EXPERIENCE WITH TREATMENT OF LOW LEVEL RADIOACTIVE LIQUID WASTE IN SLOVAK REPUBLIC

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ABSTRACT

Presented paper deals with research, experience and treatment of low level radioactive liquid waste (LLW) in Slovak Republic.

There is shortly described operation of nuclear power plants (NPP) in Slovak Republic with a focus on their safe decommissioning. The decommissioning of A-1 NPP is already ongoing project in this area in Slovak Republic. Within this project it is necessary to treat with different kind of radioactive waste.

There has been realized extensive research at our Institute with treatment of LLW and its fixation in stable matrix. Experiments have been provided especially with materials such as spent resins, sludge, concentrate and other liquid waste.

The Nuclear Regulatory Authority of the Slovak Republic, after discussion with other relevant bodies, decides to fix these wastes mainly in cement or bitumen matrix using 200 l steel drums. The fiber reinforced concrete containers (FRC) are used for disposal of these products in National Radioactive Waste Repository at Mochovce, which is already in operation. Cementation procedure is used for fixation of prepared drums with fixed LLW or fixation of solid radioactive waste in FRC.

The laboratory for analyzing of radioactive waste properties and optimization of procedures for treatment with them has been established at our institute. Also the pilot plants for incineration, cementation and bitumenation have been built. The parameters of main equipment have been optimized on laboratory experiences. These plants were also used for treatment with LLW from NPP Jaslovske Bohunice (V-1, V-2, A-1 NPP). Their performance and main parameters are presented in paper.

The best results were obtained with cementation procedure. This method allows treatment with different type of LLW and it is essential for fixation of drums or waste in FRC, what is also in accordance with the main conception of disposal of low level radioactive waste in the Slovak Republic.

Good experience with cementation procedure has been used in several other projects in Slovak Republic as well as in abroad.

VUJE Trnava assisted at commissioning of BSC (Jaslovske Bohunice Treatment and Conditionig Centre). Within the A-1 Decommissioning Project VUJE's pilot cementation plant is used for cementation of different kind of sludge, concentrates and furthermore for cementation of materials such as radioactive gravel and ashes.

The design and procurement of cementation facility for treatment with tritiated water at Salaspils, Latvia, was recently done. Its parameters and performance is described in presented paper.

In accordance with our good experience with cementation and bitumenation of LLW VUJE Trnava is already involved in design and procurement of Final Treatment Centre of Liquid Radioactive Waste at NPP Mochovce as a general contractor.

INTRODUCTION

Nowadays there are 6 units in operation in the Slovak Republic (1st and 2nd unit of V-1 NPP, 1st and 2nd unit of V-2 NPP and 1st and 2nd unit of Mochovce NPP), 2 units are under temporary stopped construction (3rd and 4th unit of Mochovce NPP). All mentioned reactors are of Russian design WWER-440, type V-230 and V-213.

The first NPP operated in Slovakia was A–1 NPP. Its reactor unit was provided with reactor cooled by CO_2, moderated by heavy water and using as fuel natural uranium (HWGCR type). It was in operation since 1972 and was finally shutdown in 1977 after accident (level 4 according to the International Nuclear Event Scale). The reactor unit is in the preparation stage for decommissioning of its components. Some auxiliary buildings have already been decommissioned to the green field. The part of turbine building is used for processing and storage of radioactive waste. All spent fuel from the A-1 NPP was transported to Russia. Low level and intermediate radioactive waste from the main production unit was partially treated and conditioned.

VUJE has been chosen as a general procurement for the A-1 NPP Decommissioning Project. The main goal of the project is to obtain radiological safe status. Besides of others it also includes following activities:

- development of suitable decontamination and dismantling equipment and techniques,
- decontamination of specified rooms, technological equipment and structures to reduce smearable contamination,
- physical, chemical and radiological characterization of radioactive waste and its amount,
- development of technologies for treatment and conditioning of different kind of radioactive waste,
- conditioning of radioactive waste in acceptable form for its disposal in National Radioactive Waste Repository.

There is considerable amount of LLW, which was produced during operation of NPP and is stored at A-1 NPP. The other radioactive waste originates from decontamination and dismantling activities. It was necessary to make a balance of all radioactive waste and its characterization. Radioactive wastes were divided into three basic groups according to their future treatment: combustible, solid and liquid waste. The Study of Complex Strategy for Treatment with

Radioactive Waste has been done [1]. Several technologies for treatment with waste were optimized in laboratory scale, with a focus on incineration of solid waste and bitumenation or cementation of liquid waste and ashes. The products of these technologies (200 l steel drums with waste fixed in bitumen or cement) are loaded into the fiber reinforced concrete containers and fixed in cement matrix. The National Radioactive Waste Repository for disposal of FRC, containing the low level waste, has been built at Mochovce and started its operation in 2002.

It is clear, that it is essential to manage cementation process for treatment with LLW in Slovak conditions. The laboratory results obtained at VUJE with cemntation of different radioactive waste have been used for design and construction of several cementation plants.

LABORATORY ANALYSES

There has been realized extensive research at our Institute with treatment of LLW and their fixation in stable matrix. The laboratory for physical, chemical and radiological characterization of radioactive waste has been established. Experiments have been carried out especially with liquid LLW such as spent resins, different kind of sludge, concentrates and other liquid waste. The laboratory provides measurement of waste properties such as:

- viscosity,
- density,
- distribution of particles,
- overall gamma activity,
- basic radiological composition,
- chemical composition.

Later the laboratory was equipped with necessary equipment for study and control of bitumention as well as cementation procedure. There is equipment such as different kind of mixers, laboratory presses, conditioning chambers, calorimeters etc.

The percentage of fixed waste salt (or fixation coefficient f) in final products must respect radiation limits for their disposal at the National Radioactive Waste Repository.

VUJE PILOT PLANTS FOR TREATMENT OF WASTE

The pilot plants for incineration, bitumenation and cementation of radioactive waste were built up at our institute. The parameters of their main equipment had been optimized on laboratory experiences. Experiments have been done with real waste and in real scale. Results were used for further improvement of design of main apparatuses and parameters of processes.

These plants were not used just for experiments, but also for treatment of LLW from Jaslovske Bohunice NPP (V-1, V-2, A-1, NPP).

Incineration Plant

Incineration plant is used for treatment of solid combustible radioactive waste, such as paper or plastic materials, used protective clothes, mops, pieces of wood etc. The waste is packed into approximately 200 l plastic or paper bags. Incineration facility consists of furnace, ducts, cooler of off-gas, bag filters and HEPA filters. The entire incineration plant operates at negative pressure to ensure that all off-gas pass through HEPA filter.

Bag of waste is put into furnace through gate on its top and placed on the furnace grid. Combustion temperature range from 750°C to 950°C according to the treated waste in bag. Off-gas is led through ducts, cooled and filtered prior to exhaust to stack. Produced ash is settled in 200 l steel drums under furnace. Samples of ash are regularly taken and evaluated prior to future treatment (indrum cementation). Temperature, pressure and flow of air are measured and controlled.

Bitumenation Plant

Remarkable consideration in research has been paid to bitumenation process [2]. Concentrates, spent resins, sludge or their combinations are treated by this technology. The pilot bitumenation plant (see Fig. 1) consists of these main apparatuses: storage tanks of bitumen, concentrate, sludge or resins, film evaporator with rotating wiping blades, filling equipment, roller conveyor, cooling chamber and lidding equipment.

Waste and bitumen are dosed to evaporator on its top, wiped by blades, concentrated and homogenized and bitumen product is discharged into 200 l steel drum fixed under the evaporator. The drum is moved to cooling chamber where bitumen product cools off.
The film evaporator with rotating wiping blades is the main apparatus of the plant. Its main parameters are following:

type:	film evaporator with wiping blades,
rotor speed:	500 min^{-1},
heating medium:	steam at temperature 185 °C,
heating surface:	2.0 m^2,
output:	120 l of evaporated water per hour,
inside diameter:	360 mm,
total length:	3500 mm.

Fig. 1 VUJE bitumenation plant

Samples of bitumen products are taken at the bottom part of evaporator and evaluated prior to future treatment. Pressure of heating steam, temperature conditions of evaporator and dosing rate of bitumen and waste are measured and controlled. Fixation coefficient is approximately $f_b = 0.4$, according treated waste and process conditions.

Cementation Plant

Cementation plant is used for fixation of spent resins, concentrate, sludge, ashes, and gravel in 200 l steel drums. It consists of dosing bin of cement, dosing tank of waste, mixing unit and fixation device of drum. Cementation formulation is prepared on results of experimental test of waste samples. Appropriate amounts of materials are loaded into dosing vessels. Ashes or gravel are loaded into drum prior to cementation. Mixing of waste, cement, additives and/or service water is performed directly in 200 l drums (indrum cementation), which is fixed under mixing unit.

The mixing unit consists of filling lid, which is fixed to the top of the drum and rotation shaft equipped with mixing blade. Materials are gradually filled into drum through filling lid while mixer shaft rotates and also move up and down inside the drum. After homogenization the filling lid is disconnected from drum, mixer shaft is pull out of the drum, samples of cement matrix are taken and the drum is removed to curing place. The different mixing blades were used, according treated waste.

Capacity of the cementation plant is approximately 3 drums/shift of cement product and water coefficient is approximately $c_w = 0.42$ for cementation of concentrate.

DESIGNED AND PROCURED FACILITIES FOR TREATMENT WITH RADIOACTIVE WASTE

Obtained experimental experience as well as experience with design and operation of plants has been used for design and procurement of other treatment plants for radioactive waste.

Cementation Plant for Treatment of Tritiated Water

The VUJE as a winner of international tender, organized by IAEA has procured the cementation facility for treatment of tritiated water at Reactor Research Centre Salaspils, Latvia.

The experimental reactor in Salaspils is a Russian design pool IRT type and was shut down on 19th June 1998. The cementation facility is part of complex system of treatment with radioactive waste. Concrete containers reinforced by steel are used for disposal of LLW in Latvia.

The cementation facility is dedicated for immobilization of tritiated water of reactor pool in cement matrix. Cement grout is used for fixation of low level solid radioactive waste, which is loaded into concrete containers. The cementation facility (see Fig. 2) is located in reactor hall and therefore the disposition restrictions had to be respected.

The capacity of cementation facility is 3.6 m^3 per shift. Cement grout is prepared in batch mixer. Cement, tritiated water and aqueous additive are dosed into the mixer. The volume of homogenize batches can range from 20 l up to 120 l. Cement is fed from cement storage bin to weighting unit by screw feeder and then to mixer. The tiritiated water is fed to mixer by pump from its existing storage tank. Service water or splashing water can be dosed instead of tritiated water. The aqueous additive is dosed to mixer from agitated storage tank. After homogenization cement grout is pumped to container, which is placed on vibration plate. Vibration plate provides that cement grout better leaks within solid radioactive waste loaded in container. Autonomous ventilation and filtration unit provides negative pressure in the whole system to avoid cross contamination. The control unit allowed automatic or manual preparation of cement grout. It is connected with remote control and archiving PC, where all dosed amounts of materials as well as other parameters of process are recorded.

The amounts of all materials are dosed according prescribed formulation and required volume of batch. Formulation is estimated on laboratory experiments. Necessary laboratory equipment and measurement procedures were part of procurement.

The cementation facility started its operation in 2002. About 40 containers with radioactive waste fixed in cement matrix were produced up to this paper submitting.

Fig. 2 The cementation facility for immobilization of tritiated water, Salaspils, Latvia.

Movable Cementation Facility

Important part of the A-1 Decommissioning project is treatment of liquid radioactive waste and sludge or gravel stored in underground storage tanks at A-1 NPP. VUJE as main procurer of this project designed the Movable Cementation Facility (MCF) for treatment of this waste. MCF employs indrum mixing. It means that all materials and waste are dosed into drum, mixed and after that mixing blade is pull out of drum. Main technological apparatuses are mounted in ISO containers and thus the whole facility is easy to transport and build up next any other tank with stored waste. MCF is also provided with equipment for fixation of waste in matrix such as SIAL or phosphate ceramic.

Technological flow sheet of MCF is shown in Fig. 3. Waste from underground tank is pumped to agitated storage tank (see pos. 1a), from which samples of sediment are taken. Cementation procedure (ratio of sediment, cement, water and plastification additive) is estimated on results of radiochemical analyses and estimation of dry residue ratio of samples. After that the sediment is pumped into operation tank (1b). The drum is fixed on roller conveyor (20), which provides its automatic movement along the facility. In the filling position (10) sediment from operation tank and if it is necessary service water are fed into drum. After that the drum is moved to and fixed in mixing position (23) below mixer unit (12). Mixer's shaft with blade is inserted to the drum and homogenizes its content. Appropriate amounts of cement and plastification additive are weighted in weighting units (3, 22) and dosed to the drum while the blade is rotating and moving up and down in the drum. Time of homogenization is approximately 30 min. Then mixer's shaft with blade is pulled out and the drum is moved to vibration and sampling position (14). Samples of grout are taken, dose rate and smearable contamination on surface of drum are measured and the drum is vibrated to achieve better compactness of content. When free water is disappeared the drum is remotely lidded (38) and then the drum is removed from conveyor to curing place.

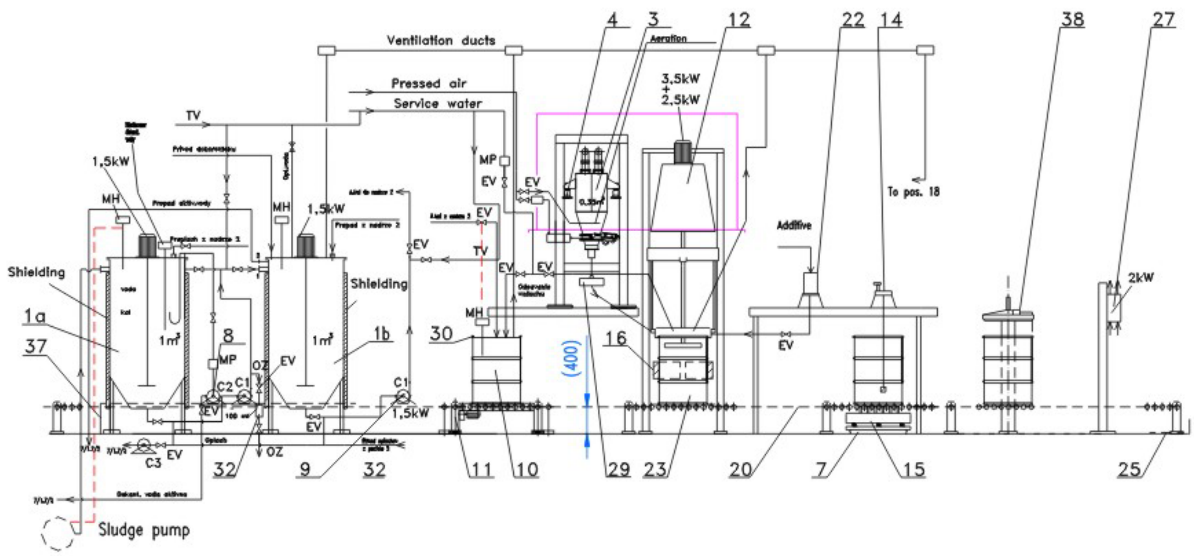

Fig. 3 Technological flow sheet of movable cementation facility

Autonomous ventilation and filtration unit provides negative pressure in the whole system. Main technological apparatuses of MCF are mounted in ISO containers and shielded. The most of technological operation are automatic and they are controlled from separate control unit. Expected output of MCF is from 3 to 4 drums per shift of cement product.

At the time of this paper submittal, the design and manufacture of main parts of MCF is in final stage. Initial mockup testing is planed in the spring 2004. After mockup tests MFC will be used for treatment of sludge stored in underground tanks of Active Water Purifying Station of A-1 NPP at Jaslovske Bohunice.

Design of Final Treatment Center for Liquid LLW in Mochovce

VUJE as an experienced company in the field of bitumenation and cementation was chosen as general contractor for design and procurement of Final Treatment Center for Liquid LLW in Mochovce (FTC, see Fig. 4).

FTC will process Liquid LLW produced at NPP Mochovce. This waste comprises radioactive concentrate, spent resins and sludge. Two main technologies will be used – bitumenation and cementation.

Treatment of concentrate will be provided by bitumenation. Salt of concentrate and bitumen will be mixed in film evaporator with rotating wiping blades and discharged into 200 l steel drums. Spent resins and sludge will be decanted, dried and mixed with bitumen. Mixture is discharged into 200 l steel drums. Samples of bitumen products are taken at the bottom part of evaporator/mixer. Drums are moved along bitumenation line on roller conveyor. After they cool off they are lidded and removed from conveyor to storage place.

Bitumenation facility consisted of these main apparatuses: storage tanks of concentrate, spent resins, sludge and additive, tanks of bitumen, film evaporator, decanter, drier, mixer, filling stations and roller conveyor.

It is supposed that five campaigns of bitumenation of concentrate and one campaign of bitumenation of resins and sludge will be done per year.

Drums with bitumen product will be loaded into fiber reinforced concrete container and grouting with cement grout. Cement grout will be prepared from mixture of cement, additive and active concentrate. Batch mixer with rotated shaft equipped with blades will be used for homogenization of cement grout. Containers with bitumen drums are placed on roller conveyor, which move them along cementation facility.

Cementation facility consisted of these main apparatuses: Storage bins of cement and additive, operation tank of concentrate, weighting and dosing systems, mixer unit, roller conveyor and vibration plate. Cementation campaign will follow after each bitumenation campaign.

The project of FTC also proposes design of control system, ventilation and filtration system, manipulation with drums and containers, sampling, measurement and regulation, decontamination and treatment with secondary waste.

All operations of bitumenation and cementation are automatic and remotely controlled. The final product of FTC will be a filled fiber reinforced concrete container, which meets limits for final disposal in the National Radioactive Waste Repository at Mochovce.

The overall capacity of FTC will be:

- 870 m^3/year of concentrate,
- 40 m^3/year spent resins and sludge.

This represents:

- 1 205 drums with bitumen product,
- 172 fiber reinforced concrete container.

At the time of this paper submittal, the initial project was submitted for approval to the Slovak Electric plc. and the main technological apparatuses of FTC are in stage of design. Realization of project should start in 2004. Initial mockup testing is planed in the spring 2006.

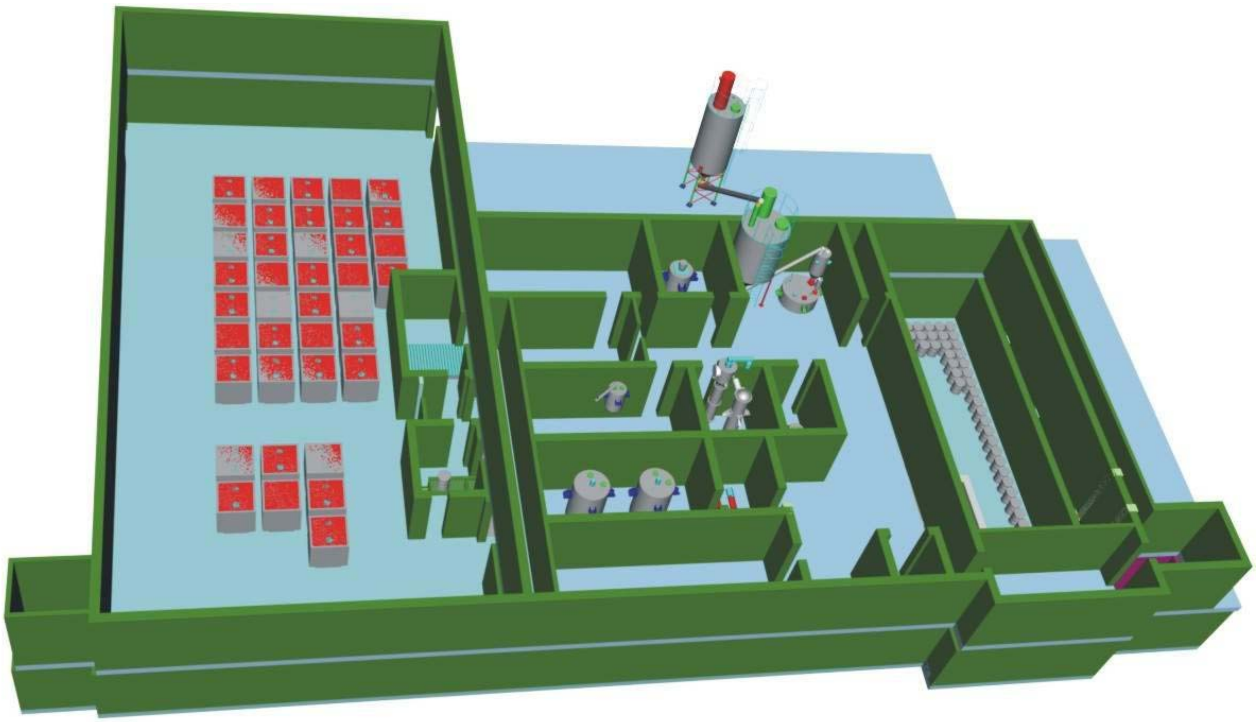

Fig. 4 Final treatment center for liquid llw at Mochovce.

CONCLUSION

Treatment with liquid LLW is based on precise physical, chemical and radiological characterization of waste. It is also important to know their properties, physical and chemical behavior during treatment procedure. The final product of any treatment procedure has to respect criteria for its final disposal. Measurement of relevant properties of final products it is therefore needful.

Laboratory tests are essential in optimization of parameters of process as well as for design of main parameters of equipment. However verification of the whole process on pilot plant is still the most useful.

Very often the radioactive waste has not been treated or conditioned up to now, but just stored. Thus it will be necessary to treat higher amounts and also different kind of them in the future. Claims on safety and economical operation of treatment plants as well as on the criteria for final disposal of waste products will rise too. Research in the field of radioactive waste treatment is therefore challenging and justifiable job.

REFERENCES

1 Hladký E., Timuľák J., Pekár A., Žatkulák M., Urbanec M., Holická Z.: „Complex strategy of treatment with radioactive waste." Study, VÚJE, Trnava, Slovak Republic, 03. 2000.

2 Breza M., Pekár A., Dávid L.: „Processing of radioactive wastes produced in Czechoslovak NPP by means of the bitumention technology." ICEM 93, ASME, Prague, Czech Republic, 1993.

3 "Improved cement solidification of low and intermediate level radioactive waste." International Atomic Energy Agency, Vienna, 1993.

4 Geser C., Hedin G., Johansson H., Torstenfelt B., Waltersten T.: "Versatile cement solidification of low and intermediate level radwaste with the moss system." WM'00 Conference, February 27-March 2, 2000, Tucson, AZ, USA.

5 F. M. Lee, The chemistry of cement and concrete, 1970.