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Thermoelectrochemical processes in lithium-ion batteries: modeling and analysis

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Abstract Problem. *Thermoelectrochemical processes in a lithium-ion battery with a graphite anode and an NMC-type cathode were investigated. A mathematical model based on a system of nonlinear partial differential equations was proposed, taking into account charge, heat, and mass transfer processes in the electrodes and electrolyte. The model incorporates electrochemical kinetics described by the Butler–Volmer equation, lithium-ion diffusion, potential distribution, and thermal effects caused by Joule heating and electrochemical reactions. Based on the numerical solution of the DFN model with an integrated thermal module, the spatiotemporal distributions of temperature, current density, and voltage under various load conditions were analyzed. The presence of temperature gradients and nonuniform current distribution affecting battery efficiency, degradation, and lifetime was established. Critical operating conditions characterized by increased internal resistance, local overheating, and reduced system stability were identified.* ***Goal.*** *To develop a mathematical model of dynamic thermoelectric processes in a lithium-ion battery and to evaluate the effectiveness of using a system of nonlinear differential equations for analyzing electrical, thermal, and diffusion phenomena.* ***Methodology.*** *A generalized DFN model with an integrated thermal module (DFN + Thermal PDE) was employed. Numerical methods were applied to solve the system of equations.* ***Results.*** *The dependences of the output voltage on temperature and load current were obtained. The influence of local overheating and nonuniform current density distribution on battery efficiency and service life was determined.* ***Originality****. A multiphysics model integrating electrical, thermal, and mass-transfer processes into a unified system was proposed.* ***Practical value.*** *The proposed model can be used for predicting the performance of lithium-ion batteries, optimizing battery design, improving thermal management systems, and enhancing the energy efficiency, reliability, and safety of battery systems.*

Keywords: *lithium-ion battery, thermoelectrochemical modeling, temperature, current density, electrochemical processes, electric vehicle, battery pack.*

Introduction

In recent years, there has been a significant increase in the use of energy storage systems based on lithium-ion batteries, particularly in electric transportation, portable electronics, and autonomous power systems operating on renewable energy sources. The expansion of these applications necessitates the development of new types of battery systems that combine high specific energy capacity, enhanced thermal stability, and long service life [1, 2].

Due to the combination of these characteristics, lithium-ion batteries are considered among the most promising components of modern energy infrastructure, providing increased autonomy and environmental sustainability of technical solutions in the field of electric power engineering.

The efficiency, reliability, and safety of battery systems are significantly influenced by internal physicochemical processes caused by the interaction of electrical, thermal, and diffusion phenomena.

In this regard, an important scientific task is the development of adequate mathematical models capable of describing the dynamics of electric potential distribution, temperature fields, and spatiotemporal variations in ion concentration within different regions of the battery cell.

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In contrast to existing approaches, this study proposes a mathematical model of dynamic thermoelectric transfer processes in a lithium-ion battery. The model is based on a system of nonlinear differential equations describing the coupled processes of charge, heat, and mass transfer in the electrodes and electrolyte.

The developed model is intended to improve the accuracy of predicting the operational characteristics of energy storage systems, as well as to optimize thermal and electrochemical processes under various operating conditions.

Analysis of publications

An analysis of contemporary scientific research indicates that one of the key directions in the development of lithium-ion batteries is the creation of adequate multiphysics models that account for the interrelation between electrochemical, thermal, and diffusion processes [3]. Studies [4–6] consider modern approaches to battery mathematical modeling, including simplified and generalized electrochemical models that provide sufficient accuracy while reducing computational costs. At the same time, a number of recent studies emphasize the importance of using pseudo-two-dimensional (P2D) models and their modifications for a more detailed description of internal processes occurring in electrodes and electrolytes.

Studies [7,8] are devoted to the analysis of thermal processes in lithium-ion batteries. It has been established that temperature nonuniformities are among the main causes of active material degradation, reduced efficiency, and the occurrence of hazardous operating conditions, including thermal runaway. Furthermore, it has been determined that even minor local overheating can significantly affect the kinetics of electrochemical reactions and accelerate battery aging processes [9–11].

Study [12] investigates the influence of operating conditions on the characteristics of traction battery packs used in electric vehicles. It was found that high current loads lead to local overheating and an increase in internal resistance, whereas low temperatures reduce available capacity and limit charging rates. In addition, researchers note the significant impact of cyclic operating modes on degradation processes, particularly lithium loss and structural changes in electrode materials [13,14].

Publications [15,16] focus on thermal management methods for battery systems. Passive, air-cooled, and liquid-cooled systems, as well as their influence on temperature field uniformity and battery performance, are considered. Particular attention is paid to the optimization of cooling channel geometry and the application of intelligent temperature control systems [17,18].

Studies [19,20] investigate combined and innovative thermal regulation systems, including the use of phase-change materials and hybrid approaches, which improve heat dissipation efficiency. Additional studies confirm the перспективність of employing nanostructured materials and thermally conductive composites to enhance the performance of such systems.

Research presented in [21,22] demonstrates the development of integrated thermoelectrochemical models capable of predicting spatiotemporal distributions of temperature, current density, and voltage in battery cells under real operating conditions. Modern approaches also involve the application of machine learning methods and digital twins to improve prediction accuracy and enable real-time model adaptation.

Thus, contemporary scientific approaches are aimed at developing comprehensive models and efficient thermal management systems that ensure improved reliability, safety, and durability of lithium-ion batteries. At the same time, further research is directed toward integrating physical models with intelligent control and optimization algorithms, thereby opening new opportunities for the advancement of electric transportation systems.

Purpose and Tasks

The aim of this study is to develop a mathematical model describing dynamic thermoelectric processes in a lithium-ion battery as an innovative approach to the analysis of multiphysics processes in energy storage systems, as well as to evaluate the effectiveness of using a system of nonlinear differential equations for investigating coupled electrical, thermal, and diffusion phenomena.

The research is focused on improving the accuracy of electrochemical process modeling in lithium-ion batteries through the integration of electrical, thermal, and mass transfer effects into a unified mathematical model. This approach makes it possible to consider the battery as a dynamic multiphysics system and provides a more adequate representation of its real operating conditions.

To achieve this aim, the following objectives must be accomplished:

– to analyze modern scientific approaches to the mathematical modeling of lithium-ion batteries and the multiphysics processes occurring within them;

– to investigate the features of electrical, thermal, and diffusion processes within a battery cell;

– to develop a mathematical model based on a system of nonlinear partial differential equations;

– to integrate thermoelectric interactions within a unified modeling framework;

– to evaluate the influence of temperature nonuniformities on the efficiency of electrochemical processes;

– to analyze the distribution of thermal loads and identify local overheating zones;

– to investigate the stability of electrode materials under varying temperature conditions;

– to assess the accuracy of predicting battery operating parameters based on the proposed model.

The obtained research results may be used to improve the efficiency of energy storage systems, enhance diagnostic and state prediction methods for lithium-ion batteries, and further develop approaches to the multiphysics modeling of electrochemical systems.

Analysis of thermoelectrochemical processes in lithium-ion batteries

Lithium-ion batteries have become widely used in modern technology as efficient electrochemical systems for energy conversion and storage. They play an important role in supplying energy to portable electronics, electric transportation systems, and other high-tech devices [23,24].

The high demand for lithium-ion batteries is determined by the combination of their operational characteristics, including high specific energy capacity compared with other electrochemical power sources, the possibility of repeated recharging, long service life, sufficient thermal stability, as well as the combination of relatively low production cost and high specific power. An additional advantage is their ability to adapt to various operating conditions and operating modes.

In this regard, particular importance is attached to tasks related to the development and implementation of modern direct-current power sources characterized by enhanced environmental sustainability, durability, and operational safety. Among such solutions, lithium-ion batteries occupy a leading position due to their high technical and economic performance indicators [25].

During operation, especially in transportation systems, battery packs are subjected to variable, pulse, and cyclic loads. This leads to a nonuniform distribution of current density and the formation of local high-temperature zones inside the electrochemical cell. Such phenomena have a spatiotemporal nature and significantly affect the operating characteristics of the battery [26].

To describe electrochemical processes, it is advisable to use kinetic relationships that make it possible to determine local reaction rates and the corresponding current densities. At the same time, it is important to take into account temperature variations in space and time, since temperature significantly affects the electrical conductivity of materials, exchange current density, ion diffusion rate, and heat generation intensity in individual regions of the battery.

Consideration of these factors is a necessary condition for adequate modeling of lithium-ion battery operating modes and for improving their operational efficiency [27,28].

Thus, there is a need to apply thermoelectrochemical models that comprehensively account for the interaction of electrical, thermal, and diffusion processes. Such an approach makes it possible to predict the formation of local overheating zones, assess the level of thermomechanical stresses arising in materials, and identify degradation processes in battery cells under real operating conditions.

A complete thermoelectrochemical model of a lithium-ion battery:

$$\begin{gathered} i_n = i_a - i_c \\ i_a = i_0 \exp\left(\frac{\alpha_a F\eta}{RT}\right) \\ i_c = i_0 \exp\left(-\frac{\alpha_c F\eta}{RT}\right) \end{gathered} \tag{1}$$

where i_n – interfacial (reaction) current density, A/m²; i_a – anodic current component (oxidation process), A/m²; i_c – cathodic current component (reduction process), A/m²; i_0 – exchange current density, A/m²; α_a – charge transfer coefficient of the anodic process, dimensionless; α_c – charge transfer coefficient of the cathodic process, dimensionless; F – Faraday constant, C/mol; η – universal gas constant, J/(mol·K); T – temperature, K.

This relationship describes the interfacial current density as the difference between the anodic and cathodic components corresponding to the forward and reverse directions of the electrochemical reaction at the electrode–electrolyte interface. Such an approach makes it possible to separately account for the contributions of oxidation and reduction processes, thereby providing a clearer physical interpretation of electrode reaction kinetics in a lithium-ion cell.

The anodic component characterizes the intensity of the oxidation process (lithium extraction from the active electrode material), whereas the cathodic component corresponds to the reduction process (lithium intercalation). The difference between these components determines the resulting reaction current, the direction of which depends on the sign of the overpotential.

The rates of both processes depend exponentially on overpotential and temperature, reflecting the activation nature of electrochemical reactions. The temperature field affects the kinetics through thermodynamic parameters, particularly the universal gas constant and the ambient temperature, which is especially important when modeling the thermal operating conditions of the battery.

The obtained results confirm that representing the current as the difference between the anodic and cathodic components provides a clearer physical interpretation of the processes while simultaneously achieving an effective compromise between model accuracy and computational cost when using a coupled DFN model with a thermal module.

Modeling thermoelectrochemical processes in a lithium-ion battery

For the analysis of the electrochemical characteristics of the investigated lithium-ion battery, a system consisting of a graphite anode and a cathode based on nickel–manganese–cobalt oxides (NMC, $LiNi_xMn_{\gamma}Co\_zO_2$) was considered. Such a configuration is one of the most suitable for application in traction vehicles. It is characterized by high energy density and stable performance during repeated charge–discharge cycles, which is critically important for the operation of batteries used in electric tractors.

The electrochemical properties of the graphite anode, the NMC cathode, as well as the lithium-ion cell based on them of the $LiNi_xMn_{\gamma}Co\_zO_2$ type, are presented in Fig. 1.

The open-circuit voltage (OCV) analysis for the graphite anode in the range from 0.1 to 0.25 V, the NMC cathode in the range from 3.0 to 4.2 V, and the lithium-ion cell formed on their basis in the range from 3.3 to 3.8 V was carried out as a function of the state of charge (SOC). The lower-right graph presents the dU/dSOC derivative dependencies for the anode and cathode, making it possible to identify regions of increased voltage sensitivity to SOC variations.

In the low-SOC region, the graphite anode demonstrates a sharp increase in voltage, whereas in the medium and high SOC ranges, the cathode exhibits several negative extrema, indicating the complex nature of electrochemical processes within this interval.

Fig. 2 presents the results of numerical simulation of thermal processes in a lithium-ion cell obtained using the complete thermoelectrochemical model (DFN + Thermal PDE). The calculations were performed for a typical discharge mode of an electric tractor battery pack, in which thermal processes are determined by the combined effects of Joule heating, electrochemical reactions, and diffusion processes in the electrodes and electrolyte.

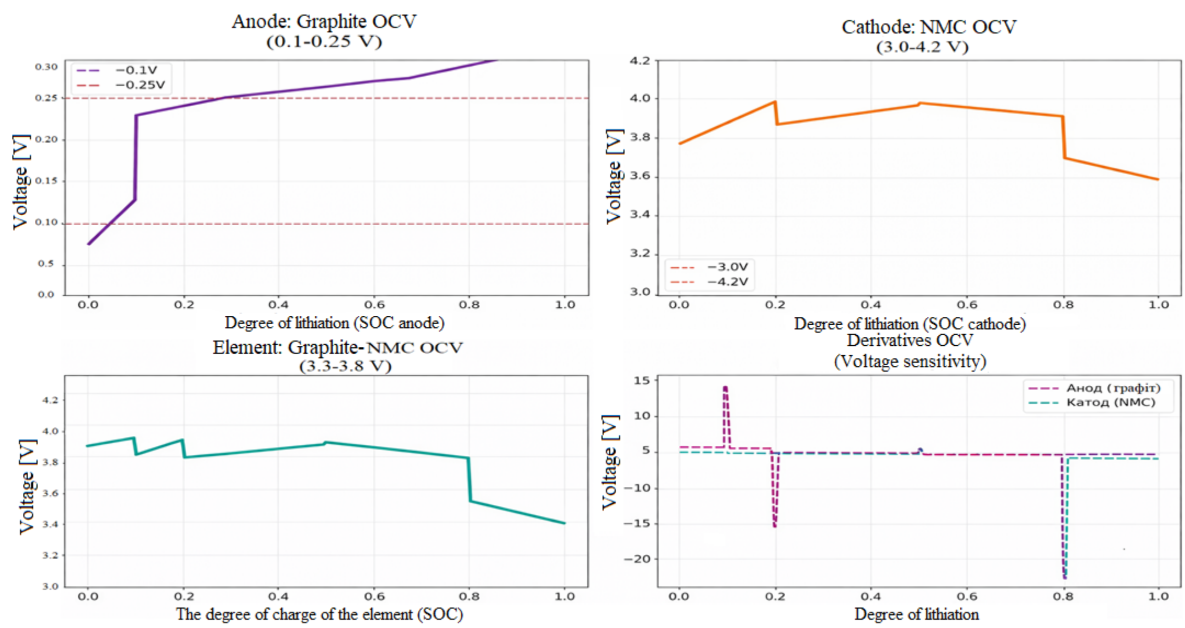

Fig.1. Electrochemical properties of a graphite anode, a cathode based on $LiNi_xMn_{\gamma}Co\_zO_2$ (NMC), and a corresponding lithium-ion battery cell

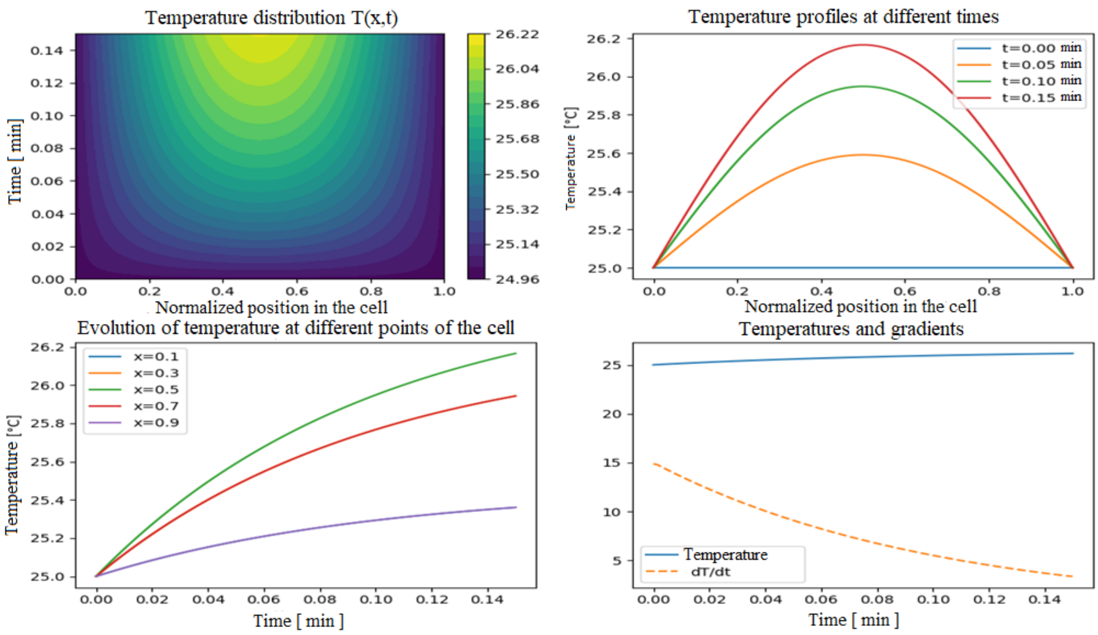

Fig.2. Spatiotemporal temperature distribution in the battery

The first graph illustrates the evolution of temperature distribution along the thickness of the electrochemical cell over time. At the initial stage of discharge, the temperature field remains nearly uniform, indicating that the system is close to a state of thermal equilibrium. As time progresses, a local temperature rise zone forms in the central part of the cell due to the increase in current density and enhanced electrochemical activity within the electrode interior. The appearance of a temperature maximum in the central region indicates limited heat dissipation efficiency at increased discharge rates, which is characteristic of high-power lithium-ion systems used in electric transportation.

The second graph demonstrates temperature profiles at different time instants along the normalized electrode coordinate. A temperature increase of approximately 1–2 °C relative to the initial conditions is observed. The symmetric profile shape with a maximum at the center confirms the dominance of heat transfer through thermal conduction in the solid phase. Small temperature gradients indicate efficient heat removal through current collectors and the cell casing.

The third graph shows the temporal variation of temperature at five characteristic points across the cell thickness. Maximum temperature values are observed in the central layers (x=0.3–0.5), whereas near the boundaries (x=0.1 and x=0.9) the temperature remains close to the initial level. Such a distribution confirms the presence of a moderate temperature gradient caused by the balance between internal heat generation and heat dissipation to the cell surface. This type of temperature field corresponds to thermally stable battery operating modes and indicates the absence of overheating during the initial discharge stages.

The final graph illustrates the coupled dynamics of temperature and its time derivative. At the initial stage of discharge, the temperature variation rate (dT/dt) is close to zero, indicating a quasi-steady-state thermal balance regime. As the discharge time increases, the temperature gradient rises due to the accumulation of Joule heat and the increase in the internal resistance of the cell. Under higher current loads or insufficient cooling system efficiency, a transition to unstable thermal regimes may occur, indicating the existence of a critical operating region.

Overall, the conducted analysis demonstrates that even under moderate current loading conditions, temperature nonuniformities are formed in lithium-ion battery cells of electric tractors, affecting local electrochemical activity and material degradation processes.

Taking these effects into account within the DFN + Thermal PDE model makes it possible to quantitatively evaluate thermal nonuniformity and optimize the cooling system of battery modules used in electric tractors. The spatiotemporal distribution of current density in the anode of the lithium-ion cell is presented in Fig. 3.

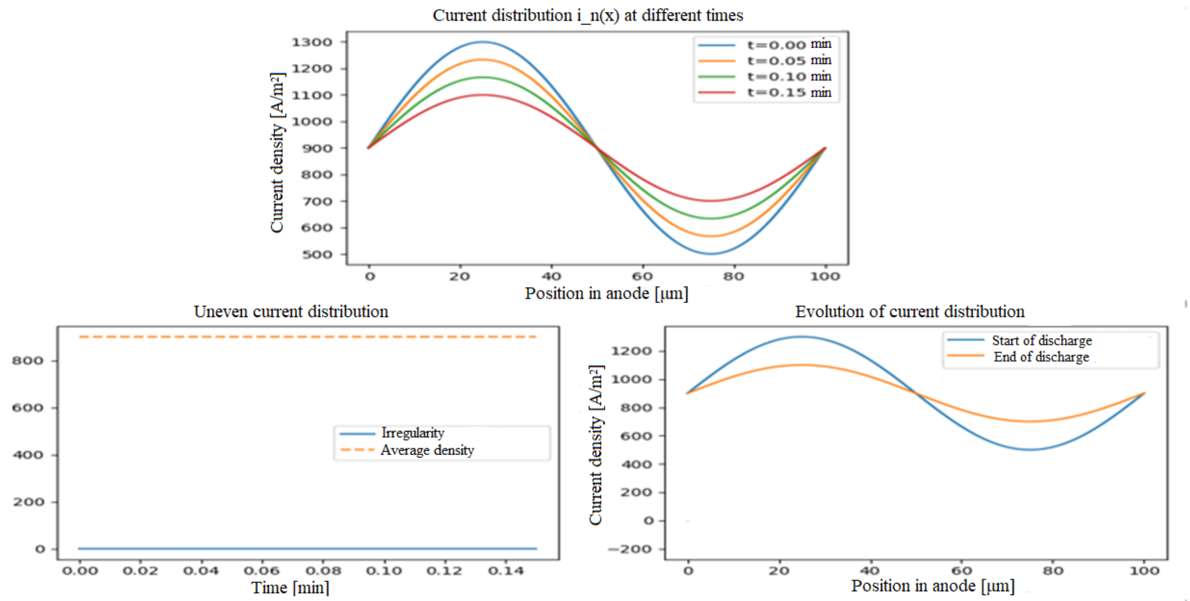

Fig.3. Spatiotemporal distribution of current density in the anode of a lithium-ion cell

As a result of the numerical solution of the thermoelectrochemical P2D model, which combines the DFN subsystem with heat transfer equations (DFN + Thermal PDE), spatiotemporal distributions of current density along the electrode were obtained. The generated plots illustrate the variation of current loading across the electrode thickness at different stages of the battery discharge process.

The analysis of the results shows that the current density distribution along the electrode thickness is moderately nonuniform and varies over time. At the initial stage of discharge ($t = 0$), a pronounced spatial profile is observed with a maximum in the internal regions of the electrode. As discharge proceeds, a gradual reduction in the amplitude of the distribution is observed, indicating a partial homogenization of current loading and a more uniform utilization of the active material.

Such dynamics reflect the characteristics of electrochemical processes in a porous electrode, where local overloading decreases over time and current density gradients become more uniform. This is consistent with the physical nature of charge and mass transport processes in the electrode under discharge conditions.

For a quantitative assessment of the degree of nonuniformity, the coefficient of variation (CV) was used, which characterizes the spatial nonuniformity of the current density distribution in(x)i_n(x)in(x). The obtained results indicate that the CV value changes only slightly over time and remains relatively stable, suggesting the absence of abrupt changes in the distribution pattern. At the same time, the average current density remains nearly constant, which corresponds to the modeling conditions and the selected discharge regime.

A comparative analysis of the initial and final current density distributions in(x) shows a reduction in the amplitude of the spatial profile. At the initial stage, the distribution exhibits more pronounced maxima and minima, whereas toward the end of discharge the profile becomes smoother, indicating a partial equalization of current loading along the electrode.

Thus, the presented results make it possible to evaluate the features of spatiotemporal current redistribution, identify zones of potential nonuniformity, and draw conclusions regarding their possible influence on thermal effects and degradation processes in electrode materials of lithium-ion battery systems.

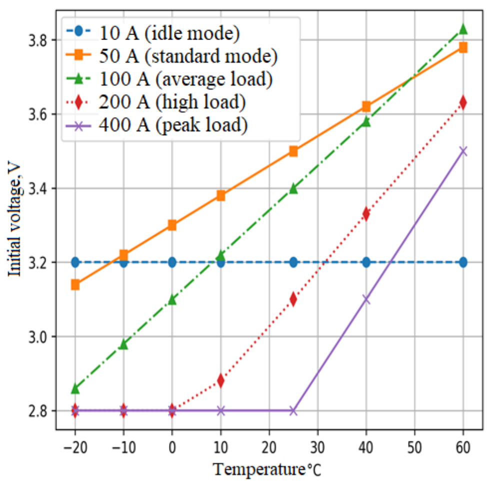

Fig.4. Dependence of the initial battery voltage on temperature

Thus, the spatiotemporal analysis of the current density distribution within the complete thermo-electrochemical DFN model, taking into account the thermal equation, makes it possible to identify the key features of the transient dynamics of the battery system under high-load operating conditions typical of electric tractors. The obtained results confirm the feasibility of considering the reverse thermodynamic coupling, as well as the spatial non-uniformity of reaction-kinetic activity, during the modeling of thermal stability and operational reliability of high-power battery systems.

Fig. 4 illustrates the dependence of the initial battery voltage on temperature under various load conditions. The graphical relationship demonstrates the influence of the temperature regime on the initial voltage of the lithium-ion cell, reflecting the degree of temperature sensitivity of electrochemical and ohmic processes.

Fig. 5 presents a heat map of the voltage drop distribution, illustrating the spatial characteristics of voltage losses within the battery cell.

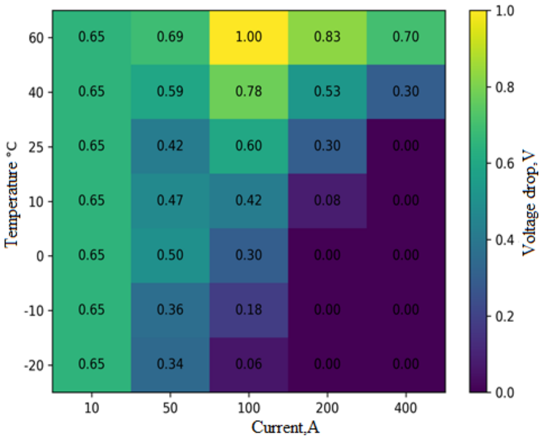

Fig. 5. Thermal map of voltage drop depending on temperature and load current

The color scale represents the level of voltage losses: regions with more intense coloration correspond to increased voltage drop caused by the combined influence of temperature factors and current magnitude.

Fig. 6 presents a three-dimensional surface characterizing the dependence of battery voltage on two independent variables, namely load current and temperature. This visualization demonstrates their mutual influence, forming a nonlinear structure of the operating range of the lithium-ion battery and determining its electrochemical properties under various operating conditions.

The analysis of the obtained surface indicates a pronounced thermally activated nature of voltage variation: at a constant load current, the voltage increases with increasing temperature. At the same time, a significant dependence on current is clearly observed, as an increase in load leads to a considerable voltage reduction, with this effect being most pronounced in the low-temperature region.

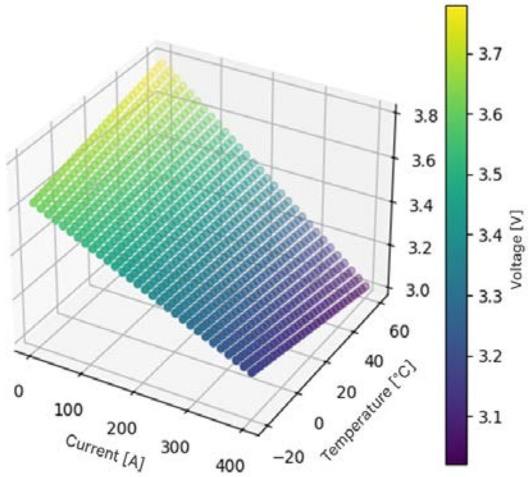

Fig. 6. Three-dimensional model of battery voltage dependence on load current and temperature

The steepest surface gradient is observed in the region combining high load current values and low temperatures, corresponding to an intensive battery voltage drop. Within this operating range, an increase in the internal resistance of the electrochemical system is observed, which may lead to reduced operational efficiency and potential instability of its characteristics. At the same time, in the region of moderate currents and elevated temperatures, a smoother voltage variation is noted, indicating more favorable conditions for electrochemical processes and lower energy losses.

Thus, the obtained three-dimensional dependence confirms the significant influence of temperature–current operating conditions on battery voltage and makes it possible to identify the regions of its most efficient operation.

The developed three-dimensional model provides a comprehensive representation of the battery operating characteristics and may be used to predict its behavior under real operating conditions, as well as for the development and optimization of battery management system algorithms.

Conclusions

The paper investigates thermoelectric processes occurring in a lithium-ion battery. The proposed model takes into account lithium-ion transport both in the electrolyte and in the solid phase of the electrodes, the distribution of electric potentials in the electrolyte and solid phases, as well as the kinetics of electrochemical reactions described by the Butler–Volmer equation. In addition, heat transfer processes accompanied by local heat generation caused by electrochemical and ohmic phenomena are considered.

The application of this model enables a detailed analysis of the spatiotemporal distributions of temperature, electric potentials, and current density within a lithium-ion cell under various operating modes and load levels. The results of numerical simulation indicate a significant influence of localized thermal loads on the efficiency and stability of electrochemical processes, lithium distribution in the electrodes and electrolyte, as well as on the dynamics of voltage and current variation within the cell.

The developed model provides the capability to predict the behavior of a lithium-ion battery under real operating conditions, taking into account transient operating modes and spatial temperature non-uniformity. The obtained results may be used for optimizing the design parameters of battery cells, developing battery management system algorithms, and improving the safety, reliability, and service life of batteries employed in electric vehicle and industrial energy applications.

Conflict of interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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Термоелектрохімічні процеси в літій-іонних акумуляторах: моделювання та аналіз

Анотація. Проблема*.* *Досліджено термоелектрохімічні процеси в літій-іонному акумуляторі з графітовим анодом і катодом типу NMC. Запропоновано математичну модель на основі системи нелінійних диференціальних рівнянь у частинних похідних, що враховує процеси перенесення заряду, тепла та маси в електродах і електроліті. У моделі реалізовано електрохімічну кінетику за рівнянням Батлера–Вольмера, дифузію іонів літію, розподіл потенціалів і теплові ефекти, спричинені джоулевим нагріванням та електрохімічними реакціями. На основі чисельного розв'язання DFN-моделі з тепловим модулем проведено аналіз просторово-часових розподілів температури, густини струму та напруги за різних режимів навантаження. Встановлено наявність температурних градієнтів і нерівномірного розподілу струму, що впливають на ефективність, деградацію та ресурс акумулятора. Визначено критичні режими роботи, які характеризуються підвищеним внутрішнім опором, локальним перегрівом і зниженням стабільності системи.* ***Мета****. Розроблення математичної моделі динамічних термоелектричних процесів у літій-іонному акумуляторі та оцінювання ефективності використання системи нелінійних диференціальних рівнянь для аналізу електричних, теплових і дифузійних явищ.* ***Методологія****.*

Використано узагальнену DFN-модель з інтегрованим тепловим модулем (DFN + Thermal PDE). Для розв'язання системи рівнянь застосовано чисельні методи. ***Результати.*** *Отримано залежності вихідної напруги від температури та струму навантаження. Виявлено вплив локального перегріву й нерівномірного розподілу густини струму на ефективність і термін служби акумулятора.* ***Оригінальність.*** *Запропоновано мультифізичну модель, що інтегрує електричні, теплові та масообмінні процеси в єдиній системі.* ***Практичне значення****. Модель може бути використана для прогнозування роботи літій-іонних акумуляторів, оптимізації конструкції, удосконалення систем терморегулювання та підвищення енергоефективності, надійності й безпеки батарейних систем.*

Ключові слова: *літій-іонний акумулятор, термоелектрохімічне моделювання, температура, густина струму, електрохімічні процеси, електромобіль, акумуляторна батарея.*

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