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COMPUTER SIMULATION OF DIFFUSION TRANSPORT PROCESSES IN MULTILAYER NANOFILMS

The difficulties associated with studying diffusion in multilayer films require the progress of contemporary modeling methods and software platforms to precisely represent phenomena, taking into account transitions between adjacent layers. In addition to the indispensable role of advanced modeling techniques and software in solving the problems of studying diffusion in multilayer films, it is extremely important to admit the key contribution of sophisticated computational approaches. In this paper, the authors attempt to merge intricate mathematical models with optimal software development methodologies to address the challenge of simulating diffusion transport processes in multilayer nanofilms computationally. Based on the experimental findings and employing the suggested model, identification was conducted utilizing the theory of state control for multicomponent systems. With the help of methods of optimal control of the state of multicomponent transport systems, the analytical solution of the model and the data of experimental observations, the distributions of diffusion coefficients for the considered components of nanofilms (samples of aluminum, molybdenum, silicon) were reproduced. Numerical simulation results were compared with experimental observations. The profiles obtained from the modeling closely match the corresponding experimental profiles, especially as the duration of multilayer formation converges to the final stages of completing the protective nanofilm multilayer formation. The maximum observed deviation does not exceed 2–3%, confirming the reliability of the mathematical model and demonstrating the practical value of the results provided. A software framework is developed for the automation of the specified calculations with the possibility of extension to other subject areas with similar tasks of identifying the key factors of the process and further numerical modeling of the time-space characteristics using the obtained results.
Key words: coefficients identification, diffusion, mathematical model, multilayer nanofilm, numerical simulation, oxide film.

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КОМП'ЮТЕРНЕ МОДЕЛЮВАННЯ ПРОЦЕСІВ ДИФУЗІЙНОГО ТРАНСПОРТУ В БАГАТОШАРОВИХ НАНОПЛІВКАХ

Проблеми, пов'язані з дослідженням дифузії в багатошарових плівках, вимагають удосконалення сучасних методів моделювання та програмного забезпечення для точного зображення явищ, враховуючи переходи між сусідніми шарами. На додаток до незамінної ролі передових методів моделювання та програмного забезпечення у розв'язанні проблем вивчення дифузії в багатошарових плівках, надзвичайно важливо визнати ключовий внесок складних обчислювальних підходів. У цій роботі автори зробили спробу поєднати складні математичні моделі з передовим досвідом розробки програмного забезпечення для розв'язання задачі комп'ютерного моделювання процесів дифузійного транспорту в багатошарових наноплівках. За результатами експериментальних даних та з використанням запропонованої моделі проведено ідентифікацію з використанням теорії контролю стану багатокомпонентних систем. За допомогою методів оптимального керування станом багатокомпонентних транспортних систем, аналітичного рішення моделі та даних експериментальних спостережень відтворено розподіли коефіцієнтів дифузії для розглянутих складових компонентів наноплівок (зразків алюмінію, молібдену, кремнію). Результати чисельного моделювання порівнюються з експериментальними спостереженнями, що відображають вміст зразка. Профілі, отримані в результаті моделювання, тісно збігаються з відповідними експериментальними профілями, особливо коли тривалість багатошарового утворення наближається до остаточного періоду завершення утворення захисної наноплівки. Максимальне спостережене відхилення не перевищує 2–3 %, що підтверджує надійність математичної моделі та вказує на практичну корисність отриманих результатів. Розроблено програмний інструментарій для автоматизації зазначених розрахунків з можливістю поширення на інші предметні області зі схожими завданнями ідентифікації ключових факторів процесу. Створено базу для подальшого чисельного моделювання часо-просторових характеристик з використанням отриманих результатів.
Ключові слова: ідентифікація коефіцієнтів, дифузія, математична модель, багатошарова наноплівка, чисельне моделювання, оксидна плівка.

Introduction

The challenges associated with investigating diffusion in multilayer films necessitate the advancement of modern modeling techniques and software frameworks to accurately depict phenomena while considering transitions between neighboring layers [1, 2]. One of the most effective approaches to thoroughly address these issues is the widely recognized integral transformation. These methods are utilized to derive solutions for diverse boundary value problems in mathematical physics pertaining to homogeneous structures, encompassing diffusion scenarios across various environments and enabling their mathematical representation.

In addition to the indispensable role of advanced modeling techniques and software frameworks in addressing the challenges of studying diffusion within multilayer films, it is crucial to recognize the pivotal contribution of sophisticated computational approaches. By utilizing cutting-edge methodologies in computer programming and software architecture, the capabilities of mathematical approaches, such as integral transformations, can be elevated to unprecedented levels of precision and efficiency. This synergy between mathematical modeling and computational innovation not only enhances our ability to accurately represent diffusion phenomena within multilayer structures but also opens new avenues for exploration and analysis.

Furthermore, the integration of modern modeling techniques with state-of-the-art computational tools facilitates a more nuanced understanding of diffusion processes across diverse material compositions and environmental conditions. By leveraging the power of numerical simulations and data-driven analyses, researchers can extract valuable knowledge on complex diffusion mechanisms that were previously inaccessible. This multidisciplinary approach enables scientists and engineers not only to solve fundamental questions in materials science and engineering but also to devise innovative strategies for optimizing the performance and functionality of multilayer films in various technological applications.

Problem formulation. In the proposed research, authors attempt to combine complex mathematical models with the best practices of software engineering to solve the problem of computer simulation of diffusion transport processes in multilayer nanofilms. The primary objectives are:

– modeling diffusion processes: to integrate complex mathematical models with contemporary software development methodologies, in domain of the diffusion within multilayer films;

– parameter identification: to employ the theory of state control for multicomponent systems to identify diffusion coefficients. It includes using methods of optimal control to analyze experimental data and accurately reproduce the distributions of diffusion coefficients for the nanofilm components;

– numerical simulation and validation: to conduct numerical simulations and compare the results with experimental observations. The target is to achieve a high degree of correspondence between the modeled and experimental profiles, particularly as the duration of multilayer formation approaches completion;

– development of a software framework: to create a software framework that automates the specified calculations and can be extended to other subject areas with similar tasks. This framework should facilitate the identification of key process factors and enable further numerical modeling of time-space characteristics based on the obtained results.

By addressing these objectives, this research seeks to overcome the limitations of existing methods and provide a robust tool for studying and optimizing the diffusion processes in multilayer films. The ultimate aim is to enhance the efficiency of experimental studies and contribute to the development of new nanomaterials with improved properties

1. Mathematical model

We propose a physical problem and corresponding mathematical model for the diffusion mass transfer process in multilayer films, based on the following multilayer medium comprised of n double layers composed

of two distinct media with differing properties (Fig. 1).

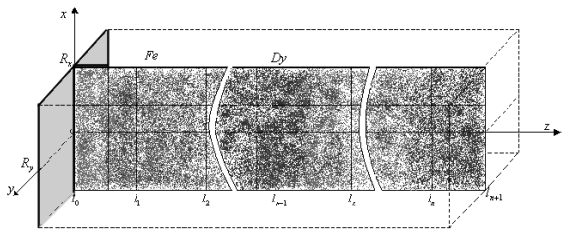

Fig.1. Schema of multilayer nanofilm

According to this representation, at each interface within the formed multicomposite, there is a mutual diffusion of components between two adjacent layers of the medium. The mechanisms governing such mutual transfer are determined by the variable gradients and rates of concentration change at the interface boundaries between layers. By incorporating changes in concentrations and their gradients over time into the boundary and interface conditions, it becomes feasible to model the mechanisms of this additional mutual transport alongside the fundamental transport equations.

When formulating a mathematical model for diffusion transfer within oxide films, a multilayer structure is taken into account. Assuming that the diffusion of atoms of constituent components of oxide films (aluminum, molybdenum, silicon) primarily drives system mixing, concentration profiles for such a multilayer system can be derived from Fick's equations, incorporating boundary conditions at the outer layers and contact conditions between successive layers. This approach provides a mathematical model describing the diffusion transfer process within a planar multilayered medium

$$\frac{\partial}{\partial z} C_k(t,x,z) + \gamma_k^2 C_k = D_0 \frac{\partial^2 C_k}{\partial x^2} + D_{z_k} \frac{\partial^2 C_k}{\partial z^2} \quad (1)$$

at domain

$$t > 0,\ x \in (0,\ R),$$
$$z : z \in \bigcup_{k=1}^{n+1} (l_{k-1},\ l_k);\ l_0 \geq 0;\ l_{n+1} = \infty,$$
$$j = \overline{1,\ 2}\ k = \overline{1,\ n},$$

where C_k is diffusion distribution; D_k is diffusion coefficient; γ_k^2 is mass distribution coefficient;

The corresponding initial and boundary conditions for the model:

$$C_k(t,\ x,\ z)\big|_{t=0} = C_{0_k}(x,\ z) \equiv C_{0_k}(z),$$
$$\left[\alpha_{11}^0 \frac{d}{dz} + \beta_{11}^0\right] C_1(t,x,z)\Big|_{z=l_0} = C_{l_0}(t,x); \quad (2)$$
$$\frac{\partial C_{n+1}}{\partial z}\Big|_{z=\infty} = 0;$$

$$\left[\begin{array}{l}\left[\alpha_{j1}^k \frac{\partial}{\partial z} + \beta_{j1}^k\right] C_k - \\ -\left[\alpha_{j2}^k \frac{\partial}{\partial z} + \beta_{j2}^k\right] C_{k+1}\end{array}\right]_{z=l_k} = 0,$$

And boundary conditions for variable x

$$\frac{\partial C_k}{\partial x}\Big|_{x=0} = 0; \quad C_k\big|_{x=R} = C_{1_k}(t,z), \quad (3)$$

where α_{ij}^k, β_{ij}^k; $k = \overline{0,n}$; $i, j = \overline{1,2}$, are coefficients determining boundary conditions and contact conditions; x, y, z are spatial coordinates.

The exact solution of the problem described by equations (1)–(3) is directly written out by applying integral Fourier transforms [3].

$$C_k(t,x,z) =$$
$$\int_0^t \int_0^R W_{l_{0,k}}(t-\tau;\ x,\ \varsigma;\ z)\ C_{l_0}(\tau,\ \varsigma) d\varsigma d\tau +$$
$$+ \sum_{k_1=1}^{n+1} \int_0^t \int_0^r \int_{l_{k_1-1}}^{l_{k_1}} H_{k,k_1}(t-\tau;\ x,\ \varsigma;\ z,\ \xi) \cdot$$
$$\cdot C_{0_k}(\varsigma,\ \xi) \cdot \delta_+(\tau) \sigma_{k_1} d\varsigma d\xi d\tau +$$
$$+ \sum_{k_1=1}^{n+1} \int_0^t \int_{l_{k_1-1}}^{l_{k_1}} W_{R_k,k_1}(t-\tau;\ x;\ z,\ \xi) \cdot$$
$$\cdot C_{1_{k_1}}(\tau,\ \xi) \sigma_{k_1} d\xi d\tau.$$

Here are Green's functions:

$$W_{l_{0,k}}(t, x; z, \xi) =$$

$$= \frac{2}{R} \sum_{m=0}^{\infty} W_{l_0,k}^{m}(t-\tau, z)(-1)^m \frac{\cos \eta_m \xi}{\eta_m};$$

$$W_{R_{k,k_1}}(t; x, \varsigma; z, \xi) =$$

$$= \frac{2}{R} \sum_{m=0}^{\infty} e^{-D_0 \eta_m^2 t} D_0 (-1)^{m+1} \eta_m \varepsilon_{k,k_1}^{m}(t; z, \xi)(-1)^m \cdot$$

$$\cdot \cos \eta_m x.$$

Cauchy's function

$$H_{k,k_1}(t; x, \varsigma; z, \xi) =$$

$$= \frac{2}{R} \sum_{m=0}^{\infty} \varepsilon_{k,k_1}^{m}(t, z, \xi) \cdot \cos \eta_m \xi \cdot \cos \eta_m x.$$

2. Experimental data and coefficients identification

According to the results of experimental data and using the proposed model, identification was carried out using the theory of state control of multicomponent systems (results obtained in [4]) (Fig. 2).

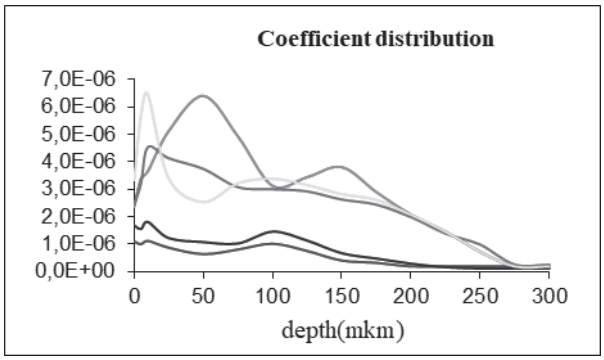

Fig. 2. Coefficients distribution at first sample

The distributions of diffusion coefficients are reproduced using methods of optimal control of the state of multicomponent transport systems, analytical solution of the model, and data of experimental observations, for the considered constituent components of nanofilms (samples of aluminum, molybdenum, silicon) (Fig. 3).

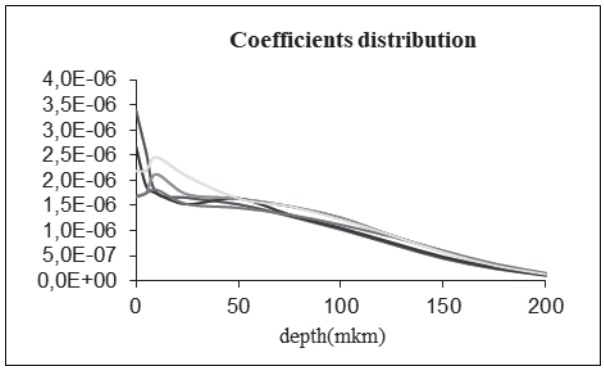

Fig. 3. Coefficients distribution at the second sample

The diffusion coefficients identified in such a way, correspond to real experimental data and are used as input parameters of the obtained mathematical solution of the model for modeling and analysis of concentration distributions of the main components of nanofilms (aluminum, molybdenum, silicon, etc.).

3. Numerical simulation

Numerical simulation results are compared with experimental observations depicting the sample content. These concentration distributions are generated for varying time intervals during the formation of the multilayer. The designated time frame corresponds to the experimental duration of three weeks. The process of forming the technological multilayer through molecular diffusion of the specified components is segmented into 5 periods, encompassing the inception of the protective multilayer from the initial period to the concluding period (Fig. 4).

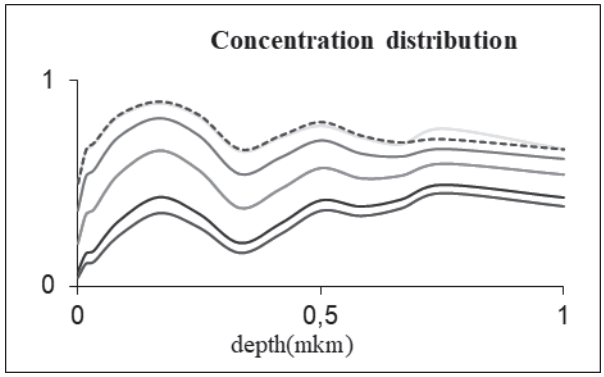

Fig. 4. Simulated concentrations for case 5

Continuous lines represent simulated data for different time durations. The dashed

line is experimentally measured data at the final duration.

As depicted in the figures above, the profiles derived from modeling closely align with the corresponding experimental profiles, particularly as the duration of multilayer formation approaches the completion period of the protective nanofilm multilayer formation. The maximum observed deviation does not surpass 2–3 %, affirming the reliability of the mathematical model and indicating the practical utility of the provided results (Fig. 5).

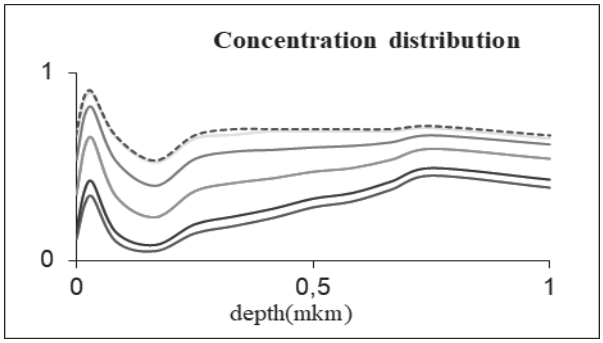

Fig. 5. Simulated concentrations

4. Software framework

The above results require complex resource-intensive calculations [5]. For this purpose, we developed a framework that automates the specified calculations with the possibility of extension to other subject areas with similar tasks of identifying the key factors of the process and further numerical modeling of the time-space characteristics using the obtained results.

The software framework in focus is implemented in Java programming language. JVM-based implementation allows us to rid of target-specific builds and follow the write-once-run-everywhere idea. At the same time, the JIT-compilation feature of JVM optimizes code execution by converting bytecode into native machine code at runtime. It allows to have runtime optimizations that are not possible with ahead-of-time compilation and are beneficial for CPU-intensive applications.

The framework design follows an object-oriented approach, and the code base is divided into major modules, such as Models, DataProcessing, Identification, and GUI. The input either can be read from the file or provided in UI. Calculated results also can be visualized or saved to the file. All components are connected with abstract interfaces, which gives the ability to easily replace one model implementation with another implementation (separation of interface and implementation).

One example is the implementation of diffusion coefficient identification, which defines the internal kinetic parameters of the process. To ascertain the coefficient distribution, we employ the gradient methods, the mathematical underpinnings of which lay in the context of parametric identification challenges in multicomponent distributed systems [6]. Tailoring our approach to the nanofilms domain, we find the method of minimum errors particularly apt. So, we picked the implementation of this method in simulations among others in the software framework. The coefficients identification algorithm is based on a gradient-identification procedure wherein, to ascertain the next approximation of the diffusion coefficient within the intraparticle space, we adhere to the following protocol (Fig. 6).

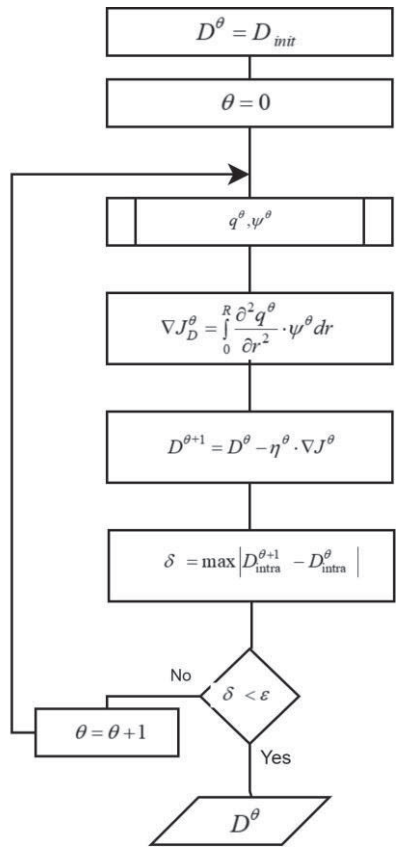

Fig. 6. Procedure for internal parameters identification

The determined distributions of diffusion coefficients within the interparticle space enable the modeling of concentration fields and integral mass distributions within the catalytic nanoporous layer with a high level of precision. As evidenced by the concentration distributions presented (Fig. 2 and Fig. 3), the profiles derived from the model closely match those obtained experimentally (Fig. 4 and Fig. 5), illustrating a noteworthy degree of consistency between the two sets of data. This alignment is further emphasized by the complete coincidence observed between the model and experimental graphs depicting the integral mass during benzene adsorption.

Conclusion

The difficulties associated with studying diffusion in multilayer films require the progress of contemporary modeling methods and software platforms to precisely represent phenomena, taking into account transitions between adjacent layers. Alongside the essential function of cutting-edge modeling techniques and software frameworks in addressing the difficulties of examining diffusion within multilayer films, it's vital to acknowledge the significant input of sophisticated computational methods.

In this paper, we endeavor to merge intricate mathematical models with optimal software development methodologies to address the challenge of simulating diffusion transport processes in multilayer nanofilms computationally. Based on the experimental findings and employing the suggested model, identification is conducted utilizing the theory of state control for multicomponent systems. Using the methods of optimal control of the state of multicomponent transport systems, the analytical solution of the model, and the data of experimental observations, the distributions of diffusion coefficients for the considered components of nanofilms (samples of aluminum, molybdenum, silicon) were reproduced. Numerical simulation results were compared with experimental observations. The profiles obtained from the modeling closely match the corresponding experimental profiles, especially as the duration of multilayer formation approaches the final stages of completing the protective nanofilm multilayer formation. The maximum observed deviation does not exceed 2–3%, confirming the reliability of the mathematical model and demonstrating the practical value of the results provided.

A software framework is developed for the automation of the specified calculations with the possibility of extension to other subject areas with similar tasks of identifying the key factors of the process and further numerical modeling of the time-space characteristics using the obtained results.

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