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Науково-виробничий журнал "Ядерна енергетика та довкілля" публікує науково-дослідні, інженерно-технічні та експертно-аналітичні розробки у сфері радіаційної фізики, ядерної, радіаційної та екологічної безпеки, радіаційного матеріалознавства, безпеки та надійності ядерно-енергетичних установок, аналітичної та радіаційної хімії, використання нанотехнологій, ІТ-технологій і промислових технологій та з інших напрямків досліджень, які можуть бути використані для розвитку ядерно-енергетичної галузі, а також інформує населення щодо результатів контролю та моніторингу стану довкілля.

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CURRENT STATUS AND ACTIVITIES TO IMPROVE ENVIRONMENTAL SAFETY OF NUCLEAR POWER

Based on the analysis of environmental problems of nuclear power industry, related to various factors of its impact on the man and the environment, three groups of tasks were identified, which will help to improve the environmental safety of nuclear energy: increase the level and degree of informativeness about the environmental conditions in the vicinity of the NPP location, organization and work on preventive preparedness for emergency response, development and implementation of technologies for reliable burial of long-lived high-level radioactive waste. The modern approaches and trends were considered that have already been implemented or scheduled for implementation to solve the problems listed above.

Key words: nuclear energy, environmental issues, environmental safety, radio-ecological monitoring, emergency response, preventive preparedness, long-lived high-level radioactive waste, waste treatment technologies

In practice, it was proved that the impact of NPP on the environment, humans and biota, in compliance with the regular operation rules is negligible. It is detected mainly at the nuclear fuel cycle (NFC) facilities associated with uranium mining, processing and production of nuclear fuel for NPP, and regeneration of uranium and plutonium from spent nuclear fuel.

However, NPP danger to humans and the environment increases dramatically in emergencies. This requires awareness of the need for strict compliance with the provisions of nuclear, radiation and environmental safety and safety culture principles.

For a long time the environmental problems of nuclear energy was pushed to the sidelines compared to its capabilities. For just a couple of decades, the nuclear power share has reached unprecedented figures in the global energy sector, which makes it very attractive for investments and planned development. In the energy balance of some countries the share of energy derived from nuclear fuel has become very significant, for example, in France, it achieved more than 75% in the total balance of electricity production.

Even the growing volume of information on environmental issues of nuclear power did not affect the pace of its development. All the arguments of the nuclear power opponents were counter-argumented by data of "nuclear experts", which showed that operating NPP releases into the environment only very small amount of radionuclides and other non-radioactive contaminants, and this amount on the level of danger (risk) has an impact on the environment several times smaller than emissions of a thermal power station of similar capacity.

All this was (and still is) consistent with the reality. Therefore, before the Chernobyl tragedy the environmental issues were rarely discussed. Over the years of operation of numerous NPP all over the world, the industry has earned a reputation as the safest one: increased safety measures ensured trouble-free and efficient operation of NPP.

However, after the accident at the Chernobyl NPP, and especially at the NPP "Fukushima-1", the environmental concerns came to the fore in the discussion of ways of the world nuclear power development. Nevertheless, even now in ensuring ecological safety of NPP declarative statements are often dominated, being far from implementation.

Until recently, all environmental problems of nuclear power were reduced mainly to difficulties in the disposal of radioactive waste (RW), including spent nuclear fuel (SNF). In fact, there are much more problems there.

Indeed, by now the impact of long-lived high-level waste LL HLW on the environment is proved in a variety of scientific papers and special measurements in the already existing spent fuel disposal facilities [1]. A problem associated with the disposal of high level radioactive waste and spent nuclear fuel has not been finally resolved yet, and still remains one of the principal ones, that determine the development of nuclear power in the world.

The inevitable problem of NPP should also be considered their impact on water bodies, which water is used for the technological needs of the plant. As an example, heated wastewater entering the hydrographic system, located in the area of NPP location, causes an increase in water temperature of water bodies, so that there is a decrease of oxygen in them, which increases the probability of blooming, and the phenomenon of thermal stress in aquatic organisms. In addition, in the normal course of NPP operation it consumes huge amounts of water, going to the cooling of the units that can upset the balance in the water system in the area of NPP location, such

as in the area of the South-Ukrainian energy complex [2.3] or the Rivne NPP [4]. Cooling towers for water cooling may even change the microclimate in the vicinity of the plant. [5]

In addition, the list of environmental consequences from the NPP impact on the environment can include:

- Changes in topography and landscape as a result of local mechanical impacts during construction;
- Multiplication of effect of ionizing radiation on the biota in the NPP location area as a result of the combined action of radiation and non-radiation factors (heat discharge, chemical pollution);
- The risk of receiving large doses of radiation by personnel, population and biota as a result of nuclear accidents and radioactive contamination of agricultural products - the main supplier of dose-forming radionuclides (including off-site contamination) in human food.

There is another problem that should be listed in addition to the above ones. It is the problem of disposal of significant quantities of radioactive waste at NPP decommissioning NPP. To solve this problem is necessary only with taking into consideration the environmental risks that accompany each step of the process.

Naturally, the degree of how the listed factors can impact on the environment is different. Nevertheless, they should all be taken into account when determining the environmental risk from the NPP at planning the ways of nuclear power development.

Summarizing the existing environmental problems of nuclear power, associated with different factors of its impact on humans and the environment, three groups of tasks can be identified which will enhance the environmental safety of nuclear power:

1. Obtaining representative information on the state of the environment in the vicinity of the location of NPP for the purpose of environmental control and minimization of the potential environmental impacts during normal and emergency modes of NPP.
2. Organization and carrying out of work on preventive preparedness for emergency response in order to reduce the environmental and radiological risks involved in accidents at a nuclear power plant.
3. Establishment and implementation of technologies of highly reliable disposal of long-lived radioactive waste.

Consider the modern approaches and trends that received approval from the world leading experts and scientists and have begun to be realized or, at any rate, is currently being planned to meet the challenges listed above. First, let's talk about how the first of the tasks mentioned above was resolved and is still ongoing.

Obtaining representative information on the environment conditions in the NPP location area. Traditionally, while ensuring the environmental safety on nuclear power plants, the focus has been paid so far to technical and technological aspects. However, in practice, to increase the level of the environmental safety of any industrial plant engineering and technological solutions is not enough. More attention should be paid to the management issues of the environmental safety that are logically related to the protection of the environment, the environment monitoring systems, as well as those that follow from the requirements of ISO 9000 and ISO 14000.

The need for the organization and functioning of the system of regulation and control of the environmental safety at nuclear power plants at all stages of their life cycle, allowing to predict, prevent, and in case of emergency to show the way of emergency response, follows from the very definition of the environmental safety. Meaning of the definition of the environmental safety is that there is no danger to the environment, and that the biosphere, human society and the state are protected from internal and external impacts, and from negative processes endangering human health, the existence of biological diversity, the sustainable functioning of ecological systems and the survival of the mankind.

The Radiation Safety Systems currently used at the NPP are not quite reasonably identified with the environmental safety systems. They are built on anthropocentric principles of radiation protection: “the environment is protected when the humans are protected” (ICRP No. 26, 1977). It is used as a guide by the organization and management of radiation monitoring at NPP. However, human safety not in all situations protects the biota. There are living organisms that are more radiosensitive than humans, or at least the same, for example, a sheep, a donkey, a dog [6]. Some terrestrial plants such as conifers have a high degree of radio-sensitivity [7]. A striking example is the “Red Forest” - a zone of complete death of conifers with partial damage to deciduous forest as a result of powerful radioactive contamination caused by the accident at the Chernobyl NPP [8].

In addition, in the existing NPP radiation safety systems the environment does not act as a system, in which the radiation dose is formed, causing the risk to life, but as some kind of indicator, which characterizes the technological parameters of the NPP emissions and discharges, and compares them with the existing levels of radiation hygienic standards. The properties of the environment and migration processes in it are not taken into account when carrying out radiation measurements in the environment and performing predictive assessments of

NPP impact on the population and the environment. As a result, it becomes problematic to solve the following important groups of radiation safety problems: getting a reliable assessment of the radiation condition in the NPP location areas for different modes of operation, the development of effective measures to protect the environment and the population in case of accidents, ecological regulation of NPP impact on human and biota and management of the environmental safety.

These problems can be solved, if in the process of establishing a radiation safety system at a particular NPP we would take into consideration, as recommended in the new publications of the ICRP No. 103 (2007), the "eco-centric" approach, according to which humans are a part of the ecosystem, and to consider the radiation safety system as part of the environmental safety system. This ensures not only the radiation safety of the population, but also the safety of the environment, which corresponds to implementation of the comprehensive biospheric approach. This approach assumes that each of the elements of the biosphere can be considered not only as an object that can be affected by radioactive contamination, but also as a source of such exposure.

So, in order to monitor and manage the process of how the radiation doses and environmental conditions are formed in the areas of NPP location for different modes of operation, one should make such changes in the currently used system of NPP radiation safety, which will complement its environmental component. It may become a radiation monitoring system of the environment area around the NPP location.

The proposed by the author of this article **NPP Radio-Ecological Monitoring system (REM)** is the result of generalization of the results of research scientists of Ukraine, Russia, Belorussia and other countries, as well as personal researches [9]. Its main provisions are stated below.

The initial postulates that were derived from many years of international experience of NPP operation, and which define the methodology of creating **NPP REM systems** are as follows:

- NPP is not only a source of radionuclides and ionizing radiation, but also the chemical and thermal pollution, and changes associated with human factors, such as the construction and urbanization of area of its location;
- a critical factor in the impact of NPP on the environment and human is the radiation, which is becoming a major cause of accidents;
- NPP system for ensuring radiation safety should be based on the principle of ecological and hygienic regulation, which is mainly intended to protect people against exposure to harmful factors, but taking into consideration ecological conditions and features of its habitat;
- the methods applied for ensuring environmental safety including the radiation safety of NPPs should take into account the behavior of different chemical substances (both stable and radioactive) in ecosystems in the area of NPP location, their characteristics and properties.

The methodology (concept, principles, methods) **of the routine and emergency SEM NPP is based on the following concept**: monitoring should be comprehensive (monitoring and control of all elements of the environment and all kinds of contaminants, regardless of the type of a source), take into account the systemic unity between "NPP + other activities + natural environment + man" and presence of feedback between the operational process at the NPP, condition of the environment and human health, which ensures compliance with the principles of ecological and hygienic standards of impact factors and provides the ability to manage the environment.

On the way of how the impact of different factors on the environment should be accounted, **an integrated NPP REM system should be** radiation-based, but environmentally focused as regards the content of studying the impact of these factors on the ecosystem, so it should be radio-ecological. It should also take into account the characteristics of geosystems, located in the zone of NPP influence (types of landscapes, landscapes geochemistry, etc.), migration conditions of environment (weather, sinks, places of accumulation, etc.), physical and chemical properties of contaminants of different nature (isomorphism, isotopic and non-isotopic analogs, and the like), the combined effect of pollutants of various nature, as well as other factors that determine the level of NPP impact on humans and the environment.

The proposed concept stipulates the following **scheme of tasks and implementation of the complex radiological environmental monitoring** in areas of NPP location, according to which the problems are to be solved as follows:

1. Observation of all impact factors and the state of the environment, assessment of the existing level of pollution of the environmental elements and the level of environmental hazard and risk assessment, identification of critical factors, ways and mechanisms of effects on humans and natural environment.

2. Modeling of the behavior of priority contaminants, particularly radionuclides, in the environment and food chain, the definition of the critical elements of the environment, the critical links in the food chain in order to predict environmental contamination and radiation burden on the population and the definition of critical

population groups, etc.

3. Environmental management (environmental safety) in the areas of NPP location.

The proposed scheme of NPP REM includes the tasks that are routine in the normal monitoring (monitoring of the impact factors and the environment, assessment of the existing level of contamination of elements of the environment, forecasting changes in the existing level of environmental pollution due to various factors). However, ecocentric approach is used as the basis to manage these tasks, which corresponds to the principle of ecological and hygienic regulating of radiation exposure factors, i.e., it makes the human a constituent part of the ecosystem. This causes the expansion of the classic information-oriented functions of monitoring, to the management-focused ones, which corresponds to modern scientific views and requirements for environmental safety. In this way the monitoring is transferred from the category of informational systems to the informational-and-controlling systems.

In our case under the control of environmental safety in accordance with the currently existing definitions [11-15] we mean a **continuous targeted cyclical process** in which, on the basis of the information received, the management body effects on the object to count negative factors, threats and risks to human and the environment, i.e., there is a feedback between a technological process and the state of the environment and human health.

One of the options of implementation of the management function of the monitoring is integration of the information obtained at all its stages, by means of expert systems [16].

In the case of municipal-level radiation accident methodology of REM on the territory, which is subject to the impact of an accidental NPP, it also should be based on ecological and hygienic principles of regulating radiation factors, and should take into account peculiarities of the atmospheric transport of an accidental release, physical geography, landscape, geochemical, demographic and other characteristics of the area, and the results obtained by conducting regular monitoring (preventive preparedness to accident response).

It is reasonable to conduct this assessment of levels of environmental contamination and radiation burden on the population for critical radionuclides (taking into account their importance), critical elements of the environment, including agrosphere as a critical industry as regards its contribution to the human dose, and for critical groups of the population. (Critical elements are the indicators that identify changes in the environment at the early stages).

Management (change of quality in order to reduce the critical level) of the environment in the event of an accident is transformed into a variety of countermeasures to minimize the levels of environmental contamination, doses on the population and the number of members of the public who appeared to be under accidental radiation exposure.

The scheme of NPP REM, which displays the proposed methodology, is shown in Figure 1. The main method of REM on the territory of NPP location is a landscape-geochemical zoning, i.e. division of the monitoring territory into homogeneous areas according to their characteristics.

This makes it possible to build a monitoring network, which will take into account not only the most adverse weather conditions, but also the terrain, landscapes and soil types, density of contamination, population, and other quantifiable characteristics of the terrain and a source of emission. There should be about 100 observation stations in the network.

The representativeness of the estimates obtained in the conduct of monitoring, is provided by the method of selection and preparation of medial samples used for the analysis of radioactive contamination.

In order to achieve the same (equal) accuracy of the measurement results, the distribution of the number of average samples for the study area should be accounted with appropriate weighting coefficients in proportion to the sizes of the area, contamination density, population, and other identifiable factors important in terms of risk.

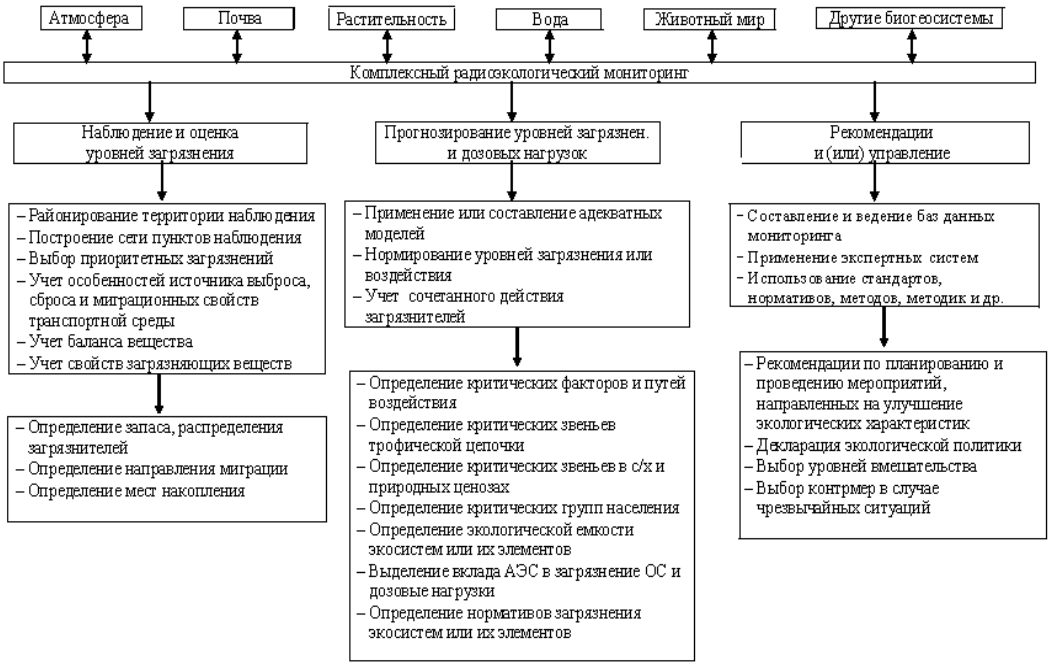

Figure 1. Scheme of integrated radiological environmental monitoring in the NPP location area. The REM system, which was based on the above methodological approaches, was successfully carried out at the Zaporozhye NPP, Chernobyl NPP (before and after the accident), Khmelnitsky NPP and Balakovo NPP, as well as at Odessa thermal NPP. In carrying out these monitoring studies of NPP representative and comparable results were obtained, which allowed reducing the errors and uncertainties in evaluation of radioecological situation in the controlled area.

It should be noted that the method of REM, based on the above methodology, including the methodology of zoning of territories around NPP and formation of a monitoring network, was developed under the guidance of Professor B.S. Priester and the staff of the NPP Department of the Odessa Polytechnic Institute in the early 80-ies of the last century. It is presented in the "Guidelines on the organization of monitoring the state of the natural environment in the area of NPP location", approved by the Ministry of Health and the State Hydrological Committee of the USSR in 1988 and endorsed by the board of Gospromatomnadzor of the Soviet Union in 1989, but, unfortunately, it still remains beyond the attention of nuclear experts of the industry.

Implementation of a radioecological monitoring system built on the basis of "ecological" methodology at NPPs would give the possibility of obtaining a representative and comprehensive information on the state of the environment in the NPP location area in all modes of its operation, and would effectively manage the environmental risks.

Nowadays, the approaches that take into account contemporary trends of increase of radiation and ecological safety on radiation hazardous objects, methodologically similar to the outlined above, are being actively implemented at Russian NPPs [17-21] and in Belarus at the construction of NPP in the town of Ostrovets. The Ukrainian NPPs conduct this work in an obsolete manner, and in some cases - even on the basis of the "Soviet" methods and recommendations, such as [22-25]. However, awareness of the need to improve the methodological, methodical, regulatory and legislative support of activities aimed at improving the NPP environmental safety, creation and introduction of new NPP environmental technologies already appears in our nuclear sector documents [26].

The proposed methodology of REM could be the base for all the NPP. On this basis methodologies, regulations, guidelines on radiological monitoring and REM management on NPP can be developed (or we can improve the existing ones). This will increase the efficiency of management of radiation and environmental safety of the plant.

Preventive Preparedness for Emergency Response. We cannot exclude the probability of accidents at NPP. This fact makes topical the problem of emergency response, which includes the need to create a response system beyond the NPP site, preventive preparedness of state authorities to accidents - notification of the population, monitoring and radiation control, implementation of emergency countermeasures, etc. These measures are "primary" in the overall system to overcome the consequences of accidents. It is urgent, but

preventive measures that will prevent the deliberate exposure of large population groups, prevent the occurrence of panic and rumors, and reduce the social and psychological effects of stress.

At the beyond design basis and severe accidents preventive emergency plans should provide for intervention levels and countermeasures to ensure effective radiation protection of the population and the environment. Methodology of development of emergency response plans on elimination of design basis accidents are adequate and complete, because it is based on many years of operating experience. But conditions of severe beyond design basis radiation accident require creativity in action planning, not only from the NPP staff, but also from special services far beyond its borders, with analysis and consideration of a large volume of geophysical, demographic and environmental information about the zone of impact of the NPP accident, which determines formation of the radiation environment and radiation exposure on the population. These actions fall within the competence of the State – the ministries of ecology, agriculture, health, and others.

The conclusion from the foregoing is the need to establish and maintain high level of **the national system to respond to potentially possible radiation accidents**. Creating such a system must begin with the organization and work on preventive preparedness for emergency response, including the development and implementation of a national system of ecological monitoring, covering the whole territory of Ukraine, and built on the basis of the ecocentric approach. Such a system would have been a source of information necessary for preparation of forward-looking assessment about the features of the formation of the radiation conditions and radiation exposure to the population in case of accident; creation of **the national unified state automated radiation monitoring system**, was in plans for nearly 30 years, but has never been implemented in reality [27]. This situation remains pending despite the fact that the analysis of the situation in terms of organization, legal, methodical, technical, information and financial support of radiation monitoring of the environment, including ARMS, has already been completed for Ukraine, and the reality of creation of the radiation monitoring and emergency response national system is confirmed the huge number of scientific papers and practical developments [27].

If we want to have domestic nuclear energy which is safe for people and the nature, we must keep in mind that the costs of training to respond and the prompt accident respond itself are much cheaper than the cost of overcoming their consequences. This is evidenced by the world's experience in liquidation of consequences of radiation accidents.

Creation and implementation of technology of reliable disposal (burial) of long-lived high-level radioactive waste. Modern nuclear power which is based on thermal neutron reactors, has a number of unresolved and pending issues affecting the pace of development, the structure and efficiency of nuclear power on a global scale. The most important of these problems are:

- Mineral and raw material base of uranium is forecast to be unable to provide sustainable long-term development of nuclear power using only thermal neutrons reactors, as they use only a small part of uranium raw materials;
- The lack of reliable technology of isolation from the biosphere of radioactive waste generated in a nuclear fuel cycle. In particular, this applies to long-lived high level radioactive waste, which includes spent nuclear fuel, if one intends to dispose it without processing, and it is declared as waste.

In the short term the only technologically achievable method capable of providing sufficient protection of the population and the biosphere from long lived HLW for several hundred years, is the disposal of waste into deep underground storage. The ideology of management of long-lived high-level waste in this case is that they are solidified, and then placed under controlled long-term storage and subsequent disposal in a deep geological formation.

In several countries in recent decades programs were developed for the construction of deep repositories for the final disposal of spent nuclear fuel or long-lived HLW (Finland, Sweden, and the United States). And although the idea of a deep underground waste disposal for the purpose of complete isolation from the biosphere seems obvious, yet not any country has begun operation of a geological storage facility.

Organization of a deep disposal facility is a very complex multi-factorial problem that needs long-term and extensive amount of scientific and engineering work. However, the main obstacle is the lack of absolute guarantees of long-term safety of such a facility, and now safety evidence for solving any environmental problems should be extremely transparent.

According to scientific concepts, the geological repository for long-loved HLW should be created in geological structures, which have long-term stability, water resistance, good sorptive properties to retain radionuclides, etc. Clay, rock salt, and such rock as granite, basalt, and tuff are considered as appropriate. Rock formation is a major natural barrier to the release of radionuclides to the biosphere. Artificial barriers are filling

(buffer material), canisters and containers in which the waste is placed, than the solid matrix as such (matrix of fuel, or a matrix, in which the incorporated high-level waste is solidified).

This multi-barrier system should isolate the waste for a long period of time - until hazardous radionuclides decay. Usually this time is estimated as 10 half-lives: for the fission products it is about 300 years, for the actinides - more than 100 thousand years. And this is the core of the problem. It is possible to predict the condition of a disposal facility and the intensity of release of nuclides from it for a few hundred years (although in this case there are questions when choosing the evolution scenario, than there are uncertainties in the description of processes, parameter estimation inaccuracies, incomplete knowledge of the geological structure and, as a result, an error in the choice of a site, etc.)

But the uncertainty in the long-term forecast of the condition of a disposal facility, and thus the release of radionuclides leads to the fact that in dealing with disposal of radioactive wastes a social and political dimension becomes an important inhibitory factor at the present stage. Confirmation is the situation with the construction of the Yucca Mountain national spent fuel repository in the USA.

Recently, experts from many countries have come to the point that the crucial issue of nuclear power – how to handle long-lived HLW – is possible to resolve only in a large-scale multi-component nuclear energy sector based on the use of fast reactors in the closed nuclear fuel cycle with deep reprocessing of irradiated nuclear fuel, extraction of individual radionuclides or their groups to return valuable raw materials into the energy cycle, the use of some useful isotopes, transmutation of particular actinides, and disposal of unused nuclides without disturbing the natural radiation balance [28].

Preserving the natural radiation balance means that after a certain historically not too long period (about 300 years), the total radiotoxicity generated from the operation of NPP and / or processing of irradiated fuel and waste being sent for disposal will not exceed the total radioactivity of uranium raw materials extracted from the earth's crust for production of NPP fuel [29].

In 2011, Russia launched a project named the **Breakthrough** (“Proryv”), consolidating the projects aimed at development of high-power reactors on fast neutrons, a closed nuclear fuel cycle technologies, as well as new fuels and materials, and also focused on achieving a new quality of nuclear power [30 - 31].

- The objective of the project is to create nuclear power complex, consolidating NPP with fast neutron reactors, production plants of regeneration (recycling) and re-fabrication of nuclear fuel, preparing all types of radioactive waste to final removal off the technological cycle, and which meets the following requirements:

- Ensuring the competitiveness of nuclear energy compared to fossil fuel of power generation, taking into account all the costs of both hydrocarbon and closed nuclear fuel cycle;

- Closure of the nuclear fuel cycle for the complete use of the energy potential of uranium raw materials;

- Consistent approach to radiation equivalent disposal of radioactive waste (with respect to the natural raw material);

- Reduction of capital costs for the construction of NPP with fast neutron reactors (at least to the level of NPP with thermal neutron reactors) due to technological and design solutions that are unique to fast reactors.

In 2014, at the site of the Siberian Chemical Plant the construction was started of a pilot demonstration power complex as a constituent part of a fast reactor with lead liquid metal coolant, BREST-OD-300 with the on-site nuclear fuel cycle, which reduces the potential risk of impacts on the environment and the population at the stages of fuel mining and milling, transportation of nuclear materials and radioactive waste disposal. As a part of the energy complex it is planned to tryout the NFC technology necessary for the future of nuclear energy.

It would be fair to say that not all the experts support the idea of the “Breakthrough” project, pointing to a number of errors and unresolved issues that may hinder its implementation. [32] Nevertheless, Russian project is still the only one in the world, aimed at a practical solution to the problem of handling long-lived HLW.

Conclusion

Generalization of existing ecological problems of nuclear energy, related to various factors of impact on people and the environment allows us to select **three groups of tasks which will help to improve the environmental safety of nuclear power**:

1. Obtaining representative information on the conditions of the environment in the vicinity of the NPP location for the purpose of environmental control and minimization of potential environmental impacts during normal and emergency modes of NPP operation.

2. Organization and carrying out of works on preventive preparedness for emergency response in order to reduce the environmental and radiological risks emerged in accidents at NPP.

3. Establishment and implementation of technologies for reliable disposal of long-lived high-level radioactive waste

The first problem can be solved through implementation on NPPs of a radiation monitoring system, built on the basis of ecological methodology that will give the possibility to obtain representative and comprehensive information on the condition of the environment in the NPP location area in all modes of its operation, and effectively manage environmental risks.

Solution of the second problem is associated with creation of a **national response system** for responding to potential possible radiation accidents, which should start with implementation in practice of the national radiation monitoring system covering the whole territory of Ukraine, built on the basis of the ecocentric approach. Such a system is a source of information necessary for the preparation of forward-looking assessment about the features of the formation of the radiation environment and radiation exposure to the population in an emergency, and creation in Ukraine of the Unified State Automated Radiation Monitoring System.

Solution of the third task is possible in principle only in a large-scale multi-component nuclear energy sector based on the use of fast reactors in the closed nuclear fuel cycle with deep reprocessing of irradiated nuclear fuel.

In conclusion, it should be emphasized that solution of environmental problems of nuclear power is vital, and it would be a terrible mistake to underestimate their seriousness. Although in this case we shall keep alive the nuclear power sector and don't close nuclear power plants: in many countries it is the only possibility to get low-cost energy and be independent from political conditions and preferences of other states.

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ADDITIONAL TECHNICAL POSSIBILITIES TO IMPROVE NPP SAFETY AND REDUCE RISK OF NEGATIVE IMPACT OF NUCLEAR POWER FACILITIES ON THE ENVIRONMENT

The article considers the causes of reactivity accidents at the conditions of an NPP unit blackout with failure of mechanical part of reactor control and protection system (RCPS). It is proposed to use high pressure turbine pumps units with friction-vortex turbine drive for supplying boric acid solution from the accumulator tank of ECCS using the energy carriers which are invariant to electric power supply in a possible accident without loss of coolant. The advantages of friction-vortex turbines over blade-type ones for the use as a feeding pump drives are shown.

Key words: reactivity accident, emergency power supply, power independent pump drives, NPP safety systems, frictional-vortex turbine.

Nuclear and radiation safety of nuclear power plants (NPP) is largely determined by the efficiency and reliability of regular (active and passive) safety systems. However, contemporary engineering means of safety systems do not fully meet the existing requirements. As it was shown by the accident at the NPP Fukushima-1 [1], among the possible NPP operational conditions is blackout of power units, which significantly limits the ability to reduce the risk of reactivity accident. As for Ukraine, according to safety analysis of Khmelnitsky NPP Unit 2, the core damage frequency at the condition of overlapping of several accident initiating events caused by full blackout is 2.4961/year [2].

Given the blackout conditions, the prescribed development of events involves emergency reactor shutdown, insertion of control rods of reactor control and protection system (RCPS) into the core with appropriate operation of electromagnetic stepping actuators. However, as it was demonstrated by the experience of events that occurred at NPP «Temelin» (Czech Republic, 2007) a situation is possible when at the emergency protection mode of reactor operation the control rods would not be inserted into the core because of distortions of guiding channels for control rods of the RCPS [3]. In such a situation, until the concentration of boric acid in the primary circuit water has not reach the level required for suppression of nuclear chain fission reaction in nuclear fuel the overheat of the core and in-vessel components may occur by reducing the coolant flow through the reactor with taking into account termination of operation of the main circulating pump units (MCP) that are power-dependent. In this case, the natural circulation of coolant, which was considered in the current designs of WWER type reactors, may not be fully effective due to changes of hydro-resistance of the reactor core. This can be enhanced by changes of the geometric parameters and forms of parts of fuel bundles, which may be caused by the same reasons that led to the distortion of guiding channels for RCPS control rods movement.

At increase of temperature of the core components - the temperature of fuel rods and fuel rod claddings, as well as water that cools the fuel assemblies (FA), the thermal-hydraulic and neutron-physical effects may occur that promote unacceptable increase of the reactor's reactivity. Indeed, when temperature of the fuel rods increases, it will cause (possibly locally) the temperature increase above the saturation temperature of the coolant, which will lead to boiling (sub-boiling) - first of all, of course, in the center of the core.

Origination of the steam fraction in coolant water can lead to undesirable positive steam effect of reactivity: when water begins to boil, the coolant density depends essentially on the amount of steam, and it is clear that the change in reactivity is proportional to reduction of the coolant density, and simultaneously it is dependent on the magnitude of the neutron flux density that increases the considered physical effect.

In general, as it is know, the steam reactivity coefficient that determines the effect of reactivity can vary over a wide range from negative to positive values depending on the composition of the core and the particular mode of reactor operation.

Large heat transfer coefficients in the "FA-coolant" system that are usually associated with boiling, could temporarily reduce temperature of the fuel. Local improvement of the heat transfer from the surface of the fuel rods to the two-phase coolant may temporarily lead to decrease of the fuel temperature in a certain local volume of the core. It is known that when temperature of the fuel decreases, it leads to narrowing of the energy absorption spectrum of neutrons of uranium isotope 238U, which is contained in the fuel composition. Therefore, at decreasing of the fuel temperature the neutrons are more likely to escape absorption at 238U resonances which leads to increase of reactivity [4].

The positive effect of the reactivity of the reactor is certainly dangerous if it cannot be compensated by the control rods insertion. Thus, this physical phenomenon which is associated with the increased reactivity of the reactor is quite a probable event and should be considered as a possible hazard (risk) of a reactivity accident. Flowchart of events and cause-effect relationships relating to these physical processes is shown in Fig. 1.

Undesirable development of the situation may be to some extent prevented by prompt emergency feeding of relatively cold coolant water with the aim to reduce its temperature within a range that promote elimination of the above effects. It is known that regular means of emergency core cooling (ECCS passive part) are designed for operation in terms of loss of coolant accidents when the pressure in the primary circuit drops to technology provided set point which is significantly less than normal working pressure.

Cooling of the core by additional feeding of the coolant using high pressure pumps (active part of ECCS) that are power-independent, becomes ineffective or at least ineffective in case of the unit blackout (with the loss of standby power)[5]. Therefore, in the considered situation that could lead to increase of the tendency of development of the reactivity accident, it's hardly possible to rely on unconditional performance of regular safety systems. So, the problem is obvious, but there is a ground to believe that it can be eliminated through the use of additional reserve means of feeding the main circulation circuit by the coolant of acceptable temperature; these means should be power-independent and functionally suitable in conditions of a blackout.

Indeed, as a means of reducing the risk of reactivity accidents we can consider turbine pump unit, providing feed water (aqueous solution) from ECCS reservoirs through the additional special dedicated feedwater line.

Fig. 1 - Block diagram of events and causes of a reactivity accident in the conditions of full blackout of an NPP Unit

It would be highly desirable that a pipe in such a power-independent pump would be adapted to use the working media of any phase state with the aim to be invariant to any media which is available at NPP (steam, water, steam mixture etc). It is clear that a conventional turbine blade, suitable for operation on only specially prepared (separated) steam, in this case would be unsuitable or ineffective.

In such a device, which can be in a "cold" (standby) mode, it would be useful to use blade-free frictional vortex turbine, such as a one with a disk rotor, as it is provided for in a Tesla turbine [6,7]. It is known that turbines of this type have high efficiency, but their efficiency can be increased using some technical improvements [8-10] and be quite sufficient in terms of technical problem when safety is valued more than performance.

The results of our experiments give reason to believe that there is some room for further modification of this class of devices. The scheme of experimental stand which can be used for further researches is shown in Fig. 2.

Preliminary experiments have shown the demonstrated suitability of gas, liquid and gas-liquid media as working fluids using multidisc model design (simulator stand design) of a rotor for frictional-vortex turbine shown in Fig. 3

In the conditions of pre-accident or accident operation of a nuclear power facility as the working fluid any media can be used: unprepared wet steam, steam-water flow, and even streams containing fractioned solids.

Selection of the working media for supplying it to the turbine pump can be made directly from the reactor circuit, as shown by the example of WWER-1000, Fig. 4.

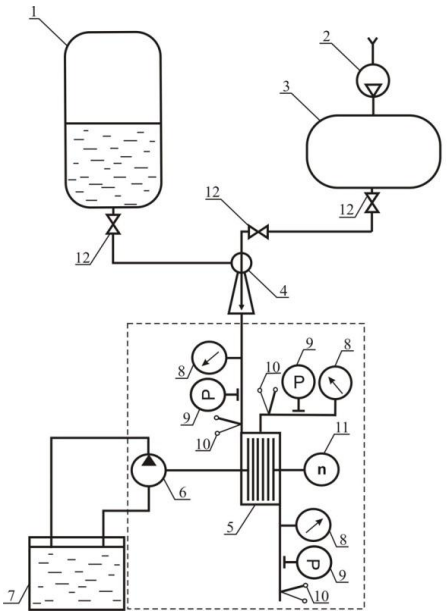

Fig. 2 - Layout of aerial and thermal-hydraulic laboratory equipment of the stand:
1 - pumped storage tank; 2 - compressor; 3 - receiver; 4 - injector; 5 - model of frictional-vortex turbine; 6 - pump; 7 - a water tank; 8 - pressure gauges; 9 - pressure sensors; 10 - thermocouples; 11 - tachometer; 12 - valves

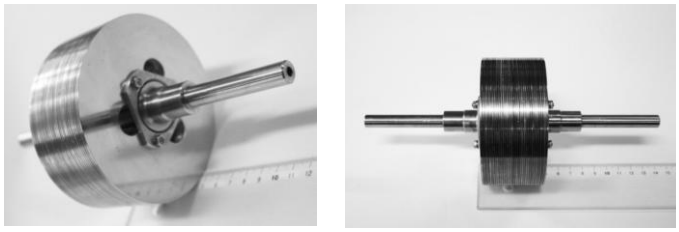

Fig. 3 – The rotor of a stand simulator of a frictional vortex turbine (assembled)

Start of this feeding unit do not require any preparatory actions and can be arranged in automatic mode by signals from sensors of thermometric passive control connected with a corresponding automatic (normally closed) valve on the pipeline of supplying the working fluid to the engine of turbine pump unit.

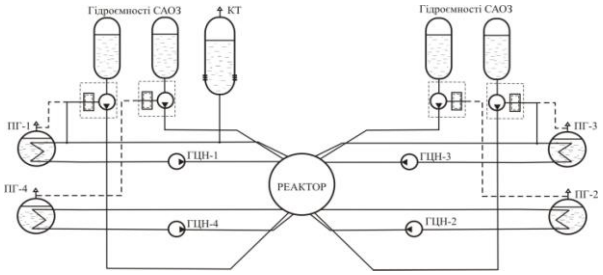

Fig. 4 - Scheme of possible backup application of energy independent drives of pumps, supplying aqueous solution of boric acid to the main circulation circuit of the reactor installation WWER-1000

The degree of passivity of this safety system is large enough, which is determined by the lack of necessity to control it by an operator, and by actual independence from specific any sources, in particular, from electric power supplying.

Application of the described system can effectively compensate dangerous trends that lead in extraordinary circumstances to reactivity nuclear power plant accident, and should enhance the NPP safety.

Conclusions

We considered physical possibility of development of a reactivity accident to an NPP unit blackout. It is

shown that this kind of event is quite possible for the equipment of modern nuclear power plants with WWER-1000 reactors. The suggested means of overcoming the existing problems by reducing risk through supplying of water media to the main circulation circuit of a reactor facility with additional back-up energy-independent pumps with turbo drives based on frictional-vortex turbines which are invariant to flows of engine fluids in any physical state. The use of the proposed technical solutions should help reduce the risk of NPP impact on the environment in case of an accident.

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RIVNE NPP IMPACT ON VOLUME ACTIVITY OF TRITIUM IN THE RIVER CTYR

The article presents results of researches of fluctuations of tritium volumetric activity in the river Styr, caused by discharges from Rivne nuclear power plant. It is shown that tritium background activity in the Styr may be exceeded ten times due to discharges from Rivne NPP. It was found that due to irregularity of tritium discharges from RNPP and fluctuations in the water level of the Styr, values of tritium volumetric activity in the river Styr may vary in a fairly large range that requires improvement of the system of monitoring tritium, both in discharges, and directly in the Styr water. Results of the studies have shown that NRBU-97 dose limits due to the critical use of water are not exceeded during tritium discharge.

Key words: volumetric activity of tritium, Rivne NPP, unbalanced water, discharge, the river Styr.

Introduction. In 1991 the United Nations Economic Commission for Europe initiated the Convention on Environmental Impact Assessment in Transboundary Context (Espoo Convention) [1], which provides for international cooperation among the Parties to the Convention in the public discussion, planning and evaluation of industrial potentially dangerous objects (including NPP) in a transboundary context. This convention was ratified by the Government of Ukraine on March 19, 1999 by the Law of Ukraine No. 534 XIV. According to Article 9 of the Convention (Research Program) the Parties pay special attention to the development or more active implementation of specific research programs, including those in accordance with paragraph 2, directed on better understanding of cause-effect relationships and their role in integrated environmental management. The researches described in the present article are fully consistent with paragraph 2. That is why the study of impact of the nuclear power plant, which has the cooling pond and discharges of unbalanced water in transboundary river Styr, is rather important.

Rivne NPP (two units of WWER-440 type, and two WWER-1000) is located in the district of the river Styr, which originates in Ukraine and falls in the river Pripyat in Belarus, and then the Pripyat in Ukraine falls in the Dnipro river. The Rivne NPP discharges into the Styr about 3.0 TBq of tritium annually [2].

Tritium, which is generated at the NPP, enters (as part of discharge water) the surface water without decontamination, and its activity is significantly greater than the activity of other radionuclides such as: 137Cs, 134Cs, 60Co, 54Mn, 60Sr. The latter is because the activity of other radionuclides is significantly reduced as a result of decontamination of waste water.

At present, radiobiological characteristics of tritium are investigated incompletely. New mechanisms of radiation exposure on humans have been revealed. Standards of tritium content in drinking water vary greatly in different countries. Therefore it is important to have reliable information on tritium contamination of surface water from the NPP, especially in a transboundary context.

The results of the studies presented in this paper are based on features of tritium discharge from the Rivne NPP, and of hydrological characteristics of the Styr in different months of the year, causing fairly significant fluctuations of tritium volume activity both during the day and throughout the year. This study covers the years 2010, 2014, and 2015.

Volume activity of tritium in water of the Styr River by the results of research expeditions in 2015. In June 2015 the authors conducted research expeditions on the Styr and took water samples for determination of tritium (Table. 1). Places of sampling are shown in Fig. 1.

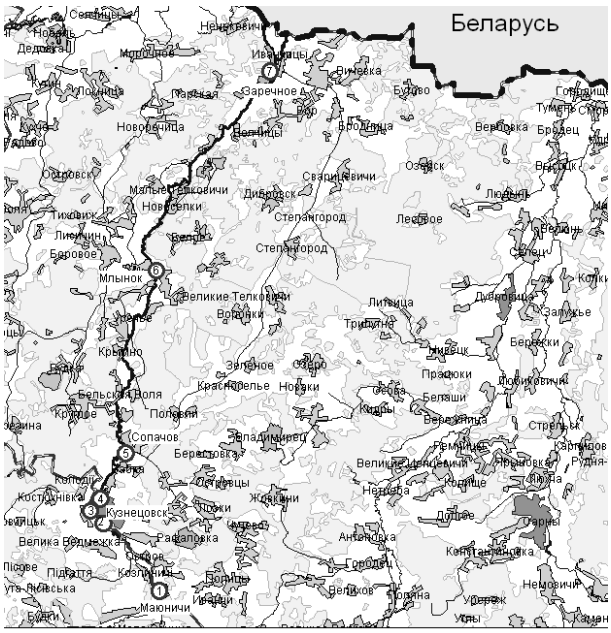

Fig. 1. Scheme of water sampling (the circles show the sampling points)

Table 1. Activity of tritium samples taken in June 2015 at the Styr river

Point No.	Description	North latitude, degrees	East longitude, degrees	Volume activity Bq/l	Notes
1	Maiunichi, bridge	51,25	25,94944	3,2	10 km upstream from RNPP
2	Kuznetsovsk	51,32917	25,84722	6	2,4 km upstream from RNPP water intake
3	Kuznetsovsk	51,31622	25,87528	135,3	The place of RNPP water discharge
4	Kuznetsovsk	51,31667	25,87528	26	50 m downstream from the point of RNPP water discharge
5	Sopachiv village	51,40233	25,89044	4,9	10 km downstream from RNPP
6	Mlynok village	51,60506	25,94403	4,4	40 km downstream from RNPP
7	Zarychchya village	51,82572	26,14542	18	70 km downstream from RNPP

Selection and preparation of water samples for measurement of tritium activity was conducted under DSU ISO 9698-2001 [3]. The volume activity of tritium measured at liquid-scintillation α-β-spectrometer Quantulus-1220-003 (Finland). The total relative error did not exceed 20%.

Directly in place of discharge of return water from the Rivne NPP the observed tritium volume activity was 135.3 Bq/l. At a distance of 10 km from discharge of tritium volume activity was equal to 4.9 Bq/l, at a distance of 40 km (v. Mlynok) - 4.4 Bq/l and on the border with Belarus - 18 Bq/l. This dependence of tritium volume activity indicates a non-uniform discharge of tritium from Rivne NPP.

So, in October 2015 we selected the water samples from the Styr River at a distance of about 40 km from the NPP (v. Mlynok) at time intervals of 1.5 hours.

The data are presented in Table. 2.

One can observe substantial dependence of tritium volume activity from the time of water sampling. In addition, the volume activity of tritium is much higher than the previous values. The average volume activity of tritium for the period from 10-00 to 19-00 on November 3 is 17.7 Bq/l, and the next day from 7am to 1pm average volume activity of tritium was 26.7 Bq / l. This dependence of tritium volume activity from the time of water sampling confirms a preliminary conclusion about the uneven discharge of tritium into the Styr River from Rivne NPP. And, one can claim significant deviations of tritium volume activity in the Styr from the background values. Fluctuations of tritium volume activity are largely caused by seasonal periods of the year. As an example, the maximum volume activity of tritium at a distance of 25-30 km downstream from the discharge point is observed in summer and autumn-winter time when the water flow in the river is minimal, which results in minimal dilution of discharges [4].

Modeling of tritium spread. When modeling the spread of tritium from Rivne NPP up to the Belarus borders we used software package PC CREAM [5]. This complex is designed to calculate radiation exposure of long-term (non-emergency) emissions and discharges of radioactive substances into rivers and seas. To evaluate the effective dose we used ICRP Publication No. 60 [6] according to the methodology [7].

There are three phases in modeling of the distribution of radionuclides in rivers [8].

The first phase - the spot of primary dilution and mixing across the depth of the channel extends to a distance of about a hundred times the depth of the river. This is the phase of initial mixing. Regarding the Styr River in the point of discharge of Rivne NPP – this is a distance of about 100 m.

Table 2. Activity of tritium in water samples of the Styr in v. Mlynok (point 6, Fig. 1)

Sample No.	Date of sampling	Time of sampling, hour, min	Volume activity Bq/l
1	03.11.2015	10:00	15,6
2	03.11.2015	11:30	16,8
3	03.11.2015	13:00	22,1
4	03.11.2015	14:30	18,8
5	03.11.2015	16:00	19,0
6	03.11.2015	17:30	13,5
7	03.11.2015	19:00	18,1
8	04.11.2015	7:00	32,3
9	04.11.2015	8:30	23,4
10	04.11.2015	10:00	27,4
11	04.11.2015	11:30	29,4
12	04.11.2015	13:00	21,1

The second phase – the phase of complete mixing, when the mixing takes place almost throughout the entire width of the river or across the cross-section of the channel. This process can occur at distances of several tens of kilometers [7, 8].

The third phase begins after the second phase, and is characterized by spread over long distances and interaction of radionuclides with suspension substances or substances that make up the bottom sediments.

In the simulation we used a "dynamic model" that takes into account hydrological parameters of the river. The whole simulation area from Rivne NPP up to the borders of Belarus territory was divided into three sections (chambers), taking into consideration the characteristics of the river.

On the 1st section of 20 km length, we modeled the spread of tritium cased by the tritium discharges from the NPP (the first and second phases of mixing). In this region there are no major tributaries.

On the 2nd section of 40 km length, we considered distribution of tritium in the river up to the confluence of the tributary – the river Stubla.

The third section, of 15 km length, covers the part of the river Styr up to the border with Belarus. Here is the confluence of the Stubla, and the Styr is divided into two horns – the Styr and the Prostyr.

Average perennial water content of the Styr in the vicinity of Kuznetsovsk city, village Mlynok and village Zarichchya related as 38:42:46 [9]. In calculations we assumed water content in sections 1-3 of the river in the same proportion.

Number of suspended solids in water - $1 \cdot 10^{-4}$ t/m^3 [10]. Speed of flushing of the upper layer of sediments in all sections is $1 \cdot 10^{-5}$ m/s [7], which is typical for small depths and small slopes of the water surface. For the Styr in the bottom the average slope of the water surface is 0.27% [11].

Simulation results of tritium distribution in 2010. According to the long-term observations of water regime in the river Styr [10] (the database as of 01 January 2007) in an extremely low-water year the water consumption in August is 0.5% of annual flow, and in March - 52.7% of annual flow. In the high-water year in August it is 1.3% of annual flow, and in March - 32.4% of annual flow. These data confirm that the distribution of annual flow by months of the year is very uneven. As a consequence of the fact that the discharge of tritium from RNPP is uneven throughout the year, the tritium volume activity should vary considerably during the year. Therefore, for the quantitative calculation of volume activity of tritium into the river Styr we need the data on the total discharge of tritium and by- month water consumption in the river Styr.

Tritium discharge for months in 2010 from RNPP according to [12] is shown in Fig. 2 (total for the year - 2660 GBq). The largest discharge was in December, and the lowest - in August. Water consumption in the river Styr in 2010 is shown in Fig. 3. The left column of the histogram is a long-term average consumption of the Styr in the city of Kuznetsovsk based on measurements at Rivne NPP [13], and right columns - the average long-term consumption of the Styr near the village Mlynok according to the State Committee for Hydrometeorology [14].

It is clear that these data vary greatly: the State Committee for Hydrometeorology's data of average perennial runoff fluctuations of the Styr water yield have much larger amplitude excursion than in our measurements near Kuznetsovsk. There are no major tributaries between the city of Kuznetsovsk and village Mlynok that could affect the water yield of the river. The calculations used the data from the State Committee for Hydrometeorology.

Calculations of volume activity of tritium for various distances downstream of the Styr are shown in Fig. 4. The background activity which is equal to 3.2 Bq/l is added to the volume activity of RNPP tritium discharge. These calculations correspond to average monthly activity of tritium that could be registered in 2010. On the first section of average monthly the volume activity varied in the range 3.4-13.4 Bq/l, and on the border with Belarus – in the range of 3.3-11.5 Bq/l.

Fig. 5 shows the calculation of volume activity of tritium in different months of the year on the border with Belarus due to only discharges from Rivne NPP. The results do not include the background activity. The lowest volume activity was observed in January, and the highest - in November.

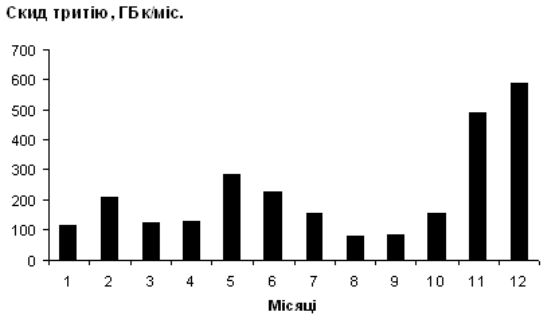

Fig. 2. Tritium discharge in 2010 from RNPP

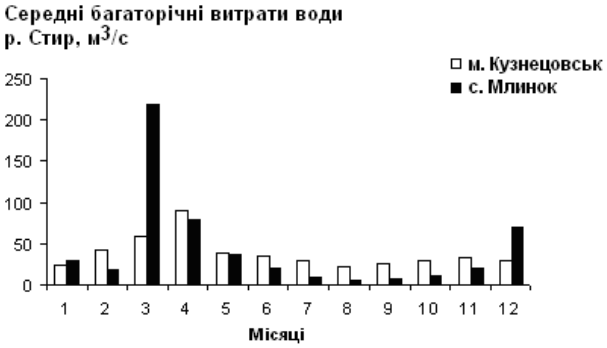

Fig. 3. Average perennial water yield of the river Styr (in m^3/s) in the city of Kuznetsovsk and village Mlynok

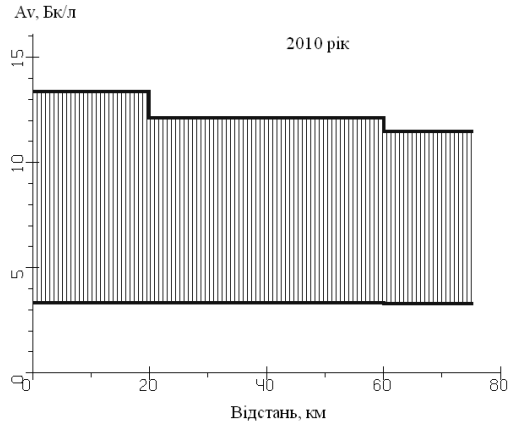

Fig. 4. The range of fluctuations of volume activity (Bq/l) of tritium at different distances from Rivne (km)

Fig. 5. Estimated volume activity of tritium at the border with Belarus in 2010 (without the background activity) in Bq/dm^3.

Contribution to maximum volume activity of tritium caused by the discharge of RNPP in November at about 8.3 Bq/l is negligible compared to the value permitted under NRBU-97 for drinking water activity which is

equal to 30 000 Bq/l [15]. According to the EU directive of 1998 the volume activity of tritium in drinking water should not exceed 100 Bq/l [16]. As it follows from the above calculations and the EU directives, the values prescribed by EU regulations are also observed. In the US standard for drinking water [17] it is allowed the tritium volume activity 20 000 pCi/l or 740 Bq/l. In Russia the allowable volume activity of tritium in drinking water is 7600 Bq / l [18].

Note that in the international practice the regulated values of acceptable tritium volume activity in drinking water, varies greatly. It is clear from the above that the most stringent standards are in the EU (100 Bq/l), less stringent they are in the United States (740 Bq/l), after that, in Russia (7600 Bq/l), and completely "soft" standards are in Ukraine (30,000 Bq/l).

The annual dose from tritium releases from the Rivne NPP is 53 nSv, which is well below the dose limit for NPP critical water use which is 10 000 nSv/year according to NRBU-97 [15]. Hence, the impact on Belarus in 2010 due to discharges of tritium from RNPP was essentially below the established dose quotas and the limit for individual effective annual dose from all types of impact 1 mSv (1000000 nSv).

From all routes of exposure of tritium in the water the main contribution to the total dose is caused by the following: consumption of fish and water (with contribution from consumption of fish around 1%).

Simulation results of tritium distribution in 2014. Fig. 6 shows the distribution of absolute discharges of tritium from RNPP monthly in 2014 [19]. This figure is also shows for comparison the data on discharges of tritium in 2010. Left transparent columns of the histograms are of 2010, and black right - of 2014

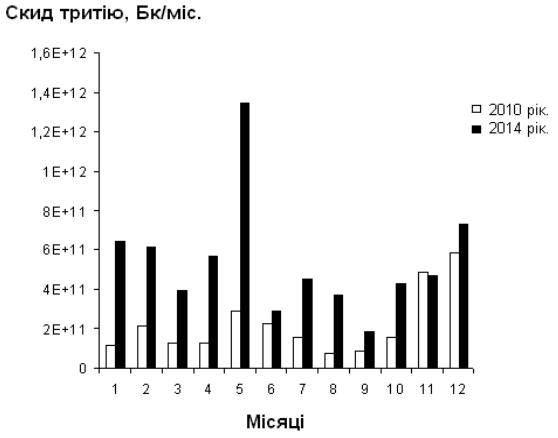

Fig. 6. Discharges of tritium from RNPP monthly in 2010 and 2014

As it follows from the data shown in Fig. 8, discharge of tritium in 2014 compared to 2010 increased 2.43 times, from $2.66 \cdot 10^{12}$ Bq to $6.47 \cdot 10^{12}$ Bq.

Water consumption (flow) in the river Styr in 2014 near the village Mlynok is shown in Fig. 7. Right black columns show the water in the river Styr near the v. Mlynok in 2014, and the left white bars show the average long-term consumption in the Styr near the v. Mlynok (given for comparison). We can see that every year, the distribution of data of the Styr consumption is significantly different. Therefore, for the correct calculation of volume activity of tritium discharged, we used only the data relating to the tested year

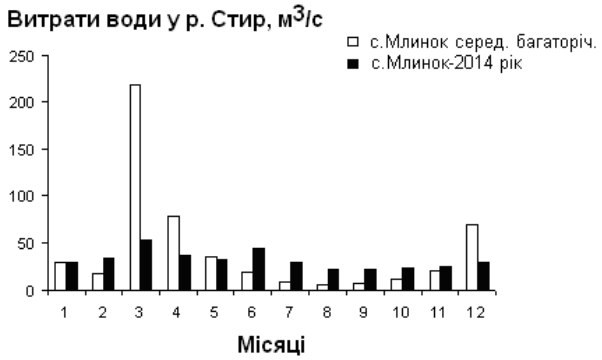

Fig. 7. The average long-term water consumption (intensity of flow) of the Styr near the village Mlynok and the data in 2014

Fig. 8 shows the calculation of volume activity of tritium discharges caused by RNPP on the border with Belarus. Tritium volume activity varied from 2.3 to 14.2 Bq/l. These calculations do not include background activity that is 3.2 Bq/l.

In 2010 the expected maximum values of volume activity of tritium on the border with Belarus were

estimated in November and July (see. Fig. 4), but in 2014 the maximum volume activity was expected in May and December (Fig. 10). The magnitude of the average volume activity mainly influenced by two factors: the value of discharge of tritium in a given month and the value of water level of the river. These two factors act in the opposite directions. If at increase in discharge the volume activity increases, then with increasing of water flow it decreases.

Fig. 9 shows the distribution of doses received at the border with Belarus from the RNPP discharge of tritium.

The total annual dose in 2014 is 90 nSv (in 2010 - 53 nSv). It should be noted that tritium discharges in 2014 were 2.43 times higher than in 2010, and the total annual dose in 2014 is 1.7 times higher than the dose of 2010. This is due to the difference in the distribution of monthly water consumption and discharges of tritium.

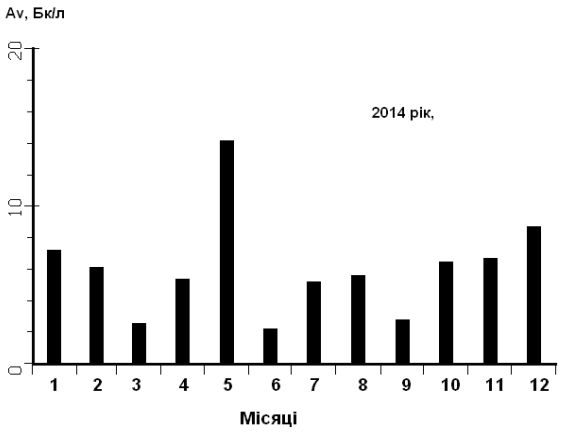

Fig. 8. Tritium volume activity on the border of Ukraine with Belorussia in 2014 due to RNPP discharge

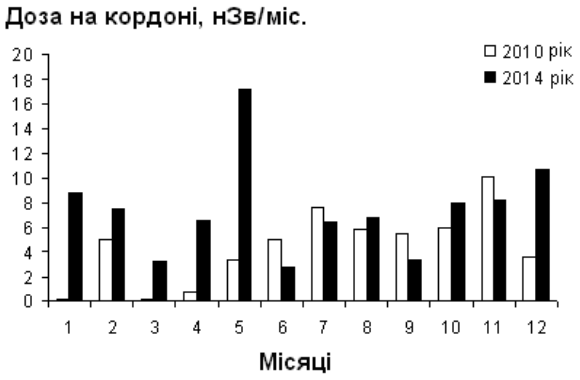

Fig. 9. Average monthly doses at the border of Ukraine due to PNPP discharge of tritium

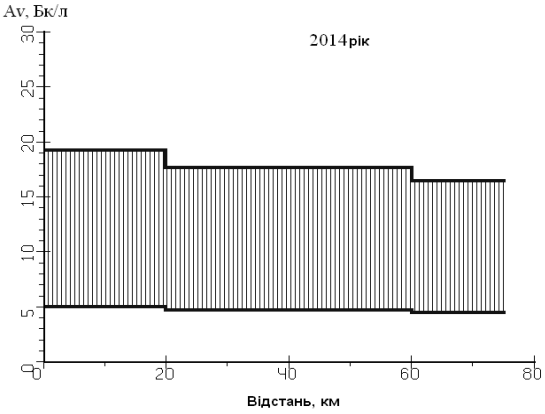

Fig. 10. The range of fluctuations of volume activity of tritium at different distances from RNPP

Calculation of tritium volume activity for various distances downstream the river Styr is shown in Fig. 10. In this Fig. the background activity of 3.2 Bq/l is added to the volume activity of Rivne NPP discharge. On the 1st

section the average monthly volume activity is varied in the range of 5.1 to 19.3 Bq/l, and on the border with Belarus it is 4.6 – 16.5 Bq/l. The comparison of data in Fig. 4 and Fig. 10 for different years indicates that as a result of uneven discharge of tritium from Rivne NPP and changing hydrological conditions in the river Styr the volume activity of tritium on the border with Belarus can vary significantly. Therefore, for correct account of impact of RNPP on contamination of the river Styr in a transboundary context it is necessary to improve monitoring of discharges of tritium.

Results of simulation of tritium distribution in 2015. Analysis of tritium contamination of the river Styr water in 2015 was complicated by the fact that we had no data on discharges of tritium RNPP by the moment of our study. But in 2015 we managed to get data on water flows in the river Styr and data of the measured volume activity of tritium.

Fig. 11 shows the value of the Styr flow near the village Mlynok in 2015. In this figure the dotted curve shows the measured data on certain days, and the solid curve – the approximation of measured data based on B-spline of the 3d level. The columns show the calculated numerical integration of the flow for each month.

Comparing data from the monthly flow of the Styr near the village Mlynok for 2014 and 2015, it should be noted that 2015 was low-water as compared to 2014. Lacking the data on discharges of tritium, we accepted that they were in 2015 the same as in 2014. Based on this assumption, we should expect that the volume activity of tritium on the border with Belarus in 2015 would be greater than in 2014.

Calculations using these data at the border with Belarus are shown in Fig. 12.

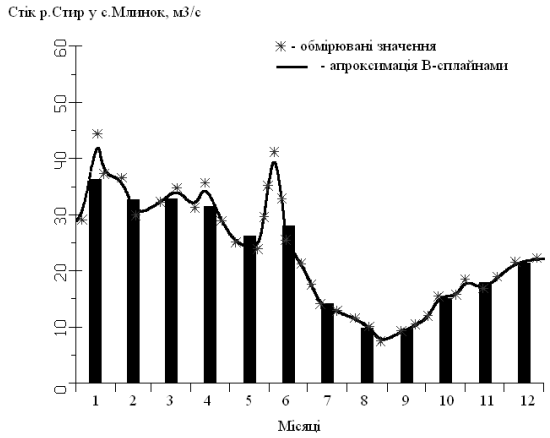

Fig. 11. Water flow of the river Styr in 2015 near the village Mlynok

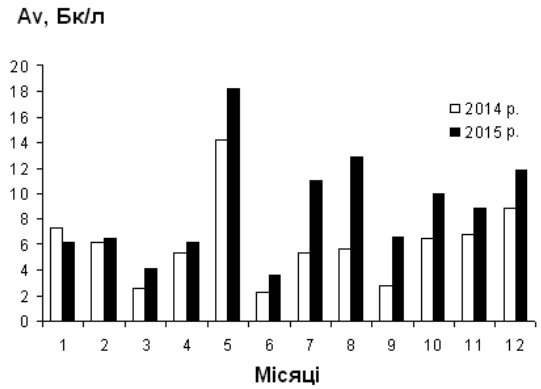

Fig. 12. Volume activity of tritium on the border with Belarus in the years 2014-2015 due to discharges from Rivne NPP

The white (transparent) left columns relate to 2014 and black right ones - to 2015. Volume activity in 2015 varied from 3.6 to 18.2 Bq/l. These calculations do not account the background activity (3.2 Bq/l). As it follows from the presented results, the tritium volume activity, due to the discharge of RNPP in 2015 from February to December, was higher than in 2014.

Fig. 13 shows the distribution of radiation doses received by the population due to discharges of tritium from RNPP during 2014-2015. In 2015, the radiation dose from February to December was higher than in 2014, and only in January it was lower. The total dose in 2015 was 128 nSv (in 2014 - 90 nSv).

Once again we can see that the values of doses are significantly determined by the distribution during the

year both of tritium discharges from RNPP and of water level in the river Styr. Therefore it is necessary to improve monitoring of discharges of tritium and of water level in the river. It is desirable to take more water samples to measure tritium and take integrated samples for a few hours or a few days, and to measure water flow at least once a day.

Calculation of tritium volume activity for various distances downstream of the river Styr is given in Fig. 14. It shows a range of monthly fluctuations of volume activity of tritium at different distances from Rivne NPP.

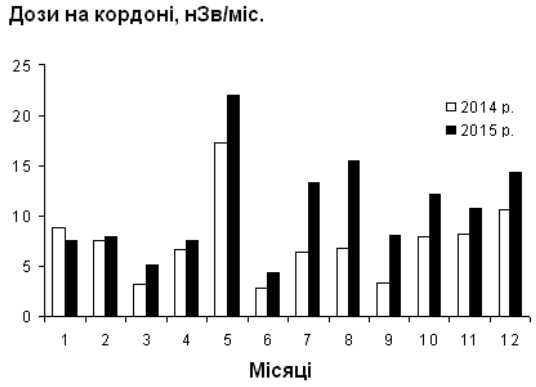

Fig. 13. Dose values at the Border with Belarus in the years 2014–2015 due to RNPP discharges

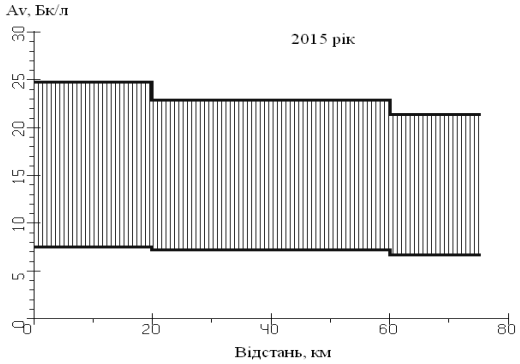

Fig. 14. A range of fluctuations of average volume activity of tritium at different distances from Rivne NPP in 2015.

In Fig. 14 the background activity (3.2 Bq/l) was added to contribution to the volume activity of RNPP discharges, i.e. the activities that can be measured directly. In the first section the average monthly volume activity was varied in the range of 7.6 to 24.8 Bq/l, and on the border with Belarus - from 6.8 to 21.4 Bq/l.

The measurement results of daily fluctuations of tritium volume activity in the river Styr. In June 2015 samples of water in the river Styr were taken, and the measurement results (including measurement errors) are shown in Fig. 15 and subsequent Figures. The relative measurement errors according to our estimates of repeated measurements do not exceed 20%. The results of measurements of volume activity in the samples are shown in Table. 1. All samples were taken during one day. In the morning the first sample was taken near the village Mayunychi - activity of tritium in this sample determines the volume of background activity in the river Styr before discharge from RNPP (3.2 Bq/l). Then sample number 2 was taken in the vicinity of the city Kuznetsovsk approximately 2.4 km above the intake. The volume activity in this sampling point is affected by tritium discharges from RNPP cooling towers and other sources of the city. Volume activity in this point is above the background. The third sample was selected directly from the discharge flow. Fourth sampling point was 50 m below the discharge (area of partial mixing of discharged water). Further samples were selected in points № 5, № 6 and № 7. The last test in point No. 7 was taken in the evening, approximately at 6 p.m. Thus, during one day there were selected samples of water throughout the whole area of the river Styr in Rivne region. For simulation of daily tritium distribution, the measurements in the river Styr the following data were used: water flow in the river Styr in June was 25.5 m^3/s (measured) volume activity of tritium in the discharge - 135.3 Bq/l (measured), water flow in the discharge of RNPP, in June 2014 - 0,55 m^3/s (assumption).

The results of simulation of tritium volume activity in the river Styr at different distances from the discharge together with the measured values are shown in Fig. 15. Negative values are distances to points upstream from the point of discharge. 0 km point is the place of discharge.

Simulation results and measurements coincide well with each other. The only difference is at 70 km from the place of discharge. We assume that the relatively higher activity in this point is due to increased activity

discharged before the beginning of our measurements. If we assume that this increased activity was discharged late in the night, than it could reach the border of Ukraine during the day, where we have registered it

These data allow us to assume that the RNPP discharge of tritium is uneven during the day. So we conducted subsequent sampling at one point at regular intervals during November 3 and 4, 2015 to explore how the tritium volume activity was changing during the day. The results of the measurements of tritium volume activity are presented in Table 2.

Calculations show that the flow of water in the village Mlynok late in October 2015 was 15.3 m^3/s, and the average flow velocity is 0.79 m/s. Therefore, the time when water from the point of discharge reached the point of measurement is approximately 14 hours. Mean volume activity of 3 November 2015 is 17.7 Bq/l (samples 1-7 in Table. 2)

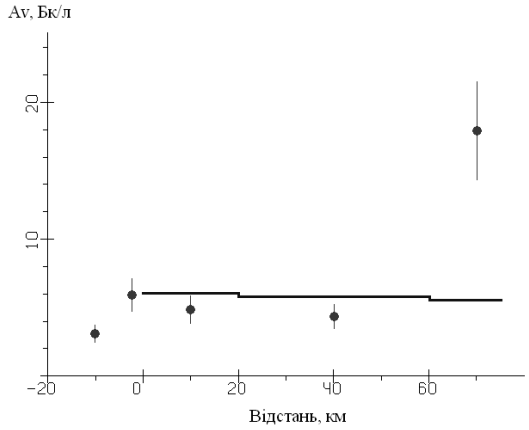

Fig. 15. The volume activity of tritium at different distances from Rivne NPP

Two different values, the discharge and the volume activity are in temporary interrelation, so for their presentation we use a coordinate system with one abscissa (time) and with two ordinates (discharge capacity and volume activity). The results of measurements are shown in Fig. 16 in the form of points with measurement errors. The time interval which is shown in the Figure, begins at 00 a.m. on November 2 and ends at 12 p.m. on November 3. The method of least squares was used for the solid line which shows the average value of volume activity during the study. Solving the inverse problem of dilution of discharge into the river Styr we can find that in the interval from 8 p.m. of November 2 to 5 a.m. on November 3 the tritium discharge of Rivne NPP was held at a speed of 0.4 MBq/s (left vertical axis and the column in Fig. 16 at which the dependence of the estimated discharge of tritium from RNPP on time is shown).

This figure also shows the time dependence of the measured volume activity of tritium in the river near the village Mlynok – this is the right vertical axis and the measured points. Measured values are shifted in time by 14 hours (time of water run) with respect to the time of discharge and are within the time period from 10:00 to 17:00 of November 3, 2015. The solid curve which runs between the points corresponds to the average value of volume activity in that day - 17.7 Bq/l.

Measurements of volume activity of tritium in the samples taken on November 4 showed that in that day the average volume activity of tritium was 26.7 Bq/l, those were samples 8-12 of 4 November, see Table 2.

Using a similar relation of volume activity measured on November 4, 2015, with a speed, we can obtain the estimated rate of RNPP tritium discharge from 17:00 to 23:00 on November 3 - the left vertical axis (rys.17) and the calculated value of the discharge speed at a specified time - 0.65 MBq/s (histogram column).

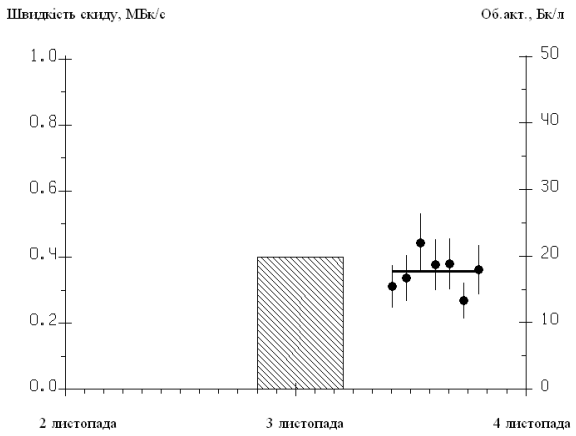

Fig. 16. Relation of RNPP tritium discharge activity and volume activity in the Styr (v. Mlynok), as of 03 November 2015

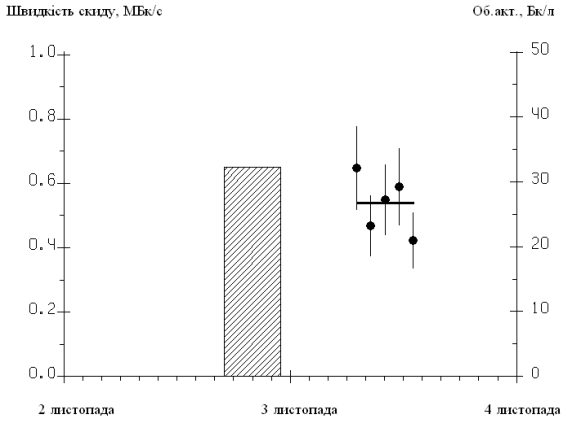

Fig. 17. Relation of RNPP tritium discharge activity and volume activity in the Styr (v. Mlynok), as of 04 November 2015.

The same figure shows the time dependence of the measured volume activity of tritium in the river near the village Mlynok - the right vertical axis and the measured points. The measured values are shifted in time by 14 hours (time of water run) with respect to time of discharge and are in the range of time from 7:00 to 13:00 of November 4, 2015. The solid curve that runs between the points corresponds to the average volume activity on that day - 26.7 Bq/l.

With this average volume activity of tritium in the village Mlynok we can expect on the border with Belarus the value of estimated volume activity of 24.8 Bq/l. Thus, at the border, according to our measurements volume activity of tritium can approximately 8 times exceed the background value (3.2 Bq/l).

According to [19] $1{,}037 \cdot 10^6$ m^3 were discharged during November 2014, i.e. the discharge rate is approximately equal to 0.46 m^3/s. Given the discharge capacity of 0.65 MBq/s volume activity of the discharged water approximately equals to 1400 Bq/l, and at the discharge capacity of 0.4 MBq/s - 870 Bq/l.

These relatively high levels of volume activity are much lower than 30,000 Bq/l permitted by NRBU-97 [15]. The more stringent restrictions are imposed by reference levels of annual water discharge. For discharges of tritium the “control level” at Rivne NPP is 2600 GBq/year [19]. Based on this value, we can find that the average permitted level of tritium discharges is 0.8 MBq/s.

In certain hours and days the discharge capacity, as it follows from the data in Fig. 17 is almost equal to the annual average permitted level. These facts show that tritium discharges from RNPP vary significantly during the day. Hence, it is necessary to improve monitoring of tritium discharges; in particular, it is desirable to take averaged samples over the certain time.

Conclusions

As a result of the research the following conclusions can be made:

1. It is shown that as a result of fluctuations in water flow of the river Styr and uneven discharges of tritium from RNPP the volume activity of tritium in water on the border of Ukraine can significantly vary.

2. As a result of observations tritium volume activity in the river Styr it was proved, that the discharge of tritium from RNPP fluctuates during the day.

3. On the border between Ukraine and Belarus in the river Styr it can be observed the activity of tritium up to 24.8 Bq/l, which is 8 times above the background activity. This increase is due to the impact of Rivne NPP.

4. The radiation doses due to discharges of tritium from Rivne NPP in the river Styr, which are caused by the consumption of drinking water are less than the limits of Radiation Safety Standards of Ukraine for critical water consumption for NPP, which is 10 μSv.

5. Due to the fluctuation of volume activity of tritium in the water of the river Styr it is necessary to monitor tritium contamination with periodicity that will keep track of these fluctuations.

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MODES OF DEGRADED HEAT TRANSFER IN SUPERCRITICAL WATER FLOW IN THE CHANNELS WITH FUEL ROD BUNDLES

Heat transfer processes in the flow of supercritical pressure water in fuel assemblies of nuclear reactors were investigated, with the analysis of conditions of degraded heat transfer. Numerical simulation was carried out of heat transfer in water flow with supercritical pressure in a seven-rod fuel assembly and a central unheated rod. In these conditions ($q/\rho w$=1, 2 kJ/kg) the mode of degraded heat transfer occurs locally in the central cells of the fuel assembly between the heated rods after reaching the critical temperature of coolant. In these cells sharp increase of temperature of walls of fuel elements and a strong azimuthal non-uniformity of the heat transfer coefficient were observed

Key words: nuclear reactor, heat transfer, fuel assembly, supercritical pressure fluid, reactor coolant.

Introduction. In recent years there are a lot of discussions of the issue of transition to supercritical parameters of the coolant in the nuclear power industry [1,2]. Each country chooses its own direction of development of nuclear power on the basis of forecasts of economic development and based on the principles of safety, competitiveness, the presence or availability of fuel resources, methods of nuclear waste disposing, and nuclear non-proliferation.

Ukraine has not come to a final decision on the choice of its nuclear fuel cycle [3] yet. According to the Energy Strategy of Ukraine till 2030 [4], the main focus will be directed on life extension of the existing units with the expired design term. It is also planned to construct new light water reactors of generation III+. In [3] the challenges are identified facing the Ukraine's nuclear fuel cycle and, in particular, it notes that for effective utilization of spent nuclear fuel the structure of nuclear power should have both thermal and fast breeder reactors.

Within the framework of the international program GIF IY (Generation IY International Forum) six innovative nuclear reactors have been chosen that can be put into operation by 2030 [5-7]:

- Fast neutron reactor with a gas (helium) cooled GFR (Gas Fast Reactor) with a closed fuel cycle;
- A high-temperature reactor with a helium-cooled and graphite-moderated VHTR (Very High Temperature Reactor);
- A reactor with water coolant under supercritical pressure SCWR (Supercritical Water Reactor);
- Fast reactor with sodium coolant SFR (Sodium Fast Reactor), a closed fuel cycle, effective burning of actinides, and reproduction of uranium-based fuel;
- Fast reactor with lead / lead-bismuth coolant LFR (Lead Fast Reactor), a closed fuel cycle, actinide burning, and efficient reproduction of nuclear fuel.
- Thermal neutron reactor MSR (Molten Salt Reactor) with circulation of fuel based on molten salts, with full processing of actinides in the fuel cycle.

Two power generation technologies with efficiency of 45-47% stand out among these technologies; they use high-temperature helium-cooled reactors, and supercritical water reactors (SCWR).

The main purpose of the use of light water reactors with supercritical pressure (SCWR) is to increase the efficiency of existing technologies of PWR (Pressurized Water Reactor). This concept was considered by reactor experts in Russia, Japan, USA, France, Canada, China and other countries in the framework of international programs for the development of fourth generation reactors. Construction of NPP with reactors of SCWR type will significantly increase plant efficiency, reduce steel intensity as compared with the existing NPP with WWER and PWR [8-10]. Nuclear power units with SCWR are characterized by the following features [9]:

- Low coolant consumption and high coolant heat in the reactor core, which reduces the power consumption of circulating pumps;
- High enthalpy of direct steam, which ensures high efficiency factor of the installation;
- Single-phase coolant, and hence, absence of modes of heat transfer crisis (nucleate boiling);
- Implementation of the one-pass cycle, leading to the compactness of the reactor system;
- The ability to use technology and equipment of the supercritical pressure units operated on fossil fuel

A possible option is a design of the core with a thermal neutron spectrum to operate in an open fuel cycle with UO2 fuel and fast neutron spectrum to operate in a closed fuel cycle with MOX-fuel and reproduction ratio ≈ 1 or higher, up to 1.2 [11]. Table 1 shows main characteristics Based on the preliminary design developments of light water reactors with supercritical pressure, made in the USA, France and Russia.

Features of the core cooling water supercritical parameters are taken into account when forming the neutron spectrum, safety analysis and evaluation of the stability of the reactor. Table 2 shows main characteristics of SCWR developed in Russia and Canada, and Japan reactor plants with thermal and fast neutron spectrum based on the preliminary design data. The reactor installation WWER-SKD (SCWR of WWER type) are considered with natural (NC) and forced (FC) coolant circulation [12].

Table 1 - Main characteristics of the light-water reactors with supercritical water pressure

Parameter	HPLWR (France)	SCWR (USA)	WWER-SKD (Russia)
Electric capacity MW	1000	1600	1700
Coolant temperature inlet/outlet ^oC	280/500	280/500	280/530
Coolant pressure, MPa	25	25	25
Diameter and the thickness of the cladding, mm	8x0,4	10,2x0,63	10,7x0,55
Packing step of fuel rods, mm	9,5	11,2	12,0
Fuel enrichment UO_2 , %	≤ 5	≤ 5	≤ 5
Average fuel burnup, MW·day/kgU	45	45	About 40
Maximum cladding temperature, ^oC	About 670		

Table 2 - Main characteristics of the reactor installations with supercritical water pressure

Parameter	WWER-SKD	WWER- SKD	CANDU-X	SCWR
Neutron spectrum	Heat, fast	Heat	Heat	fast
Heat capacity, MW	3700	1250 (NC) 2500 (FC)	2500	1650
Electric capacity MW	1700	500 (NC) и 1000 (FC)	1200	720
Efficiency factor,%	45	43	48	44
Pressure, MPa	24,5	24,5	25	25
Inlet/Outlet Temperature, ^oC	290/ 540	390/ 540	350/625	381/504
Fuel	UO_2, MOX, cermet*	UO_2, MOX	UO_2 /Th	MOX
Enrichment, %	Up to 5 (heat), Up to 20 (fast)		4	
Fuel rod diameter, mm	9-11	9-11	7	7
* - CERMET fuel with additional barrier for a single-pass installation				

In Canada, the developments are conducted of advanced heavy-channel reactors CANDU-X (Canada Deuterium Uranium -X) with supercritical water as a coolant, of electrical power from 375 to 1200 MW (Table 2). [5]

A design of SCWR reactor on fast neutrons electrical capacity of 700 MW on supercritical parameters was developed in Japan (Table 2) [9]. The core is designed for high power density at the maximum linear heat load, not exceeding 39 kW/m. For the case when the coolant flow rate is 850 kg/s, the equivalent diameter of the core is 2.1 m, which is considerably less than of PWR reactors of the same capacity.

Modes of degraded heat transfer. With the development of technology of light water reactors with supercritical coolant there is a need for clear understanding of hydraulic processes in the coolant, and the solution to the problem of creating new structural core materials. In terms of thermal physics, one of the important safety problems is the problem of determining the coefficient of heat transfer in the reactor core and safe area of thermal loads to eliminate the possibility of transition to the mode of degraded heat transfer

There are quite a lot of studies in the world concerning the fluid flow and heat transfer in the flow of supercritical fluid in pipes. However, there is still no clear understanding of the heat and mass transfer processes in the flow of the coolant at a sudden change in its physical properties [13, 14]. The sharp change in the physical properties of the coolant, depending on various factors leads to a substantial variety of heat transfer modes. In the field of supercritical coolant parameters we should distinguish the modes of normal, degraded and improved heat transfer. In normal heat transfer modes it is observed a monotonic change of wall temperature along the length of the tube, in conditions with degraded heat transfer it is observed a sharp increase in the local temperature of the wall. Such modes are very dangerous, because the resulting thermal stress can lead to distortion or destruction of shells of fuel elements (fuel rods). The parameters for which such modes can occur, significantly limit the power level of the reactor.

Numerous studies of occurrence of degraded heat transfer modes at supercritical water flow in the pipes have

shown that strong reduction of the coolant density when approaching a critical temperature causes thermal acceleration of the flow, which reduces turbulence. Due to the difference in density near the wall and in the center of the channel the buoyancy forces emerge in the flow. The combined influence of buoyancy forces and acceleration of the flow can cause deformation of the velocity profile. At lifting of the coolant flow the velocity profile becomes M-shaped with a maximum near the wall, and tangential stress at the maximum point is zero. Consequently, a locking layer is formed near the wall for heat transfer, resulting in degradation of heat transfer and raise of the wall temperature.

There is still no consensus on the determination of the boundary values of the heat flux $q_{гр}$, exceeding of which leads to the mode of degraded heat transfer, and of dependence of $q_{гр}$ from the mode and geometrical parameters. The ratio of the heat flux to the mass velocity $q/\rho w = 0.6$ kJ / kg defines the lower boundary of occurrence of modes of degraded heat transfer for flow of supercritical pressure water in pipes [15, 16]. In [15] different recommendations are given for determining the boundary of occurrence of the modes of degraded heat exchange. However, the relations describing the dependence of the heat flux density from the mass flow rate $q=f(\rho w)$, were obtained in the narrow range of parameters and, usually, for only one type of the medium.

There is extremely small amount of results of experimental investigations of heat transfer in the supercritical water flow in muti-rod assemblies [13, 14]. Some studies have noted that for the flow of supercritical pressure water in rod bundles the mode of degraded heat transfer occurs at higher values of the $q_{гр}/\rho w$ parameter, in comparison with the pipe flow.

In [13] the experimental data are presented, characterizing boundary conditions of degraded heat transfer in the fuel channel of the eight tubes with a diameter of 12 mm, wall thickness of 0.8 mm with a downward and lifting movement of the coolant. The experiments were performed at pressures of 23.6; 24.6; 26.5; 29.4 MPa in the range of variations of mass speed within 400 - 4800 kg / (m^2*s), water inlet temperature 80 - 320°C, heat flux density up to 1,6 MW/m^2. As a result, it was found that in a certain range of parameters in the section of the channel, where the average temperature of the coolant is close to pseudo-critical t_m, it is observed sharp increase of the wall temperature of a fuel rod dummy. As temperature of the heat flow increases, there is a shift of temperature peak upstream with its simultaneous increase. An analysis of the experimental data [13] made it possible to obtain the following dependences of the parameter on the mass velocity for different pressure values (Figure 1).

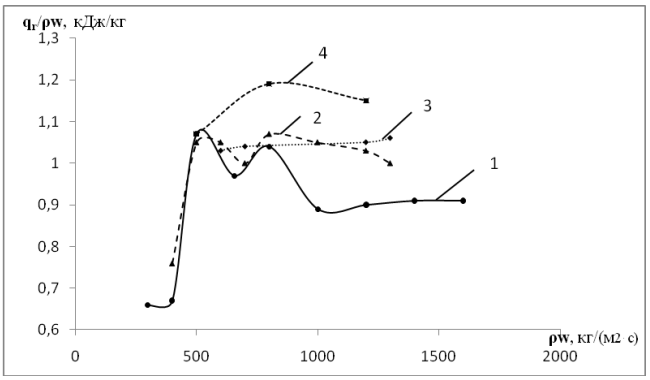

Figure 1. Change of parameter depending on the mass velocity and pressure: 1 - 23.6 Mpa; 2 - 24.6 Mpa; 3- 26.5 MPa, 4- 29.4 MPa

The figure shows that there are three areas with different influence of the mass velocity on the boundary of occurrence of the mode of degraded heat transfer in rod bundles. At low mass velocities (ρw<500 kg/(m^2s)) a significant increase is observed of the $q_{гр}/\rho w$ parameter from 0.67 kJ/kg to 1.08 kJ/kg; in the rage of 500 <ρw≤800 kg/(m^2s) it reaches its maximum value of 1.08 - 1.2 kJ / kg; and in the rage of ρw> 800 kg/(m^2 s) there is a tendency to reduction. The exception is the mode of the 26.5 MPa pressure (curve 3). In this mode, there are no experimental data in the mass velocity of 800 to 1200 kg/(m^2s). The experimental results show that for a given geometry of a bundle the $q_{гр}/\rho w$ parameter value varies between 0.67 kJ/kg - 1.2 kJ/kg. So we have not sufficient ground for talking about a substantial increase in the minimum value of the $q_{гр}/\rho w$ parameter in the bundles as compared with pipes.

The limits of occurrence of the mode of degraded heat transfer are significantly affected by the geometric characteristics of a bundle. This is evidenced by the results of calculations of hydrodynamics and heat transfer in

the flow of water in the supercritical pressure in the bundles with triangular or square packing [17-19]. Calculations were performed using both a per-channel method, and three-dimensional modeling.

In [17] the results of the calculation are presented (with the use of the FLUENT code), of heat transfer in a fuel assembly of the CANDU-X, a channel-type reactor cooled with supercritical water. The fuel assembly comprises eight ∅13.5 mm fuel rods of the central row and 35 ∅10.5 mm fuel rods in two outer rows, enclosed in a channel with the inner diameter of 102 mm and heated length of 5.77 m. The heat flux density along the channel and in its cross-section is constant - 670 kW/m^2, the pressure is 25 MPa, the mass flow rate is 860 kg/(m^2 s).

In the local area, where there is the mode of degraded heat transfer, there is an unacceptably high azimuthal non-uniformity of fuel cladding temperature. Maximum azimuthal non-uniformity of temperature $\Delta T_{\varphi,\max}$ = 306°C is observed in a cell of the central row, and the minimum 28°C - in a cell of an outer row. The maximum fuel rod surface temperature 729°C in a cell of the central row is achieved at a sectional length of the channel *z* = 1,443 m, where the average temperature of the coolant is close to the critical temperature of 384,9°C.

The results of a numerical modeling of flow and heat transfer characteristics in the square package of fuel assemblies with a fuel rod of 8 mm in diameter, 4 m long, cooled with supercritical pressure water are presented in [18]. Modal parameters correspond to the Chinese model of SCWR reactor with thermal power of 3020 MW with inlet water temperature of 280°C, the outlet temperature 510°C, the mass velocity of 1540 kg/(m^2 s), and the pressure of 25 MPa. Three types of cells were considered: a typical inner quadrangle, the cell between the fuel rods and the wall and the corner cell. For modeling it was used a modified RNG *k*-ε turbulence model, which showed the best agreement between the calculated values and the experimental data according to the heat transfer coefficient from the enthalpy of the coolant at supercritical pressure.

Calculations were carried out at constant heat flux on the wall of 600 kW/m^2 for different packing density of rods *s/d*, ranging from 1.2 to 1.6. In the inner quadrangular cells the mode of degraded heat transfer occurs when *s/d* ≥1.4 and parameter $q/\rho w$ ≥0,6 kJ/kg. Maximum heat transfer coefficient is observed in pseudo-critical coolant temperature, and then comes a sharp decrease in the heat transfer coefficient and simultaneous sharp increase in the temperature of the wall. The output section of the azimuthal non-uniformity of the wall temperature reaches 40°C and with decrease of *s/d* ratio it increases.

In the cells between the fuel rods and the channel wall the mode of degraded heat transfer occurs in a narrow gap between the core and the wall in the section where the coolant temperature reaches a critical value. In this section the azimuthal non-uniformity of the wall temperature can reach 90°C. In the corner cell of the channel the mode of degraded heat transfer also occurs in the range of pseudo-critical coolant temperature.

Modeling of heat transfer in a fuel assembly. Here we consider numerical simulation of heat transfer in the flow of water in the supercritical vertical 7-rods fuel assembly. The geometric and operational parameters of the assembly correspond to the parameters of the reactor core of WWER-SKD [1]. We consider the vertical assembly position, z-axis is directed along the axis of the channel. Rods bundle is 9 mm in diameter, with heated length of 4.05 m is located in a triangular box with a step-to-diameter ratio of the rods s/d = 1,4 (Figure 2). The central core and the canal wall is not heated, the walls of the six heated rod is given a constant heat flux. The inlet is supplied with water at a temperature of less than the critical for a given pressure

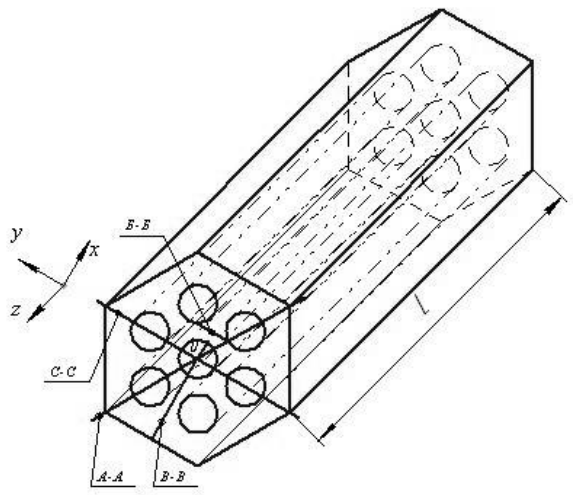

Fig 2. Fuel assembly layout

The study of fluid flow and heat transfer in the assembly was based on the RNG *k*-ε turbulence model, which includes a system of equations of conservation of mass, momentum, coolant energy, as well as the equations for the turbulent kinetic energy and energy dissipation rate [19-22]. The equations of state of water at pre- and supercritical parameters were set according to the formulations of IF-97. [23] The boundary conditions at the inlet of the fuel assembly: water temperature 563 K, speed 0.5 m/s, pressure of 25 MPa. The heat flux on the walls of the heated rods is 400 kW/m^2.

Calculations have shown that in a fuel assembly with a central non-heated rod the mode of degraded heat transfer occurs at the outlet of the assembly in the central cell in the gap between the rods (B-B section) at the parameter value > 0.8 kJ/kg. With increase of the $q/\rho w$ parameter the area of the degraded heat transfer is shifted upstream to the inlet section. So, when $q/\rho w$ = 1.2 kJ/kgs, the mode of degraded heat transfer occurs in the middle of the channel in central cells between heated rods. Figure 3 shows the variation in wall temperature along the length of the heated rod at points 1, 2.

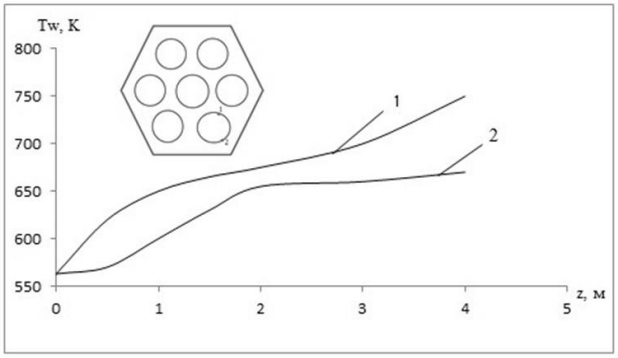

Figure 3. Distribution of wall temperature along the length of the rod

Point 1 in Figure 3 is the most heat-loaded one. In this point, the wall temperature along the length of the rod changes unevenly. On the section of the fuel assembly from the inlet to cross-section point z = 1,5 m the coolant velocity in corner cell is greater than in the center, so the temperature in point 1 increases greater than in point 2. The maximum point of the velocity profile in the angular cell is shifted to the heated rod. In cross section z=2 m maximums of velocity profile in the corner cell and the central cell virtually coincide (0.6 m/s). Starting from this section and up to going out of the assembly in the gap between the bars (cross-section B-B) the flow rate increases sharply and is 1.16 m/s at the outlet (Figure 4). The flow rate of the angular cell is changed weakly, and at the exit from the assembly the maximum velocity is 0.78 m/s.

It is interest to note that in contrast to the pipe flow, the velocity profiles in the bundle in the area of degraded heat exchange (curves 3 and 4, Figure 4) remain convex with a maximum in the middle of the gap between the rods. Formation of M-shaped velocity profile is not observed.

Figure 5 shows change of the temperature profiles in the B-B cross-section along the length of the assembly.

Coolant temperature in the section between the heated rod increases along the assembly length in sections z> 2 m, the temperature profile is strongly deformed, and there is the mode of degraded heat transfer. The wall temperature at point 1 (Figure 3) increases abruptly, and at point 2 it varies slightly.

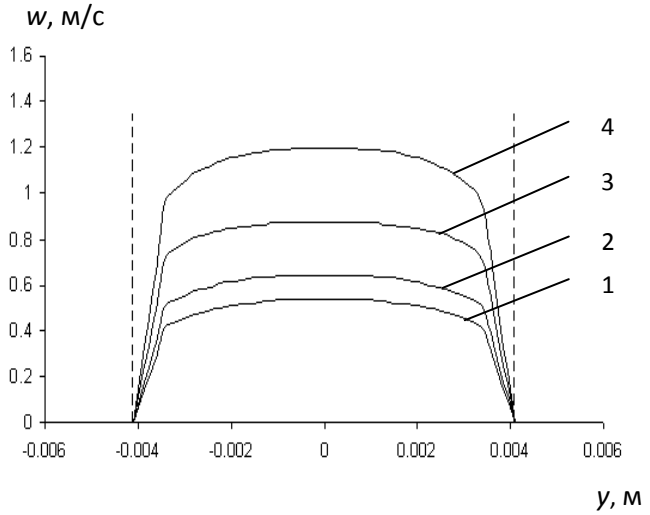

Figure 4. The velocity profiles in the cross section B-B: 1) z = 1 m, 2) z = 2 m, 3) z= 3 m, 4) z = 4 m

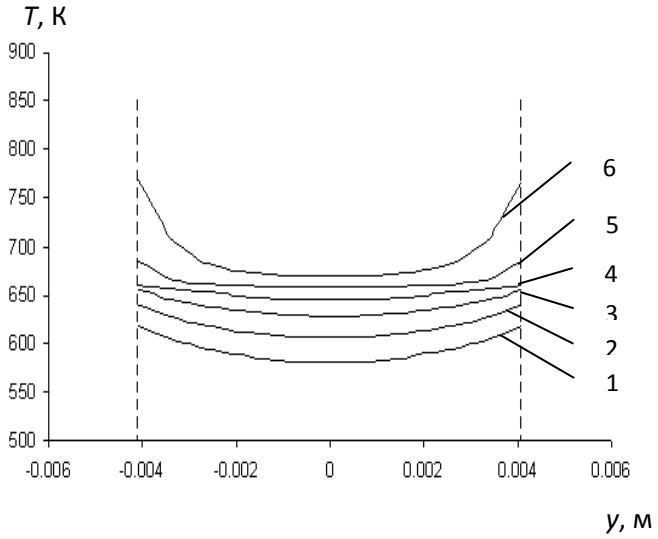

Figure 5. Temperature profiles in the cross-section B-B: 1) z = 0,5 m, 2) z = 1 m, 3) z = 1,5 m, 4) z = 2 m 5) z = 3 m, 6) z = 4 m**.**

Figure 6 shows the change along the length of the channel of heat transfer coefficient α in the cross-section passing through the point 1.

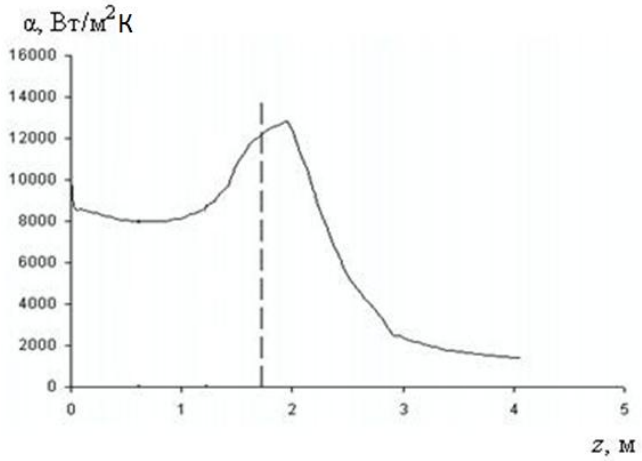

Figure 6. Changes in the heat transfer coefficient along the length of the rod

In the region where the coolant temperature reaches a critical value (dashed line), the heat transfer coefficient increases, and azimuthal unevenness of the wall temperature decreases. In the section *z*=2m temperature profile in the cross-section B-B becomes nearly uniform (curve 4, Figure 5), and the heat transfer coefficient becomes maximum. In the section z> 2 meters there is a sharp decrease in the coefficient of heat transfer, which is associated with decrease in the heat capacity in this field and thermal flow acceleration. Now, there is origination of the mode of degraded heat transfer. The wall temperature at point 1 (Figure 3) sharply increases and reaches 760 K at the outlet of the assembly, in this case unevenness of azimuthal temperature is 90 K.

Conclusions

Both experimental studies and computational simulations show that at the flow of supercritical pressure water in the channels with rod bundles in a certain range of values of the geometrical and operational parameters degraded mode of heat transfer appear, which is characterized by sharp increase of the fuel cell cladding temperature and its strong azimuthal non-uniformity. The minimum value at which the mode of degraded heat transfer may occur in the local region of the bundle is $q_{cp} / \rho w$ = 0.6 kJ / kg.

Numerical simulation of supercritical water in a 7-rods fuel assembly with unheated central core showed that at the considered conditions ($q / \rho w$ = 1.2 kJ/kg) the degraded heat transfer mode occurs locally in the cells of the central beam between heated bars after reaching critical flow temperature.

In the cells of the bundle, where the mode of degraded heat transfer emerges, we observe strong thermal acceleration of the flow, sharp decrease in the heat transfer coefficient and rise of the fuel rod wall temperature.

In the area of degraded heat transfer we observe a strong azimuthal non-uniformity of the wall temperature of the heated rods. Maximum azimuthal non-uniformity of the wall temperature at the exit of the rods is up to 90K.

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EXPERIENCE OF MULTI-YEARS HYDROBIOLOGICAL MONITORING OF NPP TECHNOECOSYSTEM

The results of researches of hydrobiological regime of water bodies of Khmelnitsky NPP techno-ecosystem obtained on the basis of long-term monitoring are presented. Positive and negative aspects of arrangement of such monitoring were considered. Optimization issues of hydrobiological monitoring, the relevance of which is increasing at Ukrainian NPPs because of introduction of EU Water Framework Directive requirements in the field of water management and environmental protection are discussed.

Key words: NPP techno-ecosystem, cooling pond, hydro-biological monitoring, background water bodies.

Close interaction of natural and engineered elements of the environment (water reservoirs, dams, canals, industrial units, etc.), and their biotic population, leads to the formation of a new type of natural-and-industrial systems. In turn, NPP cooling and technical water supply systems always have a connection with the natural water bodies, thus forming a kind of a techno-ecosystem.

All NPPs of Ukraine conduct permanent environmental monitoring of both radiation and non-radiation impact on the environment. However, in accordance with modern requirements for integrated environmental monitoring, it can be considered as a limited and incomplete. In particular, the EU Water Framework Directive (WFD) [1] regulates the use of hydrobiological indicators for environmental assessment of the aquatic bodies.

Organization of monitoring. Environmental monitoring is defined as a complex system of observations, evaluation and forecast of changes in ecosystems under the influence of anthropogenic impacts [2] and includes both biological geophysical and hydro-chemical aspects [3]. Hydrobiological monitoring is an integral part of a comprehensive environmental monitoring.

Integrated hydrochemical and hydrobiological studies of cooling ponds (CP) of thermal and nuclear power plants are carried out since the 1960s. [4]. At the initial stage, the main purpose of these studies was to investigate a state of aquatic organisms in the aspect of consequences of direct impact of power plants on the environment, especially of the so-called thermal factor contamination. The problem of reverse effects of biotic factors on technical systems (the possibility of bio-interferences and bio-caused failures in the plant's operation) did not receive sufficient attention.

Biotic factors can effects on technical systems in many ways. With vigorous "blooming" of water caused by cyanobacteria (blue-green algae) at the intensive photosynthesis the pH grows, thereby enhancing the scaling processes at heat transfer surfaces. Significant accumulation of algae in the surface layer of water reduce the penetrating solar radiation, thus reducing photosynthesis rate and oxygen saturation by other algae, including the bottom ones, which may lead to kill phenomena, and reduction of water quality in ponds.

Objects of monitoring. Among the invertebrate animals many can cause bio-hindrances in the work of equipment. They can be divided into three groups. The first group includes organisms living in the water column - plankton, including water-borne stage - zebra mussel larvae, statoblasts of bryozoans, sponges gemmules, which can settle and adhere to the substrate in one or another part of the water supply system. The second group includes the attached fouling - mollusks, bryozoans, sponges. The greatest problems in this group create dreissenidae (*Dreissena polymorpha Pall.*, and *D. bugensis Andr*.).

The third group of organisms, which can cause bio-disturbances are the organisms that enter the water supply systems with water flow. It is the developing in the intake channels fouling organisms (zebra mussels, sponges), as well as mobile gastropods and fish, higher aquatic plants and macroscopic algae [5]

Hydrobiological monitoring is necessary in two aspects: for reliable assessment of impact of industrial facilities on the environment, and for monitoring of potential or actual biological disturbances. We should note the relationship between these problems: biological disturbances may result in reduced reliability of the technical units, thereby increasing the risk of negative impact of technical facilities on the environment.

In conducting research we considered the purpose of hydro-biological monitoring of NPP techno-ecosystems as follows:

- study and control of populations and integrity of groups of aquatic organisms, which act as indicators of the condition of the aquatic environment, as well as influence on the formation of biological interference with the process equipment;
- monitoring and forecasting of the invasive process, that is possible appearance of alien organisms,

particularly dangerous from the point of view of formation bio-disturbances;

- short-term and long-term forecasting of increase or decrease the probability of biological disturbance.

Conducting the monitoring: spatial aspect. Over the period of years of research of cooling ponds the very concept of choice of observation points was changed. In 1960-1970-ies researches and permanent observation were conducted in the so-called "heated" zone, adjacent to the discharge of NPP heated water, and in "unheated" for the screening control purposes.

However, the studies have shown that the distribution of environmental conditions parameters, as well as the organisms themselves are much more complex, and depend on the design and way of operation of cooling systems.

In particular, at arrangement of extended hydrobiological monitoring (the monitoring, which is conducted on a special program with maximum coverage of observation points [6, 7]) at the objects of Khmelnitsky NPP it is expedient to carry out researches not only in cooling ponds, but also in other water bodies. Control over contour groups (benthos, periphyton) should be carried out in the area of inlet and outlet channels in the central part of the water body, in the southern part (benthos), in channels and on the dam (periphyton), as well as in shallow waters of southern part of the pond (higher aquatic plants). Studies have shown that the choice of observation points in the pelagic subsystem depends significantly on weather conditions, namely, the strength and direction of the wind, because the nature of the transfer of water masses at the man-caused circulation is constantly changing.

An important result of the monitoring, and the study of NPP techno-ecosystems was the introduction into a circle of constant observation of so-called background water bodies. The importance of their study is determined by, on one hand, the fact that their ecosystem may reflect the impact of the NPP techno-ecosystem, if any, and on the other hand, these closely located ponds and streams can be sources of undesirable, even hazardous aquatic organisms. Selection of background water bodies [7] that are hydraulically connected with techno-ecosystem but it is not directly involved, is determined by the very concept of techno-ecosystem. For example, in the Khmelnitsky NPP techno-ecosystem the background are the Gorin River and the Gniloy Rog river, several pitches in the area of the cooling pond. From the Gorin river the cooling pond periodically receives feedwater, and the Gniloy Rog river is completely accumulated in the cooling pond. Respectively, the biological material and the water of a certain quality comes with the river water. The Gniloy Rog river water before flowing into the cooling pond is characterized by a relatively high content of phenols (82 ± 6 g/dm 3), in connection with significant development of higher aquatic plants and their withering away, which defines high content of these compounds, at least in the water of the southern region. For comparison, in the water supply channel phenol content was 1.8 times lower (in 2014).

According to recent studies a negative impact of the KhNPP techno-ecosystem on the background ponds was not observed, and their bio-funds currently do not impact significantly on hydrobiological state of the cooling pond. However, some species of algae of southern origin identified in fish ponds, have been found in the cooling pond as well, where they can produce significant biomass, causing the so-called "blooming" of water. On the contrary, there are some species in the taxonomic composition of aquatic background objects that are not found in the cooling pond . In the course of monitoring on the basis of bio-indication it was found unsatisfactory condition (in terms of biological contamination) of the pitch involved in the production cycle of the KhNPP, which is the water source important consumer of group A, and for chemical water treatment.

Experience of monitoring revealed that specific tasks should be constantly adjusted in the course of monitoring, i.e., monitoring system should have elements of self-organization, and it shall be possible to make additions and changes. Thus, the emergence of invasive species can significantly change the priorities in the choice of objects of observation. Spontaneous invasion of Dreissena into the KhNPP cooling pond and significant biological noise that it caused, immediately refocused interest in the objects of observation. Control of population dynamics of *Dreissena polymorpha*, and then *D. bugensis* for several years became the main objects of monitoring.

The results obtained in the course of monitoring should be valid and reliable, because on their basis can responsible decisions are made. Typically, hydro-biological monitoring is carried out using conventional methods in Hydrobiology [8]. When monitoring the development of communities of aquatic organisms on solid underwater substrates, including hydro-installations, effective and promising are diving research methods [8]. They make it possible already at the first stages to visually select specific communities. We have developed a technique of diving study of periphyton, which was successfully used in different water bodies. A diver-researcher can rather accurately estimate the distribution of even small organisms, the degree of coverage of the substrate by attached forms, spatial complexity of communities, identify dominant forms, especially those that define the character of the community. A lot of hydrobiological information can be obtained by carrying out at technical diving works by personnel of NPP diving stations.

In this regard, attention should be paid to the need for close contact between divisions, conducting environmental monitoring and production units of the NPP. All information about biotic factors causing interference to equipment operation, should enter the hydrobiological monitoring database by a single scheme (system). The experience of our work has shown that such an organized system of accumulating data in the database storage is virtually absent.

There is a group of methods that are specific for studying communities and fouling organisms. These are the methods of experimental substrates (ES), which provide data on the composition, abundance of periphyton beyond the water supply systems in case of failure to obtain material directly in the technical systems. Thus we can perform tracking of the development of organisms that cause bio-interferences, under model conditions. To better simulate the conditions which in their general features are similar to those in water supply systems, special facilities can be created, with the same water as in the water supply system. Unlike the operating water supply systems such simulators are always available for monitoring. Their design should provide fast and reliable inspection of samples, and sampling for quantitative and qualitative analyzes. Such monitoring units are used in the power plants abroad [9].

Techno-ecosystem of each NPP is characterized by its own unique conditions, the plants have different types of cooling ponds and their morphometric characteristics, water exchange, thermal, hydro-chemical and gas regimes, the distribution of bottom sediments, etc. Three NPP in Ukraine (Khmelnitsky, Zaporizhzhya, and South-Ukrainian) use cooling ponds that are significantly differ in their parameters. On Rivne NPP for additional feed and partial discharge of heated water of the river Styr water is used. In this connection, it seems impractical to assess the state of ecosystems of various NPP using one template parameter. The WFD proposes approaches to determination of the ecological status of a water body on the basis of the assessment of changes in the ecosystem of the water body with respect to some real or conditional reference state. The concept of "good ecological status" is usually associated with the "completely or almost completely undisturbed conditions". Thus, the reference condition of the water body is the condition, which existed prior to the intervention of anthropogenic factors to this ecosystem. However, if we are dealing with an artificially created water bodies, it is not possible to select a state, which would correspond to the reference environmental conditions,. So the WSD proposes to use the concept of "ecological potential" for essentially anthropogenically modified or artificial water bodies. That is, for each techno-ecosystem the individual ecological potential should be developed – acceptable ecological state (in this case - techno-ecosystem of the NPP), in which it would be: 1) no or minimal negative impact of techno-ecosystem on the environment, and 2) it would have no negative effects of biotic and complex ecosystem factors on operation mode of NPP equipment.

Based on considerable experience of research in the cooling ponds a methodology was proposed of expert assessments, which includes four groups of factors - changes in the hydrochemical regime, hydrodynamics, general engineering and hydrothermal effects. For two NPP in Ukraine it was shown that the most ecologically dangerous factors are in the "hydrothermal" group [10]. Expert assessments should be used both for rapid assessments and for estimates of generalized influence of anthropogenic factors.

The results of many years of observations of the state of techno-ecosystem provide an opportunity to make recommendations, to give forward-looking statements regarding the development of ecosystems (succession). For example, during the preparation of materials for the "Environmental impact assessment ..." (EIA) "Feasibility Study of the construction of Khmelnitsky NPP unit 2" it has been predicted the emergence in the cooling pond of *Dreissena polymorpha* and the likelihood of interference caused by this mollusk. Further monitoring confirmed the forecast. The appeared biological disturbances were the consequence of the lack of monitoring observations in 2002-2004.

Conclusion. One of the important measures for implementation of the provisions of the EU Water Framework Directive is creation of hydro-biological monitoring system at NPP in Ukraine, which should significantly complement the existing environmental monitoring system.

Many years of experience in hydro-biological monitoring leads to the following conclusions:

- hydrobiological monitoring of techno-ecosystems, in contrast to the natural ecosystem monitoring can detect not only the nature and extent of human impact on hydroecosystems, but also the influence of biotic factors on the functioning of technical objects;
- assessment methodology used in the WFD is based on the comparative approach, so for assessment of the real state it is necessary to choose "reference" comparison of estimates; for techno-ecosystem it could be environmental potentials, or complex ecosystem characteristics that can be considered acceptable both with respect to the environmental criteria and in terms of safe and efficient operation of technical systems;
- available experience of long-term monitoring should be used at organization of an effective environmental monitoring at NPP. In particular, the formation of databases should be based not only on the data obtained from

monitoring of cooling ponds, technical reservoirs, and the NPP water supply systems, which requires the organization of a unified data accumulation program on the biological interference in the operation of the equipment.

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EVALUATION OF EQUIVALENT DOSES OF THE COSMIC RAY COMPONENTS AND DENSITIES OF THEIR FLUXES IN KYIV

By measurements in Kyiv (Ukraine) using NaI(Tl)-detector (∅20×12 cm) it has been determined the ionizing flux of the cosmic rays of (0.022±0.002) particles /(cm^2·s) and their annual absorbed dose in the air of (232±14) μSv/y. Using the detector system with two NaI(Tl) scintillators (∅7.3×7.3 cm) and a summing block of the pulse-height signals of detectors it has been evaluated the maximum muon energy as 400 GeV. With the use of three various neutron detectors it has been determined the neutron flux of the cosmic rays of (0.015±0.002) neutrons/ (cm^2·s) and the annual ambient dose of 39 μSv/y.

Key words: cosmic radiation, equivalent dose rate, neutron flux density

In our previous work [1] we studied the response spectra of high-energy heavy charged particles (HCP) of secondary cosmic radiation (SCR) and accompanying γ-quanta on NaI (Tl) detector ((∅ 20 × 12) cm, which was placed under reinforced concrete floor of a multistory building. To identify cosmic particles the measurements were conducted with screens of different thickness. There were γ-quanta with energy ~70 MeV from decay of π^0-mesons, which were probably formed near the detector, because those photons could not penetrate several meters of concrete. This indicates that the flow of HCP must contain $\pi^{\pm}$-mesons. We also calculated the energy losses and tracks of μ- and π-mesons. As it turned out, these values are slightly different for both types of HCP of the same energy as in [1] so a generic term "mesons" was used. In this article we will use their contemporary names - muons and pions.

As a follow-up and in development of [1] this work includes the following objectives: 1) to make measurements with NaI (Tl) detector (∅ 20 × 12 cm) outdoors and clarify the contribution of HCP (>10 MeV) in the equivalent dose rate (EDR) for air ; 2) experimentally confirm or disprove indirect conclusion of [1] on the existence of particles with energies >200 GeV in the flow of cosmic muons and pions; 3) to determine the neutron flux density of HCP and evaluate annual equivalent dose for Kyiv.

Equivalent dose rate (EDR) measurements of high-energy heavy charged particles (HCP)-component of cosmic radiation were carried out by detector NaI (Tl) ∅ 20×12 cm size in the open air without protection (Fig. 1, 1) and with flat-screen lead on the detector 5 cm thick and the size of 35×35 cm (Figure 1, 2), whose presence leads to full absorption of electrons with energies <500 MeV and gamma rays with energies >20 MeV (only about 4% of gamma quanta with energies of 10 MeV enters the detector). *Spectra* 1 and 2 are similar; peaks in the range of 70÷80 MeV are caused by geometrical dimensions of the detector. Reduction of the number of counts of the peak of *spectrum 2* as compared to the *spectrum 1* corresponds to the absorption of gamma rays from the decay of π^0-mesons. The same features of response spectra were observed in [1] using a thin lead screens, which slightly reduce the energy of HCP. Such methods can be used to separate responses of HCP of gamma rays and electrons. The essential difference between the response spectra is caused by a channel width of the energy scale, which for Fig. 1 is ~2 MeV /channel, and [1] - ~1 MeV /channel.

The authors [2] postulate that the *spectrum 2* is formed by muons and the difference between the *spectra* 1 and 2 corresponds to the total contribution of gamma rays and electrons. We believe that this interpretation is incorrect. According to the calculation on the formula (2), in [1] a lead screen of 5 cm thick totally absorbs muons with energies <94 MeV, except HCP that permeate through the screen and the detector. Therefore after the lead screen the particles, which completed its run in the detector (*spectrum* 1) should not appear on the 10÷100 MeV part of the *spectrum2*. This area can enter HCP with higher energies (100÷200 MeV range of *spectrum* 1), which lost their energy in the screen down to 100 MeV. However, changing of responses in the range 10÷100 MeV of *spectrum* 2 can not completely fill the range lower than <100 MeV. This means that most part of HCP as well as gamma rays and electrons penetrate the detector under the angles >50° from the vertical axis (go beyond the lead screen)

Calculation of absorbed dose rate by detector NaI (Tl) for muons, pions, electrons and gamma rays and its recalculation into EDR in air is rather easy with the use of formula (4) - (6) [1], if there is a dominance of HCP flow over the flows of electrons and gamma rays.

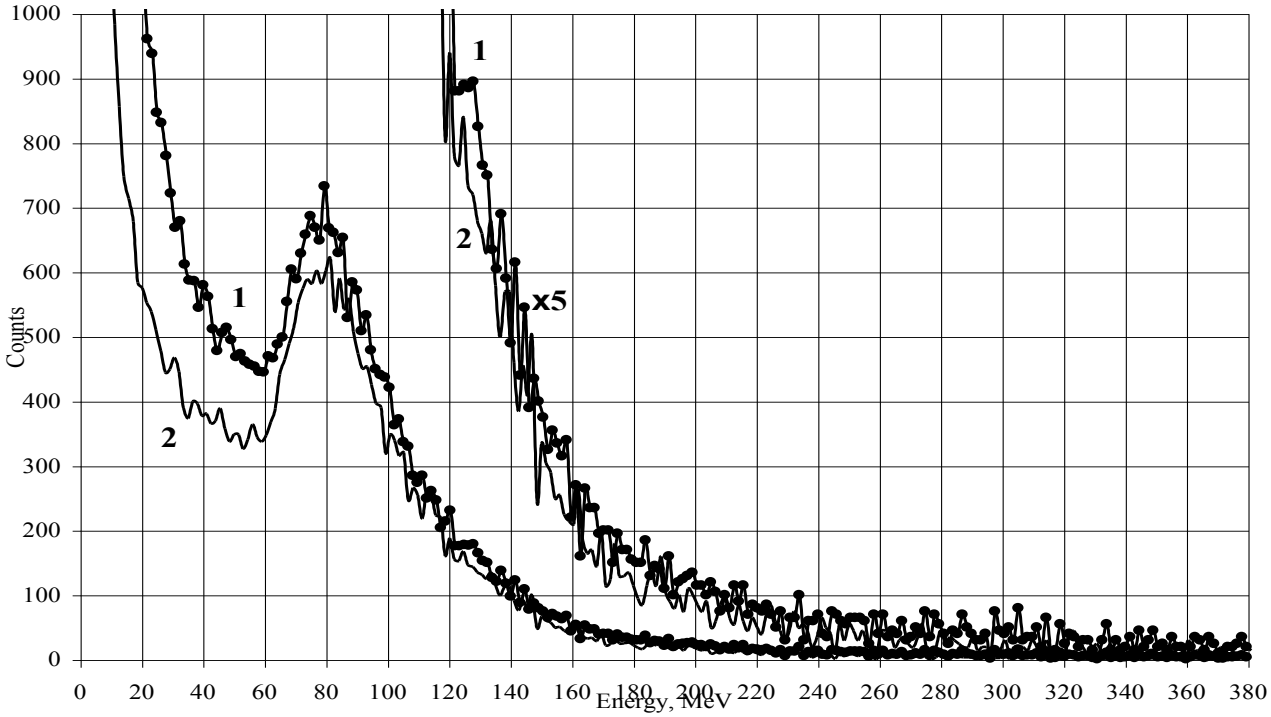

Fig. 1. Response spectra of NaI (Tl) detector for cosmic rays in the open air without protection (1) and with screen of Pb 5cm (2). Exposure time - 2 hours

This approach is not entirely true for gamma rays, because it does not takes into account the cross sections of their interaction with the environment. However, since the flow of gamma-quanta is insignificant, their contribution to $EDR_{surface}$ ($ПЕД_{пов}$) roughly corresponds to the contribution of the electrons at the registration of a photon

The contribution of secondary cosmic rays in the $ПЕД_{пов}$ is taken into account if we know their flow and power. With a thin lead screens we can evaluate these parameters and, as a consequence, to find energy dependence $K_{пов/NaI}$ both for electrons and for HCP. But there are situations where the muon disintegrates in the detector with emission of electron, which energy is unknown, but it is registered as an additive to response energy of a muon. That is, in the formula (3) from [1] the specific response energy may correspond to different HCP energies and therefore different $K_{пов/NaI}$, so the calculated value $ПЕД_{пов}$ is an approximate estimate of the dose rate in the open air, which for the energy range 10-380 MeV is (232±14) mSv/year. This value is less than the stated in [2] $ПЕД_{пов}$=(274±5) mSv/year because of different power band and methods of calculation.

At the end of this chapter we ask a reader to pay attention to some irregularities in the spectrum of 90-150 MeV (Fig. 1). If we calculate the impact of a flat screen lead in this range, the effective passage of HCP through the screen changes from $T_{кр}^{erc}$ =0,90±0,03 at the edges of that region to $T_{рез}^{erc}$ = 0,69±0,03 in its center (120-130 MeV). One can explain this deviation by insufficient statistics of measurements (although 7 statistical deviations between $T_{кр}^{erc}$ and $T_{рез}^{erc}$ demonstrate the reliability of values), or by the fact that response in the center of the interval 120-130 MeV correspond to such HCP energies when interaction with nuclei of lead is enhanced (resonance phenomenon). In this experiment the runs of high HCP were not defined: they ranges from 12 cm to 23 cm, corresponding to input kinetic energy of 40 GeV and 1 GeV. The resonant structure of the interaction is observed in reactions of $\pi^{\pm}$-mesons with protons in the nuclei of lead at input energies ~1,4 GeV with a cross section of 15 barns, and they pass through the 5-centimeter screen is $T_{рез}^{\pi}$ = 0.085 [3]. Outside this resonance (for the energy up to several tens of GeV) the cross-section falls to 1.8 bars, and the passage of pions $T_{кр}^{\pi}$ = 0.744.

It's easy to show that $T_{кр}^{erc}$, $T_{рез}^{erc}$ and the calculated $T_{кр}^{\pi}$, $T_{рез}^{\pi}$ are related as

$$\frac{T_{рез}^{erc}}{T_{кр}^{erc}} = \frac{T_{рез}^{\pi} + \frac{N_{\mu}}{N_{\pi}} T^{\mu}}{T_{кр}^{\pi} + \frac{N_{\mu}}{N_{\pi}} T^{\mu}}, \qquad (1)$$

where $T_{рез}^{erc}, T_{кр}^{erc}$ — pass of pions flow N_π through the lead screen in resonance area and at its edges, respectively; T^{μ}=0.95 — pass of muons flow N_μ through the lead screen in response area 90—150 MeV.

In the pion resonance area with the responses of 120-130 MeV (1) we get the flow ratio of muons and pions N_μ/N_π=2.19, and $N_\pi/(N_\mu+N_\pi)$=0.17. That is, in the flow of HCP pions have the contribution of (15-20)% in this region of responses.

Confirmation of the existence of high HCP in the cosmic radiation requires a separate experiment. If we create a detector system with two detectors ∅7,3×7,3 cm, located in alignment one above another we will be able to register by both detectors the losses of high HCP, the trajectory of which runs parallel or at a slight angle to the axis of the detector system. These responses coincide in time and, if they pass through adder, they give the signal amplitude approximately twice as much as the signal from one detector. This means that the detection system provides direction of inflow of HCP with the run of about 16 cm.

The authors have decided to use a simple circuit of adding the response amplitudes of two detectors, which occurs at the joint resistance (load) for amplifying the current of these detectors. But this scheme needs to prove its linearity, for which the spectra of gamma 137Cs and 60Co are used for one or two detectors (Fig. 2). The areas under the peaks of 137Cs (0.662 MeV), 60Co-I (1.173 MeV) 60Co-II (1.332 MeV) are about twice as big for the two detectors (Fig. 2, *2*), than for the one (Fig. 2, *1*). For one detector appearing of a summary peak of ^{60}Co Σ (2.505 MeV) is due to the registration of two cascade gamma quanta of ^{60}Ni after β-decay of ^{60}Co with the difference in escape angles close to zero. The coincidence in time of gamma quanta of ^{60}Co I in the first detector with ^{60}Co II in the second one, or vice versa, for the two detectors with the adder corresponds to the difference in escape angles close to 180°. To test the linearity of the adder the amplification was increased, reaching the amplitude ^{60}Co Σ of 8 V; with the energy linearity for the four peaks preserved.

A particular input of the adder is the area ^{60}Co Σ: for two detectors it is about 4 times greater (10500 counts) than the area of one detector (2350 counts). The 60Co nuclei have isotropic orientation in space and nuclei ^{60}Ni after β-decay also have isotropic distribution.

Both cascade gamma quanta are electric E2-transitions from levels that have spins I=4 and I=2, and the probability of their escape at a certain angle is described by deformed torus. Increased registration of ^{60}Co Σ in 4 times in the case of the pulse adder means that the spatial distribution of the escape of two gamma rays are identical, and the spin orientation of the lower level (I=2) is close to the spin orientation of the upper level (I=4).

So, the basic requirements for an adder are satisfied: reliable registration of the resulting signal at the coincidence in response time of two detectors; power linearity of calibration; the same total resolution of the summarized signal as for one detector.

To register the response spectra of HCP the 16 times less amplification was chosen than for the calibration of the adder. Two spectra measurements were conducted: *spectrum 2* – for two detectors (Fig. 3, *2*); and *spectrum 1* - for the lower detector (Fig. 3, 1) with a doubling of the number of responses.

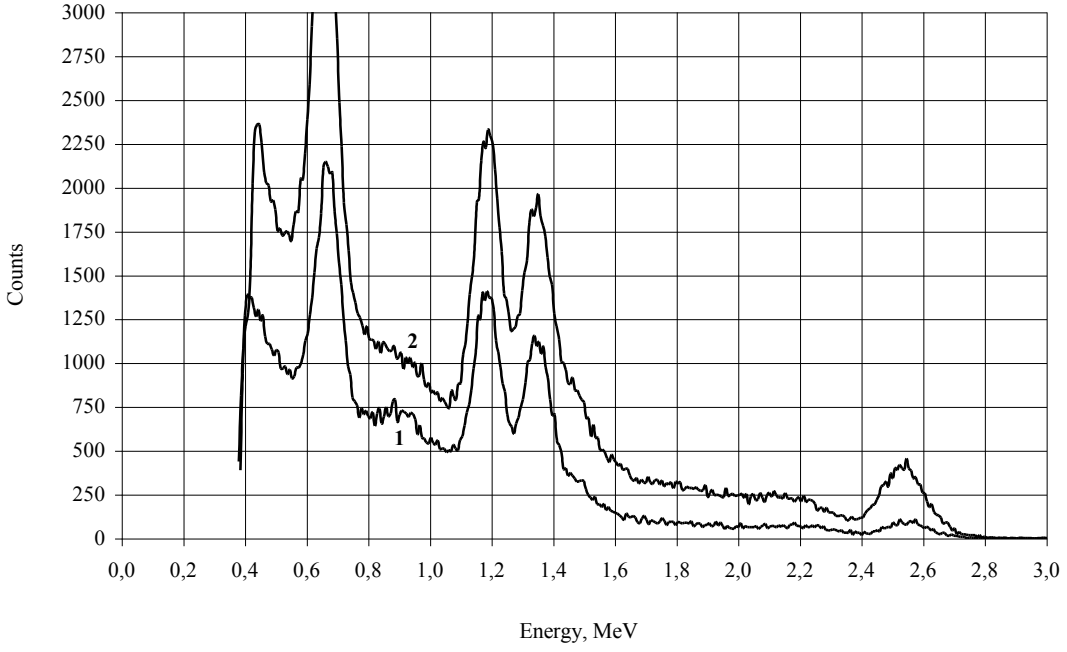

Fig. 2. Gamma spectrum of ^{137}Cs and ^{60}Co from one (1) and two (2) NaI (Tl) detectors of ∅7.3×7.3 cm for energy calibration of the adder. The exposure time - 400 seconds

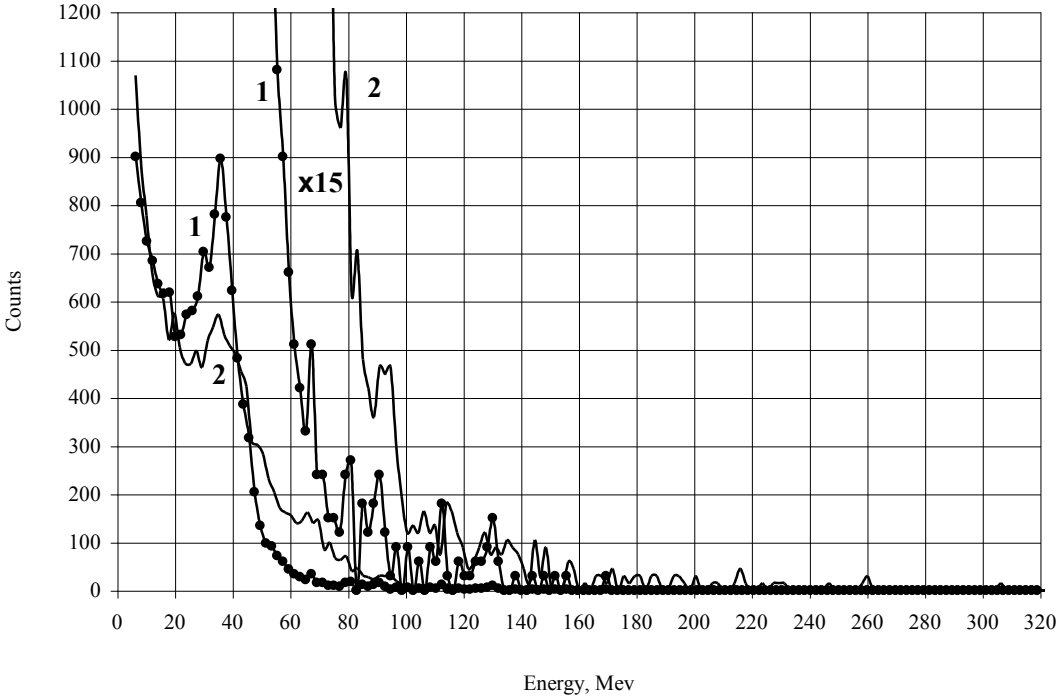

Fig. 3. Response spectra from the adder for one (1) and two (2) NaI (Tl) detector ∅7.3×7.3 cm. The exposure time - 2 hours

Let's discuss the features of these spectra. Reduction of the number of responses of *spectrum 2* in the range of 20-40 MeV can be explained by the registration of the same particle by both detectors and transfer of the total response in the region >50 MeV.

For the full energy range 6÷320 MeV the ratio of sum counts of *spectrum 2* to the sum of *spectrum 1* is 1,07±0.02, which may be the result of doubling of the number of responses from one detector instead of adding from the both ones. Most likely, this ratio should be <1 because in the *spectrum 2* at coincidence in time of pulses from two detectors we recorded one signal and, in addition, because of slight shielding of the lower detector by the upper one.

For the single-detector system there was not observed any pulses >200 MeV with an exposure time of 10 hours, and for two-detector system >320 MeV, although the adder allows recording responses up to 400 MeV. The most probable explanation is as follows: there is simultaneous registration of two events for a single particle, for example, in the first detector it loose energy of 120 MeV, and in the second - after an indefinite run there is a collision with the nucleus and its collapse ("star") with a total response of 200 MeV. Energy losses of particles can be measured by their run (8 cm for the first detector) using the equation (3) from [1]; they are 4.09 MeV·cm^2/g which corresponds to input kinetic energy of 400 GeV (Table 1).

Energy losses of HCP with kinetic energies of 150-350 MeV are as minimal as 1.28 MeV·cm^2/g. The interval of responses of 60÷70 MeV (Table 1) gives estimate energy losses less than the minimum - this means that the total run of a particle in the double-detector system is less than 16 cm, for example, at the side entering. The last response interval of 200÷320 MeV is probably created by "stars".

The authors also evaluated density of HCP flows for the registration plane of the double-detector system as 41.85 cm^2 (Table 1).

Table 1. Calculated kinetic energy and flux density of HCP with the trajectories close to the axis of the detector system

Power range MeV	Difference of *spectrum 2* and *1*, counts	Energy losses of HCP $dE/d(\rho x)$*, MeV·cm^2/g	HCP Energy *E*, GeV	HCP flux density, particles/(cm^2·s)
60—70	610	1.11	0.2	$20.2\cdot10^{-4}$
70—80	404	1.28	0.3	$13.4\cdot10^{-4}$
80—100	273	1.53	1.2	$9.1\cdot10^{-4}$
100—150	100	2.13	13.0	$3.3\cdot10^{-4}$
150—200	31	2.98	130	$1.0\cdot10^{-4}$
200—320	19	4.09**	400**	$0.6\cdot10^{-4}$
60—320	**1437**	—	—	**0.0047**

* — calculated for the middle of the selected range;
** — calculated for the maximum responses of *spectrum 1* and *2*

It is interesting to separate a part of the high-energy HCP with the trajectories close to the axis of the detector system in the full flux density registered by detectors NaI(Tl) ∅20×12 cm (spectrum 1 (Fig. 1), the total number of counts - 51135) and NaI(Tl) ∅7.3×7.3 cm (spectrum 1 (Fig. 3), the total number of counts - 7245). If detectors are considered as sphere of transverse areas 300 cm^2 and 51.4 cm^2, the flux density values are 0.024 particles/(cm^2·s) and 0.020 particles/(cm^2·s), respectively. Thus, high-energy HCP of energies 0.2÷400 GeV, which were selected by the double-detector system with in vertical orientation of the axis is approximately 1/5 of the total density of particles with energies >10 MeV (0.022±0.002) particles/(cm^2·s))

Determination of neutron flux density of secondary cosmic radiation requires consideration of effects of gamma and neutron backgrounds from spontaneous fission of natural uranium and thorium isotopes that are present in the soil or building materials (depending on the location of measurement).

To register neutrons the counters were used based on reactions of slow neutrons capture with escape of fast charged particles:

Detector NRD (USA): plastic ball ∅ 22.9 cm with BF_3-counter of neutrons;

Detector ND-5, with volume of the moderator ~5 dm3 in the form of a cylinder ∅ 16 cm and 21 cm height, filled with paraffin, ^{3}He neutron counter SNM-17;

Detector ND-10: with volume of the moderator ~10 dm3 in the form of a cylinder ∅ 16 cm and 53 cm height, filled with paraffin, ^{3}He neutron counter SNM-37

These counters record the gamma quanta at rather low thresholds (column 2 Table 2). For reliable identification of neutrons it is necessary to set a higher amplitude threshold of registering. But high-energy photons yet produce in a cathode material the electrons, which energy loss in the gaseous environment can

exceed this threshold registration. These signals are registered as neutrons, and their number N_γ (Table 2) depends on the geometrical dimensions of the counter.

Table 2. Evaluation of background radiation at registration of neutrons

Detector	**γ- background, pulses/min**	N_γ**, pulses/min**	N_{SF}**, pulses/min**
NRD (USA)	1	0.026	0.00002
ND-5	2	0.025	0.001
ND-10	3	0.057	0.001

In addition, it is necessary to evaluate the contribution of spontaneous fission neutrons N_{SF} of natural isotopes, among which the most intense source is uranium-238. To calculate the share of background the dose rate of background radiation of 0.125 μGr/h was used, which corresponded to the conditions of measurements. Then the dose rate of gamma decay chain of uranium-238 is 0.050 μGr/h [4] and the expected dose rate of photon spontaneous fission - $0{,}2\cdot10^{-6}$ μGr/h (it is taken into account that instant gamma-quanta are 8 times more intense than the chain-fission ones). Given that the latter value is equal to the kinetic energy of two spontaneous fission neutrons, and based on the assertion that the flow of neutrons of 1 n°/(cm^2·s) creates the dose rate of 0.0493 μGr/h [4], we can estimate the density of spontaneous fission neutrons as $4\cdot10^{-6}$ n°/(cm^2·s). Taking into account the cross-sectional area of the material of moderator of the neutron detector, and neutron registration efficiency (Table 3), we obtain N_{SF} evaluation (Table 2).

The calculation results indicate the following: at low natural γ-background you cannot ignore N_γ and N_{SF}. But during measurements you have to exclude the presence of equipment, which contains uranium-238 and other fission isotopes.

Measurements of neutrons of secondary cosmic radiation (SCR) N_n were conducted by the aforementioned detectors several times, with subsequent averaging (Table 3). When calculating the density of neutron flux it was taken into account the effectiveness of the registration of each detector and its error.

Weighted average neutron flux density of secondary cosmic radiation from three detectors is (0.015±0,002) n°/(cm^2·s). Equivalent dose for this flux at radiation weighting factor Q = 6 [4] is estimated at 39 μSv/year. In [2] the energy of cosmic neutrons was estimated using a set of different-sized neutron moderators, and as a result the neutrons of three groups were identified: heat – with energy up to ~ 0.4 eV (Q=5), the second - with energies 2÷3 MeV (Q=20), and the third most intense ones – in the range of ~ 100 MeV (Q=5). This gives reason to expect that the neutron EDR will be twice higher than the specified in this article.

Table 3. The neutron component of secondary cosmic radiation (SCR)

Detector	The effectiveness of the registration of neutrons, %	N_n, pulses/10 min	Flux density n°/(cm^2·s)
NRD (USA)	0.023±0.006	1.2±0,2	0.0220±0.0110
ND-5	2.10±0.31	33.5±2,9	0.0136±0.0023
ND-10	2.20±0.29	43.5±3,3	0.0168±0.0025

Conclusions

This work completed our study of the cosmic background; it that might not be initiated if any reliable publications would be currently available for Kyiv (~100 m above the sea level).

With the use of NaI(Tl) ∅ 20×12 detector the authors investigated response spectra of gamma quanta and SCR particles with energies of ~10 MeV to 380 MeV, which include electrons, muons and pions. The total density of these particles is (0.022±0.002) particles/(cm^2·s) corresponding the EDR in air (232±14) μSv/year, which is reduced by 40-50% in the measurements on the lower floors of a multistory building. In some energy response intervals we observed different composition of particles:

10÷50 MeV – electron, and possibly gamma quanta contribution, does not exceed 20% of the total flow of high-energy heavy charged particles (HCP) in this range;

70÷90 MeV – there are γ-quanta from the decay of π^0-mesons in the amount of ~15%;

120÷130 MeV - the authors argue observation of $\pi^\pm$-mesons in the amount of 15-20%.

Using the double-detector system of NaI(Tl) ∅7,3×7,3 cm with the adder, the authors have found the existence of HCP with energies ~ 400 GeV and evaluated the density of the measured flux in the range 0.2÷400 GeV at 0.005 particles / (cm^2·s) in a vertical orientation of detector system axis.

With three neutron detectors we determined the flux density of secondary space neutrons for the city of Kyiv

(~100 m above the sea level) as 0.015 ± 0.002 n°/(cm^2·s), which creates EDR of 39 mSv/year.

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CURRENT STATUS OF OVERCOMING THE CHERNOBYL NPP ACCIDENT CONSEQUENCES

This article analyzes the consequences of the accident at the Chernobyl nuclear power plant and characteristics of zones of radioactive contamination, evaluates distribution of radionuclides in different objects of the Exclusion zone. The article also outlines the actions to be implemented in 2015-2019 in accordance with the National Program of Decommissioning of Chernobyl Nuclear Power Plant and Transformation of the Chernobyl Shelter into an Ecologically Safe System.

Key words: Chernobyl accident, nuclear power plant, radionuclide contamination, radioactive isotopes, the Chernobyl Shelter.

On April 26, 1986 the accident occurred at the fourth unit of the Chernobyl nuclear power plant (NPP). This accident is estimated by experts as the greatest one in the history of nuclear power. The adverse effects of that man-made disaster is caused by the release of significant amounts of radionuclides in the environment, continued existence of the destroyed nuclear reactor (now - the Shelter), and with the Chernobyl NPP decommissioning.

The accident resulted in contamination of over 145 000 km2 territory of Ukraine, Belarus, the Russian Federation, with density of contamination by radionuclide 137Cs that exceeds 37 kBq/m2 (exceeding of this target level means that the specified area should be referred to as the contaminated one according to the legislation of these countries). The Chernobyl disaster affected 5 million people, and at the contaminated areas there were almost 5000 settlements of Belarus, Ukraine and Russia. Of these, in Ukraine - 2293 villages, towns and cities in which the population in the early 90th of the 20th century exceeded 2.6 million people [1].

In addition to Ukraine, Belarus and the Russian Federation, substantial contamination from the Chernobyl disaster experienced Sweden, Austria, Norway, Germany, Finland, Greece, Romania, Slovenia, and Switzerland.

Table 1. Comparative data on contamination of territories of Slavic republics after the Chernobyl accident [2]

Contamination level, Ci/km^2	1-5	5-15	15-40	Above 40
Total in the USSR	103200	17610	7160	3100
Belarus	29920	10170	4210	2150
Russia	39280	5450	2130	310
Ukraine	34000	1990	820	640

Other European countries were contaminated to a lesser extent. The radioactive fallout from Chernobyl NPP were reported by national departments of different countries that conducted radiation monitoring at the territory of Asia, North America, the waters of Atlantic and Pacific Oceans in the northern hemisphere.

Immediately after the accident, significant efforts have been focused on assessment of radioactive contamination and monitoring of radionuclides in products produced in the contaminated and adjacent territories. An extensive network of monitoring stations was created with hundreds of laboratories and radiological control posts. Maps of radioactive contamination were drawn as for Ukraine and for other regions of the Soviet Union on the basis of external radiation doses capacity measurements, and analysis of samples of the environment, as well as through gamma dose measurements from aircrafts.

Employees of the department of radioelement analysis of the Institute for Nuclear Research of Ukraine (one of them is the author of this article) during the years 1986-1988 on the basis of researches under the “Reper” program of soil samples from different regions of Ukraine and the Exclusion zone drawn maps on contamination of different areas. At the end of 1989 to the list, which determines the place of anti-radiation measures were added around 1500 settlements of Rivne, Zhytomyr, Kyiv and Chernihiv regions [3].

Table 2. Characteristics of radioactively contaminated zones

The exclusion zone		The zone of unconditional resettlement		The zone of guaranteed resettlement		The zone of enhanced radiation monitoring	
Number of settlements	Area, km^2	Number of settlements	Area, km^2	Number of settlements	Area, km^2	Number of settlements	Area, km^2
76	2000	86	2200	841	23300	1290	27150

During 1991-1995 according to the current legislation the territory of radioactive contamination zones was determined. Those zones covered 2293 settlements of twelve regions of Ukraine that suffered the greatest contamination from the Chernobyl disaster.

The territories classified as radioactively contaminated zones are located in 74 districts of 12 regions (Vinnytsa, Volyn, Zhytomyr, Ivano-Frankivsk, Kyiv, Rivne, Sumy, Ternopil, Khmelnytsky, Cherkasy, Chernivtsi, Chernihiv).

Now contaminated areas are home to nearly 2.2 millions of people, including more than 1.6 millions of persons in the area of enhanced radiation monitoring

At present, background radiation compared with 1986 is hundreds times lower. The countermeasures undertaken and the processes of self-purification of the environment led to a reduction of radionuclides in the environment and in agricultural production. This, in turn, led to reduction of external and internal exposure of the population. Total activity of the released substances decreased by more than 200 times, and radioactivity that remained in the soil outside the ChNPP site is more than 85% represented by cesium-137, almost 10% - by strontium-90, and the rest part – by transuranic elements, mostly by plutonium-241. It should be noted that the spatial distribution on the territory of Ukraine of strontium-90 and plutonium isotopes differs significantly from the distribution of cesium-137, because more than 60% of emitted from the reactor hard volatile elements (isotopes of transuranic elements) and strontium-90 remained inside the exclusion zone.

Since after the accident large areas were contaminated not only by short-lived radioactive isotopes, but the isotopes that have a significant half-life, the problems related to the accident will exist for many years.

The greatest danger now (and in the next decade) is from isotopes of strontium and cesium with a half life of about 30 years. The largest concentrations of cesium-137 are found in the surface layer of soil, where it enters the plants and fungi. Radioactive isotopes of plutonium and americium will be contained in the soil for hundreds of years, but their amount is small. As well as the isotopes of strontium, they are in the top layer of soil. But the ratio of activity 241Am / 239 + 240Pu increasing every year. If in 1986 this ratio was 0.13 ± 0.03, then for the next 70 years this ratio will increase 20 times by the radioactive decay of 241Pu and accumulation of 241Am. Currently, the contribution of americium-241 in total α activity is 50%. By 2060 its contribution will amount to 66.8%. Americium has high toxicity. It should be noted that the annual permitted concentration (GDK - in Ukr.) for americium in air is about 1x10-4 Bq/l, and in water - about 70-80 Bq/l [4].

The results of radiation-ecological monitoring makes it possible to assess the radiation situation and the main components of the natural and industrial environment in the exclusion zone and published in the Bulletin of the ecological state of the exclusion zone and zone of unconditional (obligatory) resettlement, which is published twice a year and sent to the concerned ministries and departments.

The main amount of radionuclides is accumulated in the Chernobyl Shelter. According to experts, there are about 180t of nuclear fuel in different forms, which contain radioactive substances with the total activity of about 340 PBq. In addition to fuel mass the Shelter contains large amount of radioactive materials that consists of core remnants of the destroyed reactor, reactor graphite, and contaminated metal and building constructions of the power unit.

Evaluation of distribution of inventories of radionuclides in various sites of the exclusion zone is summarized in Table 3.

In the years that have passed since the accident, radiological situation has undergone some positive changes, but requires continuous monitoring of the environment and the implementation of a number of measures.

Verkhovna Rada of Ukraine adopted the “National Program of the Chernobyl NPP Decommissioning and transforming of the Shelter into an ecologically safe system” [6].

Table 3. Distribution of inventories of radionuclides in various sites of the exclusion zone [6]

Object	Activity, PBq (PBq = 10^{15}Bq)			
	Total	^{137}Cs	^{90}Sr	Transuranic elements
Territory of the exclusion zone	8,13	5,5	2,5	0,13
Cooling pond	0,22	0,19	0,03	0,002

Points of radioactive waste disposal	5,49	3,6	1,8	0,09
Point of temporary localization of radioactive waste	2,14	1,4	0,7	0,04
Total	16	10,7	5	0,26
The Shelter	340	190	145	4,5

This program determines the main directions of work on the Chernobyl NPP decommissioning and transformation of the Shelter into an ecologically safe system, the amount of funding, organizational and technical problems.

The on-site radwaste of Chornobyl nuclear installations and facilities to be decommissioned include:

- first, second and third units (main building);
- spent nuclear fuel storage facility based on the "wet storage" technology (SFSF-1);
- temporary storage of liquid and solid radioactive waste.

Other objects of general purpose, auxiliary, electrical, waterworks, water cooling facilities are also subject to decommissioning.

The industrial site includes is destroyed fourth power unit (the Shelter), which is a nuclear hazardous object, and radwaste interim storage facility. Activities carried out on the Shelter are aimed at its transformation into an environmentally safe system.

Completion of decommissioning of Chernobyl NPP and transformation of the Shelter into an ecologically safe system requires about 100 years, so the Program includes immediate measures to be implement during 2009-2012, and long-term measures for 30-50 years.

Since the Chernobyl accident a significant amount of work was performed to improve the radiation condition of the Exclusion Zone: An area of over 30 hectares was decontaminated, it was collected and disposed more than 923,000 m3 of radioactive waste decontaminated materials and equipment were disposed. Only in the years 2010-2014 it were processed and transferred for disposal of low- and medium level radioactive waste, including ChNPP-origin, totaling 130,183 m3 [5].

According to Article 6 of the Law of Ukraine "On legal regime of the area contaminated by the Chernobyl disaster" [7] funding of work on overcoming the Chernobyl accident consequences should be at the expense of the State Budget. Activities implemented in the exclusion zone, are funded by two budget programs:

KPKV[1] 2408110 "Support of the environmentally safe condition in the Exclusion Zone and the zone of unconditional (obligatory) resettlement";

KPKV 2408120 "Support in safe condition of power units and the Shelter, and measures to prepare Chernobyl NPP for decommissioning".

The last five years the annual budget funding was about 1 bln. UAH. Financing was carried out both from the general and from special fund of the State Budget.

In the years 2015-2019 the measures in the exclusion zone should be implemented for:

development of a long-term (five-year) program of works to minimize the impact of the exclusion zone and radiation and nuclear hazardous objects on the population and the environment;

development of a long-term (five-year) program of decommissioning of Chernobyl power units (in replacement of the program, which repealed);

zoning of the territory with mandatory implementation of necessary research on specific areas;

creation of a modern system of integrated environmental monitoring with its constant improvement;

inventory and study of the degree of environmental hazard of points of radioactive waste disposal, implementing of measures to improve stability of these points;

continuation of construction and commissioning of the production plants for radioactive waste management (the Vector Complex);

creating a repository for long-term storage of vitrified high-level waste from reprocessing nuclear fuel of domestic NPP, which come back from the Russian Federation;

completion of construction and commissioning of a centralized repository for long-term storage of spent sources of ionizing radiation;

completion of the new safe containment which will make it possible to transform the Chernobyl Shelter into an ecologically safe system, and of storage facility of ChNPP spent nuclear fuel (ISF-2). Its construction was started in 2012, projected completion and commissioning - 2017

implementation of measures of Chernobyl NPP units decommissioning and ensuring their safety;

[1] transliterated

development and implementation of scientifically based water bodies protection measures;

implementation of measures aimed at improving fire safety in forests, reforestation;

prevention of unauthorized activities in the exclusion zone, intrusion into its territory of unauthorized persons and vehicles; control of inbound / outbound transportation of radioactively contaminated materials;

ensuring the security and physical protection of facilities for radioactive waste and spent nuclear fuel;

complete draining of the cooling pond with subsequent disposal of radioactive sediments;

dismantling of equipment of the radar station Chernobyl-2;

development of a set of measures to involve the least contaminated (rehabilitated) exclusion zone lands into civil use for production purposes;

ordering of industrial and social infrastructure of the exclusion zone and maintaining it in good condition;

development of proposals on improvement of the legal framework regulating activities in the exclusion zone with consideration of peculiarities of its operation, including land issues, through amendments to the Land Code of Ukraine [8];

implementation of measures on social protection of employees in the exclusion zone and creation of conditions that ensure the attractiveness of work at the enterprises [5].

At the condition of consistent implementation of the above measures, expectations of overall reduction of surface and underground migration of radionuclides it is not expected deterioration of radioecological situation in the exclusion zone, and it is projected that impact on the population of Ukraine and other areas will be gradually reduced down to minimum.

However, for the full implementation of the National Program of the Chernobyl NPP Decommissioning and Transformation of the Shelter into an Ecologically Safe System [6] requires significant funds. World practice shows that in most cases the costs of decommissioning correspond with the cost for new construction.

With regard to the accidental fourth unit of the Chernobyl NPP, the total cost of transformation of the Shelter into an environmentally safe system currently cannot be accurately evaluated, since there are no analogues in the world and it can be tens of billions of dollars (or hundreds of billions UAH).

A significant contribution to CNPP units decommissioning and transformation of the Chernobyl Shelter into an ecologically safe system comes from international assistance, but the main burden falls on Ukraine.

Conclusions

In the Exclusion Zone it was held unprecedented (for the peacetime) evacuation of population, economic activity collapsed, industrial and agricultural enterprises closed. Verkhovna Rada of Ukraine adopted the National Program of the Chernobyl NPP decommissioning and transformation of the Chernobyl Shelter into an ecologically safe system [7].

Since the Chernobyl accident significant amount of work has been performed to improve the radiation conditions in the Exclusion zone. The works to overcome Chernobyl accident consequences are performed at the expense of the State Budget. However, there are several factors that complicate implementation of measures under the Program:

- discrepancy between the approved in funding approved in the State Budget and the minimum necessary one;
- the limits of budget funding do not take into account inflation, which we have in the country now;
- failure or lack of capital expenditures does not allow of new equipment, and new technologies.

Nevertheless, there is the continuing implementation of measures of Chernobyl NPP decommissioning and minimization of environmental hazard in the Exclusion zone. In addition, the Ministry of Environment and Natural Resources of Ukraine continues to work towards a Chernobyl Biosphere Reserve with the total area of about 260 thousand hectares. Its creation will contribute to the development of science and disciplines related to the aftermath of the nuclear disaster.

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PROSPECTS AND PROBLEMS OF POSITIVE DISPLACEMENT PUMPS AND PISTON PUMPS APPLICATION AT NPP

The article provides analysis of the use of piston pumps at nuclear power plants. The main reasons of their use were discussed as well as conditions of their operation and the main associated problems.

Key words: nuclear power plant, cryogenic technique, pressure oscillations, piston pumps

Most of the NPP pumps, involved in the operational processes, are of blade-type, which are subdivided into centrifugal and axial ones. However, in some systems and processes the positive displacement pumps become widely implemented, which include gear, screw, reciprocating and rotary membrane. Distinctive features of these pumps is the high and ultra-high pressure (above 250 MPa) and high efficiency (Up to 95-99%) [1]. Only a low flow rate (30-50 l/s) limits their use as the main circulation pumps, which main task is to remove large thermal output, which requires intensive supply of coolant.

Despite this limitation, positive displacement pumps are used at the NPP for different purposes that do not require intensive supply of coolant, but working at high pressure. Below some techniques are presented suitable for the use of positive displacement pumps, as well as the problems of operation of these pumps.

Recently, at the Kola NPP the trial of a new treatment technology for decontamination of equipment was carried out, which applies high-speed jet of dry ice granules –a cryogenic blasting technique. Application of this technology has already increased significantly the efficiency of repair, ecology and productivity. Previously, the company applied traditional methods of metal decontamination by pneumatic and electric tools. However, it usually produces a lot of dust and airborne abrasive residues that negatively affect both the operation and maintenance personnel. The tests showed that the new method is qualitatively superior to the previously used technology. This is the friendliest way to clean metal by the known contaminants - from oil and grease - to paint and polymers. Cleaning with dry ice is environmentally safer process: it produces less dust, which reduces the negative impact on the maintenance personnel involved in such work. The positive properties of the cryogenic blasting are also the rejection of the use of water and solvents, as well as an opportunity to clean the easily damaged materials, and low labor costs [2].

Currently, specialized equipment HAMMELMANN (Germany) is used for cleaning the NPP equipment from sediments. Application of high-pressure water jet to clean these installations allows cleaning a wide range of equipment: from heat exchangers to oil tanks, from the scale and oil contamination. To perform these operations, high pressure piston pumps are used (from 40 MPa to 250 MPa with a water flow rate of 250 l/min). However, it is important to understand that the high speed of water can cause erosion damage of brass and copper tubes (Figure 1).

Fig. 1. Erosion of the HAMMELMANN working nozzle and condenser tubes after cleaning them from scale

Technologies of water-cutting of metals and other materials are being increasingly spread due to their effectiveness and quality of the cut. In contrast to plasma or oxygen cutting, cut parts made by water cutting do not require further processing of the cut area. This technology also requires high-pressure piston pumps

with pressure of more than 250 MPa [3]. Water is being formed into the jet flow with a speed of 500-800 m/s in the diamond nozzle, which at such speed of flow has about 1 month of operation life, and the speed of cutting the 50 mm thick steel is about 10 mm/s.

The use of cooling agents for dismantling the main NPP equipment [4] also requires the use of piston pumps. This is determined by the fact that efficiency of small centrifugal pumps is 60-70%, while the efficiency of piston pumps is more than 80% [1], which means that up to 30-40% of capacity of the centrifugal pump is transferred to heat, while for the piston pumps this parameter is less than 20%, which is extremely important when pumping cryogenic fluids. Thus, some technologies of NPP dismantling, requiring the use of liquid nitrogen, for example for gaining metal frost fracture and than its easier destruction by small impact force without dust, or more effective grinding of plastics, rubber, resin when cooled to the embrittlement temperature and subsequent compacting of such radioactive wastes into monolithic blocks require the use of piston pumps. Such cooling of the above materials is possible only at low temperatures, such as in a medium of gaseous or liquid nitrogen [5].

In addition to the above applications, the liquid nitrogen is applied in the power industry for removing insulation from wires and cables, and gaseous one - for controlling the pressure in the primary circuit via the pressure compensator, to displace hydrogen in power generators, as inert medium in the reactors of RBMK-type, and to create low-temperature superconductivity in reactors of Tokamak type. Thus, the cryogenic technologies converge more and closer to nuclear technology. Moreover, it is much more profitable to store and transport liquefied gases not under pressure in pressurized cylinders, but in the liquid state in Dewar vessels, and to use the positive displacement pumps. Currently, Thermal Power Plans (TPP) and NPPs apply such pumps in the oil facilities of steam turbines, and for injection of boric acid solution in the primary circuit.

The main problem of positive displacement pumps and, in particular, piston pumps is considerable vibration of the connected pipes, caused by strong pressure fluctuations inherent in these pumps. This problem is particularly acute for large diameter pipelines or flexible hoses, having a small flexural rigidity relationship to the mass per unit length of a pipe $(EI_x / \int A)$.

Another problem in the operation of such pumps is manifested in deterioration of the working conditions of the instrumentation which is responsible for accurate adjustment of the desired flow or pressure. This situation is caused by peculiarities of hydraulic pulsating flow in pipelines and in local resistances. In this case, the hydraulic resistance depends not only on the active part (A) of the total complex resistance of a pipeline $(Z = A + i\omega R)$, but also on its reactive component (R), which increases with the frequency of the oscillation process. This situation leads to large errors in the operation of measuring instruments calibrated in a stationary conditions and having a time constant greater than or equal to the period of oscillation of the hydraulic process. For example, at measurement of flow, by orifice plates, the readings of secondary devices are almost always underestimated that may cause some inconveniences such as overflow at filling of closed tanks. Using flow-meters, based on the principles of flow rotation in such cases does not allow exact measuring of flow.

When using turbine flow-meters, the frequency of the primary sensor signal is modulated by the vibration frequency of the hydraulic process, making it difficult to decipher the flowmeters signal.

As an example, during the accident at the Chernobyl NPP (1986.04.26), it was necessary to use liquid nitrogen to cool the reactor support. The liquid nitrogen was transported by five-ton refueling-thermoses, which were filled by three-line piston pumps. As a result of increased pressure oscillations the flowmeter readings were lower than actual, which lead to overflow and accidents on several tankers.

Thus, for implementation into power industry of larger positive displacement pumps and especially of piston pumps there is a need to develop effective means of reducing the amplitude of pressure fluctuations in the supply lines.

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EFFICIENCY AND TIME

The article presents a hypothesis of relation of the efficiency factor of energy conversion with time of conversion. It is shown that these parameters are related inversely. The experimental results and evaluations of performance of power equipment are presented, which confirm the suggested assumptions.

Key words: time, explosion, efficiency factor, turbine, energy, entropy

Those who wondered why the efficiency, or the output-input ratio, is calculated as the ratio of work or energy, while at selection of proper equipment we are guided by the capacity of an engine or a turbine. Indeed, the characteristic of "power" or "capacity" allows us to evaluate the extreme capabilities of a mechanism, and to draw a conclusion as to whether it is suitable or not for our goals. We never choose our engine for its "energy". We definitely need to know how much energy you can get out of it at a unit of time.

The efficiency we calculate as the ratio of mechanical work (power multiplied by time) to chemical or nuclear fuel energy. So the time is a latent parameter involved in calculating the efficiency of any engine that converts the energy of the substance into power received over a certain period of time that is required to make full use of the energy-intensive material.

In this case efficiency does not depend on the time since the same mechanical energy can be obtained by the product of a small capacity for a long time, or vice versa, high capacity for a short time.

Thus, it is possible to introduce another parameter closely related with both energy conversion process, and with efficiency factor, and it will be the time required for the energy conversion process from one type to another.

Consider several examples of processes with different energy conversion rate.

It is known that a perpetual motion machine must have efficiency equal to or close to 1 and from known natural processes such efficiency is the process of putrefaction, i.e., it is a process with the time of implementation that tends to infinity. Interestingly, for such processes the saying "More haste, less speed" or "long distance require steady pace" is quite true. We also intuitively aware that the quality of virtually any work depends on the time spent on it. The more carefully and painstaking is the work (procedure), the better is the output. If we replace the quality concept (marriage) with the concept of loss (i.e. defective goods), the level of quality loss can be quantitatively associated with time by presumably reverse function:

$$\phi = \frac{\alpha}{\tau} \qquad (1)$$

where α - loss intensity factor.

In turn, as it was noted above, to assess the effectiveness of technology processes we commonly use efficiency, defined as the ratio:

$$\eta = \frac{A_{П}}{A_{з}}, \qquad (2)$$

where A_n , $A_з$ – is useful work and expended work respectively.

Useful work can be presented through the performed work and loss, as

$$A_n = A_з - \Delta A_{nom} \qquad (3)$$

Than

$$\eta = \frac{A_{П}}{A_{з}} = 1 - \frac{\Delta A_{nom}}{A_{з}} = 1 - \phi \qquad (4)$$

However, we have seen above, that the quality of the work, or rather the loss of quality may be related with the time of work execution by the function (1). Substituting (1) into (4) we obtain:

$$\eta = 1 - \phi = 1 - \frac{\alpha}{\tau} \qquad (5)$$

I.e. if we take the equal quality of work, the efficiency, i.e. the efficiency of such processes should be inversely related to the time (Fig. 1).

These considerations can be confirmed by the estimations of characteristic times, typical for energy processes. The integrated evaluation will be aimed at confirmation of the relationship between the time of the process run and its efficiency.

As it was already noted, it is considered that the most energy efficient process is the process of decay, for which we can set the limits: at $\tau \to \infty \quad \eta \to 1$.

For all thermodynamic cycles of any power machines, the limiting efficiency according to the Carnot theorem, is determined by the ratio of the temperature of a source (T_1) and the temperature (T_2) of heat receiver [1]:

$$\eta = 1 - \frac{T_2}{T_1} \qquad (6)$$

This, in particular, explains higher efficiency factor of the turbines of thermal power plants (TPP) with initial steam temperature 540 - 560°C, η= 0.43 ÷ 0.45, while for the turbines of nuclear power plants (NPP), with initial steam temperature of about 300°C, the efficiency is about 0.32 ÷ 0.34 [2]. However, considering the same circumferential velocity of both types of turbines, defined by the frequency in the grid (50 Hz), and hence very close vapor axial velocity, the time of passing an TPP steam turbine should be greater than the time of passage of steam through an NPP turbine, since the length of turbines flowing part are respectively 4.7 m and 3.2 m. Taking the axial steam turbine velocity equal to 100 m/s we receive time of the energy conversion process correspondingly from 0.047 to 0.032 sec.

The efficiency of rocket engines is significantly lower - 0.10 ÷ 0.15, and energy conversion process is much faster (it is determined by the length of a nozzle and the speed of ejection of the combustion products).

Faster processes, e.g. the processes of the explosion can be evaluated by [3], where explosions of explosive mixtures in small volumes were studied.

The results of calculation of the energy for the reaction, converted to the amount of the experimental cell, as well as the results of the experimental determination of the explosive energy are shown in Table 1. Here, the efficiency of the explosion is calculated by the ratio between the theoretically possible energy of the reaction, and the real value of the explosion energy:

$$\eta = \frac{Q_T}{Q_Э} \qquad (7)$$

As one can see from Table 1, the energy of the explosion is a small fraction of the energy of a chemical reaction. Most of the energy is converted into heat, and the remainder goes to the pressure increase, i.e. into useful work of lifting a piston. Also we can see the dependence of explosion efficiency on the time of its occurrence.
Taking the detonation rate of explosive mixture of about 5000 m/s, the cell sectional area of 0.00724 m^2 and cells height $h = \frac{V}{0{,}00724}$, blast time will be equal $\tau = \frac{\sqrt{(h^2 + r^2)}}{5000}$ (see. Table. 1).

Table 1. Comparison of theoretical reaction energy and experimental energy of the explosion

Cell volume V, m^3	Chemical reaction energy $Q_т$, J	Experimental energy of explosion $Q_э$, J	The efficiency of explosion,, %	Detonation Time of explosive mixture τ, s
0,00006	652,7	6,5	0,996	0,0000097
0,00008	870,2	10,8	1,24	0,0000099

0,000176	1915	88,5	4,62	0,0000108
0,000186	2024	110,4	5,45	0,0000109
0,000204	2220	158,7	7,15	0,0000111
0,000214	2328	224,1	9,63	0,0000113

For clarity of presentation, let's transfer data to a generalized diagram (Fig. 1, 2). The figure shows the trend - the inverse relationship between the time of the energy conversion process and its efficiency.

It is interesting to note that Prigozhin in his article [4] makes an attempt to link the processes of entropy over time, and this dependence is also inversely proportional to the time, i.e. with increasing of time, the entropy function falls. This position is also confirmed by thermal processes - where model processes have minimum increase of entropy, in contrast to actual processes with small efficiency.

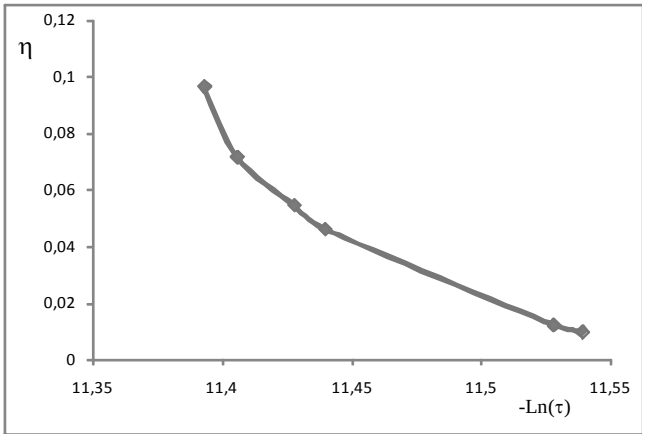

Fig. 1 Dependence of the efficiency of the explosion of detonating mixture on time

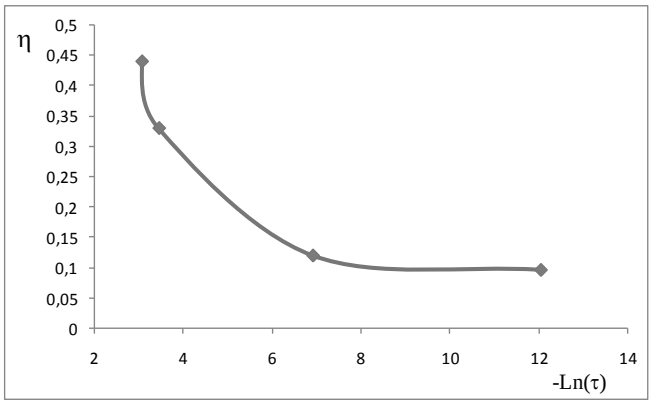

Fig. 2 Dependence of the efficiency of thermal processes on the time of their occurrence

Conclusions

It is suggested when considering the energy conversion processes to take into account the runtime of these processes.

So, an old proverb "The slower you move - the farther you go" got its experimental confirmation.

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WATER PURIFICATION FROM COMPOUNDS OF U(VI) WITH THE USE OF MAGNESIUM-IRON LAYERED DOUBLE HYDROXIDES

An assessment is provided of the effectiveness of the synthetic inorganic sorbents — magnesium-iron layered double hydroxides, for the extraction of uranium (VI) in a wide range of pH of aqueous media with a high salt content, including bicarbonate ions. It is shown that the sorption capacity of these materials depends on the ratio of cations in the structure of their brucite layer. It was found that the most selective with respect to uranium is the carbonate form of magnesium-iron layered double hydroxides with molar ratio 2:1 of $[Mg^{II}]/[Fe^{III}]$ in the brucite structure in layer.

Key words: Uranium (VI), water purification, sorption, magnesium iron layered double hydroxide.

One of the long-term and dangerous consequences of the operation of nuclear power complex is accumulation of permanently increasing volumes of liquid radioactive waste and radioactive contamination of the environmental water objects. One of the most difficult-to-extract from aqueous media and most dangerous among heavy metals and radionuclides is uranium because of its long half-life, high radioactivity and chemical toxicity. Safety indicators of drinking water for the natural uranium must not exceed $\leq$ 1 Bq/dm^3 (0.040 mg/dm^3) [1].

Among uranium compounds the uranium compounds in which the mentioned radionuclide is in the hexavalent state have the greatest technogenic impact on the environment. Even in the presence of mineral and organic- mineral suspensions of natural origin with a high adsorption capacity, uranium (VI) is characterized by considerable mobility in the environment due to the high complexing ability, formation of soluble complexes of di- and tri-carbonaturanil at the pH characteristic to natural water and sewage. This complicates the process of extraction of U (VI) from the aqueous media.

The effectiveness of purification technologies of radioactively contaminated water is directly linked to the well-being of aquatic ecosystems. To purify water from the U (VI) the methods of chemical vapor deposition, extraction, membrane separation, reverse osmosis and adsorption [2-6] are used. Among these methods the sorption one is one of the most effective and technologically simple, especially when removing trace amounts of metal ions.

To remove U (VI) from aqueous media various types of sorption materials are applied: clay minerals, zeolites, activated carbon, biomass, titanium and zirconium phosphates of amorphous and crystalline structure, titanium phosphate-silicate, etc. [6-13].

However, due to the formation of various chemical forms of U(VI) and presence in natural and waste water of complexing ligands and competing ions, it is not always possible to achieve the desired degree of purification [14], and the maximum extraction of the mentioned radionuclide is observed only within a narrow range of pH values of aqueous media.

The promising chemicals for purification of aqueous media from U(VI) are synthetic inorganic layered double hydroxides (LDHs) that are hydrotalcite-like structures and are compounds of the composition: $[(Me^{II})_{1-x}(Me^{III})_x(OH)_2]^{x+}\cdot[(A^{n-})_{x/n}\cdot mH_2O]$, where Me^{II} и Me^{III} are cations in the oxidation states 2+ and 3+, respectively, A^{n-} - anion (either organic or inorganic), n - the valence of anion, x - stoichiometric coefficient [15-17]. The said inorganic materials are characterized by large sorption chemical and radiation resistance, unlike natural or organic ion exchangers, as well as low cost [16].

Efficiency of extraction of toxicants at water purification using LDH dependent on the physical and chemical properties of the structure thereof. The sorption capacity of LDH can be increased by targeted varying at synthesis of distribution of cations in brucite layers and anions in the interlayer space.

There is a publication on extracting U(VI) by Mg,Al- and Mg,Nd-LDH with different anions in the interlayer space (NO_3^-, CO_3^{2-}, OH^-) [18]. Sorption of U(VI) from aqueous solutions using magnetic Mg, Al-LDH intercalation citrate ions was studied as well [19].

Earlier in [20-23] we showed that a sufficiently high efficient for adsorption purification of aqueous media from both cationic and anionic forms of U(VI) are Zn,Al-LDH intercalated by etilendiamintetraacetate-, and hexamethylendiamintetraacetate- and diethylenetriaminpentaacetate- and hexacyanoferrate (II) -ions.

It is known [24, 25] that iron-containing minerals are characterized by high sorption capacity with respect to

U(VI) in a weakly alkaline pH range which is especially important for the extraction of its carbonate form.

The purpose of this work is to assess the effectiveness of magnesium-iron layered double hydroxides (Mg, Fe-LDH) with different molar ratio [Mg^{II}]/[Fe^{III}] in the structure of brucite layer - 2:1, 3:1, 4:1 for the purification of aqueous media compounds of U(VI).

The methodology of the sorption experiments. For the preparation of model aqueous solutions of U(VI), sulphates and bicarbonates of sodium we used their salts $UO_2(SO_4)_2 \cdot 3H_2O$, Na_2SO_4 and $NaHCO_3$ "reagent grade".

The ionic strength (I) of aqueous solutions was created by using $NaClO_4$ "reagent grade" salt solutions.

Sorption of U(VI) was carried out under static conditions at continuous shaking for one hour (water phase volume - 50 cm^3, sample sorbents - 0,100 g/dm^3, $C_{U(VI)}=1 \cdot 10^{-4}$ mol/dm^3). After sorption of the aqueous phase was separated by centrifugation (5000 rev/min), and it was determined in the equilibrium concentration of U (VI) with arsenazo III in a strong acid medium (5.7 M HNO_3) in the visible spectrum (λ = 656 nm and thickness of cuvette *l*=2-5 cm) on the KFK-3-01 [26]. The molar ratio of the light absorption of the complex in this environment $6 \cdot 10^4$. The lower limit of detection of U(VI) is 5μg in 25 cm^3 solution, with measurement error of 5%.

Necessary values pH_0 was created by injection of solutions of 0,1 mol/dm^3 HNO_3 and 0,1 mol/dm^3 NaOH. Values pH_0 and pH of the solution after sorption ($pH_{равн}$) was measured using the ionometer I-160M.

Sorption value of U(VI) (a_s), μmol/g, and the degree of purification (*CO*, %) was calculated as follows:

$$a_s = (C_0 - C_p)\frac{V}{m}, \qquad (1)$$

$$CO = \frac{(C_0 - C_p)}{C_0} \cdot 100, \qquad (2)$$

where C_0, C_p - initial and equilibrium concentration of U(VI), μmol /dm^3; V - volume of aqueous solution, dm^3; m – sorbent sample, g.

The phase composition of the tested sorbents was determined by X-ray diffractometer DRON-2.0 with filtered cobalt radiation.

Synthesis of Mg, Fe-LDH sorbents. LDH in the carbonate form with a molar ratio [[Mg^{II}]/[Fe^{III}] 2:1 (Mg2Fe-LDH), 3:1 (Mg3Fe-LDH) and 4:1 (Mg4Fe-LDH) were obtained by precipitation from solutions of Mg and Fe nitrate by hydroxide sodium in the presence of Na_2CO_3 at constant pH. For this purpose into 1 dm^3 3-necked round bottom flask with "pockets" for the electrodes for measuring the pH and containing 300 cm3 of distilled water, while stirring with a mechanical stirrer we added 1 mol/dm^3 NaOH solution until pH 10. With vigorous stirring, we added to the flask by a peristaltic pump at 1 cm3/min simultaneously two solutions of equal volumes, the first of which contained the required amount of a mixture of 2 mol/dm3 solution of $Mg(NO_3)_2$ and 1 mol / dm3 solution of $Fe(NO_3)_3$, and the second one - the required amount of 2 mol / dm3 NaOH solution to obtain LDH and threefold (compared with the stoichiometrically necessary), Na_2CO_3. During the reaction, pH of the reaction mixture was kept at about 10±0,5 by adding 2 mol/dm^3 NaOH or HNO_3 solutions. After the precipitation was formed, the flask was corked and placed into a drying oven for 24 hours for aging the precipitations at 80°C. Then the precipitates were transferred onto glass filters #4 and washed with hot distilled water until of negative reaction of the sample with diphenylamine.

Radiographic studies of Mg, Fe-LDH sorbents. Diffraction lines being observed in powder diffraction patterns of the resulting products are shown in Table 1. It can be seen that all diffraction reflections are LDH and can be indexed in the hexagonal cells, which is consistent with published data on the crystallization of Mg, Fe synthetic hydrotalcites in the rhombohedral structure [14]. From the Table. 1 and Fig. 1 dataone can see that with increasing of iron content in Mg,Fe-LDH there is a decrease in values of basal reflections 003 and 006, which is associated with increased electrostatic interaction between positively charged layers with interplanar brusit-like carbonate anions. From the X-ray analysis of our results one can only detect a tendency of reduction of the parameter *a* of an LDH hexagonal cell with the increase of substitution of trivalent iron magnesium. It should also be noted the absence of exact matching between the observed and calculated values of the interplanar spacings for the resulting synthetic products that seem to be associated with defects in the structures of the obtained layered double hydroxides (Table. 1).

Table 1. Values of interplanar space and the Miller indices of diffraction lines of the synthesized Mg,Fe-LDH

hkl	Mg/Fe 2:1	Mg/Fe 3:1	Mg/Fe 4:1
003	7,76	7,82	7,94
006	3,862	3,891	3,957
012	2,639	2,636	2,246
015	2,364	2,341	2,379
018	1,997	1,989	2,024
116	1,556	1,556	1,559
113	1,552	1,523	1,528
116	1,440	1,442	1,445

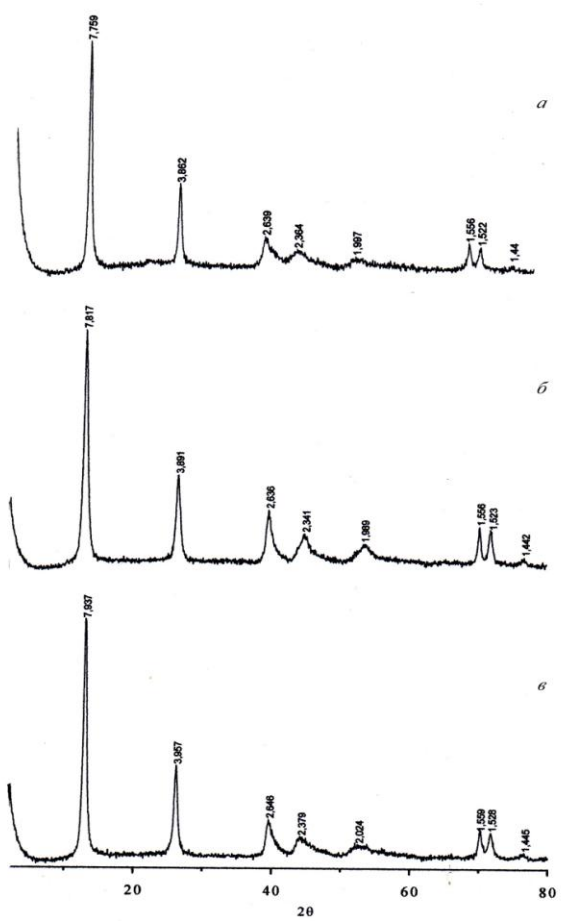

Fig. 1. The diffraction patterns of samples: а — Mg2Fe-LDH; б — Mg3Fe-LDH; в — Mg4Fe-LDH.

Effect of the aqueous media pH on U(VI) extraction by the investigated sorbents. It was studied the efficiency of U(VI) sorption at different pH from the model aqueous medium with I = 0,01 ($NaClO_4$) by carbonate forms of Mg,Fe-LDH (Fig. 2). It was found that almost complete removal of U(VI) by these sorbents is achieved over a wide range of pH_0 (2,0÷10,0), indicating that not only cationic but also anionic forms of U(VI) are absorbed, such as soluble carbonate - $[UO_2(CO_3)_2]^{2-}$, $[(UO_2(CO_3)_3]^{4-}$, resulting in a slightly alkaline pH range upon contact of the aqueous solution with air. [27] It is obvious from the data of [28], that the main mechanism of U(VI) extraction is the formation of solid iron ternary surface complexes in the interplanar space [≡S-L-U], where ≡S is a solid sorbent surface, $UO_2L_m^{n-}$ - - compensation charge - anion, a carbonate form of uranium

Perhaps the extraction of the metal ion in the acidic and neutral pH regions and the aqueous solution is alsom occurred due to isomorphous substitution of UO_2^{2+} with Mg^{2+} in a sorbent composition of [18], and binding of U(VI) with the hydroxyl groups on the surface of brucite layers.

The same as for other representatives of the LDH [20-24], Mg,Fe-LDH is characterized with typical shift of $pH_{равн.}$ values, indicating the alkaline properties of the sorbent, and probably, the anion-exchange replacement of OH anti-ions, partially compensating the positive charge of metal layers.

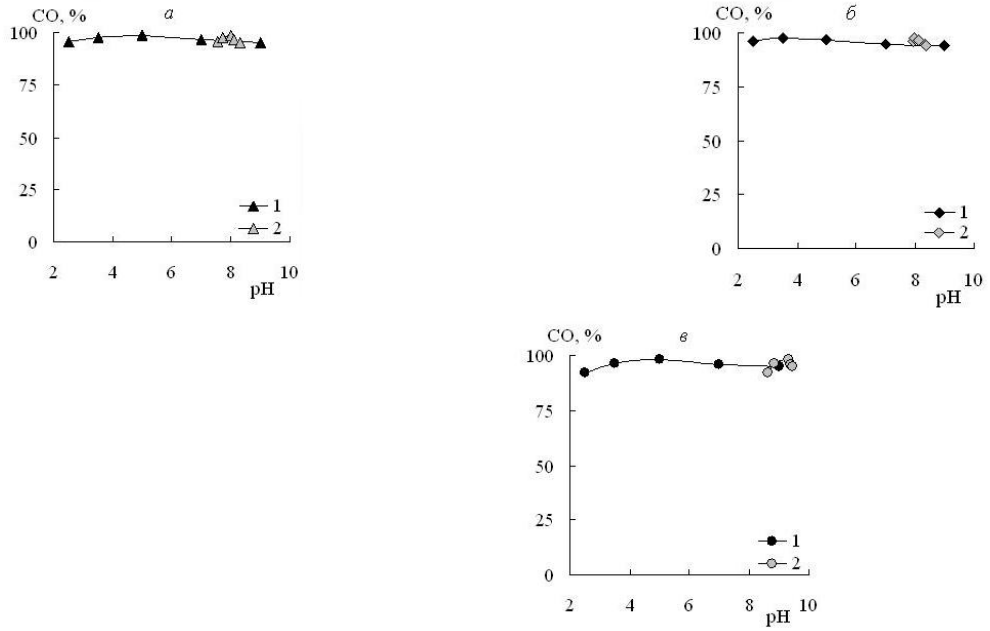

Fig. 2. Influence of pH on the extraction of U(VI) from the aqueous media (I($NaClO_4$)=0,01; $C_{U(VI)}=1\cdot10^{-4}$mol/dm^3) by the following samples *a* — Mg2Fe-LDH; *b* — Mg3Fe- LDH; *c* — Mg4Fe- LDH.
1 — pH_0; 2 — $pH_{равн}$

Sorption isotherms of U(VI) in the studied sorbents. Sorption isotherms of U(VI) were experimentally obtained for Mg, Fe-LDH sorbents (Fig. 3). They were processed according to the Langmuir and Freundlich equations, which are the most common for description of solid-liquid phase of adsorption systems (Table. 2). As one can see, these equations describe satisfactorily the experimental data in virtually entire range of concentrations. Correlation coefficients are quite high (R> 0,9). The maximum limit of adsorption U(VI), calculated from the Langmuir equation, is characteristic for Mg2Fe-LDH and is 555.6 μmol/g.

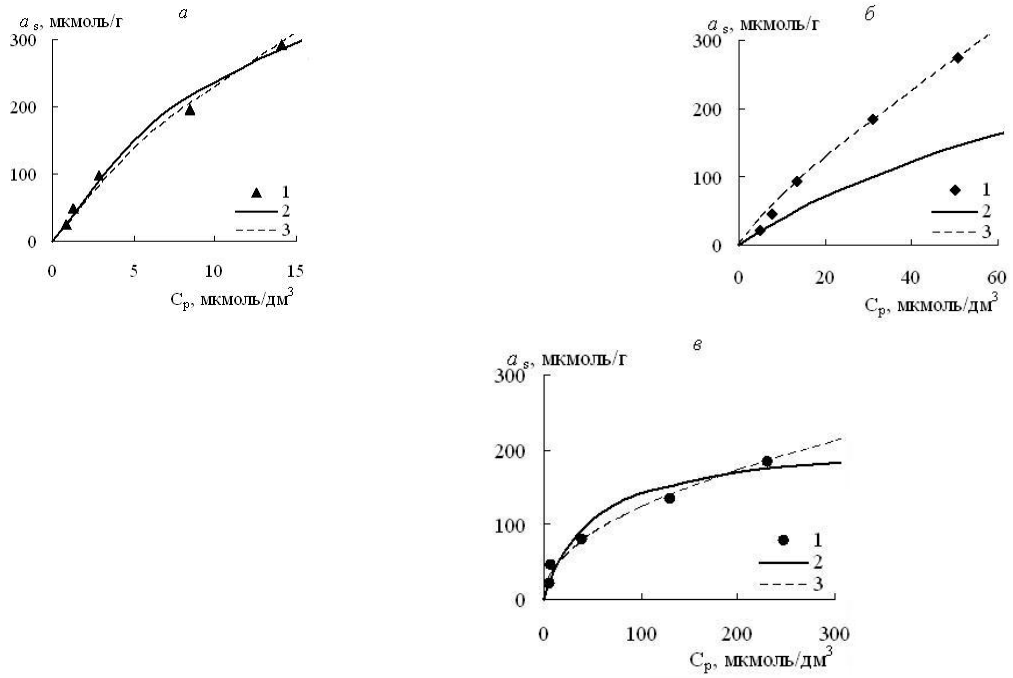

Fig. 3. Sorption isotherms of U(VI) from the aquatic media (pH_0 5,0) on the following samples:
а — Mg2Fe- LDH; *б* — Mg3Fe- LDH; *в* — Mg4Fe- LDH.
1 — experimental data; 2 — isotherms obtained on the basis of the Freundlich equation; 3 — the same for the Langmuir equation.

Table 2. Coefficients sorption equations U(VI) from the aqueous medium (I (NaClO4) = 0,01; pH0 5.0) on

samples Mg,Fe-LDH.

Sorbent	On Freundlich equation $a_s = b \cdot C_p^{1/n}$			On Langmuir equation $a_s = \frac{k \cdot a_\infty \cdot C_p}{1 + k \cdot C_p}$		
	K	*1/n*	*R**	*k*	a_∞, μmol/g	*R**
Mg2Fe- LDH	43,16	0,723	0,9960	0,074	555,6	0,9511
Mg3Fe- LDH	10,93	0,821	0,9780	0,009	454,6	0,9999
Mg4Fe- LDH	12,75	0,491	0,9366	0,020	212,8	0,9602
Note. R — correlation coefficient						

Influence of macro-components of water on investigated sorbents ability to extract U(VI). Cations and anions composition of aquatic environments can significantly affect on the adsorption purification processes from the U(VI) compounds. Given the high tendency of U(VI) to form complexes and its presence in natural and waste waters, predominantly in the anionic form, we investigated the effect of the concentration of such main macro-components of water, as cations Na +, and HCO_3^- and SO_4^{2-}- anions on the extraction of U (VI) by samples Mg, Fe-LDH (Fig. 4).

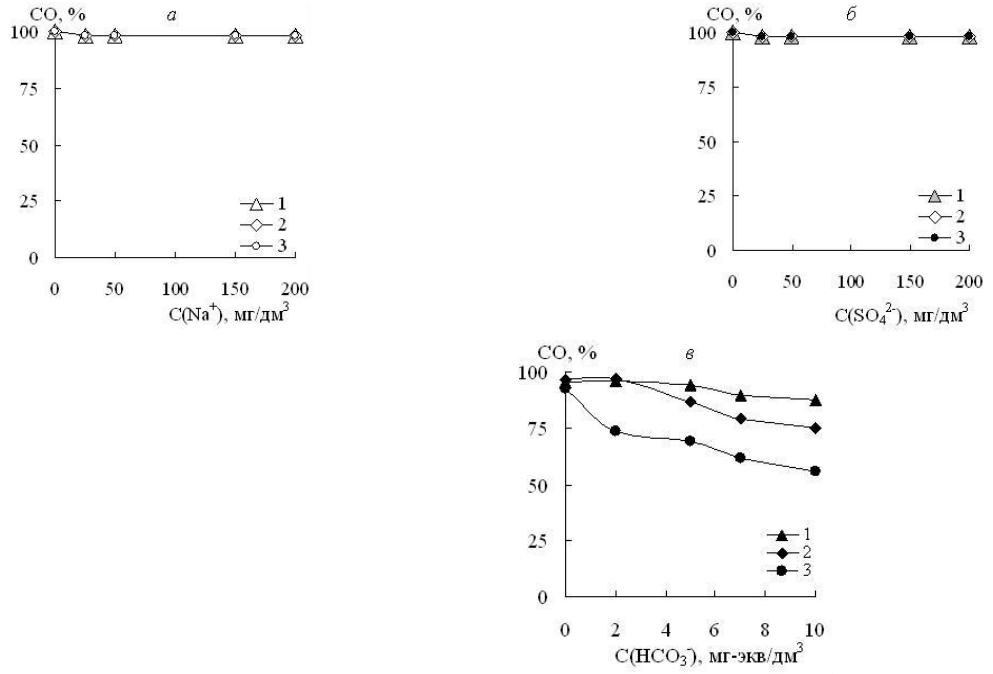

Fig. 4. Effect of the concentration of sodium ions (a), sulfate ions (b) and hydrocarbonate ions (c) on the ability to extract U(VI) from aqueous solutions (CU(VI)=1·10-4 mol/dm3) samples:
1 — Mg2Fe- LDH, 2 — Mg3Fe- LDH, 3 — Mg4Fe- LDH.

It was found that the ions of Na^+ и SO_4^{2-} did not reduce the degree of water purification from U(VI) in a wide range of concentrations (25 ÷ 200 mg / d m^3) sorbents for all samples (Fig. 4a, b).

It should be noted that U(VI) does not form complexes with HCO_3^-- anions [29], but these anions by the shift of carbon dioxide equilibrium in aqueous solution play a decisive role in the formation of solid carbonaceous anionic form of U(VI) at much lower pH than the one for emerging of CO_3^{2-} - anions (pH = 8.3) [27]. Therefore, the presence HCO_3^-- anions in the composition of aquatic media can significantly affect the sorption processes of U(VI).

As one can see from Fig. 4, at low concentrations (up to 5 mg-ekv/dm^3) HCO_3^-- anions have virtually no effect on the extraction of U(VI) by Mg2Fe-LDH samples, and at further increase in the concentration of these anions in aqueous solution, there is a slight decrease in the degree of purification of U(VI). For Mg3Fe-LDH samples it is characteristic a gradual decrease in the absorption of U(VI) even at concentrations of HCO_3^- anions > 2 mg-equiv/dm^3. At the same time when using Mg4Fe-LDH sorbents we observed more sharp (in 2 times) decrease of purification quality. This is presumably due to the fact that increase ratio $[Mg^{II}]/[Fe^{III}]$ in brusit-like layers from 2:1 to 4:1 there is a corresponding decrease of positive charge of a layer, whereby the number of fixed anionic complexes of U(VI) is reduced and sorption capacity of Mg, Fe-LDH is decreased.

Conclusions

A comparison was made of the effectiveness of synthetic inorganic sorbents based on layered double hydroxides for purification of aqueous media over a wide pH range (2,0 ÷ 10,0), including those with high salt content and the concentration of hydrogen ions from the compounds of U(VI). The obtained data demonstrate that sufficiently selective with respect to the said radionuclide is a carbonate form of Mg, Fe-LDH having a molar ratio $[Mg^{II}]/[Fe^{III}]$ in the structure of brucite layer 2: 1, which allows to recommend it for purification of not only medium-mineralized but also highly mineralized natural and waste water containing uranium

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SORBENT MATERIALS FOR PURIFICATION OF AQUEOUS MEDIA FROM THE URANIUM (VI)

The results of the analysis of the scientific publications on the use of sorption materials for purification of aqueous media from the uranium (VI) are presented. The main types of sorbents and mechanisms of uranium retrieval are considered. It is shown that layered double hydroxides are the most efficient in the sorption purification of water from cationic and anionic forms of uranium (VI).

Key words: feed-and bleed disbalance, safety, nuclear power plant, small leak, purging imbalance feeding, trend

The main element of nuclear power is uranium [1]. The most stable in aqueous environments are uranium compounds, in which it is in the hexavalent state. Increased ability of U(VI) to form complexes with ligands of natural and technogenic origin, is caused by its complex electronic configuration, and its existence in natural and waste water mainly in the form of soluble uranyl bicarbonate and tricarbonate anionic complexes causes intense mobility of this radionuclide in the environment and makes it difficult its isolation (Fig.1) [2]. In addition, U(VI) is characterized by high chemical and radiological toxicity.

Therefore, for further development of nuclear energy in Ukraine the main environmental priority is to meet the challenges of rehabilitation and protection of environmental objects against radioactive contamination, in particular in the Chernobyl NPP zone and regions of mining and milling of uranium ore and production of nuclear fuel.

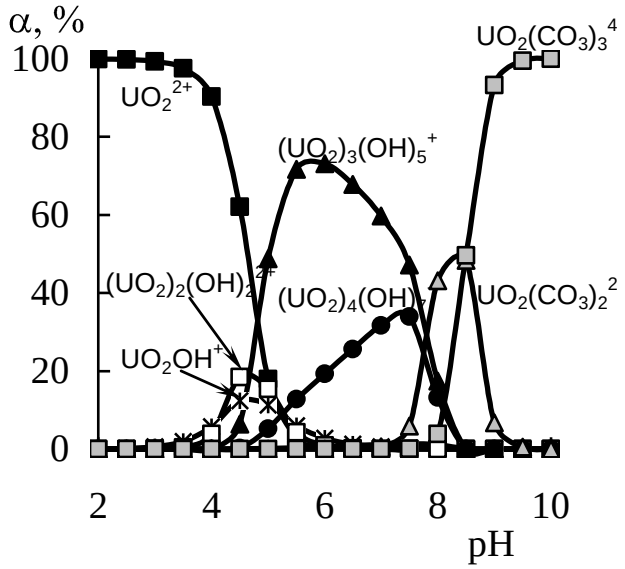

Fig. 1. Distribution of the main forms of U(VI) in aqueous solutions depending on the pH at a concentration of carbonate ions 10^{-5} mol/dm^3 [2].

To ensure this priority, the industry should be provided with high-performance technologies of environmental protection against given radionuclide.

Use of chemical vapor deposition and extraction for the removal of radionuclides from aqueous environments is not always economically (energy consumable and require the use of additional reagents) and environmentally feasible (generate large quantities of radioactive sludge), especially when purifying large volumes of water.

Sorption technique is a common and effective, especially at the stage of purification of aqueous media from radionuclides to the desired level of concentration. This process is flexible in designing of technological equipment and its operation, and allows the concentration of radionuclides and creation of environmental express-monitoring system. In addition, for the most part of sorption materials the recovery is possible through appropriate desorption process.

The effectiveness and selectivity of extraction of radionuclides depends on the nature of the sorbent matrix

and its functional groups, and presence of radionuclides and inorganic and organic components in the aquatic environment [3].

To remove these toxins from the aquatic environments different types of sorption materials are widely used: these are natural and synthetic inorganic sorbents, organic, natural and synthetic sorbents.

For production of adsorbents with the desired functional properties different approaches are used: synthesis of innovative materials, modification of the existing natural or synthetic inorganic and organic materials by implantation into a sorbent matrix of complexing groups, production of selective thin-layer, composite and hybrid sorbents based on available and cheap natural and technological products [4].

Extraction of uranium (VI) from aqueous environments by sorbents based on natural minerals. The use of natural sorbents for the purification of large volumes of aqueous media from ecotoxicants is cost-reasonable. The main advantage of these sorption materials is their low cost and availability of accessible sources of local minerals.

It is known [5, 6], that the most widespread use of natural sorbents for the removal of radionuclides and other toxic metals have become two-dimensional aluminosilicate structure - clay minerals (montmorillonite (or bentonite) [6-8], kaolinite [6], saponite [9] etc.), and three-dimensional aluminosilicate structures - zeolites [10, 11].

In [9, 12-15] it was shown that the pH value of aquatic environment significantly affects the removal of U(VI) by natural sorbents. As it can be seen from Fig. 2, in subacid and alkaline areas it is observed sharp decrease values of sorption of U(VI) in natural montmorillonite, because at low pH dissociation of hydroxyl groups on the particle surface minerals is almost completely suppressed [16, 17].

The sorption of U(VI) by saponit was investigated in [9]. It was found that efficient sorption of U(VI) in the specified natural minerals was also observed at pH = 4,0 ÷ 6,0, corresponding to sorption of only cationic form of U(VI). In acidic and alkaline aqueous solutions the sorption of uranium, as well as other natural minerals is significantly reduced.

The authors [14] presented the results of studies of sorption of U(VI) in clinoptilolite. It is shown that maximum purification of aqueous solutions (in absence of CO_2) from U(VI) is achieved at pH = 6.0, it means the removal of only positively charged forms of U(VI).

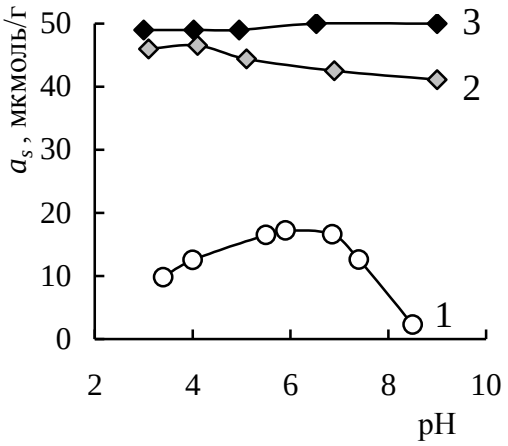

Fig. 2. Effect of pH on extraction of U(VI) from aqueous solutions of various types of sorbents:
1 - natural montmorillonite [16, 17] 2 — Zn/Al-FeCN [47]; 3 — Zn/Al-EDTA [44].

Sorbents based on natural materials not always can provide necessary degree of purification of aqueous media from ecotoxicants, especially in the presence of anionic forms, and have a number of shortcomings, including insufficient high selectivity, and complexity of their use in dynamic conditions. In this regard, to enhance the sorption capacity of natural minerals they are further modified by functionalization of their surface with organic and inorganic agents, water-soluble polymers, heat treatment, etc.

Several publications [16-18] is devoted to investigation of sorption properties of montmorillonite modified by polyetilenimine (PEI), to extract U(VI) and ions of toxic metals. It was found that the degree of sorption of U(VI) by natural forms of montmorillonite is significantly lower than in the modified PEI, where the isotherm at low concentrations is almost straight and the degree of sorption of U(VI) is more than 97% (depending on the uranium content) at pH = 5.5 [16]. However, the presence of carbonate ions reduces the sorption values of

U(VI) by PEI-modified montmorillonite (Fig. 3) [16, 17]. Therefore, for effective extraction of the specified radionuclide the authors recommend to conduct preliminary acidification of the contaminated water.

In [19] it was proposed to extract U(VI) from aqueous solutions at pH = 2,0-7,0 by kaolin modified by calcination followed by acid activation. Modified kaolin sorption capacity increases with increasing of pH. The authors argue that in the investigated range of pH only cationic forms of U(VI) are extracted in the form of uranyl ion and hydroxyforms.

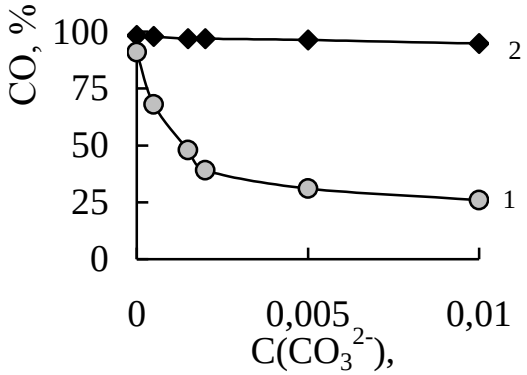

Fig. 3. Impact of the concentration of carbonate ions on the degree of extraction of U(VI) from aqueous solutions by sorbents: 1 - modified polyetilenimine montmorillonite (M-PEI [16, 17]) and 2 – Synthetic zinc-aluminum layered double hydroxide intercalated with complexone (Zn/Al-EDTA [44]).

It was also obtained and investigated the sorbent based on thermally activated (100-500°C) bentonite for the absorption of U(VI) in the range of pH 2,0-9,0 where the given material have chemical resistance [20]. It is shown that the most efficient extraction of U(VI) from aqueous solutions is observed at pH 3-4. This shows the high sorption affinity of thermally activated bentonite only to UO_2^{2+}, which is the predominant cationic form of U(VI) in the indicated range of pH.

It was studied the modification of not only clay minerals, but also zeolites with the aim to remove radionuclides and toxic metals ions from aqueous media, particularly it was studied [21] the structure and sorption properties of bentonite and zeolite modified by polyacryl-amidoxim with respect to cationic form of U(VI) - UO_2^{2+}.

Removal of uranium (VI) from aqueous solutions by bio-sorbents. The new and advanced natural organic sorbents include biosorbents that are characterized by high efficiency of removal of radionuclides and other toxic metal ions, and by low cost. The main types of sorption biomaterials are: non-living biomass (bark, lignin, shrimp, crabs shells, etc.), algae biomass and microbial biomass (bacteria, fungi, yeast). [22]

The authors [23-26] showed the use of biosorbents for purification of radioactive waste water from U(VI) and other actinides.

It was studied the possibility of extraction of U(VI) from model solutions and real water by phytosorbents FS-728, 710, 744, 745, 761, which differ by varying degrees of phosphorylation of groups [25]. It was found that the maximum specified radionuclide sorption is observed at pH $\leq$ 4.

In [26] authors investigated the possibility of removing U(VI) from aqueous solutions by a biosorbent based on bacteria *Bacillus licheniformis*. Maximum sorption capacity of this biosorbent in relation to U(VI) is achieved at a pH of 4.5-5.0, where the removal of only cationic form of U(VI) is realized in the form of uranyl ion and hyidroxoform and at optimum temperature of 25-45°C.

However biosorbents separation from the aqueous phase after sorption is complicated by formation of large quantities of radioactive sludge which require further processing. In addition, instability of the bio-sorption processes (sensitivity to pH and temperature of the water media, concentrations of cations, inorganic and organic complexing ligand present in water) and the difficulty to obtain the required amount of biomass hinders commercialization of water treatment technology by biosorbents.

Extraction of uranium (VI) from aqueous media by inorganic sorbents. In the last 10-15 years, more efforts were put into study of the use of synthetic sorbents, mostly inorganic that have mechanical, chemical and radiation resistance, for removal of radionuclides and other toxic metals.

For the selective extraction of U(VI) and other radionuclides from water titanium and zirconium phosphate - ion exchangers of amorphous and crystalline structures are used [27]. Efficient removal of U(VI) from solution is observed only in the range of pH = 3.0÷5.0, that is in range of existence of cationic forms of investigated radionuclides, but in the presence of nitrates of alkali and alkaline-earth elements the distribution coefficients of

U(VI) are reduced.

The high selectivity to U(VI) has spherical amorphous granular titanium phosphates and titanium phosphate-silicates, titanium-silicates that can be used in ion exchange columns due to optimal grain size [28, 29]. For the titanium phosphate the distribution ratio of the indicated eco-toxicant is 656÷4732 ml/g; for titanium phosphate-silicates - 488÷8175 ml/g; for titanium-silicates - 697÷4152 ml/g at pH ≈ 5.3, where U(VI) exists only in the form of positively charged forms.

To purify water solutions from U(VI) a nanoporous sorbent was developed based on zirconium oxyphosphate - $Zr_2O(PO_4)_2$ [30]. This sorbent showed high sorption capacity with respect to uranium at pH = 4.0-7.0. However it was revealed that sorption equilibrium is reached only after 48 hours, and extracting of U(VI) from aqueous solutions in air ambient (in the open systems) containing CO2 is significantly reduced at pH ≥ 7.0 due to formation of anionic soluble carbonate complexes of uranium .

Effective for the removal of radionuclides is the use of ion exchange resins [31], which provide high purification performance and should have higher absorption characteristics. However, ion exchange resins practically do not absorb colloidal forms of radionuclides, and they are expensive compared to other sorption materials.

The authors of [32] studied the adsorption properties of ion exchange resins brand Amberlite XAD-2, treated by Cyanex272, with respect to U(VI) depending on the concentration of uranium, acidity and time of extraction. Maximum extraction of U(VI) (99,7%) is at concentration of HNO_3 in aqueous solution 10^{-3} M.

In [33] it was shown that the use of ion exchange resins for purification of aqueous media of di- and tricarbonateuranil complexes of U(VI) the uranium content in treated water is less than 1 mg/dm^3, which is well below the maximum permissible concentration (40 mg/dm^3).

It should be noted that the use of ion exchange materials requires their regeneration, which can lead to a decrease in exchange capacity and to secondary contamination.

One of the most common groups of anionic inorganic sorbents is the class of layered double hydroxides (ШПГ) or hydrotalcite-like materials (synthetic anion-exchange clays). These compounds have the following composition:

$[(MeII)_{1-x}(MeIII)_x(OH)_2]_y+\cdot[(An-)_{x/n}\cdot mH_2O]$, where MeII and MeIII are cations in oxidation states +2 and +3, respectively, An- - almost any anion (organic or inorganic), n - valence of anion, x – stoichiometric coefficient, which means molar ratio [MeII] / [MeIII] (Fig. 4) [34-37].

These inorganic absorption materials, unlike natural and organic ion exchangers are more chemically and radiation resistant, and also have low cost [34]. The ability to relatively easily control the synthesis of homogeneous distribution of cations in brucite-like layers and anions in the interlayer space facilitates targeted variation in the concentration of reaction centers in a sorbent matrix to improve the selectivity of layered double hydroxides (LDH).

Heat treatment (calcination) of the mentioned layered sorbents at 350-500°C leads to creation of a homogeneous mixture of double oxides and increases their sorption capacity by increasing the number of active centers. At sorption of anions from an aqueous solution they observed regeneration of the heat-treated LDH structures.

The authors of [38] for the sorption of cationic forms of U(VI) proposed Mg/Al- and Zn/Al-LDH, containing carbonate anions in the interlayer space. It is shown that significant amounts of chloride and sulphate (15-50 g/dm^3) have no influence on the degree of water purification from U(VI) by the indicated sorbents, but hydrocarbons significantly reduce extracting of U(VI).

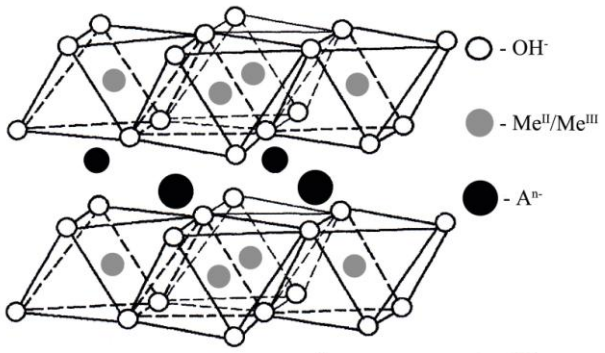

Fig. 4. Spatial drawing of structure of LDH with octahedral crystal lattice [37] where Me^{II} and Me^{III} - cations in oxidation states +2 and +3, respectively, A^{n-} - virtually any anion or anionic complex.

Promising among the LDH are organic-nonorganic hybrid types, which contain in the interlayer space complexing anions which strongly bind ions of toxic metals in the complex compounds [39, 40].

In [41, 42] the sorption of U(VI) ions and other toxic metals was investigated in Mg/Al- and Mg/Nd-LDH with different anions in the interlayer space (EDTA, CO_3^{2-}, NO_{3-}, OH^-). It was found that in the presence of CO_3^{2-} in aqueous solution in an amount of $10^{-3} \div 5 \cdot 10^{-2}$ mol/dm^3 carbonate forms of sorbent are virtually ineffective for extraction of U(VI). For the sorbent Mg/Al-EDTA the distribution coefficients K_d for U(VI) are up to 100-120 cm^3/g.

The authors of [43] argue that purification of aqueous solutions of U(VI) by a magnetic Mg/Al-LDH intercalated with citrate ions is associated mainly with the formation of metal-citrate complexes in the interlayer space of the said sorption material. It is shown that the maximum extraction of U(VI) from aqueous solutions by the mentioned LDH occurs at pH 6.0 and is up to 180 mg/g at the initial concentration of 200 mg/dm^3 and the dose of 0.05 g of the sorbent. At further increase of the pH value sorption of U(VI) is dramatically reduced as a result of increase in the proportion of negatively charged forms of the investigated radionuclide.

Recently it was reported about successful purification of aqueous media from U(VI) and other ions of toxic metals, by inorganic sorption materials based on Zn/Al-LDH intercalated with etylendyamintetra acetate - (Zn / Al-EDTA) hexametylendyamintetraacetat- (Zn/Al-HMDTA) and dyetylentryaminpentaacetat- (Zn/Al-DTSIA) and hexacyanoferat (II) -anions (Zn/Al-FeCN) [44-47]. It is shown that these sorbents are quite highly effective for the extraction of U(VI) from aqueous media, regardless of its location in natural water, unlike natural montmorillonite that removes only the cationic forms of uranyl (Fig. 2, 5) [44, 47]. At removing the U(VI) from aqueous solutions with a high content of carbonate anions it was achieved maximum degree of purification on Zn/Al-EDTA, compared with the natural mineral, modified by PEI, where at the increase in the concentration of carbonate anions in aqueous solution the degree of purification is sharply reduced (Fig. 3) [16, 44].

For comparative evaluation of effectiveness of different types of sorbents the U(VI) extraction by specified materials from the waste water of mines were analyzed [48], where the mentioned radionuclide exists at pH≈8 in a the purely absorbed forms, mainly the carbonate ones. As it can be seen from Fig. 5, the use of natural montmorillonite is ineffective for purification of the investigated aquatic solutions from U(VI) because the degree of purification is than 1% at a dose of sorbent of 6 g/dm^3; modification of the indicated mineral with PEI significantly improves the absorption of toxicant - the degree of purification is 69%. At the same time when using Zn/Al-EDTA the degree of purification of waste mine water from U(VI) is 95% even at a dose of sorbent 1 g/dm^3, with increasing the doses of sorbents based on Zn/Al-LDH to 6 g/dm^3 the efficiency water purification increases and reaches 99% for Zn/Al-EDTA, 93% for Zn/Al-HMDTA, 85% for Zn / Al-FeCN [16, 17, 44, 47].

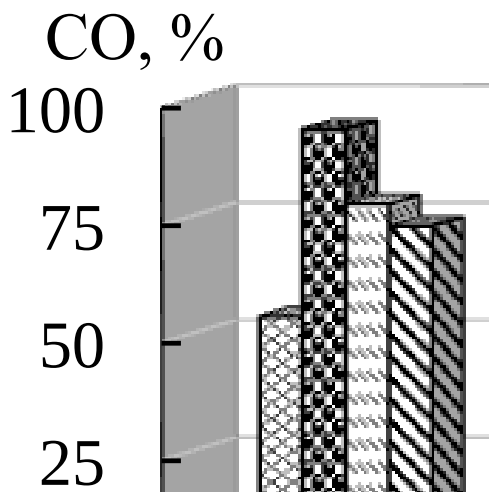

Fig. 5. Influence of dose of sorbents on the degree of extraction of U(VI) from waste mine water: 1-montmorillonite and montmorillonite PEI 2 [16, 17]; 3- Zn/Al-EDTA [44], 4- Zn/Al-HMDTA [45], 5- Zn/Al-FeCN [47].

The main advantages of the use of LDH compared to other sorption materials are: environmentally friendliness (no pollution from anionite – only CO_3^{2-} is released in aqueous solution; ease of synthesis at low temperatures; universality of chemical composition; chemical, thermal and radiation stability, and ease of use in technological operations

Conclusions

The analysis of scientific publications has shown that removal of U(VI) from aqueous solutions depends on

many factors: the forms of uranium(VI) and physics-chemical properties of water bodies in the environment. Sorption technique is the promising one at extraction of radionuclides from contaminated aquatic media, particularly in the refining stage. However, none of the listed sorption material, except the layered double hydroxides, cannot effectively purify natural water from the anionic forms of uranium (VI). Therefore, to improve the environmental situation it is an urgent task to develop highly efficient and selective sorption materials based on various forms of layered double hydroxides intended to extract ecotoxicants of different nature.

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