The Reactor Graphite Handling. Problems and Solutions – 17208

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ABSTRACT

The problem of industrial uranium-graphite reactors and NPP units decommissioning is one of the urgent tasks that require efficient and cost-effective solutions.

To date, only Russia has already shut down and subject to decommissioning more than 120 objects. World-wide, more than 260,000 ton of irradiated graphite have arisen from the commercial nuclear power operations, research reactors, and defense production reactors. Under the circumstances the problem to develop cost-effective methods of the irradiated graphite management has appeared.

Alternative disposal options for the irradiated graphite were discussed for at least 25-30 years. To date, various methods of irradiated graphite reprocessing have been proposed. They are based on the application of different physical and chemical processes, most of which involve incineration of the entire mass of graphite. However, they all have disadvantages, and do not solve the whole complex of problems.

The primary goal of the research presented here was to carry out a study aimed at the development of a more simple technology (as compared to the incineration of the entire mass of graphite) that would provide for the graphite purification and allow the possibility of its transfer to the less hazardous waste category followed by the disposal of it in a near-surface repository.

To achieve it is proposed to consider two directions, each of them reducing the cost of graphite reprocessing and the waste volume disposed of in the repository.

The first alternative is based on the principle of partitioning radionuclides of different nature from the graphite, thus allowing to transfer it into the LLW category and minimize the volume of secondary waste products.

The second option to change the reprocessing technology is the idea to use reactor graphite as the starting material to produce stable final waste form compositions intended for the long-term storage of radioactive waste including transuranic elements.

INTRODUCTION

The uranium-graphite reactors (there are only about 250 in the world) used primarily for military purposes - for production of weapons-grade materials, and later for electricity production, and to undertake work of a scientific nature. With the completion of these reactors operating materials (graphite and steel) have become waste.

To date, the Russian Federation has already stopped and shall be decommissioned more than 120 objects. (Total in the world was put into operation about 260 energy, industrial, and experimental reactors and in them is more than 260,000 tons of irradiated graphite.) Mass withdrawal problem of exploitation of industrial uranium-graphite reactors (PUGR) and NPP graphite-moderated, is quite relevant and its solution requires the search for effective and economically sound technologies [1-3]. Processing of the reactor graphite can substantially reduce the volume of waste intended for disposal and increase safety.

Considering the development prospects of different approaches to the irradiated graphite handling, the complexity of the task in question should be pointed out. The solution demands thorough and comprehensive characterization of the irradiated graphite. Indeed, the topic is addressed in the large number of publications available to date. But the analysis of the reported data [1-5] makes clear the information is insufficient and the studies should be continued.

The main reason is a great variety of reactor types used and different conditions of their operation. Moreover, the initial graphite characteristics were different due to the different composition of impurities, and hence, the irradiated graphite differs from site to site by the content and spectrum of radionuclides accumulated over the long-continued operation. Again, the emergency situations should not be forgotten, when the activity and the spectrum of radionuclides in the graphite cannot be calculated merely as a function of the accumulated activation products and the coolant used.

Knowing the level of contamination of the different sections, it is possible to organize sorting (essentially fractionation) graphite and depending on the activity of selected fractions offer different processing methods.

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RESULTS OF EXPERIMENTS AND DISCUSSION

One of the options for the reactor graphite management is to store it indefinitely in under-ground storage facilities. This can present a number of problems, both environmental and economic.

Quite obviously, the effective strategy of the reactor graphite handling should provide for the differential approach to different objects. The choice of the most effective solution requires the systematic analysis to determine the applicability of the storage or the acceptable graphite processing techniques for the following disposal in each particular case.

This approach provides a systematic comparison of options from which decisions may subsequently be made, and its main considerations are:

- risks to the workforce at nuclear sites;
- environmental risks (both in terms of damage to the natural environment and harm to the public);
- costs and social effects.

The justification of the conservation scenario was made taking into consideration if the industrial uranium-graphite reactors in the Russian Federation were placed near the existing radioactive waste disposal and burial site locations developed in the course of the former defense programs, as well as if the graphite stack of a reactor was situated below the ground level.

It should be mentioned also that the physical form of the reactor graphite itself is the most compact, and any processing of the irradiated reactor graphite gives rise to the increase in the radioactive waste volume, changes the aggregate state of the waste and increases associated risks and cost.

Therefore the long-term storage of the reactor graphite in the purpose designed repositories may at first glance seem to be one of the simplest and less expensive ways of handling it. However, this solution may cause a number of the environmental and economical problems.

This approach makes possible an objective comparison of different scenarios taking into account the associated risks and the necessary allocation of funds.

To date, various methods of irradiated graphite reprocessing have been proposed. They are based on the application of different physical and chemical processes, most of which involve incineration of the entire mass of graphite. Indeed where possible, incineration provides very high volume reduction. Depending on the activity of the ash residue, it may be simply packaged for storage and/or disposal or a further immobilization step may be required.

However, they all have disadvantages, and do not solve the whole complex of problems.

Radioactive waste (including graphite) incineration is a well developed though fairly complex technology. The equipment is sophisticated, particularly for off-gas systems which can include afterburners, heat exchangers, cold air injection, filter candles, bag houses, scrubbers and HEP A filters. This means that adoption of this process requires a substantial capital outlay, and operating and maintenance costs are likely to be high.

If graphite is processed by the incineration, the waste volume reduction becomes impossible on account of the ^{14}C isotope present in the irradiated graphite in the amount of 10^8-10^9 Bq/kg; for obvious reasons the latter cannot be removed into the atmosphere. If lime is used for CO_2 absorption, 1 m^3 (2.2 ton) of burned graphite gives 18.3 ton of calcium carbonate, which is almost an order of magnitude more than the initial graphite mass.

It is our opinion there is a possibility to find more simple technologies as compared to the graphite incineration providing the necessary decontamination of graphite and the conversion of it into a less hazardous waste category followed by the disposal of it in a subsurface repository.

The proposed solution consists in the maximum reduction of the solid waste formed, namely, the processed graphite itself and the volume of the secondary waste containing ^{14}C radionuclide in the calcium carbonate form.

The application of the method under discussion is advantageous as well if the graphite to be processed, along with the ^{14}C, ^{3}H, and ^{36}Cl activation products, is contaminated by TRU and other radionuclides from the nuclear fuel and construction materials as a result of an emergency situation.

According to the proposed technique, the result is achieved by means of the procedures listed below.

First the graphite is treated by chemical reagents to destroy its surface layer followed by the removal of the product containing the radionuclide contaminants from the graphite surface. The removal of the thin surface layer makes an effective decontamination, and the bulk radionuclide mass is removed as a separate product.

The next stage assumes the thermal treatment of the graphite at the temperature of 700÷800^{o}C.^{14}C radionuclide is immobilized by means of absorbing the yielded carbon dioxide by a suitable absorbing agent, e.g. calcium hydroxide solution.

In Fig 1 the graphite samples after the thermal treatment at 700^{o}C are shown. One of them was preprocessed before heating.

A B

Fig. 1. The appearance of granite samples after heat treatment (700^{0}**C),** A **–** treated beforehand with oxidizing agent**,** and B **-** is not subjected pretreatment.

Unlike the single-stage incineration process, the proposed technique provides conditions for the following graphite treatment, when ^{14}C and the activation products (^{60}Co and others) are accumulated in the separate product of the minimum volume; the incineration process is performed at the lower temperature. Partitioning of the radionuclide contaminants becomes possible; they can be processed and stored separately of the bulk graphite mass.

Among the other advantages of the technique it is worth to mention the simplified off-gas treatment, the increased equipment service life, and the higher personnel safety in the course of the decommissioning works. When implemented, the technique results in less amount of the secondary waste. The secondary waste in the case means the off-gas scrubber solutions and the solid waste, namely, metal-ceramic or cloth filters to be replaced and utilized.

This approach to the reactor graphite handling issue is unparalleled in the world’s practice, and the results obtained in the experimental research enabled to file an application for a patent [6].

The second option to change the reprocessing technology is the idea to use reactor graphite as the starting material to produce stable final waste form compositions intended for the long-term storage of radioactive waste including transuranic elements.

Graphite is going to provide the high chemical stability and high thermal conductivity of the material composition, that in turn guarantee the safety of radwaste storage for an unlimited period of time.

The technique eliminates the need of separate operations of the irradiated graphite processing, since it is in demand at that very object where the decommissioning activities are under way.

Application of the proposed approach gives an appreciable economic effect due to the less costs recognized as expenses for the container packaging and the new repository construction.

Summarizing all the above said, the concept proposed for consideration is capable to simultaneously solve two actual tasks as follows:

- process the accumulated radioactive waste by means of incorporating it into the stable matrices;
- use the irradiated graphite without preprocessing on site where the reactor facility decommissioning activities are under way.

Application of the proposed approach gives an appreciable economic effect due to the less costs recognized as expenses for the container packaging and the new repository construction.

The preparation of stable graphite based compositions for the radioactive waste immobilization is not an exclusive option for the reactor graphite utilization. There are reported techniques for the preparation of the heat-expanded graphite possessing the low bulk density and the high adsorption capacity. Those sorbents may just as well be used for the immobilization of the liquid radioactive waste at that very object where the decommissioning activities are under way.

It is worth to indicate one more promising direction of the possible reactor graphite application the economic assessment being very difficult to make. It is referred to the application of the reactor graphite for the fabrication of waste disposal containers. Another way is to use an unclaimed part of the reactor graphite for the construction of engineering barriers and structural elements of the radioactive repositories themselves.

CONCLUSIONS

The ultimate goal of any R&D work for an upgrade of any waste treatment technology including the reactor graphite processing is the maximum possible reduction of the final waste form volume and associated costs – the less the volume, the less the expenses for the transportation, long term storage, and disposal.

Despite the extensive studies performed in the area, neither of the options available to date prevails over the others.

The search for new advanced methods of graphite processing is made to choose the less expensive technology meeting the requirement of the ultimate isolation from the biosphere of all radionuclides present in the irradiated graphite. The choice of the graphite handling technique may be individual for each selected object of storage or disposal and for each given country. And the final decision on that choice is not to be made until after the technical assessment and the feasibility study of all the options available.

The efficient reactor graphite handling technology has to be chosen with regard to the specific activity level and the spectrum of radionuclides present. The ultimate solution depends on the applicability of either storage or processing.

The application of the partitioning principle enables to segregate the most contaminated fragments; and those after the simple decontamination procedures might be converted into the low-level radioactive waste category. This approach provides the many-fold reduction of the load born by the equipment targeted at the decontamination of the most "hot" part of the partitioned graphite, and in the long run will minimize the volume of all the secondary waste.

The utilization of the irradiated graphite as a starting material for the fabrication of compositions or items (containers) intended for the long-term storage of other radioactive waste could be considered as an alternative option for irradiated graphite handling.

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