Remediation of Old Environmental Liabilities in the Nuclear Research Institute Rez plc

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ABSTRACT

The Nuclear Research Institute Rez plc (NRI) is a leading institution in all areas of nuclear R&D in the Czech Republic. The NRI's activity encompasses nuclear physics, chemistry, nuclear power, experiments at research nuclear reactors and many other topics. The NRI operates two research nuclear reactors, many facilities as a hot cell facility, research laboratories, technology for radioactive waste (RAW) management, radionuclide irradiators, an electron accelerator, etc.

After 50 years of activities in the nuclear field, there are some environmental liabilities that shall be remedied in the NRI. There are three areas of remediation: (1) decommissioning of old obsolete facilities (e.g. decay tanks, RAW treatment technology, special sewage system), (2) treatment of RAW from operation and dismantling of nuclear facilities, and (3) elimination of spent fuel from research nuclear reactors operated by the NRI. The goal is to remedy the environmental liabilities and eliminate the potential negative impact on the environment. Based on this postulate, optimal remedial actions have been selected and recommended for the environmental remediation. Remediation of the environmental liabilities started in 2003 and will be finished in 2012. Some liabilities have already been successfully remedied.

The most significant items of environmental liabilities are described in the paper together with information about the history, the current state, the progress, and the future activities in the field of remediation of environmental liabilities in the NRI.

INTRODUCTION

The Nuclear Research Institute Rez (NRI) is a leading institution in all areas of nuclear R&D in the Czech Republic. The NRI has had a dominant position in the nuclear programme since it was established in 1955 as a state-owned research organization and it has developed to its current status. In December 1992 the NRI has been transformed into a joint-stock company.

The Institute's activity encompasses nuclear physics, chemistry, nuclear power, experiments at the research reactor and many other topics. Main issues addressed in the NRI in the past decades were concentrated on research, development and services provided to the nuclear power plants operating WWER reactors, development of chemical technologies for fuel cycle and irradiation services to research and development in the industrial sector, agriculture, food processing and medicine.

At present the research activities are mainly targeted to assist the State Office for Nuclear Safety – the nuclear safety regulating body, power plant operators and nuclear facilities contractors. Significant attention is also paid to the use of nuclear technology outside the nuclear power sector, providing a wide range of services to industry, medicine and the preparation of radiopharmaceuticals. The NRI operates two research nuclear reactors, many facilities as a hot cell facility, research laboratories, technology for RAW management, radionuclide and electron irradiators, etc.

After 50 years of activities in the nuclear field, there are many environmental liabilities that shall be remedied in the NRI. There are three areas of remediation: (1) decommissioning of old obsolete facilities (e.g. decay tanks, liquid RAW storage tanks, old RAW treatment technology, special sewage system), (2)

processing of RAW resulting from operation and dismantling of nuclear facilities and (3) elimination of spent fuel from research nuclear reactors. The goal is to remedy the environmental liabilities and eliminate the potential negative impact on the environment. Based on this postulate, optimal remedial actions have been selected and recommended for the environmental remediation.

The character of the environmental liabilities is very specific and requires special remediation procedures. The remediation of the environmental liabilities is performed by the NRI. The NRI has gained many experiences in the field of RAW management and decommissioning of nuclear facilities and will use its facilities, experienced staff and all relevant data needed for the successful realization of the remediation.

Remediation of the environmental liabilities started in 2003 and will be finished in 2012. The past activities in the frame of remediation of the environmental liabilities are described in [1, 2, 3].

PREPARATION FOR REMEDIATION

The characterization of environmental liabilities has been already carried out. The work was carried out to obtain information about the NRI site, the degree and extent of environmental pollution and potentially endangered target groups. The first stage comprised the collection of the needed information. In the framework of the first stage studying the existing data led to the identification of information that was lacking.

A risk analysis study was performed comprising the identification and characterization of potential sources of risk, potentially exposed receptors and exposure pathways, potential chemical compounds, radionuclides and media of concern. Additional information on natural conditions at the site was obtained through hydrogeological studies of the pollution and information on sources of ionizing radiation and radioactive contamination using dosimetric measurement and radiochemical analyses were included into the risk analysis. The results of the risk analysis enabled to determine the priorities of the remediation, the technical conception of the remedial actions as well as the estimation of the expenses.

The inventory of RAW resulting from remediation has been prepared. The result was approx. 1500 m^3 of RAW corresponding to approx. 600 tons.

DESCRIPTION OF ENVORINMENTAL LIABILITIES

Decay Tanks (Building No. 211/5)

The Building No. 211/5 (Decay Tanks) was designed in 1958, and had been in use since 1961. The building was designed for storage and decay of concentrated short-lived RAW, but also RAW containing long-lived radionuclides have been shipped there. All transfer of RAW has been stopped in 1990.

The building is submerged in the terrain on three sides. It contains two cylindrical tanks (length 9.5 m, diameter 3 m, weight approx. 10 metric tons), each with a capacity of 63 m³. One serves for RAW storage, the second one serves as a reserve tank. The tanks are made from structural steel jacketed by stainless steel inside the vessel. The design life of these tanks is now exceeded. They are placed into two separate concrete bunkers located partially below ground. The walls of the bunkers are 1 m thick. Above the bunkers are located a masonry building with tank inlet pipes, ventilation equipment and equipment for taking of water samples from tanks. The drawing of the Building No. 211/5 is provided in the Figure 1.

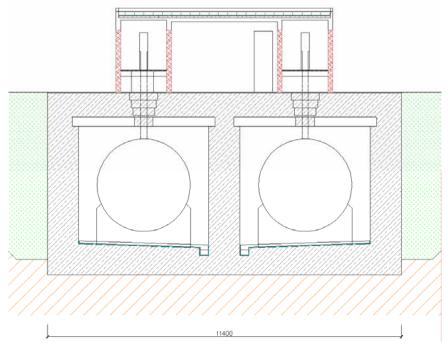


Fig. 1. Building No. 211/5 – Decay tanks (section)

The tanks are equipped with two openings, which served for RAW receipt and for collection of water samples. Tank A contains 4.5 m³ of liquid with an activity of 0.5 MBq/l. The main identified radioisotopes are Cs-137 and ¹⁵²Eu. Tank B contains 2.5 m³ of solid RAW and also 8 m³ of liquid with an activity of 21 MBq/l. The main identified radioisotopes are Co-60 and Cs-137. However, there is anecdotal evidence that up to 2 g Pu-239 may also have been deposited in the tank at some time. The solid RAW consist of tins containing irradiated samples residues of irradiated measuring probes and impact and stretching test bodies from the reactor vessels and tins containing fission material from spent fuel. According to measurements made inside the tank B, the maximum dose rate is above the pile of solid RAW, approx. 2 Gy/h. The surrounding bunkers contain water with an activity less than 20 Bq/l. The leakage and spillage from the storage tanks and direct irradiation from in-situ material were identified as the main risks to the environment and/or to employees.

The remediation procedure will be as follows:

1. The liquid RAW from tanks will be removed. A facility equipped with technology for removal of RAW and its processing is being constructed. The underground bunker will be used as a hot cell. A penetration through the concrete ceiling of the bunker will be made and a removable shielding will be installed. The control room of the hot cell will be placed in front of the bunker. A penetration through the wall of the bunker will be made and a shielding window and manipulators will be installed. Then a head of the tank B will be cut off and a technology box will be installed. This box will be hermetically connected with the tank and equipped with technology for RAW manipulation, characterization and fragmentation.

2. Solid RAW will be removed and directly processed in the technology box. Then RAW packed in special cases will be loaded into disposal units and send for disposal.

3. The tanks will be decontaminated by high-pressure water jetting and abrasive blasting and then will be dismantled by special fragmentation equipment. The RAW will be either released into the environment or disposed. The building will be decontaminated.

The construction of the facility started in 2006 and will be finished in 2007; then removal and processing of RAW will start. Decontamination and fragmentation of the tanks and decontamination of the building will be carried out in 2009.

Liquid RAW storage tanks (Building No. 211/3)

Three steel tanks of the same design as the decay tanks described above are located in concrete underground bunkers. The tanks served for receiving liquid RAW from a research reactor. The tanks are aged beyond their design life. All three tanks are contaminated by fission and corrosion products, mainly Cs-137, Co-60 and Sr-90. Leakage or spillage of these tanks was identified as the main environmental risk.

According to the original project the remediation procedure would comprise decontamination and dismantling of tanks and processing of RAW. Then new tanks for storage of liquid RAW would be installed. Now a new concept is being prepared. The tanks will be decontaminated and after the investigation of the tanks` integrity, the polyethylene lining will be installed inside the tanks. Then the resources for fragmentation and RAW processing and installation of new tanks will be saved.

Decommissioning of tanks started in 2004 and will be finished in 2007. Decontamination of tanks is being performed now. Surface of one tank was contaminated by a bituminous product from decontamination of a bituminisation unit performed in the past. Application of dry ice blasting was proposed but this could lead to spreading of contamination. Then a method utilized the organic solvents was proposed but the amount of the solvent could be rather greater. Also the protection of workers could be demanding. So then a very simple method was used – application of mineral oils to soften the bituminous layer. Then the layer was removed manually and a very small amount of organic solvent was applied to complete the decontamination.

Old RAW treatment technology (Building No. 241)

The old RAW technology comprises the evaporation unit, storage tanks and a set of mixed-bed filters. The technology was in operation since 1962 and was shut down in 1992. The total amount of the equipment to be decommissioned corresponds approximately to 50 metric tons of steel. The equipment is contaminated with fission and corrosion products, mainly with Cs-137, Co-60 and Sr-90.

The remediation procedure will comprise dismantling of the equipment after decontamination and processing of RAW. Decommissioning of technology started in 2004 and will be finished in 2008.

The decommissioning of a condenser used for cooling of vapors from evaporation could serve as a typical example of dismantling of the old treatment technology in the Building No. 241. The condenser with a diameter of 0.7 m and a length of 4 m was made from carbon steel (shell) and brass (heat exchange tubes). Abrasive cutting wheel and hydraulic shears were used for fragmentation. It was decided to treat and condition RAW from condenser fragmentation for disposal because of difficult decontamination (strong corrosion of steel parts). Decontamination of brass tubes would be possible but with respect to its inner diameter it would be difficult and time consuming to prove that the release levels were met. The process of fragmentation is shown in the Figure 2.



Fig. 2. Fragmentation of the condenser

Contaminated equipment (Building No. 250)

The Building No. 250 houses radiochemical laboratories, hot and semihot cell complexes, rabbit systems and auxiliary equipment. Two laboratories called "Alpha halls" contain eight sets of wall boxes and a number of glove boxes. The boxes were used for handling with alpha radionuclides (U, Np, Pu, Am) and are now significantly contaminated. The total volume of boxes is approx. 80 m³. Contamination with alpha radionuclides was identified as the main risk to employees.

The remediation procedure will comprise dismantling of the equipment after preliminary decontamination and processing of RAW. The dismantlement of the equipment started in 2004 and will be finished in 2010.

Dry ice blasting was used for decontamination of a semihot cell used for experimental reprocessing of spent fuel in the past. The semihot cell made from cast iron was partially decontaminated in the past and then its internal and external surfaces were covered with coating to prevent spreading of contamination. The coating was partly removed by a paint remover at first and then dry ice blasting was applied for decontamination. Dry ice blasting was also used for decontamination of building surfaces – removing of contaminated coatings. Very small amount of secondary RAW was generated and dispersion of contamination was limited.

Special Sewage System

The Special Sewage System was used for transfer of liquid RAW from various facilities (research reactors, radiochemical laboratories) to a RAW processing facility. The system consisted of a stainless steel pipe network with a total length of 410 m situated in an underground concrete corridor. The integrity of the system has never been tested. The system was contaminated by fission and corrosion products, mainly by Cs-137, Co-60 and Sr-90. Leakage of waste water from piping was identified as the main risk to the environment.

The remediation procedure started with a removal of soil and opening of the corridor. The pipes and other steel components (valves, fittings, etc.) have been removed and sent for treatment. The total amount of

contaminated metal parts was approx. 20 metric tons. Limited parts of the surface of the concrete corridor were contaminated because of small leakages. The contaminated surfaces were removed and RAW processed.

A standard mechanical saw was used for fragmentation of pipes before decontamination. The pipe parts as joints and flanges and corroded parts were sent for conditioning. High-pressure water jetting was used for preliminary decontamination of pipes. Then the ultrasonic bath with decontamination solutions was used. Decontamination was successful in most cases; some pipes were mechanically decontaminated by a special one-purpose instrument (an abrasive rotating device). External contamination of pipes was measured by a standard contamination instrument and contamination inside the pipes was measured by a special tube detector. Approximately 90 % of pipes were released into the environment.

The decommissioning started in 2004 and was finished in 2005. Then a new sewage system has been installed. The old concrete corridor was used for installation of a new system equipped with a leakage monitoring system after corridor renovation.

RAW stored in the Reloading Site (Building No. 211/6)

The Building No. 211/6 was initially constructed as a temporary reloading site to handle conditioned RAW but later was used also for storage of various RAW before treatment. RAW is stored in 8 concrete boxes each with dimensions of $5.5 \times 8 \times 4 \text{ m}$ (1400 m³ total capacity). The bases of the boxes are 4 m below ground level and are drained to four closed sumps. The building has a steel roof. The total volume of stored RAW is approximately 600 m³. An incomplete inventory is available; this gives only a very general description of the RAW contained in the boxes. The RAW is contaminated mainly with Cs-137, Co-60 and Sr-90. Leakage of liquid waste in boxes, wash-off of contamination from RAW by rainwater and direct irradiation from in-situ material were identified as the main risks to the environment and/or to employees.

The hall above the Reloading Site with a crane and auxiliary technology (see the Figure 3) was constructed in 2004. RAW will be sorted and transported for processing (fragmentation, decontamination and conditioning). Then RAW will be disposed or released into the environment. The treatment of RAW will be performed in 2008 – 2012. Then the building will be decontaminated and used as a new RAW store after reconstruction.



Fig. 3. Building No. 211/6 – Reloading Site with a new hall

RAW stored in the Red Rock Storage Site

Storage of RAW at the "Red Rock Storage Site" started in 1988. The stored waste includes RAW arising from reconstruction of the VVR-S research reactor (primary circuit, ventilation system, etc.) stored in ISO shipping containers and old technology equipment for RAW processing (heat exchangers, tanks, filters).

The view at the storage site is shown in the Figure 4. The total storage area is 300 m^2 . The total amount of RAW is approx. 90 metric tons. The RAW is contaminated mainly with Cs-137, Co-60 and Sr-90. Rain wash-off from contaminated equipment to soil and groundwater and irradiation from in-situ material were identified as the main risks to the environment and/or to employees.



Fig. 4. Red Rock storage site

RAW will be transported for processing (fragmentation, decontamination and conditioning). RAW will be disposed or released into the environment. The processing of RAW will be performed in 2008 – 2012.

RAW from archive program

RAW consists of residues of irradiated surveillance specimens and experimental samples of steel for reactor pressure vessels. The surveillance specimens are stored in closed metal vessels with a diameter of 130 mm and height of 170 mm. In a distance of 0.5 m the surface dose rate does not exceed 0.2 Gy/h. The total number of vessels with the specimens is in order of hundreds. The RAW is stored in hot cells in the Building No. 250.

In 1996 a small part of RAW was transported in a special transport container to the High Level Waste Store (HLWS), where the facility for storage has been constructed. It consists of a system of shielded nests made from lead located in a storage box. A part of RAW has already been processed and sent for disposal. The system has been found as unsatisfactory mainly from the point of view of radiation protection, that is why a new system of RAW management was designed in 2002. A new system for RAW handling has already been constructed in hot cells. It serves for characterization, sorting and repacking of the RAW into new vessels. The vessels will be put into special steel disposal containers. Processing of RAW started this year and will be finished in 2008.

Experimental irradiation channels

The experimental irradiation channels made from aluminium and stainless steel were used in the LVR-15 research reactor. The total volume was 2 m³. The maximum dose rate in a distance of 1 m was 100 mGy/h. Direct irradiation from in-situ material was identified as the main risk to employees. In 1996 the channels have been fragmented under water and then put into the shielded containers and after conditioning were transported for disposal or for further storage in the High Level Waste Store (HLWS) according to the surface dose rate higher than 1 mSv/h (one of limits for disposal into the repository). The total amount of containers with volume of 200 l was 37, approximately a half has been sent for disposal directly and a half has been sent for storage.

Spent nuclear fuel from research reactors

The EK-10 nuclear fuel was used in the VVR-S research reactor from 1957 until 1974. The fuel EK-10 was made up of rods of a 10 % enriched uranium dioxide-magnesium alloy in aluminium cladding. One EK-10 assembly consists of 16 rods. In 1974, a new fuel element was introduced with 80 % enrichment – IRT-2M. This consisted of a number of concentric square rings of uranium/aluminium alloy fuel/metal clad on either side with aluminium. In 1996 a similar fuel, still referenced as IRT-2M, was introduced which had 36 % enrichment but used uranium dioxide.

In years 1969 – 1975, EK-10 spent fuel was removed from the initial away-from-reactor (AFR) pools to the Reloading Site (Building No. 211/6). EK-10 spent fuel was held in dry concrete storage containers. It was only a provisional storage that did not meet the criteria for safe storage of spent fuel. That is why the spent fuel was transferred to the HLWS in years 1996 – 1997. The IRT-2M fuel has been moved out of the initial AFR pool into the HLWS pool between the years 1996 – 2003. Now 240 IRT-2M and 16 EK-10 spent fuel assemblies are being stored in the pool and 190 EK-10 assemblies in a dry storage.

The actual state of the EK-10 spent fuel stored in dry storage containers was not known. The most serious circumstance was that the storage containers were not hermetically sealed. This fact connected with a fuel cladding corrosion could lead to release of radionuclides outside the storage containers. There also existed a possibility of a loss of integrity of storage containers, because of their age and materials used for construction of containers. According to the period of storage, the character of construction materials of

containers (carbon steel drum filled with concrete, carbon steel liner) and the possible interaction of the container material with aluminium cladding the cladding corrosion was probable.

In 2002 a new concept for EK-10 spent fuel management was designed. A new hot cell has been constructed in the HLWS and EK-10 spent fuel are being packed into stainless steel cases and hermetically sealed (welded). The packed spent fuel is stored in a special vault. Spent fuel packing started this year and will be completed in 2007.

The spent fuel will be transported for reprocessing to the Russian Federation in the frame of a joint programme called the Global Threat Reduction Initiative (GTRI) of the US National Nuclear Security Administration (NNSA) in 2007. The mission will be a joint effort between the United States, the Czech Republic, Russian Federation and the International Atomic Energy Agency. The transport of spent fuel from the Czech Republic will serve as a pilot project in the frame of the initiative for realization of spent fuel transport from another countries.

TECHNOLOGY FOR REMEDIATION

Selection of technology for remediation

The process of selection of technology for decommissioning is one of the most important aspects of decommissioning activities. This process has strong impact on the whole decommissioning process, including radiation protection and RAW management. Radiation protection is the most important factor. The level of contamination of RAW from remediation is up to tens of MBq/cm², and dose rate is up to tens of mGy/h. There are also another important parameters – type of material, its thickness, accessibility of technology, etc. The evaluation of possible use of various technologies for fragmentation and decontamination with aim to facilitate management of generated RAW and release into the environment was performed together with economic evaluation.

The NRI are being equipped with new technological equipment for fragmentation and decontamination. The standard industrial technologies with small modifications are being purchased and also special technologies (e.g. technologies for decontamination, measurement, hot cells, etc.) are being developed with assistance of external subcontractors.

Fragmentation and decontamination

The list of methods used for fragmentation and decontamination is provided in the Table I.

Table I. List of methods used for fragmentation and decontamination

Fragmentation	Decontamination
Power hydraulic shears	Vacuuming (vacuum cleaner with HEPA filter)
Mechanical saw	High-pressure water jet
Abrasive cutting wheel	Chemical decontamination
Oxy acetylene cutting	Foam decontamination
Plasma arc cutting	Ultrasonic decontamination in a special bath
In-situ mechanical milling (fragmentation of tanks, remote controlled) – in testing	Dry ice blasting
	Grit blasting (in-situ, in box) - considered

In-situ mechanical milling will be used mainly for fragmentation of tanks, but it can be also used for planar object. In the NRI, there are mainly two types of tanks made from structural steel with thickness of 12 mm (bottom 14 mm) jacketed by stainless steel inside the vessel with thickness of 2 mm with volume of 10 m³ (weight 4000 kg, length 3500 mm, diameter 2000 mm) or 63 m³ (weight 9700 kg, length 9500 mm, diameter 3000 mm). The tanks are located within the bunkers and the space around is very limited (from 500 to 1000 mm), mainly under tanks (from 300 to 600 mm). Sludge will be removed before fragmentation and the tanks will be preliminary decontaminated. One purpose remote controlled in-situ mechanical milling will be used for fragmentation of tanks. The cutting rate of the device is max. 250 mm, the kerf width is max. 16 mm. The tanks will be fragmented on rings with width of 400 mm and then segmented by hydraulic shears. The device will produce secondary RAW in a form of small chips, almost no airborne contamination will be produced. Now the device is in the phase of testing.

Technology for RAW treatment

The evaporation unit is used for treatment of aqueous liquid RAW. The concentrate is cemented into 2001 drums by means of a batch-type cementation unit. The processing of pressable solid RAW consists of indrum, low pressure compaction (into 1151 drums) and embedding of the 1151 drum within a 2161 drum with concrete.

Release of RAW into the environment

The total amount of solid RAW resulting from remediation for processing will be approx. 1500 m³, at least one third is expected for release into the environment after treatment. It will require special equipment for measurement. The equipment owned by WADE Company will be used for measurement for release into the environment. The device called MERLIN (MEasuring ReLeasing INstrument) is used for release of RAW mainly from the Dukovany NPP. These are two mobile units - measuring and control one. There are three multi-channel spectrometers of gamma radiation with semiconductor HPGe detectors (analyse of gamma radiation within the energy scope of 50 to 2000 keV) and two plastic scintillators (analyse of beta radiation within the energy scope of 100 to 3000 keV) in a special shielded tunnel made of 10 cm of lead and 1 cm of stainless steel. For every batch of material, the key radionuclides are determined, which can be taken into consideration according to the character of the operation and material released to the environment. The activities of the directly measured radionuclides are determined on the basis of the measured spectra of gamma radiation; the activities of the other monitored radionuclides (e.g. Pu-239, Sr-90) are assessed in relation to the correlated radionuclides (e.g. Co-60, Cs-137). The reduction of the natural background (daughter products of radon) is ensured thorough maintaining the overpressure in the measuring shielded chamber. It is also necessary to take samples, measure the samples in laboratories and carry out radiochemical analyses. It allows to precise the important radionuclides characteristic for the workplace and correlation. The measurement is performed as official measurement and the authorized person carries it out. Approximately 600 tons of material (both metallic and nonmetallic) was released into the environment in last four years. Measurement of RAW from remediation of environmental liabilities will be very complicated because RAW are very heterogeneous and many correlations must be assessed according to the character of groups of RAW.

Disposal of RAW

Approximately 1000 m³ of solid RAW resulting from remediation are expected for disposal. The standard system of solid RAW processing consists of fragmentation and conditioning by cementation into 200 l drums. Then the drums are sent for disposal into the repository. A new concept has been prepared for disposal of big fragments of contaminated technological equipment directly into the disposal cells of the repository. It will be advantageous from the point of view of radiation protection because it will require less fragmentation operations. It will be also less time consuming and many resources will be saved.

FINANCING

The Ministry of Finance of the Czech Republic finances the remediation of the environmental liabilities. The total expenses for remediation of the old environmental liabilities in the NRI will be approx. 900 millions CZK, it corresponds approx. to 40 millions US\$.

CONCLUSION

The character of the environmental liabilities in the NRI is very specific and requires special remediation procedures. The remedial activities are being carried out on the high level of radiation protection and up to now there was no extraordinary event or accident.

The experience gained during the remediation process will be used for future activities connected mainly with the decommissioning not only facilities operated by the NRI (research reactors, radiochemical laboratories, hot cell complexes, etc.), but also facilities operated by other companies (workplaces with ionizing radiation sources, nuclear facilities, nuclear power plants).

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