This study defines fire hazard parameters for the Panasonic NCR18650B ($LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$) lithium-ion battery (LIB). The task to obtain high-quality fire extinguishing substances and materials to prevent the spread of combustion implies determining the appropriate data experimentally. In particular, establishing the thermophysical characteristics and time dependence of the change in temperature indicators for the Panasonic NCR18650B ($LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$) LIB depending on different fire extinguishing substances is a relevant issue that is resolved in this work.

Based on the results of experimental studies, it was determined that the time of occurrence of the critical temperature inside LIB (~ 170°C) exposed to an external energy source (~ 300°C) is ~ 400 s. The effectiveness of the use of water and carbon dioxide (CO_2) on the effectiveness of reducing (cooling) the temperature of the internal filling was experimentally established. Accordingly, the time for reducing the battery temperature to 20°C with water is 400 s; when using CO_2, it is 280 s.

The mathematical model reasonably describes the cooling process of the LIB internal filling and accordingly verifies the experimental results of the study. The proposed mathematical model makes it possible to predict the complete cooling of the LIB depending on the type of extinguishing agent and the initial temperature of the substance. Additionally, the LIB heat transfer coefficients α ($W/m^2{\cdot}°C$) exposed to the action of water and CO_2 were established, which are 20 and 50, respectively.

The results make it possible to devise effective fire extinguishing agents and an algorithm for their application, in particular, to set the parameters of the extinguishing time and the required volume of the extinguishing agent in accordance with the power and type of battery. Additionally, the mathematical model built can be used for other types of LIBs with already known thermophysical characteristics

Keywords: lithium-ion cell, cooling efficiency, temperature reduction, extinguishing agents, water, carbon dioxide

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EXPERIMENTAL DETERMINATION OF THE EFFECT OF FIRE-EXTINGUISHING AGENTS ON A DECREASE IN THE TEMPERATURE INDICATORS OF CYLINDRICAL LITHIUM-ION BATTERIES

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1. Introduction

Alternative sources of electricity generation and storage play an important role at the present stage of human development. Among the significant number of technical means for storing electricity, it is necessary to single out lithium-ion batteries (LIBs). A distinctive characteristic of LIBs is their ability to accumulate a large amount of electrical energy, store it for a long time and, if necessary, power various electrical appliances. Thus, today, almost everything that surrounds us can operate from LIB-based batteries, from small household appliances to vehicles (cars, trucks, buses).

Fire statistics and frequent cases of burning of electric vehicles [1–3] indicate the existence of a problem related to fire hazard of LIB-based batteries. On the other hand, the burning of LIBs raises the issue of determining the immediate extinguishing tactics and installing fire extinguishing agents to effectively extinguish such fires, which in most cases require the involvement of significant material and human resources.

Given the complexity of extinguishing batteries with LIB, the basic tactical method of such extinguishing is direct immersion of the electric vehicle in a container with water. In some cases, it is even recommended to simply secure the scene and only localize the fire until its complete self-extinguishing.

A general analysis of LIB fire statistics reveals that annually in technologically developed countries such as China, the USA, the Netherlands, and others there is an increase in cases of LIB fires of various types by at least 20%. This is especially true for electric scooters and electric bicycles.

An additional problem in determining the fire hazard and further obtaining optimal fire extinguishing agents for LIBs is the constant improvement and modification of the chemical composition of such LIBs. LIB manufacturers are trying to constantly improve their products in order to obtain better electrical performance of the element, while deliberately not highlighting certain thermophysical or other indicators that may have an impact, in particular, on the issue of the fire hazard of the element. Perfect knowledge of the comprehensive characteristics of LIB makes it possible to correctly predict its behavior at elevated temperatures, its behavior under the influence of various fire extinguishing agents, etc.

Accordingly, defining a scientifically sound basis for determining the thermal characteristics of LIB and the features of their behavior on various fire extinguishing agents is a relevant issue and is the basis for further scientific research. In addition, establishing and revealing the dependence of the effect of fire extinguishing agents on the temperature reduction of LIB to prevent the occurrence of a thermochemical reaction in the cell and prevent further spread of combustion is fundamental for assessing their fire safety. Additionally, the results to be obtained could serve to further define the basic technical parameters for stationary fire extinguishing systems for electricity storage and accumulation systems.

2. Literature review and problem statement

In work [4], using computer simulation and a mathematical model, the optimal geometry and design of the "cooling jacket" for the cylindrical LIB assembly were determined. The study reported the temperature indicators of the cylindrical cell, taking into account the different geometry of the cell's cooling channels. The work took into account temperature indicators only in the range from 25°C to 40°C, which understandably does not reflect the critical temperatures preceding the occurrence of a thermochemical reaction. It is clear that the scientific interest of the study is precisely in determining the maximum possible decrease in the operating temperatures of the cell, and not in preventing the occurrence of LIB self-ignition due to an uncontrolled increase in temperature to critical indicators. At the same time, study [4] emphasizes the importance of highlighting and studying the issue of LIB thermoregulation.

The results from [5] show that depending on the type of chemical filling of the LIB cathode ($LiCoO_2$, $LiFePO_4$, $LiNi_2O_4$), the fire hazard of the cell is reduced or increased due to the different temperature of its destruction. Accordingly, the optimal composition of the LIB cathode should be resistant to decomposition under the influence of elevated temperatures. Additional parameters of LIB, such as the electrolyte, are also quite often not resistant to the effects of high temperatures. All these indicators ultimately lead to changes in the direct thermophysical parameters of the cell and are individual for each LIB. Accordingly, the research for each type and type of LIB should be individual in each case.

An option for overcoming the difficulties associated with determining the optimal chemical composition of LIB in a battery and reducing the operating temperature of cells is to analyze the results reported in [6, 7].

In [6], it was determined that passive battery temperature control systems are ineffective in cases where the LIB temperature exceeds the optimal operating temperature. On the other hand, active cooling methods do not take into account the temperature difference in the battery cells. Accordingly, a hybrid approach to controlling the temperature indicators of LIB is the most effective. It was shown that the overall optimal temperature for LIB is 55°C, which in turn depends on the correlation of the internal resistance of the cell with the state of charge and the thermal conductivity of the cell. However, questions regarding the determination of LIB indicators and the influence of various substances on the reduction of the temperature indicators of the cell under elevated temperatures remained unresolved.

In [7], a microchannel liquid cooling battery thermostat for an electric vehicle was investigated. A mathematical model was built to determine the heat transfer value taking into account the electrochemical composition of LIB. It was found that the presence of active cooling does not have full effect because there is uneven temperature indicators of the battery cells. This result indicated the need to improve the existing cooling system. Similarly to work [6], attention in [7] is only on the optimal parameters of LIB operation without taking into account the increase in temperature indicators above 55°C and above.

Work [8] reports initial studies on determining the effectiveness of using water with additives when extinguishing a car battery. The authors established improved fire-extinguishing efficiency of water with the addition of a foaming agent when extinguishing an electric vehicle battery. Thus, based on the results of experimental studies, it was established that a foaming agent solution (water with 5% foaming agent F-500) reduces the burning temperature of the battery to ±80°C 30 s faster than regular water. An indisputable limitation of the study is the determination of the overall effect of the fire-extinguishing agent on the burning temperature of the battery. Accordingly, the study did not take into account the issues of possible re-ignition of LIBs and the temperature of their direct heating and ignition from neighboring cells.

In study [9], additional results were obtained regarding the fire hazard of LIB. The authors solved the scientific problem of determining the parameters of cylindrical and package LIB after exposure to ultra-low temperatures, in particular immersion in liquid nitrogen (LN_2) at a temperature of –196°C for five minutes. During the experimental studies, both the general performance indicators of LIB and the possibility of LIB ignition after its puncture with a sharp object and mechanical destruction were determined. Accordingly, based on the results of experimental studies, it was established that in none of the LIBs considered, combustion occurred after their mechanical damage. The corresponding result of the study is relevant for LIBs that are not under direct threat of ignition and can serve to prevent the occurrence of LIB combustion. Accordingly, the issues of direct cooling and cessation of the thermochemical reaction were not considered in the cited work.

According to the results from [10], it was found that out of sixteen manufacturers, twelve primarily recommend using water as the main extinguishing agent; the situation is similar to the use of powder extinguishing agents. The application of foam and CO_2 for extinguishing LIB is recommended by nine and ten manufacturers, respectively. At the same time, sand, nitrogen, and even halon are recommended for use. Additionally, the authors analyzed and took into account the fact that the burning of LIB can be attributed simultaneously to three fire classes out of five, namely: A, C, and D. Accordingly, the use of most fire extinguishing agents is quite logical and justified. However, the work does not provide direct results of the effectiveness and comparison of the use of most fire extinguishing agents during the burning of LIB.

Works [11, 12] build on the studies reported in [9, 10]. The authors have somewhat expanded the range of possible fire extinguishing agents and analyzed their impact and fire extinguishing properties during the burning of LIB. According to the results of the studies, it is water (sprayed water), water with the addition of a foaming agent that have the best cooling and fire extinguishing properties. However, no comparisons with other types of fire extinguishing agents were made.

As regards further studies on determining the effectiveness of using various fire extinguishing agents, one should pay attention to the results from [13]. The study reports the results of experimental studies on the effectiveness of using water, powder fire extinguishing agents, and the innovative liquefied gas Novec 1230 ($C_6F_{12}O$) during the extinguishing of LIB. The study gives results of a decrease in the combustion temperature and the fire extinguishing effect of the specified substances. At the same time, the effectiveness of using the corresponding substances was determined by stopping the spread of combustion along the cylindrical cells of LIB due to a decrease in the surface temperature of adjacent cells. As indicated in the cited work, the direct effectiveness of fire extinguishing agents for cooling a burning LIB can be placed in the following sequence: ABC dry powder > BC dry powder > water spray > Novec 1230. However, water best cooled the adjacent LIB that was exposed to thermal effects from the burning cell. Accordingly, the difference in temperature reduction on the adjacent LIB cell was 40.68°C, as opposed to 4.4–19.7°C for powder agents, and 3.6°C for Novec 1230.

In most cases, the attention of scientists has been drawn to three main fire extinguishing agents that have shown their effectiveness. In particular, these are water (sprayed and finely sprayed), water with foam additives (F-500, compressed nitrogen foam), and a variety of gas compounds (CO_2, LN_2, $C_6F_{12}O$) with a cooling effect.

Thus, in work [14], the issue of extinguishing (cooling) of cylindrical LIB was studied and, in general, the tests showed practically identical results of fire extinguishing efficiency of sprayed water on LIB of different charge degree. In particular, the temperature of the cell fell within 200÷250 s by 100÷125°C. The authors reported a practical result on determining the time of temporary stop of the thermochemical reaction in LIB, which was about 25 s when reaching the critical temperature of LIB of 175°C. However, the issues of influence on the retarding effect of the thermochemical reaction of other fire extinguishing agents, in particular the metered supply of water and aqueous solutions, remained unexplained.

In [15], the results of fire extinguishing efficiency of non-typical fire extinguishing agents are described; their effect was studied. Thus, the authors established the fire extinguishing efficiency of a mixture of water and liquid nitrogen (H_2O + LN), as well as liquid nitrogen and "dry water" (LN + $C_6F_{12}O$). The studies were conducted on a prismatic LIB (NCM, 106 Am/h) which was heated by thermal plates. The results of experimental studies showed that the use of mixtures of substances (H_2O + LN та LN + $C_6F_{12}O$) for 10 s gives a short-term cooling effect and a decrease in the temperature of LIB within 99.92÷105.6°C. The use of pure liquid nitrogen gives an instant drop in the flame temperature to –163.93°C but, after that, there is a return of temperature indicators to 283.5°C, and a further decrease to 181°C.

In study [16], the extinguishing efficiency of a prismatic iron phosphate battery using compression foam at different supply pressures was considered. According to the results of research and data analysis, it was found that the surface temperature was only 11% of the initial temperature. Such results may be caused primarily by the failure to achieve a cooling effect directly from the internal filling of LIB. As a result, the use of foam only stops intense burning, but does not stop the thermochemical reaction itself.

However, the results from [17] show a change in the temperature of a batch LIB by almost 300°C within 25÷30 s. This difference in results is explained by the use of different types of LIBs and, of course, the intensity of the supply of the extinguishing agent. According to the results of the work, the issue of the effectiveness of using the appropriate fire extinguishing agent only for batch LIBs was resolved. At the same time, the thermophysical parameters and chemical composition of each LIB presented in the work were not taken into account. Additionally, the internal heating temperature of the batch LIB was not established, most likely due to the conceptual difference in the structure of batch and cylindrical LIBs.

Taking into account our review of the related literature, it can be stated that water is a universal and most effective fire extinguishing agent for extinguishing LIBs. On the other hand, cooling fire extinguishing agents such as LN, CO_2, LN_2 show the least efficiency while the use of powders is generally questionable since they have a different principle of action.

The common aspect of studies [8, 9, 14–17] is that the researchers determined the basic criterion for the effectiveness of using fire extinguishing agents to be the temperature of the surface of LIB. Studies [16, 17] focused only on the issue of suppression of flame combustion and did not take into account the probability of further heating of LIB due to the continuation of the thermochemical reaction.

Considering the fact that the spread of combustion in the battery occurs due to an increase in the internal temperature of the cell, determining the thermophysical indicators of the cell is one of the key tasks in this area. Taking into account that the studies that we reviewed do not report results of changes in the temperature of the internal filling of cylindrical LIB depending on the action of the fire extinguishing agent, there is a need to establish patterns and define the corresponding indicators.

3. The aim and objectives of the study

The purpose of our study is to determine the effectiveness of using fire extinguishing agents to reduce the internal temperature of a cylindrical LIB at the occurrence of an irreversible thermochemical reaction (combustion).

To achieve the goal, the following tasks were set:

– to experimentally evaluate temperature indicators of the internal filling with various substances of the cylindrical battery Panasonic NCR18650B ($LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$);

– to model mathematically the LIB cooling process with subsequent determination of the corresponding thermophysical parameters (heat transfer coefficient) under the condition of using various fire extinguishing agents and time indicators of cell cooling.

4. The study materials and methods

The object of our study is fire hazard parameters for the lithium-ion battery Panasonic NCR18650B ($LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$).

The principal hypothesis of the study assumes that a cylindrical LIB, in particular Panasonic NCR18650B ($LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$) made in Japan, is a source of significant fire hazard because of an increase in the internal temperature of the cell, so cooling fire extinguishing agents, accordingly, can slow down the increase in the internal temperature.

According to the hypothesis, the action of the basic fire extinguishing agents should have an inhibitory effect on the course of the chemical reaction inside the LIB through a decrease in the direct internal temperature of the cell.

Considering the fact that it is not possible to obtain an increase in the internal temperature inside the cell through the formation of a short circuit inside the cell and the beginning of a chemical reaction, under the conditions of this experimental study the increase in temperature inside the LIB was achieved using an external high-temperature source.

In order to perform the study on assessing the temperature indicators of the LIB internal filling, an experimental setup was used, described in detail in [18]. The use of an appropriate bench makes it possible to solve the practical part of the experiment, in particular establishing temperature indicators inside a full-fledged LIB with internal filling.

Thus, a sample of the cylindrical Panasonic NCR18650B battery discharged to 100% was prepared for research. Accordingly, a completely fire-safe cell with an internal filling corresponding to the working sample was obtained. After that, a through hole with a diameter of 2 mm was drilled in the experimentally prepared sample along its entire length for subsequent placement of thermocouples: Fig. 1.

The temperature change was recorded throughout the experiment using chromel-alumel thermocouples with the ability to record temperature values from –80 to 1200°C. The readings from the thermocouples and their further processing were provided by a secondary device, the PVI-111 regulator-measuring device. Two thermocouples were installed inside the lithium-ion battery, and two more were installed on its surface to control the temperature. The laboratory bench for heating the LIB from an external source of thermal radiation (flame) and further recording the temperature values of the LIB internal filling exposed to the action of fire extinguishing agents is shown in Fig. 2.

The source of open combustion (flame) was two gas burners powered by a gas cylinder. To ensure uniform heating of the entire area of the LIB, the cell itself was placed in an additional metal housing; respectively, the burners carried out primary heating of the metal housing, which created the effect of a "furnace". The flame temperature of the gas burner was 750–800°C. Thermocouples TC-1 and TC-2 were placed directly inside the LIB, and TC-3 and TC-4 on the surface of the cell. The LIB itself was fixed to a tripod using special fasteners. The cell was heated until the temperature inside the LIB reached a value of 170–250°C. Considering that the onset of a thermochemical reaction in the cell begins with the temperature reaching 170°C, there is no need to heat it up additionally. However, to verify the impossibility of ignition of the LIB at 100% discharge and reaching an internal temperature above 170°C, the heating of the cell was continued to a temperature of 250°C (±30°C). After heating, the LIB was cooled using a carbon dioxide fire extinguisher for 10 s, a water jet with a flow rate of 0.024 l/s (a volume of 2.67 l) and direct immersion of the LIB in a vessel with a volume of 0.89 l. Accordingly, during cooling, the temperature indicators of the LIB internal filling were recorded.

Fig. 1. Sample of the Panasonic NCR18650 lithium-ion battery with internal filling and holes for placing thermocouples

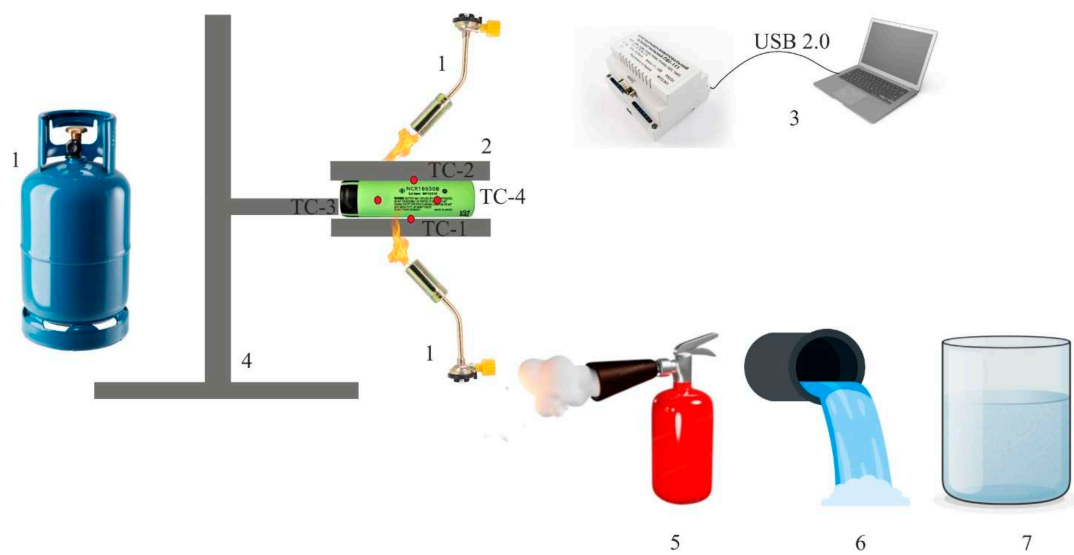

Fig. 2. Diagram of a laboratory bench for heating a cylindrical LIB from an external source (flame) and subsequent recording of temperature indicators: 1 — burners with a gas cylinder; 2 — additional metal housing for placing LIB; 3 — PVI—111 regulator-measuring device with the ability to connect to a personal computer with thermocouples (TS-1, TS-2, TS-3, TS-4); 4 — tripod for fixing the cell; 5 — carbon dioxide fire extinguisher; 6 — water flow (water supply) from the vessel; 7 — vessel with water

3 LIBs were prepared for experimental studies, which were directly used for each stage of our study.

5. Results of determining the effectiveness of using fire extinguishing agents to reduce the internal temperature of Panasonic NCR18650B

5. 1. Results of experiments to determine the effect of fire extinguishing agents on the temperature of the internal filling of Panasonic NCR18650B

The results from determining experimentally the effectiveness of cooling the internal filling of LIB using a carbon dioxide fire extinguisher (CO_2) are shown in Fig. 3.

It is worth noting that during the experiment, several attempts were made to carry it out. In particular, the LIB was heated to a temperature of 260°C (first attempt) and 165°C (second attempt), which was due to the technical features of the experiment. However, the repeated execution of the study made it possible to obtain reasonable indicators and data.

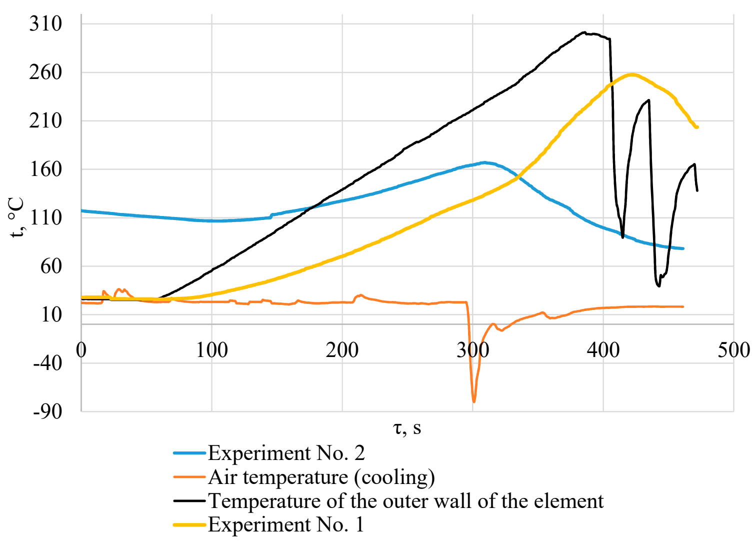

Fig. 3. Temperature indicators of the internal filling of Panasonic NCR18650 using CO_2

According to the results of our study (Fig. 3), in both cases, after reaching the appropriate temperature of the cell and supplying the extinguishing agent (CO_2), the drop in the internal temperature did not occur instantly. The delay in reducing the temperature indicators in both cases was ±5 s, after which the temperature in the middle of the cell dropped. The temperature of the extinguishing agent at the peak value was –80°C. First of all, the extinguishing agent was supplied in pulses for 10 s, as evidenced by the temperature fluctuations of the LIB surface, which practically dropped by 120°C, but after stopping the supply of the extinguishing agent, it rapidly returned to the previous value.

In the second stage of the study, the LIB was subjected to heating and cooling with water, in particular, in the first case, the LIB was poured with water, simulating cooling by a water jet at volumetric flow rate $Q = 0.024$ l/s. In the second case, the LIB was directly immersed in a container with water at room temperature. The results from determining the temperature indicators of the internal filling of the Panasonic NCR18650 when using water (immersion of the cell in water) are shown in Fig. 4.

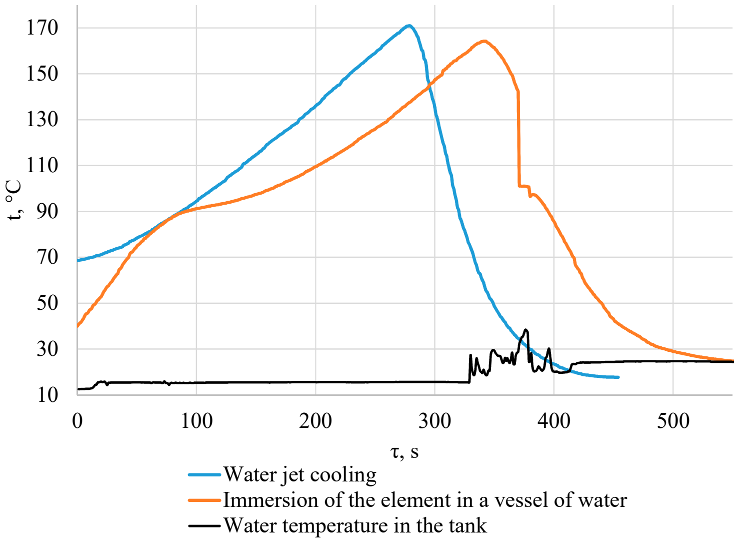

Fig. 4. Temperature indicators of the internal filling of Panasonic NCR18650 using water

From Fig. 4 it is obvious that the cooling rate of the LIB internal filling is different compared to CO_2 and occurs almost instantly. In the first variant of the experiment, water was supplied until the internal temperature of the cell reached 30°C. During the immersion of the LIB in a container with water, in the first seconds there was an instant decrease in temperature indicators by 54.9°C, and there was also a short-term increase in the water temperature to 37.1°C (in the immediate vicinity of the LIB). In the following seconds, the temperature of the LIB gradually decreased, while the temperature of the water increased, and the experiment was stopped after reaching the temperature equilibrium of water and the LIB at 24.4°C.

5. 2. Mathematical justification and determining the heat transfer coefficients of the inner shell of Panasonic NCR18650B

To obtain a complete mathematical description of the process of the influence of the fire extinguishing agent on the cooling of the LIB case after combustion, mathematical modeling of the process was carried out. For the mathematical description of the above process, the differential equation of heat conductivity was used. Since the lithium-ion battery has the shape of a cylinder, the equation of heat conductivity was considered for a cylindrical coordinate system

$$c\rho\frac{\partial t(r,\tau)}{\partial\tau}=\frac{1}{r}\frac{\partial}{\partial r}\left(r\lambda\frac{\partial t(r,\tau)}{\partial r}\right), r\in\left[0,r_n\right], \tau>0. \quad (1)$$

For a complete mathematical description of the process of preheating and subsequent cooling of the battery case, a 2-stage simulation was carried out. At the first stage, it is necessary to carry out the heating process under the influence of an external temperature of 300°C (according to the methodology and results of the experimental study). In this case, a boundary condition of the third kind must be added to equation (1), i.e., it is assumed that the heat exchange between the protective metal case and the surface of the LIB case occurs according to the Newton-Richmann heat exchange law

$$-\lambda\frac{\partial t}{\partial r}(r_n,\tau)=\alpha\left(t(r_n,\tau)-300\right). \qquad (2)$$

In this case, heat transfer coefficient α was taken to be 30 W/m°C.

The ambient temperature was taken as the initial condition, which is 20°C

$$t(r,0)=t_0=20°\text{C}, \qquad (3)$$

where $t(r, \tau)$ – temperature, °C; r – radius, m; τ – time, s; c – specific heat capacity of the material, J/(kg·°C); ρ – density of the material, kg/m^3; λ – thermal conductivity of the material, W/(m·°C); α – heat transfer coefficient, W/(m^2·°C).

The problem stated (1) to (3) fully describes the process of both heating and cooling in multilayer cylindrical structures. The only difference is the specification of the boundary condition (2) and the initial condition (3). The boundary condition describes the change in the ambient temperature in the surface layer of the structure, and the initial condition determines the temperature of the structure before the start of the heating (cooling) process.

The structure of the derived explicit exact formulas makes it possible to quickly and effectively calculate the temperature field distribution in a multilayer cylindrical structure, which includes the following stages:

1) input of initial data (coordinates of the layers, their coefficients of thermal conductivity, specific heat capacity, density, initial condition);

2) finding function $u_i(r, \tau)$ for each interval;

In work [19], it was established that for each layer of the structure the function $u(r,\tau)=300$;

3) calculation of the roots (ω_k) of the corresponding characteristic equation of the problem for eigenvalues [20]

$$\det\left[P+Q\cdot\widetilde{B}(r_2,r_0,\omega)\right]=0,$$

where the following is indicated:

$$P=\begin{pmatrix}0 & 1\\ 0 & 0\end{pmatrix},\quad Q=\begin{pmatrix}0 & 0\\ \alpha r_n & 1\end{pmatrix},$$

$$\widetilde{B}(r_2,r_0,\omega)=\prod_{i=1}^{2}\widetilde{B}_{i-1}(r_i,r_{i-1},\omega),$$

$$\widetilde{B}_{i-1}(r_3,r_{i-1},\omega)=\begin{pmatrix}b_{11}^i & b_{12}^i\\ b_{21}^i & b_{22}^i\end{pmatrix},$$

$$b_{11}^i=\frac{\pi\beta_i r_{i-1}\left(J_1(\beta_i,r_{i-1})Y_0(\beta_i,r_i)-J_0(\beta_i,r_i)Y_1(\beta_i,r_{i-1})\right)}{2},$$

$$b_{12}^i=\frac{\pi\left(J_0(\beta_i,r_{i-1})Y_0(\beta_i,r_i)-J_0(\beta_i,r_i)Y_0(\beta_i,r_{i-1})\right)}{2\lambda_i},$$

$$b_{21}^i=\frac{\pi\lambda_i\beta_i^2 r_i r_{i-1}\left(J_1(\beta_i,r_i)Y_1(\beta_i,r_{i-1})-J_1(\beta_i,r_{i-1})Y_1(\beta_i,r_i)\right)}{2},$$

$$b_{22}^i=\frac{\pi\beta_i r\left(J_1(\beta_i,r_i)Y_0(\beta_i,r_{i-1})-J_0(\beta_i,r_{i-1})Y_1(\beta_i,r_i)\right)}{2},$$

where J_0, J_1, N_0, N_1 are the Bessel and Neumann functions of zero and first order, respectively;

4) finding eigenfunctions $R_k(r, \omega_k)$ for each interval [19]:

$$\mathbf{R}_{k0}(r,\omega_k)=\tilde{B}_0(r,r_0,\omega_k)\cdot(1,\ \ 0)^T=(b_{11}^0,\ \ b_{21}^0)^T,$$

$$\mathbf{R}_{ki}(r,\omega_k)=\tilde{B}_1(r,r_1,\omega_k)\cdot\tilde{B}(r_1,r_0,\omega_k)\cdot(1,\ \ 0)^T=$$
$$=\tilde{B}_1(r,r_1,\omega_k)\cdot(b_{11}^0,\ \ b_{21}^0)^T;$$

5) finding the Fourier coefficients for the expansion of the initial condition [20]

$$f_k=\frac{1}{\|R_k\|^2}\sum_{i=0}^{n-1}c_i\rho_i\int_{r_i}^{r_{i+1}}\left(20-u(r,0)\right)R_{ki}(r,\omega_k)r\,\mathrm{d}r;$$

6) finding the Fourier coefficients for the expansion of function $\partial/\partial\tau\cdot u_i(r, \tau)$

$$\gamma_k=\frac{1}{\|R_k\|^2}\sum_{i=0}^{n-1}c_i\rho_i\int_{r_i}^{r_{i+1}}\left(\frac{\partial u(r,\tau)}{\partial\tau}\right)R_{ki}(r,\omega_k)r\,\mathrm{d}r;$$

7) finding function $v_i(r, \tau)$ [19]

$$v(r,\tau)=\sum_{k=1}^{\infty}\left[f_k\cdot e^{-\omega_k\tau}-\int_0^{\tau}e^{-\omega_k(\tau-s)}\gamma_k(s)\,\mathrm{d}s\right]\cdot R_k(r,\omega_k)=$$
$$=\sum_{i=0}^{n-1}v_i(r,\tau)\cdot\theta_i,$$

where f_k and γ_k are the Fourier coefficients of the initial condition expansion and function $\partial/\partial\tau\cdot u_i(r, \tau)$, respectively;

8) using the reduction method to find the temperature field distribution on each of the intervals $t_i(r,\tau) = u_i(r,\tau) + v_i(r,\tau)$ [19]

$$t(r,\tau)=\psi(\tau)+$$
$$+\sum_{i=0}^{n-1}\sum_{k=1}^{\infty}\left[f_{ki}\cdot e^{-\omega_k\tau}-\int_0^{\tau}e^{-\omega_k(\tau-s)}\gamma_{ki}(s)\,\mathrm{d}s\right]\cdot R_{ki}(r,\omega_k)\theta_i..$$

LIB was considered as a two-layer structure with the following thermophysical characteristics given in Table 1.

Table 1

Thermophysical characteristics of Panasonic NCR18650B for mathematical modeling to determine the heat transfer coefficients of the inner shell of the cell

Parameter	Dimensionality	Value
Internal filling of the cell (cathode and anode)		
λ_0	W/m· °C	324
c_0	J/kg· °C	677
ρ_0	kg/m^3	4743
Body (steel shell) of the cell		
λ_1	W/m· °C	56
c_1	J/kg· °C	470
ρ_1	kg/m^3	7800

Mathematical modeling of the heating process of LIB filling was carried out using the direct method of calculating the non-stationary temperature field. The direct method includes the use of the reduction method, the application of the concept of quasi-derivatives, the method of eigenfunctions, and the method of expansion into Fourier series. The implementation of the direct method of calculating the temperature field was carried out in the computer algebra system Maple 13. The simulation of the heating process was carried out for 400 s, the results of which are shown in Fig. 5.

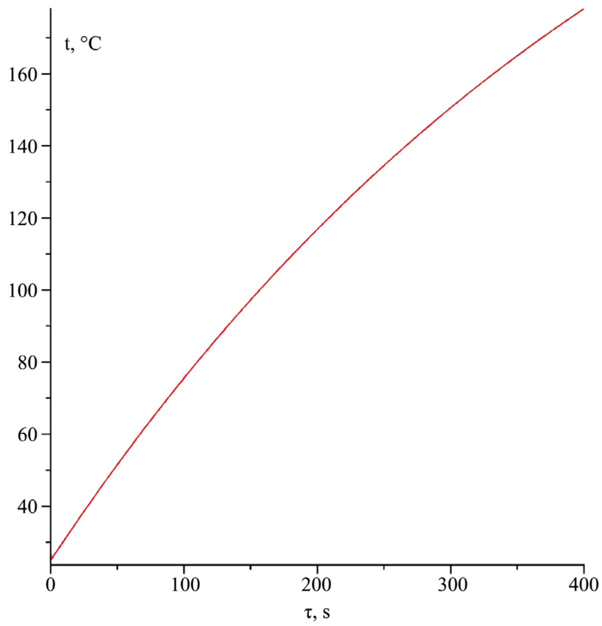

Fig. 5. Graphical representation of the heating process of Panasonic NCR18650 using mathematical apparatus

After that, the second stage of mathematical description of the process was prepared, during which the LIB was cooled with carbon dioxide and water. The initial temperature field distribution was taken as the temperature field distribution that was along the LIB thickness at the time of heating for 400 s.

After conducting the appropriate mathematical calculations, the LIB cooling charts were constructed using various fire extinguishing agents.

Carbon dioxide with a temperature of –80°C was used as the first fire extinguishing agent. During the simulation of the cooling process, it was established that the heat exchange coefficient between carbon dioxide and the LIB body is $\alpha = 50$ (W/m^2 °C).

Similarly, the LIB cooling process with water was carried out taking into account the water temperature of 20°C and the heat exchange coefficient was set to $\alpha = 200$ (W/m^2 °C).

The results of simulating the cooling inside the LIB under the influence of carbon dioxide and water are shown in Fig. 2.

In order to further determine the effectiveness of using fire extinguishing agents to reduce the temperature indicators of the LIB internal filling, the basic indicators of experimental research and mathematical modeling were analyzed; they are given in Table 2.

In accordance with the data in Table 2, it is possible to unequivocally state that the efficiency of using CO_2 for cooling LIB and preventing the spread of combustion in the battery is doubtful. The average change in the temperature of the internal filling was only 40°C, which, given the occurrence of a thermochemical reaction in the temperature range of 80 ÷ 160°C, is not very effective.

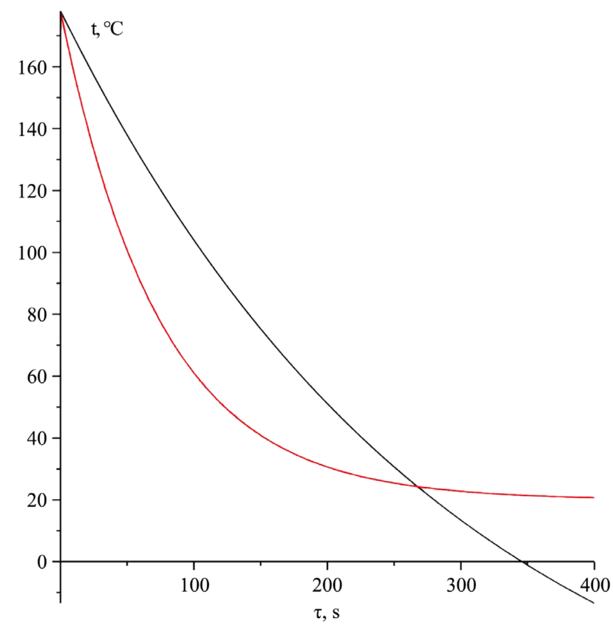

Fig. 6. Mathematical modeling of the cooling process of the internal filling of Panasonic NCR18650 using water (red curve) and a carbon dioxide fire extinguisher (black curve)

The mathematical apparatus used makes it possible to simulate the cooling of the internal filling to 0°C (we assume the cessation of the thermochemical reaction) and different initial temperatures of the extinguishing agent. For example, the simulation was carried out at an initial temperature of 190°C (Fig. 7).

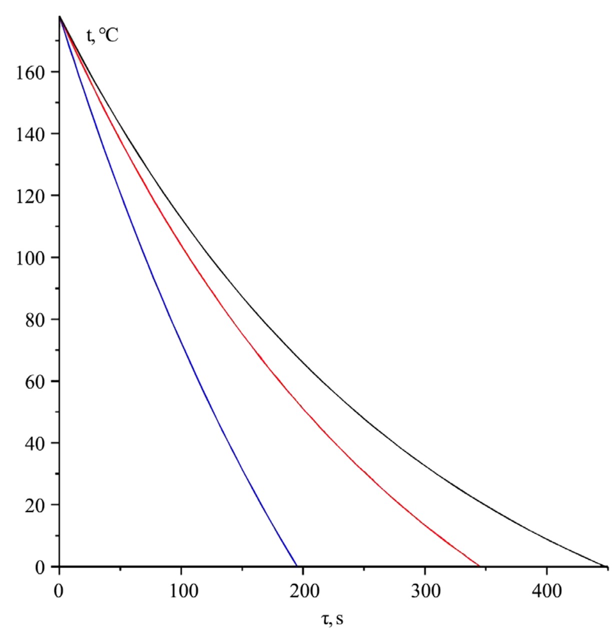

Fig. 7. Comparing the cooling rate of the internal filling of Panasonic NCR18650 using fire extinguishing agent at different initial temperatures: black – 50°C; red – 80°C; blue – 190°C

Table 2

Main indicators for the effectiveness of using fire extinguishing agents

No.	Experiment	Temperature change	Temperature drop (experiment), °C/s	Temperature drop (simulation), °C/s
1	CO_2	36.8 ÷ 43.50	0.68 ÷ 0.88	0.85 ÷ 0.32
2	Water	131.7	1.61	1.88 ÷ 0.14
3	Immersion	120.5*	1.18*	–

*Note: * – it is necessary to take into account that first of all the temperature of LIB dropped sharply by 54.9°C, amounting to 2.98°C/s over 19 s.*

Mathematical modeling with subsequent construction of a graphical dependence shows that under such conditions the cooling time of the internal filling is as follows: carbon dioxide – 50°C – 450 s to 0°C; carbon dioxide – 80°C – 345 s to 0°C; carbon dioxide – 190°C – 195 s to 0°C.

6. Discussion of results of determining the effectiveness of reducing the internal temperature of Panasonic NCR18650B

A special feature of our experimental studies is establishing the patterns of changes in the temperature of the LIB internal filling during its heating and cooling with fire extinguishing agents. This approach makes it possible to exclude from the experiment secondary temperature indicators caused directly by the temperature of the flame and combustion products. Additionally, the obtained temperature parameters of the cooling of the LIB internal filling will make it possible to determine the heat transfer coefficient characteristic of the use of the corresponding fire extinguishing agent.

The results of experimental studies and mathematical description of the process showed the advantage of water as a fire extinguishing agent that effectively reduces the internal temperature of LIB. However, further comparison of the cooling rate of the LIB internal filling shows that water reduces the temperature indicators to 20°C within 400 s, and carbon dioxide reaches the corresponding indicators in 280 s (Fig. 3). At the same time, it should be noted that the rate of decrease in temperature of the LIB internal filling when using water drops to 50°C during the first 120 s (Fig. 4) in contrast to carbon dioxide.

Additionally, it is necessary to take into account the fact that the decrease in temperature when using CO_2 did not occur instantly, and therefore the effectiveness of using CO_2 to prevent and limit the spread of combustion in the battery is questionable. This is confirmed by the amplitude decay of temperature indicators.

The corresponding desired effect can be achieved with a long-term supply of CO_2, which may be economically unprofitable and a technically complex process. The average temperature change in both cases was 120°C from the initial temperature of the LIB internal filling. However, it is worth noting that due to the different nature of the substances, water has a primarily better cooling effect. Carbon dioxide, on the contrary, is primarily unable to quickly cool LIB but, after some time, the temperature drop accelerates.

Quite unexpectedly, better cooling results were achieved when a normalized stream of water was supplied to the cell. The corresponding effect can be explained by the constant water temperature in contrast to the temperature of the water in the vessel, which nevertheless increased by taking heat from LIB. Presumably, the use of water with a lower initial temperature and a larger volume caused a better effect of lowering the temperature, but if a larger amount was cooled simultaneously, the result of the experiment may be repeated. At the same time, the use of CO_2 and similar compounds with a cooling effect has an advantage over water and aqueous solutions since it does not harm material values as water does.

The disadvantage of our experimental studies relates to the fact that the experiments were conducted using single samples of LIB, which does not fully reflect the nature of the combustion of the battery and its subsequent cooling.

Mathematical modeling of the process of reducing the temperature indicators of the internal filling of LIB made it possible to obtain accurate time indicators of reducing temperature indicators. Accordingly, the general nature of the cooling efficiency of LIB was confirmed by solving and applying equations (1) to (3).

The use of the Newton-Richmann heat transfer law (1) made it possible to define the uncertain thermophysical parameters of the Panasonic NCR18650B ($LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$) LIB given in Table 1, in particular the thermal conductivity of the material and the heat transfer coefficient.

Unlike the referenced studies [11–15], the numerical indicators of the heating and cooling time of the LIB internal filling were established based on the results of the experiments and mathematical modeling. Additionally, it was possible to predict the cooling time of the internal temperature of LIB depending on the action of different fire extinguishing agents with different initial temperatures.

Our mathematical model could be used for various types of cylindrical LIBs with already known corresponding thermophysical parameters. However, it will not be relevant for batch or prismatic LIBs; it can be considered a limitation of its use. Accordingly, further scientific research should be aimed at obtaining the possibility of conducting a mathematical calculation to determine the change in the internal temperature of prismatic and package LIBs.

In addition, the question remains unsolved whether it is sufficient to lower the cell temperature to 20°C, since repeated spontaneous ignition of the cell may occur [21], especially in the case of electric vehicles burning. The use of carbon dioxide or other substances with a temperature below zero, other than water, makes it possible to quickly lower the LIB temperature to a critical one (0°C and below) from the point of view of stopping the thermochemical reaction. Accordingly, such a statement automatically makes it possible not to exclude such fire extinguishing agents as CO_2, LN, LN_2 from the list of effective and possible to use.

The established time, temperature, and thermophysical parameters of the influence of fire extinguishing agents on change in the temperature indicators of the LIB internal filling are a reasonable addition to the existing knowledge base on the techniques and methods for extinguishing LIBs. In the future, the resulting values could serve as an addition during the design of mobile or stationary fire extinguishing systems in terms of determining and establishing consumption indicators and defining the type of fire extinguishing agents.

7. Conclusions

1. The use of CO_2 as a fire extinguishing agent for LIB is ineffective since it causes only a short-term cooling ef-

fect. The decrease in the internal temperature of the cell occurs slowly, by insignificant values of indicators, and with some time delay. Water, unlike CO_2, is the most effective fire extinguishing agent since its effect on reducing the temperature indicators of the internal filling of LIB is three times higher than when using a carbon dioxide fire extinguisher.

2. Based on the mathematical modeling, it was established that the heat transfer coefficient of LIB is $\alpha = 50$ (W/m°C) when using carbon dioxide and the total time for cooling the cell to 20°C is 280 s. The heat transfer coefficient when using water is $\alpha = 200$ (W/m°C), and the total time for lowering the temperature indicators is 400 s.

Conflicts of interest

The authors declare that they have no conflicts of interest in relation to the current study, including financial, personal, authorship, or any other, that could affect the study, as well as the results reported in this paper.

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Data availability

All data are available, either in numerical or graphical form, in the main text of the manuscript.

Use of artificial intelligence

The authors confirm that they did not use artificial intelligence technologies when creating the current work.

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