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THE PROBLEMS OF UTILIZING GRAPHITE OF STOPPED GRAPHITE-URANIUM REACTORS

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A list of radioactive nuclides, the activity of which forms the main part of total activity of graphite stack and graphite elements of the construction of stopped industrial graphite-uranium reactors has been defined. The analysis of activity part contributed by these nuclides at different moments of time after stopping reactor was carried out. A set of construction graphite elements, in which there is a possibility of self-sustaining release of the energy stored (Wigner's energy) was determined. It was stated that the most value of the Wigner's energy is achieved in graphite constructions operated in low-temperature region or at high values of flux densities of damaging neutrons and concurrent gamma radiation.

Introduction

The question on removing nuclear plants with commercial carbon-uranium reactor (CCUR) from the service constitutes a complex of problems connected with the choice of optimal ways and methods of handling with stored radioactive waste (RAW). Among the whole mass of stored RAW spent graphite of CCUR occupies a special place. After long irradiation graphite has not taken on any properties for useful application. Therefore, irradiated graphite falls in the category of unused RAW and requires individual approach to the choice of methods in its treatment. It is explained by many factors:

1. Reactor graphite has a unique crystal structure and is characterised by porosity, which defines its properties and behaviour at irradiation.
2. Graphite stack is a basic element of CCUR core not to be replaced during the operation life and it has the most accumulated neutron fluence among the RAW.
3. Graphite of CCUR stack blocks and bushes has a number of features in size, isotope composition of radioactive pollution and character of radioactive nuclides distribution both in stack volume and in separate graphite details. Radioactive contamination of graphite details is, first of all, determined by induced activity (mainly ^{60}Co, ^{3}H, ^{14}C) due to impurity activation contained in original material. In this case ^{14}C, forming 95 % of graphite activityб is involved into biological chains. In addition to activation products, graphite activity is defined by radioactive nuclides (^{137}Cs, ^{90}Sr, ^{154}Eu etc.), formed in the stack

as a result of coolant leakage and falling of fuel fragments into stack.

4. Graphite is a fire-risk material with high specific combustion value (~8 kkal/g, ignition point amounts for ~700 °C). This fact is dramatized by the presence of stored energy (the Vigner's energy) in irradiated graphite.

From the moment of CCUR stopping «Siberian chemical combine» (SCC, Seversk town) I-1, EI-2, ADE-3 (1990–1992) in the course of concept development on removing of CCUR from the service the work on studying radiation characteristics, physical-chemical properties and stored energy of spent graphite has been done [1–5].

Radiation characteristics of graphite in stopped CCUR

To solve the problem involved the necessity of developing ways of irradiated graphite treatment the probing for the purpose of defining distribution of radiation fields in graphite stacks of stopped CCUR of SCC [6]. Stack probing allowed us to obtain photon field distribution at stack height and radii as well as the presence of neutron fields in all stacks. The regions with increased density of neutron flux were revealed. In the same regions high values of dosage rate were noted. These changes showed that approximately in 2,5 % of stack volume the magnitude of dosage rate exceeds 1000 R/h, in 34 % – the magnitude of dosage rate lies in the range 10...1000 R/h, in the rest of stacks it is to 10 R/h. Presence of similar regions is explained by incidents at which fragments of fuel material fall into graphite stack. Since all incidents took place at the initial period of reactor operation, these fragments were exposed to long radiation (more than 22 years), that caused the formation of fission product and transuranium elements in separate regions of stacks. On the bases of the data on stack probing irradiated fuel quality in each stack of stopped CCUR of SCC was estimated.

Calculations made before the reactor stopping showed that nuclide composition of graphite radioactive contamination was limited by some radioactive nuclide – ^{3}H, ^{14}C, ^{55}Fe, ^{60}Co, which were formed, mainly, due to neutron activation of graphite impurities. Results of stack probing showed that it is far from being so. It was necessary to know the total composition of graphite radioactive contamination. It could be done only by means of spectrometric analysis of graphite samples from different stack details. Various techniques and devices were developed to select graphite probes from practically all kinds of stack details [6].

Probes of graphite bushes were selected both from those extracted from the stacks and those extracted from the storage. The samples for radiospectrometric analysis were made from these probes.

The results of analysis showed that 95 % of all activity of irradiated stack graphite amounted for ^{14}C. Though graphite average specific stack activity amounted approximately $6{,}9\cdot10^{8}$ Bc/kg, nevertheless, in separate regions it could be significantly higher. Only for ^{137}Cs activity of approximately 3,5 % of stack volume amounted $10^{8}...10^{10}$ Bc/kg.

Study of activity distribution in thickness of graphite stack details showed that on the surface the activity is 3...5 times higher than that in the volume. As a rule, the given difference is conditioned by the presence of fission products and actinolites (up to ^{244}Cm) on the surface of radioactive nuclides.

Distribution of fission, activation products and transuranium elements in graphite stack occurred depending on individual ability for sorption, diffusion and migration of the radioactive nuclides under the influence of operational factors such as temperature, pressure etc. In some cells basic radioactive contamination was concentrated in the joints between graphite blocks along the column height and in different defects of surface.

By means of estimation of changes in radiation characteristics in time of main constructions as well as in the analysis of cut metal samples of different types of alloys it was determined that in the initial period of exposure activity of gamma-emitting nuclides in metal CCUR constructions was mainly defined by ^{51}Cr, ^{59}Mn and ^{60}Co. In 8–12 months the basic dose-forming nuclide was ^{60}Co. Calculations made for CCUR showed that gamma-radiation intensity in metal constructions at 100-year exposure decreased sufficiently, but maximum value of dosage rate in metal constructions will not exceed 0,01 R/h in 100 years.

In graphite stacks in the regions of irradiated fuel particle localization in the period of exposure (to 3 years) the spectrum of gamma-radiation was defined by radiation of short-lived fission products: Ce^{144}, Ru^{106}, Cs^{134}, Eu^{155} etc. In the subsequent time interval (from 3 to 50 years) it was defined mostly by nuclide gamma-radiation Co^{60} and Cs^{137}, in the less degree Eu^{154}. After 50 years of exposure radiation environment will be formed only by gamma-radiation of long-lived fission product Cs^{137}. The dosage rate in these local regions can achieve 100 R/h after 100 years of exposure.

Thus, after a long period of exposure radiation environment in CCUR reactor space I-1, EI-2 and ADE-3 changed significantly due to natural decomposition with regard to short-lived activation and fission products. Residual activity will be mainly accounted for graphite stack. In this case the determining role will be played by activity of long-lived fission products and transuranium elements.

Thus, radiation environment of graphite stack will influence the concept development and technical project on removing reactor from the service.

The obtained data on activity of graphite details, summary activity of stacks, isotope composition of activity, activity distribution over the stack volume and separate details were applied in composition of radiation passports and other documents as well as they make possible to conclude on nuclear safety of all stopped CCUR of SCC.

The work in given direction continues to obtain more complete information and improvement of calculation and experimental methods in radiation examination.

The analysis of general radiation environment shows that the work on complete disassembly of basic highly-active reactors is not appropriate at the moment for technical and economical reasons. The most optimal variant is that of removing from the service providing for suspended disassembly of reactor constructions. To increase the safety of stopped CCUR it is necessary to improve the existing and create the additional safety barriers that should prevent from migration of radiation nuclides with different chemical properties and maintain the properties within the time of potential ecological danger of long-lived radiation nuclides.

At present graphite stacks of stopped CCUR at SCC are not disassembled and are in reactor space. Safety barriers separate graphite stacks from the environment, they prevent from radiation nuclides flowing out. In radial direction it is:

- silumin inserts;
- metal core shroud;
- internal and external walls of lateral protection (20 mm);
- concrete molding of mounting space (1500 mm);
- concrete walls of reactor cavity (2000 mm);
- concrete wall of the building (1000 mm).

In the axial direction safety barriers are support-protecting constructions, burial of protective constructions (3000 mm), concrete molding of lower constructions etc.

Graphite stored energy of stopped CCUR at SCC

From the safety point of view the potential danger presents the possibility of temperature rise within the storage of irradiated reactor graphite due to self-sustaining release of stored energy. As native and foreign investigations show, the value of stored energy and possibility of its self-sustaining release depends strictly on the temperature of graphite irradiation. It was supported by the studies carried out at SCC.

It was shown that the magnitude of stored energy achieves its maximum value in upper and lower parts of graphite bush set of reactor cells I-1 operating in proton condition. Bushes of its stack are the most «low-temperature» details among the details of graphite stacks in all stopped CCUR of SCC. The results showed that for graphite details of this reactor the self-sustaining release of stored energy is possible for graphite of upper and lower part of bush set. In this case the value of stored energy amounts approximately 220...280 kal/g for upper bushes of operating cells, but for upper bushes of control and safety system (SCS) cells and cells with enriched material (EM) it is approximately 400 kal/g. According to the results of investigation and estimation in possibility of stored energy release it is necessary, first of all, to extract I-1 from the CCUR stack:

- 2–3 upper inserts from operating cells;
- Complete sets of inserts of SCS and EM.

High value of the Wigner's energy for upper inserts and SCS inserts is conditioned by the fact that in the radiation temperature range 100...300 °C in the graphite content there is a large quantity of non-equilibrium interstices and vacancies, which define the changes in parameters of crystal lattice and corresponding graphite properties. In this range decrease in temperature of radiation and flux density of accompanying gamma-radiation, causing decrease in thermal and radiation γ-anneal, results in increasing the concentration of accumulated defects [7]. Graphite inserts of EM cells are employed at higher values of flux densities of damaging neutrons (more than 180 keV) and accompanying γ-radiation that results in high value of the Wigner's energy.

In determining stored energy of ADE-3 CCUR stack details similar recommendations were given only for upper inserts of SCS and EM, since due to higher temperature of ADE-3 CCUR stack inserts the self-sustaining release of stored energy is possible only for graphite of these inserts. This problem is more actual for graphite of I-1 CCUR insert graphite than for that of ADE-3 and EI-2, both by the reason of more inserts for which self-sustaining release of stored energy is possible and owing to large mass of inserts themselves.

Among all graphite details of stopped CCUR of SCC self-sustaining release of stored energy is possible for:

- 12...15 % mass of all I-1 CCUR inserts;
- 1...2 % mass of all EI-2 and ADE-3 CCUR inserts.

Research shows that for other part of graphite mass of stack graphite at stopped CCUR of SCC the self-sustaining release of stored energy is impossible. In the opposite case to provide safety at removing from the service of CCUR would be rather difficult, since stack inserts could be easily extracted, but the blocks are not change parts.

Ways of graphite utilization in stopped CCUR

«The concept of removing commercial reactor installations from operation» developed in Atomic Energy Department in 1990 proposes three variants: «Conservation», «Entombment» and «Abandonment», which correspond to three stages according to MAGATE classification («Storage under control», «Limited use of platform», «Unlimited use of platform) and three techniques applied in the USA («Safety storage», «Burial at place» and «Removal»). All variants differ from each other just in the number of capital outlays for their performance and values of dose loading for personnel and are reduced to one item: finally the reactor installation should be buried according to the requirements imposed on burial of radioactive wastes, moreover, any variant of removing from operation should allow for complete disassembling of reactor installation.

To fulfil «The Decisions C1-483 of NTS Section № 1» of Atomic Department from 1990 by the decision ratified by the chief of 4 GNTU for SCC reactors the concept variant of removing from operation – «Burial at place» is adopted. The given variant consists in the following: after complete removing of fuel, reactor aftercooling and complex engineering and radiation examination the reactor installation (including graphite stack) is set in long-term stability state, i. e.:

1. All non-activated equipment is demonstrated.
2. Additional safety barriers for prevention from radiation nuclides out of reactor are produced:
   2.1. Reactor bottom is concreted by waterproofing concrete, making support of main supporting iron.
   2.2. Lateral metal constructions are filled with concrete.
   2.3. All structural openings in concrete reactor cavity are sealed.
   2.4. All cavities of reactor space are filled with mixtures of natural materials on the basis of concrete clays.
3. Upper part of the reactor is sealed by means of demountable ferror-concrete spans for protection from fire, blast wave, impact and etc, and in this way the reactor is buried for 100 years. Radioactive equipment and systems out of concrete reactor cavity are disassembled after the exposure of 30...50 years.

The given solution is equivalent to Stage 2 in terms of MAGATE classification. Similar solutions distinguished for only time exposure, are accepted nearly in all countries of the world. Thus, for example, for Hanford reactor in the USA the time of exposure 75 years is accepted, for commercial reactors in Great Britain – 100 years. Long time of exposure allows for development of the most optimal ways and methods of radioactive construction treatment.

According to «The perspective plan ...» the work on making I-1, EI-2, and ADE-3 of CCUR continuously stable within the reactor cavity is supposed to be completely finished in 2010.

As it was mentioned above, more serious ganger is graphite inserts, stored at storages of solid wastes. At the moment the most perspective ways of spent graphite insert treatment are combustion and sealing by means of flowing clay mud.

At present different ways of graphite combustion are suggested: traditional way; in boiling bed; by gas laser, as well as graphite gasification by means of superheated steam (pyrolysis). As it was estimated by experts, combustion of spent graphite will produce, as a result, radioactive wastes prepared for long-term burial of 1...2 % volume of initial graphite content. All listed techniques have one essential disadvantage: in combustion of graphite gaseous radioactive product $^{14}CO_2$ is produced. To fix this product one can turn it into solid chemically inert compounds. For this purpose, for example, calcium and magnesium carbonates are supposed to be used. The main disadvantage of such method of utilization consists in increase of waste content.

French investigations and developments [8] have shown that the solution on spent graphite combustion is acceptable from the point of view of radiation safety. The pilot installation for combustion in boiling bed of ground graphite of 30...50 kg/h capacity was developed and tested. α-emitters and other radiation nuclides contained in ^{60}Co, ^{137}Cs, ^{55}Fe, graphite could be reliably caught by filters, but ^{14}C and ^{3}H are released into atmosphere. At combustion of ground powder of 1000 tons of graphite per year in boiling bed 4 times more ^{14}C than at operation of one VVER-440 reactor and 2 times less than in operation of a recycling fuel plant will be released into atmosphere. That is the value of radioactive carbon release will be at the level typical for objects of atomic industry.

The proposed technology of combustion in boiling bed developed by «Framatome» (France) firm ensures reliable isolation of nearly all radiation nuclides in graphite from the environment apart from ^{14}C. Owing to high mobility, as a result of atmospheric processes ^{14}C is transmitted over long distances and, being oxidized to $^{14}CO_2$, it is involved into natural carbon cycle together with common carbon dioxide through photosynthesis.

SCC and Geological Institute of RAS have considered the practical possibility of sealing in solid waste store by means of flowing clay mud. As a result of generation of clay cement mass by this method one can exclude possible radiation nuclide release out of concrete constructions both in gaseous and ion-solution forms.

Scientific-Research and Design Institute of Technology (SRDIT) has developed the technique of filling the stores with clay mud for experimental-industrial tests. RAO conservation in the suggested way in geological environment is not only ecologically friendly, but also the most economically reasonable.

The results of mathematical simulation of radiation nuclide transport in compositions on the clay basis from Tomsk region deposits and experimental study of their properties allow us to make a conclusion on the fact that given compositions have high adsorptivity with respect to radiation nuclides, preserve the physical properties over hundred years, stability in construction material behaviour in filler environment, sufficient bearing resistance. Compositions and ways of preparation of these compositions for construction of additional safety barriers in existing RAO storages of SCC reactor production has been determined.

At site № 2 of SCC a breadboard construction of the store was developed. At this store the technique of additional safety barriers proposed by SRDIT will be tested by means of pumping flowing clay mud. The results of studying properties of recommended clay compositions allow for suggestion that the clay monolith produced as a result of pumping mud into breadboard model will possess high migration-resistance and filtration-resistance characteristics. In the case of successful tests it will be possible to apply this technology for creation of additional safety barriers in the existing RAO stores of SCC reactor operation.

At present, other techniques of irradiated graphite treatment are being considered. They are to be thoroughly justified in terms of ecological, technological and economical factors.

Conclusion

The main trends in modification of additional safety barrier production by means of pumping flowing mud solution into the storage applicable to existing RAO storage of SCC reactor production has been grounded. The basic reason for choosing the given modification is the fact that RAO conservation is environmentally friendly and economically reasonable in geological surrounding. It was stated that for preventing from self-sustaining stored energy release from CCUR core in first place 2–3 graphite upper bushes of operating cells should be removed as well as the complete set of bushes from CM3 and OM cells. Composition and way of preparing compositions on the basis of Tomsk clays was determined. They provide the following properties:

- high adsorptivity with regard to different radioactive nuclides;
- conservation of properties during several hundreds of years;
- stable behavior of construction materials in the filler environment;
- sufficient bearing resistance.

The question on adsorptivity of these clays is still an open question. The Institute of Physical Chemistry of RAS is engaged in its solution. As a result of this work the preservative for RAO storage and stopped reactors will be selected.

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GAMMA-SPECTROMETRIC CONTROL METHOD OF ACTIVITY AND NUCLIDE COMPOSITION OF LOW-ACTIVE SOLID RADIOACTIVE WASTE

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The gamma-spectrometric control method of low-active solid radioactive waste, based on direct measurement of activity and nuclide composition has been developed. The measurements were carried out in the geometry of standard steel container of 200 l. volume, where low-active wastes were placed. To take into account distribution non-homogeneities of solid waste over the geometry measured a special rotating platform was used, the low-active radioactive wastes being placed on it. Metrological certification was performed and the main errors of this method for 95 % of confidence probability were determined.

Introduction

For enterprises of atomic industry increase in safety processes of radioactive waste treatment is one of the perspective ways of development. The crucial moment in solving the safety problems is development and introduction of modern methods in radiation survey permitting for determination of basic characteristics (activity and radioactive nuclide composition) at all stages of treatment [1, 2].

At present radiation survey of solid radioactive waste (SRW) of low and middle activity produced in the process of operation at most atomic enterprises is performed by measurement of dosage rate and value of surface radioactive contamination. As a rule, metrologically certified techniques of SRW activity and nuclide composition considering distribution non-homogeneities of solid waste activity over the measured geometry is absent.

The purpose of the present work is to develop gamma-spectrometric control method of low-active SRW activity and nuclide composition including the corresponding methodological and metrological equipment.