Modeling Diffusion Transport Mechanisms in Multilayer Nanofilms Using Computer Simulations

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Abstract

In this paper, the authors strive to integrate complex mathematical models with optimal software development practices to tackle the challenge of computationally simulating diffusion transport processes in multilayer nanofilms. Drawing on the experimental findings and using the proposed model, identification was performed using the theory of state control for multicomponent systems. Utilizing optimal control methods for multicomponent transport systems, the analytical solution of the model, and experimental observation data, the diffusion coefficient distributions for the components of the nanofilms (samples of aluminum, molybdenum, silicon) were reproduced. The numerical simulation results were compared with experimental observations. The profiles generated from the simulations closely align with the experimental profiles, particularly as the multilayer formation process approaches the final stages of completing the protective nanofilm layers. The maximum observed deviation does not exceed 2–3 %, confirming the reliability of the mathematical model and highlighting the practical value of the results presented. A software framework was developed to automate the specified calculations, with the potential for extension to other subject areas that involve similar tasks of identifying key process factors and further numerical modeling of time-space characteristics using the obtained results.

Keywords

Coefficient identification, diffusion, mathematical model, multilayer nanofilm, numerical simulation, oxide film

1. Introduction

The challenges of investigating diffusion in multilayer films necessitate advancing 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

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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, the 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 multi-component 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.

2. 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 (Figure 1).

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.

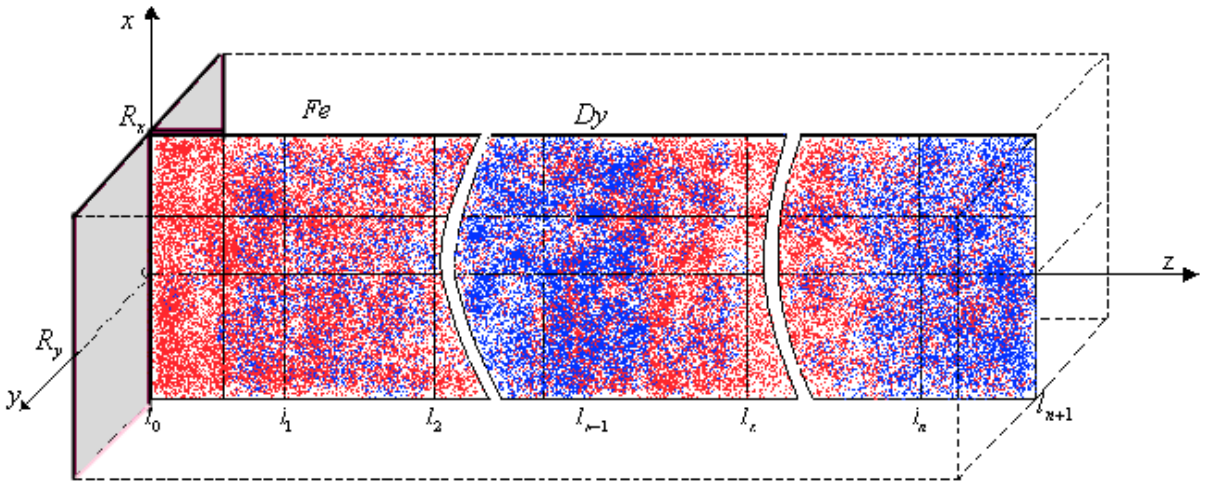

Figure 1: Schema of multilayer nanofilm.

This approach provides a mathematical model describing the diffusion transfer process within a planar multilayered medium

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

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:

$$C_k(t,x,z)\big|_{t=0}=C_{0_k}(x,z)\equiv C_{0_k}(z), \tag{2}$$
$$\left[\alpha_{11}^0\frac{d}{dz}+\beta_{11}^0\right]C_1(t,x,z)\big|_{z=l_0}=C_{l_0}(t,x);$$
$$\frac{\partial C_{n+1}}{\partial z}\big|_{z=\infty}=0;$$
$$\left[\left[\alpha_{j1}^k\frac{\partial}{\partial z}+\beta_{j1}^k\right]C_k-\left[\alpha_{j2}^k\frac{\partial}{\partial z}+\beta_{j2}^k\right]C_{k+1}\right]_{z=l_k}=0,$$

And boundary conditions for variable x

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

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

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

3. Experimental data and coefficient 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]) (Figure 2).

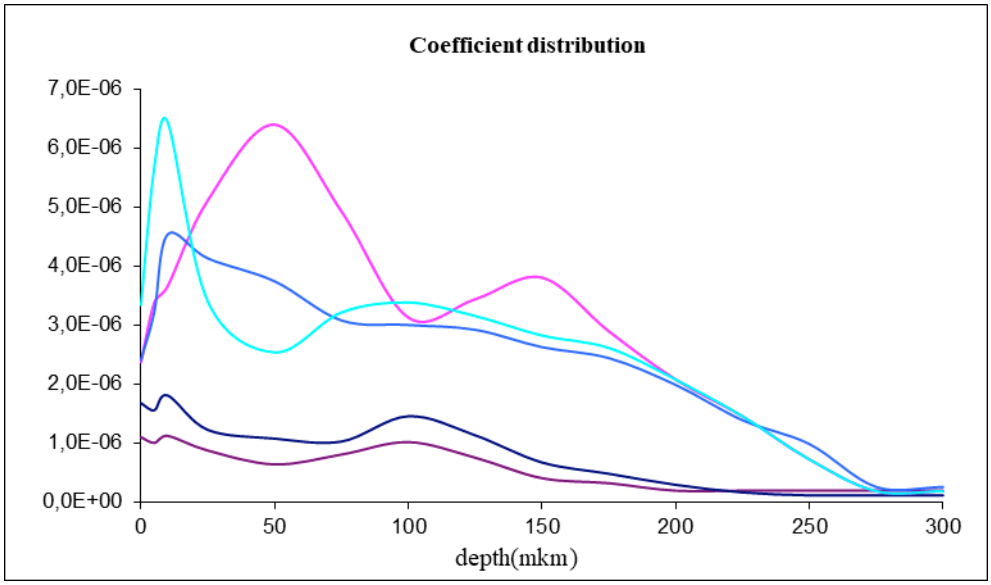

Figure 2: Coefficients distribution at first sample

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) (Figure 3).

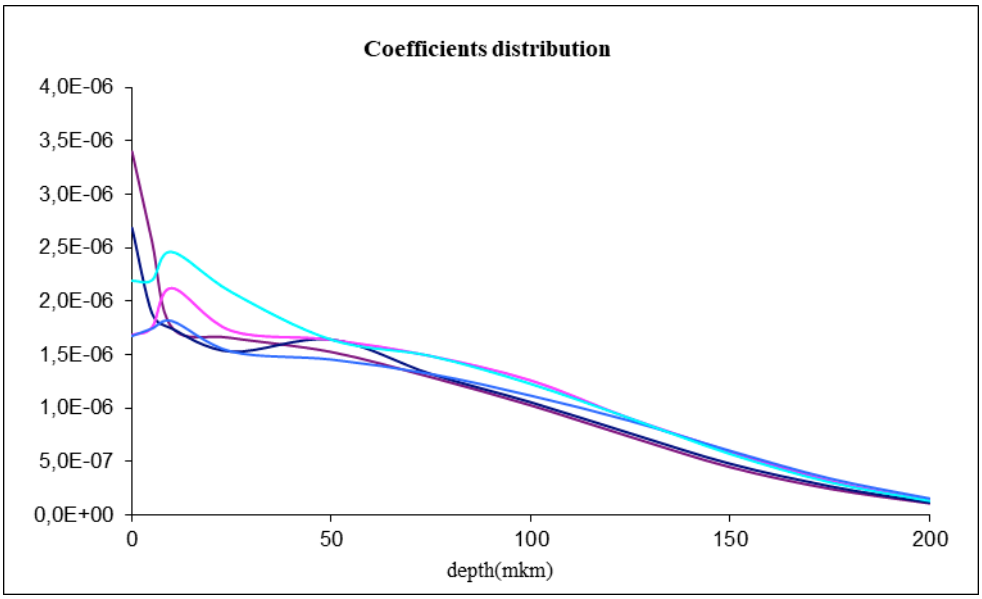

Figure 3: Coefficients distribution at the second sample

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.).

4. 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 (Figure 4).

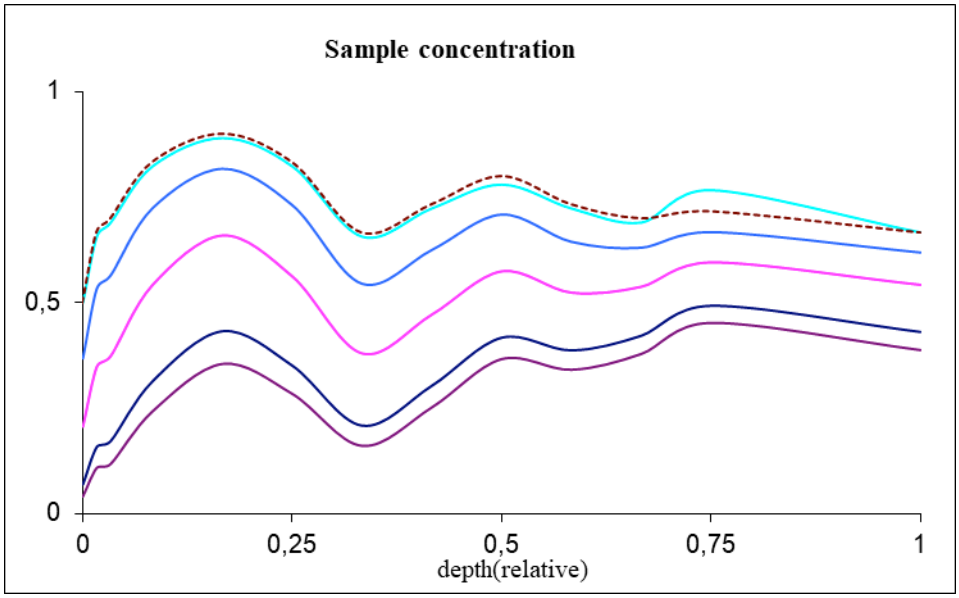

Figure 4: Simulated concentrations for case 5

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 (Figure 5 and Figure 6).

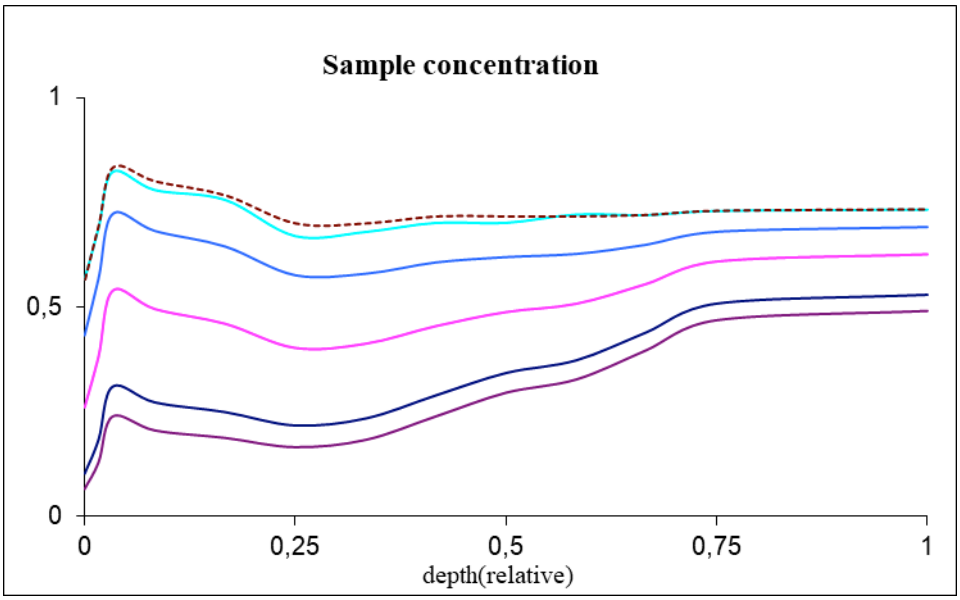

Figure 5: Simulated concentrations for case 3

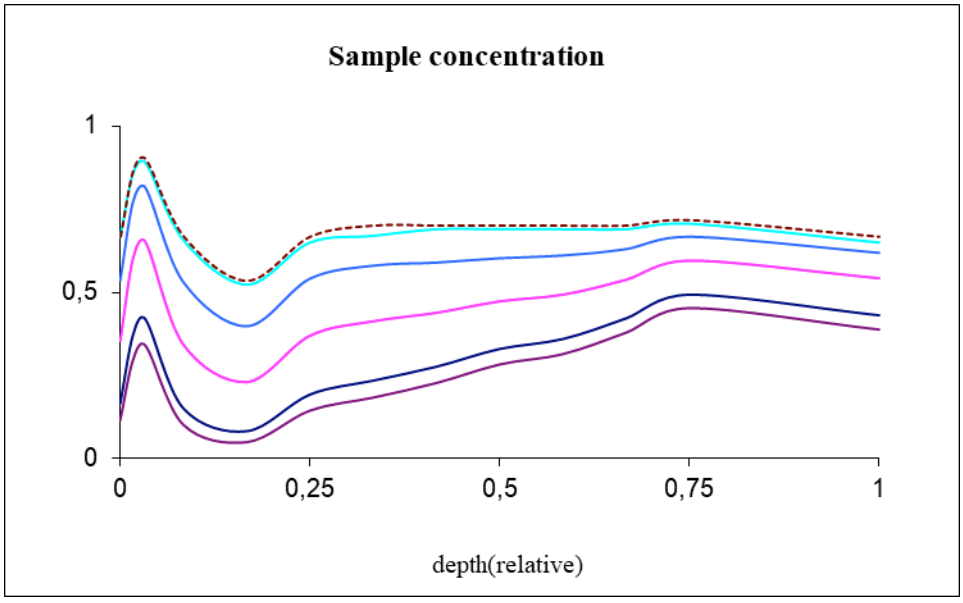

Figure 6: Simulated concentrations for case 1

5. 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. Also, using Java language allows to implement performance-effective algorithms to speed up the calculus process [6].

The software tools based on the framework are developed as a stand-alone, so it is self-sufficient and uses the resources of the system on which it is running to perform calculations, and does not require connection to external servers or systems.

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) [7].

The hierarchy of classes for the visualization of modeling results is based on the DrawArea class, which contains the basic elements for displaying graphics (Figure 7). Consequently, the graphical user interface can be adapted to the needs of the user and can show visualizations of the modeled parameters in the desired directions.

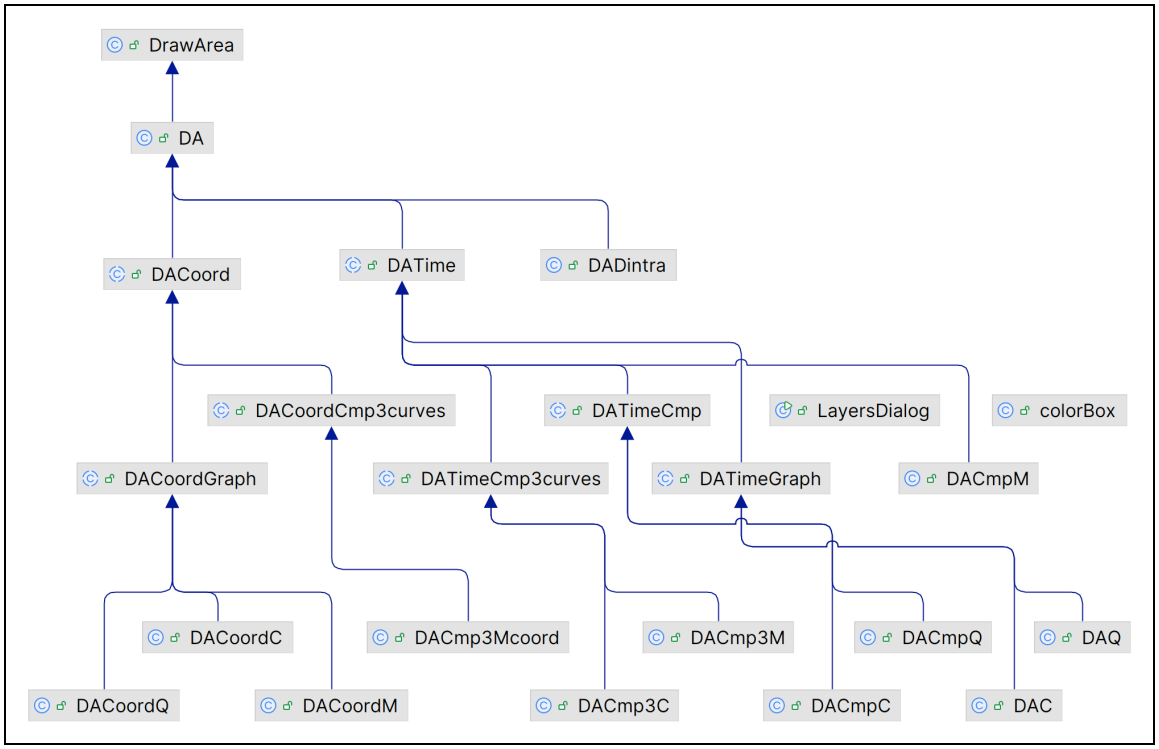

Figure 7: Procedure for internal parameters identification

For modeling implementation, the other hierarchy of classes is designed and begins with the DMTBase class (Figure 8). The hierarchy contains models of different complexity and is compatible with visualization classes (Figure 7) through the implementation of the DMTDraweble interface. Such an approach allows you to experiment, quickly replace one model with another, and find out how changes in model parameters or features will affect changes in the dynamics of certain kinetic characteristics of the modeled system.

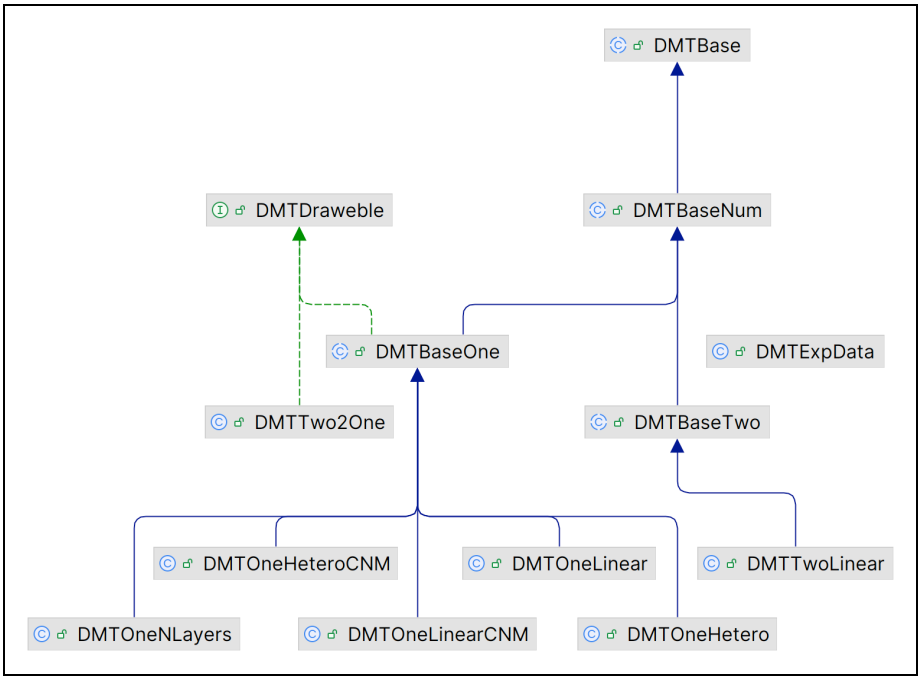

Figure 8: Procedure for internal parameters identification

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 [8]. 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 coefficient 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 (Figure 9).

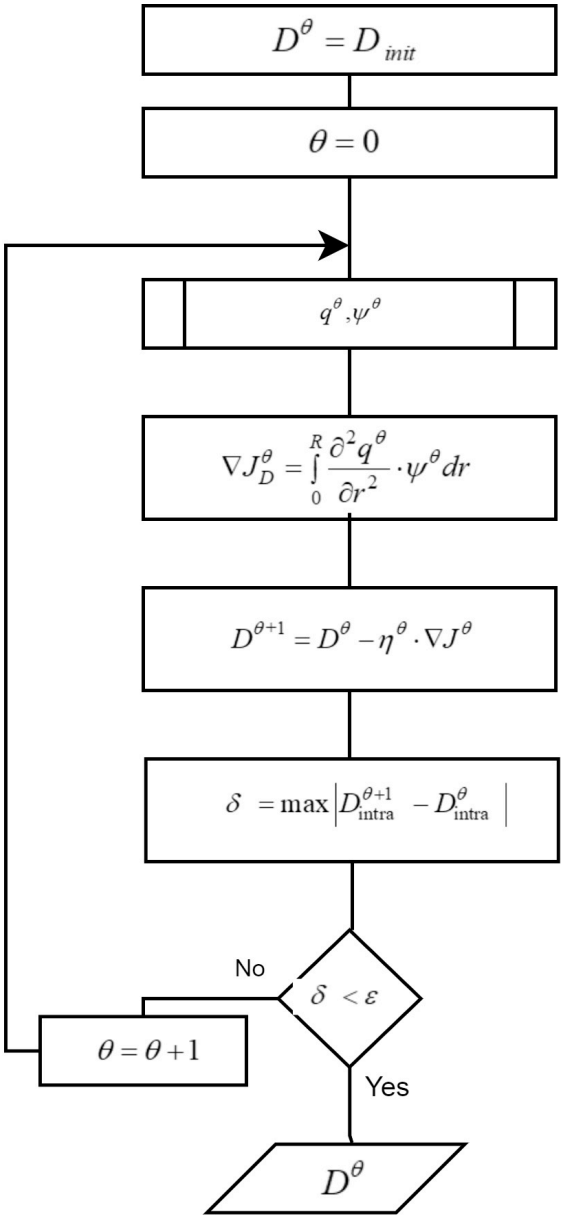

Figure 9: Procedure for internal parameters identification

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 (Figure 2 and Figure 3), the profiles derived from the model closely match those obtained experimentally (Figure 4 and Figure 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.

6. 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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