Lessons Learned from RAW Treatment in the Slovak Republic – Minimization for Final Disposal - 8423

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Abstract

This paper is referring about the utilization of technologies for the treatment and conditioning of low and intermediate level RAW from operation and decommissioning of nuclear facilities in Slovakia. This experience represents more than 116 reactor years of NPP operation, mainly of NPPs equipped with VVER 440 reactors, 30 years of decommissioning activities, 27 years of development and operation of technologies for the treatment and conditioning of RAW and 7 years of LLW and ILW final repository operation.

These technologies are located in two localities: Jaslovske Bohunice and Mochovce. The complex treatment and conditioning center (cementation, bituminisation, incineration, vitrification, fragmentation and compacting) for almost all types of radioactive waste is located in Jaslovske Bohunice NPP site. The treatment and conditioning center for liquid radioactive waste (cementation and bituminisation) and the surface type repository for LLW and ILW final disposal are located in Mochovce area. The treated waste forms are disposed to repository in cubical Fiber Reinforced Concrete (FRC) containers.

The experience from the phase of technology development and the phase of technology modifications for various types of RAW, the experience from long term operation of technologies and the experience from transportation of original and packed wastes are described in this paper.

The method of optimally combined technology utilization in order to maximize the radionuclide inventory at the same time with respect of disposal safety limitations of repository is described, too.

The significant RAW volume reduction for final disposal was achieved through mediation of the combination of treatment and conditioning technologies. The disposal of treated RAW in cubic FRC containers allowed the optimal utilization of volume and radiological capacity of LLW and ILW repository in Mochovce and the fulfillment of determined safety requirements at the same time.

1. INTRODUCTION

Construction and further development of nuclear power engineering in the Slovak Republic led gradually to operation of six units with WWER 440 reactor and one unit with KS-150 HWGCR reactor from which HWGCR and one unit with WWER 440 reactor are decommissioned at present time. This operational and decommissioning experience represents more than 116 reactor years of NPP operation, 30 years of decommissioning activities, 27 years of development and operation of technologies for the treatment and conditioning of RAW and 7 years of LLW and ILW final repository operation.

Operation and decommissioning of nuclear facilities are closely connected with generation of a significant volume of radioactive waste of various composition and radioactivity levels. Since 1972, when the first nuclear rector was initiated to the service in Slovakia, there have been produced approximately 36 000m^3 of liquid and solid RAW, from that nearly 6 800m^3 RAW comes from decommissioning of NPP A-1, which service was closed in 1977 as a result of accident, which was classified according to INES by level 4. Relatively large amount RAW coming from NPP A-1 with higher contents of fission products and transuranium elements has evoked the necessity of intensive development of technology for their processing. Between 1980 and 1993 basic technologies of treatment RAW by bituminisation, cementation, incineration, vitrification and compacting were developed and put into operation in Czechoslovakia.

Conception for radioactive waste disposal was formulated by Resolution of the Government of the SR No. 190/94 Coll. This concept determined that for final disposal of LLW and short-lived ILW radioactive waste the near surface repository should be upgraded and finished up at Mochovce site. Operation of repository started in 1999. Wastes, which do not meet criteria for disposal in Mochovce will be stored temporarily at the NPP sites. Construction of an integral storage facility in Jaslovské Bohunice is assumed by the year 2012.

2. SUMMARY BALANCE OF RADIOACTIVE WASTE

A complex balance of radioactive waste and basic knowledge of their physical-chemical parameters and activity is an essential information for any considerations on a usability of technological procedures for their treatment and conditioning, and thus for formulation of their safety management strategy.

Radioactive waste in nuclear power plants can be divided into the following two large groups:

a) Waste from nuclear power plant operation
b) Waste from nuclear power plant decommissioning.

The summary balance of operational radioactive waste includes operational waste from NPP V-1, V-2 and EMO 1,2 generated by the year 2005 is given in Table 1. A forecast of operational RAW generation deals with an expected time of operation and an average annual generation of individual RAW types determined on the basis of real data on RAW generation during past years.

Table 1 Operational RAW – Present and Future Production

Waste Type	Generated by 2005		Future Production	
	Quantity [m³]	Total Activity [GBq]	Quantity [m³]	Total Activity [GBq]
RA- concentrate	26,300	27,960	7,030	3,698
Low-active sorbets	149	175	167	167
Medium-active sorbets	420	4,200	258	2,580
Oil	6	0.6	12	1
Ventilation filters	610	610	717	717
Combustible RAW	1,150	38	3 460	105
For cementation	65	2.5	146	2
For high-pressure pressing	345	21	332	19
Metal large-sized	172	3.0	613	9
Other	13	0.25	0	
High-level RAW stored in a storage facility (mogilnik)	App.7.0 (19t)	34,000	7 (19.6t)	36,650
TOTAL	**29,227**	**66,991.4**	**12,742**	**43,948**

Radioactive waste from A-1 NPP decommissioning represents a specific problem considering high activity of some waste types and presence of increased concentration of radionuclides especially with long lived especially alpha nuclides. The summary balance of RAW generated during the decommissioning work of A-1 NPP by the year 2005 is presented in Table 2.

Table 2 Radioactive waste from A-1 NPP decommissioning

Waste Type	Quantity [m³]	Total Activity *Beta, Gamma* [GBq]	Total Activity *Alpha* [GBq]
Chrompik	12.8	3.04 E+6	30
Liquid RAW	127.1	912.7	0.063
Dowtherm	23.5	1,770	0.31
Oil	40.2	1,3	0.007
Sludge- geopolymer	6.9	1.4 E+5	125.4
Highly active sludge from hot cell (Vitrification)	App. 0.8	1 000 (N)	10 (N)
Sludge - cementation	293.6	1.4 E+5	85
Filter components – total	App. 255 (37.4 t)	App. 15 (N)	App. 0.15 (N)

Metal scrap from PC	194.4 t	App. 100 (N)	1.0 (N)
Used PDS(storage of spent fuel cans)	138.7 t	2,800	14.0 (N)
Empty PDS (storage of spent fuel cans)	7.8 t	App. 70	0.7 (N)
Metal ingots from re-melting	3 t	App. 0.013	1.0 E-5 (N)
Glass product from the vitrification line (VICHR)	1.5 (3.2 t)	2.1 E+4	2.0 E+2 (N)
Metal scrap from processing	1,274 t	124.2	0.1
Compactable waste	453	36.8	0.035
Combustible waste	88.3 t	41.4	0.005 (N)
soil + concrete	7,400 t	74 (N)	0.01 (N)
Spent ion exchangers, sand and bio sorbent	2.0	270	0.35
Building waste (concrete waste)	1,263 t	6.33	0.006
Cement product in barrels	561 barrels	132	0.09
Bituminous product in barrels	2,052 barrels	2,000	2.17
Ash from an incinerator	29 barrels	3.4	
Contaminated soil, metal cables, concrete waste, gravel, asphalt, concrete	5,361 barrels	54	
Sorbets from a long-term repository water purification	3 barrels	9.0 E+4	9.0 (N)
Ash from an incinerator in building 808	15.6 t	7.3	0.003
Heat exchanger of an evaporator stored in building 41	0.4 m³ (1 t)	0.14	
Fixed sludge from a long-term repository (SIAL)	0.14 t	140 (N)	1.4 (N)
Cemented chromo sulphuric acid	4.5 t (21 barrels)	2.5	0.16
Active coal (SV)	1.0 t	0.2 (N)	2.0 E-4(N)

N- Activity not measured, the value determined by a qualified estimation

Forecast of decommissioning RAW generation in future is given in the Table 3 and 4.

Table 3 Radioactive waste from stage 2 of A-1 NPP decommissioning

Waste Type	**Quantity [m³] (t)**	**Total Activity [GBq]**
Liquid RAW	44,519	800
Solid combustible RAW	284	1.4
Solid compactable waste	4,334	347
Solid RAW from grinding	65	33
Ventilation filters	672	40
Slag from re-melting (Cement product)	854 t	854
Reactor vessel fragmented parts	216 t	350,000

Other highly active metal scrap	1,619 t	32,380
Cartridges with a vitrification product	3.3 (423 pc)	12,000
Shielded ion exchanger columns	10 (50 columns)	7,200

Table 4 Radioactive waste from V-1, V-2 and Mochovce NPPs decommissioning [1]

Waste Type	Quantity $[m^3]$ (t)	Total Activity [GBq]
Liquid RAW (concentrate)	9,428	83,500
Spent ion exchangers	70	7,0
Solid combustible RAW	228	2,3
Solid compactable waste	847,5	68
Small metal scrap	1,190 t	95
Pellets from high-pressure metal scrap	550 t (378 m^3)	44
Ventilation filterst	7.5	1.5
Sand cementation (final product)	31 m^3	1.5
Anthracite + coal – cementation	40	0,3
Fragmented highly active reactor parts	744 t	C-14:4.3 E+4 Ni-59:4.8 E+5 Ni-63:5.3 E+7
Low and medium active reactor parts	2,046 t	8.5 E+3
Ingots from re-melting	5,000 t	638
Concrete cementation (slag, activated concrete and concrete from grinding)	3,718 m^3 of a final product	1,859

[1] V-1 NPP decommissioning after its operation end in 2006-2008, and future decommissioning of V-2 NPP and Mochovce NPP after their lifetimes

3. OPERATION OF TREATMENT AND CONDITIONING FACILITIES

3.1 Radioactive Waste Treatment at Bohunice

In the locality of Jaslovské Bohunice is installed several technologies which enable proceeding the following processes of conditioning and treatment of RAW:

- incineration of solid and liquid RAW,
- over concentration of liquid RAW,
- cementation of concentrated liquid RAW,
- cementation of sludge and gravels in 200 dm^3 barrels,
- high pressure compacting of solid RAW,

- bituminisation of liquid RAW,
- sorting of solid RAW,
- purification of low-active waste water,
- vitrification of liquid RAW and inorganic resins,
- fragmentation and decontamination of metal RAW,
- fixation of sludge and sediments into the geopolymer matrix,

All sorts of RAW, which have been produced by the operation of the nuclear power plants V1, JE V2, solid RAW produced by the operation NPP EMO, RAW originated from decommissioning NPP A-1 and institutional RAW are being treated by equipments of conditioning and treatment of RAW. Whereas the capacity of processing technology does not cover the requirement of the fluency final conditioning and disposal RAW in the repository, a storage accommodation for temporary holding of treated and conditioned RAW has been built up near by the equipments and in the object JE A-1.

The Bohunice processing center is the main place for the treatment and conditioning of liquid and solid RAW in Slovakia. It is the facility where all RAW from NPP operation and decommissiong and institutional RAW which can be disposed in National Repository of Radioactive Waste at Mochovce are treated and conditioned to the form for final disposal. The facility was built between 1993 and 1999. The facility consists of sorting, incineration, high pressure equipment (supercompactor), equipment for liquid RAW over concentration and cementation. There is a place where conditioned RAW are placed into the Fiber Reinforced Concrete Containers and filled up by active cement grout. The table 5 proves the efficiency of these technologies.

Table 5 Summary of treated waste

Processing Technology	**Specific parameter**	**Production of conditioned RAW**							
		2000	2001	2002	2003	2004	2005	2006	2007
Cementation	FRC	35	144	203	210	232	243	241	208
Supercompaction RAW	T	37	85,7	108	112,2	126	122,2	124	147,7
Incineration - solid RAW	T	10,3	51	75	91,3	76,2	62,8	100,7	38,6
Incineration - liquid RAW	dm^3	660	3956	5093	12400	4200	3874	17523	11600
Concentration	m^3	28,5	220	323	355,8	374,5	394,4	513	494

During the operation of the technological equipment the most technical problems were occurred in the waste incineration part, where effected by higher concentration of Cl^- anions being found in the incineration waste comes to an excessive corrosion of piping in the nod responsible for the gas cleaning, that invoke the necessity of their renewal. The air-servicing filtration system (the system of pre-filters and filters of exhausting gases) beyond the wet wash-basins was restored. The project changes on the nod of releasing and ash treating by addition of additive was realized so that it could have been treated on supercompactor. Reparations of the casement wall of the nettle chamber were made simultaneously.

The operation on the „concentration“ equipment is rather stable, without occurrence of disturbances, which would require a long- run layoff.

The equipment for cementation is the limited place of the whole conception of RAW treatment. It is in operation almost during the whole year, because of the individual Fiber Reinforced Concrete Containers are filled by cement grout prepared by this equipment. It works without greater operation problems. Besides the cementation of condensed concentrate from NPP V-1, V-2 it is used also for treatment of the washing liquid from the waste incineration facility and for some liquid RAW from A-1 NPP decommissioning. The barrels filled with bitumen products containing radioactive salt and compacted solid RAW (pellets) are set into the Fibre Reinforced Concrete Containers and embedded with active cement grout. Barrels with gravel fixed in cement matrix and barrels with sludge fixed in cement or geopolymer matrix are also embedded to the containers.

The super compactor provides the reduction of volume of solid compactable RAW. It is usable approximately on a half of its capacity. Operating failures do not have any significant effect on the production. The supercompactor claims occasional maintenance of oil hydraulic system.

3.2 Final Treatment Centre Mochovce

The second centre for the treatment and conditioning of RAW in Slovakia is located in the area of the Nuclear Power Plant in Mochovce. In this centre following technologies are installed:

- bituminisation of concentrate into 200 dm^3 barrels
- bituminisation of resins and sludge mixture into 200 dm^3 barrels
- overconcentration of concentrates for the needs of cementation
- embedding of fixed liquid RAW packed into 200 dm^3 barrels with active cement grout into the fibre reinforced concrete containers
- logistic technologies (for transport of solid RAW from the NPP Mochovce for processing in Bohunice waste treatment facilities in ISO containers and receipt of RAW treatment product in 200 dm^3 barrels from the Jaslovské Bohunice).

The Final Treatment Centre in Mochovce (FTC) is designed first of all for the treatment of liquid RAW materials (concentrates, spent resins with sludge) originated from NPP Mochovce operation. Fibre Reinforced Concrete Containers with 200 dm^3 steel drums with bitumen product and filled up with active cement grout made from concentrates are used for final disposal in the National Radioactive Waste Repository at Mochovce.

FTC is operating on a campaign schedule. Five campaigns of bituminisation of concentrates and one campaign of bituminisation of resin and sludge are conducted per year. The overall capacity of FTC for concentrates is 870 m^3/year and for spent resin and sludge mixture is 40 m^3/year which represents the amount of 1.205 drums with bitumen product and 172 filled Fibre Reinforced Concrete Containers.

The technology of bituminisation on Thin Film Evaporator is installed for the treatment of radioactive concentrates. The designed capacity of evaporator is 200 dm^3/hour of evaporated water. The feeding rate of concentrate and bitumen is adjusted in order to obtain the bitumen product with salt content 40% (w.) The bitumen product is discharged from the bottom nozzle of the evaporator into 200 dm^3 steel drums. The drums are automatically capped and the activity of ^{137}Cs a ^{60}Co is measured. Vapour from the evaporator is routed into a condenser and the condensate is collected in a distillate tank. The condensate is reused after cleaning on activated carbon.

The bituminisation of spent resins and sludge is carried out on discontinuous batch type facility. This facility consists of decanter, dryer and mixer-homogeniser. The sediment is isolated in decanter from water phase and consequently flows down by gravity into the dryer heated by steam. The dried sediment of spent resins and sludge is mixed with melted bitumen in batch type mixer equipped with rotating blades. After gradual adding of sediment and short mixing the mixture is the bitumen product discharged to 200 dm^3 drums similarly like bitumen product from TFE. The ratio of bitumen-sediment-Polyethylene is adjusted in order to achieve the 40 % (w.) portion of resins and sludge in bitumen product.

The cement grout is prepared by mixing of over concentrate (salt content: 400 - 450 g/dm^3) with lime and zeolite-cement mixture in a cement grout mixer equipped by rotating blades. The over concentrate is prepared by evaporation on circulation steam heated evaporator.

A FRC container loaded with bitumen product drums or drums originated from A-1 NPP is then filled with cement grout dosed by pump. An ultrasonic probe monitors the level of cement grout in container. During filling a sample of cement grout can be taken. After each grout batch loading, the FRC is lifted and vibrated on a vibration plate.

The filled FRC containers are cured on roller conveyor and the plugs are sealed using fiber reinforced concrete mixture. This container meets all limits for final disposal so after checking procedure, it is transported to the National Radioactive Waste Repository at Mochovce.

Logistic technologies are devoted and equipped with technology for transport of solid RAW from the NPP Mochovce for processing in Bohunice (incineration, compaction) in ISO containers and transport and receipt of RAW from A-1 NPP decommissioning in 200 dm^3 drums for to combine conditioned operational RAW from NPP Mochovce with conditioned RAW from A-1 NPP decommissioning to achieve higher avail of volume capacity of the National Radioactive Waste Repository at Mochovce.

FTC has recently successfully finished the active tests and is ready for trial operation star-up.

4. FINAL DISPOSAL IN MOCHOVCE REPUBLIC REPOSITORY OF RAW

Mochovce Radioactive Waste Republic Repository (RAW RR) is a multi-barrier repository of above ground type intended for final disposal of solid and solidified radioactive waste generated at operation and decommissioning of nuclear power plants, in research institutes, laboratories and hospitals in the Slovak Republic. The aim of waste disposal in this repository is its safety long-term isolation from the environment.

RAW RR premises are situated in the vicinity of Mochovce Nuclear Power Plant premises. The repository consists of a set of disposal boxes orders in rows and double rows. The first double row is sheltered by steel hall. Boxes are made from reinforced concrete with a 600 mm thick wall. Within stage I, 2 double rows were constructed, i.e. 80 disposal boxes (1 row = 20 boxes). Totally 90 fibre-concrete containers with a unit volume 3.1 m^3 can be disposed in one box. As results from the abovementioned, the total capacity equals to 7,200 containers with the total volume approximately 22,320 m^3.

4.1 Safety limits for RAW disposal

The fundamental criterion for final disposal of the radioactive waste is non-exceeding the limits and conditions of repository operation, i.e. safety and operational restrictions. Safety limits and conditions are connected directly with the safety analyses and operational limits are based on structure of technological components and characterize their function and usage or exclude some phenomena that do not relate to the safety analyses directly.
Current methods of long-term safety demonstration for repository come out from scenario development in the future. In the post-closure safety of Mochovce repository the following two types of scenario are assumed:

A. The evolution scenario assumes severe failure of the top cover of the repository so that infiltrating water saturates repository and gradual degradation of barriers occurs. The released radionuclides are assumed to migrate through the bottom clay layer and be transported via the aquifer to well near repository and small lake water.

B. The intrusion scenarios include construction scenario that assume that a road and multistorey building is constructed so that a part of the repository is subject to excavation after institutional period. However, as ICRP recommended to asses the exposure to the population around the site, a residence scenario is also considered.

The radiological protection criteria for members of the public were set by the Ministry of Health of the Slovak Republic as the radiation protection regulatory authority for both types of scenario:

- Effective dose for individuals from public for evolution scenario (scenario where probability will be equal 1) must be less than 0,1 mSv/yr during each year after institutional period
- Effective dose for individuals from public for each intrusion scenario (scenario where probability will less than 1) must be less than 1 mSv/yr.

These limits were taken into account during the derivation of the acceptance criteria for the radionuclides contained in the waste. Generally, calculation endpoint of disposal facilities performance assessment is radiological impact on humans and environment. In that case, starting points of assessment are the waste activity concentrations and inventory activity. In Mochovce repository safety analysis, end points are both the concentration per package and total activity values for following 19 radio nuclides: ^{14}C, ^{59}Ni, ^{63}Ni, ^{79}Se, ^{90}Sr, ^{93}Mo, ^{93}Zr, ^{94}Nb, ^{99}Tc, ^{107}Pd, ^{126}Sn, ^{129}I, ^{135}Cs, ^{137}Cs, ^{151}Sm, ^{238}Pu, ^{239}Pu, ^{241}Am. On the other hand, radiological protection criteria are the starting points of the calculation. This approach was developed and applied because the actual inventory that will be disposed of is highly uncertain.

Beside criteria which confine activities of radio nuclides in the waste form and repository important from the point of safety, part of the limits and conditions is type of waste form, its structural stability, leachability, thermal and radiation effects, the possibility of gas generation, microbial degradation and inception of a critical state, the content of corrosive, explosive and self-ignition materials, free liquids and complexing agents, corrosion resistance and surface contamination of waste packages and dose rate, dimensions, mass and labeling.

4.2 Packaged forms of RAW

Only radioactive wastes fixed in proper matrix in Fiber Reinforced Concrete Containers can be stored in the repository, integrity of the containers is guaranteed at least for 300 years. Functional FRC presents an important barrier against water penetration from box to waste and against migration of radionuclides from waste form. Due to high mechanical resistance this barrier has a significant task in providing the stability of entire repository. Strict strength and tightness requirements are imposed on the containers. They have to be resistant against repeated temperature changes, and have to demonstrate radiation stability also after filling with modified wastes. The containers have a cubic shape with height of 1,7m and minimum wall thickness of 11,5cm, internal volume of 3,1 m^3. Weight of empty container is 4300kg.

Set of disposable waste packages had been approved by regulatory authority on the basis of the operator proposal, i.e. according outputs of waste management system valid at the time of facility commissioning in 1999 year:

- containers filled homogeneously by cemented wastes,
- containers filled by drums with bituminised liquid wastes,
- containers filled by drums with waste metal pieces,
- containers filled by pellets from high pressure compaction of solid, non-combustible waste

Cementation is used for macro-encapsulation of drums and pallets in the FRC. Increasing experiences from conditioning of radioactive waste by new technologies provided new products like:

- solidification of radioactive chrompik into glass matrix of boric-silicate type,
- encapsulation of sludge into inorganic silicate (geopolymer) or cement matrix directly in the drums,
- ash homogenized with paraffin and high pressure compacted.

These new products together with their combinations increased the number of approved waste packages on current number of 19.

1.260 containers (17,5 % from the total volume) were disposed of in the repository from the starting of the Mochovce repository operation to the end of the year 2006 (see Table 6). From the disposed wastes, operational wastes predominate and radioactive waste from decommissioning of A-1 has minimal portion. One container with average weight of 3.983 kg of conditioned waste contains on average 4,1 drums and 3,55 pellets of compacted solid waste.

Table 6 Summary of waste disposed of in the Mochovce repository during the years 2001-2006

Year	Number of containers	Weight-operational waste [t]	Weight-waste from NPP A-1 [t]	Drums [pieces]	Pellets [pieces]
2001	115	553,85		537	550
2002	214	917,57	3,8	1.023	841
2003	240	880,78	107,30	1.130	783
2004	218	781,58	140,54	1.081	888
2005	238	855,4	124,0	1.214	994
2006	228	823,8	166,8	1.093	1.154

Table 7 shows the total activity of disposed containers and percentage of two most important radionuclides in total activity during years 2001-2006. The major contribution (91,5-98,0 %) to the total activity comes from ^{137}Cs. Contribution of other radionuclides is negligible.

Table 7 Total activity received and percentage of two most important radionuclides in the Mochovce repository during the years 2001-2006.

Year	Total activity [Bq]	% from total received activity (1st nuclide)	% from total received activity (2nd nuclide)	Total activity per container [Bq]
2001	1,53E+12	97,4 ^{137}Cs	1,20 ^{90}Sr	1,33E+10
2002	2,99E+12	97,4 ^{137}Cs	1,94 ^{14}C	1,40E+10
2003	2,41E+12	91,5 ^{137}Cs	7,11 ^{14}C	1,00E+10
2004	2,24E+12	98,0 ^{137}Cs	1,24 ^{90}Sr	1,03E+10
2005	1,09E+13	96,5 ^{137}Cs	3,10 ^{90}Sr	4,58E+10
2006	4.34E+12	96,2 ^{137}Cs	4,03 ^{90}Sr	1.90E+10

5. CONCLUSION

1. All installed and operated technologies for RAW treatment and conditioning and their mutual combination provide technological platform for processing of versatile spectrum of operational and decommissioning RAW.

2. Important result of common processing of used the technological procedures (equipments) in conjunction with final disposal of RAW is the reduction of the total volume of RAW.

3. The combination of using operating technologies considerably contributes to the optimal utilization of capacity of the National Repository of RAW whereas all of radiological and other limits are observed.

4. The most important criteria in the strategy of RAW processing are the proper choice of technology for each waste stream. All technologies assigned for RAW processing into the final form for disposal are sufficiently developed and are ready to be used for the processing of recent and future inventory of RAW. Nevertheless, the improvement and adaptation of these technologies is necessary for conditioning of some actual waste streams.