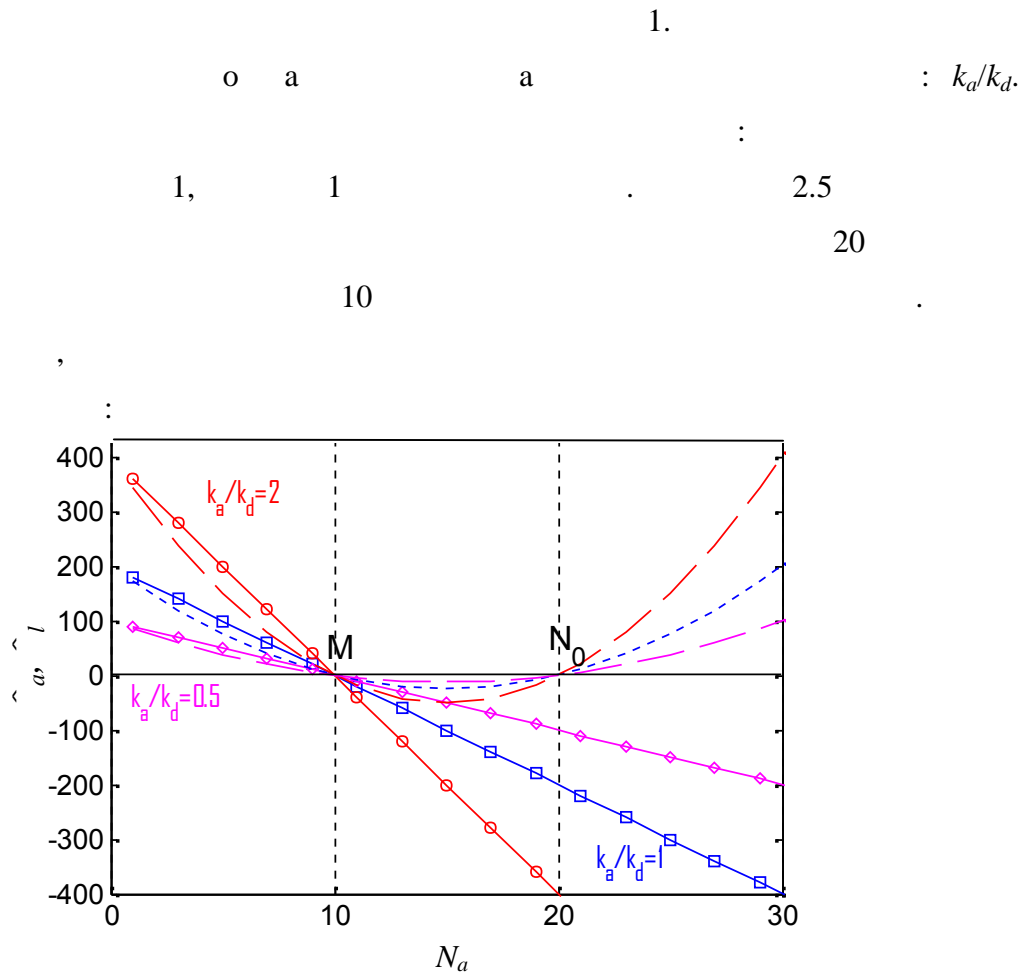
(1.1) — (1.6)

(1.7)

$$\hat{d} = k_d N_a. \tag{2.21}$$

$$\hat{a} = k_a (N_0 - N_a)(M - N_a). \quad (2.22)$$

$$\hat{l} = k_a N_0 (M - N_a). \quad (2.23)$$



Сл. 2.5 Адсорпционе брзине према егзактном моделу (цртане испрекиданим линијама) и према апроксимативном моделу (цртане симболима и пуним линијама) при чему је константа равнотеже: већа од 1 (црвени пар: испрекидана линија и линија са круговима), једнака 1 (плави пар: тачкаста линија и линија са квадратима) и мања од 1 (ружичасти пар: тачка-црта крива и линија са ромбовима)

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 2.5,  
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 0 .  
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 (  $N_0 \gg$  ) ,  
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 0 ,  
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(  
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### 3.2

(1.3) , (Riccati)

(1.4) , :

$$N_a(t) = \frac{N_0 M_a [1 - e^{-k_a(x-s)t}]}{x - S e^{-k_a(x-s)t}} \tag{3.1}$$

(1.3). (1.5) (1.6) :

$$x = \frac{1}{2} \left[ \frac{k_d}{k_a} + N_0 + M_a + \sqrt{\left( \frac{k_d}{k_a} + N_0 + M_a \right)^2 - 4N_0 M_a} \right] \tag{3.2}$$



$$S = \frac{1}{2} \left[ \frac{k_d}{k_a} + N_0 + M_a - \sqrt{\left( \frac{k_d}{k_a} + N_0 + M_a \right)^2 - 4N_0M_a} \right] \quad (3.3)$$

( )

(

),

:

$$N_a(t) = N_{a,\infty} + \sum_m N_{a,t,m} e^{-t/\tau_m} \quad (3.4)$$

(3.4),

$\tau_m$ ,

(3.1).

$$\begin{aligned} k_{II} = k_a(x - S) &= \frac{k_a}{2} \left[ \frac{k_d}{k_a} + N_0 + M_a + \sqrt{\left( \frac{k_d}{k_a} + N_0 + M_a \right)^2 - 4N_0M_a} \right] \\ &\quad - \frac{k_a}{2} \left[ \frac{k_d}{k_a} + N_0 + M_a - \sqrt{\left( \frac{k_d}{k_a} + N_0 + M_a \right)^2 - 4N_0M_a} \right] \quad (3.5) \\ &= k_a \sqrt{\left( \frac{k_d}{k_a} + N_0 + M_a \right)^2 - 4N_0M_a} = \sqrt{(k_d + k_a(N_0 + M_a))^2 - 4k_a^2 N_0 M_a} \end{aligned}$$

:

$$N_a(t) = \frac{xS [1 - e^{-k_{II}t}]}{x - S e^{-k_{II}t}} = S [1 - e^{-k_{II}t}] \frac{1}{1 - \frac{S}{x} e^{-k_{II}t}} \quad (3.6)$$

(3.6)

( 1, x )

$$N_a(t) = S \left[ 1 - e^{-k_{II}t} \right] \sum_{m=0}^{\infty} \left( \frac{S}{X} \right)^m e^{-mk_{II}t} \quad (3.7)$$

(3.4).

$$N_a(t) = S + \sum_{i=1}^{\infty} (S - X) \left( \frac{S}{X} \right)^m e^{-mk_{II}t} \quad (3.8)$$

t , , (X

$$M). \quad (3.8)$$

$$(3.5) \quad (3.8)$$

$$( k_{II} ) \quad (3.5) \quad k_{II}$$

:

$$\sqrt{k_d^2 + (k_a N_0 + k_a M_a)^2 + 2k_d^2 k_d (N_0 + M_a) - 4k_a (k_a N_0 M_a)} = \quad (3.9)$$

$$\sqrt{k_d^2 + k_a^2 (N_0 - M_a)^2 + 2k_d k_a (N_0 + M_a)}$$

, ( )

### 3.3

(Laplace)

[34], [35], [52],

[31]).

[39],

[3],

(2.10)

$$\frac{dN_{a,i}}{dt} = k_{l,i} \left( M_{a,i} - \sum_{j=1}^r \frac{i}{j} N_{a,j} \right) - k_{d,i} N_{a,i} \quad i = 1, \dots, r \quad (3.10)$$

(Laplace).

$$\mathcal{L} \left( \frac{dN_{a,i}}{dt} \right) = k_{l,i} M_{a,i} \mathcal{L}(u) - \sum_{j=1}^r \frac{k_{l,i}}{j} \mathcal{L}(N_{a,j}) - k_{d,i} \mathcal{L}(N_{a,i}) \quad i = 1, \dots, r \quad (3.11)$$

$\mathcal{L}$  (Heaviside) ,  $u = u(t)$  ,  $t < 0$  ( ).

$$\mathcal{L}\left(\frac{dN_{a,i}}{dt}\right) = s\mathcal{N}_{a,i}(s) - N_{a,i}(t_0) \quad \mathcal{L}(N_{a,i}) = \mathcal{N}_{a,i} \quad t_0 = 0, \quad (3.12)$$

$$N_{a,i}(0) = 0 \quad \mathcal{L}\left(\frac{1}{s}\right) = 1/s \quad (3.18)$$

$$s\mathcal{N}_{a,i}(s) = \frac{k_{l,i}M_{a,i}}{s} - \sum_{j=1}^r \frac{k_{l,i}}{j} \mathcal{N}_{a,j}(s) - k_{d,i}\mathcal{N}_{a,i}(s) \quad i = 1, \dots, r \quad (3.13)$$

$$\mathcal{N}_{a,i}(s) = \frac{P_i(s)}{Q_i(s)} = \frac{N_\infty}{s} + \sum_{j=1}^{m_{i,1}} \frac{A_{i,j}}{(s+r_j)^{n+1}} + \sum_{j=1}^{m_{i,2}} \frac{B_{i,j}}{s^2 + s_j^2} + \sum_{j=1}^{m_{i,3}} \frac{C_{i,j}s}{s^2 + x_j^2} + \sum_{j=1}^{m_{i,4}} \frac{D_{i,j}\tilde{S}_j}{(s-u_j)^2 + \tilde{S}_j^2} \dots \quad i = 1, \dots, r \quad (3.14)$$

(3.14)  $r, s, x, u, B, C, D, N_\infty, \tilde{S}$   $i$   $i$   $(r)$   $(s, x)$   $(u)$  ,

$Q_i$

$P_i/Q_i$

$P_i/Q_i$

(3.14)

(3.14) :

$$\begin{aligned}
N_{a,i}(t) = & N_{stac}u(t) + \sum_{j=1}^{m_{i,1}} A_{i,j} \frac{t^n}{n!} e^{-\Gamma t} u(t) + \sum_{j=1}^{m_{i,2}} B_{i,j} \sin(S_j t) u(t) \\
& + \sum_{j=1}^{m_{i,3}} C_{i,j} \cos(X_j t) u(t) + \sum_{j=1}^{m_{i,4}} D_{i,j} \sin(\check{S}t) e^{ut} u(t) \dots
\end{aligned}
\tag{3.15}$$

$i = 1, \dots, r$

$u(t)$  ( , )

(2.5),

$$\frac{dN_a}{dt} = k_a N_0 (M_a - N_a) - k_d N_a = k_l (M_a - N_a) - k_d N_a \tag{3.16}$$

$$s\mathcal{N}_a(s) - N_a(0) = k_l \left( \frac{M_a}{s} - \mathcal{N}_a(s) \right) - k_d \mathcal{N}_a(s) \quad (3.17)$$

$$\mathcal{N}_a(s)(s + k_l + k_d) = \frac{k_l M_a}{s} \quad (3.18)$$

$$\mathcal{N}_a(s) = \frac{k_l M_a}{s(s + k_l + k_d)} \quad (3.19)$$

$$\begin{aligned} N_a(t) &= \mathcal{L}^{-1} \{ \mathcal{N}_a(s) \} = \mathcal{L}^{-1} \left\{ \frac{k_l M_a}{s(s + k_l + k_d)} \right\} = \mathcal{L}^{-1} \left\{ \frac{k_l M_a}{k_l + k_d} \frac{k_l + k_d}{s(s + k_l + k_d)} \right\} \\ &= \frac{k_l M_a}{k_l + k_d} \mathcal{L}^{-1} \left\{ \frac{k_l + k_d}{s(s + k_l + k_d)} \right\} = \frac{k_l M_a}{k_l + k_d} \left( 1 - e^{-(k_l + k_d)t} \right) u(t) \end{aligned} \quad (3.20)$$

### 3.3.1

(3.10)

$$\begin{aligned}
\frac{dN_{a_1}}{dt} &= k_{l_1} (M - N_{a_1} - N_{a_2}) - k_{d_1} N_{a_1} \\
\frac{dN_{a_2}}{dt} &= k_{l_2} (M - N_{a_1} - N_{a_2}) - k_{d_2} N_{a_2}
\end{aligned} \tag{3.21}$$

$$\begin{aligned}
s\mathcal{N}_{a_1}(s) - N_{a_1}(0) &= k_{l_1} \left( \frac{M}{s} - \mathcal{N}_{a_1}(s) - \mathcal{N}_{a_2}(s) \right) - k_{d_1} \mathcal{N}_{a_1}(s) \\
s\mathcal{N}_{a_2}(s) - N_{a_2}(0) &= k_{l_2} \left( \frac{M}{s} - \mathcal{N}_{a_1}(s) - \mathcal{N}_{a_2}(s) \right) - k_{d_2} \mathcal{N}_{a_2}(s)
\end{aligned} \tag{3.22}$$

$$\begin{aligned}
\mathcal{N}_{a_1}(s)(s + k_{l_1} + k_{d_1}) + k_{l_1} \mathcal{N}_{a_2}(s) &= \frac{k_{l_1} M}{s} \\
k_{l_2} \mathcal{N}_{a_1}(s) + \mathcal{N}_{a_2}(s)(s + k_{l_2} + k_{d_2}) &= \frac{k_{l_2} M}{s}
\end{aligned} \tag{3.23}$$

$$\begin{aligned}
\mathcal{N}_{a_1}(s) &= \frac{Mk_{l_1}(s + k_{d_2})}{s \left[ s^2 + s(k_{l_1} + k_{d_1} + k_{l_2} + k_{d_2}) + (k_{l_2}k_{d_1} + k_{l_1}k_{d_2} + k_{d_1}k_{d_2}) \right]} = \frac{Mk_{l_1}(s + k_{d_2})}{s \left[ s^2 + bs + c \right]} \\
\mathcal{N}_{a_2}(s) &= \frac{Mk_{l_2}(s + k_{d_1})}{s \left[ s^2 + s(k_{l_1} + k_{d_1} + k_{l_2} + k_{d_2}) + (k_{l_2}k_{d_1} + k_{l_1}k_{d_2} + k_{d_1}k_{d_2}) \right]} = \frac{Mk_{l_2}(s + k_{d_1})}{s \left[ s^2 + bs + c \right]}
\end{aligned} \tag{3.24}$$

$$b = k_{l_1} + k_{d_1} + k_{l_2} + k_{d_2} \quad c = k_{l_2}k_{d_1} + k_{l_1}k_{d_2} + k_{d_1}k_{d_2}$$



$$b^2-4c$$

$$( \quad )$$

$$(3.23)$$

$$\mathcal{N}_{a_1}(s) = \frac{Mk_{l_1}(s+k_{d_2})}{s[s^2+bs+c]} = Mk_{l_1} \left[ \frac{A_1}{s} + \frac{B_1}{(s+r)} + \frac{C_1}{(s+s)} \right]$$

$$\mathcal{N}_{a_2}(s) = \frac{Mk_{l_2}(s+k_{d_1})}{s[s^2+bs+c]} = Mk_{l_2} \left[ \frac{A_2}{s} + \frac{B_2}{(s+r)} + \frac{C_2}{(s+s)} \right] \quad (3.25)$$

$$\begin{aligned} r &= \frac{1}{2} \left[ -(k_{l_1} + k_{d_1} + k_{l_2} + k_{d_2}) + \sqrt{(k_{l_1} + k_{d_1} + k_{l_2} + k_{d_2})^2 - 4(k_{l_2}k_{d_1} + k_{l_1}k_{d_2} + k_{d_1}k_{d_2})} \right] \\ s &= \frac{1}{2} \left[ -(k_{l_1} + k_{d_1} + k_{l_2} + k_{d_2}) - \sqrt{(k_{l_1} + k_{d_1} + k_{l_2} + k_{d_2})^2 - 4(k_{l_2}k_{d_1} + k_{l_1}k_{d_2} + k_{d_1}k_{d_2})} \right] \end{aligned} \quad (3.26)$$

$$1, B_1, C_1, \quad 2, B_2, C_2$$

$$\begin{aligned}
A_1 &= \frac{k_{d_2}}{c}, & B_1 &= \frac{c+k_{d_2}(r-b)}{c(s-r)}, & C_1 &= \frac{c+k_{d_2}(s-b)}{c(r-s)} \\
A_2 &= \frac{k_{d_1}}{c}, & B_2 &= \frac{c+k_{d_1}(r-b)}{c(s-r)}, & C_2 &= \frac{c+k_{d_1}(s-b)}{c(r-s)}
\end{aligned}
\tag{3.27}$$

(3.25) ( , )

:

$$\begin{aligned}
N_{a_1}(t) &= Mk_{l_1} [A_1 + B_1 e^{-rt} + C_1 e^{-st}] \\
N_{a_2}(t) &= Mk_{l_2} [A_2 + B_2 e^{-rt} + C_2 e^{-st}]
\end{aligned}
\tag{3.28}$$

### 3.3.2

(3.10)

$$\begin{aligned}
 \frac{dN_{a_1}}{dt} &= k_{l_1} (M - N_{a_1} - N_{a_2} - N_{a_3}) - k_{d_1} N_{a_1} \\
 \frac{dN_{a_2}}{dt} &= k_{l_2} (M - N_{a_1} - N_{a_2} - N_{a_3}) - k_{d_2} N_{a_2} \\
 \frac{dN_{a_3}}{dt} &= k_{l_3} (M - N_{a_1} - N_{a_2} - N_{a_3}) - k_{d_3} N_{a_3}
 \end{aligned} \tag{3.29}$$

$$\begin{aligned}
 s\mathcal{N}_{a_1}(s) - N_{a_1}(0) &= k_{l_1} \left( \frac{M}{s} - \mathcal{N}_{a_1}(s) - \mathcal{N}_{a_2}(s) - \mathcal{N}_{a_3}(s) \right) - k_{d_1} \mathcal{N}_{a_1}(s) \\
 s\mathcal{N}_{a_2}(s) - N_{a_2}(0) &= k_{l_2} \left( \frac{M}{s} - \mathcal{N}_{a_1}(s) - \mathcal{N}_{a_2}(s) - \mathcal{N}_{a_3}(s) \right) - k_{d_2} \mathcal{N}_{a_2}(s) \\
 s\mathcal{N}_{a_3}(s) - N_{a_3}(0) &= k_{l_3} \left( \frac{M}{s} - \mathcal{N}_{a_1}(s) - \mathcal{N}_{a_2}(s) - \mathcal{N}_{a_3}(s) \right) - k_{d_3} \mathcal{N}_{a_3}(s)
 \end{aligned} \tag{3.30}$$

$$\begin{aligned}
N_{a_1}(t) &= Mk_{l_1} \left[ A_1 + B_1 e^{-\Gamma t} + C_1 e^{-S t} + D_1 e^{-X t} \right] \\
N_{a_2}(t) &= Mk_{l_2} \left[ A_2 + B_2 e^{-\Gamma t} + C_2 e^{-S t} + D_2 e^{-X t} \right] \\
N_{a_3}(t) &= Mk_{l_3} \left[ A_3 + B_3 e^{-\Gamma t} + C_3 e^{-S t} + D_3 e^{-X t} \right]
\end{aligned} \tag{3.31}$$

$$P_3(s) = s^3 + c_2 s^2 + c_1 s + c_0 = (s - r)(s - s)(s - x).$$

$$\begin{aligned}
c_2 &= k_{l,1} + k_{l,2} + k_{l,3} + k_{d,1} + k_{d,2} + k_{d,3} \\
c_1 &= k_{l,1}(k_{d,2} + k_{d,3}) + k_{l,2}(k_{d,1} + k_{d,3}) + k_{l,3}(k_{d,2} + k_{d,1}) \\
&\quad + k_{d,1}k_{d,2} + k_{d,2}k_{d,3} + k_{d,1}k_{d,3} \\
c_0 &= k_{l,3}k_{d,1}k_{d,2} + k_{l,1}k_{d,3}k_{d,2} + k_{l,2}k_{d,1}k_{d,3} + k_{d,3}k_{d,1}k_{d,2}
\end{aligned} \tag{3.32}$$

$$A_1 = \frac{-k_{d,2}k_{d,3}}{rsx} \quad A_2 = \frac{-k_{d,1}k_{d,3}}{rsx} \quad A_3 = \frac{-k_{d,2}k_{d,1}}{rsx} \tag{3.33}$$

r,

$$B_1 = \frac{(r + k_{d,2})(r + k_{d,3})}{r(r - s)(r - x)} \quad B_2 = \frac{(r + k_{d,1})(r + k_{d,3})}{r(r - s)(r - x)} \quad B_3 = \frac{(r + k_{d,2})(r + k_{d,1})}{r(r - s)(r - x)} \tag{3.34}$$

s,

$$C_1 = \frac{(s + k_{d,2})(s + k_{d,3})}{s(s - r)(s - x)} \quad C_2 = \frac{(s + k_{d,1})(s + k_{d,3})}{s(s - r)(s - x)} \quad C_3 = \frac{(s + k_{d,2})(s + k_{d,1})}{s(s - r)(s - x)} \tag{3.35}$$

x,

$$D_1 = \frac{(x + k_{d,2})(x + k_{d,3})}{x(x-r)(x-s)} \quad D_2 = \frac{(x + k_{d,1})(x + k_{d,3})}{x(x-r)(x-s)} \quad D_3 = \frac{(x + k_{d,2})(x + k_{d,1})}{x(x-r)(x-s)} \quad (3.36)$$

### 3.4

SPP

SPP

SPP

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$n_e$ ,

$n_A$ .

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,  $c$ ,

,  $c_0$ ,

:

$$n = \frac{c_0}{c} = \sqrt{\frac{\sim V}{\sim_0 V_0}} = \sqrt{\sim_r V_r} \tag{3.37}$$

$\mu_r$

$V_r$

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( ),  
 SPP  
 (...)  
 ),

(EMA, Effective Medium Approximation),

[59].

je  $V_{\max}$

$V_{ad}$   
 $N_a$

[60], [61]:

$$n_{eff} = \frac{1}{V_{max}} [(V_{max} - V_{ad})n_e + V_{ad}n_A] = n_e + (n_A - n_e) \frac{V_{ad}}{V_{max}}. \quad (3.38)$$

$$n_{eff} = n_e + (n_A - n_e) \frac{N_a V_{1ad}}{M V_{1ad}} = n_e + \frac{(n_A - n_e)}{M} N_a = n_e + \Delta n_{eff}. \quad (3.39)$$

$n_{eff}$

$$\Delta n_{eff} = n_{eff} - n_e = \frac{(n_A - n_e)}{M} N_a = w N_a. \quad (3.40)$$

RIU,

(refractive index unit).

$$w = \frac{(n_A - n_e)}{M}. \quad (3.41)$$

$$\Delta n_{eff} = \sum_{i=1}^r N_{a,i} \frac{(n_{Ai} - n_e)}{M} = \sum_{i=1}^r w_i N_{a,i} \quad (3.42)$$

$$\Delta n_{eff} = \sum_{i=1}^r N_{a,i} \frac{(n_{Ai} - n_e)}{M_{a,i}} = \sum_{i=1}^r w_i N_{a,i} \quad (3.43)$$



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( (intrinsic)  
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## 4.

### 4.1

([62]–[65],

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) [66]–[72]. M

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[65], [73].

([37], [70]).

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([70], [73]).

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## 4.2

[70],

[73].

([71], [74], [75]).

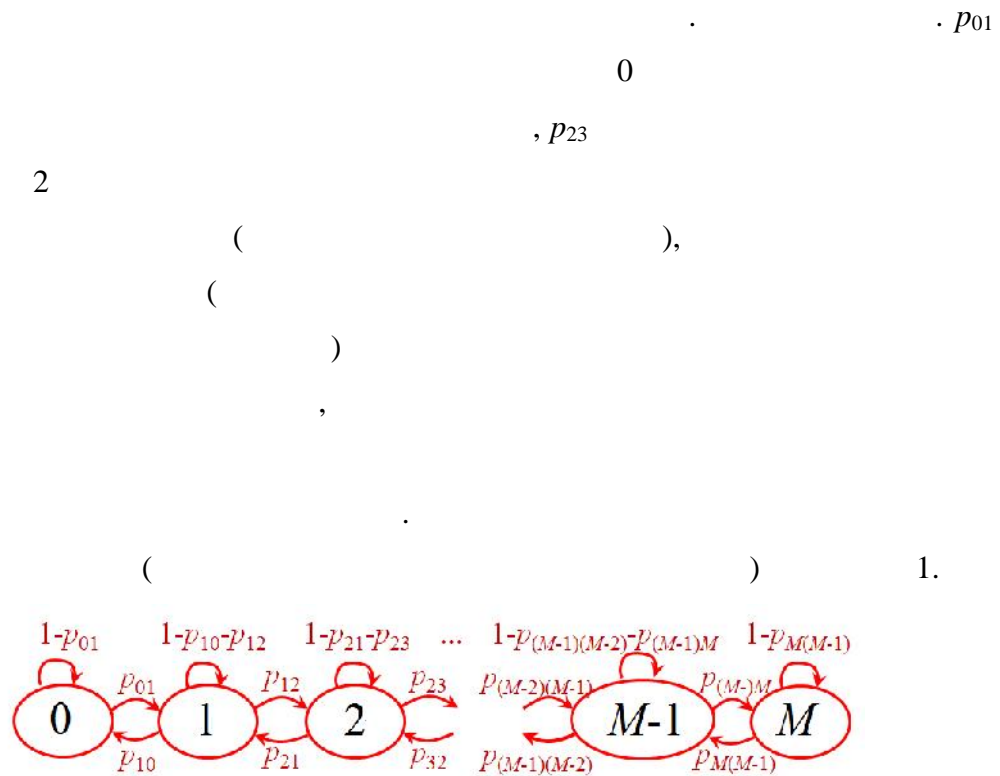
[72].

(0, 1, ... ).

Ma  
(Markov chain).  
{0, ..., M}.

$\Delta t$

$N_a$   
(  
+ 1  
 $N_a$   
 $\Delta t$ ,  
:  
:  
)  
( 4.1).



Сл. 4.1 Дијаграм стања за број адсорбованих честица на површини. Кругови су стања, број у кругу је број адсорбованих молекула тог стања а уз стрелице су назначене вероватноће да систем из стања на почетку стрелице током интервала  $\Delta t$  пређе у стање на крају стрелице.

( [67], [73] )

). [37]

$P_{N_a}(t + \Delta t)$

$t + \Delta t$

$N_a$

$\Delta t$

$\Delta t$

:

$$P_{N_a}(t + \Delta t) = P_{N_a-1}(t)p(N_a - 1 \rightarrow N_a) + P_{N_a+1}(t)p(N_a + 1 \rightarrow N_a) + P_{N_a}(t)p(N_a \rightarrow N_a) \quad (4.1)$$

$$P_{N_a-1}(t) \quad t \quad N_a - 1, \\ P_{N_a+1}(t) \quad t \quad N_a + 1.$$

:

$$P_{N_a}(t + \Delta t) = P_{N_a-1}(t)p(N_a - 1 \rightarrow N_a) + P_{N_a+1}(t)p(N_a + 1 \rightarrow N_a) + P_{N_a}(t)[1 - p(N_a \rightarrow N_a - 1) - p(N_a \rightarrow N_a + 1)] \quad (4.2)$$

,

$$W, \quad \Delta t. \quad :$$

$$p(N_a - 1 \rightarrow N_a) = W(N_a - 1 \rightarrow N_a)\Delta t \quad (4.3)$$

(4.2)

$$\frac{P_{N_a}(t + \Delta t) - P_{N_a}(t)}{\Delta t} = P_{N_a-1}(t)W(N_a - 1 \rightarrow N_a) + P_{N_a+1}(t)W(N_a + 1 \rightarrow N_a) - P_{N_a}(t)[W(N_a \rightarrow N_a - 1) + W(N_a \rightarrow N_a + 1)] \quad (4.4)$$

:

$$\frac{dP_{N_a}(t)}{dt} = P_{N_a-1}(t)W(N_a - 1 \rightarrow N_a) + P_{N_a+1}(t)W(N_a + 1 \rightarrow N_a) - P_{N_a}(t)[W(N_a \rightarrow N_a - 1) + W(N_a \rightarrow N_a + 1)] \quad (4.5)$$

$N_a$

$N_a$ .

(

појављују под називом линеарни односно нелинеарни модел), те ће и стохастичка анализа касније бити дата одвојено. За решавање главне једначине, овде је то једначина (4.5), у оба случаја се користи генеришућа функција вероватноћа.

Генеришућа функција вероватноћа (у наставку – ГФВ) је дефинисана за дискретне случајне променљиве као бесконачан степени ред ([37], [63], [65], [67]), у извођењима датим касније користимо тај израз:

$$F(s, t) = \sum_{N_a=0}^{\infty} P_{N_a}(t) s^{N_a} = P_0(t) + P_1(t)s + P_2(t)s^2 + P_3(t)s^3 + \dots \quad |s| \leq 1 \quad (4.6)$$

Променљива  $s$  је бездимензиона величина која служи да обезбеди конвергенцију (зато је неопходно ограничење по апсолутној вредности). На основу генеришуће функције вероватноћа могу се добити све вероватноће у сваком тренутку као и сви моменти посматране стохастичке променљиве. Нпр. први извод генеришуће функције по променљивој  $s$  је

$$\frac{\partial F(s, t)}{\partial s} = P_1(t) + 2sP_2(t) + 3P_3(t)s^2 + \dots = \sum_{N_a=1}^{\infty} N_a P_{N_a}(t) s^{N_a-1} \quad |s| \leq 1 \quad (4.7)$$

У извођењима која следе појављиваће се суме у којима фигуришу  $P_{N_a-1}(t)$  и  $P_{N_a+1}(t)$ , у којима такође можемо препознати парцијални извод ГФВ за случајну променљиву  $N_a$ , јер важи:

$$\begin{aligned} \frac{\partial F(s, t)}{\partial s} &= \sum_{N_a=0}^{\infty} N_a P_{N_a}(t) s^{N_a-1} = \sum_{N_a=0}^{\infty} (N_a - 1) P_{N_a-1}(t) s^{N_a-2} \\ &= \sum_{N_a=0}^{\infty} (N_a + 1) P_{N_a+1}(t) s^{N_a} \quad |s| \leq 1 \end{aligned} \quad (4.7a)$$

У случају када у суми фигурише  $P_{N_a-1}(t)$ , подразумевамо да је вероватноћа  $P_{-1}(t)$  у првом члану  $-P_{-1}(t)/s^2$  једнака нули јер је то вероватноћа да систем буде у непостојећем стању сви остали чланови се уклапају у развој из израза (4.7).

Први извод генеришуће функције по променљивој  $s$  у тачки  $s = 1$  представља средњу тј. очекивану вредност случајне променљиве у времену



$$\overline{N_a} = E\{N_a(t)\} = \left. \frac{\partial F(s,t)}{\partial s} \right|_{s=1} \quad (4.8)$$

s

$$\frac{\partial^2 F(s,t)}{\partial s^2} = \sum_{N_a=0}^{\infty} N_a(N_a-1)P_{N_a}(t)s^{N_a-2} \quad |s| \leq 1 \quad (4.9)$$

= 1,

$$\left. \frac{\partial^2 F(s,t)}{\partial s^2} \right|_{s=1} = \sum_{N_a=0}^{\infty} N_a^2 P_{N_a}(t) - \sum_{N_a=0}^{\infty} N_a P_{N_a}(t) = \overline{N_a^2} - \overline{N_a} \quad (4.9)$$

:

$$D\{N_a(t)\} = \overline{N_a^2} - \overline{N_a}^2 = \left. \frac{\partial^2 F(s,t)}{\partial s^2} \right|_{s=1} + \left. \frac{\partial F(s,t)}{\partial s} \right|_{s=1} - \left( \left. \frac{\partial F(s,t)}{\partial s} \right|_{s=1} \right)^2 \quad (4.10)$$

:

$$u\{N_a(t)\} = \sqrt{\frac{D\{N_a(t)\}}{E\{N_a(t)\}^2}} \quad (4.11)$$

(

)

$$P_{N_{a1}, N_{a2}}(t)$$

$N_{a1}$

$N_{a2}$

:

$$F(s_1, s_2, t) = \sum_{N_{a1}=0}^{\infty} \sum_{N_{a2}=0}^{\infty} P_{N_{a1}, N_{a2}}(t) s_1^{N_{a1}} s_2^{N_{a2}} \quad |s_1|, |s_2| \leq 1 \quad (4.12)$$

$$\frac{\partial F(s_1, s_2, t)}{\partial s_1} = \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M N_{a1} P_{N_{a1}, N_{a2}}(t) s_1^{N_{a1}-1} s_2^{N_{a2}} \Big|_{s_1, s_2=1} = \overline{N_{a1}} \quad (4.13)$$

$$\frac{\partial F(s_1, s_2, t)}{\partial s_2} = \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M N_{a2} P_{N_{a1}, N_{a2}}(t) s_1^{N_{a1}} s_2^{N_{a2}-1} \Big|_{s_1, s_2=1} = \overline{N_{a2}} \quad (4.14)$$

$$\frac{\partial^2 F(s_1, s_2, t)}{\partial s_1 \partial s_2} = \frac{\partial^2 F(s_1, s_2, t)}{\partial s_2 \partial s_1} = \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M N_{a1} N_{a2} P_{N_{a1}, N_{a2}}(t) s_1^{N_{a1}-1} s_2^{N_{a2}-1} \Big|_{s_1, s_2=1} = \overline{N_{a1} N_{a2}} \neq \overline{N_{a1}} \cdot \overline{N_{a2}} \quad (4.15)$$

(4.5),

$$P_{N_a}(t), P_{N_a-1}(t), P_{N_a+1}(t)$$

### 4.3.

[70])

([37],

$$P(N_g, N_a, t) = \dots$$

(4.2),

( $N_g, N_a$ )

$$P_{N_g, N_a}(t + \Delta t) = P_{N_g+1, N_a-1}(t) p(N_a - 1 \rightarrow N_a) + P_{N_g-1, N_a+1}(t) p(N_a + 1 \rightarrow N_a) + P_{N_g, N_a}(t) [1 - p(N_a \rightarrow N_a - 1) - p(N_a \rightarrow N_a + 1)] \quad (4.16)$$

$$p(N_a - 1 \rightarrow N_a) = k_a (N_g + 1) [M - (N_a - 1)] \Delta t$$

$$p(N_a + 1 \rightarrow N_a) = k_d (N_a + 1) \Delta t$$

$$p(N_a \rightarrow N_a + 1) = k_a N_g (M - N_a) \Delta t$$

$$p(N_a \rightarrow N_a - 1) = k_d N_a \Delta t$$

(4.17)

$k_a$

$k_d$

$$P(N_g, N_a, t + \Delta t)$$

(4.17)

$$\frac{P_{N_g, N_a}(t + \Delta t) - P_{N_g, N_a}(t)}{\Delta t} = P_{N_g+1, N_a-1}(t) k_a (N_g + 1) [M - (N_a - 1)] + P_{N_g-1, N_a+1}(t) k_d (N_a + 1) - P_{N_g, N_a}(t) [k_a N_g (M - N_a) + k_d N_a] \quad (4.18)$$

$$\frac{dP_{N_g, N_a}(t)}{dt} = P_{N_g+1, N_a-1}(t)k_a(N_g+1)[M - (N_a - 1)] \quad (4.19)$$

$$+ P_{N_g-1, N_a+1}(t)k_d(N_a+1) - P_{N_g, N_a}(t)[k_a N_g(M - N_a) + k_d N_a]$$

(4.12),

$$F(s_g, s_a, t) = \sum_{N_g=N_0}^{N_0-M} \sum_{N_a=0}^M P_{N_g, N_a}(t) s_g^{N_g} s_a^{N_a} \quad |s_g|, |s_a| \leq 1 \quad (4.20)$$

$$(4.19) \quad s_g^{N_g} s_a^{N_a} \quad N_g \quad N_a$$

$$\begin{aligned} \frac{d}{dt} \sum_{N_g=N_0}^{N_0-M} \sum_{N_a=0}^M P_{N_g, N_a}(t) s_g^{N_g} s_a^{N_a} = \\ k_a \sum_{N_g=N_0}^{N_0-M} \sum_{N_a=0}^M (N_g+1)[M - (N_a - 1)] P_{N_g+1, N_a-1}(t) s_g^{N_g} s_a^{N_a} \end{aligned} \quad (4.21)$$

$$+ k_d \sum_{N_g=N_0}^{N_0-M} \sum_{N_a=0}^M (N_a+1) P_{N_g-1, N_a+1}(t) s_g^{N_g} s_a^{N_a}$$

$$- \sum_{N_g=N_0}^{N_0-M} \sum_{N_a=0}^M [k_a N_g(M - N_a) + k_d N_a] P_{N_g, N_a}(t) s_g^{N_g} s_a^{N_a}$$

(4.13) – (4.15),

(4.21)

:

$$\frac{d}{dt} \sum_{N_g=N_0}^{N_0-M} \sum_{N_a=0}^M P_{N_g, N_a}(t) s_g^{N_g} s_a^{N_a} = k_a M s_a \sum_{N_g=N_0}^{N_0-M} \sum_{N_a=0}^M (N_g+1) P_{N_g+1, N_a-1}(t) s_g^{N_g} s_a^{N_a-1}$$

$$- k_a s_a^2 \sum_{N_g=N_0}^{N_0-M} \sum_{N_a=0}^M (N_g+1)(N_a-1) P_{N_g+1, N_a-1}(t) s_g^{N_g} s_a^{N_a-2}$$

$$+ k_d s_g \sum_{N_g=N_0}^{N_0-M} \sum_{N_a=0}^M (N_a+1) P_{N_g-1, N_a+1}(t) s_g^{N_g} s_a^{N_a} - k_a M s_g \sum_{N_g=N_0}^{N_0-M} \sum_{N_a=0}^M N_g P_{N_g, N_a}(t) s_g^{N_g-1} s_a^{N_a}$$

$$+k_a s_g s_a \sum_{N_g=N_0}^{N_0-M} \sum_{N_a=0}^M N_g N_a P_{N_g, N_a}(t) s_g^{N_g-1} s_a^{N_a-1} - k_d s_a \sum_{N_g=N_0}^{N_0-M} \sum_{N_a=0}^M N_a P_{N_g, N_a}(t) s_g^{N_g} s_a^{N_a-1}$$

(4.22)

(4.22)

( )

$$\frac{dF}{dt} = k_a M s_a \frac{\partial F}{\partial s_g} - k_a s_a^2 \frac{\partial^2 F}{\partial s_g \partial s_a} + k_d s_g \frac{\partial F}{\partial s_a} - k_a M s_g \frac{\partial F}{\partial s_g} + k_a s_g s_a \frac{\partial^2 F}{\partial s_g \partial s_a} - k_d s_a \frac{\partial F}{\partial s_a} \quad (4.23)$$

$$\frac{\partial F}{\partial t} = (s_a - s_g) \left[ k_a M \frac{\partial F}{\partial s_g} - k_a s_a \frac{\partial^2 F}{\partial s_g \partial s_a} - k_d \frac{\partial F}{\partial s_a} \right] \quad (4.24)$$

$$\frac{\partial}{\partial t} \frac{\partial F}{\partial s_a} =$$

$$(s_a - s_g) \frac{\partial}{\partial s_a} \left[ k_a M \frac{\partial F}{\partial s_g} - k_a s_a \frac{\partial^2 F}{\partial s_g \partial s_a} - k_d \frac{\partial F}{\partial s_a} \right] + \left[ k_a M \frac{\partial F}{\partial s_g} - k_a s_a \frac{\partial^2 F}{\partial s_g \partial s_a} - k_d \frac{\partial F}{\partial s_a} \right] \quad (4.25)$$

$$s_g = 1:$$

$$\left. \frac{\partial}{\partial t} \frac{\partial F}{\partial s_a} \right|_{s_a=s_g=1} = k_a M \left. \frac{\partial F}{\partial s_g} \right|_{s_a=s_g=1} - k_a s_a \left. \frac{\partial^2 F}{\partial s_g \partial s_a} \right|_{s_a=s_g=1} - k_d \left. \frac{\partial F}{\partial s_a} \right|_{s_a=s_g=1} \quad (4.25)$$

$$\frac{d\overline{N}_a}{dt} = k_a M \overline{N}_g - k_a \overline{N}_g \overline{N}_a - k_d \overline{N}_a \quad (4.26)$$

,  $N_g$   $N_a$

$$P_{N_g, N_a}(t) \approx P_{N_g}(t) P_{N_a}(t) \quad (4.27)$$

$$\overline{N_g N_a} = \overline{N_g} \cdot \overline{N_a} \quad (4.27)$$

(4.26)

$$\frac{d\overline{N}_a}{dt} = k_a \overline{N}_g (M - \overline{N}_a) - k_d \overline{N}_a \quad (4.28)$$

) : (4.24)

$$s_g = = 1,$$

$$\frac{\partial^2 F(s_a, s_g, t)}{\partial s_a^2} = \sum_{N_g=0}^{M-N_{a1}} \sum_{N_a=0}^M N_a (N_a - 1) P_{N_{a1}, N_{a2}}(t) s_a^{N_a-2} s_g^{N_g} \Big|_{s_a, s_g=1} = \overline{N_a^2} - \overline{N_a} \quad (4.29)$$

(4.24)

$s_a,$

$$\frac{d\overline{N_a^2}}{dt} + 2[k_d + k_a(N_0 - \overline{N_a})]\overline{N_a^2} = k_a MN_0 - [k_d + k_a(2MN_0 - N_0 - M)]\overline{N_a} - k_a(2M - 1)\overline{N_a^2} \quad (4.30)$$

$\overline{N_a}$  je

(3.9)-(3.11)

$$N_a(t) = \frac{N_0 M [1 - e^{-k_a(x-s)t}]}{x - s e^{-k_a(x-s)t}} \quad (4.31)$$

:  $k_a$

$$x = \frac{1}{2} \left[ \frac{k_d}{k_a} + N_0 + M + \sqrt{\left( \frac{k_d}{k_a} + N_0 + M \right)^2 - 4N_0 M} \right] \quad (4.32)$$

$$s = \frac{1}{2} \left[ \frac{k_d}{k_a} + N_0 + M - \sqrt{\left( \frac{k_d}{k_a} + N_0 + M \right)^2 - 4N_0 M} \right] \quad (4.33)$$

$$D\{N_a\} = \overline{N_a^2} - \overline{N_a}^2 = \frac{N_0 M [1 - e^{-k_a(x-s)t}]}{x - s e^{-k_a(x-s)t}} \left( 1 - \frac{N_0 [1 - e^{-k_a(x-s)t}]}{x - s e^{-k_a(x-s)t}} \right) \quad (4.34)$$

$$D\{N_a\} = \overline{N_a^2} - \overline{N_a}^2 = \overline{N_a} \left( 1 - \frac{\overline{N_a}}{M} \right) \quad (4.35)$$

$$u\{N_a\} = \sqrt{\frac{D\{N_a\}}{\overline{N_a^2}}} = \sqrt{\left( \frac{1}{\overline{N_a}} - \frac{1}{M} \right)} \quad (4.36)$$

( )

## 4.4

### 4.4.1

$$p(N_a - 1 \rightarrow N_a) = k_l (M - (N_a - 1)) \Delta t \quad (4.37)$$

$$p(N_a + 1 \rightarrow N_a) = k_d (N_a + 1) \Delta t \quad (4.38)$$

$$p(N_a \rightarrow N_a) = 1 - k_l (M - N_a) \Delta t - k_d N_a \Delta t \quad (4.39)$$

$k_l$  ,  $k_d$  ,  $\Delta t$

$$\begin{aligned} \frac{dP_{N_a}(t)}{dt} = & P_{N_a-1}(t) k_l [M - (N_a - 1)] \\ & + P_{N_a+1}(t) k_d (N_a + 1) \\ & - P_{N_a}(t) [k_l (M - N_a) + k_d N_a] \end{aligned} \quad (4.40)$$

$P_{N_a}(t)$ ,  $P_{N_a-1}(t)$   $P_{N_a+1}$

$$(4.40) \quad s^{N_a}$$

$N_a$  0

$$\sum_{N_a=0}^{\infty} s^{N_a} \frac{dP_{N_a}(t)}{dt} = \sum_{N_a=0}^{\infty} s^{N_a} P_{N_a-1}(t) k_l [M - (N_a - 1)]$$



$$+ \sum_{N_a=0}^{\infty} s^{N_a} P_{N_a+1}(t) k_d (N_a + 1) - \sum_{N_a=0}^{\infty} s^{N_a} P_{N_a}(t) [k_l (M - N_a) + k_d N_a] \quad (4.41)$$

$$\begin{aligned} \frac{d \left( \sum_{N_a=0}^{\infty} s^{N_a} P_{N_a}(t) \right)}{dt} &= M k_l s \sum_{N_a=0}^{\infty} s^{N_a-1} P_{N_a-1}(t) - k_l s^2 \sum_{N_a=0}^{\infty} s^{N_a-2} P_{N_a-1}(t) (N_a - 1) \\ &+ k_d \sum_{N_a=0}^{\infty} s^{N_a} P_{N_a+1}(t) (N_a + 1) \\ &- k_l M \sum_{N_a=0}^{\infty} s^{N_a} P_{N_a}(t) + (k_l - k_d) s \sum_{N_a=0}^{\infty} s^{N_a-1} P_{N_a}(t) N_a \end{aligned} \quad (4.42)$$

(0, 1, ... ) ,

$$\begin{aligned} \frac{dF(s,t)}{dt} &= M k_l s F(s,t) - k_l s^2 \frac{\partial F(s,t)}{\partial s} + k_d \frac{\partial F(s,t)}{\partial s} \\ &- k_l M F(s,t) + (k_l - k_d) s \frac{\partial F(s,t)}{\partial s} \end{aligned} \quad (4.43)$$

$$\frac{\partial F(s,t)}{\partial t} = M k_l (s-1) F(s,t) - (s-1) (k_d + k_l s) \frac{\partial F(s,t)}{\partial s} \quad (4.44)$$

(4.43) o (4.44) :

$$F(1,t) = 1 \quad F(s,0) = 1 \quad (4.45)$$

(4.44)

(4.45)

— , , ...  
∞,

(4.6),

$$N_a \quad (0, \dots)$$

$$F(s, t) = P^M = [1 + Q]^M = [1 + (s-1)G]^M \quad (4.46)$$

$P \quad Q$

$G$

(4.44)

(4.46)

$G$

(4.44),

$$\frac{\partial F(s, t)}{\partial t} = MP^{M-1}(s-1) \frac{\partial G}{\partial t} \quad (4.47)$$

$$\frac{\partial F}{\partial s} = MP^{M-1} \left[ \frac{\partial G}{\partial s} (s-1) + G \right] \quad (4.48)$$

(4.46),

(4.48),

(4.47)

(4.44),

a

o

(-1) :

$$\cancel{M} P^{M-1} \cancel{(s-1)} \frac{\partial G}{\partial t} = k_l \cancel{M} \cancel{(s-1)} P^M - \cancel{(s-1)} (k_d + k_l s) \cancel{M} P^{M-1} \left[ \frac{\partial G}{\partial s} (s-1) + G \right] \quad (4.49)$$

a o

a

$P^{M-1}$

:

$$\frac{\partial G}{\partial t} = k_l P - (k_d + k_l s) \left[ \frac{\partial G}{\partial s} (s-1) + G \right] \quad (4.50)$$

$P,$

(4.50)

$$\frac{\partial G}{\partial t} = k_l [1 + (s-1)G] - (k_d + k_l s) \left[ \frac{\partial G}{\partial s} (s-1) + G \right] \quad (4.51)$$

,

$G$

$$\frac{\partial G}{\partial t} = k_l [1 + (s-1)G] - (k_d + k_l s) G \quad (4.52)$$

$$\frac{\partial G}{\partial t} = k_l(1-G) - k_d G \quad (4.53)$$

$$G \quad (4.53)$$

:

$$\frac{dN_a}{dt} = k_l(M - N_a) - k_d N_a \quad (4.54)$$

$$N_a(t) = \frac{k_l M}{k_l + k_d} \left[ 1 - e^{-(k_l + k_d)t} \right] \quad (4.55)$$

$$(4.55) \quad G \quad :$$

$$G(t) = \frac{k_l}{k_l + k_d} \left[ 1 - e^{-(k_l + k_d)t} \right] \quad (4.56)$$

$$(4.56)$$

$$(4.46),$$

$$(4.44)$$

$$(4.45).$$

$$F(s,t) = \left[ 1 + (s-1) \frac{k_l}{k_l + k_d} \left[ 1 - e^{-(k_l + k_d)t} \right] \right]^M \quad (4.57)$$

$$, \quad , \quad . \quad b \quad -$$

[76]:

$$[a+b]^M = \sum_{N_a=0}^M \binom{M}{N_a} a^{M-N_a} b^{N_a} \quad (4.58)$$

$$(4.57)$$

$$(4.6),$$

$$F(s,t) = \left[ \frac{k_l e^{-(k_l + k_d)t} + k_d}{k_l + k_d} + s \frac{k_l (1 - e^{-(k_l + k_d)t})}{k_l + k_d} \right]^M =$$

$$\sum_{N_a=0}^M \binom{M}{N_a} \left( \frac{k_l e^{-(k_a+k_d)t} + k_d}{k_l + k_d} \right)^{M-N_a} \left( \frac{k_l (1 - e^{-(k_l+k_d)t})}{k_l + k_d} \right)^{N_a} s^{N_a} \quad (4.59)$$

(4.6) (4.59)

$N_a$

:

$$P_{N_a}(t) = \binom{M}{N_a} \left( \frac{k_l e^{-(k_a+k_d)t} + k_d}{k_l + k_d} \right)^{M-N_a} \left( \frac{k_l (1 - e^{-(k_l+k_d)t})}{k_l + k_d} \right)^{N_a} \quad (4.60)$$

(4.59)

(4.8)

$$\overline{N_a} = \frac{k_l M}{k_l + k_d} (1 - e^{-(k_l+k_d)t}) \quad (4.61)$$

( ) (4.10)

(4.11):

$$D\{N_a(t)\} = \frac{M k_l (1 - e^{-(k_l+k_d)t}) (k_d + k_l e^{-(k_l+k_d)t})}{(k_l + k_d)^2} = \overline{N_a} \left( 1 - \frac{\overline{N_a}}{M} \right) \quad (4.62)$$

$$u\{N_a(t)\} = \sqrt{\frac{k_d + k_l e^{-(k_l+k_d)t}}{k_l M (1 - e^{-(k_l+k_d)t})}} = \sqrt{\frac{1}{\overline{N_a}} - \frac{1}{M}} \quad (4.63)$$

(

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(4.46)

$$F(s, t) = \left[ 1 + \frac{k_l}{k_l + k_d} (1 - e^{-(k_l+k_d)t}) (s-1) \right]^M = \left[ 1 + \frac{\overline{N_a}}{M} (s-1) \right]^M \quad (4.64)$$

4.4.2

$$\begin{aligned}
 P_{N_{a1}, N_{a2}}(t + \Delta t) &= P_{N_{a1}-1, N_{a2}}(t) p((N_{a1}-1) \rightarrow N_{a1}) \\
 &\quad + P_{N_{a1}, N_{a2}-1}(t) p((N_{a2}-1) \rightarrow N_{a2}) \\
 &\quad + P_{N_{a1}+1, N_{a2}}(t) p((N_{a1}+1) \rightarrow N_{a1}) \\
 &\quad + P_{N_{a1}, N_{a2}+1}(t) p((N_{a2}+1) \rightarrow N_{a2}) \\
 &+ P_{N_{a1}, N_{a2}}(t) \left[ 1 - p(N_{a1} \rightarrow (N_{a1}-1)) - p(N_{a1} \rightarrow (N_{a1}+1)) \right. \\
 &\quad \left. - p(N_{a2} \rightarrow (N_{a2}-1)) - p(N_{a2} \rightarrow (N_{a2}+1)) \right] \tag{4.65}
 \end{aligned}$$

$$\begin{aligned}
p((N_{a1}-1) \rightarrow N_{a1}) &= k_{l1}(M - (N_{a1}-1) - N_{a2})\Delta t & p((N_{a1}+1) \rightarrow N_{a1}) &= k_{d1}(N_{a1}+1)\Delta t \\
p((N_{a2}-1) \rightarrow N_{a2}) &= k_{l2}(M - N_{a1} - (N_{a2}-1))\Delta t & p((N_{a2}+1) \rightarrow N_{a2}) &= k_{d2}(N_{a2}+1)\Delta t \\
p(N_{a1} \rightarrow (N_{a1}+1)) &= k_{l1}(M - N_{a1} - N_{a2})\Delta t & p(N_{a1} \rightarrow (N_{a1}-1)) &= k_{d1}N_{a1}\Delta t \\
p(N_{a2} \rightarrow (N_{a2}+1)) &= k_{l2}(M - N_{a1} - N_{a2})\Delta t & p(N_{a2} \rightarrow (N_{a2}-1)) &= k_{d2}N_{a2}\Delta t
\end{aligned}
\tag{4.66}$$

$$\begin{aligned}
\frac{dP_{N_{a1},N_{a2}}(t)}{dt} &= P_{N_{a1}-1,N_{a2}}(t)k_{l1}(M - (N_{a1}-1) - N_{a2}) \\
&\quad + P_{N_{a1},N_{a2}-1}(t)k_{l2}(M - (N_{a1}-1) - N_{a2}) \\
&\quad + P_{N_{a1}+1,N_{a2}}(t)k_{d1}(N_{a1}+1) + P_{N_{a1},N_{a2}+1}(t)k_{d2}(N_{a2}+1) \\
&\quad - P_{N_{a1},N_{a2}}(t)(k_{d1}N_{a1} + k_{l1}(M - N_{a1} - N_{a2}) + k_{d2}N_{a2} + k_{l2}(M - N_{a1} - N_{a2}))
\end{aligned}
\tag{4.67}$$

$N_{a1}, \dots, 0$  :

$$\begin{aligned}
\frac{d \sum_{N_{a1}=0}^M s_1^{N_{a1}} P_{N_{a1},N_{a2}}(t)}{dt} &= \sum_{N_{a1}=0}^M s_1^{N_{a1}} P_{N_{a1}-1,N_{a2}}(t)k_{l1}(M - (N_{a1}-1) - N_{a2}) \\
&\quad + \sum_{N_{a1}=0}^M s_1^{N_{a1}} P_{N_{a1},N_{a2}-1}(t)k_{l2}(M - (N_{a1}-1) - N_{a2}) \\
&\quad + \sum_{N_{a1}=0}^M s_1^{N_{a1}} P_{N_{a1}+1,N_{a2}}(t)k_{d1}(N_{a1}+1) + \sum_{N_{a1}=0}^M s_1^{N_{a1}} P_{N_{a1},N_{a2}+1}(t)k_{d2}(N_{a2}+1) \\
&\quad - \sum_{N_{a1}=0}^M s_1^{N_{a1}} P_{N_{a1},N_{a2}}(t)(k_{d1}N_{a1} + k_{l1}(M - N_{a1} - N_{a2}) + k_{d2}N_{a2} + k_{l2}(M - N_{a1} - N_{a2}))
\end{aligned}
\tag{4.68}$$

$0 \quad M - N_{a1}$

$$\frac{d \left[ \sum_{N_{a2}=0}^{M-N_{a1}} s_2^{N_{a2}} \sum_{N_{a1}=0}^M s_1^{N_{a1}} P_{N_{a1},N_{a2}}(t) \right]}{dt} =$$

$$\begin{aligned}
& \sum_{N_{a2}=0}^{M-N_{a1}} s_2^{N_{a2}} \sum_{N_{a1}=0}^M s_1^{N_{a1}} P_{N_{a1}-1, N_{a2}}(t) k_{l1} (M - (N_{a1} - 1) - N_{a2}) \\
& + \sum_{N_{a2}=0}^{M-N_{a1}} s_2^{N_{a2}} \sum_{N_{a1}=0}^M s_1^{N_{a1}} P_{N_{a1}, N_{a2}-1}(t) k_{l2} (M - (N_{a1} - 1) - N_{a2}) \\
& + \sum_{N_{a2}=0}^{M-N_{a1}} s_2^{N_{a2}} \sum_{N_{a1}=0}^M s_1^{N_{a1}} P_{N_{a1}+1, N_{a2}}(t) k_{d1} (N_{a1} + 1) \\
& + \sum_{N_{a2}=0}^{M-N_{a1}} s_2^{N_{a2}} \sum_{N_{a1}=0}^M s_1^{N_{a1}} P_{N_{a1}, N_{a2}+1}(t) k_{d2} (N_{a2} + 1) - \\
& \sum_{N_{a2}=0}^{M-N_{a1}} s_2^{N_{a2}} \sum_{N_{a1}=0}^M s_1^{N_{a1}} P_{N_{a1}, N_{a2}}(t) (k_{d1} N_{a1} + k_{l1} (M - N_{a1} - N_{a2}) + k_{d2} N_{a2} \\
& \quad + k_{l2} (M - N_{a1} - N_{a2}))
\end{aligned} \tag{4.69}$$

(4.12) – (4.15).

$$\begin{aligned}
\frac{dF(s_1, s_2, t)}{dt} &= k_{l1} M \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M P_{N_{a1}-1, N_{a2}}(t) s_1^{N_{a1}} s_2^{N_{a2}} \\
& - k_{l1} \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M (N_{a1} - 1) P_{N_{a1}-1, N_{a2}}(t) s_1^{N_{a1}} s_2^{N_{a2}} - k_{l1} \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M N_{a2} P_{N_{a1}-1, N_{a2}}(t) s_1^{N_{a1}} s_2^{N_{a2}} \\
& + k_{l2} M \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M P_{N_{a1}, N_{a2}-1}(t) s_1^{N_{a1}} s_2^{N_{a2}} - k_{l2} \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M N_{a1} P_{N_{a1}, N_{a2}-1}(t) s_1^{N_{a1}} s_2^{N_{a2}} \\
& - k_{l2} \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M (N_{a2} - 1) P_{N_{a1}, N_{a2}-1}(t) s_1^{N_{a1}} s_2^{N_{a2}} \\
& + k_{d1} \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M (N_{a1} + 1) P_{N_{a1}+1, N_{a2}}(t) s_1^{N_{a1}} s_2^{N_{a2}} \\
& + k_{d2} \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M (N_{a2} + 1) P_{N_{a1}, N_{a2}+1}(t) s_1^{N_{a1}} s_2^{N_{a2}} \\
& + (k_{l1} + k_{l2} - k_{d1}) \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M N_{a1} P_{N_{a1}, N_{a2}}(t) s_1^{N_{a1}} s_2^{N_{a2}}
\end{aligned}$$

$$\begin{aligned}
& + (k_{l1} + k_{l2} - k_{d2}) \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M N_{a2} P_{N_{a1}, N_{a2}}(t) s_1^{N_{a1}} s_2^{N_{a2}} \\
& - (k_{l1} + k_{l2}) M \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M P_{N_{a1}, N_{a2}}(t) s_1^{N_{a1}} s_2^{N_{a2}}
\end{aligned} \tag{4.70}$$

$$\begin{aligned}
\frac{dF(s_1, s_2, t)}{dt} &= k_{l1} M s_{a1} \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M P_{N_{a1}-1, N_{a2}}(t) s_1^{N_{a1}-1} s_2^{N_{a2}} \\
& - k_{l1} s_1^2 \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M (N_{a1} - 1) P_{N_{a1}-1, N_{a2}}(t) s_1^{N_{a1}-2} s_2^{N_{a2}} \\
& - k_{l1} s_1 s_2 \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M N_{a2} P_{N_{a1}-1, N_{a2}}(t) s_1^{N_{a1}-1} s_2^{N_{a2}-2} \\
& + k_{l2} M s_2 \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M P_{N_{a1}, N_{a2}-1}(t) s_1^{N_{a1}} s_2^{N_{a2}-1} \\
& - k_{l2} s_1 s_2 \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M N_{a1} P_{N_{a1}, N_{a2}-1}(t) s_1^{N_{a1}-1} s_2^{N_{a2}-1} \\
& - k_{l2} s_2^2 \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M (N_{a2} - 1) P_{N_{a1}, N_{a2}-1}(t) s_1^{N_{a1}} s_2^{N_{a2}-2} \\
& + k_{d1} \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M (N_{a1} + 1) P_{N_{a1}+1, N_{a2}}(t) s_1^{N_{a1}} s_2^{N_{a2}} \\
& + k_{d2} \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M (N_{a2} + 1) P_{N_{a1}, N_{a2}+1}(t) s_1^{N_{a1}} s_2^{N_{a2}} \\
& + (k_{l1} + k_{l2} - k_{d1}) s_1 \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M N_{a1} P_{N_{a1}, N_{a2}}(t) s_1^{N_{a1}-1} s_2^{N_{a2}} \\
& + (k_{l1} + k_{l2} - k_{d2}) s_2 \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M N_{a2} P_{N_{a1}, N_{a2}}(t) s_1^{N_{a1}} s_2^{N_{a2}-1}
\end{aligned}$$



$$-(k_{l1} + k_{l2})M \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M P_{N_{a1}, N_{a2}}(t) s_1^{N_{a1}} s_2^{N_{a2}} \quad (4.71)$$

(4.71)

$$\begin{aligned} \frac{dF(s_1, s_2, t)}{dt} &= k_{l1} M s_1 F(s_1, s_2, t) - k_{l1} s_1^2 \frac{\partial F(s_1, s_2, t)}{\partial s_1} - k_{l1} s_1 s_2 \frac{\partial F(s_1, s_2, t)}{\partial s_2} \\ &+ k_{l2} M s_2 F(s_1, s_2, t) - k_{l2} s_1 s_2 \frac{\partial F(s_1, s_2, t)}{\partial s_1} - k_{l2} s_2^2 \frac{\partial F(s_1, s_2, t)}{\partial s_2} \\ &+ k_{d1} \frac{\partial F(s_1, s_2, t)}{\partial s_1} + k_{d2} \frac{\partial F(s_1, s_2, t)}{\partial s_2} + (k_{l1} + k_{l2} - k_{d1}) s_1 \frac{\partial F(s_1, s_2, t)}{\partial s_1} \\ &\quad + (k_{l1} + k_{l2} - k_{d2}) s_2 \frac{\partial F(s_1, s_2, t)}{\partial s_2} - (k_{l1} + k_{l2}) M F(s_1, s_2, t) \end{aligned} \quad (4.72)$$

(4.44)

$$\begin{aligned} \frac{dF(s_1, s_2, t)}{dt} &= (k_{l1} M s_1 + k_{l2} M s_2 - (k_{l1} + k_{l2}) M) F(s_1, s_2, t) \\ &+ \left( (k_{l1} + k_{l2} - k_{d1}) s_1 + k_{d1} - k_{l1} s_1^2 - k_{l2} s_1 s_2 \right) \frac{\partial F(s_1, s_2, t)}{\partial s_1} \\ &+ \left( (k_{l1} + k_{l2} - k_{d2}) s_2 + k_{d2} - k_{l2} s_2^2 - k_{l1} s_1 s_2 \right) \frac{\partial F(s_1, s_2, t)}{\partial s_2} \end{aligned} \quad (4.73)$$

$$\begin{aligned} \frac{\partial F(s_1, s_2, t)}{\partial t} &= M \left[ k_{l1} (s_1 - 1) + k_{l2} (s_2 - 1) \right] F(s_1, s_2, t) \\ &+ \left[ k_{l1} s_1 (1 - s_1) + k_{l2} s_1 (1 - s_2) + k_{d1} (1 - s_1) \right] \frac{\partial F(s_1, s_2, t)}{\partial s_1} \\ &+ \left[ k_{l1} s_2 (1 - s_1) + k_{l2} s_2 (1 - s_2) + k_{d2} (1 - s_2) \right] \frac{\partial F(s_1, s_2, t)}{\partial s_2} \end{aligned} \quad (4.74)$$

$$\begin{aligned}
F(s_1, s_2, t) &= [1 + G_1(s_1 - 1) + G_2(s_2 - 1)]^M = [(1 - G_1 - G_2) + G_1s_1 + G_2s_2]^M \\
&= \sum_{N_{a1}=0}^M \sum_{N_{a2}=0}^{M-N_{a1}} \binom{M}{N_{a1}, N_{a2}} (1 - G_1 - G_2)^{M-N_{a1}-N_{a2}} (G_1s_1)^{N_{a1}} (G_2s_2)^{N_{a2}}
\end{aligned} \tag{4.75}$$

(4.79)

$$F(s_1, s_2, 0) = 1 \quad F(1, 1, t) = 1 \tag{4.76}$$

$G_1 \quad G_2$

(2.5),

$$\frac{dN_{a,i}}{dt} = k_{l,i}N_{f,i} - k_{d,i}N_{a,i} = k_{l,i} \left( M - \sum_{j=1}^2 N_{a,j} \right) - k_{d,i}N_{a,i} \quad i = 1, 2 \tag{4.77}$$

(4.75)

$$P_{N_{a1}, N_{a2}}(t) = \frac{M!}{N_{a1}!N_{a2}!(M - N_{a1} - N_{a2})!} (1 - G_1 - G_2)^{M-N_{a1}-N_{a2}} G_1^{N_{a1}} G_2^{N_{a2}} \tag{4.78}$$

(4.78)

(4.67)

4.4.3

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$$N_0 \gg N_a).$$

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$$\frac{dP_{N_{a1}, \dots, N_{ar}}(t)}{dt} = \sum_{i=1}^r P_{N_{a1}, \dots, N_{ai}-1, \dots, N_{ar}}(t) k_{li} \left( M - \sum_{j=1}^r N_{aj} + 1 \right) \quad (4.79)$$

$$+ \sum_{i=1}^r P_{N_{a1}, \dots, N_{ai}+1, \dots, N_{ar}}(t) k_{di} (N_{ai} + 1) - P_{N_{a1}, \dots, N_{ar}}(t) \left\{ \sum_{i=1}^r k_{li} \left( M - \sum_{j=1}^r N_{aj} \right) + \sum_{i=1}^r k_{di} N_{ai} \right\}$$

,

$$F(s_1, \dots, s_r, t) = [1 + G_1(s_1 - 1) + \dots + G_r(s_r - 1)]^M = \left[ G_f + \sum_{i=1}^r G_i s_i \right]^M \quad (4.80)$$

$$= \sum_{N_{a1} + \dots + N_{ar} + N_{af} = M} \binom{M}{N_{a1}, \dots, N_{ar}, N_{af}} (G_f)^{N_{af}} \prod_{i=1}^r (G_i s_i)^{N_{ai}}$$

$$G_f = \left( 1 - \sum_{i=1}^r G_i \right) \quad N_{af} = M - \sum_{i=1}^r N_{ai} \quad (4.81)$$

$$F(s_1, s_2, \dots, 0) = 1 \quad F(1, 1, \dots, t) = 1 \quad (4.82)$$

$$(4.80)$$

$$P_{N_{a1}, \dots, N_{ar}}(t) = \frac{M!}{N_{af}! \prod_{i=1}^r N_{ai}!} G_f^{N_{af}} \prod_{i=1}^r G_i^{N_{ai}} \quad (4.83)$$

$$(4.83) \quad (4.79).$$

$$\begin{aligned} & \frac{dP_{N_{a1}, \dots, N_{ar}}(t)}{dt} = \frac{M!}{N_{af}! \prod_{i=1}^r N_{ai}!} \left\{ G_f^{N_{af}} \frac{d}{dt} \left( \prod_{i=1}^r G_i^{N_{ai}} \right) + \left[ \frac{d}{dt} (G_f^{N_{af}}) \right] \left[ \prod_{i=1}^r G_i^{N_{ai}} \right] \right\} \\ & = \frac{M!}{N_{af}! \prod_{i=1}^r N_{ai}!} \left\{ G_f^{N_{af}} \sum_{j=1}^r N_{aj} G_j^{N_{aj}-1} \frac{dG_j}{dt} \prod_{\substack{i=1 \\ i \neq j}}^r G_i^{N_{ai}} + \left[ \prod_{i=1}^r G_i^{N_{ai}} \right] N_{af} G_f^{N_{af}-1} \frac{dG_f}{dt} \right\} \\ & = \frac{M!}{N_{af}! \prod_{i=1}^r N_{ai}!} \left\{ G_f^{N_{af}} \sum_{j=1}^r N_{aj} G_j^{N_{aj}-1} \frac{dG_j}{dt} \prod_{\substack{i=1 \\ i \neq j}}^r G_i^{N_{ai}} - \left[ \prod_{i=1}^r G_i^{N_{ai}} \right] N_{af} G_f^{N_{af}-1} \sum_{j=1}^r \frac{dG_j}{dt} \right\} \\ & = \frac{M! G_f^{N_{af}} \prod_{i=1}^r G_i^{N_{ai}}}{N_{af}! \prod_{i=1}^r N_{ai}!} \left\{ \sum_{j=1}^r \frac{N_{aj}}{G_j} \frac{dG_j}{dt} - \frac{N_{af}}{G_f^{N_{af}}} \sum_{j=1}^r \frac{dG_j}{dt} \right\} \quad (4.84) \end{aligned}$$

$$(4.84) \quad (4.83)$$

$$\frac{dP_{N_{a1}, \dots, N_{ar}}(t)}{dt} = P_{N_{a1}, \dots, N_{ar}}(t) \left\{ \sum_{j=1}^r \frac{N_{aj}}{G_j} \frac{dG_j}{dt} - \frac{N_{af}}{G_f^{N_{af}}} \sum_{j=1}^r \frac{dG_j}{dt} \right\} \quad (4.85)$$

$$(4.79)$$

$$(4.83).$$

$$P_{N_{a1} \dots N_{ai-1} \dots N_{ar}}(t) = \frac{M! G_f^{N_{af}+1} G_1^{N_{a1}} \dots G_i^{N_{ai}-1} \dots G_r^{N_{ar}}}{N_{a1}! \dots (N_{ai}-1)! \dots N_{ar}! (N_{af}+1)!} = \frac{N_{ai} G_f}{(N_{af}+1) G_i} P_{N_{a1} \dots N_{ar}}(t) \quad 1 < i \leq r \quad (4.86)$$

$$P_{N_{a1} \dots N_{ai+1} \dots N_{ar}}(t) = \frac{M! G_1^{N_{a1}} \dots G_i^{N_{ai}+1} \dots G_r^{N_{ar}} G_f^{N_{af}-1}}{N_{a1}! \dots (N_{ai}+1)! \dots N_{ar}! (N_{af}-1)!} = \frac{G_i N_{af}}{G_f (N_{ai}+1)} P_{N_{a1} \dots N_{ar}}(t) \quad 1 \leq i < r \quad (4.87)$$

(4.84)-(4.87) (4.79)

$$P_{N_{a1} \dots N_{ar}}(t) \left\{ \sum_{i=1}^r \frac{N_{ai}}{G_i} \frac{dG_j}{dt} - \frac{N_{af}}{G_f^{N_{af}}} \sum_{i=1}^r \frac{dG_i}{dt} \right\} = \sum_{i=1}^r \frac{N_{ai} G_f}{(N_{af}+1) G_i} P_{N_{a1} \dots N_{ar}}(t) k_{li} \left( M - \sum_{j=1}^r N_{aj} + 1 \right) + \sum_{i=1}^r \frac{G_i N_{af}}{G_f (N_{ai}+1)} P_{N_{a1} \dots N_{ar}}(t) k_{di} (N_{ai}+1) - P_{N_{a1} \dots N_{ar}}(t) \left\{ \sum_{i=1}^r k_{li} \left( M - \sum_{j=1}^r N_{aj} \right) + \sum_{i=1}^r k_{di} N_{ai} \right\} \quad (4.88)$$

(4.88)

:

$$\sum_{i=1}^r \left( \frac{N_{ai}}{G_i} - \frac{N_{af}}{G_f} \right) \frac{dG_1}{dt} = \sum_{i=1}^r k_{li} \left[ \left( M - \sum_{j=1}^r N_{aj} + 1 \right) \frac{N_{ai} G_f}{(N_{af}+1) G_i} - \left( M - \sum_{j=1}^r N_{aj} \right) \right] + \sum_{i=1}^r k_{di} \left( \frac{G_i N_{af}}{G_f} - N_{ai} \right) \quad (4.89)$$

(4.89)

$r$

:

$$\left( \frac{N_{ai}}{G_i} - \frac{N_{af}}{G_f} \right) \frac{dG_i}{dt} = k_{li} G_f \left\{ \frac{N_{ai}}{G_i} - \frac{N_{af}}{G_f} \right\} - k_{di} G_i \left\{ \frac{N_{ai}}{G_i} - \frac{N_{af}}{G_f} \right\} \quad 1 < i < r \quad (4.90)$$

(4.90)

:

$$\frac{dG_i}{dt} = k_{li} G_f - k_{di} G_i \quad 1 < i < r \quad (4.91)$$

(4.91)

$M,$

:

$$\frac{dN_{a,i}}{dt} = k_{l,i} \left( M - \sum_{j=1}^r N_{a,j} \right) - k_{d,i} N_{a,i} \quad i = 1, \dots, r \quad (4.92)$$

$$(4.79) \quad \left( \dots \right)$$

#### 4.4.4

$$(2.10).$$

$i$ -je

$$M_{a,i} = \chi_i S.$$

$$\begin{aligned} \frac{dN_{a,i}}{dt} &= k_{l,i} \left( M_{a,i} - \sum_{j=1}^r \frac{i}{j} N_{a,j} \right) - k_{d,i} N_{a,i} \\ &= k_{l,i} \left( S - \sum_{j=1}^r \frac{N_{a,j}}{j} \right) - k_{d,i} N_{a,i} \end{aligned} \quad \begin{matrix} i = 1, \dots, r \\ (4.93) \end{matrix}$$

$$(4.93)$$

$$\frac{dN_{a,i}}{dt} = k_{l,i} \left( S - \sum_{j=1}^r \frac{N_{a,j}}{j} \right) - k_{d,i} \frac{N_{a,i}}{i} \quad i = 1, \dots, r \quad (4.94)$$

$$N_{ai}/\chi_i,$$

$$\frac{d_{ni}}{dt} = k_{l,i} \left( S - \sum_{j=1}^r n_j \right) - k_{d,i} n_i \quad i = 1, \dots, r \quad (4.95)$$

(4.92),

(2.5).

,

(

)

:

$N_a$

$n_i$ ,

.

(

,

...)

( $N_{ai}, M$ )

( $n_i, M$ ).

(

$n_i$

$\chi_i$ .

1, 2, 3 4

$(0, 1/\chi_i, 2/\chi_i, \dots)$ .

#### 4.4.4.1

(4.66),



(4.93)

2

$$\frac{dN_{a,i}}{dt} = \frac{k_{l,i}}{i} (S_{1,2} - N_{a1} - N_{a2}) - k_{d,i} N_{a,i} \quad i=1,2 \quad (4.96)$$

$$\underline{M} = S_{1,2} \quad \underline{k}_{l,1} = \frac{k_{l,1}}{2} \quad \underline{k}_{l,2} = \frac{k_{l,2}}{1} \quad (4.97)$$

t.

$$\begin{aligned} P((N_{a1}-1) \rightarrow N_{a1}) &= \underline{k}_{l,1} (\underline{M} - (N_{a1}-1)_{2} - N_{a2-1}) \Delta t \\ P((N_{a2}-1) \rightarrow N_{a2}) &= \underline{k}_{l,2} (\underline{M} - N_{a1-2} - (N_{a2}-1)_{1}) \Delta t \\ P(N_{a1} \rightarrow (N_{a1}+1)) &= \underline{k}_{l,1} (\underline{M} - N_{a1-2} - N_{a2-1}) \Delta t \\ P(N_{a2} \rightarrow (N_{a2}+1)) &= \underline{k}_{l,2} (\underline{M} - N_{a1-2} - N_{a2-1}) \Delta t \\ P((N_{a1}+1) \rightarrow N_{a1}) &= k_{d1} (N_{a1}+1) \Delta t \\ P((N_{a2}+1) \rightarrow N_{a2}) &= k_{d2} (N_{a2}+1) \Delta t \\ P(N_{a1} \rightarrow (N_{a1}-1)) &= k_{d1} N_{a1} \Delta t \\ P(N_{a2} \rightarrow (N_{a2}-1)) &= k_{d2} N_{a2} \Delta t \end{aligned} \quad (4.98)$$

(4.98)

(4.65)

:

$$\begin{aligned} P(N_{a1}, N_{a2}, t + \Delta t) &= P(N_{a1}-1, N_{a2}, t) \underline{k}_{l,1} (\underline{M} - (N_{a1}-1)_{2} - N_{a2-1}) \Delta t \\ &+ P(N_{a1}, N_{a2}-1, t) \underline{k}_{l,2} (\underline{M} - N_{a1-2} - (N_{a2}-1)_{1}) \Delta t \\ &+ P(N_{a1}+1, N_{a2}, t) k_{d1} (N_{a1}+1) \Delta t + P(N_{a1}, N_{a2}+1, t) k_{d2} (N_{a2}+1) \Delta t \\ &+ P(N_{a1}, N_{a2}, t) \left( 1 - k_{d1} N_{a1} \Delta t - \underline{k}_{l,1} (\underline{M} - N_{a1-2} - N_{a2-1}) \Delta t - k_{d2} N_{a2} \Delta t \right. \\ &\quad \left. - \underline{k}_{l,2} (\underline{M} - N_{a1-2} - N_{a2-1}) \Delta t \right) \end{aligned} \quad (4.99)$$

$$P(N_{a1}, N_{a2}, t), \quad t,$$

$$\begin{aligned} \frac{dP(N_{a1}, N_{a2}, t)}{dt} = & P(N_{a1}-1, N_{a2}, t) \underline{k}_{l,1} (\underline{M} - (N_{a1}-1) - N_{a2}) \\ & + P(N_{a1}, N_{a2}-1, t) \underline{k}_{l,2} (\underline{M} - N_{a1} - (N_{a2}-1)) \\ & + P(N_{a1}+1, N_{a2}, t) k_{d1} (N_{a1}+1) + P(N_{a1}, N_{a2}+1, t) k_{d2} (N_{a2}+1) \\ & - P(N_{a1}, N_{a2}, t) \left( k_{d1} N_{a1} + (\underline{k}_{l,1} + \underline{k}_{l,2}) (\underline{M} - N_{a1} - N_{a2}) + k_{d2} N_{a2} \right) \end{aligned} \quad (4.100)$$

$$(4.100) \quad s_1^{N_{a1}} \quad N_{a1}$$

$$\begin{aligned} & \frac{d}{dt} \sum_{N_{a1}=0}^{\infty} s_1^{N_{a1}} P(N_{a1}, N_{a2}, t) = \\ & \sum_{N_{a1}=0}^{\infty} s_1^{N_{a1}} P(N_{a1}-1, N_{a2}, t) \underline{k}_{l,1} (\underline{M} - (N_{a1}-1) - N_{a2}) \\ & \quad + \sum_{N_{a1}=0}^M s_1^{N_{a1}} P(N_{a1}, N_{a2}-1, t) \underline{k}_{l,2} (\underline{M} - N_{a1} - (N_{a2}-1)) \\ & + \sum_{N_{a1}=0}^{\infty} s_1^{N_{a1}} P(N_{a1}+1, N_{a2}, t) k_{d1} (N_{a1}+1) + \sum_{N_{a1}=0}^M s_1^{N_{a1}} P(N_{a1}, N_{a2}+1, t) k_{d2} (N_{a2}+1) \\ & - \sum_{N_{a1}=0}^{\infty} s_1^{N_{a1}} P(N_{a1}, N_{a2}, t) \left( k_{d1} N_{a1} + (\underline{k}_{l,1} + \underline{k}_{l,2}) (\underline{M} - N_{a1} - N_{a2}) + k_{d2} N_{a2} \right) \end{aligned} \quad (4.101)$$

$$(4.101) \quad s_2^{N_{a2}} \quad N_{a2} \quad N_{a2}$$

$$\frac{d}{dt} \left[ \sum_{N_{a2}=0}^{\infty} s_2^{N_{a2}} \sum_{N_{a1}=0}^{\infty} s_1^{N_{a1}} P(N_{a1}, N_{a2}, t) \right] =$$

$$\begin{aligned} & \sum_{N_{a2}=0}^{\infty} s_2^{N_{a2}} \sum_{N_{a1}=0}^{\infty} s_1^{N_{a1}} P(N_{a1}-1, N_{a2}, t) \underline{k}_{l,1} (\underline{M} - (N_{a1}-1) - N_{a2}) \\ & + \sum_{N_{a2}=0}^{\infty} s_2^{N_{a2}} \sum_{N_{a1}=0}^{\infty} s_1^{N_{a1}} P(N_{a1}, N_{a2}-1, t) \underline{k}_{l,2} (\underline{M} - N_{a1} - (N_{a2}-1)) \\ & + \sum_{N_{a2}=0}^{\infty} s_2^{N_{a2}} \sum_{N_{a1}=0}^{\infty} s_1^{N_{a1}} P(N_{a1}+1, N_{a2}, t) k_{d1} (N_{a1}+1) \end{aligned}$$

$$\begin{aligned}
& + \sum_{N_{a2}=0}^{\infty} s_2^{N_{a2}} \sum_{N_{a1}=0}^{\infty} s_1^{N_{a1}} P(N_{a1}, N_{a2} + 1, t) k_{d2} (N_{a2} + 1) \\
& - \sum_{N_{a2}=0}^{\infty} s_2^{N_{a2}} \sum_{N_{a1}=0}^{\infty} s_1^{N_{a1}} P(N_{a1}, N_{a2}, t) (k_{d1} N_{a1} + k_{d2} N_{a2} + (\underline{k}_{l,1} + \underline{k}_{l,2})) (\underline{M} - N_{a1} - N_{a2})
\end{aligned} \tag{4.102}$$

(4.12) – (4.15)

$$\begin{aligned}
\frac{dF(s_1, s_2, t)}{dt} &= \underline{k}_{l,1} \underline{M} \sum_{N_{a2}=0}^{\infty} \sum_{N_{a1}=0}^{\infty} P(N_{a1} - 1, N_{a2}, t) s_1^{N_{a1}} s_2^{N_{a2}} \\
& - \underline{k}_{l,1} - 2 \sum_{N_{a2}=0}^{\infty} \sum_{N_{a1}=0}^{\infty} (N_{a1} - 1) P(N_{a1} - 1, N_{a2}, t) s_1^{N_{a1}} s_2^{N_{a2}} \\
& - \underline{k}_{l,1} - 1 \sum_{N_{a2}=0}^{\infty} \sum_{N_{a1}=0}^{\infty} N_{a2} P(N_{a1} - 1, N_{a2}, t) s_1^{N_{a1}} s_2^{N_{a2}} \\
& + \underline{k}_{l,2} \underline{M} \sum_{N_{a2}=0}^{\infty} \sum_{N_{a1}=0}^{\infty} P(N_{a1}, N_{a2} - 1, t) s_1^{N_{a1}} s_2^{N_{a2}} \\
& - \underline{k}_{l,2} - 2 \sum_{N_{a2}=0}^{\infty} \sum_{N_{a1}=0}^{\infty} N_{a1} P(N_{a1}, N_{a2} - 1, t) s_1^{N_{a1}} s_2^{N_{a2}} \\
& - \underline{k}_{l,2} - 1 \sum_{N_{a2}=0}^{\infty} \sum_{N_{a1}=0}^{\infty} (N_{a2} - 1) P(N_{a1}, N_{a2} - 1, t) s_1^{N_{a1}} s_2^{N_{a2}} \\
& + k_{d1} \sum_{N_{a2}=0}^{\infty} \sum_{N_{a1}=0}^{\infty} (N_{a1} + 1) P(N_{a1} + 1, N_{a2}, t) s_1^{N_{a1}} s_2^{N_{a2}} \\
& + k_{d2} \sum_{N_{a2}=0}^{\infty} \sum_{N_{a1}=0}^{\infty} (N_{a2} + 1) P(N_{a1}, N_{a2} + 1, t) s_1^{N_{a1}} s_2^{N_{a2}} \\
& + \left( (\underline{k}_{l,1} + \underline{k}_{l,2}) - 2 - k_{d1} \right) \sum_{N_{a2}=0}^{\infty} \sum_{N_{a1}=0}^{\infty} N_{a1} P(N_{a1}, N_{a2}, t) s_1^{N_{a1}} s_2^{N_{a2}} \\
& + \left( (\underline{k}_{l,1} + \underline{k}_{l,2}) - 1 - k_{d2} \right) \sum_{N_{a2}=0}^{\infty} \sum_{N_{a1}=0}^{\infty} N_{a2} P(N_{a1}, N_{a2}, t) s_1^{N_{a1}} s_2^{N_{a2}} \\
& - \left( \underline{k}_{l,1} + \underline{k}_{l,2} \right) \underline{M} \sum_{N_{a2}=0}^{\infty} \sum_{N_{a1}=0}^{\infty} P(N_{a1}, N_{a2}, t) s_1^{N_{a1}} s_2^{N_{a2}}
\end{aligned} \tag{4.103}$$

$$\begin{aligned}
\frac{dF(s_1, s_2, t)}{dt} &= \underline{k_{l,1}} \underline{M} s_1 \sum_{N_{a2}=0}^{\infty} \sum_{N_{a1}=0}^{\infty} P(N_{a1}-1, N_{a2}, t) s_1^{N_{a1}-1} s_2^{N_{a2}} \\
&- \underline{k_{l,1}} \underline{2} s_1^2 \sum_{N_{a2}=0}^{\infty} \sum_{N_{a1}=0}^{\infty} (N_{a1}-1) P(N_{a1}-1, N_{a2}, t) s_1^{N_{a1}-2} s_2^{N_{a2}} \\
&- \underline{k_{l,1}} \underline{1} s_1 s_2 \sum_{N_{a2}=0}^{\infty} \sum_{N_{a1}=0}^{\infty} N_{a2} P(N_{a1}-1, N_{a2}, t) s_1^{N_{a1}-1} s_2^{N_{a2}-2} \\
&+ \underline{k_{l,2}} \underline{M} s_2 \sum_{N_{a2}=0}^{\infty} \sum_{N_{a1}=0}^{\infty} P(N_{a1}, N_{a2}-1, t) s_1^{N_{a1}} s_2^{N_{a2}-1} \\
&- \underline{k_{l,2}} \underline{2} s_1 s_2 \sum_{N_{a2}=0}^{\infty} \sum_{N_{a1}=0}^{\infty} N_{a1} P(N_{a1}, N_{a2}-1, t) s_1^{N_{a1}-1} s_2^{N_{a2}-1} \\
&- \underline{k_{l,2}} \underline{1} s_2^2 \sum_{N_{a2}=0}^{\infty} \sum_{N_{a1}=0}^{\infty} (N_{a2}-1) P(N_{a1}, N_{a2}-1, t) s_1^{N_{a1}} s_2^{N_{a2}-2} \\
&+ k_{d1} \sum_{N_{a2}=0}^{M-N_{a1}} \sum_{N_{a1}=0}^M (N_{a1}+1) P(N_{a1}+1, N_{a2}, t) s_1^{N_{a1}} s_2^{N_{a2}} \\
&+ k_{d2} \sum_{N_{a2}=0}^{\infty} \sum_{N_{a1}=0}^{\infty} (N_{a2}+1) P(N_{a1}, N_{a2}+1, t) s_1^{N_{a1}} s_2^{N_{a2}} \\
&+ \left( (\underline{k_{l,1}} + \underline{k_{l,2}}) \underline{2} - k_{d1} \right) s_1 \sum_{N_{a2}=0}^{\infty} \sum_{N_{a1}=0}^{\infty} N_{a1} P(N_{a1}, N_{a2}, t) s_1^{N_{a1}-1} s_2^{N_{a2}} \\
&+ \left( (\underline{k_{l,1}} + \underline{k_{l,2}}) \underline{1} - k_{d2} \right) s_2 \sum_{N_{a2}=0}^{\infty} \sum_{N_{a1}=0}^{\infty} N_{a2} P(N_{a1}, N_{a2}, t) s_1^{N_{a1}} s_2^{N_{a2}-1} \\
&- (\underline{k_{l,1}} + \underline{k_{l,2}}) \underline{M} \sum_{N_{a2}=0}^{\infty} \sum_{N_{a1}=0}^{\infty} P(N_{a1}, N_{a2}, t) s_1^{N_{a1}} s_2^{N_{a2}}
\end{aligned} \tag{4.104}$$

$$\begin{aligned}
\frac{\partial F(s_1, s_2, t)}{\partial t} &= \\
&\underline{k_{l,1}} \underline{M} s_1 F(s_1, s_2, t) - \underline{k_{l,1}} \underline{2} s_1^2 \frac{\partial F(s_1, s_2, t)}{\partial s_1} - \underline{k_{l,1}} \underline{1} s_1 s_2 \frac{\partial F(s_1, s_2, t)}{\partial s_2} \\
&+ \underline{k_{l,2}} \underline{M} s_2 F(s_1, s_2, t) - \underline{k_{l,2}} \underline{2} s_1 s_2 \frac{\partial F(s_1, s_2, t)}{\partial s_1} - \underline{k_{l,2}} \underline{1} s_2^2 \frac{\partial F(s_1, s_2, t)}{\partial s_2} \\
&+ k_{d1} \frac{\partial F(s_1, s_2, t)}{\partial s_1} + k_{d2} \frac{\partial F(s_1, s_2, t)}{\partial s_2} + \left( (\underline{k_{l,1}} + \underline{k_{l,2}}) \underline{2} - k_{d1} \right) s_1 \frac{\partial F(s_1, s_2, t)}{\partial s_1}
\end{aligned}$$

$$+ \left( (\underline{k}_{l,1} + \underline{k}_{l,2})_1 - k_{d2} \right) s_2 \frac{\partial F(s_1, s_2, t)}{\partial s_2} - (\underline{k}_{l,1} + \underline{k}_{l,2}) \underline{M} F(s_1, s_2, t) \quad (4.105)$$

$$\begin{aligned} \frac{\partial F(s_1, s_2, t)}{\partial t} &= \underline{M} \left[ \underline{k}_{l,1} (s_1 - 1) + \underline{k}_{l,2} (s_2 - 1) \right] F(s_1, s_2, t) \\ &+ \left[ \underline{k}_{l,1} {}_2s_1(1 - s_1) + \underline{k}_{l,2} {}_2s_1(1 - s_2) + k_{d1}(1 - s_1) \right] \frac{\partial F(s_1, s_2, t)}{\partial s_1} \\ &+ \left[ \underline{k}_{l,1} {}_1s_2(1 - s_1) + \underline{k}_{l,2} {}_1s_2(1 - s_2) + k_{d2}(1 - s_2) \right] \frac{\partial F(s_1, s_2, t)}{\partial s_2} \end{aligned} \quad (4.106)$$

(4.106)

(4.74)

## 4.5

[37],

: (4.61) – (4.63).

4.4.2, (4.65) – (4.77) .

1

$$\begin{aligned}
 & 1 \quad , \\
 & \frac{\partial F(s,t)}{\partial t} = Mk_l(s-1)F(s,t) - (s-1)(k_d + k_l s) \frac{\partial F(s,t)}{\partial s}, F(1,t) = 1 \quad F(s,0) = 1 \\
 & F(s,t) = \left[ 1 + (s-1) \frac{N_a}{M} \right]^M = [1 + (s-1)G]^M \\
 & P_{N_a}(t) = \binom{M}{N_a} \left( 1 - \frac{N_a}{M} \right)^{M-N_a} \left( \frac{N_a}{M} \right)^{N_a} = \binom{M}{N_a} (1-G)^{M-N_a} (G)^{N_a} \\
 & N_a = \overline{N_a} = \frac{k_l M}{k_l + k_d} (1 - e^{-(k_l + k_d)t}) = MG \\
 & D = \frac{k_l M (1 - e^{-kt}) (k_d + k_l e^{-kt})}{(k_l + k_d)^2} = \overline{N_a} \left( 1 - \frac{\overline{N_a}}{M} \right) \\
 & u = \frac{\sqrt{D\{X(t)\}}}{\sqrt{E\{X(t)\}^2}} = \sqrt{\frac{\overline{N_a} (1 - \overline{N_a} / M)}{\overline{N_a}^2}} = \sqrt{\frac{[k_d + k_l e^{-kt}]}{Mk_l (1 - e^{-kt})}} \quad ,
 \end{aligned}$$


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$$\begin{aligned}
 & 2 \quad , \\
 & F(s_1, s_2, t) = \left[ 1 + \frac{N_{a1}}{M} (s_1 - 1) + \frac{N_{a2}}{M} (s_2 - 1) \right]^M, F(s_1, s_2, 0) = 1 \quad F(1, 1, t) = 1 \\
 & P_{N_{a1}, N_{a2}}(t) = \frac{M!}{N_{a1}! N_{a2}! (M - N_{a1} - N_{a2})!} (1 - G_1 - G_2)^{M - N_{a1} - N_{a2}} G_1^{N_{a1}} G_2^{N_{a2}}
 \end{aligned}$$

$$\frac{d\overline{N}_{a1}}{dt} = k_{l1} (M - \overline{N}_{a1} - \overline{N}_{a2}) - k_{d1} \overline{N}_{a1}$$

$$\frac{d\overline{N}_{a2}}{dt} = k_{l2} (M - \overline{N}_{a1} - \overline{N}_{a2}) - k_{d2} \overline{N}_{a2}$$

$$\overline{N}_{a1} = N_{a1} \quad D\{N_{a1}\} = \overline{N}_{a1} (1 - \overline{N}_{a1} / M) \quad u\{N_{a1}\} = \sqrt{1 / \overline{N}_{a1} - 1 / M}$$

$$\overline{N}_{a2} = N_{a2} \quad D\{N_{a2}\} = \overline{N}_{a2} (1 - \overline{N}_{a2} / M) \quad u\{N_{a3}\} = \sqrt{1 / \overline{N}_{a3} - 1 / M}$$

3 ,

$$F(s_1, s_2, t) = [1 + G_1(s_1 - 1) + G_2(s_2 - 1) + G_3(s_3 - 1)]^M,$$

$$F(s_1, s_2, s_3, 0) = 1 \quad F(1, 1, 1, t) = 1$$

$$P_{N_{a1}, N_{a2}, N_{a3}}(t) = \frac{M!}{N_{af}! \prod_{i=1}^3 N_{ai}!} G_f^{N_{af}} \prod_{i=1}^3 G_i^{N_{ai}} \quad G_f = \left(1 - \sum_{i=1}^3 G_i\right) \quad N_{af} = M - \sum_{i=1}^3 N_{ai}$$

$$\overline{N}_{a1} = N_{a1} = MG_1 \quad D\{N_{a1}\} = \overline{N}_{a1} (1 - \overline{N}_{a1} / M) \quad u\{N_{a1}\} = \sqrt{1 / \overline{N}_{a1} - 1 / M}$$

$$\overline{N}_{a2} = N_{a2} = MG_2 \quad D\{N_{a2}\} = \overline{N}_{a2} (1 - \overline{N}_{a2} / M) \quad u\{N_{a2}\} = \sqrt{1 / \overline{N}_{a2} - 1 / M}$$

$$\overline{N}_{a3} = N_{a3} = MG_3 \quad D\{N_{a3}\} = \overline{N}_{a3} (1 - \overline{N}_{a3} / M) \quad u\{N_{a3}\} = \sqrt{1 / \overline{N}_{a3} - 1 / M}$$

$$G_1(t) = \frac{-k_{l,1} k_{d,2} k_{d,3}}{\underline{z}_1 \underline{z}_2 \underline{z}_3} + e^{\underline{z}_1 t} \frac{k_{l,1} (\underline{z}_1 + k_{d,2}) (\underline{z}_1 + k_{d,3})}{\underline{z}_1 (\underline{z}_1 - \underline{z}_2) (\underline{z}_1 - \underline{z}_3)}$$

$$+ e^{\underline{z}_2 t} \frac{k_{l,1} (\underline{z}_2 + k_{d,2}) (\underline{z}_2 + k_{d,3})}{\underline{z}_2 (\underline{z}_2 - \underline{z}_1) (\underline{z}_2 - \underline{z}_3)} + e^{\underline{z}_3 t} \frac{k_{l,1} (\underline{z}_3 + k_{d,2}) (\underline{z}_3 + k_{d,3})}{\underline{z}_3 (\underline{z}_3 - \underline{z}_1) (\underline{z}_3 - \underline{z}_2)}$$

$$G_2(t) = \frac{-k_{l,2} k_{d,1} k_{d,3}}{\underline{z}_1 \underline{z}_2 \underline{z}_3} + e^{\underline{z}_1 t} \frac{k_{l,2} (\underline{z}_1 + k_{d,1}) (\underline{z}_1 + k_{d,3})}{\underline{z}_1 (\underline{z}_1 - \underline{z}_2) (\underline{z}_1 - \underline{z}_3)}$$

$$+ e^{\underline{z}_2 t} \frac{k_{l,2} (\underline{z}_2 + k_{d,1}) (\underline{z}_2 + k_{d,3})}{\underline{z}_2 (\underline{z}_2 - \underline{z}_1) (\underline{z}_2 - \underline{z}_3)} + e^{\underline{z}_3 t} \frac{k_{l,2} (\underline{z}_3 + k_{d,1}) (\underline{z}_3 + k_{d,3})}{\underline{z}_3 (\underline{z}_3 - \underline{z}_1) (\underline{z}_3 - \underline{z}_2)}$$

$$G_3(t) = \frac{-k_{l,3} k_{d,2} k_{d,1}}{\underline{z}_1 \underline{z}_2 \underline{z}_3} + e^{\underline{z}_1 t} \frac{k_{l,3} (\underline{z}_1 + k_{d,2}) (\underline{z}_1 + k_{d,1})}{\underline{z}_1 (\underline{z}_1 - \underline{z}_2) (\underline{z}_1 - \underline{z}_3)}$$

$$+ e^{\underline{z}_2 t} \frac{k_{l,3} (\underline{z}_2 + k_{d,2}) (\underline{z}_2 + k_{d,1})}{\underline{z}_2 (\underline{z}_2 - \underline{z}_1) (\underline{z}_2 - \underline{z}_3)} + e^{\underline{z}_3 t} \frac{k_{l,3} (\underline{z}_3 + k_{d,2}) (\underline{z}_3 + k_{d,1})}{\underline{z}_3 (\underline{z}_3 - \underline{z}_1) (\underline{z}_3 - \underline{z}_2)}$$

$\underline{z}_1, \underline{z}_2, \underline{z}_3$

$P_3(s)$

$$: \quad P_3(s) = s^3 + c_2s^2 + c_1s + c_0 = (s - \underline{z}_1)(s - \underline{z}_2)(s - \underline{z}_3)$$

$$c_2 = k_{l,1} + k_{l,2} + k_{l,3} + k_{d,1} + k_{d,2} + k_{d,3}$$

$$c_1 = k_{l,1}(k_{d,2} + k_{d,3}) + k_{l,2}(k_{d,1} + k_{d,3}) + k_{l,3}(k_{d,2} + k_{d,1}) \\ + k_{d,1}k_{d,2} + k_{d,2}k_{d,3} + k_{d,1}k_{d,3}$$

$$c_0 = k_{l,3}k_{d,1}k_{d,2} + k_{l,1}k_{d,3}k_{d,2} + k_{l,2}k_{d,1}k_{d,3} + k_{d,3}k_{d,1}k_{d,2}$$

$$\frac{dN_{a,i}}{dt} = k_{l,i}N_{f,i} - k_{d,i}N_{a,i} = k_{l,i} \left( M - \sum_{j=1}^r N_{a,j} \right) - k_{d,i}N_{a,i} \quad i = 1, \dots, r$$

$$F(s_1, \dots, s_r, t) = [1 + G_1(s_1 - 1) + \dots + G_r(s_r - 1)]^M = \left[ G_f + \sum_{i=1}^r G_i s_i \right]^M$$

$$= \sum_{N_{a1} + \dots + N_{ar} + N_{af} = M} \binom{M}{N_{a1}, \dots, N_{ar}, N_{af}} (G_f)^{N_{af}} \prod_{i=1}^r (G_i s_i)^{N_{ai}}$$

$$F(s_1, s_2, \dots, 0) = 1 \quad F(1, 1, \dots, t) = 1$$

$$G_f = \left( 1 - \sum_{i=1}^r G_i \right) \quad N_{af} = M - \sum_{i=1}^r N_{ai}$$

(4.77)

$$P_{N_{a1}, \dots, N_{ar}}(t) = \frac{M!}{N_{af}! \cdot \prod_{i=1}^r N_{ai}!} G_f^{N_{af}} \prod_{i=1}^r G_i^{N_{ai}}$$

$$\overline{N_{a1}} = N_{a1} = MG_1 \quad D\{N_{a1}\} = \overline{N_{a1}} \left( 1 - \overline{N_{a1}} / M \right) \quad u\{N_{a1}\} = \sqrt{1 / \overline{N_{a1}} - 1 / M}$$

...

...

...

$$\overline{N_{ar}} = N_{ar} = MG_r \quad D\{N_{ar}\} = \overline{N_{ar}} \left( 1 - \overline{N_{ar}} / M \right) \quad u\{N_{ar}\} = \sqrt{1 / \overline{N_{ar}} - 1 / M}$$

$$\frac{d\overline{N_{a1}}}{dt} = k_{l1} \left( S_1 - \overline{N_{a1}} - \frac{1}{2} \overline{N_{a2}} \right) - k_{d1} \overline{N_{a1}}$$

$$\frac{d\overline{N_{a2}}}{dt} = k_{l2} \left( S_2 - \frac{2}{1} \overline{N_{a1}} - \overline{N_{a2}} \right) - k_{d2} \overline{N_{a2}}$$



1

$$\frac{dN_a}{dt} = k_a N_0 M - (k_d + k_a (M + N_0)) N_a + k_a N_a^2$$

$$\frac{d\overline{N}_a}{dt} = k_a N_0 M - (k_d + k_a (M + N_0)) \overline{N}_a + k_a \overline{N}_a^2$$

### 4.6

SPP

$$\Delta n_{eff} = N_a \frac{(n_A - n_e)}{M} = w N_a \quad (4.107)$$

( $n_e$ ),  
 $w$  :

$$w = \frac{(n_A - n_e)}{M} \quad (4.108)$$

[77]:

$$\Delta n_{eff} = \sum_{i=1}^r N_{a,i} \frac{(n_{Ai} - n_e)}{M} = \sum_{i=1}^r w_i N_{a,i} \quad (4.109)$$

$r$  ( 0 )

$$(M+1)^2$$

[78]:

$$\{0, w_1, w_2, w_1 + w_2, \dots, Mw_1, \dots, Mw_2, \dots, M(w_1 + w_2)\} \quad (4.110)$$

$$E\{\Delta n_{eff}\} = \sum_{i=1}^r w_i E\{N_{a,i}\} \quad (4.111)$$

$$D\{\Delta n_{eff}\} = E\{(\Delta n_{eff})^2\} - (E\{\Delta n_{eff}\})^2 \quad (4.112)$$

$$(4.111) \quad (4.112)$$

$$u\{\Delta n_{eff}\} = \sqrt{\frac{D\{\Delta n_{eff}\}}{E\{\Delta n_{eff}\}^2}} \quad (4.113)$$

#### 4.6.1

$$1 \quad (3.32),$$

$$E\{\Delta n_{eff}\} = E\{wN_a\} = wE\{N_a\} = wMG \quad (4.114)$$

SPP

(

(4.10)

( ) (4.57).

= 1 :

$$\left. \frac{\partial F(s,t)}{\partial s} \right|_{s=1} = \frac{\partial}{\partial s} [1+(s-1)G]^M \Big|_{s=1} = M [1+(s-1)G]^{M-1} \bar{G} \Big|_{s=1} = MG = \bar{N}_a \quad (4.115)$$

= 1 :

$$\left. \frac{\partial^2 F(s,t)}{\partial s^2} \right|_{s=1} = \frac{\partial}{\partial s} \left\{ M [1+(s-1)\bar{G}]^{M-1} \bar{G} \right\} \Big|_{s=1} = \bar{G}^2 M (M-1) \quad (4.116)$$

:

$$D\{N_a\} = \left. \frac{\partial^2 F(s,t)}{\partial s^2} \right|_{s=1} + \left. \frac{\partial F(s,t)}{\partial s} \right|_{s=1} - \left( \left. \frac{\partial F(s,t)}{\partial s} \right|_{s=1} \right)^2 = \quad (4.117)$$

$$G^2 M (M-1) + MG - M^2 G^2 = MG(1-G) = \bar{N}_a \left( 1 - \frac{\bar{N}_a}{M} \right)$$

:

$$u\{N_a\} = \sqrt{\frac{D\{N_a\}}{E\{N_a\}^2}} = \sqrt{\frac{\bar{N}_a \left( 1 - \frac{\bar{N}_a}{M} \right)}{\bar{N}_a^2}} = \sqrt{\frac{1}{\bar{N}_a} - \frac{1}{M}} \quad (4.118)$$

:

$$E\{\Delta n_{eff}\} = w\bar{N}_a \quad D\{\Delta n_{eff}\} = w^2 D\{N_a\} = w^2 \bar{N}_a \left( 1 - \frac{\bar{N}_a}{M} \right)$$

$$u\{\Delta n_{eff}\} = \sqrt{\frac{1}{\bar{N}_a} - \frac{1}{M}} \quad (4.119)$$

4.6.2

–

:

$$E \{ \Delta n_{eff} \} = E \{ w_1 N_{a1} + w_2 N_{a2} \} = w_1 M G_1 + w_2 M G_2 = w_1 \overline{N_{a1}} + w_2 \overline{N_{a2}} \quad (4.120)$$

$$(4.112)$$

$$\begin{aligned} D \{ \Delta n_{eff} \} &= E \{ (w_1 N_{a1} + w_2 N_{a2})^2 \} - (w_1 \overline{N_{a1}} + w_2 \overline{N_{a2}})^2 \\ &= w_1^2 \overline{N_{a1}^2} + w_2^2 \overline{N_{a2}^2} + 2w_1 w_2 \overline{N_{a1} N_{a2}} - (w_1^2 \overline{N_{a1}^2} + w_2^2 \overline{N_{a2}^2} + 2w_1 w_2 \overline{N_{a1} N_{a2}}) \end{aligned} \quad (4.121)$$

$$= w_1^2 D \{ N_{a1} \} + w_2^2 D \{ N_{a2} \} + 2w_1 w_2 (\overline{N_{a1} N_{a2}} - \overline{N_{a1}} \overline{N_{a2}}) \quad (4.15)$$

$$\begin{aligned} \overline{N_{a1} N_{a2}} &= \left. \frac{\partial^2 F}{\partial s_1 \partial s_2} \right|_{s_1=s_2=1} = \left. \frac{\partial^2}{\partial s_1 \partial s_2} [(1-G_1-G_2) + s_1 G_1 + s_2 G_2]^M \right|_{s_1=s_2=1} = \\ &= \left. \frac{\partial}{\partial s_2} \left\{ M [(1-G_1-G_2) + s_1 G_1 + s_2 G_2]^{M-1} G_2 \right\} \right|_{s_1=s_2=1} = \\ &= M(M-1) [(1-G_1-G_2) + s_1 G_1 + s_2 G_2]^{M-2} G_1 G_2 \Big|_{s_1=s_2=1} \\ &= M(M-1) G_1 G_2 \neq \overline{N_{a1}} \cdot \overline{N_{a2}} \end{aligned} \quad (4.122)$$

$$D \{ \Delta n_{eff} \} = w_1^2 \overline{N_{a1}} \left( 1 - \frac{\overline{N_{a1}}}{M} \right) + w_2^2 \overline{N_{a2}} \left( 1 - \frac{\overline{N_{a2}}}{M} \right) + \frac{2w_1 w_2}{M} \overline{N_{a1} N_{a2}} \quad (4.123)$$

:

$$\begin{aligned} u \{ \Delta n_{eff} \} &= \sqrt{\frac{w_1^2 M G_1 (1-G_1) + w_2^2 M G_2 (1-G_2) + 2w_1 w_2 M G_1 G_2}{(w_1 M G_1 + w_2 M G_2)^2}} \\ &= \sqrt{\frac{w_1^2 M G_1 + w_2^2 M G_2 + 2w_1 w_2 M G_1 G_2 - w_1^2 M G_1^2 - w_2^2 M G_2^2}{(w_1 M G_1 + w_2 M G_2)^2}} \\ &= \sqrt{\frac{w_1^2 M G_1 + w_2^2 M G_2}{(w_1 M G_1 + w_2 M G_2)^2} - \frac{1}{M}} \end{aligned} \quad (4.124)$$

4.6.3

–

$$E \{ \Delta n_{eff} \} = w_1 M G_1 + w_2 M G_2 + w_3 M G_3 \quad (4.125)$$

$$\begin{aligned} D \{ \Delta n_{eff} \} &= E \left\{ (w_1 N_{a1} + w_2 N_{a2} + w_3 N_{a3})^2 \right\} - (w_1 \overline{N_{a1}} + w_2 \overline{N_{a2}} + w_3 \overline{N_{a3}})^2 \\ &= w_1^2 \overline{N_{a1}^2} + w_2^2 \overline{N_{a2}^2} + w_3^2 \overline{N_{a3}^2} + 2w_1 w_2 \overline{N_{a1} N_{a2}} + 2w_1 w_3 \overline{N_{a1} N_{a3}} + 2w_3 w_2 \overline{N_{a3} N_{a2}} \\ &\quad - \left( w_1^2 \overline{N_{a1}^2} + w_2^2 \overline{N_{a2}^2} + w_3^2 \overline{N_{a3}^2} + 2w_1 w_2 \overline{N_{a1} N_{a2}} + 2w_1 w_3 \overline{N_{a1} N_{a3}} + 2w_3 w_2 \overline{N_{a3} N_{a2}} \right) \end{aligned} \quad (4.126)$$

$$\begin{aligned} &= w_1^2 D \{ N_{a1} \} + w_2^2 D \{ N_{a2} \} + w_3^2 D \{ N_{a3} \} + 2w_1 w_2 (\overline{N_{a1} N_{a2}} - \overline{N_{a1}} \overline{N_{a2}}) \\ &\quad + 2w_1 w_3 (\overline{N_{a1} N_{a3}} - \overline{N_{a1}} \overline{N_{a3}}) + 2w_3 w_2 (\overline{N_{a3} N_{a2}} - \overline{N_{a3}} \overline{N_{a2}}) \end{aligned}$$

$$\overline{N_{a1} N_{a2}} = \frac{\partial^2 F}{\partial s_1 \partial s_2} \Big|_{s_1=s_2=1} = \frac{\partial^2}{\partial s_1 \partial s_2} \left[ (1 - G_1 - G_2 - G_3) + s_1 G_1 + s_2 G_2 + s_3 G_3 \right]^M \Big|_{s_1=s_2=1} =$$

$$\begin{aligned} &\frac{\partial}{\partial s_2} \left\{ M \left[ (1 - G_1 - G_2 - G_3) + s_1 G_1 + s_2 G_2 + s_3 G_3 \right]^{M-1} G_1 \right\} \Big|_{s_1=s_2=1} = \\ &M (M - 1) \left[ (1 - G_1 - G_2 - G_3) + s_1 G_1 + s_2 G_2 + s_3 G_3 \right]^{M-2} G_1 G_2 \Big|_{s_1=s_2=1} \end{aligned} \quad (4.127)$$

$$= M (M - 1) G_1 G_2 \neq \overline{N_{a1}} \cdot \overline{N_{a2}}$$

$$N_{a3} N_{a2} \quad (4.127)$$

$$\overline{N_{a3} N_{a2}} = \frac{\partial^2 F}{\partial s_3 \partial s_2} \Big|_{s_3=s_2=1} = \frac{\partial^2}{\partial s_3 \partial s_2} \left[ (1 - G_1 - G_2 - G_3) + s_1 G_1 + s_2 G_2 + s_3 G_3 \right]^M \Big|_{s_3=s_2=1} =$$

$$\begin{aligned} &\frac{\partial}{\partial s_2} \left\{ M \left[ (1 - G_1 - G_2 - G_3) + s_1 G_1 + s_2 G_2 + s_3 G_3 \right]^{M-1} G_3 \right\} \Big|_{s_3=s_2=1} = \\ &M (M - 1) \left[ (1 - G_1 - G_2 - G_3) + s_1 G_1 + s_2 G_2 + s_3 G_3 \right]^{M-2} G_3 G_2 \Big|_{s_3=s_2=1} \end{aligned} \quad (4.128)$$

$$= M (M - 1) G_3 G_2 \neq \overline{N_{a3}} \cdot \overline{N_{a2}}$$

$$N_{a1} N_{a3} \quad (4.127)$$

$$\begin{aligned}
\overline{N_{a1}N_{a3}} &= \frac{\partial^2 F}{\partial s_1 \partial s_3} \Big|_{s_1=s_3=1} = \frac{\partial^2}{\partial s_1 \partial s_3} \left[ (1 - G_1 - G_2 - G_3) + s_1 G_1 + s_2 G_2 + s_3 G_3 \right]^M \Big|_{s_1=s_3=1} = \\
&\frac{\partial}{\partial s_3} \left\{ M \left[ (1 - G_1 - G_2 - G_3) + s_1 G_1 + s_2 G_2 + s_3 G_3 \right]^{M-1} G_1 \right\} \Big|_{s_1=s_3=1} = \\
&M(M-1) \left[ (1 - G_1 - G_2 - G_3) + s_1 G_1 + s_2 G_2 + s_3 G_3 \right]^{M-2} G_1 G_3 \Big|_{s_1=s_3=1} \quad (4.129) \\
&= M(M-1) G_1 G_3 \neq \overline{N_{a1}} \cdot \overline{N_{a3}}
\end{aligned}$$

:

$$\begin{aligned}
D\{\Delta n_{eff}\} &= w_1^2 M G_1 (1 - G_1) + w_2^2 M G_2 (1 - G_2) + w_3^2 M G_3 (1 - G_3) \\
&- 2M [w_1 w_2 G_1 G_2 + w_1 w_3 G_1 G_3 + w_3 w_2 G_3 G_2] \quad (4.130)
\end{aligned}$$

$$(4.125) - (1.130)$$

$$3.3.2, \quad (3.31) - (3.36),$$

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MATLAB, Mathematica, Mapple .

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a

MATLAB 2012b symbolic toolbox,

Mathworks.

```

>> syms M k1 k2 kd1 kd2 y1 y2 Na1 Na2
>> [y1,y2]=dsolve('Dy1==k11*M-k11*y1-k11*y2-kd1*y1','Dy2==k12*M-k12*y2-kd2*y2-kd2*y2','y1(0)=0','y2(0)=0')
y1 =
(exp(-t*(kd1 + k11))*exp(-t*(kd2 + 2*k12))*(kd2*exp(t*(kd2 + 2*k12))*(M*k11^2 + M*kd1*k11 - M*kd2*k11 - M*k11*k12)/((kd1 + k11)*(kd1 - kd2 + k11 - 2*k12)) - (M*k11*exp(kd1*t + k11*t)*(kd1 - kd2 + k11 - kd2))/((kd1 + k11)*(kd1 - kd2 + k11 - 2*k12))) - kd1*exp(t*(kd2 + 2*k12))*(M*k11^2 + M*kd1*k11 - M*kd2*k11 - M*k11*k12)/((kd1 + k11)*(kd1 - kd2 + k11 - 2*k12)) - (M*k11*exp(kd1*t + k11*t)*(kd1 - kd2 + k11 - kd2))/((kd1 + k11)*(kd1 - kd2 + k11 - 2*k12))) - kd1*exp(t*(kd2 + 2*k12))*(M*k11^2 + M*kd1*k11 - M*kd2*k11 - M*k11*k12)/((kd1 + k11)*(kd1 - kd2 + k11 - 2*k12))) + 2*k12*exp(t*(kd2 + 2*k12))*(M*k11^2 + M*kd1*k11 - M*kd2*k11 - M*k11*k12)/((kd1 + k11)*(kd1 - kd2 + k11 - 2*k12)) - (M*k11*exp(kd1*t + k11*t)*(kd1 - kd2 + k11 - kd2))/((kd1 + k11)*(kd1 - kd2 + k11 - 2*k12))) + k11*exp(t*(kd1 + k11))*(M*k12)/(kd2 + 2*k12) - (M*k12*exp(kd2*t)*exp(2*k12*t))/(kd2 + 2*k12))/((kd1 - kd2 + k11 - 2*k12))
y2 =
-exp(-t*(kd2 + 2*k12))*((M*k12)/(kd2 + 2*k12) - (M*k12*exp(kd2*t)*exp(2*k12*t))/(kd2 + 2*k12))

```

Сл. 5.1 Процедура за решавање Лагергренових једначина у временском домену програмским пакетом MATLAB, 3-компонентна меша.

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```

>> syms M k1 k2 kd1 kd2 y1 y2 Na1 Na2 w
>> [Na1,Na2]=solve('w*Na1=k11*(M/w-Na1-Na2)-kd1*Na1','w*Na2=k12*(M/w-Na1-Na2)-kd2*Na2',Na1,Na2)
Na1 =
(M*k11*(kd2 + w))/(w*(kd1*kd2 + kd1*k12 + kd2*k11 + kd1*w + kd2*w + k11*w + k12*w + w^2))
Na2 =
(M*k12*(kd1 + w))/(w*(kd1*kd2 + kd1*k12 + kd2*k11 + kd1*w + kd2*w + k11*w + k12*w + w^2))
>> ilaplace(Na1,w)
ans =
(M*k11*k12)/(kd1*kd2 + kd1*k12 + kd2*k11) - (M*k11*k12*exp(-w*(kd1/2 + kd2/2 + k11/2 + k12/2))*cosh(w*(kd1^2/4 - (kd1*kd2)/2 + (kd1*k11)/2 - (kd1*k12)/2 + kd2^2/4 - (kd2*k11)/2 + (kd2*k12)/2 + k11^2/4 + (k11*k12)/2 + k12^2/4 - (kd1*kd2)/2 + (kd1*k11)/2 - (kd1*k12)/2 + kd2^2/4 - (kd2*k11)/2 + (kd2*k12)/2 + k11^2/4 + (k11*k12)/2 + k12^2/4)^(1/2))*(kd1/2 + kd2/2 + k11/2 + k12/2 - (M*k11*k12^2 + M*k11*k12*kd2 - M*kd1*k11*k12)/(M*k11*k12))/((kd1^2/4 - (kd1*kd2)/2 + (kd1*k11)/2 - (kd1*k12)/2 + kd2^2/4 - (kd2*k11)/2 + (kd2*k12)/2 + k11^2/4 + (k11*k12)/2 + k12^2/4)^(1/2))/(kd1*kd2 + kd1*k12 + kd2*k11)
>> ilaplace(Na2,w)
ans =
(M*k12*k12)/(kd1*kd2 + kd1*k12 + kd2*k11) - (M*k12*k12*exp(-w*(kd1/2 + kd2/2 + k11/2 + k12/2))*cosh(w*(kd1^2/4 - (kd1*kd2)/2 + (kd1*k11)/2 - (kd1*k12)/2 + kd2^2/4 - (kd2*k11)/2 + (kd2*k12)/2 + k11^2/4 + (k11*k12)/2 + k12^2/4)^(1/2)) - (sinh(w*(kd1^2/4 - (kd1*kd2)/2 + (kd1*k11)/2 - (kd1*k12)/2 + kd2^2/4 - (kd2*k11)/2 + (kd2*k12)/2 + k11^2/4 + (k11*k12)/2 + k12^2/4)^(1/2))*(kd1/2 + kd2/2 + k11/2 + k12/2 - (M*k12*k11^2 + M*k11*k12*kd1 - M*kd2*k11*k12)/(M*k11*k12))/((kd1^2/4 - (kd1*kd2)/2 + (kd1*k11)/2 - (kd1*k12)/2 + kd2^2/4 - (kd2*k11)/2 + (kd2*k12)/2 + k11^2/4 + (k11*k12)/2 + k12^2/4)^(1/2))/(kd1*kd2 + kd1*k12 + kd2*k11)

```

Сл. 5.2 Софтверско решавање Лагергренових једначина у комплексном домену, двокомпонентни аналит, окружење MATLAB



w.

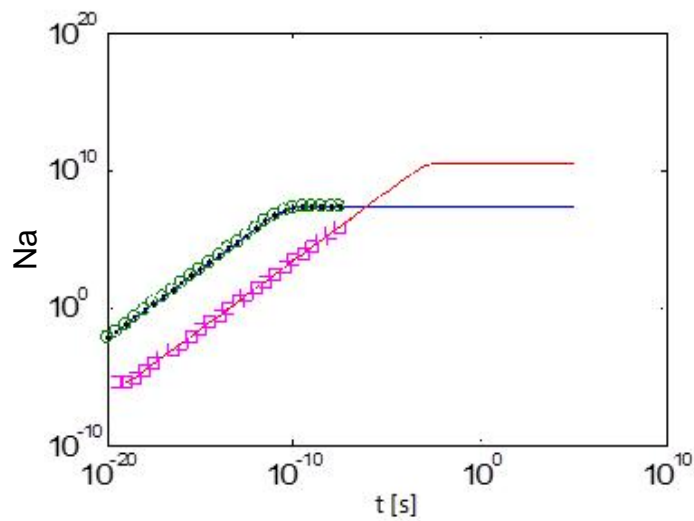
5.3

1 Pa (

)

0.1 mPa (

),



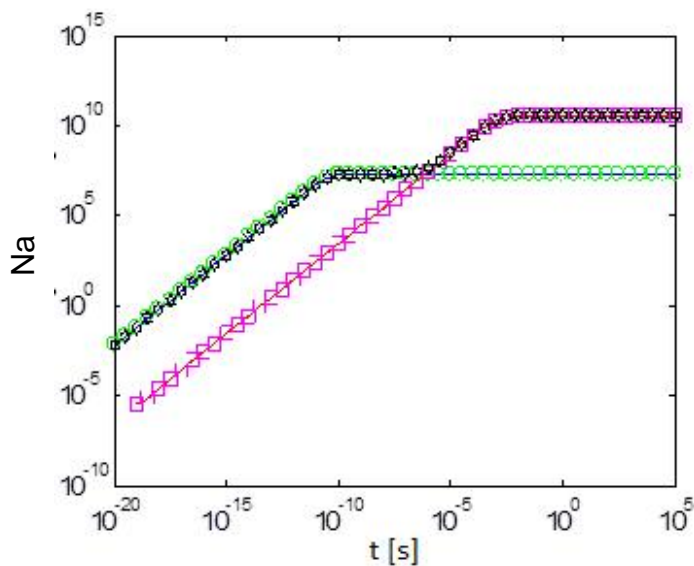
Сл. 5.3 Временска еволуција броја адсорбованих молекула двокомпонентне смеше. Метан на парцијалном притиску од 1 Pa и бензен на парцијалном притиску од 0.1 mPa Линеје су добијене на основу аналитичких израза а симболи на основу решења Symbolic Toolbox пакета MATLAB

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. 5.5

```
>> syms M kd1 kd2 kd1 kd2 y1 y2 Na1 Na2 z1 z2
>> Na1=(kd1*M*(s+kd2))/(s*(s-z1)*(s-z2));
>> Na2=(kd2*M*(s+kd1))/(s*(s-z1)*(s-z2));
>> ilaplace(Na1)
ans =
(exp(t*z1)*(M*kd2*kd1 + M*kd1*z1))/(z1*(z1 - z2)) - (exp(t*z2)*(M*kd2*kd1 + M*kd1*z2))/(z2*(z1 - z2)) + (M*kd2*kd1)/(z1*z2)
>> ilaplace(Na2)
ans =
(exp(t*z1)*(M*kd1*kd2 + M*kd2*z1))/(z1*(z1 - z2)) - (exp(t*z2)*(M*kd1*kd2 + M*kd2*z2))/(z2*(z1 - z2)) + (M*kd1*kd2)/(z1*z2)
```

Сл. 5.4 Решавање факторизованих Лагергренових једначина у комплексном домену помоћу Symbolic Toolbox, MATLAB – 2-компонентна смеша.



Сл. 5.5 Временске еволуције за број адсорбованих молекула према изразима у којима фигуришу експоненцијалне функције уместо хиперболичких

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MATLAB

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• ( *dsolve*)

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5.6

```

>> syms M k1 k2 kd1 kd2 N1 N2 eqn1 eqn2 N01 N02
>> eqn1='DN1=k1*(N0-N1)*(M-N1)-kd1*N1';
>> N1=dsolve(eqn1,'N1(0)=0')
N1 =
(kd1 + M*k1 + N0*k1 + tan(((t - (atan((kd1*i + M*k1*i + N0*k1*i)/(M^2*k1^2 - 2*M*N0*k1^2 +
2*M*kd1*k1 + N0^2*k1^2 + 2*N0*kd1*k1 + kd1^2)^(1/2))*i)/(M^2*k1^2 - 2*M*N0*k1^2 +
2*M*kd1*k1 + N0^2*k1^2 + 2*N0*kd1*k1 + kd1^2)^(1/2))*(M^2*k1^2 - 2*M*N0*k1^2 + 2*M*kd1*k1 +
N0^2*k1^2 + 2*N0*kd1*k1 + kd1^2)^(1/2)*i)/2)*(M^2*k1^2 - 2*M*N0*k1^2 + 2*M*kd1*k1 + N0^2*k1^2
+ 2*N0*kd1*k1 + kd1^2)^(1/2)*i)/(2*k1)
>> simplify(N1)
M/2 + N0/2 + kd1/(2*k1) + (tan(atan((kd1*i + M*k1*i + N0*k1*i)/(M^2*k1^2 - 2*M*N0*k1^2 +
2*M*kd1*k1 + N0^2*k1^2 + 2*N0*kd1*k1 + kd1^2)^(1/2)) + (t*(M^2*k1^2 - 2*M*N0*k1^2 +
2*M*kd1*k1 + N0^2*k1^2 + 2*N0*kd1*k1 + kd1^2)^(1/2)*i)/2)*(M^2*k1^2 - 2*M*N0*k1^2 +
2*M*kd1*k1 + N0^2*k1^2 + 2*N0*kd1*k1 + kd1^2)^(1/2)*i)/(2*k1)

```

Сл. 5.6 Рикатијева једначина решена помоћу Symbolic Toolbox, MATLAB 2012a

(3.9)

:

$$N_a(t) = \frac{N_0 M \left[ 1 - e^{-k_a(x-s)t} \right]}{x - s e^{-k_a(x-s)t}} \quad (5.1)$$

,

,

,

s

$$s = \frac{1}{2} \left[ \frac{k_d}{k_a} + N_0 + M - \sqrt{\left( \frac{k_d}{k_a} \right)^2 + (N_0 - M)^2 + 2 \frac{k_d}{k_a} (N_0 + M)} \right] \quad (5.2)$$

(

(5.1),

.  $N_0 \gg M$ ,

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(5.2)

[55]

$$s = \frac{1}{2} \left[ \frac{4N_0M}{\left( \frac{k_d}{k_a} + N_0 + M \right) + \sqrt{\left( \frac{k_d}{k_a} \right)^2 + (N_0 - M)^2 + 2 \frac{k_d}{k_a} (N_0 + M)}} \right] \quad (5.3)$$

,

## 5.2

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### 5.2.1 K

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5.7

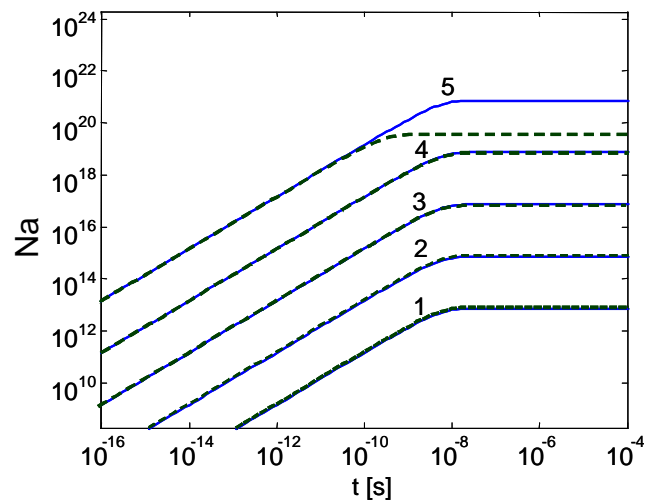
3 dm<sup>3</sup>,

1 mm<sup>2</sup>

50 kPa,

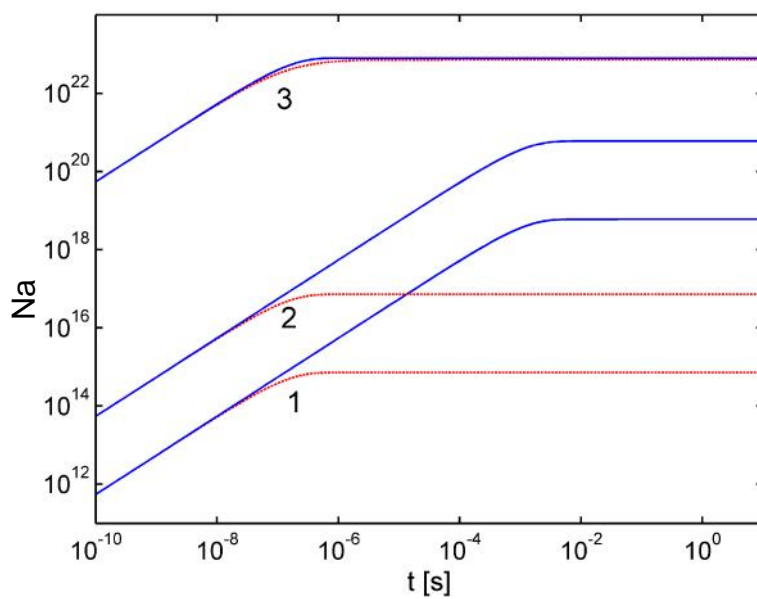
100 m<sup>2</sup>

dm<sup>3</sup>.



Сл. 5.7 Временска еволуција броја молекула угљен монооксида у плазмонском сензору са активном површином од злата. Пуне линије добијене су линеарним моделом, испрекидане нелинеарним. Парови линија обележени бројевима 1, 2, 3, 4 и 5 одговарају, редом, следећим активним површинама: 1mm<sup>2</sup>, 100mm<sup>2</sup>, 10<sup>4</sup> mm<sup>2</sup>, 1m<sup>2</sup> и 100m<sup>2</sup>. Притисак: 50 кРа, запремина 3 dm<sup>3</sup>, температура 300К

5.7, (5)  
 (5.7, .5)



Сл. 5.8 Временска зависност броја адсорбованих молекула бензена на активној површи од злата од  $10^4 \text{ m}^2$ , на собној температури, у комори од  $3 \text{ dm}^3$ , пуне линије: линеаран модел, испрекидане: нелинеаран у функцији од притиска. Парови линија 1, 2 и 3 одговарају притисцима од  $10^{-3} \text{ Pa}$ ,  $0.1 \text{ Pa}$  и  $10^5 \text{ Pa}$ , редом.

5.8.

$$1 N_0 = 7.25 \cdot 10^{14},$$

$$2 N_0 = 7.25 \cdot 10^{16},$$

$$3 N_0 = 7.25 \cdot 10^{22}.$$

$$= 6.25 \cdot 10^{22}.$$

:

(

:

5.9

$$6.25 \cdot 10^{14}$$

5.9

$$N_0 \quad 2.17 \cdot 10^{12},$$

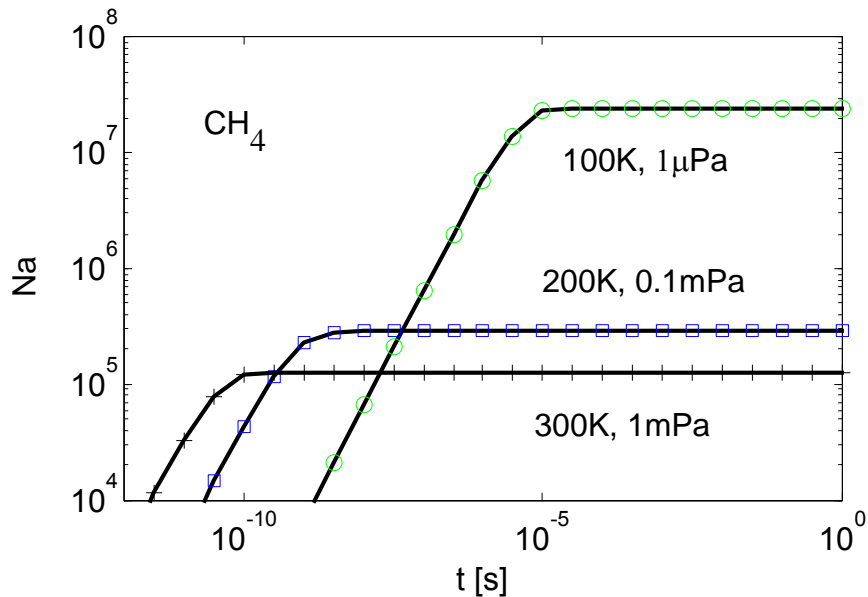
$$N_0 \quad 1.09 \cdot 10^{14},$$

+

$$N_0 \quad 7.25 \cdot 10^{14}.$$

5.9

( $N_0$ )



Сл. 5.9 Временска еволуција броја молекула метана адсорбованих на површину од злата са  $6.25 \cdot 10^{14}$  адсорпционих места у комори од  $3 \text{ dm}^3$ , рачуната према линеарном (пуне линије) и нелинеарном моделу (симболи) за различите притиске и температуре.



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$$N_0 \gg M, \quad (5.3)$$

$$S \cong \frac{1}{2} \left[ \frac{4N_0M}{\left(\frac{k_d}{k_a} + N_0\right) + \left(\frac{k_d}{k_a} + N_0\right)} \right] = \frac{k_a N_0 M}{k_d + k_a N_0} = \frac{k_l M}{k_d + k_l} \quad (5.4)$$

$$k_d/k_a$$

$$k_a N_0 M / k_d,$$

[55].

(3.13).

$$k_{II} = k_a \sqrt{\left(\frac{k_d}{k_a} + (N_0 + M)\right)^2 - 4N_0M} = k_a \sqrt{\left(\frac{k_d}{k_a}\right)^2 + (N_0 - M)^2 + 2\frac{k_d}{k_a}(N_0 + M)}$$

$$= k_a \sqrt{\left(\frac{k_d}{k_a}\right) \left[ \left(\frac{k_d}{k_a}\right) + 2(N_0 + M) \right] + (N_0 - M)^2} \quad (5.5)$$

$$N_0 \gg M, \quad (5.5)$$

$$\sqrt{k_d^2 + k_a^2 (N_0 - M)^2 + 2k_d k_a (N_0 + M)} \xrightarrow{N_0 \gg M} (k_d + k_a N_0) = k_d + k_l = k_L \quad (5.6)$$

$$, \quad k_d/k_a$$

$$, \quad k_d/k_a \gg 2(N_0 + M),$$

, a to je  $k_d$ .

	5.9.	5.9
	$N_0$	$2.17 \cdot 10^{12}, \quad k_d/k_a$
, $5.5 \cdot 10^{19}$ .		$N_0$
$k_d/k_a \quad 2.38 \cdot 10^{23}$ .	+	$1.09 \cdot 10^{14},$
$k_d/k_a \quad 3.56 \cdot 10^{24}$ .		$N_0 \quad 7.25 \cdot 10^{14},$
		$= 6.25 \cdot 10^{14},$

(  
(2.17) – (2.20)

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## 5.2.2

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w, je a (3.32), 3.

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$$\begin{aligned}
\ddagger_{\Delta n_{eff}}^2 &= w^2 \overline{N_a} \left\{ 1 - \frac{\overline{N_a}}{M} \right\}, & \ddagger_{\Delta n_{effL}}^2 &= w^2 \overline{N_{aL}} \left\{ 1 - \frac{\overline{N_{aL}}}{M} \right\} \\
u_{\Delta n_{eff}} &= \sqrt{\left( \frac{1}{\overline{N_a}} - \frac{1}{M} \right)}, & u_{\Delta n_{effL}} &= \sqrt{\left( \frac{1}{\overline{N_{aL}}} - \frac{1}{M} \right)}
\end{aligned} \tag{5.7}$$

$$N_0 \gg M$$

$$k_d/k_a$$

$$, k_d/k_a \gg 2(N_0 + M).$$

...)

$$N_0 \gg M \quad N_a \text{ je}$$

$$p \gg p_1 \quad p_1 = k_B T M / V \quad (5.8)$$

$$k_d/k_a \gg 2(N_0+M)$$

$$p \ll p_2 \quad p_2 = \frac{t_c \sqrt{f m k_B T / 2}}{r_s \dagger_0 e^{E_d / (RT)}} - \frac{k_B T M}{V} \quad (5.9)$$

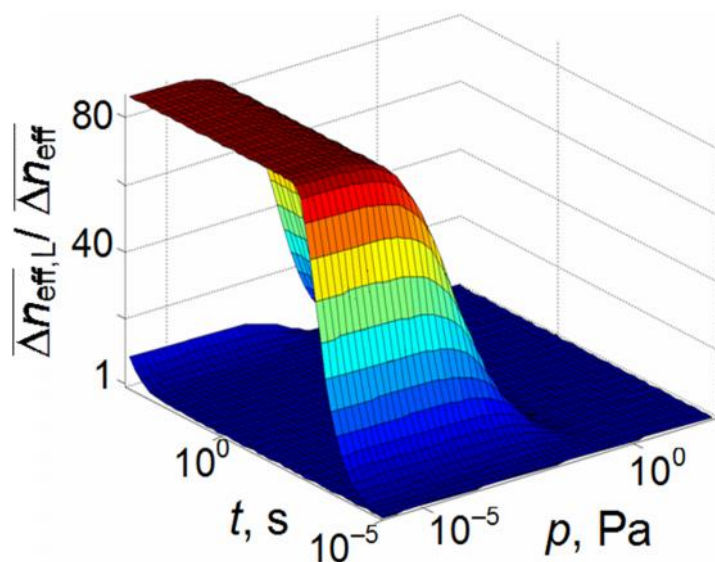
5.10

$$\begin{aligned} & \text{ : } = 100 \quad ( \quad ), \quad = 300 \\ & ( \quad ), \quad = 10^{-6} \text{ m}^2 \quad 3 \cdot 10^{-6} \text{ m}^3, \end{aligned}$$

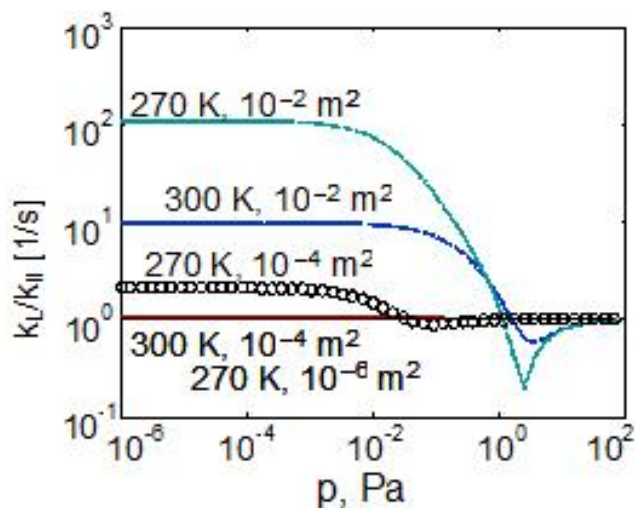
5.10

5.10

5.11



Сл. 5.10 Однос временских еволуција првог момента промене индекса преламања за линеарни и нелинеарни модел за распон притисака од  $10^{-6}$  до 100 Pa, активну површ од  $A = 10^{-6} \text{ m}^2$ , и запремину  $3 \cdot 10^{-6} \text{ m}^3$ . Шарени слој:  $T = 100 \text{ K}$ , плави слој  $T = 300 \text{ K}$ .



Сл. 5.11 Однос инверзних временских константи члановима прелазног одзива код линеарног и нелинеарног модела у зависности од притиска за различите температуре и активне површи сензора,  $V = 3 \cdot 10^{-6} \text{ m}^3$ .

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(5.7)

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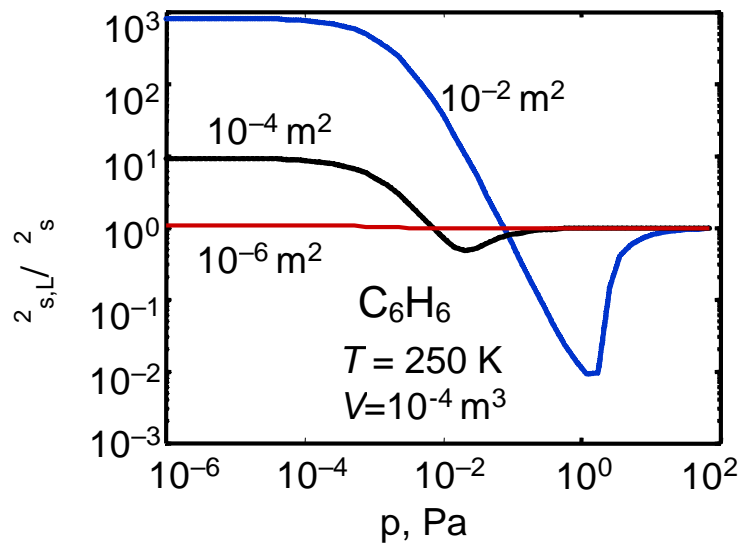
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Сл. 5.12 Однос стационарних вредности варијанси броја адсорбованих честица линеарног и нелинеарног модела.

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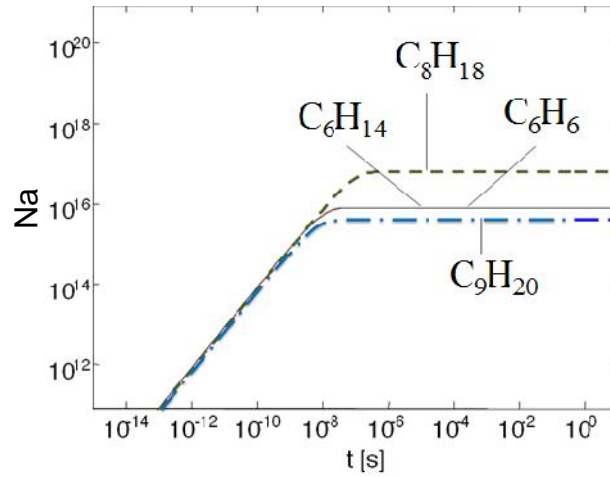
$10^{-6} \text{ m}^2$ .

### 5.3

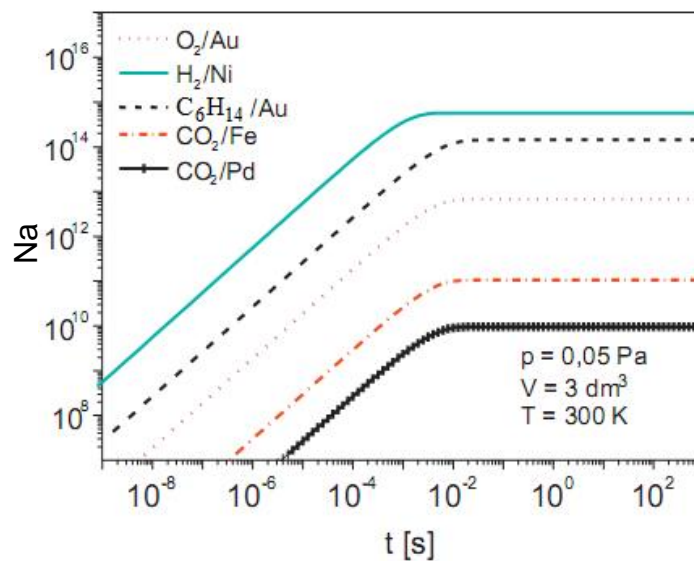
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[79].



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Сл. 5.13 Временска еволуција броја адсорбованих честица: површина  $10^{-3} \text{ m}^2(\text{Au})$ ,  $T = 300 \text{ K}$ ,  $V = 0.003 \text{ m}^3$ ,  $p = 0.5 \cdot 10^5 \text{ Pa}$ , за хексан (пуна линија), бензен (тачкаста, преклопљена са пуном), октан (испрекидана) и декан (црта-тачка).



Сл. 5.14 Временска еволуција броја адсорбованих молекула при монокомпонентној монослојној адсорпцији за 5 независних ситуација: црна испрекидана линија – хексан на злату, љубичаста тачкаста – кисеоник на злату, светло плава – водоник на никлу, црвена тачка-црта – угљен диоксид на гвожђу и црна са симболом | – угљен диоксид на паладијуму.

5.14

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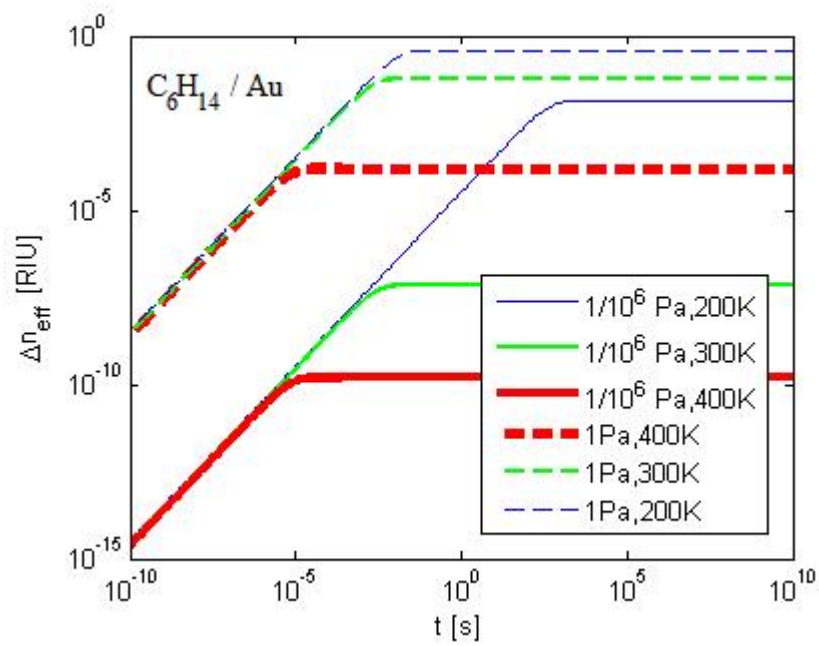
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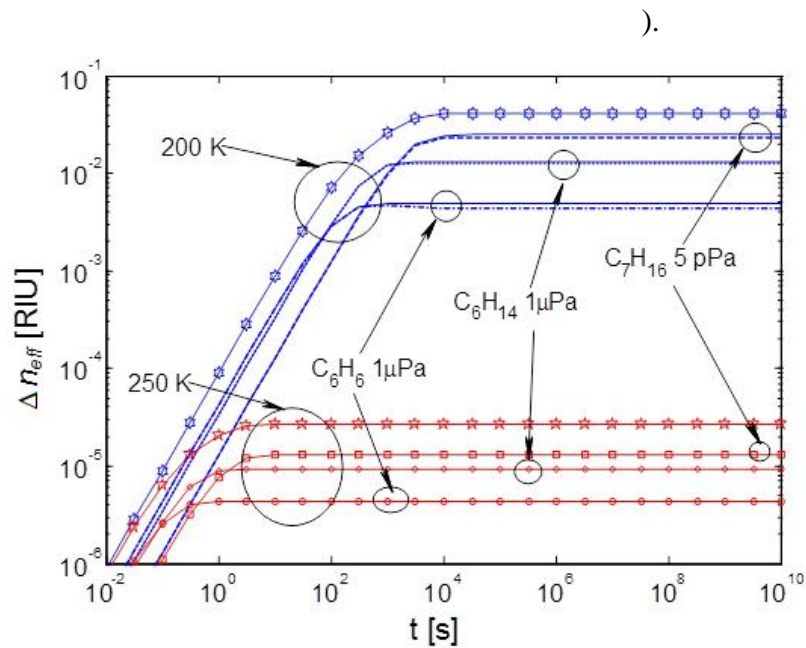
5.15 [80],

. RIU (refractive index unit).

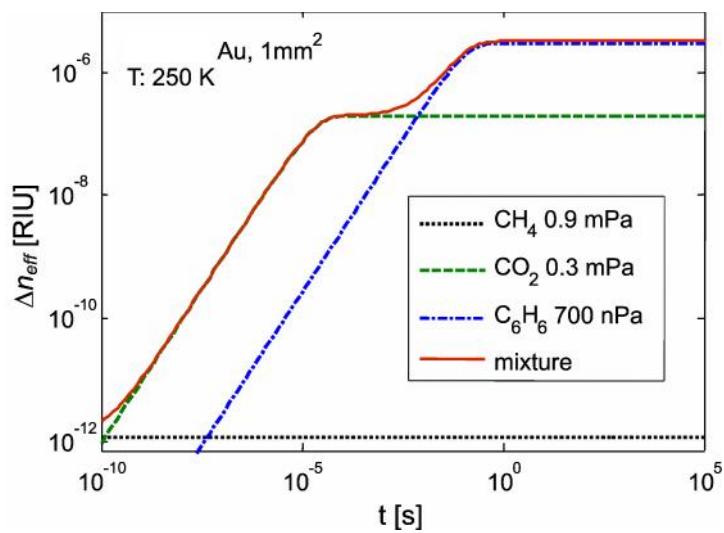


Сл. 5.15 Временска еволуција промене индекса преламања услед адсорпције хексана на злату за различите температуре и притиске.

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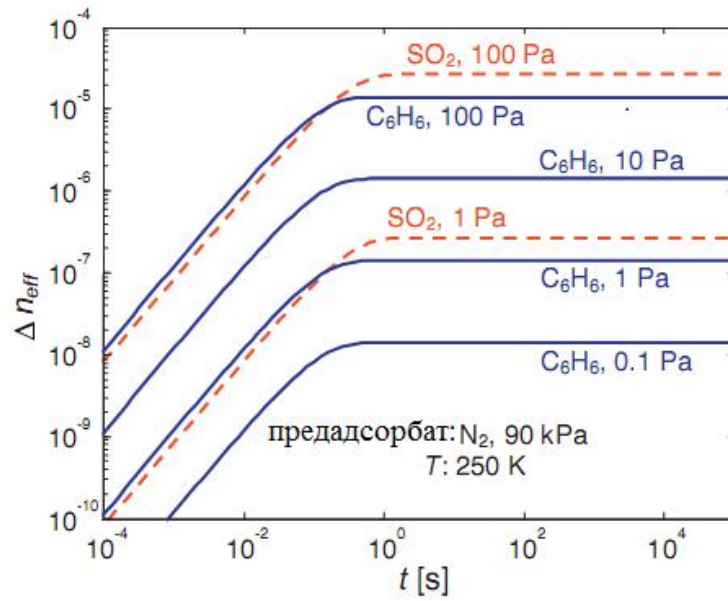
Сл. 5.16 Временска еволуција промене индекса преламања услед адсорпције трокомпонентне мешавине хексана, хептана и бензена на злату за различите температуре и притиске



Сл. 5.17 Временска еволуција промене индекса преламања услед адсорпције трокомпонентне мешавине метана, бензена и угљен диоксида на злату за различите температуре и притиске

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 / : 40% 50%. 5.17  
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 (90 kPa) ,  
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 $10^{-6} \text{ m}^2$ ,  $3 \cdot 10^{-6} \text{ m}^3$ .  
 1000 [17]  
 $10^{-8} \text{ RIU}$ .  
 5.18  
 ,  
 100 Pa 5.18.



Сл. 5.18 Временска еволуција промене индекса преламања услед адсорпције сумпор диоксида или бензена на злату, у ситуацији када је позадински гас азот на притиску 90 kPa

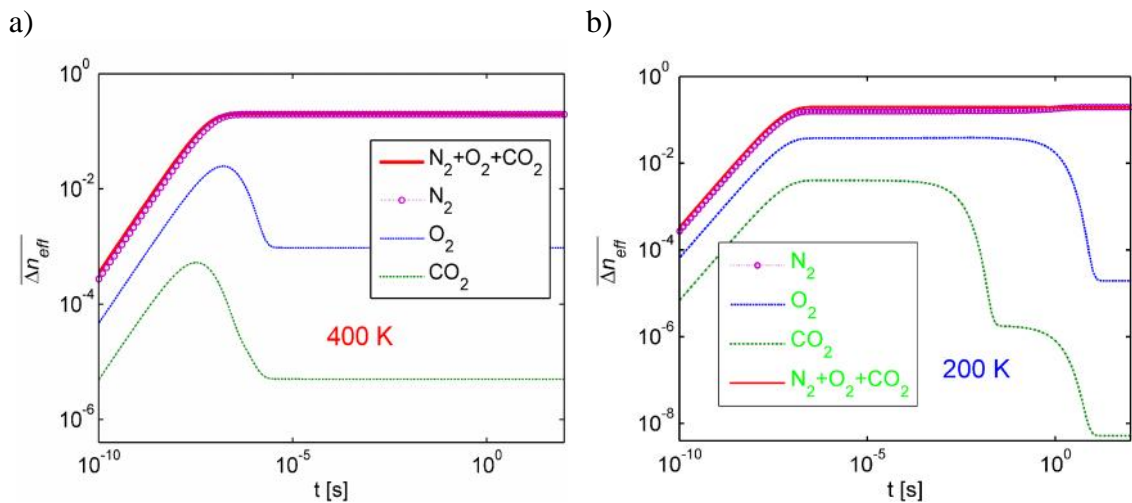
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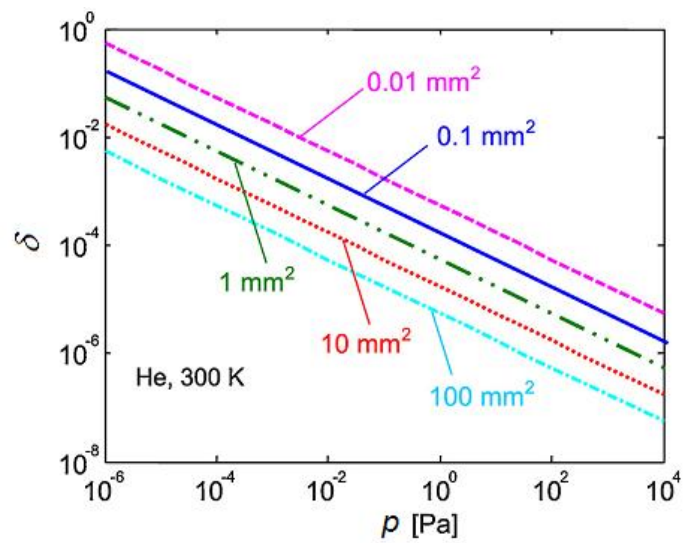
...

5.19

( : 2%  
 , 20% 78%  $10^5$  Pa)  
 (200 400 ).



Сл. 5.19 Средња вредност промене индекса преламања услед адсорпције синтетичке атмосфере (2%  $\text{CO}_2$ , 20%  $\text{O}_2$  и 78%  $\text{N}_2$  на  $10^5$  Pa) на злату за а)  $T = 200$  K и б)  $T = 400$  K. [81]

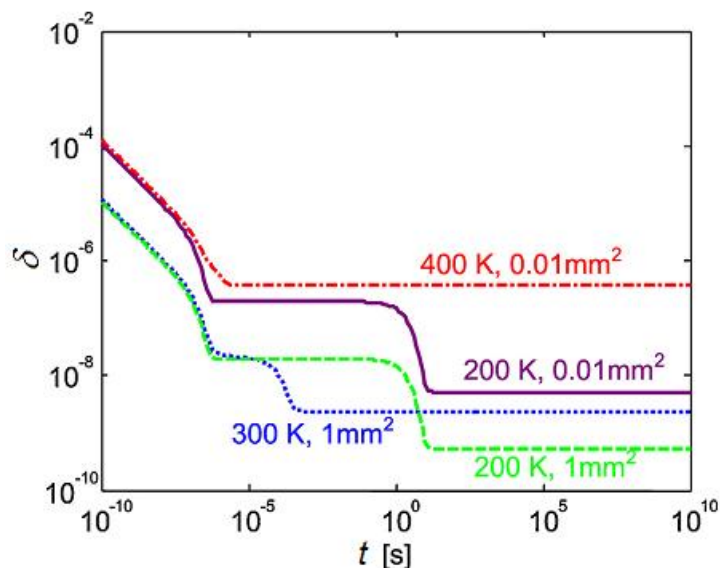


Сл. 5.20 Релативне флукуације промене индекса преламања у зависности од притиска приликом детекције хелијума помоћу сензора различитих активних површина при  $T = 300$  К

( 5.21,

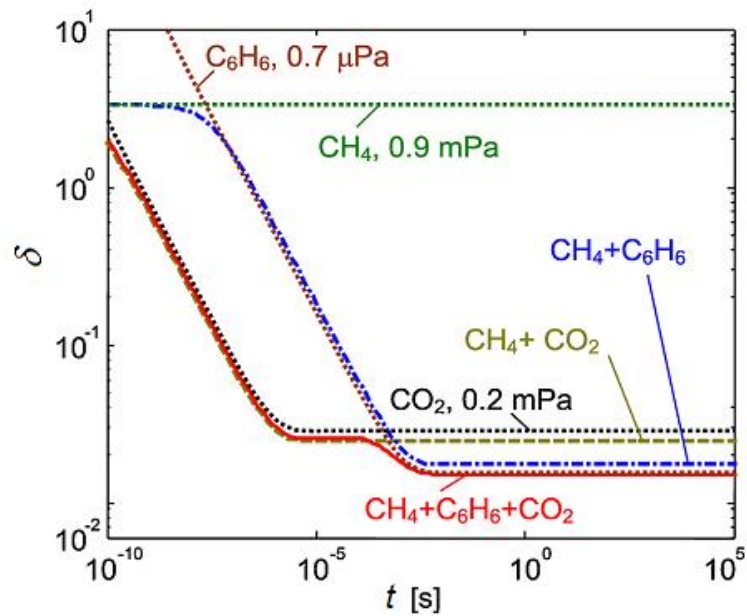


5.22.

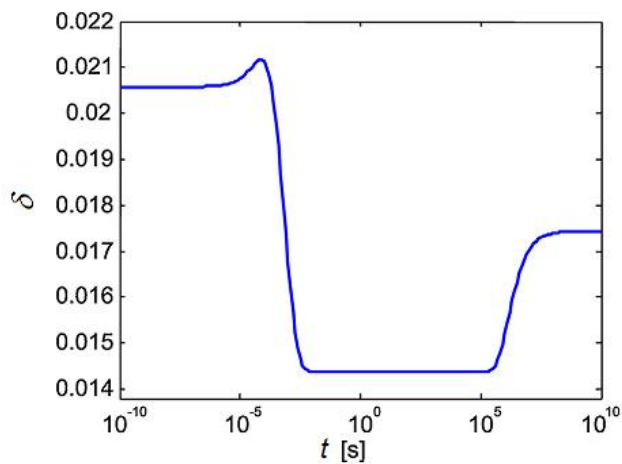


Сл. 5.21 Временска еволуција релативних флукуација промене индекса преламања услед адсорпције синтетичке атмосфере (2% CO<sub>2</sub>, 20% O<sub>2</sub>, 78%N<sub>2</sub>, 10<sup>5</sup> Pa) на златну површ за разне температуре и активне површине сензора

( 0.9 mPa)  
(700 nPa)



Сл. 5.22 Временска еволуција релативних флукуација промене индекса преламања услед адсорпције 3 гаса (метан, угљен диоксид, трагови бензена) у случајевима: када су сами (тачкасте криве: метан зелена, бензен тамноцрвена, угљен диоксид црна), када граде двокомпонентну мешавину (испрекидана браон за  $CH_4 + CO_2$ , црта-тачка плава за  $CH_4 + C_6H_6$ ) и када су сва три заједно (пуна црвена). Златна површ  $0.01 mm^2$ , собна температура



Сл 5.23 Временска еволуција релативних флукуација промене индекса преламања услед присуства угљен диоксида на  $0.3 mPa$  са траговима бензена ( $2.1 ppm$ )

5.23

(2.1 ppm)

5.23,

0.3 mPa

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$k_B T M / V$

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(  $k / k_d$  )

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 [82] – [89]. [56], [79],  
 [82] /  
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 [55]. [80]  
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 [89]

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[79] – [89]:

- [55] O. Jakšić, Ž. Čupić, Z. Jakšić, D. Randjelović, and L. Kolar-Anić, “Monolayer Gas Adsorption in Plasmonic Sensors: Comparative Analysis of Kinetic Models,” *Russ. J. Phys. Chem.*, vol. 87, no. 13, pp. 2134–2139, 2013.
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$$\frac{dN_a}{dt} = k_a(N_0 - N_a)(M_a - N_a) - k_d N_a \quad (.1)$$

$$\frac{dN_a}{dt} = k_a(N_0 - N_a)(M - N_a) - k_d N_a \quad (.2)$$

$$\begin{aligned} \frac{dN_a(t)}{dt} &= k_a(N_0 - N_a)(M - N_a) - k_d N_a \\ &= k_a N_a^2 - [k_a(N_0 + M) + k_d] N_a + k_a N_0 M \end{aligned} \quad (.3)$$

(Riccati)

2.24,

23,

2.106,

(J. A. Dean and N. A. Lange, *Lange's Handbook of Chemistry*, 15th ed.

McGraw-Hill, 1999):

$$\int \frac{dx}{a + 2bx + cx^2} = \frac{1}{2\sqrt{b^2 - ac}} \ln \frac{\sqrt{b^2 - ac} - b - cx}{\sqrt{b^2 - ac} + b + cx} + C \quad [b^2 - ac > 0] \quad (.4)$$

C

:

$$\int \frac{dx}{a + 2bx + cx^2} = \frac{1}{2\sqrt{b^2 - ac}} \ln \left( \frac{-cx + b - \sqrt{b^2 - ac}}{cx + b + \sqrt{b^2 - ac}} \right) \quad (.5)$$

$$\int \frac{dx}{a + 2bx + cx^2} = \frac{1}{\sqrt{4b^2 - 4ac}} \ln \left( \frac{-2cx + 2b - \sqrt{4b^2 - 4ac}}{2cx + 2b + \sqrt{4b^2 - 4ac}} \right) \quad (.6)$$

$$\int \frac{dx}{ax^2 + b + c} = \frac{1}{\sqrt{b^2 - 4ac}} \ln \left( -\frac{2ax + b - \sqrt{b^2 - 4ac}}{2ax + b + \sqrt{b^2 - 4ac}} \right) \quad (\text{A.7})$$

У изразу (A.7) се појављују коренови једначине  $ax^2 + bx + c = 0$

$$x_1 = \frac{-b + \sqrt{b^2 - 4ac}}{2a} \quad x_2 = \frac{-b - \sqrt{b^2 - 4ac}}{2a} \quad (\text{A.8})$$

Сада, (A.7) и (A.8) дају

$$\int \frac{dx}{ax^2 + b + c} = \frac{1}{a(x_1 - x_2)} \ln \left( -\frac{x - x_1}{x - x_2} \right) = \frac{1}{a(x_1 - x_2)} \ln \left( \frac{x_1 - x}{x - x_2} \right) \quad (\text{A.9})$$

Израз (A.9) користимо за решавање једначине (A.3) која је облика

$$\frac{dx}{dt} = ax^2 + bx + c \quad (\text{A.10})$$

Почевши од

$$\int \frac{dx}{ax^2 + b + c} = \int dt + C, \quad (\text{A.11})$$

применимо (A.9) и добије се

$$\frac{1}{a(x_1 - x_2)} \ln \left( \frac{x_1 - x}{x - x_2} \right) = t + C_0 \quad (\text{A.12})$$

Затим:

$$\ln \left( \frac{x_1 - x}{x - x_2} \right) = a(x_1 - x_2)(t + C_0) \quad (\text{A.13})$$

Константа  $C_0$  се добија из услова да за  $t = 0$ ,  $x$  такође мора бити 0.

$$C_0 = \frac{\ln(-x_1/x_2)}{a(x_1 - x_2)} \quad (\text{A.14})$$

Тако, 
$$\ln \frac{x_1 - x}{x - x_2} = a(x_1 - x_2)(t + C_0) \quad (\text{A.15})$$

$$x_1 - x = (x - x_2)e^{a(x_1 - x_2)(t + C_0)} = (x - x_2)e^{a(x_1 - x_2)t} e^{a(x_1 - x_2)C_0} \quad (.16)$$

$$(.14) \quad C_0.$$

$$x_1 - x = (x - x_2)e^{a(x_1 - x_2)(t + C_0)} = (x - x_2)e^{a(x_1 - x_2)t} e^{\frac{a(x_1 - x_2) \ln(-x_1/x_2)}{a(x_1 - x_2)}} = (x - x_2)e^{a(x_1 - x_2)t} \left( -\frac{x_1}{x_2} \right) \quad (.17)$$

$$(.17)$$

$$x_2 e^{-a(x_1 - x_2)t} \quad (.18)$$

$$x_2 (x_1 - x) e^{-a(x_1 - x_2)t} = (x_2 - x) x_1 \quad (.19)$$

$$x_2 x_1 e^{-a(x_1 - x_2)t} - x x_2 e^{-a(x_1 - x_2)t} = x_2 x_1 - x x_1 \quad (.20)$$

$$x (x_2 e^{-a(x_1 - x_2)t} - x_1) = x_2 x_1 (e^{-a(x_1 - x_2)t} - 1) \quad (.21)$$

$$x (x_2 e^{-a(x_1 - x_2)t} - x_1) = \frac{x_1 x_2 (1 - e^{-a(x_1 - x_2)t})}{x_1 - x_2 e^{-a(x_1 - x_2)t}} \quad (.22)$$

$$(.3) \quad (.10)$$

2

2

$x$	$a$	$b$	$c$
$N_a$	$k_a$	$-[k_a(N_0 + M) + k_d]$	$k_a N_0 M$

$$(.8)$$

$$x_1 = \frac{-b + \sqrt{b^2 - 4ac}}{2a} = \frac{1}{2} \left[ -\frac{b}{a} + \sqrt{\left(\frac{b}{a}\right)^2 - \frac{4c}{a}} \right]$$

$$x_2 = \frac{-b - \sqrt{b^2 - 4ac}}{2a} = \frac{1}{2} \left[ -\frac{b}{a} - \sqrt{\left(\frac{b}{a}\right)^2 - \frac{4c}{a}} \right], \quad (.23)$$

$$x = \frac{1}{2} \left[ \frac{k_d}{k_a} + N_0 + M + \sqrt{\left(\frac{k_d}{k_a} + N_0 + M\right)^2 - 4N_0M} \right] \quad (.24)$$

$$s = \frac{1}{2} \left[ \frac{k_d}{k_a} + N_0 + M - \sqrt{\left(\frac{k_d}{k_a} + N_0 + M\right)^2 - 4N_0M} \right] \quad (.25)$$

(Vieta)

$$\begin{aligned} x_1 x_2 &= \frac{1}{2} \left[ -\frac{b}{a} + \sqrt{\left(\frac{b}{a}\right)^2 - \frac{4c}{a}} \right] \frac{1}{2} \left[ -\frac{b}{a} - \sqrt{\left(\frac{b}{a}\right)^2 - \frac{4c}{a}} \right] = \\ &= \frac{1}{4} \left[ \left(-\frac{b}{a}\right)^2 - \left(\sqrt{\left(\frac{b}{a}\right)^2 - \frac{4c}{a}}\right)^2 \right] = \frac{1}{4} \left[ \left(\frac{b}{a}\right)^2 - \left(\left(\frac{b}{a}\right)^2 - \frac{4c}{a}\right) \right] = \frac{c}{a} \end{aligned} \quad (.26)$$

$$x_1 + x_2 = \frac{1}{2} \left[ -\frac{b}{a} + \sqrt{\left(\frac{b}{a}\right)^2 - \frac{4c}{a}} \right] + \frac{1}{2} \left[ -\frac{b}{a} - \sqrt{\left(\frac{b}{a}\right)^2 - \frac{4c}{a}} \right] = -\frac{b}{a}$$

1, (.26)

$$x_1 x_2 = \frac{c}{a} = N_0 M \quad (.27)$$

$$x_1 + x_2 = -\frac{b}{a} = \frac{k_d}{k_a} + N_0 + M$$

x and s,

$$N_a(t) = \frac{N_0 M \left[ 1 - e^{-k_a(x-s)t} \right]}{x - s e^{-k_a(x-s)t}} \quad (.28)$$

(.27)

s



$$S = \frac{N_0 M}{\frac{1}{2} \left[ \frac{k_d}{k_a} + N_0 + M + \sqrt{\left( \frac{k_d}{k_a} + N_0 + M \right)^2 - 4N_0 M} \right]} \quad (.29)$$

(.29)

$$\frac{dN_a}{dt} = k_L (N_{a,\infty} - N_a) = k_L N_{a,\infty} - k_L N_a. \quad (.1)$$

(W. E. Boyce and R. C. Diprima, *Elementary Differential Equations and Boundary Value Problems*, John Wiley & Sons, Incorporated, 2001.):

$$\frac{dy}{dx} + P(x)y = Q(x). \quad (.2)$$

$$y = e^{-\int P(x)dx} \int e^{\int P(x)dx} Q(x) dx. \quad (.3)$$

$$P = -k_L, \quad Q = k_L N_{a,\infty}.$$

$$e^{-\int k_L dt} = e^{k_L t + c_1} = c_2 e^{k_L t}. \quad (.4)$$

$$(c_2 = 1)$$

$$e^{k_L t} \frac{dN_a}{dt} + e^{k_L t} (k_l + k_d) N_a = e^{k_L t} k_L N_{a,\infty}. \quad (.5)$$

:

$$\frac{d}{dt} [e^{k_L t} N_a] = e^{k_L t} k_L N_{a,\infty} \quad (.6)$$

$$\int \frac{d}{dt} [e^{k_L t} N_a] = \int e^{k_L t} k_L N_{a,\infty} dt \quad (.7)$$

$$e^{k_L t} N_a = \frac{k_L N_{a,\infty}}{k_L} e^{k_L t} + c_3 \quad (.8)$$

$$N_a(t) = N_{a,\infty} + c_3 e^{-k_L t} \quad (.9)$$

$$c_3 \quad N_a(0) = 0, \quad :$$

$$\boxed{N_a(t) = N_{a,\infty} (1 - e^{-k_L t})} \quad (.10)$$

(.10)

$$N_a(t \rightarrow \infty) = N_{a,\infty}.$$

$$\frac{dN_a}{dt} = k_l (M - N_a) - k_d N_a = k_l M - (k_l + k_d) N_a. \quad (.11)$$

$$P \quad Q, \quad (k_l + k_d), a \quad Q \quad k_l M. \quad P$$

$$N_a(t) = \frac{k_l M}{k_l + k_d} (1 - e^{-(k_l + k_d)t}) \quad (.12)$$

(.10) (.12), je

$$\boxed{N_{a,\infty} = \frac{k_l M}{(k_l + k_d)}} \quad (.13)$$

**(Laplace)**

(Pierre Simon

Laplace, 1749-1827)

,  $f(t)$ ,  
 $F(s)$

$$F(s) = \mathcal{L}\{f(t)\} = \int_0^{\infty} e^{-st} f(t) dt \quad (s = \sigma_0 + i\omega, \sigma_0 > 0, t \geq 0)$$

$f(t)$ .  $f$   $F(s)$  je " " " " "  
 $[0, +\infty)$ .  $F$   $t < 0$

e o o e o

e a a:

$$\mathcal{L}^{-1}\{F(s)\} = \frac{1}{2\pi i} \int_{\sigma_0 - i\infty}^{\sigma_0 + i\infty} e^{st} F(s) ds \quad \sigma_0 > s_0$$

.  $s_0$  je

$F(s)$ .

### **Линеарност**

$$\mathcal{L}\left\{\sum_{k=1}^n a_k f_k(t)\right\} = \sum_{k=1}^n a_k F_k(s)$$

### **Сличност**

$$a > 0, \quad \mathcal{L}\{f(at)\} = \frac{1}{a} F\left(\frac{s}{a}\right), \quad \mathcal{L}\{f(t)\} = F(s)$$

### **Диференцирање оригинала**

$$a > 0 \quad \mathcal{L}\{f(t)\} = F(s), \quad \mathcal{L}\{f'(t)\} = sF(s) - f(0)$$

### **Диференцирање слике**

$$\text{А о} \quad \mathcal{L}\{f(t)\} = F(s), \quad \mathcal{L}\{tf(t)\} = -F'(s).$$

$$\mathcal{L}\{t^n f(t)\} = (-1)^n F^{(n)}(s)$$

### **Теорема померања**

$$\mathcal{L}\{e^{s_0 t} f(t)\} = F(s - s_0)$$

### **Теорема кашњења**

$$\mathcal{L}\{f(t - \dagger)\} = -e^{s\dagger} F(s), \quad \dagger \geq 0$$

### **Лапласова трансформација конволуције функција**

$$\mathcal{L}\{f * g\} = \mathcal{L}\{f\} \mathcal{L}\{g\}$$

$$(f * g)(x) = \int_{-\infty}^{+\infty} f(x-t)g(t)dt = \int_{-\infty}^{+\infty} f(t)g(x-t)dt$$

$x(t) = \mathcal{L}^{-1}\{X(s)\}$	$X(s) = \mathcal{L}\{x(t)\}$
$u(t - \dagger)$	$e^{-\dagger s}$
$u(t)$	1
$\frac{(t - \dagger)^n}{n!} e^{-r(t - \dagger)} u(t - \dagger)$	$\frac{e^{-\dagger s}}{(s + r)^{n+1}}$
$\frac{t^n}{n!} u(t)$	$\frac{1}{s^{n+1}}$
$\frac{t^q}{\Gamma(q+1)} u(t)$	$\frac{1}{s^{q+1}}$
$u(t)$	$\frac{1}{s}$
$u(t - \dagger)$	$\frac{e^{-\dagger s}}{s}$
$t \cdot u(t)$	$\frac{1}{s^2}$
$\frac{t^n}{n!} e^{-rt} u(t)$	$\frac{1}{(s + r)^{n+1}}$
$e^{-rt} u(t)$	$\frac{1}{s + r}$
$(1 - e^{-rt}) u(t)$	$\frac{r}{s(s + r)}$
$\sin(\check{S}t) u(t)$	$\frac{s}{s^2 + \check{S}^2}$

$\sin(\check{S}t)u(t)$	$\frac{\check{S}}{s^2 + \check{S}^2}$
$\sinh(rt)u(t)$	$\frac{r}{s^2 - r^2}$
$\cosh(rt)u(t)$	$\frac{s}{s^2 - r^2}$
$e^{\check{r}t} \sin(\check{S}t)u(t)$	$\frac{\check{S}}{(s - \check{r})^2 + \check{S}^2}$
$e^{\check{r}t} \cos(\check{S}t)u(t)$	$\frac{\check{S}}{(s - \check{r})^2 + \check{S}^2}$
$t^{n-1}u(t)$	$s^{-(n+1)/n} \Gamma\left(1 + \frac{1}{n}\right)$
$\ln\left(\frac{t}{t_0}\right)u(t)$	$-\frac{t_0}{s} [\ln(t_0 s) + \chi]$
$J_n(\check{S}t)u(t)$ (Bessel)	$\frac{\check{S}^n \left(s + \sqrt{s^2 + \check{S}^2}\right)^{-n}}{\sqrt{s^2 + \check{S}^2}}$
$I_n(\check{S}t)u(t)$ (Bessel)	$\frac{\check{S}^n \left(s + \sqrt{s^2 - \check{S}^2}\right)^{-n}}{\sqrt{s^2 - \check{S}^2}}$
$Y_0(rt)u(t)$ (Bessel)	$-\frac{2 \sinh^{-1}(s/r)^{-n}}{f \sqrt{s^2 + \check{S}^2}}$
$\operatorname{erf}(t)u(t)$	$\frac{e^{s^2/4} (1 - \operatorname{erf}(s/2))}{s}$

(Heaviside)

$$\begin{aligned}
F(s_1, s_2, t) &= [1 + G_1(s_1 - 1) + G_2(s_2 - 1)]^M = [(1 - G_1 - G_2) + G_1s_1 + G_2s_2]^M \\
&= \sum_{N_{a1}=0}^M \sum_{N_{a2}=0}^{M-N_{a1}} \binom{M}{N_{a1}, N_{a2}} (1 - G_1 - G_2)^{M-N_{a1}-N_{a2}} (G_1s_1)^{N_{a1}} (G_2s_2)^{N_{a2}} \quad , ( .1)
\end{aligned}$$

$$P_{N_{a1}, N_{a2}}(t) = \frac{M!}{N_{a1}!N_{a2}!(M - N_{a1} - N_{a2})!} (1 - G_1 - G_2)^{M-N_{a1}-N_{a2}} G_1^{N_{a1}} G_2^{N_{a2}}. \quad ( .2)$$

$$\begin{aligned}
\frac{dP_{N_{a1}, N_{a2}}(t)}{dt} &= \frac{M!}{N_{a1}!N_{a2}!(M - N_{a1} - N_{a2})!} \\
&\left\{ (1 - G_1 - G_2)^{M-N_{a1}-N_{a2}} N_{a1} G_1^{N_{a1}-1} \frac{dG_1}{dt} G_2^{N_{a2}} \right. \\
&\quad \left. + (1 - G_1 - G_2)^{M-N_{a1}-N_{a2}} G_1^{N_{a1}} N_{a2} G_2^{N_{a2}-1} \frac{dG_2}{dt} \right. \\
&\quad \left. + G_1^{N_{a1}} G_2^{N_{a2}} (M - N_{a1} - N_{a2}) (1 - G_1 - G_2)^{M-N_{a1}-N_{a2}-1} \left( -\frac{dG_1}{dt} - \frac{dG_2}{dt} \right) \right\} \quad ( .3)
\end{aligned}$$

( .3)

( .2).

$$\frac{dP_{N_{a1}, N_{a2}}(t)}{dt} = P_{N_{a1}, N_{a2}}(t) \left\{ \frac{N_{a1}}{G_1} \frac{dG_1}{dt} + \frac{N_{a2}}{G_2} \frac{dG_2}{dt} - \frac{(M - N_{a1} - N_{a2})}{(1 - G_1 - G_2)} \left( \frac{dG_1}{dt} + \frac{dG_2}{dt} \right) \right\} \quad ( .4)$$

-

( .2).

$$\begin{aligned}
P_{N_{a1}-1, N_{a2}}(t) &= \\
&\frac{M!}{(N_{a1}-1)!N_{a2}!(M - N_{a1} + 1 - N_{a2})!} (1 - G_1 - G_2)^{M-N_{a1}+1-N_{a2}} G_1^{N_{a1}-1} G_2^{N_{a2}}
\end{aligned}$$

$$= \frac{N_{a1}(1-G_1-G_2)}{(M-N_{a1}+1-N_{a2})G_1} P_{N_{a1},N_{a2}}(t) \quad (.5)$$

$$P_{N_{a1},N_{a2}-1}(t) = \frac{N_{a2}(1-G_1-G_2)}{(M-N_{a1}+1-N_{a2})G_2} P_{N_{a1},N_{a2}}(t) \quad (.6)$$

$$P_{N_{a1}+1,N_{a2}}(t) = \frac{M!(1-G_1-G_2)^{M-N_{a1}-1-N_{a2}} G_1^{N_{a1}+1} G_2^{N_{a2}}}{(N_{a1}+1)!N_{a2}!(M-N_{a1}-1-N_{a2})!} \\ = \frac{G_1(M-N_{a1}-N_{a2})}{(1-G_1-G_2)(N_{a1}+1)} P_{N_{a1},N_{a2}}(t) \quad (.7)$$

$$P_{N_{a1},N_{a2}+1}(t) = \frac{G_2(M-N_{a1}-N_{a2})}{(1-G_1-G_2)(N_{a2}+1)} P_{N_{a1},N_{a2}}(t) \quad (.8)$$

$$\frac{dP_{N_{a1},N_{a2}}(t)}{dt} = P_{N_{a1}-1,N_{a2}}(t)k_{l1}(M-(N_{a1}-1)-N_{a2}) \\ + P_{N_{a1},N_{a2}-1}(t)k_{l2}(M-(N_{a1}-1)-N_{a2}) \\ + P_{N_{a1}+1,N_{a2}}(t)k_{d1}(N_{a1}+1) + P_{N_{a1},N_{a2}+1}(t)k_{d2}(N_{a2}+1) \\ - P_{N_{a1},N_{a2}}(t)(k_{d1}N_{a1} + k_{l1}(M-N_{a1}-N_{a2}) + k_{d2}N_{a2} + k_{l2}(M-N_{a1}-N_{a2})) \quad (.9)$$

(.3)-(.8) ( .9).

$$P_{N_{a1},N_{a2}}(t) \left\{ \frac{N_{a1}}{G_1} \frac{dG_1}{dt} + \frac{N_{a2}}{G_2} \frac{dG_2}{dt} - \frac{(M-N_{a1}-N_{a2})}{(1-G_1-G_2)} \left( \frac{dG_1}{dt} + \frac{dG_2}{dt} \right) \right\} = \\ k_{l1}(M-(N_{a1}-1)-N_{a2}) \frac{N_{a1}(1-G_1-G_2)}{(M-N_{a1}+1-N_{a2})G_1} P_{N_{a1},N_{a2}}(t) \\ + k_{l2}(M-N_{a1}-(N_{a2}-1)) \frac{N_{a2}(1-G_1-G_2)}{(M-N_{a1}+1-N_{a2})G_2} P_{N_{a1},N_{a2}}(t) \quad (.10)$$

$$+ k_{d1}(N_{a1}+1) \frac{G_1(M-N_{a1}-N_{a2})}{(1-G_1-G_2)(N_{a1}+1)} P_{N_{a1},N_{a2}}(t) \\ + k_{d2}(N_{a2}+1) \frac{G_2(M-N_{a1}-N_{a2})}{(1-G_1-G_2)(N_{a2}+1)} P_{N_{a1},N_{a2}}(t) \\ - (k_{d1}N_{a1} + k_{l1}(M-N_{a1}-N_{a2}) + k_{d2}N_{a2} + k_{l2}(M-N_{a1}-N_{a2})) P_{N_{a1},N_{a2}}(t)$$

(.10) :

$$\left\{ \left( \frac{N_{a1}}{G_1} - \frac{(M-N_{a1}-N_{a2})}{(1-G_1-G_2)} \right) \frac{dG_1}{dt} + \left( \frac{N_{a2}}{G_2} - \frac{(M-N_{a1}-N_{a2})}{(1-G_1-G_2)} \right) \frac{dG_2}{dt} \right\} =$$



$$\begin{aligned}
& k_{l1} \left\{ \frac{N_{a1}(1-G_1-G_2)}{G_1} - (M - N_{a1} - N_{a2}) \right\} \\
& + k_{l2} \left\{ \frac{N_{a2}(1-G_1-G_2)}{G_2} - (M - N_{a1} - N_{a2}) \right\} \\
& + k_{d1} \left\{ \frac{G_1(M - N_{a1} - N_{a2})}{(1-G_1-G_2)} - N_{a1} \right\} + k_{d2} \left\{ \frac{G_2(M - N_{a1} - N_{a2})}{(1-G_1-G_2)} - N_{a2} \right\}
\end{aligned} \tag{.11}$$

$$\begin{aligned}
& \left( \frac{N_{a1}}{G_1} - \frac{(M - N_{a1} - N_{a2})}{(1-G_1-G_2)} \right) \frac{dG_1}{dt} = \\
& k_{l1}(1-G_1-G_2) \left\{ \frac{N_{a1}}{G_1} - \frac{(M - N_{a1} - N_{a2})}{(1-G_1-G_2)} \right\} - k_{d1}G_1 \left\{ \frac{N_{a1}}{G_1} - \frac{(M - N_{a1} - N_{a2})}{(1-G_1-G_2)} \right\}
\end{aligned} \tag{.12}$$

$$\begin{aligned}
& \left( \frac{N_{a2}}{G_2} - \frac{(M - N_{a1} - N_{a2})}{(1-G_1-G_2)} \right) \frac{dG_2}{dt} = \\
& k_{l2}(1-G_1-G_2) \left\{ \frac{N_{a2}}{G_2} - \frac{(M - N_{a1} - N_{a2})}{(1-G_1-G_2)} \right\} - k_{d2}G_2 \left\{ \frac{N_{a2}}{G_2} - \frac{(M - N_{a1} - N_{a2})}{(1-G_1-G_2)} \right\}
\end{aligned} \tag{.13}$$

(.12) (.13)

$$\begin{aligned}
\frac{dG_1}{dt} &= k_{l1}(1-G_1-G_2) - k_{d1}G_1 \\
\frac{dG_2}{dt} &= k_{l2}(1-G_1-G_2) - k_{d2}G_2
\end{aligned} \tag{.14}$$

(.14)

$M,$

:

$$\frac{dN_{a,i}}{dt} = k_{l,i} \left( M - \sum_{j=1}^r N_{a,j} \right) - k_{d,i} N_{a,i} \quad i=1,2 \tag{.15}$$

(.1)

(  
).

$$\begin{aligned} \frac{dF(s_1, s_2, t)}{dt} &= \underline{M} \left[ \underline{k}_{l,1}(s_1 - 1) + \underline{k}_{l,2}(s_2 - 1) \right] F(s_1, s_2, t) \\ &+ \left[ \underline{k}_{l,1} s_1(1 - s_1) + \underline{k}_{l,2} s_1(1 - s_2) + k_{d1}(1 - s_1) \right] \frac{\partial F(s_1, s_2, t)}{\partial s_1} \quad (.1) \\ &+ \left[ \underline{k}_{l,1} s_2(1 - s_1) + \underline{k}_{l,2} s_2(1 - s_2) + k_{d2}(1 - s_2) \right] \frac{\partial F(s_1, s_2, t)}{\partial s_2} \end{aligned}$$

(.1) 1.

$$\begin{aligned} \frac{d}{dt} \frac{\partial F(s_1, s_2, t)}{\partial s_1} &= \underline{M} \left[ \underline{k}_{l1}(s_1 - 1) + \underline{k}_{l2}(s_2 - 1) \right] \frac{\partial F(s_1, s_2, t)}{\partial s_1} + \underline{M} \underline{k}_{l1} F(s_1, s_2, t) \\ &+ \left[ \underline{k}_{l1} s_1(1 - s_1) + \underline{k}_{l2} s_1(1 - s_2) + k_{d1}(1 - s_1) \right] \frac{\partial^2 F(s_1, s_2, t)}{\partial s_1^2} \quad (.2) \\ &+ \left[ \underline{k}_{l1} s_2 - 2\underline{k}_{l1} s_1 + \underline{k}_{l2} s_2(1 - s_2) - k_{d1} \right] \frac{\partial F(s_1, s_2, t)}{\partial s_1} \\ &+ \left[ \underline{k}_{l1} s_2(1 - s_1) + \underline{k}_{l2} s_2(1 - s_2) + k_{d2}(1 - s_2) \right] \frac{\partial^2 F(s_1, s_2, t)}{\partial s_2 \partial s_1} - \underline{k}_{l1} s_2 \frac{\partial F(s_1, s_2, t)}{\partial s_2} \end{aligned}$$

$$1 = 2 = 1$$

$$\begin{aligned} \frac{d\overline{N_{a1}}}{dt} &= \\ 0 \cdot \overline{N_{a1}} + \underline{M} \underline{k}_{l1} + 0 \cdot \frac{\partial^2 F(s_1, s_2, t)}{\partial s_1^2} + \left[ \underline{k}_{l1} s_2 - 2\underline{k}_{l1} s_1 - k_{d1} \right] \overline{N_{a1}} + 0 \cdot \frac{\partial^2 F(s_1, s_2, t)}{\partial s_2 \partial s_1} - \underline{k}_{l1} s_2 \overline{N_{a2}} \\ &= \underline{M} \underline{k}_{l1} - \left( \underline{k}_{l1} s_2 + k_{d1} \right) \overline{N_{a1}} - \underline{k}_{l1} s_2 \overline{N_{a2}} \end{aligned}$$

(.3)

( .1) 2:

$$\begin{aligned}
 \frac{d}{dt} \frac{\partial F(s_1, s_2, t)}{\partial s_2} &= \underline{M} \left[ \underline{k}_{l1}(s_1 - 1) + \underline{k}_{l2}(s_2 - 1) \right] \frac{\partial F(s_1, s_2, t)}{\partial s_2} + \underline{M} \underline{k}_{l2} F(s_1, s_2, t) \\
 &+ \left[ \underline{k}_{l1} \underline{s}_1 (1 - s_1) + \underline{k}_{l2} \underline{s}_1 (1 - s_{a2}) + k_{d1}(1 - s_1) \right] \frac{\partial^2 F(s_1, s_2, t)}{\partial s_1 \partial s_2} - \underline{k}_{l2} \underline{s}_1 \frac{\partial F(s_1, s_2, t)}{\partial s_1} \quad ( .4) \\
 &+ \left[ \underline{k}_{l1} \underline{s}_2 (1 - s_1) + \underline{k}_{l2} \underline{s}_2 (1 - s_2) + k_{d2}(1 - s_2) \right] \frac{\partial^2 F(s_1, s_2, t)}{\partial s_2^2} \\
 &+ \left[ \underline{k}_{l1} \underline{s}_1 (1 - s_1) + \underline{k}_{l2} \underline{s}_1 - 2\underline{k}_{l2} \underline{s}_2 - k_{d2} \right] \frac{\partial F(s_1, s_2, t)}{\partial s_2}
 \end{aligned}$$

$$\underline{s}_1 = \underline{s}_2 = 1$$

$$\begin{aligned}
 \frac{d \overline{N_{a2}}}{dt} &= \\
 0 \cdot \overline{N_{a2}} + \underline{M} \underline{k}_{l2} + 0 \cdot \frac{\partial^2 F(s_1, s_2, t)}{\partial s_1 \partial s_2} - \underline{k}_{l2} \underline{s}_2 \overline{N_{a1}} + 0 \cdot \frac{\partial^2 F(s_1, s_2, t)}{\partial s_2^2} + \left[ \underline{k}_{l2} \underline{s}_1 - 2\underline{k}_{l2} \underline{s}_2 - k_{d2} \right] \overline{N_{a2}} \\
 &= \underline{M} \underline{k}_{l2} - \underline{k}_{l2} \underline{s}_2 \overline{N_{a1}} - \left( \underline{k}_{l2} \underline{s}_1 + k_{d2} \right) \overline{N_{a2}} \quad ( .5)
 \end{aligned}$$

( .3) ( .5)

$$\begin{aligned}
 \frac{d \overline{N_{a1}}}{dt} &= \underline{k}_{l1} \left( \underline{M} - \underline{s}_2 \overline{N_{a1}} - \underline{s}_1 \overline{N_{a2}} \right) - k_{d1} \overline{N_{a1}} \\
 \frac{d \overline{N_{a2}}}{dt} &= \underline{k}_{l2} \left( \underline{M} - \underline{s}_2 \overline{N_{a1}} - \underline{s}_1 \overline{N_{a2}} \right) - k_{d2} \overline{N_{a2}} \quad ( .6)
 \end{aligned}$$

:

$$\frac{\partial F(s,t)}{\partial t} = Mk_l(s-1)F(s,t) + (k_d + (k_l - k_d)s - k_l s^2) \frac{\partial F(s,t)}{\partial s} \quad (.1)$$

$$\frac{d}{dt} \left( \frac{\partial F(s,t)}{\partial s} \right) = \frac{\partial}{\partial s} \left( k_d + (k_l - k_d)s - k_l s^2 \right) \cdot \frac{\partial F(s,t)}{\partial s} + (k_d + (k_l - k_d)s - k_l s^2) \frac{\partial^2 F(s,t)}{\partial s^2} + k_l M \frac{\partial}{\partial s} (s-1) \cdot F(s,t) + k_l M (s-1) \frac{\partial F(s,t)}{\partial s} \quad (.2)$$

$$\frac{d\overline{N}_a}{dt} = ((k_l - k_d) - 2k_l) \cdot \overline{N}_a + 0 \cdot \frac{\partial^2 F(s,t)}{\partial s^2} \Big|_{s=1} + k_l M F(s,t) \Big|_{s=1} + 0 \quad (.3)$$

(PGF) = 1 1

$$\frac{d\overline{N}_a}{dt} = -(k_l + k_d) \cdot \overline{N}_a + k_l M \quad (.4)$$

$$y' + p(x)y = q(x) \quad (.5)$$

$$y = e^{-\int p(x)dx} \left( c + \int q(x) e^{\int p(x)dx} dx \right) \quad (.6)$$

$$y = e^{-pt} \left( c + \int q e^{pt} dt \right) = e^{-pt} \left( c + \frac{q}{p} e^{pt} \right) = c e^{-pt} + \frac{q}{p} \quad (.7)$$

:  $t = 0,$

$$0 = c + \frac{q}{p} \quad c = -\frac{q}{p} \quad y = \frac{q}{p}(1 - e^{-pt}) \quad (.8)$$

$$\overline{N}_a = \frac{k_l M}{k_l + k_d} (1 - e^{-(k_l + k_d)t}) \quad (.9)$$

:

$$\frac{\partial F(s,t)}{\partial t} = (k_d + (k_l - k_d)s - k_l s^2) \frac{\partial F(s,t)}{\partial s} + k_l M (s-1) F(s,t) \quad (.10)$$

:

$$\begin{aligned} \frac{d}{dt} \left( \frac{\partial F(s,t)}{\partial s} \right) = & \\ & ((k_l - k_d) - 2k_l s) \cdot \frac{\partial F(s,t)}{\partial s} + (k_d + (k_l - k_d)s - k_l s^2) \frac{\partial^2 F(s,t)}{\partial s^2} \quad (.11) \\ & + k_l M \cdot F(s,t) + k_l M (s-1) \frac{\partial F(s,t)}{\partial s} \end{aligned}$$

, (.2)

$$\begin{aligned} \frac{d}{dt} \left( \frac{\partial^2 F(s,t)}{\partial s^2} \right) = & -2k_l \frac{\partial F(s,t)}{\partial s} + ((k_l - k_d) - 2k_l s) \cdot \frac{\partial^2 F(s,t)}{\partial s^2} + \\ & + ((k_l - k_d) - 2k_l s) \frac{\partial^2 F(s,t)}{\partial s^2} + (k_d + (k_l - k_d)s - k_l s^2) \frac{\partial^3 F(s,t)}{\partial s^3} \end{aligned}$$

$$+k_l M \frac{\partial F(s,t)}{\partial s} + k_l M \frac{\partial F(s,t)}{\partial s} + k_l M (s-1) \frac{\partial^2 F(s,t)}{\partial s^2} \quad (.12)$$

$$= 1$$

$$\left. \frac{d}{dt} \left( \frac{\partial^2 F(s,t)}{\partial s^2} \right) \right|_{s=1} = -2k_l \frac{\partial F(s,t)}{\partial s} \Big|_{s=1} - 2(k_l + k_d) \cdot \frac{\partial^2 F(s,t)}{\partial s^2} \Big|_{s=1} + 2k_l M \frac{\partial F(s,t)}{\partial s} \Big|_{s=1} \quad (.13)$$

$$(y' + p(x)y = q(x)),$$

$$\left. \frac{d}{dt} \left( \frac{\partial^2 F(s,t)}{\partial s^2} \right) \right|_{s=1} = -2k_l \frac{\partial F(s,t)}{\partial s} \Big|_{s=1} - 2(k_l + k_d) \cdot \frac{\partial^2 F(s,t)}{\partial s^2} \Big|_{s=1} + 2k_l M \frac{\partial F(s,t)}{\partial s} \Big|_{s=1} \quad (.14)$$

$q$

( PGF  $s$  1) .

$$k = k_l + k_d \quad p = 2k \quad q = \frac{2k_l^2 M (M-1)}{k_l + k_d} (1 - e^{-kt}) \quad (.15)$$

$$\begin{aligned} \left. \frac{\partial^2 F(s,t)}{\partial s^2} \right|_{s=1} &= e^{-pt} \left( c + \int q e^{pt} dt \right) \\ &= e^{-2kt} \left( c + \int \frac{2k_l^2 M (M-1)}{k_l + k_d} (1 - e^{-kt}) e^{2kt} dt \right) = \\ &= c e^{-2kt} + e^{-2kt} \frac{2k_l^2 M (M-1)}{k_l + k_d} \left( \int e^{2kt} dt - \int e^{kt} dt \right) \quad (.16) \\ &= c e^{-2kt} + e^{-2kt} \frac{2k_l^2 M (M-1)}{k_l + k_d} \left( \frac{e^{2kt}}{2k} - \frac{e^{kt}}{k} \right) = \\ &= c e^{-2kt} + \frac{k_l^2 M (M-1)}{(k_l + k_d)^2} - \frac{2k_l^2 M (M-1)}{(k_l + k_d)^2} e^{-kt} \end{aligned}$$

$$D = \frac{\partial^2 F(s,t)}{\partial s^2} \Big|_{s=1} + \frac{\partial F(s,t)}{\partial s} \Big|_{s=1} - \left( \frac{\partial F(s,t)}{\partial s} \Big|_{s=1} \right)^2 \quad (.17)$$

$$= 1, \\ D = ce^{-2kt} + \frac{k_l^2 M(M-1)}{(k_l + k_d)^2} - \frac{2k_l^2 M(M-1)}{(k_l + k_d)^2} e^{-kt} + \frac{k_l M}{k_l + k_d} (1 - e^{-kt}) \\ - \left( \left( \frac{k_l M}{k_l + k_d} \right)^2 (1 - 2e^{-kt} + e^{-2kt}) \right) \quad (.18)$$

$$D = \left( c - \left( \frac{k_l M}{k_l + k_d} \right)^2 \right) e^{-2kt} + \frac{k_l^2 M(M-1)}{(k_l + k_d)^2} + \frac{k_l M}{k_l + k_d} - \left( \frac{k_l M}{k_l + k_d} \right)^2 \\ - \left( \frac{2k_l^2 M(M-1)}{(k_l + k_d)^2} + \frac{k_l M}{k_l + k_d} - 2 \left( \frac{k_l M}{k_l + k_d} \right)^2 \right) e^{-kt} \quad (.19)$$

$$c = \frac{\cancel{\left( \frac{k_l M}{k_l + k_d} \right)^2}}{\cancel{\left( \frac{k_l M}{k_l + k_d} \right)^2}} - \frac{k_l^2 M(M-1)}{(k_l + k_d)^2} - \frac{\cancel{k_l M}}{\cancel{k_l + k_d}} + \frac{\cancel{\left( \frac{k_l M}{k_l + k_d} \right)^2}}{\cancel{\left( \frac{k_l M}{k_l + k_d} \right)^2}} \\ + \left( \frac{2k_l^2 M(M-1)}{(k_l + k_d)^2} + \frac{\cancel{k_l M}}{\cancel{k_l + k_d}} - 2 \frac{\cancel{\left( \frac{k_l M}{k_l + k_d} \right)^2}}{\cancel{\left( \frac{k_l M}{k_l + k_d} \right)^2}} \right) \quad (.20)$$

$$c = \frac{k_l^2 M(M-1)}{(k_l + k_d)^2} \quad (.21)$$

$$D = \left( \frac{k_l^2 M(M-1)}{(k_l + k_d)^2} - \left( \frac{k_l M}{k_l + k_d} \right)^2 \right) e^{-2kt} + \frac{k_l^2 M(M-1)}{(k_l + k_d)^2} + \frac{k_l M}{k_l + k_d} - \left( \frac{k_l M}{k_l + k_d} \right)^2 \\ - \left( \frac{2k_l^2 M(M-1)}{(k_l + k_d)^2} + \frac{k_l M}{k_l + k_d} - 2 \left( \frac{k_l M}{k_l + k_d} \right)^2 \right) e^{-kt} \quad (.22)$$

$$D = -\frac{k_l^2 M}{(k_l + k_d)^2} e^{-2kt} - \frac{k_l^2 M}{(k_l + k_d)^2} + \frac{k_l M}{k_l + k_d} (1 - e^{-kt}) + 2 \frac{k_l^2 M}{(k_l + k_d)^2} e^{-kt} \quad (.23)$$

$$\begin{aligned} D &= \frac{k_l M}{k_l + k_d} (1 - e^{-kt}) - \frac{k_l^2 M}{(k_l + k_d)^2} (1 - 2e^{-kt} + e^{-2kt}) \\ &= \frac{k_l M}{k_l + k_d} (1 - e^{-kt}) - \frac{k_l^2 M}{(k_l + k_d)^2} (1 - e^{-kt})^2 \\ &= \frac{k_l M}{k_l + k_d} (1 - e^{-kt}) \left( 1 - \frac{k_l}{k_l + k_d} (1 - e^{-kt}) \right) \\ &= \frac{k_l M}{(k_l + k_d)^2} (1 - e^{-kt}) (k_d + k_l e^{-kt}) \end{aligned} \quad (.24)$$

$$u = \sqrt{\frac{D\{X(t)\}}{E\{X(t)\}^2}} = \sqrt{\frac{\frac{k_l M}{(\cancel{k_l + k_d})^2} (1 - e^{-kt}) (k_d + k_l e^{-kt})}{\left( \frac{k_l M}{\cancel{k_l + k_d}} (1 - e^{-kt}) \right)^2}} = \sqrt{\frac{[k_d + k_l e^{-kt}]}{M k_l (1 - e^{-kt})}} \quad (.25)$$



$$\begin{aligned}
\frac{dF(s_1, s_2, t)}{dt} &= M \left[ k_{l1}(s_1 - 1) + k_{l2}(s_2 - 1) \right] F(s_1, s_2, t) \\
&+ \left[ k_{l1}s_1(1 - s_1) + k_{l2}s_1(1 - s_{a2}) + k_{d1}(1 - s_1) \right] \frac{\partial F(s_1, s_2, t)}{\partial s_1} \\
&+ \left[ k_{l1}s_2(1 - s_1) + k_{l2}s_2(1 - s_2) + k_{d2}(1 - s_2) \right] \frac{\partial F(s_1, s_2, t)}{\partial s_2}
\end{aligned} \tag{.1}$$

1.

$$\begin{aligned}
d \frac{\partial F(s_1, s_2, t)}{\partial s_1} &= M \left[ k_{l1}(s_1 - 1) + k_{l2}(s_2 - 1) \right] \frac{\partial F(s_1, s_2, t)}{\partial s_1} + M k_{l1} F(s_1, s_2, t) \\
&+ \left[ k_{l1}s_1(1 - s_1) + k_{l2}s_1(1 - s_2) + k_{d1}(1 - s_1) \right] \frac{\partial^2 F(s_1, s_2, t)}{\partial s_1^2} \\
&+ \left[ k_{l1} - 2k_{l1}s_1 + k_{l2}(1 - s_{l2}) - k_{d1} \right] \frac{\partial F(s_1, s_2, t)}{\partial s_1} \\
&+ \left[ k_{l1}s_2(1 - s_1) + k_{l2}s_2(1 - s_2) + k_{d2}(1 - s_2) \right] \frac{\partial^2 F(s_1, s_2, t)}{\partial s_2 \partial s_1} - k_{l1}s_2 \frac{\partial F(s_1, s_2, t)}{\partial s_2}
\end{aligned} \tag{.2}$$

$$s_1 = s_2 = 1$$

$$\begin{aligned} \frac{d\overline{N_{a1}}}{dt} &= \\ 0 \cdot \overline{N_{a1}} + Mk_{l1} + 0 \cdot \frac{\partial^2 F(s_1, s_2, t)}{\partial s_1^2} + [k_{l1} - 2k_{l1} - k_{d1}] \overline{N_{a1}} + 0 \cdot \frac{\partial^2 F(s_1, s_2, t)}{\partial s_2 \partial s_1} - k_{l1} \overline{N_{a2}} & \quad (.3) \\ &= Mk_{l1} - (k_{l1} + k_{d1}) \overline{N_{a1}} - k_{l1} \overline{N_{a2}} \end{aligned}$$

(.1)

2:

$$\begin{aligned} \frac{d}{dt} \frac{\partial F(s_1, s_2, t)}{\partial s_2} &= M [k_{l1}(s_1 - 1) + k_{l2}(s_2 - 1)] \frac{\partial F(s_1, s_2, t)}{\partial s_2} + Mk_{l2} F(s_1, s_2, t) \\ &+ [k_{l1}s_1(1 - s_1) + k_{l2}s_1(1 - s_2) + k_{d1}(1 - s_1)] \frac{\partial^2 F(s_1, s_2, t)}{\partial s_1 \partial s_2} - k_{l2}s_1 \frac{\partial F(s_1, s_2, t)}{\partial s_1} \quad (.4) \\ &+ [k_{l1}s_2(1 - s_1) + k_{l2}s_2(1 - s_2) + k_{d2}(1 - s_2)] \frac{\partial^2 F(s_1, s_2, t)}{\partial s_2^2} \\ &+ [k_{l1}(1 - s_1) + k_{l2} - 2k_{l2}s_2 - k_{d2}] \frac{\partial F(s_1, s_2, t)}{\partial s_2} \end{aligned}$$

$$1 = 2 = 1$$

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$$\begin{aligned} \frac{d\overline{N_{a2}}}{dt} &= \\ 0 \cdot \overline{N_{a2}} + Mk_{l2} + 0 \cdot \frac{\partial^2 F(s_1, s_2, t)}{\partial s_1 \partial s_2} - k_{l2} \overline{N_{a1}} + 0 \cdot \frac{\partial^2 F(s_1, s_2, t)}{\partial s_1^2} + [k_{l2} - 2k_{l2} - k_{d2}] \overline{N_{a2}} & \quad (.5) \\ &= Mk_{l2} - k_{l2} \overline{N_{a1}} - (k_{l2} + k_{d2}) \overline{N_{a2}} \end{aligned}$$

(.3) (.5)

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$$\frac{d\overline{N_{a1}}}{dt} = k_{l1} (M - \overline{N_{a1}} - \overline{N_{a2}}) - k_{d1} \overline{N_{a1}} \quad ( .6)$$

$$\frac{d\overline{N_{a2}}}{dt} = k_{l2} (M - \overline{N_{a1}} - \overline{N_{a2}}) - k_{d2} \overline{N_{a2}}$$

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## Биографија аутора

Олга Јакшић (девојачко Башовић) је рођена 19. децембра 1968. године у Београду. Након основне школе завршила је математичку гимназију „Вељко Влаховић“ у Београду. Електротехнички факултет у Београду је уписала школске 1987/1988, а дипломирала школске 1992/1993. године. Дипломски рад под називом „Испитивање особина статистички синхронизабилних кодова“ је одбранила на Електротехничком факултету у Београду 1993. године. Магистарски рад под називом „Шум у микроелектромеханичким системима“ је одбранила на Електротехничком факултету у Београду 2005. године.

Од 1. јуна 1993. до 1. фебруара 2006. године је била запослена у Центру за микроелектронске технологије и монокристале Института за хемију, технологију и металургију у Београду.

Просветним радом се бавила од 1. фебруара 2006. до 1. септембра 2011. године у Електротехничкој школи „Никола Тесла“ у Панчеву.

Научно-истраживачким радом као званичном професијом се поново бави од 1. септембра 2011. године у Центру за микроелектронске технологије и монокристале Института за хемију, технологију и металургију у Београду. Ангажована је на пројекту технолошког развоја који финансира Министарство просвете, науке и технолошког развоја републике Србије (назив пројекта: "Микро, нано-системи и сензори за примену у електропривреди, процесној индустрији и заштити животне средине", број пројекта ТР32008) у оквиру кога проучава моделе адсорпционо-десорпционих процеса и шум у микро и нано (електро) механичким системима.

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*Orza Jakunt*

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*Orza Lakmut*

