ABSTRACT

During BN-350 reactor operations and also during the initial stages of decommissioning, cesium traps were used to decontaminate the reactor’s primary sodium coolant. Two different types of carbon-based trap were used – the MAVR series, low ash granulated graphite adsorber (LAG) contained in a carrier designed to be inserted into the reactor core during shutdown; and a series of ex-reactor trap accumulators (TAs) which used reticulated vitreous carbon (RVC) to reduce Cs-137 levels in the sodium after final reactor shutdown. In total four MAVRs and seven TAs were used at BN-350 to remove an estimated cumulative 755 TBq of cesium. The traps, which also contain residual sodium, need to be immobilized in an appropriate way to allow them to be consigned as waste packages for long term storage and, ultimately, disposal. The present paper reports on the current status of the implementation phase of immobilization, with particular reference to the work done to date on the trap accumulators, which have the most similarity with the cesium traps used at other fast reactors.

INTRODUCTION

Failures in highly rated fast reactor fuel pins can result in fission product contamination of the primary sodium coolant and cause consequential difficulties for plant operation and decommissioning. A number of the world’s fast reactors have therefore made use of cesium traps to remove the main contaminant of concern (principally the cesium isotopes Cs-137 and Cs-134). These traps represent a difficult challenge during reactor decommissioning, as the combination of high concentration of radioactivity with residual chemically reactive and potentially flammable alkali metals renders them unsuitable for consignment as waste without processing and immobilization.

During the different periods of BN-350 reactor operation two types of cesium traps were used: in-core devices designed by the Research Institute of Atomic Reactors (NIIAR), Russia (MAVR series, on the basis of low ash granulated graphite) and stationary devices jointly designed by the Institute of Atomic Energy (IAE NNC), Kazakhstan, Idaho National Laboratory (INL), USA and MAEC Kazatomprom, Kazakhstan (on the basis of RVC carbon material) [1]. The design of these cesium traps is shown in Fig. 1.

At present, 4 spent cesium traps of the MAVR series and 7 spent cesium traps developed jointly by IAE NNC, INL and MAEC Kazatomprom are stored at BN-350. The specific

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1 MAVR – Russian acronym meaning “compact adsorber for the removal of radioactivity”.
activity of cesium-137 in the NIIAR trap with maximum activity of cesium is equal to $2.6 \times 10^{13}$ Bq/kg. The maximum specific activity in one of the stationary traps is $8.9 \times 10^{13}$ Bq/kg. In terms of specific activity of cesium, the traps are comparable with the activity of spent fuel elements cooled for 10 years - $\sim 10^{14}$ Bq/kg.

![Diagram of Trap Accumulator design and Design of MAVR adsorber]

**Trap Accumulator design**

**Design of MAVR adsorber**

Both types of cesium trap have been designed and fabricated to withstand 50 years storage at BN-350, but they cannot be placed in long-term storage without being converted into a fire-protected and chemically inactive form to meet Kazakhstani regulations regarding radioactive wastes handling. As a result, it has been necessary to develop the technology to condition the cesium traps for long term storage and convert them into radioactive waste packages acceptable for long term storage in Kazakhstan.

The objectives of this immobilization project were:

- To identify and validate a suitable process for disposition of the cesium traps and their contents.
- To immobilize the traps and make them ready for long term waste storage.
The project to immobilize the accessible cesium traps at BN-350 is funded principally by the UK Government Department of Energy and Climate Change, under a continuation of International Science and Technology Center (ISTC) Project K-512. This project had previously been constituted with US funding to design, install and operate the trap accumulators, based on US designs used on the EBR-II reactor. For the immobilization phase, the Kazakhstan organizations, NTSC and MAEC Kazatomprom, undertook the work advised by UK and US technical collaborators, Nuvia Limited and Idaho National Laboratory (INL). Initial option studies began in 2004, soon after completion of TA operations. Immobilization of the traps is scheduled to be completed by the end of 2011.

REGULATORY BASIS FOR CONDITIONING SPENT CESIUM TRAPS

In order to condition spent cesium traps for their long-term storage / disposal it is necessary to have available waste acceptance criteria for placing conditioned cesium traps into the site for long term storage or disposal of radioactive wastes.

At present, Kazakhstan has no licensed site for long-term storage or disposal of radioactive wastes. Completely defined waste acceptance criteria could be developed only for an already operating disposal site or for a waste package which is fully characterized and for which a safety analysis report had been developed for its complete lifetime. For future RW disposal sites which have only a partial concept for their long-term management, only preliminary waste acceptance criteria could be developed. These criteria should conservatively determine requirements for the conditioned packages of radioactive wastes. Later, waste acceptance criteria could become less and less conservative during the development of the concept for radioactive wastes disposal site management up to the moment when they become the final criteria for operating the disposal site.

Kazakhstan has experience in developing waste acceptance criteria for disposal of some categories of radioactive wastes. Requirements related to the criteria for acceptance of low and medium level radioactive wastes for their disposal in near-surface disposal sites are specified in the regulatory document “Safety rules for near-surface disposal of radioactive wastes” (SRNDRW). These rules set requirements for near-surface disposal of solid radioactive waste packages and are aimed at excluding unacceptable risks of causing damage to health of the population and environment, now and in the future. These rules have already been developed but are not yet in force and are progressing through the extensive process of approval between ministries of the Republic of Kazakhstan.

SRNDRW formulates criteria for acceptance of radioactive wastes for near-surface disposal. It requires that radioactive wastes acceptable for near-surface disposal shall have the following properties:

Aggregative state: Wastes accepted for disposal should be only in the solid form. If wastes initially were liquid, they should be conditioned to a solid low leachability state (less than $10^{-3} \text{ g/cm}^2 \times \text{days}$ for Cs-137 and Sr-90).

Density: Wastes should be compacted in order to minimize their volume and to reduce the surface area over which leaching can occur.
**Radionuclide composition and waste activity:** Radionuclide composition of wastes, specific and total activity of radionuclides in the packages (maximum and average values for disposal site) should meet requirements established by the design of the disposal site.

Limits on specific activity of radionuclides are established on the basis of estimations of the radiological effect of the disposal site on the whole population, taking into account possible scenarios of radiation impact on individuals from the population during operation of the site, its decommissioning, and after its closure. In the estimations it is necessary to take into account that accidental or deliberate entrance of persons from the population to the disposal site during the established period of control may happen.

Specific activity should be averaged over the whole volume of the waste package. At the same time, the specific activity of the package for long-lived alpha-sources should be limited to 4000 kBq/kg and the averaged activity for all waste packages should not exceed 400 kBq/kg.

**Thermal stability:** Wastes should be stable against degradation arising from residual heat after disposal and any effect of external heat sources.

**Package Design:** Design of all packages (weight, volume, form and size) shall correspond to the designed storage and transportation conditions and be easy to operate.

**Mechanical strength:** Wastes and containers shall have sufficient mechanical strength to hold form and withstand rough usage.

The compressive strength limit for cemented wastes shall not be less than 50 atmospheres, including after exposure to a cumulative radiation dose of $10^6$ Gy.

A thermal destruction test shall consist of 30 cycles of heating to $+60^\circ C$ and cooling to $-40^\circ C$.

Packages should withstand vertical loads corresponding to the design of the near-surface radioactive waste disposal site.

**Stability:** Stability of wastes may be provided by the form of wastes or by the package. Medium level radioactive wastes shall maintain their stability during a period of not less than 300 years.

**Toxicity:** Content of chemically toxic, poisonous, pathogenic and infectious substances in wastes should be determined with adequate accuracy and limited as much as possible.

**Low dispersion ability:** In order to avoid surface contamination during handling of packages, they shall not contain dispersible wastes (dust, powder) and light contaminated materials (paper, polyethylene film) should be briquetted.

**Surface radioactive contamination:** The level of radioactive contamination on the external surface of packages shall not exceed dose limits established by the regulatory documents of Kazakhstan for service personnel.

**Chemical stability:** Wastes shall not contain strong oxidants, chemically and corrosion-active and unstable substances. Wastes shall not decompose and release gases and fumes. In case of the presence of organic or self-ignitable materials in wastes, they shall be processed into explosion- and flame-proof form. The amount of free liquid shall
not exceed 0.5% of the waste package volume. Liquids shall have pH from 4 to 11. For liquid wastes solidified in cement, free liquid shall have minimal pH= 9.

**Chemical compatibility:** The content of stable complexing substances used, for example, in decontamination, as well as possible chemical transformations in wastes that may increase their migration ability in future, shall be taken into account at the stage of preparation of wastes for disposal.

Medium level radioactive wastes containing long-lived radionuclides (with half-lives of more than 30 years), except transuranics, with total specific alpha activity more than $10^4$ kBq/kg, and/or transuranic radionuclides with total specific activity more than $10^2$ kBq/kg, shall be disposed in such a way that their upper level is located at a depth of not less than 5 meters from the daylight surface, or shall be provided with shielding barriers that have a service life not less than 500 years.

Another regulatory document “Safety requirements for collection, processing and storing of radioactive wastes” (SRCPS-2003) was put into force in Kazakhstan on August 1, 2003, to ensure safety when handling radioactive wastes. This document sets safety assurance requirements for the systems of collection, processing, conditioning and storing of low- and medium-level radioactive wastes from atomic energy utilizing bodies, where these wastes are formed and/or processed, including specialized radioactive waste processing plants and radioactive waste disposal plants.

SRCPS-2003 sets requirements for conditioned packages of radioactive wastes, namely:

The radioactive waste package must prevent unacceptable spread of radionuclides into the environment and must not contain:

- strong oxidants and chemically unstable substances;
- poisonous, pathogenic and infectious substances;
- biologically active substances;
- highly inflammable, explosive or fire-risk substances;
- substances that can detonate or decompose with explosion;
- substances that enter into exothermic reaction with water accompanied with explosion;
- substances that contain or are capable of generating toxic gases, vapors or sublimates;

The content of free liquid in the package must not exceed 0.5% of the package volume.

The requirements specified in the regulatory documents SRNDRW and SRCPS-2003 could be considered as preliminary requirements for acceptance of low and medium level radioactive wastes to be disposed in the near-surface disposal sites. In the future, they could be extended to the level of preliminary waste acceptance criteria for other classes of radioactive wastes which do not currently have requirements for their long-term management in Kazakhstan.

Processing of radioactive wastes without having an operational disposal site for accepting these wastes can be implemented based on the preliminary waste acceptance criteria established in accordance with the requirements for the conceptual disposal site. The type of disposal site such as surface site, near-surface site, or a site in deep geological
formations should be determined in advance before developing such preliminary waste acceptance criteria.

The BN-350 Cesium Trap Conditioning Program assumes that, after being conditioned, the spent cesium traps will be placed into a future near-surface disposal site for which preliminary waste acceptance criteria will have been developed as specified in the regulatory documents SRNDRW and SRCPS-2003. Such an approach allowed the start of works on Cesium Trap Conditioning Program before the creation of a licensed disposal site in the Republic of Kazakhstan. An important point in this case is to avoid the risk of costly work to re-condition cesium traps in the future to meet the final criteria for the operating disposal site.

CONSIDERATION OF OPTIONS FOR CONDITIONING OF TRAPS FOR LONG TERM STORAGE OR DISPOSAL

As the first stage of the Cesium Trap Conditioning Program a Technical Options Study of cesium trap disposal was implemented [2]. Following analysis, three options were chosen as most favorable for further consideration:

- Pack the traps into canisters identical to the packed fuel canisters and send them to secure storage near the “Baikal” facilities located within the former nuclear test site in Semipalatinsk, Kazakhstan.
- Drain sodium, fill-in the traps with some inert material (such as lead, plastic or other) without removal of sodium residues and send them to interim storage and eventual disposal.
- Drain sodium, process residues, extract the sorbent, pack it in sealed containers and send them to interim storage and eventual disposal.

More detailed consideration of these options made it possible to choose the priority directions for further scientific investigations. As a priority technology that required additional experimental verification, it was proposed to consider conditioning of all the BN-350 cesium traps by filling with lead or lead-bismuth (Pb-Bi) alloy in accordance with the option “Drain sodium, fill-in the traps with some inert material without removal of sodium residues and send them to interim storage and eventual disposal”. Advantages of this technology over other ones are as follows: the technology has already been used previously in Russia [3, 4], relative safety of technological procedures, minimum volumes of radioactive waste; the technology is simple and economically effective; the conditioned packages are environmentally safe for long-term storage or disposal.

It was shown that upon filling the traps with lead it is possible to dispose of eight of the eleven traps immediately as medium level solid radioactive waste packages in a near-surface radioactive waste disposal site. The remaining two traps of the MAVR series and one Trap Accumulator can be disposed within approximately 40-50 years upon the decay of Cs-137 in the traps down to acceptable levels. Taking into account that currently in the Republic of Kazakhstan no near-surface radioactive waste disposal facility has yet been constructed, such a postponed solution is considered appropriate.

Considering the technology of trap filling with lead as the preferred option, evaluations for safety substantiation were performed. It was shown that release of radionuclides into gas cavities and release to atmosphere during trap conditioning, as well as sodium and
cesium leaching from the lead compound on contact with water, would correspond to safety requirements and requirements for handling radioactive waste. A program of experiments aimed at verification of calculations performed in substantiation of the proposed technology was proposed.

The next stage of the Cesium Trap Conditioning Program included the experimental program to do the necessary testing and qualification of the selected options, focused on the options of filling with lead or Pb-Bi.

EXPERIMENTAL SUBSTANTIATION OF OPTIONS FOR CONDITIONING OF TRAPS FOR LONG TERM STORAGE AND SELECTION OF PREFERRED OPTION

Experimental substantiation of options for conditioning of traps for long term storage and selection of the preferred option has been successfully accomplished [5]. The following set of experiments has been carried out: filling of a full-scale cesium trap mock-up with sodium followed by its draining from the mock-up to determine the optimal regimes of draining; filling in bench-scale cesium trap mock-ups with sodium and cesium, followed by sodium draining and filling with lead or Pb-Bi alloy at different temperatures and filling rates to choose the optimal regimes for filling BN-350 spent cesium traps; implementation of leachability tests to determine the rate of cesium/sodium release from the filling materials into water.

It was demonstrated that more than 80% of the sodium could be drained from the Trap Accumulators as the initial phase of their conditioning. This drained sodium can subsequently be processed together with the bulk sodium from the reactor primary coolant circuit.

The experiments on small scale mock-ups demonstrated that lead filling at 350°C gave good filling and wettability for both RVC and LAG. After cooling down the filled mock-ups, some small cavities were found in the samples, due possibly to shrinkage of the filler during solidification as well as by gas release from the sorbents during the fill, see Fig. 2.

These small cavities are not considered detrimental to the effectiveness of the immobilization of the activity in the traps. Sodium was found to be distributed rather
homogeneously in the filler with some preference to segregate at the grain boundaries, resulting in high brittleness of the filler.

Filling in mock-ups with lead-bismuth at 250-280°C was carried out and good filling and wettability of both RVC and LAG was demonstrated. However, a decrease of fill temperature down to 150-180°C resulted in problems with the filling procedure. It was only possible to fill one mock-up loaded with RVC. After performing cutting/slitting of the mock-up it was found that a plug had formed in the upper part of the inlet valve, which contained an increased concentration of bismuth and sodium. Other mock-ups could not be filled up with the lead-bismuth eutectic alloy\(^2\) at 150-180°C due to creation of similar plugs in the inlet piping to the mock-ups. It appears likely that a compound with increased melting point was created and this was not distributed homogeneously in the upper part of the alloy as had occurred at the higher (250°C) filling temperature. Instead it had floated to the surface in the form of a foam, plugging the valves at this temperature. An electron microscope photograph of the plug is represented in Fig. 3. The elemental composition of this plug was determined by means of scanning electron microscope type SEM JSM-6480LA and elevated concentrations of bismuth and sodium were found in the plug: Bi – 53,4% (mass); Pb – 9,2% (mass); Na – 15,4% (mass).

When filling the mock-ups with lead-bismuth alloy there was found to be a redistribution of lead and bismuth concentrations while it mixed with residual sodium in the mock-up. The ratio of lead-bismuth was about 1:1 at the inlet to the mock-up while when alloy was passing through the mock-up this ratio changed to 3÷5:1 where the bismuth concentration was decreased by several times. Thus, one may conclude that inter-metallic compounds of bismuth and sodium were created and these had tried to float to the upper part of the mock-up. Nevertheless, the filling/wettability of RVC with the alloy also appeared to be good for this case.

\(^2\) The eutectic composition is approximately 44.5% Pb/55.5% Bi by weight and has a melting point of 123.5°C.
The rate of cesium release from the sorbents into the gas phase was acceptably low during filling with both lead and lead-bismuth alloy. The maximum cesium release fraction from RVC was $1.6 \times 10^{-4}$, while this value for LAG was $10^{-5}$ when filling mock-ups with lead at 350$^\circ$C. Cesium release from RVC/LAG into the lead/lead-bismuth matrix did not exceed 15% in the filled mock-ups but the activity was retained within the solidified filler metal matrix.

The rate of Cs$^{137}\text{leachability}$ meets the Kazakhstani requirements for the whole mock-ups immediately after a fill procedure, while the rate of Cs$^{137}$ leachability for the half-sectioned mock-ups achieves the allowable value after seven days of leaching.

As a result of the experimental program it was identified that filling spent cesium traps with lead at 350$^\circ$C is the preferred option for their conditioning. Using the eutectic alloy of Pb-Bi was concluded not to be reasonable for conditioning the cesium traps. There are two main reasons: redistribution of lead and bismuth concentrations within the bench scale cesium trap mock-ups during the filling procedure, accompanied by the creation of bismuth-sodium inter-metallic compounds with high melting temperatures, which could lead to cesium trap plugging during conditioning cesium traps in the future and the comparatively high cost of this alloy. These disadvantages are not sufficiently counterbalanced by the advantages arising from the possibility of filling cesium traps at temperatures significantly lower than the lead melting point (327$^\circ$C). Initially, there was a belief that conditioning cesium traps at lower temperatures would result in a significant and desirable decrease of cesium release from the sorbent. However, this effect was not found to be very pronounced at the temperatures 250-280$^\circ$C, while further decrease of fill temperature caused the formation of plugs in the traps and in the system.

**CONDITIONING BN-350 SPENT CESIUM TRAPS**

During the current stage of the Cesium Trap Conditioning Program it is planned to condition all 7 stationary cesium traps and only one MAVR trap, which is currently located in the reactor hall of BN-350, by means of lead filling. Extraction of the three other MAVRs from the on-site solid waste storage trenches followed by their conditioning is to be postponed until the time when the full scale works of retrieving and conditioning BN-350 solid radioactive wastes conditioning will commence.

This scope of work is naturally divided into three distinct tasks. In the first task the Design Package of documentation for conditioning both stationary cesium traps and in-core cesium traps was developed. The second task includes procurement and fabrication of necessary equipment, its installation and check-out. After final installation of mechanical and electrical components, equipment will be set to work and commissioned and an operational readiness review will be performed. The third task will involve conditioning of all 7 TAs and one MAVR trap.

Design activities started with some preliminary analysis. As was mentioned in the previous section of this paper, the bench scale cesium trap mock-ups were successfully filled with lead at 350$^\circ$C and then demonstrated acceptable properties to meet preliminary waste acceptance criteria for their disposal. Nevertheless, when proceeding to the actual cesium traps scale factors could be an issue. While it was comparatively easy to maintain homogeneous temperatures across the bench scale mock-ups during the filling procedure,
it was unclear if homogeneity could be realized for the full scale cesium traps. It was also necessary to determine what temperature distribution should be maintained across the full scale cesium trap to avoid plugging during trap filling.

There was performed a simulation of filling Trap Accumulators with lead at different temperatures of the melt and trap in order to identify the critical regimes for conditioning TAs with lead [6]. A simple two-dimensional axially symmetric model was developed to perform thermo-hydraulic simulations of filling, see Fig. 4. The Lead-gas system was considered as a two-phase liquid with non-mixable phases. Solidification of the lead was simulated by changes in its viscosity; at temperatures below the melting point of lead its viscosity was assumed to be increased by several orders of magnitude.

Fig. 4. Two-dimensional axially symmetric model of TA (red colour – lead, blue colour- gas) at start of fill

Several options for filling the cesium trap with lead were considered: when lead is supplied into a cesium trap homogeneously heated up to 350°C; when filling a cold cesium trap at 27°C and when the trap case is heated up to 350°C, while the RVC bed and inlet pipe are cold and have a temperature of 27°C. Simulations demonstrated that a full scale cesium trap homogeneously heated up to 350°C could be filled completely within 20-30s without problems. Filling a cold cesium trap at 27°C results in filling only the bottom part of the trap where lead is solidified and plugs the trap. However, if a cesium trap is filled with the temperature of the trap case $T_{\text{trap case}} = 350^\circ\text{C}$, temperature of RVC bed $T_{\text{RVC}} = 27^\circ\text{C}$ and temperature of inlet pipe $T_{\text{pipe}} = 27^\circ\text{C}$ it could be filled completely with lead within 120s without any blockage due to solidified lead. For this option, during the filling procedure one can see some cold areas inside the RVC material but the energy capacity preserved in the melt is enough to heat up these areas and after a couple of minutes the cesium trap is completely filled with lead. Kinetics of cesium trap lead filling for these different options are shown in Fig. 5.
Fig. 5. Kinetics of cesium trap lead filling at different temperatures of trap components, demonstrating changes in the filler density during the filling procedure.
Additionally, the possibility of heating a cesium trap up to the temperature necessary for sodium draining immediately followed by lead filling was investigated. This was done in case the internal cesium trap heaters originally installed inside the biological shielding had been damaged during operations.

Thermal calculations demonstrated that cesium traps could be heated together with the biological shielding up to the necessary temperature of 350°C within 60 hours by means of external 5kW heaters covered by 100mm of thermal insulation. The temperature distribution field after 60 hours of heating is shown in Figure 6.

![Temperature distribution across cesium trap and biological shielding after 60 hours of heating.](image)

Similarly simulations have been done for the MAVR type cesium trap to substantiate conditioning of a full scale MAVR by means of lead filling.

At the present time the Design Package for conditioning TAs and the MAVR is complete. The Design Package was reviewed and approved by all parties involved in the immobilization project realization. Fabrication drawings related to installations for conditioning the cesium traps have been issued for manufacture. The Safety Analysis Report has been developed and approved. Analysis of potentially hazardous factors during operation of the installations has been undertaken. Design accidents/emergency situations were considered, together with measures required to mitigate the consequences of these events. Exposure/ inhalation doses for operations personnel and public were calculated for both normal operations of the installations and in the case of design accidents/emergency situations. Works for installing the equipment to condition the BN-
350 spent cesium traps are currently underway. Immobilization of the traps is scheduled to be completed by the end of 2011.

CONCLUSIONS

The BN-350 Cesium Trap Conditioning Program was organized in three main phases:

- Option studies and technical investigations – resulting in the selection of either lead (Pb) filling, as used by Russia for conditioning of MAVRs, or lead-bismuth (Pb-Bi) filling techniques.

- Confirmation of the preferred option for immobilization by means of bench scale trials of the selected process. The results showed that, despite the potential benefits from lower filling temperatures, the lead-bismuth system had potential problems with plugging due to the formation of inter-metallic compounds of sodium and bismuth. Hence the decision was made to use a combined approach of sodium draining (for the TAs), followed by lead filling which allowed the lead to displace any residual sodium remaining. An important finding was that there was a low rate of release of cesium into the ventilation system during filling, with a correspondingly high efficiency of retention of cesium within the lead-carbon matrix. Leachability of Cs-137 from test pieces was also shown to be acceptably low.

- A three year implementation phase, currently in progress, to immobilize the single available MAVR (the remaining units are currently held in the site waste storage vaults) and the seven trap accumulators.

The approach follows on from the lead filling process originally developed in Obninsk for the Russian-designed MAVR-type traps and is now being applied to the ex-reactor RVC-type traps. Immobilization of the traps is scheduled to be completed by the end of 2011.

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