Design & Development of Facility for Production of Active Cs-137 Source Pencils for Blood Irradiator

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Abstract:

The search for an option for a radioactive source, alternative to Co-60, is on for quite some time. Abundance of Cs-137, a high yield radionuclide, in the nuclear waste has prompted the idea of separation of the same from the nuclear waste and making it suitable for medical and industrial applications. This will give the societal benefits in addition to revenue generation. Longer half life of Cs-137 (30 Yrs) is an added advantage of using this isotope over Co-60. Hence, as a special emphasis in Indian Nuclear Waste Management programme, a facility has been designed, installed and commissioned for production of Cs-137 source pencils in vitrified form to be used as source pencils for blood irradiation.

Introduction

In India Co-60 is the isotope presently being used for medical applications. It needs frequent replacement because of its short half life (5.27 Years) resulting in multiple handling, transportation and loading-unloading operations. Further, in Indian scenario, production of Co-60 also leads to reactivity losses and in turn revenue losses to the nuclear power reactors. However, Cs-137, the major constituent of the nuclear waste, is a suitable substitute for Co-60 as a radioactive source because of its longer half life (30 yrs). Nuclear Recycle Group has given special emphasis on utilization of Cs-137 for such applications. This will give societal benefits in addition to the revenue generation and will also address issues related to disposal aspects of High Level Radioactive Waste.

Though traditionally Cs as Cesium Chloride (CsCl) powder has been used in radiation source applications elsewhere in the world, CsCl is highly soluble in water and the powder can get easily dispersed resulting in release of activity under accidental conditions. It is thus desirable to use Cs in immobilized form, with good product characteristics, so that high degree of safety can be ensured during its use in public domain.

Historical Background

In accordance with the Policy of Closed Fuel Cycle, India had developed an expertise in vitrification of High Level Nuclear Waste (HLW) and industrial scale vitrification plants have been set up at WIP-Tarapur, WIP-Trombay and AVS-Tarapur for vitrification of the HLW. Further, during refinement of Indian Waste Management Policy, emphasis was given on separation of actinides and high yield radio-nuclides like Cs-137 from the nuclear waste. Ion Exchange & Solvent Extraction Processes had been indigenously developed for separation of Cs-137. Separation of Cs-137, major heat generating radionuclide in HLW, will be advantageous in terms of reduced radio-toxicity of the remaining waste and reduced heat load during long term storage of such waste. In-spite of having expertise in vitrification of nuclear waste, the main hurdle in manufacturing of Cs-137 source pencils was the controlled pouring of the vitrified glass in stainless steel pencils.

Initial efforts for making Cs-137 source pencils were based on the concept of preparation of small spherical beads and putting them inside the stainless steel pencils. However, difficulty in making uniform beads due to corrosive nature of the vitrified product and presence of voids inside the source pencil, the bead making concept was not pursued further. Subsequently, it was decided to make the Cs source pencils based on the concept of direct controlled pouring of accurately measured vitrified product into stainless steel pencils.
Evolution of Glass Pouring Technology

The simplified block diagram of the activities involved in production of Cs-137 source pencils is shown in Fig. 1. Among all, the controlled pouring of vitrified product into the pencils is one of the most important activities. Mainly three design approaches were attempted progressively for controlled pouring of the vitrified product into the pencils as shown in Fig. 2:

a) Glass Bead Method

Initial concept of making Cs Source pencils was based on the preparation of 1-2 mm dia. glass beads to be filled into the pencils. The nozzles having 1 mm and 1.5 mm diameter opening were used for making the beads as shown in Fig. 2(a). However, it was observed that the method was not a practical solution because of the following:

i) Very high temperature requirement at the tip of nozzle to initiate the glass pouring.
ii) After few cycles of operation, the nozzle bore was getting enlarged due to the glass corrosion.

b) Freeze Valve Method

In this method, the vitrified mass is poured by energizing the bottom freeze plug directly into the source pencils ref. Fig.2 (b), a process similar to being followed at WIP. Multiple trials were carried out with different sets of temperature cycles. However, it was found to be very difficult to achieve the control over the glass viscosity and quantity being poured. Moreover, glass thread formation, which was not acceptable, and difficulty in predicting the start and end points of glass pouring were the shortcomings of this method.

c) Mechanical Plug Method

In order to achieve a fine control on pouring, the process pot design was modified with provision of a mechanical plug and seat type arrangement. Subsequently, the design was fine tuned through progressive modification of the plug and the seat (refer Fig.2 c, d and e). In this design, the plug was actuated through a pneumatic actuator to start / stop the pouring of vitrified product into the pencils placed on Product Trolley. This design has resulted in the most successful option among all the approaches tried for controlled pouring with which the pencils can be filled appropriately without any spillage of the vitrified product.

Applications & Advantages of Cs-137

Cs-137 Source Pencils will have following applications:

- Blood Irradiation.
- Food Irradiation.
- Brachytherapy.
- Sterilization of Medical Equipment.

At present, BARC has targeted the development of Cs-137 Source pencils for Blood Irradiation application only. Advantages of Cs-137 as Radioactive Source over Co-60 Sources are:

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**Fig. 1: Block Diagram of the Facility**

**Fig. 2: Progressive Designs of Process Pot**
• Reduced frequency of Source replenishment.
• Less handling and transportation.
• Less shielding requirement.
• Ready availability in nuclear waste.
• Reduction in radio-toxicity of remaining HLW.
• Reduction in heat load of remaining HLW, thereby resulting in reduced or no cooling requirements.

Present Facility

Technology Development Division (TDD) has rigorously worked on the critical aspects like design of mechanical plug based induction heated furnace which will ensure the controlled pouring of vitrified product at a temperature of around 950°C in SS pencils of 23 mm OD and 204 mm in length. The size of the pencil was dictated by the Co-60 source presently being used in the Blood Irradiators in India so as to keep the other dimensional parameters unchanged. The plant scale demonstration facility was designed to achieve the goal of direct controlled pouring of accurately measured vitrified product into these pencils. Subsequently, fabrication of all the required equipment and power supplies were completed and the demonstration facility was ready in March 2010. Full scale trials with various composition of glasses and simulated waste were carried out. After gaining the required confidence, through innumerable inactive trials, it was planned to install the plant scale facility for production of active source pencils.

Initially, feasibility studies were carried out in order to install the Cs Pencil making facility at the existing radioactive cells of WIP, Trombay. Subsequent to the feasibility studies, actual site measurements were carried out inside the radioactive cells. Further, the cell worthy set up was designed consisting of structure for Induction Furnace, compatible Inconel 690 Process Pot & Susceptor, Pneumatic actuation system, Product Trolley along with the Indexing arrangement, Load cell with appropriate shielding and flexible shafts to facilitate the placement of drive motors outside the hot cell. The main system, designed in a modular concept, with necessary remote handling interface has been installed in Area 19 of WIP, Trombay. WIP cask loading station has also been modified to facilitate the transfer of the active pencils to BRIT cask. The design has also gone through the safety approval processes through PLSC, ULSC and OPSRC route ensuring safety in all the stages of operations.

In general, the process of making Cs source pencils consists of vitrification of the separated Cs radioisotopes, dispensing of accurately measured Cs glass into the pencils, seal welding of the lid to the filled pencil, decontamination and transportation of the pencils to BRIT for end use in medical applications.

Sourcing of Cs-137 from HLW

Cesium-137, in appreciable quantity, is present in the high level radioactive waste. The presently available sulphate bearing waste has 2-3 Ci/l of cesium concentration. Apart from Cs, this high-level liquid waste at Trombay is characterized by the presence of strontium, actinides, high concentration of uranium and sodium. This waste is acidic (1-1.2 M HNO₃) in nature with average density of 1.2 gm/cm³. The waste is subjected to partitioning by three cycles of solvent extraction to remove different radionuclides. The solvent extraction process involves, i) removal of U using a solution of 30% TBP in dodecane, ii) removal of Cs using Calix Crown Ether solution having concentration of 0.03 (M) CC6 in 50% iso-decyl alcohol and dodecane and iii) removal of Actinides and Sr using TEHDGA solution having concentration of 0.4 (M) TEHDGA in 15% iso-decyl alcohol and dodecane.

The raffinate from first cycle, lean in uranium, after adjustment of acidity is contacted with calyx crown ether solution to recover Cs from the stream at a processing rate of 100 lph. The Cesium lean HLW (raffinate) serves as feed for the third cycle for removal of actinides & strontium. The loaded organic, after second cycle, is subjected to stripping with DMW. The stripped product from second cycle, essentially a cesium rich solution, is stored. This purified Cs-137 product solution is further evaporated, concentrated and fed for vitrification of cesium in glass matrix for making source pencils. The composition of the purified feed solution is given in Table 1.
Vitrification

Cesium nitrate solution after evaporation in existing evaporator is fed to the melter for vitrification process. The processing steps involved in vitrification are evaporation, calcination, glass melt formation, soaking and draining. During vitrification Cesium nitrate of concentration 20 Ci/litre is added to melter as the feed solution in controlled rate. The vitrification is carried out as per the standard operating procedure of WIP with existing system based on predetermined temperature profile. Approximately 6 kg of vitrified product is poured in the Cs pencil process pot. The composition and properties of the vitrified product are shown in Table 2.

Table 1: Composition of Purified feed solution

<table>
<thead>
<tr>
<th>Component</th>
<th>Concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td>HNO₃</td>
<td>3 M</td>
</tr>
<tr>
<td>Cs¹³⁷ (as CsNO₃)</td>
<td>20 Ci/L</td>
</tr>
</tbody>
</table>

Table 2: Composition & Properties of Vitrified Product

<table>
<thead>
<tr>
<th>Oxides</th>
<th>Wt (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO₂</td>
<td>28.8</td>
</tr>
<tr>
<td>B₂O₃</td>
<td>25.2</td>
</tr>
<tr>
<td>Na₂O</td>
<td>18.0</td>
</tr>
<tr>
<td>CaO</td>
<td>7.2</td>
</tr>
<tr>
<td>Fe₂O₃</td>
<td>9.0</td>
</tr>
<tr>
<td>TiO₂</td>
<td>1.8</td>
</tr>
<tr>
<td>Cs₂O</td>
<td>10.0</td>
</tr>
<tr>
<td>Laboratory pouring temp, °C</td>
<td>850</td>
</tr>
<tr>
<td>Pouring temp in plant scale, °C</td>
<td>900-950</td>
</tr>
<tr>
<td>Density, g/cc</td>
<td>2.5–2.7</td>
</tr>
<tr>
<td>Na leach rate, g/cm²/d</td>
<td>1.46 x 10⁻⁴</td>
</tr>
</tbody>
</table>

Table 3: Standard Specifications of Source Pencils

<table>
<thead>
<tr>
<th>Sr. No.</th>
<th>Description</th>
<th>Details</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Inner Pencil Dimensions</td>
<td>Ø23 mm, 204 mm long</td>
</tr>
<tr>
<td>2</td>
<td>Active Length</td>
<td>194 mm</td>
</tr>
<tr>
<td>3</td>
<td>Active Volume</td>
<td>70 CC</td>
</tr>
<tr>
<td>4</td>
<td>Sp. Activity of glass</td>
<td>1.78 Ci/g</td>
</tr>
<tr>
<td>5</td>
<td>Activity Content per Pencil</td>
<td>300 Ci</td>
</tr>
<tr>
<td>6</td>
<td>Lengthwise Activity variation</td>
<td>&lt; 20 %</td>
</tr>
<tr>
<td>7</td>
<td>Outer Pencil dimensions</td>
<td>Ø25.5 mm, 210 mm long</td>
</tr>
<tr>
<td>8</td>
<td>Surface Contamination of outer Pencil</td>
<td>&lt; 185 Bq</td>
</tr>
</tbody>
</table>
**Induction Furnace**

The induction heating furnace for cesium pencil facility comprises of Power supplies, Capacitor bank, High frequency bus bar and Coil assembly. This furnace has two (02) zones viz. 20 KW & 5 KW with separate power supply as shown in Table 4.

<table>
<thead>
<tr>
<th>Location</th>
<th>Power</th>
<th>Frequency</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zone-I</td>
<td>20 KW</td>
<td>5 KHz</td>
</tr>
<tr>
<td>Zone-II</td>
<td>5 KW</td>
<td>10 KHz</td>
</tr>
</tbody>
</table>

The capacitor bank for this furnace is made up of two sets of capacitors suitable for tuning 20 KW & 5 KW coils. The capacitor leads and copper connectors are water cooled and the electrical connections between capacitor bank and coil assembly is made up of high frequency bus bar.

**Product Trolley (Area 19)**

Product trolley is meant for receiving and positioning pencils below the induction furnace. The drives for the trolley and indexing are given through one of the wall penetration plugs through flexible shafts. This arrangement will enhance the operational life of the drive motors.

After re-melting of Cs loaded vitrified product its pouring into the source pencils of specified dimensions is one of the most important aspects of this facility. Small quantity and high activity of the vitrified product to be poured into the pencil have added to the criticality of the Product Trolley Design.

The Product Removal system mainly comprises of a locating trolley equipped with a pencil holding assembly and an indexing drive. The trolley has linear motion and the pencil holding assembly, mounted on the trolley, has rotary indexing motion. The trolley has been planned below the induction furnace and the pencil holding assembly can accommodate twelve (12) pencils. During filling of the pencil it has the provision of weight measurement of glass being poured. The material of construction of the Product Trolley is Stainless Steel (SS 304 L & SS 410).

The main features of the product trolley are:

1. Accurate positioning of the pencil below drain nozzle of the furnace through highly précised linear movement and indexing.
2. Minimum distance between Pencil top and the drain nozzle.
3. Accurate measurement of the glass being poured into the pencil.
Welding & Decontamination Station (Area 21)

The Welding & DC station has been planned in Area 21 of WIP. The decontamination baths, leak test baths and the Remote Welding Unit (RWU) have been installed on a platform within the reach envelopes of Incell crane and wall mounted pair of Three Piece Manipulators.

The RWU is a special purpose autogenous pulse TIG welding unit meant for the remote welding of the lids of inner and outer pencil. The unit mainly comprises of a remotely replaceable welding head and the Power Supply. Considering the small size of the pencil to be welded, a rotary chuck has been provided along with a stationary electrode holder for this purpose. The camera and a suitable optical viewing system have been provided to check the weld quality.

Tests for Cs-137 Source Pencil

In order to deploy, Cs-137 Pencil as a radiation source in public domain, AERB has prescribed the following mandatory tests to be carried on each and every filled pencil:

a) Leak testing as per AERB/SS/3(Rev.-1) standard: Finished sealed source to be immersed (at least 5 cm below the level) in a water bath, which is maintained at a temperature between 90°C - 95°C. Presence of leakage, if any, by way of bubble formation are to be observed for at least 2 minutes.

b) Loose contamination testing as per AERB/SS/3(Rev.-1) standard: Loose contamination on the sealed source is to be checked by taking swipe sample from outer surface of the pencil with a tissue paper. If the detected activity is less than 185 Bq, the sealed source is considered to be free from surface contamination.

Cask Support Platform

Cask Support Platform facilitates the accommodation of the BRIT cask in WIP Cask loading station for transfer of the source pencils, in specially designed transfer cages, from WIP Cell to BRIT Cask. Seal welded, tested and decontaminated source pencils have been planned to be transferred from the Area 21 to the BRIT Cask, placed outside, through this loading platform ref. Fig No. 8.
Remote Handling & Dismantling

In view of very high specific activity of Cs-137 glass to be handled and radioactivity inventory inside the facility, remote operation and maintenance features have been considered while designing the systems of the facility. The dismantling scheme has been designed with the features of remote removal of each and every subsystems of the facility.

Master slave three piece manipulators having 20 kg payload capacity have been deployed to assist the in-cell crane for handling the source pencils inside the cell. The maintenance scheme has been prepared based on the remote removal of the trolley top as well as the full trolley itself. The furnace system has been designed with features such as remote removal of the process pot, susceptor and the thermocouples.

Production of Cs-137 Source Pencil

After completion of the installation work, three (03) incell trial runs were carried out to ensure accuracy of the mechanical setup and also to re-establish the control parameters along with the heating cycle. Finally, on 19th May, 2015 the active Cs loaded glass collected in the Cs Process Pot was re-melted in the Cs furnace as per the established temperature profile. After completion of the soaking process, the Cs glass was poured into the pencils (10 Nos.). Multistage pouring was done to fill the accurate quantity of Cs glass (170 g) in each pencil with the assistance of real time load cell data and timer. These freshly filled pencils were allowed to cool and subsequently transferred to the Welding & DC station wherein the lids were welded to these pencils. After passing the leak test, the pencils were encapsulated in the outer pencil and welding of the lids of these outer pencils was carried out. These outer pencils, now called as source pencils, were further leak tested and decontaminated as per the AERB guidelines before transferring and loading in the BRIT Cask.

On 1st June, 2015 Director BARC handed over ten (10) Cs-137 source pencils, having total activity of 2500 Ci, to the CE, BRIT. India has become the first country in the world to deploy Cs-137 vitrified radioactive sources in commercial domain.
Acknowledgement

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