



DEVELOPMENT OF A REFERENCE DYNAMIC MODEL OF AMMONIA SYNTHESIS PROCESS UNDER UNCERTAINTY

Khalilov A.J. [0000-0002-6646-2174]

Senior lecturer of the Department of Agronomy Navoi State University of Mining and Technologies

Annotation. In this article, research on the control effects of the technological process of ammonia synthesis under conditions of uncertainty, the output coordinates and the structure of interrelationships between the disturbances are analyzed, and the generalized equations for the output coordinate of the control object are presented. A generalized dynamic structural diagram of the control object corresponding to these equations is created. Also, on the basis of the results of the conducted experiments, a standard dynamic model of the control object was developed in the technological process of ammonia synthesis in the conditions of uncertainty, and the identification of the control object was carried out based on the time and frequency characteristics.

Key words: dynamic modeling, transfer function, control object, identification, time characteristics, frequency characteristics, reference model.

Annotatsiya. Maqolada noaniqlik sharoitida ammiak sintezi texnologik jarayonining boshqaruv ta'sirlari, chiqish koordinatalari va g'alayonlar orasidagi o'zaro munosabatlar strukturasi haqida tadqiqotlar tahlil qilinib, boshqaruv ob'ektning chiqish koordinatasi uchun umumlashgan tenglamalar keltirilgan. Boshqaruv ob'ektining ushbu tenglamalarga mos keladigan umumlashtirilgan dinamik struktur sxemasi yaratilgan. Shuningdek, o'tkazilgan tajribalar natijalari asosida olingan ma'lumotlar negizida noaniqlik sharoitida ammiak sintezi texnologik jarayonida boshqaruv ob'ektining etalonli dinamik modeli ishlab chiqildi va vaqt hamda chastotali xarakteristikalari asosida boshqaruv ob'yekti indentifikatsiyasi amalga oshirilgan.

Kalit so'zlar: dinamik modellashtirish, uzatish funksiyasi, boshqaruv ob'yekti, identifikatsiya, vaqt xarakteristikalari, chastotali xarakteristikalar, etalonli model.

Аннотация. В данной статье исследованы эффекты управления технологическим процессом синтеза аммиака в условиях неопределенности, проанализированы выходные координаты и структура взаимосвязей между возмущениями, а также представлены обобщенные уравнения для выходной координаты объекта управления. Создается обобщенная динамическая структурная схема объекта управления, соответствующая этим уравнениям. Также на основе результатов проведенных экспериментов была разработана типовая динамическая модель объекта управления в технологическом процессе синтеза аммиака в условиях неопределенности и проведена идентификация объекта управления по времени и частотные характеристики.

Ключевые слова: динамическое моделирование, передаточная функция, объект управления, идентификация, временные характеристики, частотные характеристики, эталонная модель.

Introduction

Ammonia synthesis stands as a cornerstone of modern industrial processes, playing an indispensable role across various sectors, including agriculture, pharmaceuticals, and chemical manufacturing. Globally, it stands as one of the most produced chemicals, with a pivotal position in sustaining agricultural productivity through the production of fertilizers and as a vital component in the synthesis of numerous chemicals essential for modern life. This pivotal role underscores the necessity for continual advancements in process understanding and optimization to meet the evolving demands of society and industry [1-4].

The significance of accurate dynamic models in chemical processes cannot be overstated. While static models provide valuable insights into steady-state behavior, they often fall short in capturing the intricate dynamics inherent in complex chemical reactions, such as those involved in ammonia synthesis. Understanding system behavior under uncertainty is particularly critical in optimizing process efficiency and ensuring operational



stability. Uncertainties in input parameters, such as feedstock composition, temperature, and pressure, can significantly impact process performance and product quality, necessitating the development of dynamic models capable of accommodating these uncertainties [6-10].

Hence, the primary motivation of this study is to address the pressing need for accurate dynamic models in the realm of ammonia synthesis. By developing a comprehensive dynamic model that accounts for uncertainties in input parameters and operating conditions, this research aims to enhance our understanding of the dynamic behavior of the ammonia synthesis process. Through this endeavor, we seek to not only optimize process efficiency but also contribute to the development of robust control strategies and risk management techniques [11-18].

The overarching objective of this study is therefore to develop a dynamic model for the ammonia synthesis process that effectively captures its dynamic behavior under uncertainty. By integrating advanced modeling techniques with rigorous uncertainty analysis, we aim to provide a versatile tool for engineers and researchers to simulate and optimize the ammonia synthesis process in diverse operating conditions and scenarios. Through this research, we aim to pave the way for enhanced process control, improved operational efficiency, and sustainable production practices in the realm of ammonia synthesis.

Methodology

Dynamic model chemical processes. The dynamic model for the ammonia synthesis process under uncertainty is developed based on fundamental chemical engineering principles and reaction kinetics. The mathematical formulation of the model involves representing the mass and energy balances of the reactor system, considering the complex network of reactions involved in ammonia synthesis, including the Haber-Bosch process.

The model incorporates the kinetics of individual reactions, accounting for factors such as temperature, pressure, and reactant concentrations. The rate equations for each reaction are derived from established kinetic models available in literature [1-2]. These rate equations are integrated into a system of ordinary differential equations (ODEs) to describe the time evolution of reactant and product concentrations within the reactor.

These rate equations describe the rate of formation or consumption of reactants and products as a function of temperature, pressure, and reactant concentrations. For example, the rate equation for the Haber-Bosch reaction can be expressed as:

$$r_{NH_3} = k \cdot P_{NH_3}^{(eq)} - k \cdot P_{N_2} \cdot P_{H_2}^3$$

where r_{NH_3} represents the rate of ammonia formation, k is the rate constant, $P_{NH_3}^{(eq)}$ is the equilibrium partial pressure of ammonia, P_{N_2} and P_{H_2} are the partial pressures of nitrogen and hydrogen, respectively.

These rate equations are integrated into a system of ordinary differential equations (ODEs) to describe the time evolution of reactant and product concentrations within the reactor over time. The ODEs are solved numerically using appropriate numerical integration techniques, such as the Runge-Kutta method, to simulate the dynamic behavior of the system.

The mathematical formulation also considers the effects of uncertainties in input parameters, such as feedstock composition, temperature, pressure, and kinetic rate constants. Uncertainties are incorporated into the model using probabilistic methods, such as Monte Carlo simulation or Latin hypercube sampling, to generate random samples from probability distributions assigned to uncertain parameters.

The rate of each elementary step in the reaction mechanism is governed by kinetic rate laws that describe the dependence of reaction rates on reactant concentrations,



temperature, and catalyst activity. For example, the rate of ammonia formation can be described by the Langmuir-Hinshelwood rate law [1]:

$$r_{NH_3} = k \cdot \frac{k_{N_2} \cdot P_{N_2} \cdot k_{H_2}^3 \cdot P_{H_2}^3}{(1 + k_{N_2} \cdot P_{N_2} + K_{H_2} \cdot P_{H_2}^3)^2}$$

where r_{NH_3} represents the rate of ammonia formation, k is the rate constant, K_{N_2} and K_{H_2} are the adsorption equilibrium constants for nitrogen and hydrogen, and P_{N_2} and P_{H_2} are the partial pressures of nitrogen and hydrogen, respectively.

Overall, the mathematical formulation of the dynamic model aims to accurately capture the complex kinetics and dynamics of the ammonia synthesis process, while also accounting for uncertainties to provide robust predictions under varying operating conditions.

Mass and Energy Balances. The mathematical model incorporates mass and energy balances to describe the conservation of mass and energy within the reactor system. The mass balance equation accounts for the accumulation and depletion of nitrogen, hydrogen, and ammonia species, considering reaction kinetics and mass transfer effects.

The mass balance equation for the Haber-Bosch process in ammonia synthesis can be expressed as [3]:

$$\frac{d[NH_3]}{dt} = r_{NH_3}V$$

where NH_3 represents the concentration of ammonia, r_{NH_3} denotes the rate of ammonia formation, and V signifies the volume of the reactor.

The energy balance equation accounts for the heat transfer and chemical reaction within the system [3]:

$$C_p \frac{dT}{dt} = \dot{Q} + \Delta H_{rxn} r_{NH_3} V$$

where C_p represents the heat capacity of the system, T is the temperature, \dot{Q} denotes the heat flux, and ΔH_{rxn} represents the heat of reaction.

Similarly, the energy balance equation governs the distribution of heat within the reactor, accounting for heat generation due to reaction kinetics and heat transfer through the reactor walls.

Kinetic Rate Laws for Ammonia Formation. The kinetics of ammonia formation are described using rate laws that capture the dependency of reaction rates on reactant concentrations, temperature, and pressure. The rate of ammonia synthesis is typically modeled using the Langmuir-Hinshelwood mechanism, which accounts for the adsorption of reactant molecules onto the catalyst surface and subsequent surface reactions leading to product formation. The rate constants and adsorption parameters are determined based on experimental data and theoretical considerations.

The rate of ammonia formation follows the kinetics described by the Haber-Bosch mechanism [4]:

$$r_{NH_3} = k_f[N_2][H_2] - k_r[NH_3]$$

where k_f and k_r represent the forward and reverse rate constants, respectively, and $[N_2]$ and $[H_2]$ denote the concentrations of nitrogen and hydrogen, respectively.

Boundary Conditions and Initial Values. Boundary conditions specify the behavior of the system at the boundaries of the reactor domain, while initial values define the initial state of the system at the start of the simulation. Boundary conditions may include specifying inlet concentrations and temperatures of reactants, as well as outlet conditions such as pressure and flow rates. Initial values are typically set based on experimental or theoretical estimates of reactor startup conditions.

Boundary conditions specify the initial and boundary values of concentrations and temperature within the reactor system. For instance, at the reactor inlet:

$$[N_2] = [N_2]_{in} \quad [H_2] = [H_2]_{in} \quad T = T_{in}$$

and at the reactor outlet:

$$[NH_3] = [NH_3]_{out} \quad T = T_{out}$$

Initial values for concentrations and temperature are typically specified as [1]:

$$[N_2]_0 = [N_2]_{in} \quad [H_2]_0 = [H_2]_{in} \quad [NH_3]_0 = 0 \quad T_0 = T_{in}$$

Development of a dynamic model of ammonia synthesis process under uncertainty. The research on the control effects of the technological process of ammonia synthesis under conditions of uncertainty, the output coordinates and the structure of interrelationships between the disturbances is analyzed in [5], and the generalized equations for the output coordinate of the control object are given as follows:

$$y^{[k]} = y^{u[i][k]} + y^{f[j][k]}, \quad i = \overline{1,4}, \quad j = \overline{1,3}, \quad k = \overline{1,4}$$

here the following designations are included for the control object:

$y^{[k]}$ - output coordinate (catalyst temperature) in the kth layer of the ammonia synthesis column;

$y^{u[i][k]}$ – the component of the output coordinate due to the i-control effect (current consumption passing through the bypass valve);

$y^{f[j][k]}$ – output coordinate component based on the j-turbulent impact condition [5].

A generalized structural scheme of the control object corresponding to these equations is presented in Figure 1.

In Figure 1, $W_o^{u[i][k]}$ is the transfer function representing the influence of the i-control effects on the k-control channel of the ammonia synthesis technological process under conditions of uncertainty, and the transfer function $W_o^{f[j][k]}$ is the influence on the k-control channel based on the j-turbulent influence conditions, respectively.

For this model, the transfer functions of the technological process of ammonia synthesis under uncertainty can be given as follows:

$$W_o^{u[k][j]}(z) = \frac{\sum_{h=1}^{k_o^{u[k][j]}+1} b_h^{u[k][j]} \cdot z^{-h-d_o^{u[k][j]}}}{1 - \sum_{h=1}^{n_o^{u[k][j]}} a_h^{u[k][j]} \cdot z^{-h}}, \quad k = \overline{1,4}, \quad j = \overline{k,4}$$

Results

Provide simulation results demonstrating the dynamic behavior of the ammonia synthesis process under different scenarios of uncertainty.

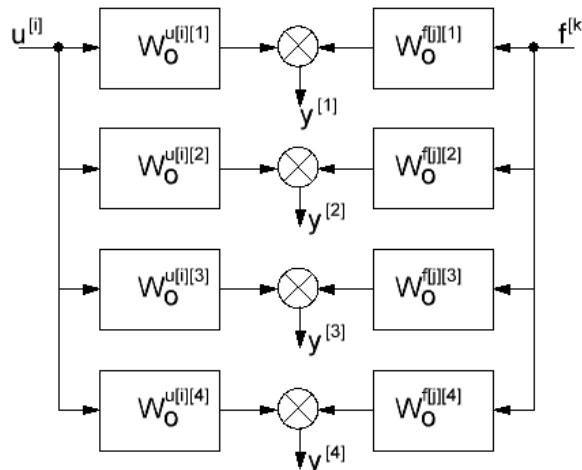


Figure 1. A generalized structural scheme of the control object.

From these transfer functions, we construct a structural dynamic scheme of the ammonia synthesis column model for the technological process of ammonia synthesis under conditions of uncertainty and a scheme of control objects identification (Figure 2):

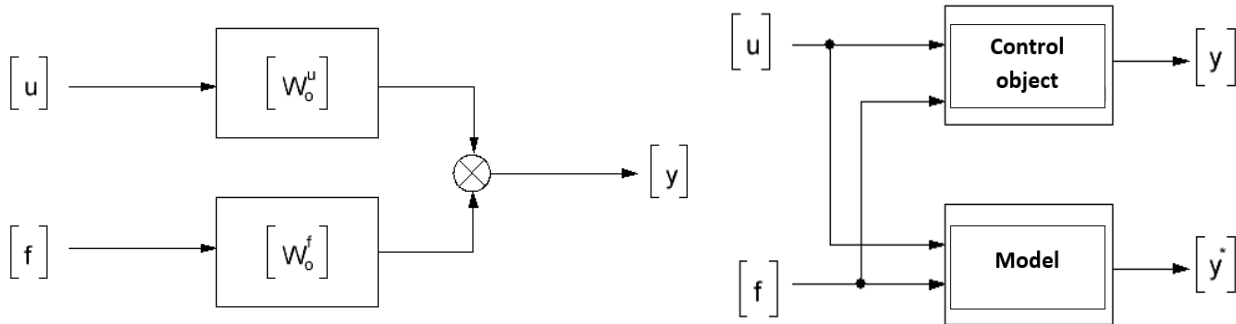


Figure 2. Structural schemes of the control object.

Based on these structural schemes, the inputs of the control object and the model in the technological process of ammonia synthesis under conditions of uncertainty are given the same impact signal and turbulent impact signals.

The main goal of control object identification is the transfer function $W_o^{u[i][k]}$ representing the influence of the i -control effects of the technological process of ammonia synthesis on the k -control channel under conditions of uncertainty, and the transfer function on the k -control channel based on the j -turbulent influence conditions, respectively the transfer function $W_o^{f[j][k]}$ representing the effect is to minimize such a set of coefficients that the difference between the experimental output coordinates of the real process $[y]$ and the result of the dynamic model $[y^*]$ for the transfer functions is minimized:

$$[\Delta] = |[y] - [y^*]| \rightarrow \min$$

On the basis of the data obtained on the basis of the results of the conducted experiments, a standard dynamic model of the control object in the technological process of ammonia synthesis was developed in the conditions of uncertainty.

Simulation Results: On the basis of the data obtained on the basis of the results of the conducted experiments, a reference dynamic model of the control object in the technological process of ammonia synthesis was developed in the conditions of uncertainty. The time characteristics of this reference dynamic model were obtained using the functions of the Linear Analysis extended package included in Matlab [19-23]:

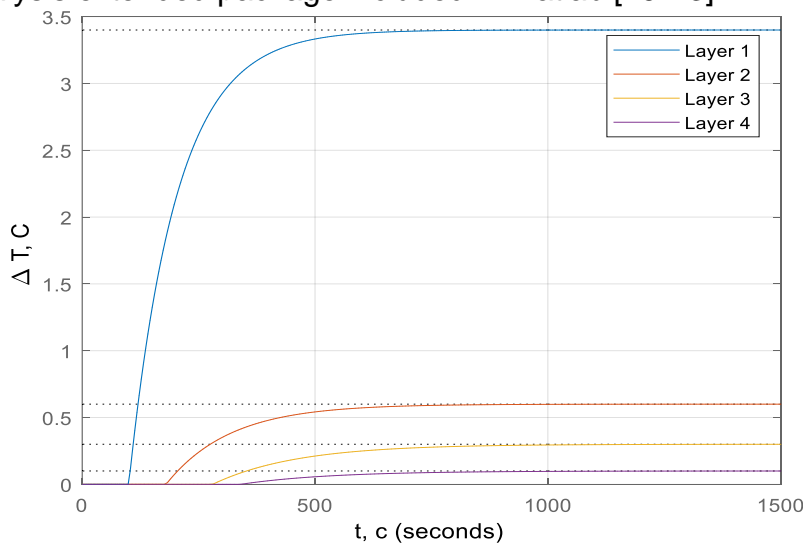


Figure 3. Dynamics of catalyst temperature change by layer during the technological process of ammonia synthesis when the first bypass valve is fully opened.

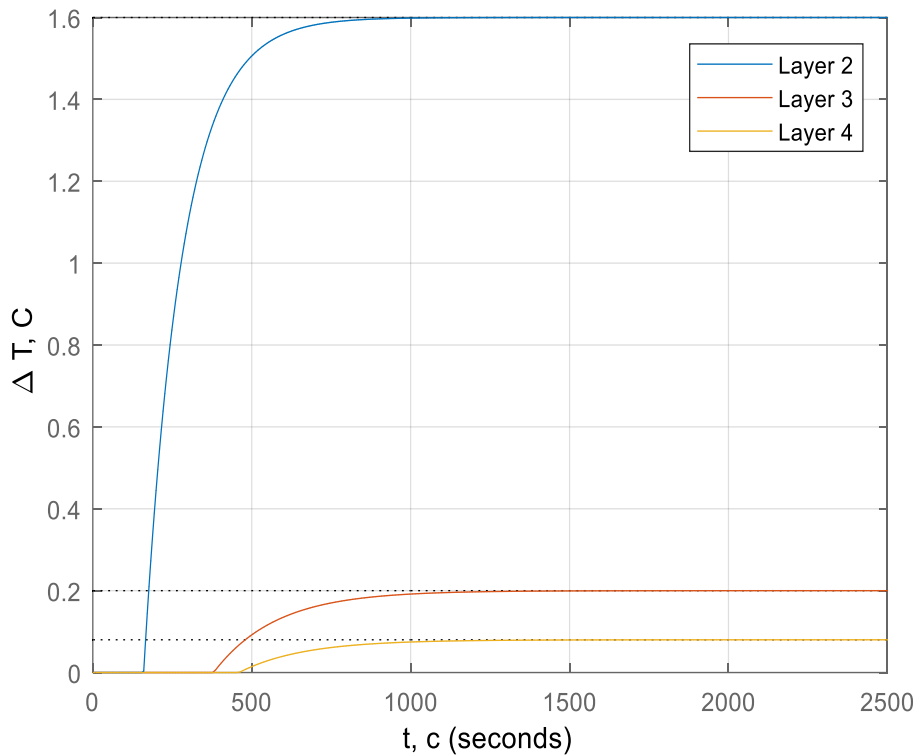


Figure 4. Dynamics of catalyst temperature change by layer in the process of ammonia synthesis with the second bypass valve fully open.

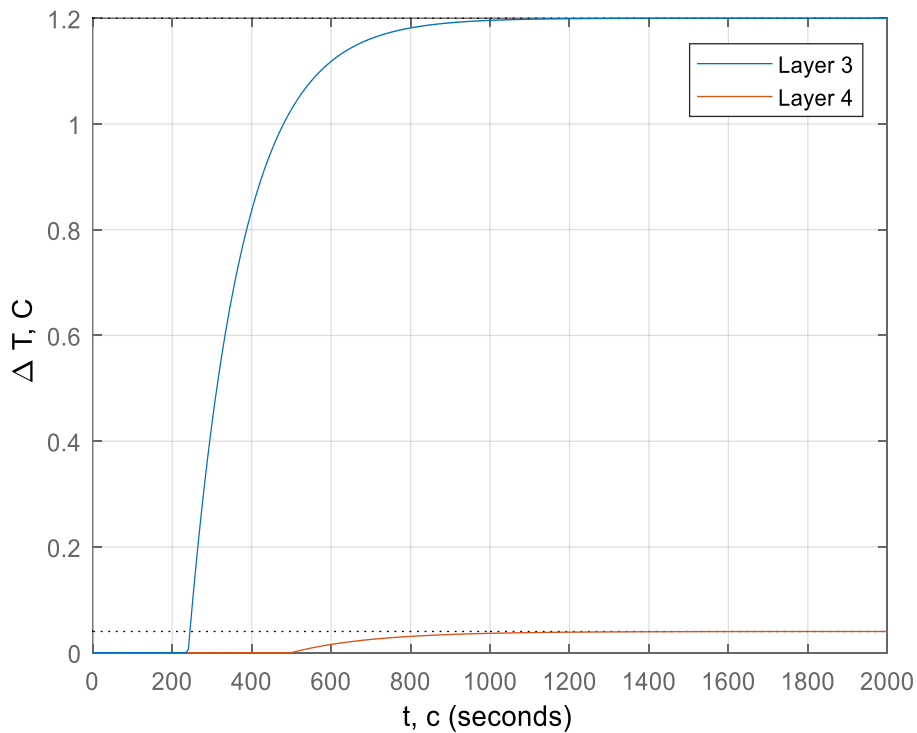


Figure 5. Dynamics of catalyst temperature change by layer in the technological process of ammonia synthesis when the third bypass valve is fully opened.

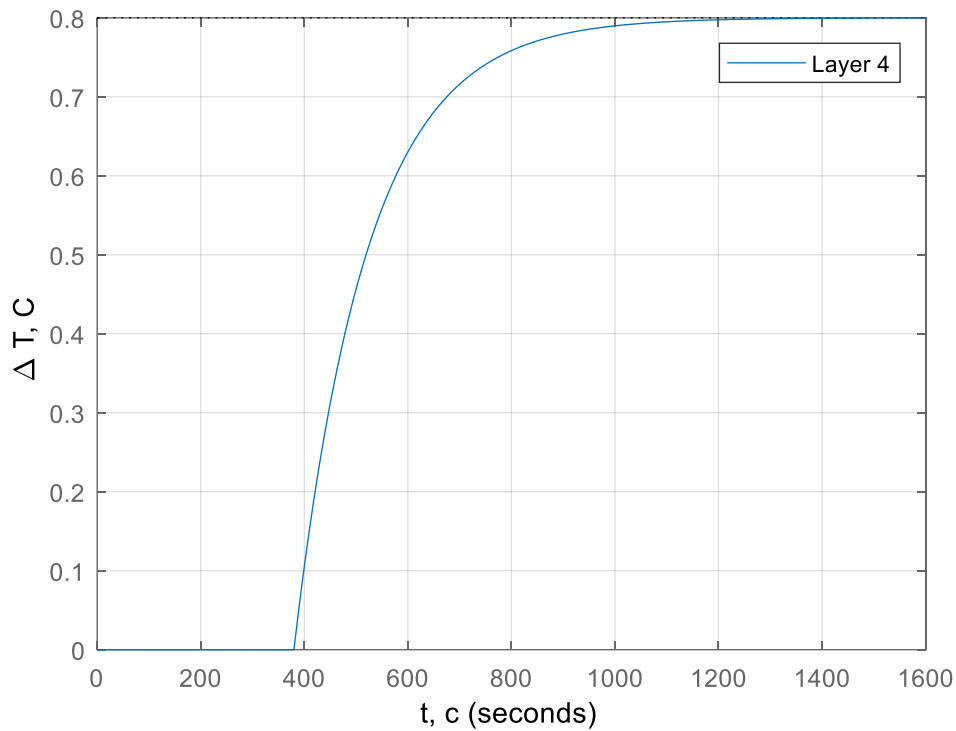


Figure 6. Dynamics of catalyst temperature change by layer during the process of ammonia synthesis with the fourth bypass valve fully open.

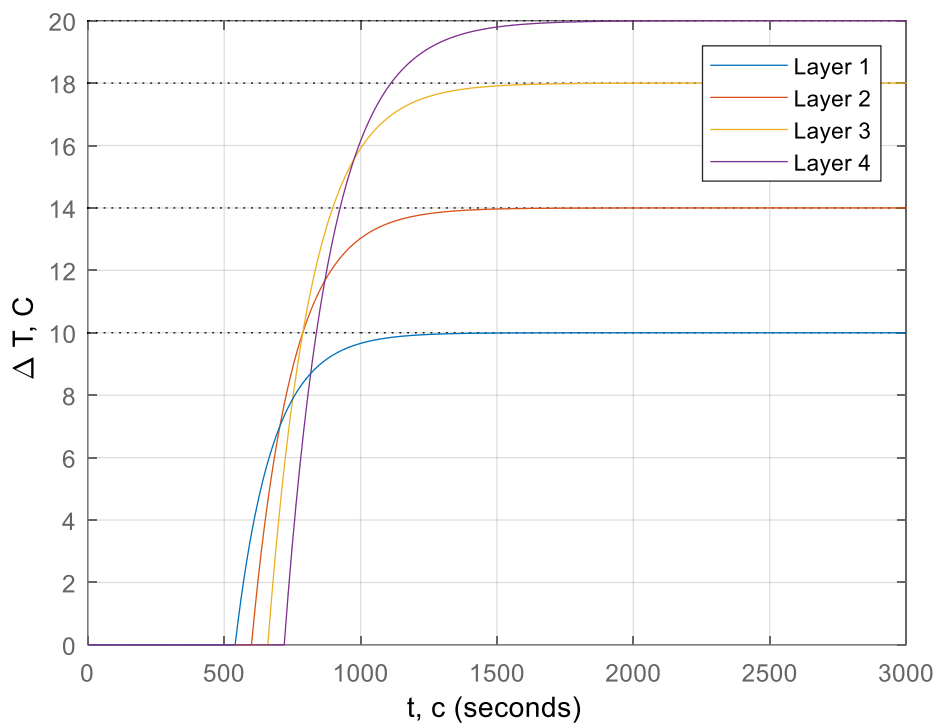


Figure 7. Dynamics of catalyst temperature changes across layers in an ammonia synthesis column as a result of a 10% increase in the nitrogen-hydrogen mixture ratio.

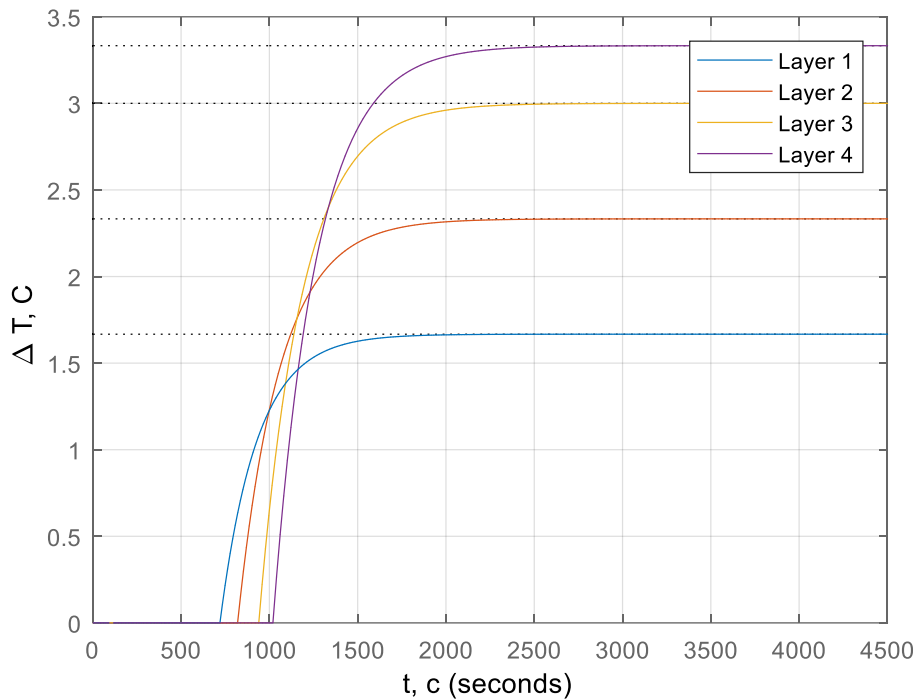


Figure 8. Dynamics of catalyst temperature changes across layers in an ammonia synthesis column as a result of a 10% increase in ammonia concentration.

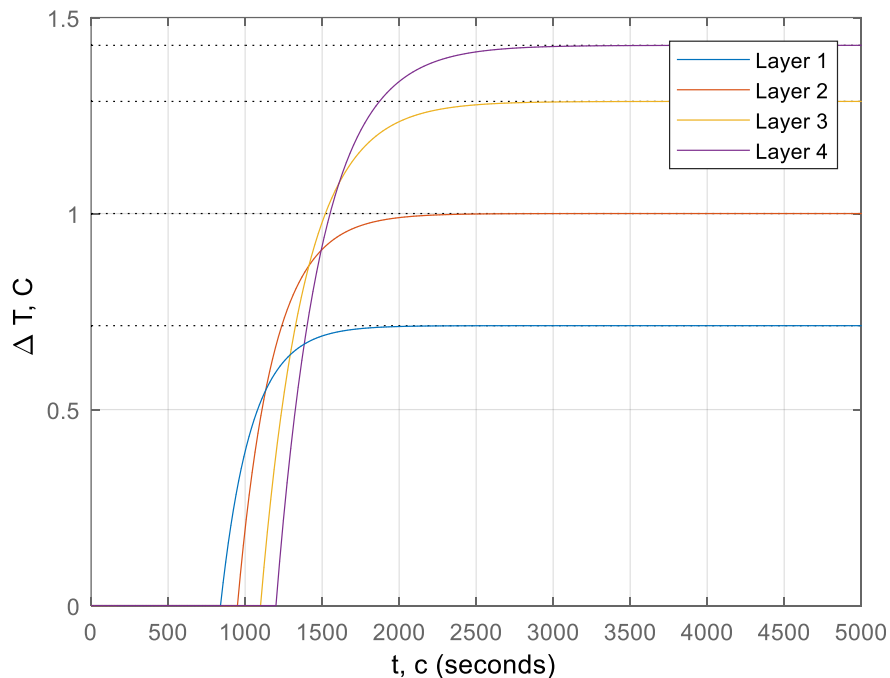


Figure 9. Dynamics of catalyst temperature changes across layers in an ammonia synthesis column due to an increase in the concentration of inert compounds.

The frequency characteristics of this reference dynamic model were obtained using the functions of the Linear Analysis extended package included in Matlab:

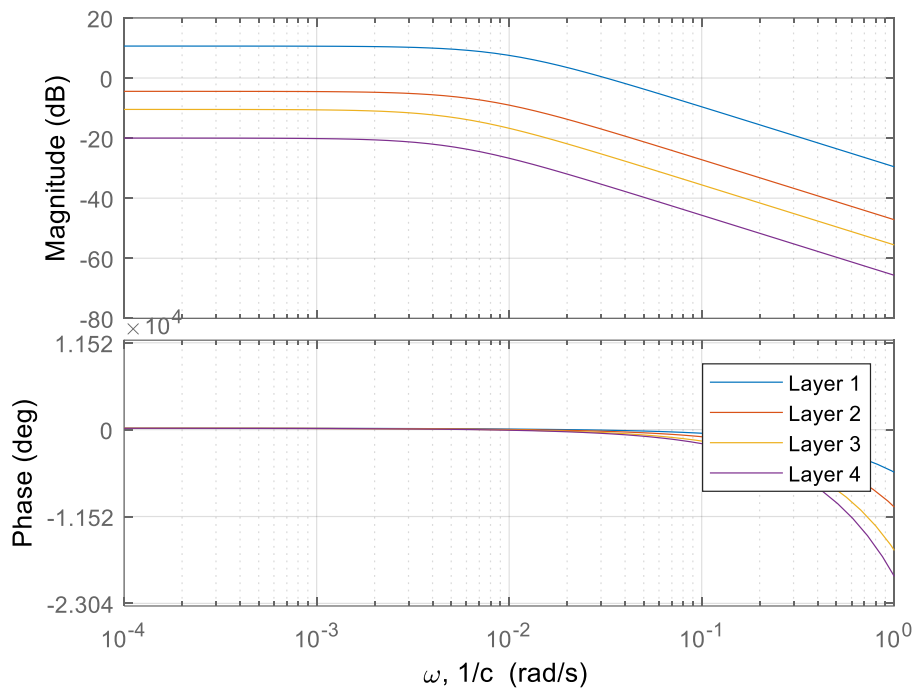


Figure 10. Frequency characteristics of temperature deviations in the technological process of ammonia synthesis as a result of harmonic changes of the first bypass valve.

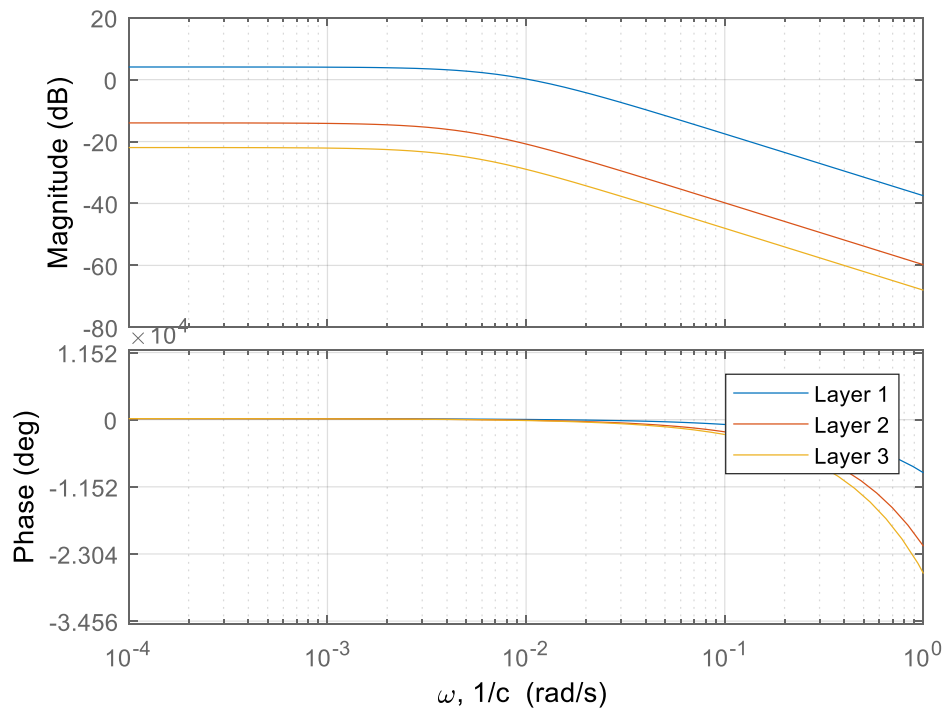


Figure 11. Frequency characteristics of temperature deviations in the technological process of ammonia synthesis as a result of harmonic changes of the second bypass valve.

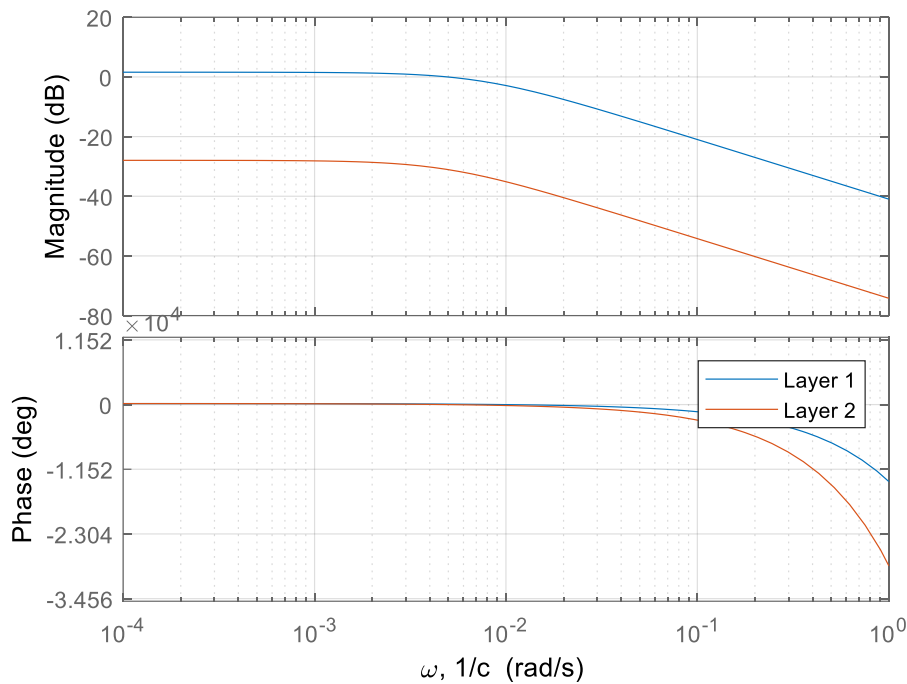


Figure 12. Frequency characteristics of temperature deviations in the technological process of ammonia synthesis as a result of harmonic changes of the third bypass valve.

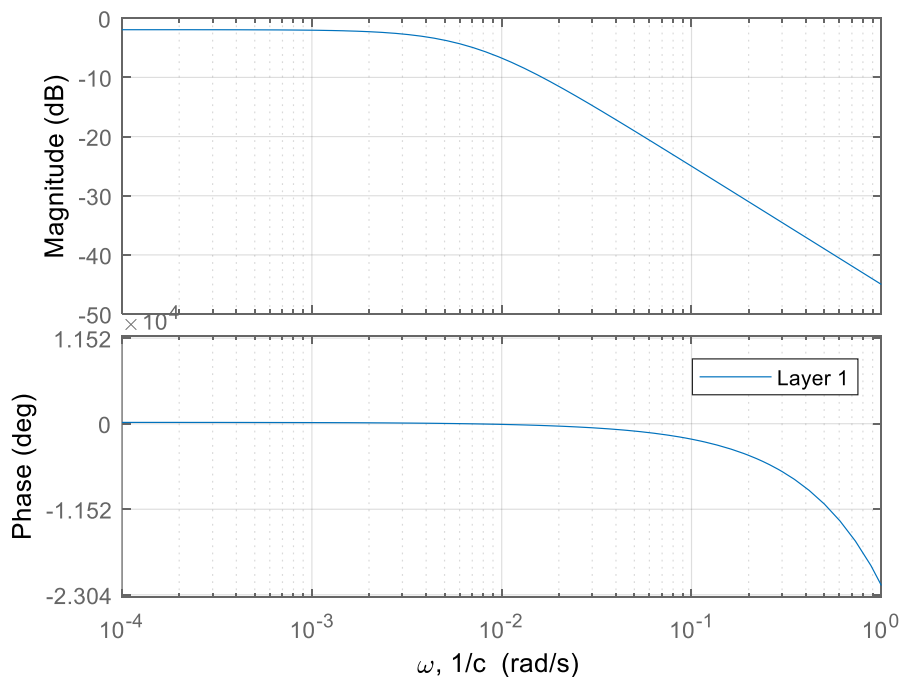


Figure 13. Frequency characteristics of temperature deviations in the technological process of ammonia synthesis as a result of harmonic changes of the fourth bypass valve.

Discussion

Interpretation of Results: The simulation results obtained from the developed dynamic model of the ammonia synthesis process offer profound insights into various facets



crucial for process optimization, control strategies, and risk assessment. Firstly, in terms of process optimization, the dynamic model allows for a more comprehensive understanding of how different operational parameters interact over time. By considering the dynamic behavior of the system, it becomes possible to identify optimal operating conditions that can enhance both the efficiency and yield of ammonia production. For instance, the model elucidates the time-dependent effects of temperature, pressure, and catalyst activity on the synthesis reaction kinetics, enabling the formulation of strategies to maximize production while minimizing energy consumption and raw material usage.

Furthermore, regarding control strategies, the dynamic model provides a valuable tool for designing robust control systems capable of effectively regulating the synthesis process under varying operating conditions and disturbances. By accurately capturing the transient responses and nonlinear dynamics inherent in the system, the model facilitates the development of advanced control algorithms, such as model predictive control (MPC), adaptive control, or cascade control, to maintain process stability and optimize performance in real-time.

In terms of risk assessment, the dynamic model offers insights into the potential consequences of deviations from optimal operating conditions or unexpected disturbances on process safety and reliability. By simulating various scenarios and assessing their impacts on key performance metrics, such as ammonia yield, reactor temperature, and pressure profiles, the model enables proactive risk mitigation strategies to be implemented, including alarm systems, emergency shutdown procedures, and contingency plans, to prevent or mitigate potential hazards and ensure operational integrity.

Comparison with Existing Models: A comparative analysis between the proposed dynamic model and existing static or deterministic models reveals significant advantages in capturing system dynamics and uncertainties. Unlike static models, which assume steady-state conditions and neglect temporal variations in process variables, the dynamic model incorporates time-dependent equations that accurately represent the transient behavior of the synthesis reactor over time. This enables a more realistic simulation of dynamic processes such as startup/shutdown procedures, load changes, and disturbances, which are essential for assessing the dynamic performance and controllability of the system.

Moreover, compared to deterministic models that rely on precise input data and deterministic assumptions, the dynamic model explicitly accounts for uncertainties and variability inherent in the synthesis process, such as fluctuations in feed composition, catalyst activity, and ambient conditions. By incorporating probabilistic distributions or stochastic parameters, the model provides a more realistic representation of the inherent variability in process inputs and outputs, allowing for robust sensitivity analysis and probabilistic risk assessment.

Overall, the proposed dynamic model offers a superior framework for understanding and analyzing the complex dynamics and uncertainties associated with the ammonia synthesis process, thereby enhancing its predictive accuracy, reliability, and applicability in practical engineering applications.

Limitations and Future Directions: Despite its advancements, the current study has several limitations that warrant consideration for future research. Firstly, the dynamic model may still rely on simplifying assumptions or empirical correlations for certain process kinetics or thermodynamic properties, which could introduce uncertainties or inaccuracies in the simulation results. Future efforts should focus on improving the fidelity of the model by incorporating more rigorous kinetic models, accurate thermodynamic data, and validated experimental data to enhance its predictive accuracy and robustness.

Additionally, the dynamic model may overlook certain process complexities or nonlinearities that could affect its performance under extreme operating conditions or unforeseen scenarios. Future research directions could involve the development of more sophisticated modeling techniques, such as computational fluid dynamics (CFD), multi-scale



modeling, or hybrid modeling approaches, to capture the intricacies of mass and heat transfer, fluid dynamics, and chemical reactions within the synthesis reactor more comprehensively.

Furthermore, the applicability of the dynamic model may be limited by computational constraints or data availability, particularly for real-time process monitoring, control, and optimization in industrial settings. Future studies should explore strategies to enhance the computational efficiency and scalability of the model, such as parallel computing, reduced-order modeling, or data-driven approaches, to enable its seamless integration into process control systems and decision-making frameworks.

In conclusion, by addressing these limitations and pursuing the aforementioned research directions, future studies can advance the state-of-the-art in dynamic modeling of ammonia synthesis processes, thereby facilitating more efficient, reliable, and sustainable production of ammonia for various industrial applications.

Conclusion

In summary, this study has presented the development of a dynamic model for the ammonia synthesis process, considering uncertainties inherent in the system. Through rigorous simulation and analysis, key insights have been gained into the dynamic behavior of the process, highlighting the importance of dynamic modeling in capturing transient phenomena and uncertainties that static models fail to address adequately. The dynamic model offers a more comprehensive understanding of the synthesis process, enabling optimization strategies and control measures to be devised to enhance efficiency and reliability.

The developed dynamic model holds significant practical implications for industrial applications within the realm of ammonia synthesis. By accurately representing the dynamic behavior and uncertainties of the process, the model serves as a valuable tool for process engineers and operators in optimizing plant performance, maximizing ammonia yield, and minimizing energy consumption and raw material usage. Moreover, the insights gained from the dynamic model facilitate the design and implementation of robust control strategies to maintain process stability and ensure safe and efficient operation under varying operating conditions and disturbances. Overall, the adoption of dynamic modeling techniques in industrial settings can lead to improved process efficiency, reliability, and profitability in ammonia production facilities.

In conclusion, the development of a dynamic model for the ammonia synthesis process represents a significant advancement in the field of chemical engineering. By accounting for the dynamic nature and uncertainties inherent in the process, dynamic modeling offers a more realistic and insightful approach to understanding and optimizing complex chemical processes like ammonia synthesis. As the demand for ammonia continues to rise across various industrial sectors, the significance of dynamic modeling in enhancing process efficiency, reliability, and sustainability cannot be overstated. Moving forward, continued research and development in dynamic modeling techniques will be essential for further advancing our understanding and optimization of chemical processes, ultimately driving innovation and progress in the field of chemical engineering.

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