NEW SYSTEM INSTALLED FOR NUCLIDE REMOVAL AND BORON RECOVERY AT THE PAKS NPP IN HUNGARY

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ABSTRACT

A new radioactive liquid waste treatment system is in commissioning phase at the Paks Nuclear Power Plant in Hungary. Fortum Nuclear Services Ltd from Finland has designed and supplied a NUclide REmoval System (NURES) and a boron recovery system. Other parts of the system were supplied by the Paks NPP, and the whole system is implemented in cooperation between the parties. The system is going to treat evaporator concentrates and other radioactive liquids. The purpose of the system is to recover boron for reuse and to separate target radionuclides (Cs and corrosion products) allowing release of the purified liquid.

The NURES system was originally developed to remove cesium from evaporator concentrates at the Loviisa Nuclear Power Plant in Finland, where it has been successfully operated since 1991. For the application of Paks NPP, the original NURES-technology has for the first time been complemented with boron recovery system. The boron that is dissolved into liquid waste will be re-used in primary coolant.

The new radioactive liquid waste treatment system consists of separate subsystems: the Boron Recovery System, the NUclide REmoval System (NURES), a system to destroy organic complexes, the Ultrafiltration System and a transport system for liquids.

Compared to conventional treatment methods, such as direct solidification with cement or bitumen, the new system minimises the volume of interim storage and the amount of waste to be disposed of and allows considerable savings in waste management costs. The system reduces the volume of the existing and future liquid wastes effectively. Depending on the quality of original waste the overall volume reduction factor for evaporator concentrates is expected to vary in the range of 40 to 90. The amount of secondary waste is dominated by the waste outcoming from the Ultrafiltration System. The commissioning of the new system is scheduled to take place in the shift of 2003/2004.

Today, industrial applications of the same technology in other countries cover also treatment of spent fuel reprocessing liquids, reactor coolant liquids, spent fuel pool waters, floor drains, groundwater and laboratory wastes and a wide range of target nuclides.

INTRODUCTION

Typical methods for the treatment of radioactive liquid wastes include direct solidification with cement or bitumen for final disposal, and purification of liquids with conventional ion exchangers or zeolites, and solidification of ion exchange materials for final disposal. Low selectivity of ion exchange materials has limited the applications of the conventional ion exchangers to liquids that contain only very low salt concentrations.

Highly selective ion exchange materials have given new possibilities for the treatment of radioactive waste liquids. Even liquids that have a very high salt concentration, like evaporator concentrates, can be treated with this technology.

Three highly selective ion exchangers, CsTreat®, SrTreat® and CoTreat, have been introduced into the market. These materials are totally inorganic and they are available in different grain sizes for both column and precoat applications. Selectivity factors of these new ion exchangers are on much higher levels than for the other commercially available materials. Unlike conventional resins and some new inorganic/organic composite resins or membranes, these ion exchangers can also be used in the expanding area of applications where there is the requirement for totally inorganic ion exchangers.

In power plant operations, when the goal is to minimize total costs from collection of radioactive liquids to their final disposal, savings are often obtained when smaller amount of liquid is coming for solidification. As an example, at the Loviisa NPP in Finland [1] the use of a new nuclide removal system has caused only a fraction of the costs, what the cementation of over 900 m^3 of evaporator concentrates would have cost.

Efficient use of highly selective ion exchange materials gives possibilities to optimize treatment of liquid effluents in a new way. Conventionally, part of the liquid wastes are treated with a moderate to low degree of purification and discharged, which still causes remarkable radioactive releases into the environment. The rest of the liquids are solidified for final disposal. New type of treatment with highly selective ion exchangers gives a possibility to change this practice. In many cases the amount of water to be released can be increased at the same time as the activity release is decreased and the amount of final waste to be disposed of is also decreased. This is possible even for high salt concentrates. Thus, many of the liquids that nowadays are solidified directly can be purified for release, provided that the chemical character of these liquids does not limit the release. On the other hand, liquids that contain high levels of radioactivity can be purified to lower level. Economic benefit from this kind of system modification can be evaluated only on the case by case basis.

In some systems, like in demineralizer, the savings may come from reduction of the volume of the ion exchanger. For example, if zeolite is used instead of CsTreat® for removal of cesium, even in moderately low salt concentration the amount of zeolite can be thousands times bigger than the amount of CsTreat®. The demineralizer system itself can be very advantageous compared to evaporation and cementation. For example, in Callaway NPP in USA, the use of CsTreat® in the demineralizer system has reduced the liquid radwaste processing costs, causing remarkable savings [2].

Reclassification of liquids can give high savings, if vitrification or even construction of a new system can be avoided by selective removal of radionuclides. In the project in JAERI [3], relatively high concentrations of Cs-137 and Sr-90 did not allow direct removal of an 11 m^3 batch of liquid to be transported for normal waste treatment at the Tokai-mura site. When this amount of liquid was treated with 20 columns, each containing 0.8 liters of CsTreat® and 0.8 liters of SrTreat®, purified liquid could be treated with the normal waste liquid treatment at the site.

SELECTIVE REMOVAL OF RADIONUCLIDES

CsTreat®, SrTreat® and CoTreat are relatively new products in commercial market and they represent highest selectivity factors in the market. Table I gives the comparison of the selectivity factors for different ion exchangers and shows, why CsTreat® has been regarded as the most powerful ion exchanger for cesium on the market. These materials are manufactured in Finland by Fortum Nuclear Services Ltd, and they have found several applications around the world [4-7].

CsTreat® was originally developed for treatment of high salt (total salt content 350 g/L) evaporator concentrates at the Loviisa NPP in Finland but has later found several applications for the treatment of other types of waste solutions, too. This ion exchanger is 100 % inorganic and its efficiency is based on its material, hexacyanoferrate, and it is very efficient for removal of cesium. Typical use of CsTreat® is in granular form in column operation. Column sizes from less than one liter to 250 liters have been used. Normal grain size is 0.25-0.85 mm, but sizes 0.15-0.25 mm and 0.85-2 mm have been used in test operations. Latest development is the use of powder, <0.15 mm, in precoat applications.

SrTreat® and CoTreat are 100 % inorganic ion exchangers based on titanium oxides. They are developed for removal of strontium and cobalt and other corrosion products, respectively. Typical use of SrTreat® and CoTreat is in granular form in column operation. Column sizes from less than one liter to 12 liters have been used. Normal grain size is 0.30-0.85 mm.

Table I Selectivity coefficients for Cs/Na exchange in commercial ion exchangers [4]

Ion Exchange Material	Concentration of Na (mol/L)	Selectivity coefficient $k_{Cs/Na}$
Sulphonic acid resin	not known	<10
Zeolite (mordenite)	0.1	450
Resorcinol-formaldehyde resin	6.0	11,400[a)]
Silicotitanate (CST)	5.7	18,000
CsTreat®	5.0	1,500,000

a) Selectivity coefficient calculated from measured distribution coefficient of 5450 ml/g.

CESIUM REMOVAL FROM EVAPORATOR CONCENTRATES

In 1991 at the Finnish Loviisa NPP, CsTreat® was for the first time commercially used for cesium removal. Since that time it has been in use for the treatment of high salt (total salt content 350 g/L) evaporator concentrates [1]. Average processing capacity of the media has been over 13 000 L/kg and the decontamination factor (DF) for Cs-137 has been above 1,000. Remarkable savings have been obtained in the Loviisa NPP in the waste treatment costs by use of CsTreat®. Until now, over 900 m^3 of evaporator concentrates have been treated with 112 liters of CsTreat®. Volume reduction factor (VRF), calculated for the treated waste volume, was over 8000.

BORON AND NUCLIDE REMOVAL SYSTEM FOR PAKS NPP

General Description

Paksi Atomerőmű Részvénytársaság, the operator of Paks NPP (PWR, VVER-440) in Hungary, is currently implementing with Fortum Nuclear Services Ltd a new radioactive liquid waste treatment system.

The new system will be a part of the liquid waste treatment system of Paks NPP, and it is located in the auxiliary building 1. The technology consists of several subsystems: boron recovery system, cesium removal system, a system to destroy organic complexes, an ultrafiltration system and connecting pipelines, new connecting pipelines to the existing tanks, pumps and existing tanks and equipment (see Fig. 1).

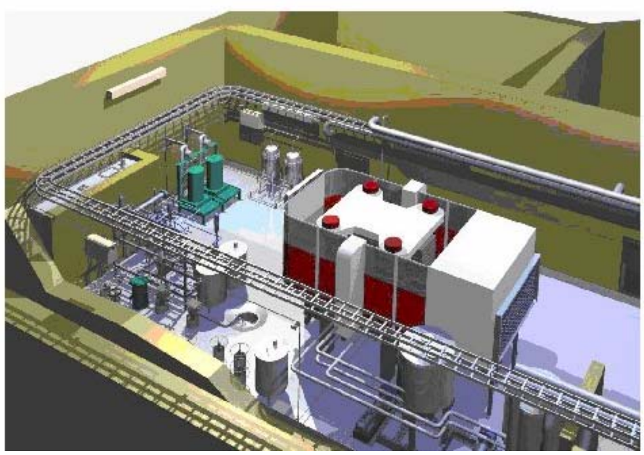

Fig. 1 The new liquid waste treatment system of Paks NPP

The boron recovery system is used for separation of boron from evaporator concentrates for reuse. Boron will be separated into a solid alkaline borate cake, interim-stored, and can be reused as boron feed.

The ultrafiltration system is used for removal of particulate material from liquids. The principle of operation includes separation of liquid with ionic material with big volume from small volume with concentrated particulate material.

Cesium isotopes, Cs-134 and Cs-137, are removed with an own purification system. Cesium removal is combined with the ultrafiltration system to form a bigger Nuclide Removal System. Their capacities are adjusted to match each other.

A new subsystem to destroy organic complexes is used to release radionuclides, especially cobalt, into ionic form for easier removal.

The overall treatment capacity of the system is up to 240 l/h. Decontamination factor for caesium is to be better than 1000 when DF factor is calculated as the ratio of caesium concentration in the input per caesium concentration in the output.

The existing liquid waste system is complemented with a new pumping system, new pipelines etc, to make it possible that all types of wastes, e.g. liquids and sludge, can be transported for further treatment in this new treatment system or for example in a cementation system.

Fortum Nuclear Services' delivery was designed, fabricated and installed, and it will be commissioned according to the ASME/Hungarian nuclear standards. Quality assurance and control is based on Fortum Nuclear Services' standards. The Hungarian authorities have performed inspections at the different stages of the project.

This new system does not have connections to the emergency and operation systems of the reactors, and it does not affect the safety of Paks NPP. The new added part of the process system is designed in such a way that in emergency situations - malfunction of components, malfunction of important measurements, and leaks - the system stops operating. Should, however, the system not stop operating, the most important alarms go to the control room, where operators can stop the operation of the system. If needed, original operation of the whole TW-system can be returned by closing the operation of the new system. In this case, the new system will be separated from the original system. Releases from Paks NPP will take place through the existing routes.

The activity data from Paks NPP have been taken as design values. The system has been designed for treatment of existing and future liquid wastes. The amount of liquid waste, what is generated in normal operation of Paks NPP within one year is about 250 m^3 altogether from 4 blocks.

Boron Recovery System

The Boron Recovery System will separate boron from the evaporator concentrates. For crystallization pH is adjusted under controlled conditions in the tank. Separation of alkaline borate is carried out by a highly efficient pressure filtration unit, recovering a relatively dry and clean cake of alkaline borate. The capacity of the pressure filtration unit is 1 m^3/h of liquid. The system is automatically controlled [8].

At least 70 % (wt) of boric acid content of the original waste shall be recovered in a reusable form and quality in solid form. Filtrate will include some 20 g/l of boron and it is taken for further treatment. After the boron recovery the liquid is led to the Nuclide Removal System.

Nuclide Removal System

After boron recovery Cs-134 and Cs-137 are removed from the waste liquid with NURES using CsTreat®. The capacity of the system is up to 240 l/h with two parallel ion exchange columns. Decontamination factor for cesium will be better than 1000 when DF factor is calculated as the ratio of cesium concentration in the input of the cesium removal system to cesium concentration in the output of the system. The system operates with automatic control. The automatization and electric supply system are common for the ultrafiltration and cesium removal system. The capacity of the system can be easily increased by adding ion exchanger units. Designed volume reduction factor will be about 8000. (in the case of Loviisa NPP it has been over 8000.)

As auxiliary materials the cesium removal system needs only acid for pH control, if pH control is needed. Otherwise the liquid is led directly to the cesium removal system. Purified liquid goes to the storage tank and from there to the control tank for release.

Performance of CsTreat® and CoTreat for removal of cesium from evaporator concentrates and for removal of corrosion products from pool water has been tested.

Complex Destroying System

After cesium removal the liquid is treated in a special system to destroy organic complexes. When radioactive elements are released from complexes the liquid is led to the Ultrafiltration System which is used for removal of particulate materials from liquids. From the Ultrafiltration System the liquid can be led to the NURES for removal of ionic corrosion products with CoTreat.

Ultrafiltration System

The ultrafiltration system is used for removal of particulate material from liquids. The principle of operation includes separation of liquid with ionic material with big volume from small volume with concentrated particulate material. The system is automatically controlled. Automation and activity measurements are common for the combination of the ultrafiltration system and cesium removal system. The liquid is purified in this system and after this ionic corrosion products can be removed with CoTreat in NURES, or liquid can be collected for release.

In the ultrafiltration test at the Paks NPP the Decontamination Factor (DF) for nuclides Mn, Co, Nb, Zr and Ag altogether as an average has been higher than 100, giving that less than 1 % of particle bound activities will be in purified liquid. Maximum flow rate in purification to reach this DF is 240 l/h.

The new liquid waste treatment system will give an overall volume reduction factor in the range of 40 to 90, depending on the quality of original waste. All above mentioned systems are connected to the liquid waste treatment systems of the Paks NPP.

Operation Personnel

Operation of the new liquid waste treatment system does not require new persons. Process experts, who work in auxiliary buildings, and reactor experts, and specialists of waste management, as well as shift specialists, can be trained for operation of the new system. In each shift there has to be at least one person who can operate the system. Normally, the system does not require continuous supervision. In one shift the parameters of the system are monitored.

When boron recovery system is operated, in transportation of recovered alkaline borate two persons are needed for each three hours to remove boron. For transportation of spent cartridges and membranes two persons are needed.

Total need for operation persons is one person for supervision of the system, using 1,5 hours in an operation day, and two persons for operation of the boron recovery system, using totally 4 hours in an 8 hour operation time.

The reliable and safe operation of the implemented system will be supported by the experience gained on similar installations at Loviisa nuclear power plant and other locations.

APPLICATIONS FOR DIFFERENT TYPES OF LIQUIDS

Applications for floor drain waters

At the Olkiluoto NPP in Finland, 240 m^3 of low-salt water was purified with one 12 liter CsTreat® column, giving a volume reduction factor (VRF) of about 20,000. Depending on the flow rate the decontamination factor was from 100 to over 1,000.

In July 1996, a 250 liter bed of CsTreat® was installed at the Callaway NPP (PWR), Missouri, USA in the demineralizer system to replace evaporator [4]. During about three years, over 3,000 m^3 of low-salt floor drain water was treated with this bed. Ionic cesium was removed to a non-detectable level. VRF was about 12,000.

Applications for cooling and pond waters

In 1996, 760 m^3 of various waste waters, originating from the operation of ex-Soviet naval training reactors, were purified in Paldiskij, Estonia using one 12 liter CsTreat® column. No sign of bed exhaustion was observed when the treatment campaign was completed, which means that the processing capacity of CsTreat® exceeded 100 m^3/kg [9], and a VRF was over 8,300.

In 1996-97, a transportable NURES container, utilizing 12 liter CsTreat® and SrTreat® columns, prefilters and a carbon filter, was used in Murmansk, Russia to treat radioactive wastewaters from nuclear-powered icebreakers [9, 10]. These liquids contained up to 4.5 g/l of NaCl. Maximum DF's for Cs-137 and Sr-90 have been 1,000 and 5,000, respectively, average being

635 and 102 respectively. The NURES system appeared to be efficient for the removal of other radionuclides, e.g. Co-60 and Sb-125, too.

In 2000, the first application at the USDOE's site with CsTreat® was started at Savannah River Site. About 20 000 m^3 of contaminated water of the old disassembly pool was successfully decontaminated to demonstrate the efficiency of these materials.

Application for reprocessing liquids

In the summer of 1997, CsTreat® and SrTreat® were taken into use at Japan Atomic Energy Research Institute (JAERI) Tokai site for removal of Cs-137 and Sr-90 from alkaline reprocessing waste effluent. The original concentrations of 74 MBq/L of both Cs-137 and Sr-90 were reduced by DFs of over 1000 [3]. The project ended with good results in the year 2000.

CONCLUSIONS

After the bid invitation and an international competition the Paks NPP selected the technology from Fortum Nuclear Services Ltd for its new liquid waste treatment system. The system has been fully designed and licensed. Implementation of the system started in 2000 in cooperation of the Paks NPP and Fortum Nuclear Services Ltd. The implementation will be finished and the system tested and commissioned in the shift of 2003/2004.

The Paks NPP had the goal to reach very good volume reduction for its waste volumes to save in storage and disposal costs. According to the design of the new system and according to the experience from other applications of the same technology, there are reasons to expect that the new system will fulfill the expectations with good margin, being finally demonstrated during the test run after implementation.

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