MANAGEMENT OF RADIOACTIVE WASTE IN NUCLEAR POWER: HANDLING OF IRRADIATED GRAPHITE FROM WATER-COOLED GRAPHITE REACTORS

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INTRODUCTION

One of the topical issues of nuclear power today is decommissioning of power, production and research nuclear reactors with expired lifetime. In addition to technical, economic and social matters decommissioning involves ecologically safe confining of radioactive waste (radwaste) being generated, in particular, during dismantling of highly radioactive reactor components.

The efforts in the area of radwaste handling technology include ecological safe handling of irradiated graphite during decommissioning of water-cooled graphite-moderated channel reactors (WGR), namely: AM at the First NPP, AMB-100 and AMB-200 at the first stage of Beloyarsk NPP, RBMK-1000 at Leningrad, Smolensk and Kursk NPP, production and research reactors where graphite is used as moderator.

AN OVERVIEW OF IRRADIATED GRAPHITE HANDLING PROBLEM

As a result of decommissioning of water-cooled graphite-moderated reactors, a large amount of radwaste in the form of graphite stack fragments is generated (on average 1500-2000 tons per reactor). That is why, it is essentially important, though complex from the technical point of view, to develop advanced technologies based on up-to-date remotely-controlled systems for unmanned dismantling of graphite stack containing high-active long-lived radionuclides and for conditioning of irradiated graphite (IG) for the purposes of transportation and subsequent long-time and ecologically safe storage either on NPP sites or in special-purpose geological repositories.

Since 1988 the enterprises of RF Minatom have conducted R&D activities on investigation and evaluation of radiation levels in the graphite stacks of channel reactors. The attempts have been made for development of IG handling technologies.

Radioactivity of reactor graphite stacks at the time of power unit shutdown for decommissioning is caused by the following factors:

- activation of graphite and impurities containing therein as a result of neutron irradiation;
- graphite contamination with the activation products of purging gas circulated in the stack;
- graphite contamination with radionuclides in case of coolant leaks into the stack;
- graphite contamination with non-radioactive products in case of coolant-to-stack leaks and subsequent activation of these products with neutrons;
- graphite contamination with fuel composition and fission products in case of accidents involving fuel damage;
- graphite contamination with radioactive products of corrosion and erosion of the structures in case of reciprocal contact of graphite and the structural elements such as pressure tubes, rods, supports, etc.

So far it is not possible to provide reliable predictions of reactor graphite radioactivity after its long-time irradiation during the reactor operation. The comprehensive studies are required for evaluation of graphite radioactivity both by calculations and experiments. Available techniques developed for this purpose are based on sampling the graphite, measuring radionuclide activity and assessment of graphite activity in the whole stack. However, one shall be very careful when extrapolating the results obtained at one reactor to another reactor, since the reactor operating conditions as well as composition of impurities in graphite may differ considerably.

MAIN RADIATION AND NUCLEAR HAZARDOUS CHARACTERISTICS OF THE GRAPHITE STACK (UNITS 1 AND 2 OF BELOYARSK NPP TAKEN AS ILLUSTRATIVE EXAMPLES)

The main characteristics critical for radiation and nuclear hazards of the graphite stack are as follows:

• graphite stack is contaminated with nuclear fuel got there as a result of the accidents;

• graphite mass is 992 tons, total activity – $6x10^4$ Ci (at the time of unit shutdown);

• fuel mass in the reactor stack amounts to 100-140 kg, as estimated by IPPE and RDIPE, respectively;

• γ-radiation dose rate in the stack cells varies from 4 to 4300 R/h, prevailing values being in the range from 50 to 100 R/h.

TRADITIONAL WAYS OF RADWASTE HANDLING

1. IG conditioning

In general case, radwaste conditioning means waste processing so as to achieve pre-determined properties, parameters, characteristics or a new state. Typical requirements to radwaste conditioning include reduction in volume, dehumidification (dry material), formation of solid radwaste (SRW) from liquid radwaste, fractionation, i.e. separation of high-active waste from low-active waste, etc.

2. Bituminization

Bituminization technology for radwaste processing is used in many countries with well-developed nuclear power. This technology is especially useful in such countries where dry territories suitable for radwaste storage (e.g., deserts, dry sands, etc.) are unavailable. This technology allows to make bitumen blocks that can be stored in ground and in other places not protected from underground water [1].

The main advantage of bituminization is more efficient protection against radionuclide transfer into the environment as compared to other methods of radwaste processing (for instance, cementing), other things being equal. Water resistance of bitumen compounds is high and is characterized by leaching rate from $1x10^{-6}$ to $1x10^{-4}$ g/cm^2 day.

3 Cementing

Cementing technology for processing low-active radwaste is popular in the countries where dry solids and beds free from underground water are available.

Cementing is carried out at ambient temperature and with the use of simple equipment. The main disadvantage of cementing technology is high leaching rate of radionuclides (by two orders of magnitude higher than for bituminization), other things being equal. Another significant factor is the presence in cement of water molecules, groups HO^-, OH^- and other radioactively unstable components whose radiolytic decomposition leads to generation of gaseous products, including hydrogen.

4. Confining in polymeric matrix

In recent years the investigations conducted in some countries were focused on exploration of synthetic resins and polymers as the possible binders for graphite radwaste [2].

In respect to IR handling technology, the end product will have the following characteristics:

• resistance to irradiation, no less than 10^{10} rad;

• resistance to high temperature;

• heat resistance;

• resistance to leaching in natural, distilled and sea water corresponds to leaching rate of bitumen compounds;

• high mechanical strength, for instance, compression strength is over 700 kg/cm^2. Though cost of synthetic resins is higher than cost of cement and bitumen, this is not a prevailing factor when the cost of graphite radwaste processing is under consideration, since the volume of the storage building can be reduced (because of smaller volume of containers with conditioned IG) and auxiliary equipment is simple.

5. On IG disposal by means of incineration

Incineration of IG has an important advantage over any other methods of graphite handling after dismantling, because the amount of radwaste intended for disposal will be significantly reduced as a result of incineration. Thus, thousands tons and cubic meters of IG can be substituted with low-mass and low-volume radwaste in the form of ash which are delivered for conditioning and disposal.

However, there are limitations to the use of incineration technology in the industrial scale, namely:

• release of radioactive ^{14}C and other radionuclides in the atmosphere;

• incomplete purification of CO_2 from radioactive impurities during incineration.

These limitations come from the environmental concerns. However, a large portion of irradiated graphite could be incinerated. This covers IG used as neutron reflector in WGR. In this graphite the content of ^{14}C and other radionuclides is by an order of magnitude smaller than for IG used in the reactor core. Besides, in the reflector graphite there are practically no fuel spills. In case of WGR mass of the reflector graphite is approximately equal to that of the reactor core graphite with account for a large number of pressure tube penetrations in the reactor core stack.

ALTERNATIVES FOR THE PROBLEM SOLUTION, THEIR BRIEF DESCRIPTION AND MAIN CHARACTERISTICS

1. Current status of IG handling problem in Russia

As commonly accepted in Russia and abroad, the graphite stacks of gas- and water-cooled graphite-moderated reactors planned for decommissioning shall be preliminary cooled during 50 to 100 years in the reactor cavity isolated from the environment. Priority of this concept relegates to the background the efforts on development of cost-efficient technologies for safe handling of IG. As a result, technology of IG handling mastered in the industrial scale is not available in Russia today. At the same time the practical solutions for the so-called sleeve graphite handling problem are demanded now both in Russia and abroad. For example, about 4 thousand tones of irradiated sleeve graphite which have to be disposed after preliminary processing were accumulated in France for six types of reactors at the Chinon, St Laurent and Bugey sites. Only three production WGR in Russian Sibirsky integrated chemical works accumulated about 5 thousand tones of graphite sleeves removed from the reactor stack in the course of their re-tubing after in-pile operation.

In terms of IG handling technology two lines were identified: long-term storage of conditioned IG and IG disposal by means of incineration.

2. Graphite processing by impregnation with solidifying polymeric compounds

As shown by the estimation of activity of IG from decommissioned WGR stacks, irradiated graphite can not be placed in the available shallow ground repositories without additional processing. It is needed for reliable confining of radionuclides in the graphite blocks, prevention of radionuclide transfer to the environment. This can be achieved by impregnation of a graphite block with highly radiation resistant solidifying polymeric compounds.

RDIPE has developed proposals on graphite handling for AMB-100 reactor at Beloyarsk NPP. RDIPE proposals can set the basis for development of enlarged production process with the following stages: dismantling of the graphite stack, immobilization of IG in radiation-resistant polymeric matrix and its placing in shielding casks for subsequent transportation to the site of long-term storage and/or disposal.

This method envisages the following sequence of operations: graphite blocks are evacuated in the sealed volume at room temperature. Liquid polymeric compound is then supplied under pressure; macropores of the graphite blocks are filled and solidified resin provides safe confining of radionuclides in graphite. Impregnation is made concurrently with pouring the graphite blocks in the transfer cask (i.e. a container loaded with IR and used in the initial stage of processing) which shall be removed from the reactor cavity in the course of graphite stack dismantling.

Interim (buffer) storage of the transfer casks with conditioned IR prior to their transportation to the design long-term storage is required so as to give time for harmonization of the schedules of stack dismantling, the number of casks with conditioned IG and the schedule for cask delivery to the design long-term storage.

The casks with conditioned IG shall be stored in a special-purpose design storage facility where long-term environmentally safe storage (disposal) conditions can be provided.

3. Technology for handling IG from RBMK reactors

VNIPIET (All-Russian Research and Design Institute for Power Technology) has developed the technology for conditioning irradiated graphite GR-280 to be applied in the industrial scale during decommissioning of RBMK NPP power units.

In accordance with the technology, RBMK IG can be conditioned by means of inorganic binder added to concrete (slag-stone) containers so as to form monolithic stmctures. Strength of solidified product obtained by this technology shall be no less than 5 MPa and leaching rate no more than $1\text{-}10^{-3}$ cm/day.

Handling of irradiated graphite includes the following stages and operations:

- removal of stack IG from the reactor cavity and placing it into the transfer cask which is unloaded from the bottom;
- transportation of the transfer cask with IG to radwaste conditioning facility at the NPP site,
- IG reloading from the transfer cask to one-trip shielding cask and filling the empty volume in this cask (making the monolithic structure);
- installation of a cover and making the one-trip shielding cask air-tight, decontamination (if required) and radiation monitoring of its surface;
- examination of radiation parameters, issuing the certificate to the cask with IG;
- temporary (buffer) storage of casks with IG at the NPP site,
- assembling of package sets with IG;
- transportation of package sets to radwaste disposal site;
- long-time underground storage of shielding casks in geological repository;
- sending the containers for interim storage.

CONCLUSION

Specific cost of graphite immobilization in radiation-resistant polymeric matrix amounts to ~ 2600 USD per 1 t of graphite, whereas specific cost of immobilization in slag-stone containers with inorganic binder (cement) is ~ 1400 USD per 1 t of graphite. On the other hand, volume of conditioned IG radwaste subject for disposal, if obtained by means of the first technology, is 2-2.5 times less than the volume of radwaste generated by means of the second technology. Besides, duration of radwaste processing, including IG conditioning, package sets transportation and disposal is much shorter, if performed in accordance with the first technology. Thus, the final choice of graphite immobilization technology shall be made in the course of a feasibility study on decommissioning of water-cooled graphite-moderated reactors and with account for the strategy of father graphite radwaste handling in the region of NPP location.

INFERENCE

It can be concluded from the above that advanced methods for graphite radwaste handling are available nowadays. Implementation of these methods will allow to enhance environmental safety of nuclear power that will benefit its progress in the future.

REFERENCES

[1]. Harell, V., Krause, H. / *Bituminization of radwaste in Karlsruhe Nuclear Research Center. Experience in operation of the facility and related investigations.* /Proceedings of the seminar on bituminization of low- and medium-active radwaste. Antwerp, 1976.

[2]. Fitzgerald, Ch. et al. /A possibility of radwaste introduction into bitumen and polyethylene. *Atomnaya tekhnika za rubezhom*, 1971, No.8,, p. 27-34 (in Russian).