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New optical method to study oxygen activity in flowing liquid

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The necessity to study the oxygen activity of aqueous media in the flowing state to solve various problems has been substantiated. It has been found that emitted γ-quanta as a result of decay of ^{16}N nuclei with energy more than 9 MeV cause the formation of additional color centers and reversible optical defects in the fiber. Their appearance leads to increased radiation-induced losses in the optical fiber, which reduce the power of laser radiation transmitted through it. A new optical method has been developed that makes it possible to study the nature of oxygen activity change by the number of γ-quanta emitted by ^{16}N nuclei when the liquid moves along the pipeline. For the first time, the nature of the change in the spectral distribution of the number of ^{16}N nuclei emitted as a result of oxygen activity was investigated, and its features in the current flow were determined. In order to realize long-term studies of oxygen activity, a method was proposed to restore optical properties of fiber in the presence of background radioactive radiation.

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Introduction

The development of nuclear power engineering and the need for research in order to refine techniques for controlling chain reactions in new generation nuclear reactors has posed a number of complex problems [1–4] before scientists, one of which —is the study of the nature of changes in the oxygen activity of liquid media used as coolant in nuclear reactors as they flow through the pipeline.

At present, no control of the oxygen activity state in the coolant flow is implemented [1,4] at nuclear power plants. The content of ^{131}I is monitored, as it specifies reactor area poisoning with xenon. In some cases, the activity of corrosion products is monitored. In new reactor models, reference nuclides are monitored, primarily ^{60}Co, since it characterizes fuel tightness. In the recent reactor models at nuclear power plants, continuous monitoring of coolant leakage value from the primary circuit to the secondary circuit is implemented based on the registration of γ-radiation of isotope ^{16}N in the steam. This makes it possible to prevent a nuclear power plant accident in the steam generator area [5]. The operating experience of this system demonstrated that this method is not applicable to study oxygen activity of the coolant in the flow (especially for a long time).

Data on the nature of changes in oxygen activity is necessary to determine the state (or control the state) of the coolant, which is necessary to ensure safe operation of the reactor. In addition, nuclear power specialists suggest that data on the change in the oxygen activity of the coolant over time will provide new information on the nature of chain reactions [1,6]. This is necessary for the development of new reactor models.

Therefore, the purpose of this paper is to develop a new method to study oxygen activity in the coolant flow.

Method to study oxygen activity in coolant flow

Oxygen activity $^{16}O(n, p)^{16}N$ is associated with interaction of oxygen nuclei within the composition of one part of the coolant —water, with neutrons with energy greater than 9 MeV, which are present in the reactor area in large quantities due to a chain reaction. This interaction results in the formation of a ^{16}N nucleus, which is highly unstable. After nucleus formation, its decay starts. Its half-life is about 7.1 s. Decay of ^{16}N is accompanied by γ-radiation with energy of $E_\gamma \approx 6.2$ MeV.

Specially designed ionization chambers are used in scientific laboratories to study this phenomenon, but their dimensions and design features preclude their use in the reactor room. It is also extremely difficult to use some industrial devices to study the oxygen activity, for example „AZOT–16–PG“, in the reactor room, which are designed to determine the leakage of coolant between the primary and secondary circuits by recording γ-quanta in steam.

Therefore, we developed the following method. Previous studies have shown that under the influence of γ-radiation the optical fiber darkens (radiation-induced losses increase, α_s) [7–9]. To measure α_s, the classical formula is used

$$\alpha_s = -10 \lg(P_{\text{out}}/P_{\text{in}})/l, \quad (1)$$

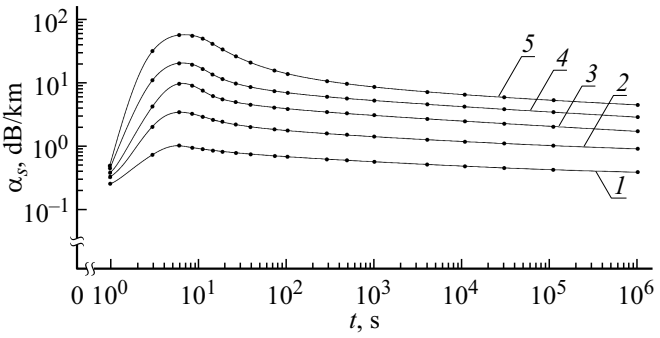

Figure 1. Variation of optical losses α_s from time t in single-mode fiber at $T = 294.2$ K. Laser radiation wavelength $\lambda = 1550$ nm. The graphs *1, 2, 3, 4* and *5* correspond to different concentrations of germanium oxide in the optical fiber core SiO_2-GeO_2 (in %): 0, 1.5, 4, 10, 20.

where P_{in} — power of laser radiation injected into the optical fiber, P_{out} — power at the output of the optical fiber, l — length of the optical fiber.

The effect of α_s increase is based on the formation of electron-hole pairs. By knocking out oxygen, the electron takes its place in a cyclic spatial structure [7,8,10]. Different color centers and „electron" compounds with different lifetimes [7–12] are formed.

The conducted studies [7–13] have shown that if exposure dose is low, the number of formed color centers and „electron" compounds is not large. There is no degradation in the glass mesh. Transparency of optical fibers is preserved long enough. The fiber darkens slightly. As the radiation exposure dose increases, the number of these formations increases, and the fiber darkening rate of the fiber increases. Losses α_s increase. Fig. 1 shows the results of the study of α_s variation from time t under short-term exposure to γ-radiation, duration of which is about 6–7 s (the exposure dose of the optical fiber is 100 Gy), for different composition of the optical fiber core. These studies were carried out on an experimental bench, design of which and the principle of measurements are similar to the experimental setup and measurements presented in [13].

The obtained results show that varied alloying of the optical fiber core (concentration of germanium oxide GeO_2 therein), makes it possible to change the sensitivity of the fiber to the influence of γ-radiation and set the operating point when recording γ-radiation at the dependence area $\alpha_s(t)$ with maximum slope steepness (fig. 1). This will allow in some cases to register bursts of oxygen activity from the decay of a small number of nuclei ^{16}N.

After γ-radiation exposure ceases, optical properties of the fiber are restored over time. The studies have shown that the processes of transparency restoration in the optical fiber after exposure to γ-radiation are extremely slow (the process may last longer than 10^7 s) [8–15]. In some cases (high exposure doses of glass [8]) the restoration of optical properties will not occur for 38 years. It means that the destruction (deformation) of the glass mesh also took place during the formation of a large number of color centers. Relaxation of color centers was slowed down most likely due to destruction (glass does not lighten). Radiation of high power (in some cases more than 2.5 W) at different wave lengths does not pass through the glass.

In contrast to numerous experimental studies related to the influence of γ-radiation on the optical fiber, in the special area of the reactor room, where the coils with the optical fiber are located, the dose rate is comparable to that affecting the fiber located on the satellite in orbit [8]. There will be no destruction of the glass mesh at such exposure doses in the fiber. Therefore, it is possible to use the developed method using additional laser radiation, which is also used by other scientists 13,16,17 in studies to clean the fiber from color centers and „electron" compounds.

Fig. 2 shows as an example the process of controlling relaxation processes of color centers and „electron" compounds in the optical fiber after cessation of pulse exposure to γ-radiation with exposure dose of 100 Gy with preservation of radioactive background.

The results obtained enabled us to establish that increased alloying of the optical fiber core with germanium oxide, the optical fiber cleaning rate under the influence of forced laser radiation rises.

When oxygen-activated coolant flows through the pipeline, additional γ-quanta are emitted to the „natural" background of radioactive radiation. Previous studies on the experimental model of Brest-300 reactor have shown that this addition to the exposure dose influencing the optical fiber has a pulsed nature and appears in a certain zone of the pipeline due to uneven distribution of neutrons with energies greater than 9 MeV in the coolant, which, when interacting with oxygen nuclei, included in the coolant, form ^{16}N nuclei as a result of reaction (n, p). Studies of oxygen activity in Brest-300 reactor have shown that the maximum time interval between the occurrence of these γ-radiation bursts in a fixed pipeline area is less than 250 s.

Therefore, the developed process of controlling the relaxation processes in the optical fiber will allow contin-

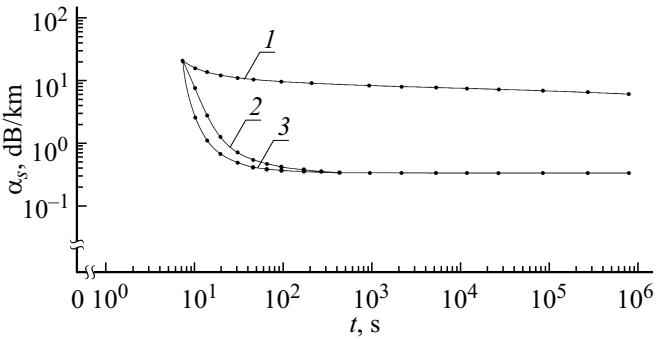

Figure 2. Variation of optical losses α_s from time t in single-mode fiber with core SiO_2-GeO_2 (alloying 10.0%) at $T = 294.3$ K. Laser radiation wavelength $\lambda = 1550$ nm. Graphs *1, 2* and *3* correspond to pulse laser radiation capacity at wave length $\lambda = 547$ nm (in mW): 0, 140, 380.

uous studies of the oxygen activity of the coolant in the pipeline. Without the use of fast „cleaning“ the optical fiber will darken. Sensitivity of the optical fiber to new exposures to γ-radiation will decrease. Using additional laser radiation makes it possible to return the operating point after registration of γ-radiation from oxygen activity to the dependence area $\alpha_s(t)$ with the maximum slope steepness (fig. 1). This ensures reproducibility of sensitivity in repeated measurements.

In addition, our previous studies of γ-radiation influence on the formation of radiation-induced losses in the optical fiber [15–17] allowed us to establish that at certain laser radiation capacity P_{in}, an equilibrium between the two processes (formation of new and relaxation of existing color centers and „electron“ compounds) can be achieved in the absence of other factors that significantly affect $\alpha_s(t)$ when exposed to γ-radiation. In this case, the detectable laser radiation capacity at the optical fiber output P_{out} remains unchanged. This means that the following relationship is satisfied:

$$P_{\text{in}} = P_{\text{out}} + P_{\text{rel}}, \tag{2}$$

where P_{rel} — the laser radiation capacity spent to relax a certain number (N) of color centers and „electron“ compounds that cause the optical signal in the fiber to attenuate.

Let us introduce the γ-quanta power P_γ, which was spent to form N new color centers and „electron“ compounds in the optical fiber. This capacity may be determined by the following formula:

$$P_\gamma = E/t_\gamma, \tag{3}$$

where E —energy to form N new color centers and „electron“ compounds, t_γ — time of optical fiber exposure to γ-quanta.

Let us assume that ^{16}N nucleus decay causes γ-quanta emission to all areas in the same manner. In this case, they interact evenly with the optical fiber, which is wound in a single layer on the insulation of the pipeline through which the coolant flows. According to the papers [7,10,12,13] and quantum field theory [18,19], in the first approximation, when one γ-quantum interacts with the optical fiber, one color center or one „electron“ compound is formed. In this case, the value E can be calculated using the following relationship:

$$E = SN\,h\nu/\sigma = SN\,hc/(\lambda\sigma), \tag{4}$$

where σ — interaction (scattering) cross section of γ-quantum with oxygen atoms O_2 in optical fiber, S — area of interaction of optical fiber with γ-quanta, N — number of emitted γ-quanta, λ — wave length of γ-quantum radiation.

The interaction area S may be determined by the following formula:

$$S = \pi\, D_p n_f d_f/2, \tag{5}$$

where D_p — diameter of the pipeline with thermal insulation, n_f — the number of optical fiber coils, d_f — diameter of the optical fiber.

When a ^{16}N nucleus decays, emission of γ-quanta corresponds to 7F-line of the spectrum ($\lambda = 0.254$ nm). Then the value of N may be determined using the following relationship:

$$N = \sigma\lambda t_\gamma (P_{\text{in}} - P_{\text{out}})/(Shc). \tag{6}$$

In relationship (6), values of the laser radiation capacities P_{in} and P_{out} are measured. The value σ is set experimentally for each fiber type, as it will depend on the alloying of the optical fiber core with GeO_2 germanium oxide, and the temperature of the optical fiber T_f. For example, for optical fiber alloyed with 1.5% GeO_2 germanium oxide, at $T = 295.3$ K $\sigma \approx 4.7 \cdot 10^{-15}$ cm^2. This value agrees well with the data obtained by other scientists [20,21]. This confirms the adequacy of the σ measurement technique we used to determine N using formula (6). Apart from monitoring of temperature values (of coolant, T_c, and T_f) when researching oxygen activity, it is necessary to monitor coolant flowrate q, since value t_γ is determined based on the following ratio: $t_\gamma = \pi L_{\text{ef}} d_p^2/4q$ (L_{ef} — effective length of pipeline, d_p — its diameter). Therefore, the measurement function q also must be implemented in the fiber optic system we are developing.

Fiber-optic system to study oxygen activity in coolant flow

We have developed a fiber-optic system to investigate the parameters of oxygen activity. It structural scheme is shown in fig. 3. The system design completely excludes the contact of the measuring elements with the coolant. The *2, 3* and *25* optical fiber coils are placed on top of the thermal insulation of the it1 pipe. This scheme of *2, 3* and *25* coil placement suggests two possible options for oxygen activity studies. These are fixed frames (distance L is fixed, see fig. 3) or movable frames — distance L varies. In this case, the distance between the L_1 coils may also change (fig. 3). The latter extends the functionality of the system we developed.

It should be noted that during NPP operation it is extremely difficult to change L distance, and this is not always expedient. The function of changing L and L_1 is necessary in the case of various studies on the experimental model of the reactor, since the value of coolant flow q in a number of experiments should be changed in a large range (at least by a factor of 100).

Let us note the main points of operation of the fiber-optic system developed by us. From the transmitting optical module *6* radiation with $\lambda = 1550$ nm (radiation capacity P_m is controlled from 0.1 to 20 mW) through an optical divider *9* ($N = 2$) arrives to inputs of multiplexers *12* and *13*. Wave length $\lambda = 1550$ nm for measuring losses $\alpha_s(t)$ of the laser radiation is chosen for the following reasons. With increasing λ the Rayleigh scattering coefficient (RSC) decreases. In addition, the „tails“ of phonon and electron absorption in the $1.0-1.6\,\mu$m wave length region do not contribute significantly to the optical losses whose

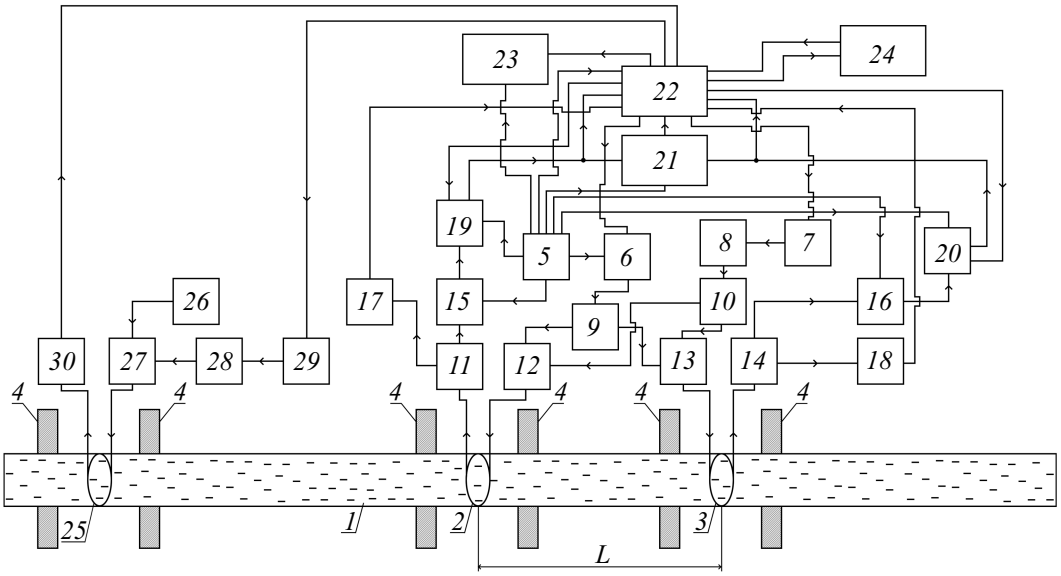

Figure 3. Structural circuit diagram of fiber optic system: *1* — pipeline with coolant; *2, 3* and *25* — coils with optic fiber (sensors); *4* — protective radiation shields; *5* — multi-functional power supply driver; *6* and *26* — transmitting optic modules; *7* and *29* — multifunctional power supply drivers; *8* and *28* — semiconductor laser; *9* and *10* — optical dividers (1/2); *11–14* and *27* — multiplexers; *15, 16* and *30* — photoelectric detectors; *17* and *18* — optic power meters; *19* and *20* — comparators; *21* — logic device; *22* — processing and control device; *23* — indication device; *24* — central computer.

data are used to study the changing nature of the coolant oxygen activity.

Other inputs of these multiplexers receive laser radiation with $\lambda = 457$ nm from a diode-pumped semiconductor laser *8* (SSP−ST−457−F). The radiation capacity P_L is controlled from 1 to 1000 mW. The multifunctional power supply driver *7* allows laser *8* to operate in both continuous and pulsed mode. Wave length from 440−485 nm laser emission range (blue part of the spectrum) to control the relaxation rate of the color centers and „electron" compounds is chosen for the following reasons. On the one hand, with long optical fiber lengths, it is necessary to provide the smallest RSC value so that the additional laser radiation will clean the glass to the maximum. On the other hand, the lower limit of laser radiation at $T = 307$ K for effective cleaning of E'centers is $\lambda = 217$ nm [11]. All other color centers, which change the optical properties of the laser radiation passing through the fiber, have loss regions in the longer wave length portion of the spectrum [7]. When cleaning the fiber, it is desirable to reduce losses of all kinds. Given that we have a much higher temperature than in [11], laser radiation with $\lambda = 457$ nm was chosen, since there is a possibility that it could trap the „tail" of the E'-center spectrum, since it shifts and expands at higher temperatures.

In the areas where the coils *2, 3* and *26* are located, the temperature for basic nuclear power plant operating modes varies from 343 to 593 K, depending on the reactor model. In the case of emergency operating modes, the temperature may increase to 1300 K or higher. This will destroy the developed system because it uses high-temperature-resistant (up to 980 K) single-mode fiber (Fiberguide, USA) with a copper-nickel coating.

It is also necessary to take into account that the optical fiber length l can be 1000 m and more. Therefore, it is desirable to shift towards increasing λ to reduce RSC. It should be noted that in manufactured industrial lasers at $\lambda = 457$ nm it is possible, using a multifunctional power supply driver, to provide laser power tuning from units of mW to 1 W in continuous mode, and in pulsed — to much higher than that.

The outputs of multiplexers *12* and *13* are connected to the optical fiber coils *2* and *3*. The laser radiation of two wave lengths from *2* and *3* optical fiber coils is delivered to the outputs of *11* and *14* multiplexers (inputs of these multiplexers are connected to the inputs of *17* and *18* power meters and *15* and *16* photoelectric detectors/). The power meters *17* and *18* are exposed to laser radiation with $\lambda = 457$ nm, the photoelectric detectors *15* and *16* — laser radiation with $\lambda = 1550$ nm. Power meters are required to monitor the optical fiber state (to determine the radiation induced losses α_s) using (1). In our developed fiber-optic system design l — this is the distance from the multiplexer *12* or *13* output to the multiplexer *11* or *14* output. In addition, this system implements the ability to measure the value of q. For this purpose, from 15 and *16* photoelectric detectors, voltage that contains laser capacity information with $\lambda = 1550$ nm is fed to the input of *19* and *20* comparators. The other input of the comparators is set to a level (voltage value U_c), and when it is reached, actuation takes place, and 1 is formed at the output of comparators (before that it was zero). Fig. 4 shows

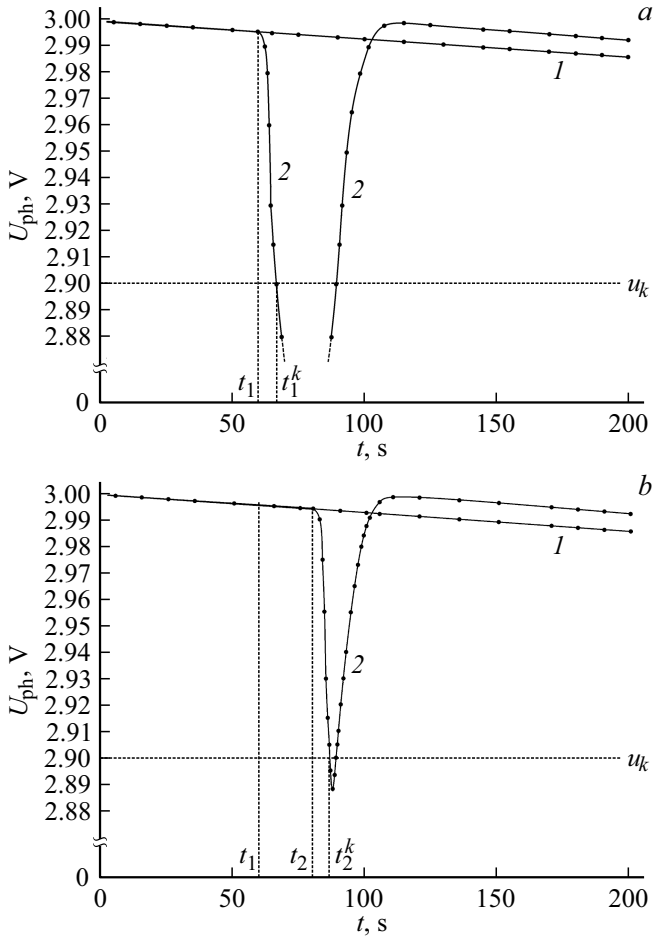

Figure 4. Variation of voltage U_{ph} from time t at photoelectric detector outputs: *a* — *15*, *b* — *16* (fig. 3). The graph *1* corresponds to the case of oxygen activity absence in the coolant in the area where the optical fiber coil is located. The graph *2* — when the coolant with oxygen activity (a burst of γ-quanta) passes through this zone.

dependence of voltage variations of U_{ph} at the outputs of the two photoelectric detectors *15* and *16* on time t.

Voltage value U_{ph}, which is determined by the laser radiation capacity detected at the photoelectric detectors *15* and *16* in case of fiber exposure only to background radiation and weak bursts of γ-quanta from the coolant flow decreases slowly and for $t = 250$ s does not reach the value U_c, which is set on the second inputs of the comparators *19* and *20* (fig. 4). Therefore, voltage value at the output of the comparators does not change and corresponds to zero. The inputs of the microchip 74135 (741S35), which excludes „OR“, which is part of the logic device *21*, receive zeros from the outputs of the comparators. The output *21* generates zero, which is fed to the *22* control and processing device. At the moment of time t_1 (appearance of large number of γ-quanta from decay of ^{16}N nuclei in coil area *2*, fig. 3) α_s value drastically rises, U_{ph} values on the *15* photoelectric detector decrease, and after some time reaches U_c at the second input of the comparator *19*. The comparator actuates, and high level corresponding to logical unit appears on its output, which is fed to one of the inputs of the microchip, which excludes „OR“. A signal corresponding to high level (unit) is generated on the output *21*. After some time Δt this process happens in the second arm of the circuit (coil *3*). After the second input of the microchip that excludes „OR“receives a signal from the comparator output *20* corresponding to logical unit, the output of the microchip, which excludes „OR“, will generate a signal corresponding to logical zero. As a result, a rectangular pulse with duration $\Delta t = t_2^k - t_1^k$ is generated at the output *21*, which corresponds to the coolant segment flow time with length L (fig. 3). This is the marking principle of measuring fluid flow rate or flow velocity in pipelines, which is the most accurate compared to others [23–26].

Then the value of the coolant flow rate q is determined using the following relationship:

$$q = \frac{\pi d_p^2}{4} \frac{L}{\Delta t}, \tag{7}$$

where d_p^2 — the inner diameter of the pipeline.

When a rectangular pulse arrives in *22*, a command is formed therein, which triggers the power supply driver *7*, and radiation from the semiconductor laser *8* with $\lambda = 457$ nm is delivered through the divider *10* to the multiplexer *12* and *13* inputs, and then to the optical fiber. This radiation destroys the color centers and „electron“ compounds produced under effect of γ-radiation. The relaxation process becomes much faster than the formation of new color centers and „electron“ compounds even in the presence of new oxygen activity in the optical fiber coil area. The laser radiation capacity $\lambda = 457$ nm that passed through the optical fiber of *2* and *3* coils is measured using *17* and *18*. Then information from *17* and *18* is fed to *22*, where α_s value is determined using (1). When reaching a certain value α_s, which is set beforehand (corresponding to the state of the optical fiber before the appearance of powerful bursts of γ-quanta in the zone of its placement), the power supply driver *7* is switched off.

During the restoration of properties (transparency of the optical fiber) the semiconductor laser with $\lambda = 1550$ nm operates in continuous mode. At a certain point in time the voltage from the photoelectric detectors will be greater than the reference voltage at the other two inputs of the comparators. The comparators will return to their original state (the output of the comparator will have a voltage level of zero). This change at the comparators will happen simultaneously, because the process of optical fiber properties restoration in two coils is identical. After that the system we developed is ready for a new flow q measurement.

To investigate the oxygen activity of the coolant, a coil *25* was installed. There are two variants of its arrangement. First, the *25* coil is placed in front of the *4* (upstream coil *2*) and between the two *27* and *28* screens. In the second case,

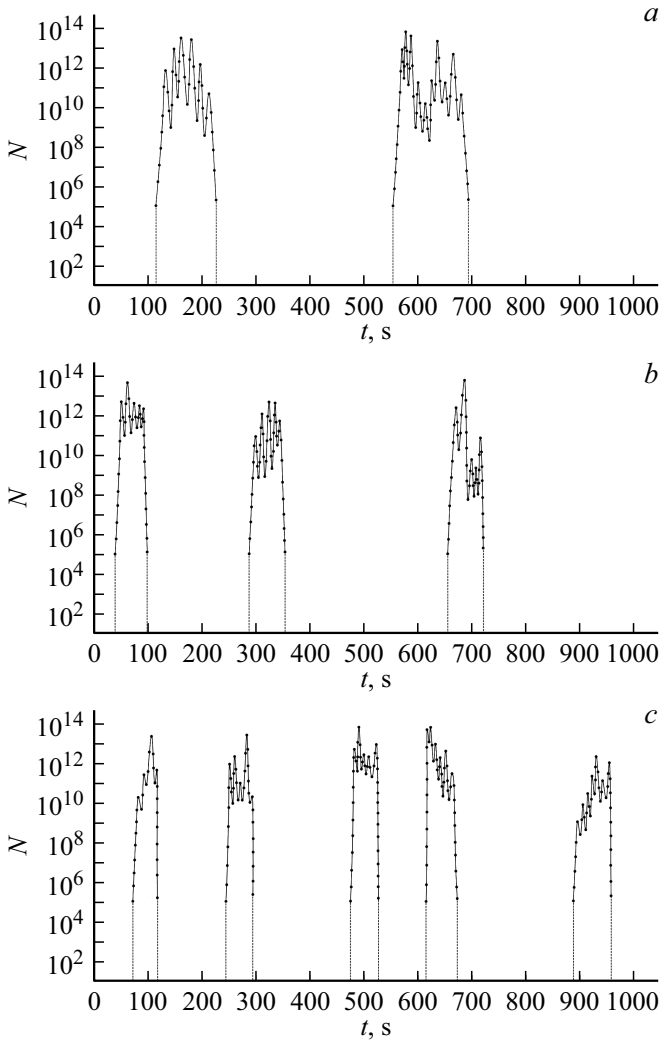

Figure 5. Variation of the number N of detected γ-quanta of oxygen activity from time t. The graphs in fig. *a, b* and *c* correspond to the coolant flow q (in m^3/s): 0.169, 0.339, 0.678.

the *25* coil can be placed downstream the screen, which is placed behind the *3* coil. The first case is preferable because it gives additional information about the laser radiation capacity, which can be used later to measure the flow rate q. The formula (6) is used to determine the number of γ-quanta N.

Fig. 5 shows as an example the dependence of N change in the coil *25* placement area on time t for different values of q. A plutonium nitride-filled solution $(H_20 + H_3BO_3)$ was used as the coolant at temperature of $T_c = 960$ K.

Analysis of the results obtained in fig. 5 shows that the oxygen activity in the coolant is randomly distributed over time. The maximum interval between its bursts in the coolant flow obtained earlier by other scientists is confirmed. It was found that by adjusting q, it is possible to partially control the distribution function N in the γ-quanta detection area of the optical fiber coils.

It should be noted that the fiber-optic system developed by us allows us to determine the number of emitted quanta in the range from 10^5 to 10^{14} in contrast to the previously developed devices, which measure the change of N in the range of not more than a factor of 1000. This fact significantly expands the possibilities of studying the oxygen activity in the coolant flow.

Conclusion

The results obtained have shown that our proposed new method and the fiber-optic system developed for its implementation make it possible to conduct studies of oxygen activity in the coolant flow under various operating modes of the reactor. The data on the nature of changes in oxygen activity allows us to monitor the condition of the coolant. This is especially important for various experimental studies, as well as for the development of recommendations to improve the design of operating reactors.

Comparison of the results of q coolant measurements with the help of the fiber-optic system developed by us with the results of its flow rate measurements by electromagnetic and Coriolis flowmeters demonstrated high efficiency of our proposed method of measuring q (the measurement error is not more than 1.5%). In the currently used industrial flowmeters for measuring q the measurement error is more than 5%. This result allows us and other scientists to clarify a number of factors in our research which are related to the effect of changes in the coolant flow rate q on the number of γ-quanta N emitted in a given time interval, especially at different values of T_c.

The question of optical fiber degradation, which in case of long-term studies of oxygen activity will necessarily occur and lead to inability of using this fiber for measurements (therefore, its replacement will be required, presumably it can be done the necessary number of times during the reactor operation), is not considered in the present paper in detail. The long-term studies that are necessary for this purpose have not yet been carried out. This will be the subject of our further research.

Conflict of interest

The authors declare that they have no conflict of interest.

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