

POLSKA AKADEMIA NAUK

INSTYTUT MASZYN PRZEPEŁYWOWYCH

**TRANSACTIONS
OF THE INSTITUTE OF
FLUID-FLOW MACHINERY**

PRACE

INSTYTUTU MASZYN PRZEPEŁYWOWYCH

103



GDAŃSK 1997

THE TRANSACTIONS OF THE INSTITUTE OF FLUID-FLOW MACHINERY

exist for the publication of theoretical and experimental investigations of all aspects of the mechanics and thermodynamics of fluid-flow with special reference to fluid-flow machines

*

PRACE INSTYTUTU MASZYN PRZEPIYWOWYCH

poświęcone są publikacjom naukowym z zakresu teorii i badań doświadczalnych w dziedzinie mechaniki i termodynamiki przepływów, ze szczególnym uwzględnieniem problematyki maszyn przepływowych

Wydanie publikacji dofinansowane zostało przez PAN ze środków DOT uzyskanych z Komitetu Badań Naukowych


EDITORIAL BOARD – RADA REDAKCYJNA

ZBIGNIEW BILICKI * TADEUSZ GERLACH * HENRYK JARZYNA
JAN KICIŃSKI * JERZY KRZYŻANOWSKI (CHAIRMAN – PRZEWODNICZĄCY)
WOJCIECH PIETRASZKIEWICZ * WŁODZIMIERZ J. PROSNAK
JÓZEF ŚMIGIELSKI * ZENON ZAKRZEWSKI

EDITORIAL COMMITTEE – KOMITET REDAKCYJNY

EUSTACHY S. BURKA (EDITOR-IN-CHIEF – REDAKTOR NACZELNY)
JAROSŁAW MIKIELEWICZ
EDWARD ŚLIWICKI (EXECUTIVE EDITOR – REDAKTOR) * ANDRZEJ ŻABICKI

EDITORIAL OFFICE – REDAKCJA

Wydawnictwo Instytutu Maszyn Przepływowych
Polskiej Akademii Nauk
ul. Gen. Józefa Fiszera 14, 80-952 Gdańsk, skr. poczt. 621,
 (0-58) 46-08-81 wew. 141, fax: (0-58) 41-61-44,
e-mail: esli@imppan.imp.pg.gda.pl

ISSN 0079-3205

JAROSŁAW KACZMAREK¹

Remark on the need of formulation of molecular catalysts mechanics

In this paper the need of formulation and a form of mechanics of molecular catalysts is discussed. The molecular catalyst is viewed as a molecule which is a catalyst of some chemical reactions and during this process its composition does not undergo a change. The aim of mechanics of molecular catalysts is to investigate a state of such a molecule or their ensembles. Thus, the catalyst is considered to be a stable object with respect to its composition and can be controlled by a system of chemical reactions. Two levels of description are suggested. The first one is the most elementary and is based on the quantum mechanics. The second one is more averaged and could be useful for phenomenological description of some chosen properties of the system.

1. Introduction

Catalysts can be classified with respect to conditions in which they act [1]. Consequently, we have homogeneous catalysts if they find themselves in the same place with substrates and no interfaces appear. On the other hand, we have heterogeneous catalysts which are separated from substrates by some interfaces. Furthermore, there is a category of catalysts called enzymes. They consist of large organic molecules and create colloidal suspension.

Especially interesting property of catalysts is their stability of composition during complicated chemical processes. This is important, for instance, in living systems. Complex molecular catalysts are objects which are controlled by chemical reactions and can create structures just owing to their stability.

An appropriate example of such a case is myosin which interacts with chemical reactions during the cross-bridge cycle. As a result of this, the monomer of myosin can deform and create a force in contractile actin-myosin unit [2], [3]. At Fig. 1. [3] a scheme of a myosin monomer is shown. There, the flexible regions are dark. The part marked by S1 interacts chemically with actin and ATP. Consequently, a force and motion are generated.

An interesting example is also a virus tail sheath which deforms in the way

¹Institute of Fluid-Flow Machinery, Department of Machinery Dynamics, ul. Fiszerza 14, 80-952 Gdańsk

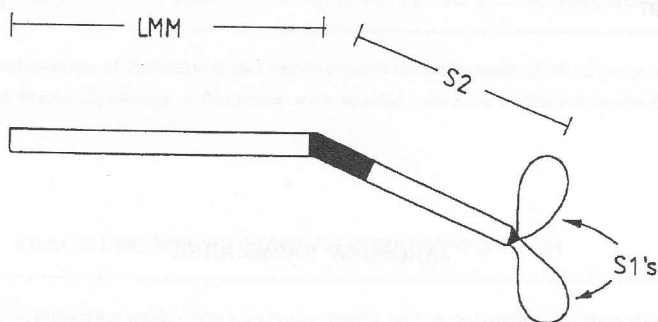


Fig. 1. Myosin monomer. The flexible regions are the dark regions between S1's and S2 and between S2 and LMM.

which is compatible with definition of the martensitic transformation. Therefore, this example is widely discussed in metallurgy for underlining of universality of the definition of the martensitic transformation [5]. At Fig. 2. and Fig. 3. [5] a shape transformation of such a structure is illustrated. It is seen that the dominant part of deformation is a shear deformation. This is just in accordance with the definition of the martensitic transformation.

The role of considered catalysts in living organisms is considerable. On the other hand, processes which are responsible for mentioned phenomena take place on molecular or on submolecular level. In general, they are very complicated. However, with the help of discussed catalysts we could attain interesting technical possibilities. A good example of interesting technical aspects for such a catalyst is just myosin molecule. Myosin and actin in contractile unit are controlled by chemical reactions. This form of control gives considerable dynamics of motion.

Similar, to some degree, systems are applied today for generation of motion. They are based on shape memory alloys and are applied in robotics, for instance. Hitachi Electrical Co. made a robot hand which is the same size as a human hand with 13 degree of freedom where shape memory alloy TiNi is used [4]. However, some difficulties appears with fast cooling in order to generate motion. Evidently, the contractile actin myosin units controllable by chemical reactions are much more attractive because of its dynamics and good controllability. However, they are extremely complicated from technological and theoretical point of view. Therefore, the purposes of theoretical investigations related to such systems have to be significantly confined in this paper.

Accordingly, in the first step, the term of molecular catalyst is introduced. This means that we consider only one, perhaps large, molecule which is not in any solution. It can catalyze a system of chemical reactions and is stable with respect to its own composition.

A mechanical description of behaviour of such a system is an interesting problem for investigations. Processes which are essential in evolution of such a mole-

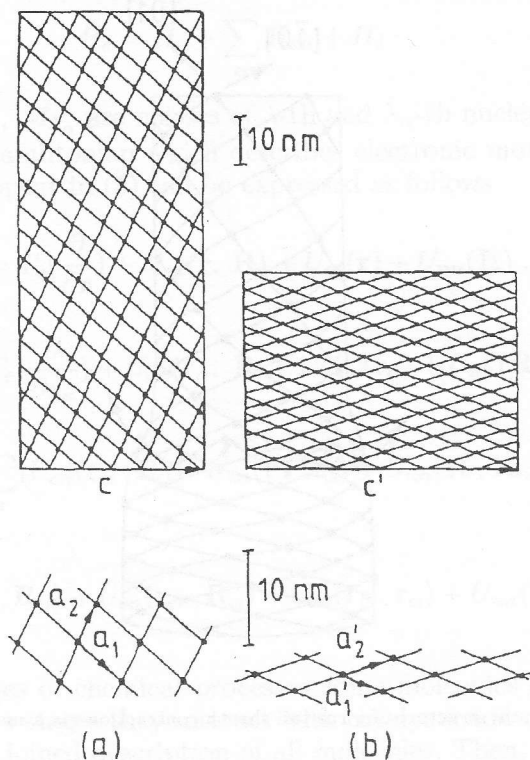


Fig. 2. Two dimensional lattice structure of virus tail sheath in (a) the metastable parent phase and (b) the martensitic product phase with corresponding lattice vectors.

cular catalyst indicate that the most elementary description should be given with the help of quantum mechanics or should be something more averaged only.

The aim of the present paper is to suggest only a possible form of mechanics of molecular catalysts. This aim is so confined since extremely complicated phenomena are involved in processes in question.

2. A concept of molecular catalysts mechanics

Let us note that quantum mechanical description related to chemical reactions are a subject of a large branch of our knowledge called chemical dynamics [6]. Therefore, an approach based on chemical dynamics should be just this elementary level of description. However, in some cases, we are interested in averaged to some degree, features of behaviour of molecular catalysts. It takes place, for instance, in deformed catalysts such as myosin or virus tail sheath. Therefore, more averaged models should also be introduced.

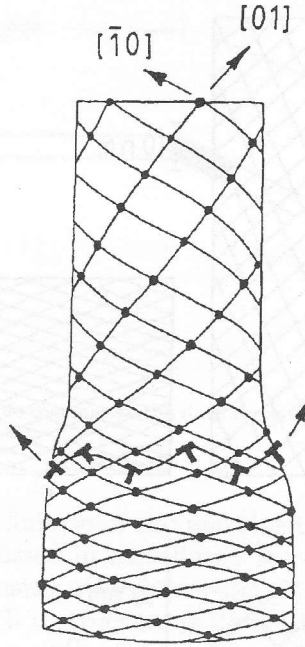


Fig. 3. Cylindrical crystal structure of virus tail sheath contraction via a martensitic transformation.

Let us consider a molecule which is the catalyst considered. Coordinates related to nuclei are given by vectors $\mathbf{R} = \{\mathbf{R}_\lambda\}$, $\lambda = 1, 2, \dots, \Lambda$, $\mathbf{R}_\lambda = \{R_{\lambda 1}, R_{\lambda 2}, R_{\lambda 3}\}$ and coordinates related to electrons are determined by vectors \mathbf{r} .

Let us consider also some other molecules which can react with the catalyst molecule. Then, the coordinates connected with nuclei of the α -th molecule are marked by $\mathbf{R}_\alpha = \{\mathbf{R}_{\lambda\alpha}\}$, $\lambda_\alpha \in \{1, 2, \dots, A\}$ and similarly as before, coordinates accompanied by electrons are given by vectors \mathbf{r}_α .

The total hamiltonian which describes the system of introduced molecules is assumed in the form

$$H_T = \bar{T}_n + \bar{H}_e, \quad (1)$$

where

$$\bar{T}_n = T_n + \sum_{\alpha} T_{n\alpha}, \quad (2)$$

$$T_n = \sum_{\lambda, i} \left(-\frac{\hbar^2}{2M_\lambda} \right) \frac{\partial^2}{\partial R_{\lambda i}^2}, \quad (3)$$

$$T_{n\alpha} = \sum_{\lambda_\alpha, i} \left(-\frac{\hbar^2}{2M_{\lambda_\alpha}} \right) \frac{\partial^2}{\partial R_{\lambda_\alpha i}^2}, \quad (4)$$

$$\bar{H}_e = H_e + \sum_{\alpha} H_{e\alpha} + H_I \quad (5)$$

and $\bar{h} = \frac{h}{2\pi}$ while M_{λ} , $M_{\lambda_{\alpha}}$ are masses of λ -th and λ_{α} -th nucleus respectively.

Thus, \bar{H}_e is the hamiltonian which describes electronic motion. Parts of the hamiltonian which appear in (5) can be expressed as follows

$$H_e = T_e\left(\frac{\partial}{\partial \mathbf{r}}\right) + U_{en}(\mathbf{r}, \mathbf{R}) + U_{ee}(\mathbf{r}) + U_{nn}(\mathbf{R}), \quad (6)$$

$$H_{e\alpha} = T_e\left(\frac{\partial}{\partial \mathbf{r}_{\alpha}}\right) + U_{en}(\mathbf{r}_{\alpha}, \mathbf{R}_{\alpha}) + U_{ee}(\mathbf{r}_{\alpha}) + U_{nn}(\mathbf{R}_{\alpha}), \quad (7)$$

$$H_I = \sum_{\alpha} (U_{en}(\mathbf{r}_{\alpha}, \mathbf{R}) + U_{en}(\mathbf{r}, \mathbf{R}_{\alpha}) + U_{nn}(\mathbf{R}, \mathbf{R}_{\alpha})) + \\ + \sum_{\alpha \neq \beta} (U_{en}(\mathbf{r}_{\alpha}, \mathbf{R}_{\beta}) + U_{en}(\mathbf{r}_{\beta}, \mathbf{R}_{\alpha}) + U_{ee}(\mathbf{r}_{\beta}, \mathbf{r}_{\alpha}) + U_{nn}(\mathbf{R}_{\beta}, \mathbf{R}_{\alpha})). \quad (8)$$

During different stages of chemical processes some molecules can be viewed as separated but in some stages they interact with catalysts and each other.

Let us discuss a joined description of all molecules. Then, for simplicity, let $\bar{\mathbf{r}} = \{\mathbf{r}, \mathbf{r}_{\alpha}\}$ and $\bar{\mathbf{R}} = \{\mathbf{R}, \mathbf{R}_{\alpha}\}$. Many phenomena in molecule suggest that the Born-Oppenheimer approximation is broken down; to cite a few: nonradiative transitions in polyatomic molecules [7], the Jahn-Teller effect [8], vibronic coupling effects in molecular spectroscopy [9], predissociation and perturbation in the spectra of diatomic molecules [10] and charge transfer collisions of atoms and molecules [11].

In the most general case, we are interested in total state of the catalyst molecule. Therefore, mentioned phenomena suggest that such a state should be considered with the help of the non-adiabatic description.

Let $\psi_k = \psi_k(\bar{\mathbf{r}}, \bar{\mathbf{R}})$ be k -th eigenvector of the electronic hamiltonian \bar{H}_e . It means that the following equation is fulfilled

$$\bar{H}_e \psi_k(\bar{\mathbf{r}}, \bar{\mathbf{R}}) = \epsilon(\bar{\mathbf{R}}) \psi_k(\bar{\mathbf{r}}, \bar{\mathbf{R}}). \quad (9)$$

Then, the total wave function in nonadiabatic approximation can be assumed in the form

$$\Psi(\bar{\mathbf{r}}, \bar{\mathbf{R}}) = \sum_k \psi_k(\bar{\mathbf{r}}, \bar{\mathbf{R}}) \chi_k(\bar{\mathbf{R}}) \quad (10)$$

and the Schrödinger equation for our system of molecules is given as

$$H_T \Psi \equiv (\bar{T}_n + \bar{H}_e) \Psi = E_T \Psi. \quad (11)$$

After detailed calculations we obtain the well known in literature [12] equation

$$\sum_j [\bar{T}_n + \epsilon_j(\bar{\mathbf{R}}) - E_T] \chi_j + \sum_\lambda \left(-\frac{\hbar^2}{2M_\lambda}\right) \int \sum_{j,k} \psi_j^* \frac{\partial^2}{\partial \mathbf{R}_\lambda^2} \psi_k d\bar{\mathbf{r}} \chi_k + \\ + \sum_\lambda \left(-\frac{\hbar^2}{M_\lambda}\right) \int \sum_{j \neq k} \psi_j^* \frac{\partial}{\partial \mathbf{R}_\lambda} \psi_k d\bar{\mathbf{r}} \frac{\partial}{\partial \mathbf{R}_\lambda} \chi_k = 0. \quad (12)$$

It is assumed that the catalyst molecule can react with the remaining molecules or with part of them. Furthermore, some of α -th molecules can react between themselves. However, the catalyst molecule is the same before and after the sequence of reactions. Consequently, it is a general assumption that the equation (12) describes just such a situation. It means that initial conditions are chosen in a way which guarantee required processes.

The main problem is related to the state of this molecule during the sequence of reactions and later.

Let us assume that at an initial instant, states of molecules are approximately described by separate for each of them hamiltonians. Thus,

$$H\psi = E\psi, \quad H_\alpha\psi_\alpha = E_\alpha\psi_\alpha. \quad (13)$$

Then, $\Psi(t_0) = \psi(t_0) \prod_\alpha \psi_\alpha(t_0)$. It is assumed also that $\Psi(t_0)$ satisfies conditions which secure that the desirable sequence of reactions will take place. The total energy of the system is $E_T(t_0) = E(t_0) + \sum_\alpha E_\alpha(t_0)$. After the sequence of reactions at an instant T_r , we can have the situation

$$E(T_r) = \int \psi^*(T_r) H \psi(T_r) d\mathbf{r} d\mathbf{R} > E(t_0). \quad (14)$$

In this case the catalyst molecule has obtained an excitation as a result of mentioned reactions. Then, the main problem is related to the evolution of excited state of this molecule. Similar problems are discussed in photochemistry (see for instance [13], [14]).

Similarly, $E(T_r)$ can be less than $E(t_0)$. Both cases can induce complicated processes which lead to changing properties of the molecular catalyst. In particular, these changes can consist in transformation of shape of molecule as in the case of myosin.

Let us note that for organic molecules full solution of equation (12) is extremely complicated. Some view on this problem can be worked out with the help, for example, [15], [6].

Difficulties in solution of equation (12) for too large systems suggest that as a primary step of investigations should be finding out possible small organic molecule which is a catalyst for possible simple chemical reactions and next to develop description and calculations as far as possible for this prototype.

In this place, methods of chemical dynamics [6] should be applied and developed. This direction of investigations is viewed to be the most elementary one.

In order to analyse only some chosen properties of more complicated systems a dimensionally reduced description can be appropriate. We assume first that such a description is based on known wave function evolution $\Psi(\bar{\mathbf{r}}, \bar{\mathbf{R}}, t)$. However, it is expected that such simplified models can be useful also on this more averaged level by some phenomenological assumptions.

Let us consider a set of admissible wave functions $\mathcal{M}_\psi = \{\Psi(\bar{\mathbf{r}}, \bar{\mathbf{R}})\}$ for our molecular system. Let $T = [t_0, t_0 + T_k]$ be a time interval and let $(\mathcal{M}_\psi \times T) = \{\Psi(\bar{\mathbf{r}}, \bar{\mathbf{R}}, t) : t \in T, \Psi(\bar{\mathbf{r}}, \bar{\mathbf{R}}, t) \in \mathcal{M}_\psi, \Psi(\bar{\mathbf{r}}, \bar{\mathbf{R}}, t_0) = \psi(\bar{\mathbf{r}}, \bar{\mathbf{R}}) \prod_\alpha \psi_\alpha(\bar{\mathbf{r}}_\alpha, \bar{\mathbf{R}}_\alpha)\}$.

The evolution of function Ψ is determined by equation (12). Thus, with the help of this equation an infinite dimensional dynamical system DSI is introduced.

Behaviour of complex molecular system is very complicated. Sometimes, we are interested in some characteristic features of this behaviour only. Then, a simplified dimensionally reduced description would be convenient. Such a situation appear when we try to describe a deformation of the complex molecule. Let us consider previously discussed deformable virus tail sheath illustrated at Fig. 2. and Fig. 3. Observed there deformation process indicates on similarity with objects which are usually described by relatively simple continuum models. Therefore, we should discuss also a dimensional reduction of DSI into a finite dimensional dynamical system.

Continuum models which are derived by means of dimensional reduction of a finite-dimensional dynamical systems are discussed in [16]. Method of simplification of description considered in this paper is similar to that from [16].

Consequently, let us consider a system of variables $\mathbf{d} = \{\mathbf{d}_i\}$, $i \in I$ where $I = \{1, 2, \dots, N\}$ and N be a number of subsystems of our molecule. Let $\mathcal{M} = \{\mathbf{d}\}$ be a set of all admissible values of these variables. We can discuss an evolution function $\chi(\mathbf{C}, \mathbf{d}_0, \mathbf{f})(t)$ which depends on unidentified at the moment parameters \mathbf{C} , external forces \mathbf{f} and $\mathbf{d}(t_0) = \mathbf{d}_0$.

We can define the space $(\mathcal{M} \times T)_{Cf} = \{\chi(\mathbf{C}, \mathbf{d}_0, \mathbf{f})(t) : t \in T, \mathbf{d}_0 \in \mathcal{M}\}$. Let us note also that forces create a space \mathcal{F} . They are also processes which create a space $(\mathcal{F} \times T) = \{\mathbf{f}(t) : t \in T\}$.

The dimensional reduction is introduced here by means of a map $\pi_T : (\mathcal{M}_\psi \times T) \rightarrow (\mathcal{M} \times T)_f \times (\mathcal{F} \times T)$, where $\pi_T = \{\tilde{\pi}_T, \tilde{\pi}_{Tf}\}$, $\tilde{\pi}_T : (\mathcal{M}_\psi \times T) \rightarrow (\mathcal{M} \times T)_f$, $\tilde{\pi}_{Tf} : (\mathcal{M}_\psi \times T) \rightarrow (\mathcal{F} \times T)$, $(\mathcal{M} \times T)_f = \{\mathbf{d}(t) : \mathbf{d}(t) = \tilde{\pi}_T \Psi, \Psi \in (\mathcal{M}_\psi \times T)\}$.

The space $(\mathcal{M} \times T)_f$ is a space of processes related to the catalyst molecule, $(\mathcal{F} \times T)$ represents interactions induced by chemical reactions related to each of α -th molecules where $\alpha = 1, 2, \dots, A$ and any other external forces.

The evolution function $\chi(\mathbf{C})$ of catalyst molecule is unidentified at this moment since it depends on a parameters \mathbf{C} . Therefore, we will indicate a possible way of specification of these parameters.

As it was previously assumed the molecule is divided on N parts. Each i -th part, where $i \in I$, consists of a given and unchanged number of atoms. For each i -th part, we introduce some variables with index i .

Let us introduce the following variables $\mathbf{a} = \{a_{mi}\}$, $\xi = \{\theta, \eta\}$, $\theta = \{\theta_i\}$, $\eta = \{\eta_{\beta i}\}$, $\varrho = \{\varrho_{\gamma i}\}$, $m \in I_m$, $\beta \in I_\beta$, $\gamma \in I_\gamma$, $i \in I$. The variable \mathbf{a} is slowly varying.

It can be, for instance, a measure of deformation which would be accepted for example, for virus tail sheath transformation. The variable ξ represents averaged effects which are connected with more quickly varying processes. θ is a counterpart of temperature and η represents variables related to nonequilibrium states. ϱ is a variable which is connected with external interactions following from reacting molecules on the catalyst molecule. Introduced sets of indices are established during defining kind of variables and characterizing their number.

With the help of these variables we can introduce the following functions: $V(\mathbf{C}_V, \mathbf{a}, \xi, \varrho)$, $K(\mathbf{C}_K, \dot{\mathbf{a}})$, $H_i(\mathbf{C}_{Hi}, \mathbf{a}, \xi, \varrho)$, $Q_{\delta ij}(\mathbf{C}_{Q\delta ij}, \mathbf{a}, \xi, \varrho)$, $i \neq j$, $i, j \in I$, $\delta \in I_\delta$, $A_{\beta i}(\mathbf{C}_{A\beta i}, \mathbf{a}, \xi, \varrho)$, $\beta \in I_\beta$, $R_{\gamma i}(\mathbf{C}_{R\gamma i}, \mathbf{a}, \xi, \varrho)$, $\gamma \in I_\gamma$, $N_{\mu k}(\mathbf{C}_{N\mu k}, \mathbf{a}, \xi, \varrho)$, $\mu \in I_\mu$, $k \in I_k \subset I$. Above mentioned functions depend on constants $\mathbf{C} \in \mathcal{C}$, where \mathcal{C} is a set of all admissible constants of this kind. The function V represents potential energy with respect of slowly varying variable \mathbf{a} . K is the kinetic energy related to the variable \mathbf{a} . H_i are forces conjugate with variables θ_i . $Q_{\delta ij}$ is a function which represents an amount of energy transformed from i -th part into j -th part of our system. $N_{\mu k}$ represents an amount of energy which is transformed from external molecules into k -th part of catalyst molecule. $A_{\beta i}$, $R_{\gamma i}$ are functions which describe evolution of variables $\eta_{\beta i}$ and $\varrho_{\gamma i}$.

By means of these functions we can postulate an expression for energy of our molecule in dimensionally reduced description in the following form

$$E = V + H_i \theta_i + K, \quad (15)$$

where the summation convention is applied.

The balance of energy equation is postulated in the form

$$\dot{E} = f_{mi} \dot{a}_{mi} + \sum_{\mu, k} N_{\mu k} + \sum_{\varrho i} r_{\varrho i}, \quad \varrho \in I_\varrho. \quad (16)$$

Thus, f_{mi} , $N_{\mu k}$, $r_{\varrho i}$ play a role of external interactions of our catalyst molecule with external ones.

Let

$$\frac{\partial K}{\partial \dot{a}_{mk}} \approx \frac{\partial K}{\partial \dot{a}_{mk}}(0) + \frac{1}{2} \frac{\partial^2 K}{\partial \dot{a}_{mk} \partial \dot{a}_{nj}}(0) \dot{a}_{nj} = M_{mknj} \dot{a}_{nj},$$

where $\frac{\partial K}{\partial \dot{a}_{mk}}(0)$ is assumed to be equal to zero. Furthermore, let $\sum_{\delta, i \neq j} Q_{\delta ij} = 0$. Then, using (15) we can transform the balance energy equation (16) into the form

$$\begin{aligned} & \left[\frac{\partial V}{\partial a_{mi}} - f_{mi} + M_{njmi} \dot{a}_{nj} \right] \dot{a}_{mi} + \left(\frac{\partial V}{\partial \theta_i} + H_i \right) \dot{\theta}_i + \\ & + \dot{H}_i \theta_i + \frac{\partial V}{\partial \eta_{\beta i}} \dot{\eta}_{\beta i} + \frac{\partial V}{\partial \varrho_{\gamma i}} \dot{\varrho}_{\gamma i} + \sum_{\delta, i \neq j} Q_{\delta ij} - \sum_{\varrho i} r_{\varrho i} - \sum_{\mu k} N_{\mu k} = 0. \end{aligned} \quad (17)$$

It is assumed that the equation is valid for arbitrary time processes. Then, the following system of equations is postulated with the help of (17)

$$\dot{a}_{mi} = b_{mi}, \quad (18)$$

$$M_{njmi} \dot{b}_{nj} = -\frac{\partial V}{\partial a_{mi}} + f_{mi}, \quad (19)$$

$$\dot{H}_i \theta_i + \frac{\partial V}{\partial \eta_{\beta i}} \dot{\eta}_{\beta i} + \frac{\partial V}{\partial \varrho_{\gamma i}} \dot{\varrho}_{\gamma i} + \sum_{\delta, i \neq j} Q_{\delta ij} - \sum_{\rho i} r_{\rho i} - \sum_{\mu k} N_{\mu k} = 0. \quad (20)$$

We assume also the evolution equations for η and ϱ as

$$\dot{\eta}_{\beta i} = A_{\beta i}(\mathbf{C}_{A\beta i}, \mathbf{a}, \xi, \varrho), \quad (21)$$

$$\dot{\varrho}_{\gamma i} = R_{\gamma i}(\mathbf{C}_{R\gamma i}, \mathbf{a}, \xi, \varrho), \quad (22)$$

and the equation

$$H_i = -\frac{\partial V}{\partial \theta_i}. \quad (23)$$

Equations (18)-(22) determine the form of evolution function $\chi(\mathbf{C})(t)$ for our dimensionally reduced system with $\mathbf{d} = \{a_{mi}, b_{mi}, \theta_i, \eta_{\beta i}, \varrho_{\gamma i}\}$ and $\mathbf{f} = \{f_{mi}, r_{\rho i}, N_{\mu k}\}$. Determination of the constants \mathbf{C} completes determination of the evolution function. Consequently, we should give a procedure for doing it.

For convenience let us consider a more general space of processes $C \equiv C(\mathbf{d}_0) = \{\varphi(t) : \varphi : T \rightarrow \mathcal{M}, \varphi(t_0) = \mathbf{d}_0 \in \mathcal{M}\}$. Let us introduce a metric $\rho : C \times C \rightarrow R^+ \cup \{0\}$ on this space. Let $i : (\mathcal{M} \times T)_f \times (\mathcal{F} \times T) \rightarrow C$ and $i_C : (\mathcal{M} \times T)_{Cf} \times (\mathcal{F} \times T) \rightarrow C$ be injections of processes into space C which are obtained firstly by calculation based on the nonreduced system (12) and secondly by assuming \mathbf{C} and calculations with the help of equations (18)-(22).

Using introduced injections we are able to define the following function

$$h(\mathbf{d}_0) = \inf_{\mathbf{C} \in \mathcal{C}} \rho(i_C \chi(\mathbf{C}, \mathbf{d}_0, \mathbf{f}), i_{\pi_T} \Psi). \quad (24)$$

By \mathbf{C}^* we denote the constant $\mathbf{C} \in \mathcal{C}$ which realizes a minimum for the function h . Consequently, $\mathbf{C}^* = \mathbf{C}^*(\mathbf{d}_0)$ depends on \mathbf{d}_0 . A satisfactory approximation should have the property that \mathbf{C}^* displays weak dependence on the initial conditions. Thus, we assume an averaged value of this constant

$$\bar{\mathbf{C}} = Av\{\mathbf{C}^* : \mathbf{C}^* = \mathbf{C}^*(\mathbf{d}_0), \mathbf{d}_0 \in \mathcal{M}\}, \quad (25)$$

where Av means an averaging procedure. At this moment the evolution function is fully determined.

Equations (18)-(23) could also be introduced phenomenologically by assumption appropriate constants \mathbf{C} . This happens only if sufficient number of experimental facts would be known.

Summing up these considerations, let us notice that we have obtained next two possibilities in modelling the molecular catalyst. The first one is in fact multiscale.

Then, the more averaged model is derived with the help of the most elementary one. The second possibility rest on introducing phenomenological assumptions related to C and based on experiments.

The model given by equations (18)-(23) can be viewed as a step for description of larger complexes of molecular catalysts by further averaging.

3. Final remarks

In the paper a concept of mechanics of molecular catalysts is discussed. The need for such mechanics follows from meaning of such objects. They can perform miscellaneous functions and are controllable by chemical reactions. Their key meaning rests on stability of their composition during chemical processes. Therefore, they can create more complex structures which coexist with chemical reactions.

Molecular and submolecular levels of processes which appear in these molecules suggest quantum mechanical description on the way related to chemical dynamics. Complicated phenomena and complexity of equations suggest necessity to look for an appropriate molecule which is relatively small and which is a catalyst for relatively simple reactions. Such a molecule would be convenient prototype for developing descriptions and calculations.

In order to describe only some chosen properties of evolution of catalysts discussed, averaged models are also introduced. They can be created with the aid of known evolution of the wave function if it is the case. If not, models of this kind could be introduced by phenomenological assumptions on variables and form of functions V , K , H , Q , N , A , R without any dependances on unknown constants.

This level of description applied, for instance, to myosin-actin contractile unit, perhaps would be a good way for description of mechanics of cytogel. Model of cytogel based on mechanochemistry is introduced in [17]. Then, discussed here concept for modelling the cytogel would be a complementary for the mechanochemistry approach.

At this stage a more complicated situation when the molecule is in a solution is not discussed.

Manuscript received in September 1996

References

- [1] Bond G. C.: *Heterogeneous catalysis: Principles and applications*, Clarendon Press, Oxford 1974.
- [2] Goody R. S., Holmes K. C.: *Biochimica et Biophysica Acta*, 726(1983), 13-39.

- [3] Highsmith S.: *Biochimica et Biophysica Acta*, 639(1981), 31-39.
- [4] Otsuka K., Shimizu K.: *Int. Met. Rev.* **31**(1986), 93-114.
- [5] Cohen M.: *Trans. of the Japan Inst. of Met.*, **29**, 8(1988), 609-624.
- [6] Baer M.: *Theory of chemical reaction dynamics*, CRC Press Inc., 1985.
- [7] Bixon M., Jortner J.: *J. Chem. Phys.*, **48**(1968), 715.
- [8] Jahn H. A., Teller E.: *Proc. Roy. Soc. (London)*, **A161**(1937), 220.
- [9] Koppel H., Domcke W., Cederbaum L.: *Adv. Chem. Phys.*, **57**(1984), 59.
- [10] Lefebvre-Brion H., Fields R. W.: *Perturbations in the spectra of diatomic molecules*, Academic Press 1986.
- [11] Sidis V.: *Diabatic potential energy surface for charge transfer processes* in M. Baer, Chenk-Yiu NG (eds.) *State-selected and state-to-state ion molecule reaction dynamics*, John Wiley & Sons 1992.
- [12] Lengsfeld B.H.: *Nonadiabatic interactions between potential energy surfaces: theoeer and applications* in M. Baer, Chenk-Yiu NG (eds.) *State-selected and state-to-state ion molecule reaction dynamics*, John Wiley & Sons 1992.
- [13] Turbo N.J.: *Modern molecular photochemistry*, The Benjamin/Cummings Publishing Co. Inc. 1978.
- [14] Cowan D. O., Drisko R. L.: *Elements of organic photochemistry*, Plenum Press, New York, 1976.
- [15] Epiotis N. D.: *Theory of organic reactions*, Springer Verlag 1978.
- [16] Kaczmarek J.: *A formulation of continuum mechanics as a dimensional reduction of a finite-dimensional dynamical system*, *Arch. Mech.*, **49**(1997), 49-66.
- [17] Oster G. F., Odell G. M.: *Physica* **12D**(1984), 333-350.

Uwagi o potrzebie sformułowania mechaniki katalizatorów molekularnych

Streszczenie

W pracy przedyskutowano potrzebę sformułowania mechaniki katalizatorów molekularnych. Pojęcie katalizatora molekularnego związane jest ze stabilną, ze względu na skład chemiczny, molekułą, która bierze udział w reakcjach chemicznych. W rezultacie molekuly tego typu mogą tworzyć większe struktury, które są stabilne podczas ich oddziaływania z reakcjami chemicznymi. Celem mechaniki katalizatorów molekularnych jest badanie stanu tego typu molekul podczas oddziaływania z wymienionymi reakcjami. Rozważa się dwa poziomy opisu, pierwszy – bardziej elementarny – związany jest z mechaniką kwantową, drugi – bardziej uśredniony – umożliwia wprowadzenie założeń fenomenologicznych.