

RADIOLOGICAL CHARACTERIZATION ACTIVITIES DURING THE PARTIAL DISMANTLING OF THE IRT – SOFIA RESEARCH REACTOR FACILITIES

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A radiological characterization methodology has been developed and implemented for assessment of radiological parameters during the partial dismantling of IRT – 2000 research reactor facilities at the *Nuclear Scientific Experimental and Educational Centre* (NSEEC). To assess the radiological situation in the places of dismantling activity a number of measurements were carried out prior and during the dismantling activities and after their accomplishment. Samples of different kind were taken and analyzed to provide a reliable database of information on quantities, type and distribution of radionuclides, comprising activation or contamination products, generated during the operation of the reactor.

Key words: radiological characterization, research reactor, partial dismantling.

1. INTRODUCTION

Radiological Characterization of the nuclear reactor facilities is the process of defining the quantities, type and the places of location of radionuclides, comprising activation or contamination products, generated by neutron activation of materials during the reactor operation. Generally, the radiological characterization of the facilities involves a survey of existing data, calculations, *in situ* measurements, sampling and analyses, and final radiological survey. According to the requirements for characterization of radiological and other hazards in nuclear facilities recommended in the IAEA Safety Standards WS-R-5 [1] for preparation of the final plan of partial dismantling, the extent and type of radioactive material (irradiated and contaminated structures and components) at the facilities shall be determined by means of a detailed characterization survey and on the basis of records collected during the operational period.

The research reactor IRT-2000 in Sofia operated in the past at 2 MW for more than 28 years is undergoing major reconstruction aimed to its conversion to a low-power reactor in accordance with a decision of the Bulgarian Government. In

this regard after preliminary radiological characterization (2000–2004) and delivery of the *Spent Nuclear Fuel* (SNF) back to the country of origin in 2008, during the period 2009–2010 a partial dismantling of the reactor equipment has been performed. The associated activities have been carried out in the following sequence:

- Removal of reactor internal systems;
- Removal of peripheral systems;
- Preservation of biological shielding and the aluminium cladding of the reactor pool;
- Preservation of the reactor building for further use.

All the activities during the partial dismantling of equipment of the IRT-2000 reactor have been conducted safely following the procedures described in a previously prepared detailed *Dismantling Plan* (DP) in accordance with the regulations and observing the ALARA principle.

The radionuclides generated in the course of nuclear reactor operation are the sources for different streams of radioactive waste (RAW). Depending on their origin these radionuclides can be distributed over two groups: fission and neutron activation products. The fission products comprise a complex mixture of radionuclides of different chemical elements, including noble gases, which have different physical and chemical properties and different behaviour in the fuel and radioactive media of the nuclear reactor. The activation products, which have the main contribution to the generated RAW, are coupled predominantly with the behaviour of the construction materials (including their corrosion products) during neutron irradiation.

The deposition of fission and activation products in the coolant of the *Primary Circulation Loop* (PCL) within the reactor vessel and the reactor internal devices can be called conditionally primary contamination. All other contamination of equipment is formed as a result of the migration and redistribution of the radionuclides *via* different mechanisms – dissolution and crystallization, evaporation and condensation, sorption, diffusion and chemical interactions. The rate of the processes and the degree to which they take place depend on the physicochemical properties of the transport media and the boundary surfaces and also on the ambient conditions (most of all on the temperature). This contamination is denoted as secondary contamination.

The primary contamination is volume contamination since it spreads to some degree or other in the entire volume of the water. The secondary contamination can be conditionally divided into volume and surface contamination. Typical surface contaminated materials are metals (when there are no activated constructive elements), glass, some non-porous polymers, and surfaces with protective covering layers. The surface contamination can be conditionally divided into fixed (non-removable) and unfixable (removable).

Radioactively contaminated objects, which can no longer be used for their purpose and at the same time are contaminated with radionuclides above the norms of regulation (exemption levels), are classified as RAW.

The aim of this work is to present the experience of IRT professionals in the field of radiological characterization activities during the partial dismantling of the research reactor IRT-2000 facilities.

2. EXPERIMENTAL WORK

The radiological characterization of the premises and facilities of IRT-2000 reactor was made in accordance with the Bulgarian legislation as well as the IAEA safety standards and recommendations [1–8]. A number of documents such as *Methodology for radiological characterization of structures, systems and components in the partial dismantling*; *Methodology for sampling of nuclear facility's structures, systems and components*, programs for radiological investigation of *Primary Circulation Loop (PCL)*, *Reactor Pool (RP)* and *Reactor Hall (RH)* facilities etc. were developed well in advance.

The characterization of the reactor facilities was made before the partial dismantling operations, during the course of different stages of activities and after the end of entire work.

2.1. SAMPLING

The main radionuclide in a nuclear reactor that dominates within the timescale of the first 50 years is ^{60}Co , being the main source of gamma radiation. For longer periods, the dominating radionuclides are ^{59}Ni and ^{14}C since their lifetimes are greater than 100 years. The main radionuclide responsible for the dose exposure from the pipelines is ^{60}Co . The materials obtained during the implementation of dismantling of the IRT-2000 reactor systems are separated mainly into:

- Activated materials due to the impact of the neutron flux from the reactor core to which they have been exposed during the operation;
- Radioactive contaminated materials;
- Toxic materials;
- Non radioactive wastes.

To assess the radiological situation at the places of dismantling activity a number of measurements were carried out. Different type of samples were taken and analyzed to provide a reliable database of information on the quantity and type of radionuclides and their distribution:

- Smears (dry or wet) – for assessment of fixed or non-fixed contamination and determination of specific activity of radionuclides;
- Material samples (solid or liquid) – metal segments from reactor systems; concrete; steel; glass, flakes from the inner surface, graphite of the TC, personal means of protection etc.;
- Water samples – from reactor coolant facilities.

Sampling points, number of samples, sample materials and analyses were discussed and described in *Methodology for radiological characterization of constructions, systems and components from partial dismantling of IRT-2000*.

Sampling places were determined after preliminary measurements of surface contamination of the corresponding facilities. Samples were taken from reactor pool internals (ejector, plate above the core, pipelines, experimental channels etc.), primary circulation loop (heat exchangers, rotary pumps etc.), spent fuel storage pool, control and safety system (control rods, channel of automatic regulator, ionization chambers etc.), reactor hall facilities etc.

The sample collection was performed carefully (at a distance) by use of special devices to ensure the maximal radiation protection of the operators. Metal segments were taken by means of cutting equipment and were put into labeled hermetic plastic bags. Liquid samples were taken by special plastic devices and were put into labeled hermetic glass bottles. The samples investigated for C^{14} content were packed in plastic vessels.

All the necessary information and parameters of the collected samples were noted in a special document/logbook. The document contains the full description of: sample type; sample weight; sampling location; time of sampling; sampling technique; method for analyses, results etc.

2.2. METHODS

Measurements of surface contamination were made with portable dosimetric equipment (Figure 1): a radiometer FH40G-L10 for alpha, beta and gamma contamination (ZnS scintillation detector with a total area of 100 cm^2 ; efficiency for ^{241}Am of 40 %, for ^{60}Co of 17 % and for $^{90}\text{Sr}/^{90}\text{Y}$ of 56 %), a portable device for surface alpha and beta contamination BERTHOLD LB 1210 C (gas flow proportional counter MZ 100 with efficiency for ^{241}Am of 22 % and possibility for ^{14}C detection); a portable gamma spectrometer of type IDENTIFINDER and a portable device MICROCONT 2 for alpha and beta surface contamination.

Sample activity characterization were carried out by Gamma spectrometry (including ^{137}Cs , ^{60}Co , ^{152}Eu), *Liquid Scintillation Spectrometry* - LSS (^{14}C , ^{90}Sr) and Total alpha-beta counting.



Fig. 1 – Portable dosimetric equipment – IDENTIFINDER, radiometer FH40G-L10 and MICROCONT.

The gamma spectrometry measurements were carried out using HPGe – GMX 50P4 coaxial detector with a Beryllium window (Ortec type) with 50 % counting efficiency and energy resolution 2.3 KeV at 1332 KeV (^{60}Co). For efficiency calibration of the detector certified calibration standards (standard geometries – Marinelli-1000, Marinelli-450 and TB-50) with wide energy range purchased from Amersham were prepared in matrix materials of different densities. The sample measuring time was between 12 and 48 hours. The obtained spectra were processed with the computer code WINNER. The ^{137}Cs specific activity was measured by its main γ - line of 661.66 keV. The specific activity of ^{60}Co was obtained using the lines of 1173.24 and 1332.50 keV. ^{152}Eu was determined from the lines of 121.78, 344.280, 443.980, 778.900, 964.130 and 1408.010 keV.

^{14}C content was measured by Liquid Scintillation Counting on a PERKIN ELMER - Tri CARB analyzer. Samples were prepared according the standard methodology of the French national organization for standardization AFNOR - AFNOR NF M60-320 standard (chemical destruction with distillation; burning in tubular furnace). ^{14}C content was obtained using the peak of 156 keV (E_{max} , measurement time 7200 s, INSTA SCINT GEL PLUS scintillation cocktail, standard materials - SRS 74360-745(Qty 3.779 E3 Bq). The applied radiochemical separation technique is effective and useful but it can not easily be used for analysis of large numbers of samples due to a long experimental time required.

The total alpha-beta activity of the samples was measured by MPS-9406-ultralowlevel alpha-beta-counter. The smear swabs (wet and dry) were measured directly - without any pretreatment of the samples.

Total beta activity of liquid samples was determined using universal measuring device UVJ-01 (MK-30 Measuring Chamber) with a Geiger detector (0.23 cps/Bq efficiency for $\text{Sr}^{90}/\text{Y}^{90}$) in lead shielding (background ≤ 2 cps).

2.3. PRELIMINARY RADIOLOGICAL CHARACTERIZATION

For specifying the type, quantity and location of contamination products, including corrosion, erosion and fission products deposited on the contaminated

surfaces as well as for determination of specific and total activity for the materials activated by the neutron flux on the reactor vessel, the internal reactor facilities and the concrete of the biological shielding, all the facilities were assessed preliminary by the following main activities:

- Examination and analysis of all records and data from the history of IRT-2000 operation;
- Selection, development and implementation of a method for calculation of component activation;
- Development of a plan for measurements and sampling;
- Execution of the measurements, sampling and smears;
- Discussion of the obtained data;
- Comparison of the calculated data with the experimental results.

To assess the RP the measurements were carried out in full of water pool, by a hermetic detector and after emptying of the pool – by direct measurements through the protective cabin. A number of samples (material samples and smears) were taken from the *Automatic Regulation (AR) rod* (3 tube sectors from the channel with 2 cm length were cut by hack-saw – at around 30 cm from the channel bottom); *vertical channel of Manual Regulator (MR) /north/* (3 tube sectors from the channel with 2 cm length were cut by hack-saw at a distance of 30 cm from the channel base); *cover of one of the vertical experimental channels /west/* (2 shaving pieces were taken from the surface by means of chisel and hammer); *cover steel hasps* (part of it, around 1 cm); *two preliminary selected graphite assemblies from the east side of the reactor core* (Assemblies were withdrawn by means of manipulator from the reactor core and measurements of their activity were carried out above the water level) etc. All the samples were put into labeled hermetic plastic bags and analyzed.

Additionally, a graphite block from the TC inside it was taken out. The block was there during the whole period of operation of IRT-2000. It was taken a sample from the graphite block for gamma spectrometry analysis. Samples (material samples and smears) were taken also from the reactor pool equipment.

The calculations, being a part of preliminary characterization activities, were realized according to the IAEA recommendations, which have the goal to ensure the quality and reliability of the results. The analysis was based on the original documents, which contain: data of the reactor system geometry; reactor core geometry and fuel loading; characteristics of the nuclear fuel (geometry and material composition of the fuel assemblies); detailed description of the constructing elements that are liable to be dismantled (geometry, material composition and mass); operational history of the reactor; data for the neutron flux; literary sources and reference books as regards the neutron interaction cross-sections for the elemental composition of the reactor system components.

2.4. RADIOLOGICAL CHARACTERIZATION DURING DISMANTLING ACTIVITIES

For radiological characterization during dismantling activities were collected about 300 smears (dry or wet) and 100 material samples (solid or liquid) - metal segments from reactor systems; concrete; steel; glass, flakes from the inner surface, graphite of the TC, personal means of protection etc.

Sampling places were determined after preliminary measurements of surface contamination of the facilities and associated areas. Samples were taken from RP facilities (ejector, plate above the core, pipelines, experimental channels etc.), PCL facilities (heat exchangers, rotary pumps etc.), spent fuel storage pool, management and protection system (MPS) equipment (ruling rod of MPS, channel of AR, ionization chambers etc.), reactor hall facilities etc. The additional samples were collected from the facilities where the measured radiation doses were higher than the permissible limits determined by the basic norms of radiation protection.

2.5. FINAL RADIOLOGICAL SURVEY

The definitive radiological survey was carried out according to final characterization plan. Detailed procedures were developed during the work progress. Sampling was carried out at different points on a sampling grid covering entire investigated areas (walls, floors, ceilings, equipment surfaces etc.). All surfaces were separated into small parts marked A, B, C, D etc. and each part was investigated by means of portable dosimetry and radiometry devices. The sample points were determined after surface contamination measurements. Material samples and smears were taken from the places with measured values higher than the permissible limits.

3. RESULTS AND DISCUSSION

3.1. PRELIMINARY RADIOLOGICAL CHARACTERIZATION

The results of the preliminary characterization served as an initial basis for specifying the type of the technique implemented in the dismantling, instruments and devices (remote, semi-remote, manual methods), the necessity of decontamination and the choice of appropriate methods, assurance of radiation protection of the employees and the environment, the RAW classification after the dismantling, the requirements for manipulation (treatment, transportation and storage of the waste) during the dismantling, dose exposure, assessment of the risk, assessment and selection of a scenario with the purpose of assuring conformity with the ALARA principle, the kind of the protective means (devices for the employees carrying out the dismantling) etc.

Some of the results obtained during the preliminary characterization of IRT-2000 facilities are given in Tables 1–4.

Measurements of the AR rod activity were carried out while it was slowly withdrawn up from the vertical channel № 17. After that the AR channel was dismantled and withdrawn, left to drain out from water, and was laid on a plastic sheet on the platform. Its activity was measured at the bottom part with a dosimeter device. The measured value of gamma dose rate in the most active part at a distance of 0.1 m (at 30 cm from its bottom part) was under 80 $\mu\text{Sv/h}$.

Table 1

Results from automatic regulation rod measurements

Distance, [m]	Measured activity [$\mu\text{Sv}\cdot\text{h}^{-1}$]
4.0	15
3.0	20
2.0	55
1.0	300
Withdrawn upper part at the channel level	600

The vertical channel MR /north/ was dismantled released from the fixation to the reactor platform and was taken on the platform. The measured value did not exceed 150 $\mu\text{Sv/h}$ in its most active part (at 30 cm from its bottom part), at a distance from the detector of 0.1 m.

The data from the gamma spectrometric analyses of material samples (tube sectors from the AR and MR channels) is given in Table 2.

Table 2

Data from gamma spectrometric measurements of metal samples - automatic regulation rod (AR); vertical channel of Manual regulator (MR), [$\text{MBq}\cdot\text{kg}^{-1}$]

Sample	Eu-154	Cs-137	Co-60
AR 1.1	2.65 ± 0.21	0.68 ± 0.05	61 ± 5
AR 1.2	2.81 ± 0.22	0.27 ± 0.03	70 ± 6
AR 1.3	2.84 ± 0.22	0.39 ± 0.04	62 ± 6
MR 1.1	2.9 ± 0.3	0.48 ± 0.05	58 ± 5
MR 1.2	2.6 ± 0.2	0.38 ± 0.03	54 ± 6
MR 1.3	2.5 ± 0.2	0.38 ± 0.03	64 ± 6

The data obtained from analyses of graphite blocks (from the TC inside) are presented in Tables 3 and 4.

Table 3

Results from measurements of two graphite blocks, [$\mu\text{Sv}\cdot\text{h}^{-1}$]

Assembly 1	Measured activity	Assembly 2	Measured activity
Upper end	10	Upper end	6
Middle	110	Middle	120
Bottom end	180	Bottom end	160

Table 4

Data from gamma spectrometric measurements of the graphite block, [$\text{Bq}\cdot\text{kg}^{-1}$]

Sample	Mass, [g]	Eu-152	Co-60
Graphite	233	130 ± 11	650 ± 60

3.2. RADIOLOGICAL CHARACTERIZATION DURING THE PARTIAL DISMANTLING

Radiological characterization during dismantling activities gives information on the type and quantity of radioactive materials arising from the dismantling operations. Some of the analyses were performed directly on the sample material while others were made after preliminary sample preparation. Data were obtained for all dismantled facilities - PCL (premise 103), RP, reactor hall facilities etc. Results obtained from gamma spectrometry measurements of pipelines, material samples from biological shield, shutter block (TC), different metal details, wall and floor materials collected from dismantled equipment are given in Figure 2.

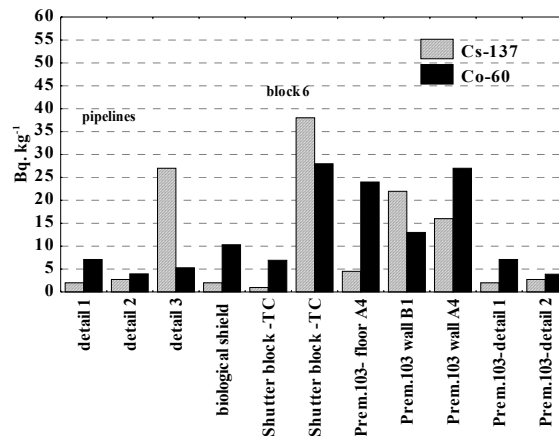


Fig. 2 – Results from gamma spectrometry measurement of the material samples.

The ^{137}Cs and ^{60}Co specific activities of all studied samples vary between 2 and $60 \text{ Bq}\cdot\text{kg}^{-1}$ except for samples collected from two areas at the floor of premise

103 (PCL) where the measured value for ^{137}Cs was 362 Bq.kg^{-1} and 2.62 KBq.kg^{-1} for ^{60}Co . The mean ^{137}Cs and ^{60}Co specific activities obtained for concrete, wall and floor collected at the PCL are presented in Table 5.

Table 5

Data for material samples taking during the partial dismantling activity, [Bq.kg^{-1}]

Sample	Cs-137	Co-60
S-1 (concrete)	2.6 ± 0.2	$8.6 \pm 0,7$
S-2 (concrete)	20.3 ± 1.8	59.5 ± 5.4
S-3 (PCL walls)	19.0 ± 1.3	20.0 ± 1.6
S-4 (PCL floor)	4.5 ± 0.4	24.0 ± 1.8

Data obtained for the graphite material collected from the TC (obtained by chemical destruction with distillation technique and by burning procedure) show a high specific activity of ^{14}C . In Table 6 are displayed data for measured ^{14}C content in four graphite material samples collected from the TC.

Table 6

Results obtained for the graphite material collected from the Thermal column

Sample	Applied procedure	^{14}C
1	chemical destruction with distillation, 0.1 g	55600
2	chemical destruction with distillation, 0.1 g	55200
3	burning in tubular furnace 1 g	59900
4	burning in tubular furnace 1g	60600

All the other results for investigated samples (smears and materials) are low and showed the lack of surface contamination. Only few contaminated details were detected during the whole operation period. Contaminated details were treated chemically. New samples were collected and analyzed after decontamination treatment. Contaminated materials after decontamination were removed and treated as a radioactive waste.

3.3. FINAL RADIOLOGICAL SURVEY

About 30 smears and 10 material samples collected from walls, floor, ceiling and equipment which were not liable to dismantling were analyzed to evaluate the radiation status of PCL. The results show the lack of contamination (Table 7).

Table 7

Data for total alpha- and beta- activity measured in smears collected at PCL facilities, [Bq.cm⁻²]

Sample	Total alpha activity	Total beta activity
1	2.87 E-4	9.5 E-3
2	1.24 E-4	2.09 E-2
3	1.61 E-4	5.57 E-3
4	1.23 E-4	2.68 E-3
5	7.33 E-5	1.74 E-3
6	4.95 E-5	1.81 E-3
7	8.60 E-4	9.2 E-3
8	7.83 E-5	8.03 E-3

Radiation status of RP facilities was evaluated by analyzing of about 50 smears and 2 material samples. Smears were collected from the RP surfaces (bottom, walls, channels and details). Some of the data are presented in Figure 3. RP facilities material samples (Al alloy) were taken from the lead plate cover. The results showed the lack of contamination except in 2 points of the floor. Contaminated materials were decontaminated or removed and classified as RAW.

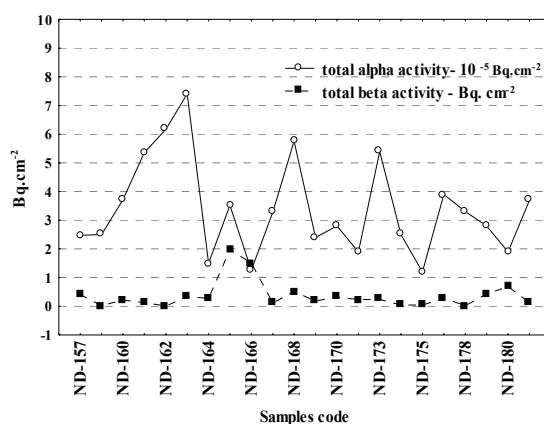


Fig. 3 – Total alpha- and beta- activity in smears collected from RP facilities.

Final characterization of the reactor hall was made by dividing the hall of 7 sectors (ceiling, floor, walls, biological shielding surfaces, etc.). Each of these sectors was separated into small parts to complete the sampling grid. During the radiological survey were analyzed about 70 smears collected from the hall facilities. The results of total alpha-beta activity measured for smears taken during the radiological characterization of the reactor hall were close to the background values. Some of the data for total alpha- and beta- activity as well as for gamma emitting ¹³⁷Cs, ⁶⁰Co and ¹⁵²Eu nuclides are presented in Tables 8 and 9.

Table 8

Data for total alpha- and beta- activity measured in smears collected at the reactor hall, [Bq.cm⁻²]

Sample	Total alpha activity	Total beta activity
1	7.81 E-5	6.46 E-4
2	1.11 E-4	5.32 E-3
3	8.67 E-5	6.25 E-3
4	1.36 E-4	8.48 E-3
5	4.92 E-5	5.92 E-4
6	9.89 E-5	1.86 E-3
7	7.40 E-5	2.73 E-3
8	2.50 E-5	6.70 E-4

Table 9

Data from gamma spectrometry measurements in samples collected at the reactor hall [Bq.kg⁻¹]

Sample	¹³⁷ Cs	⁶⁰ Co	¹⁵² Eu
1. floor material	362 ± 36	2616 ± 244	< 5
2. wall material	< 4.5	24 ± 3	< 5
3. floor material	16 ± 3	27 ± 4	< 3
4. floor material	14 ± 2	58 ± 6	< 5
5. floor material	8540 ± 830	85190 ± 7930	3270 ± 150
6. floor material	7163 ± 695	39340 ± 3664	39340 ± 3664
7. floor material	9240 ± 890	74320 ± 6920	1240 ± 70

All dismantled equipment and the generated during the dismantling activities solid radioactive waste (mainly metals - steel, aluminum and iron as well as small amounts of graphite, concrete, rubber and plastics) were categorized, sorted, packed and sealed in special reinforced concrete containers for transportation and storage of RAW. The categorization was performed in accordance with the acting Bulgarian Regulations [9, 10].

The collective equivalent dose during the reactor equipment partial dismantling and the radiological characterization activities can be assessed to 12–15 mSv. Obtained collective doses are:

- Dismantling of reactor internal systems – 2,47 man mSv;
- Dismantling of primary cooling loop equipment – 123 man μSv;
- Dismantling at the reactor site – 80 man μSv;
- Dismantling of the thermal column – 2,41 man mSv;
- On the site for secondary processing – 271 man μSv.

The obtained doses for workers vary between 0.001 and 0.972 mSv.

4. CONCLUSION

The results of the radiological characterization served as a basis for making management decisions related to specification of appropriate techniques, tools, appliances and devices used to perform the dismantling of reactor equipment and the selection of appropriate methods for decontamination, risk assessment and insurance the radiation protection of workers and the environment. Data from the final radiological characterization were used to assess the situation after accomplishment of all dismantling and decontamination activities. In addition, data from the measurements and analyses were used for the classification of RAW after appropriate procedural arrangements according to the plan for dismantling.

The strict following of the Bulgarian legislation and the IAEA safety standards realized in the partial dismantling Plan ensured that the radiological characterization at the research reactor IRT-2000 (prior, during and after the dismantling operations) has been accomplished smoothly and safe by minimizing the risks to the personnel and to the environment by minimizing the radioactive waste.

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