“The function of education is to teach one to think intensively and to think critically. Intelligence plus character—that is the goal of true education.”

– Dr. Martin Luther King, Jr.

Dream, Dream, Dream.

Dreams transform into thoughts and thoughts result in action.
BRIT

Bulletin - 2017

'Scientific Magazine of
Board of Radiation & Isotope Technology, DAE'

GOVERNMENT OF INDIA
CONTENTS

Brief Communications

1. Radioactive ion beams of $^{111}\text{In}$ using ECR plasma sputtering method 1-7
   *Vaishali Naik, Sankha Chattopadhyay et.al.*

2. Thermal analysis of Panaromic batch irradiator (PANBIT) package 8-15
   *Vaishali Yadav et.al.*

Original Research Articles

1. Preparation of ready-to-use therapeutic doses of clinical grade $^{177}\text{Lu}$-DOTA-TATE for PRRT. 16-21
   *Anupam Mathur et.al.*

   *Manoj Sharma, Vaishali Yadav et.al.*

3. New ways of producing Tc-99m Generators: Purification & recycling methodology of Molybdenum. 33-41
   *Sankha Chattopadhyay et.al.*

General / Feature Articles

1. Calibration Laboratory – A Necessity. 42-44
   *S.B. Kamble, B.K. Pathak et.al.*

2. Report on radiotracer study of Fly-ash disposal into mine void in Quarry No. 4 of Jagannath OCP for M/s. Bhushan Steel Ltd. 45-61
   *Vivek N. Yelgaonkar et.al.*

Scientific Events

62
Radioactive ion beams of $^{111}$In using ECR plasma sputtering method

Vaishali Naik,¹ Mahuwa Bhattacharjee,¹ D. Lavanya Kumar,¹ Prasanta Karmakar,¹ S. K. Das,² Debasish Banerjee,² Sankha Chattopadhyay,³ Luna Barua,³ Sujata Saha Das,³ Asit Kumar Pal,³ Arup Bandyopadhyay,¹ and Alok Chakrabarti¹

¹Variable Energy Cyclotron Centre (VECC), Homi Bhabha National Institute (HBNI), Kolkata
²Radiochemistry Laboratory, Bhabha Atomic Research Centre (BARC), Kolkata
³Radiopharmaceuticals Laboratory, Regional Centre, Board of Radiation and Isotope Technology (BRIT), Kolkata

Abstract

Radioactive ion beams of $^{111}$In (Indium-111, half-life 2.8 days) have been produced using the plasma sputtering method in an electron cyclotron resonance (ECR) ion source at the RIB facility of Variable Energy Cyclotron Centre. Indium isotopes were first produced by bombarding a natural silver target with a 32 MeV, 40 μA alpha particle beam from the K-130 cyclotron. After radio-chemical separation, about 25 mCi In-Chloride was deposited on an aluminum electrode and inserted in the plasma chamber of the ECR. Indium ions produced by ion induced sputtering in the plasma were extracted from the ion source, isotopically separated, and a pure $^{111}$In beam was measured at the focal plane of the separator. The measured $^{111}$In beam intensity was $2.67 \times 10^5$ particles/s for beam energy of 5 keV.

Introduction

Surface implantation of radioactive ion beams is a widely recognized technique for online wear diagnostics.¹ Exploitation of the potential of this technique for industrial and medical devices is the subject of ongoing research and development in various laboratories.²-⁴ Nanoscale surface implantation of energetic ion beams of $^{111}$In (Indium-111) into ultra-high molecular weight polyethylene (UHMWPE) polymers, the most common bio-material used in medical prostheses, has been effectively used for studying non-linear tribological processes during wear-in.⁵ It is also widely used in Perturbed Angular Correlation (PAC) spectroscopy—a hyperfine inter-action study that provides information on the structure and dynamics of the local surroundings of
the probe atom with a high degree of sensitivity.  

Depending on the application and the material of interest such as metals, polymers, or ceramics, techniques such as radioactive ion implantation (RII)\(^8,9\) or plasma immersion ion implantation (PIII) of radioisotopes\(^10\) have been investigated. RII has the advantage that one can implant a pure isotope beam at a chosen depth even in a chemically incompatible lattice, whereas PIII offers a flexible and inexpensive albeit less cleaner alternative.

We have set up a dedicated low energy radioisotope beam (RIB) line at the RIB facility of Variable Energy Cyclotron Centre (VECC), Kolkata.\(^11\) A 2.45 GHz electron cyclotron resonance (ECR) ion source that can also be used as a plasma reactor has been installed at the facility. The aim is to have a versatile setup offering the advantage of RII as well as PIII so that a wider R&D programme can be targeted. Metallic ion beams of beta-stable isotopes have been produced using the ECR plasma sputtering technique and routinely used for surface science studies. In this report, results on the development of \(^{111}\text{In}\) radioactive ion beams are presented.

Indium is easily ionized in surface ionization ion sources, and radioactive ion beams with good intensity have been obtained, e.g., at CERN-ISOLDE.\(^12\) Also, molecular \(^{111}\text{In}\text{^{16}O}\) ion beams have been developed using a cesium- sputtering, negative ion source for ion-implantation studies.\(^13\) However, the ECR ion source has the advantage that it can be used as a source for both RII and PIII.\(^14,15\) The ECR ion source utilizes electron cyclotron resonance to create high density plasma in which neutral atoms get ionized by multiple impact ionization by energetic electrons. The ion source can be operated in a very stable regime over long periods because there are no filaments involved.

**Experimental Details**

**A. Considerations for using sputtering technique for radioactive ion beams of Indium**

ECR is a very versatile and robust ion source that is well suited for the ionization of gaseous elements. For non-volatile elements and metals, the oven technique is routinely used with ECR. In this technique, a suitable compound or pure metal of the element of interest is evaporated in a resistively heated oven attached to the plasma chamber of the ion source. The rate of evaporation is controlled by varying the heating power of the oven. For elements such as indium with a low melting point (157°C), controlling the heating power becomes difficult because indium metal quickly becomes liquid and easily flows out of the sample tube. Short circuit between heating filaments and holder is a common problem. For this reason, even for stable isotope beams, e.g., \(^{115}\text{In}\), the oven technique with ECR has met with limited success in previous studies. A more sophisticated method for the control of oven heating power and possible use of indium oxide with suitable support gas was suggested for the stable operation of \(^{119}\text{In}\) beams.\(^16\)

In the oven, typically few grams of feed material is needed. A large fraction of evaporating material is deposited on the oven and plasma chamber walls. For this reason, the oven technique is not suitable for radioactive ion beams since the quantity of radioactive isotope “feed” will be very
small and one cannot afford to lose the material on the oven or chamber walls. The alternative is to use ion-beam induced sputtering in ECR plasma. The method basically involves inserting a solid sample of the element of interest inside the plasma chamber. The sample is negatively biased and placed just outside of the ECR zone. Plasma is created by a supporting gas such as argon. The Argon ions from the plasma get accelerated toward the sample and sputter the solid material into the plasma from its surface. The sputtered atoms get ionized in the ECR plasma.

The sputtering technique has been used earlier for the production of radioactive ion beams such as $^7$Be and $^{18}$F, $^{17}$, $^{18}$ and $^{32}$P. With careful tuning of parameters, very stable RIB with reasonably high intensity could be obtained with quite a low consumption rate of the sputtered sample. Success of this technique has prompted us to try the same method for the production of RIB of indium.

B. ECR ion source and low energy separator

The ECR ion source, M1000 from M/s Pantechnik based on the GANIL design, operates at a resonance frequency of 2.45 GHz and is optimized for the production of ions with a 1+ charge state. The magnetic mirror field is produced using two permanent magnet FeNdB rings. The double walled, water cooled stainless steel plasma chamber is 85 mm in diameter and is fitted with four radial ports that may be used for plasma diagnostics, insertion of sputter targets, etc. The RF power is fed axially via a waveguide fitted with a quartz window for vacuum sealing. A removable aluminum liner of 1 mm wall thickness is inserted in the plasma chamber to protect its walls from contamination. The ion source floats at a potential of up to 30 kV, and the ion beam is extracted at ground potential through a 7 mm aperture plasma electrode. A three-electrode extraction system is used for focusing and transporting the beam in the low energy beam transport line. The low energy separator beamline consists of a 90° analyzing magnet, an electrostatic einzel lens, a beam diagnostics chamber, and a UHV target chamber for low energy experiments. A schematic layout of the facility is shown in Figure 1.

C. Optimization of ECR plasma sputtering for non-radioactive samples

Prior to the production of radioactive ion beams, test experiments aimed at optimizing sputtering parameters were conducted for beta-stable $^{115}$In beams. Sputtering sample of indium oxide (melting point 1910°C) in a form of 25 mm diameter and 2 mm thick pellet was placed on an aluminum sample holder electrically isolated from the plasma chamber.

The weight of the sample was measured at this time and found to be 1.8771 g. The sample was inserted from one of the radial ports, and the position of the
sample was adjusted remotely. Parameters like microwave power, sputtering bias voltage, and argon gas pressure were adjusted to maximize the beam current on the Faraday cup (FC) after the separator. A typical measured spectrum for a 5 keV extracted beam (beam current measured on the Faraday cup by varying the magnetic field of the dipole magnet) is shown in Figure 2.

![Figure 2: Typical mass spectrum at the separator focal plane for an extraction voltage of 5 kV](image)

Measured beam current for $^{115}$In$^{1+}$ was around 100 nA. It may be noted that due to limitations of the present analyzing magnet, the ECR extraction voltage has been kept at 5 kV for these measurements. The sputtering bias voltage was varied, and it was found that at about 900 V, the intensity of the extracted beam was maximum. With 150 W of RF power applied, the discharge current at this bias voltage was measured around 9 mA. The trial run continued for a cumulative duration of 15 beam hours at the end of which the sample was weighed again at 1.8726 g. The consumption rate for this sample was estimated to be 0.3 mg/h.

After getting a reasonably good $^{115}$In beam, the next step was to sputter a 15 mm diameter chemically electro-deposited sample. This is to mimic the method by which the radioactive indium sample would be made. The plasma chamber aluminum liner was replaced, and the chamber was flushed with argon prior to beam tuning. A background spectrum was measured with the sample pulled out to ascertain that no indium ions are measured from the recycled atoms in the ion source. For the chemically deposited sample, a smaller beam current of around 3 nA was measured at the FC for $^{115}$In$^{1+}$, and however steady current was delivered for 8 h.

D. Cyclotron production of $^{111}$In in target and sample preparation for production of RIB

An electroplated natural silver target (200 μm thick) on a hemispherical copper base was bombarded with a 32 MeV, 40 μA alpha particle beam from the K-130 cyclotron at VECC. After 24 h of irradiation, the target was cooled for another 24 h to allow the short lived isotopes such as $^{109}$In (4.2 h) and $^{110}$In (4.9 h) to decay. After this, the target was taken out from the cyclotron Dee (Figure 3); contact dose was measured and remotely dismantled from the target holder system (Figure 4). The irradiated target in a 30 mm thick lead container was transferred...
to a lead shielded fume-hood for chemical processing and was processed for 48 h after the end of bombardment (EOB). $^{111}$In was chemically processed following the modified method used for the separation of $^{111}$In from a Cd target. The target was dissolved in nitric acid; the nitrate residue obtained after evaporation was converted into bromide by repeated evaporations to dryness from a hydrobromic acid (HBr) solution. Finally the residue was taken up in a minimum volume of 8M HBr solution. The resultant solution was cooled and extracted with peroxide free di-isopropyl ether (DIPE). $^{111}$In was back extracted from DIPE with concentrated hydrochloric acid (HCl). The product was finally evaporated to dryness and taken up with desired volume of 0.01M HCl.

The activity of the separated $^{111}$In was determined by using a calibrated dose calibrator (ISOMED 50, Hans Walischmiller GmbH, Dresden, Germany). The acidic solution containing the carrier-free activity of $^{111}$In was then evaporated to dryness, and the activity was brought into neutral medium by adding water. This activity was then dried on a 30 mm diameter aluminum rod used as the sample holder in the ECR chamber. Total deposited activity on the rod at this time was 25 mCi.

**Production of Radioactive Ion Beams of $^{111}$In**

The radioactive sample was brought to the RIB facility site and inserted in the ECR plasma chamber through one of the four radial ports. The ion source parameters were tuned based on the previous offline sputtering experiments. The separator was set for transporting the 1+ beam to the Faraday cup placed after the separator. The characteristic gamma-rays from radioactive decay of the RI beam stopped on the FC were measured using a high purity germanium (HPGe) detector placed nearby. Gamma rays at 171 keV and 245 keV corresponding to the decay of $^{111}$In were clearly identified (Figure 5). The detector spectrum was precalibrated using $^{152}$Eu as the standard source. $^{152}$Eu was also used for calculating the detector efficiency in the geometry of the experimental setup used, and typical efficiencies in this energy range were measured to be a few times $10^3$. A DC bias of 400V was applied to the indium sample, and RF power in the ion source was kept at 115W.
For a trial experiment aimed at optimizing the PAC setup, the $^{111}\text{In}^{1+}$ beam with energy of 5 keV was implanted on a TiO$_2$ single crystal sample placed on the FC for around 13 h with an average activity of 40 cps registered over this time for the 171 keV and 245 keV peaks. The relative gamma intensities for the transition corresponding to 171 keV and 245 keV are 0.9 and 0.94, respectively. From the total number of counts, irradiation time, and knowledge of the gamma intensities along with detector efficiency, the intensity of RIB being implanted on the sample was calculated to be about $2.67 \times 10^5$ particles/s in this experiment resulting in a cumulative dose of around $1.25 \times 10^{10}$ particles implanted on the sample over the period of irradiation.

**Summary and Outlook**

Radioactive ion beams of $^{111}\text{In}$ have been produced using plasma ion sputtering in a 2.45 GHz ECR ion source. Indium ions thus produced were extracted from the ion source, isotopically separated, and a pure $^{111}\text{In}$ beam was measured at the focal plane of the separator. Typical beam intensity obtained was around $2.67 \times 10^5$ pps for beam energy of 5 keV, and the measured cumulative dose was around $10^{10}$ particles on the implantation sample.

Although the intensity achieved is decent enough, given the simplicity of the technique and a low consumption rate, we believe for practical application in RII studies, the intensity still needs an enhancement of one order of magnitude for a better signal to noise ratio. In our present setup, the ECR ion source optimally produces $1^+$ ions, and the extraction voltage has been kept at 5 kV because the existing analyzing magnet is not powerful enough. Also, the strict regulatory requirement regarding handling higher levels of activity has necessitated using lower activity samples from only one radial port for these trial runs. The setup is being upgraded with a new magnet that will enable extraction at higher voltages. Dedicated shielding arrangement is being made that will allow us to insert 2 or 3 sputter samples in the plasma chamber. Arrangement for plasma immersion ion implantation is also being made by modifying the extraction chamber to insert the substrate from the axial extraction port. Implantation studies and PAC measurements will commence once this upgrade has been implemented.

**Acknowledgments**

The authors would like to thank Dr. Tapati Kundu Roy for the preparation of indium oxide samples and Dr. Dirtha Sanyal for help in the detector setup for gamma-ray measurements. The authors thank Shri G. Ganesh, Chief Executive, BRIT, for his support in the work of production of $^{111}\text{In}$. The authors also thank the staff of I & E section, VECC for their help in fixing and dismantling of the silver target and the staff of Regional Center, BRIT, Kolkata for their help in the chemical separation and counting work. The authors also thank the staff of K-130 cyclotron operations group for their effort in alpha-particle beam delivery for the experiments. Finally the authors thank the staff of Heath Physics Unit, BARC, VECC, for their help in monitoring the on-site dose after sample irradiation.
References


Thermal analysis of Panoramic Batch Irradiator (PANBIT) Package

Vaishali Yadav, VVS Prasad, Piyush Srivastava, Vishnu Verma*

Engineering Group, Board of Radiation & Isotope Technology
*Reactor Safety Division, Bhabha Atomic Research Centre

Abstract

For source replenishment the PANBIT (Panoramic Batch Irradiator) FLASK along with decayed source is transported to hot cell. The transportation of the flask requires that it qualifies the TYPE B(U) requirements as stated in national and international regulations. The flask is transported in Shock absorber as TYPE B (U) Package.

One of the accident conditions that the TYPE B(U) Package has to withstand is FIRE Test where in the Package is subjected to a thermal environment characterized by parameters like temperature, time, surface absorptivity, emissivity, solar insolation etc. Also, along with these parameters the code specifies that a post fire analysis is to be conducted to study the effect of residual heat in the package. The response of the Package, when subjected to above requirement, is analyzed using CFD software. For the first time a 3D Model is used with detailed features to study the response. The temperature distribution in the package is evaluated to study the structural and radiation shielding integrity of the Package.

Key words – Type B(U), Fire test, Post fire test, CFD steady state analysis, CFD transient analysis

Introduction to the PANBIT Package

Board of Radiation & Isotope Technology has designed a PANBIT for batch type gamma irradiation. This is Category–II Irradiator wherein 100 kCi CO-60 source is stored in PANBIT Flask. During irradiation source cage is raised from Source cavity of storage FLASK to irradiation position for gamma exposure. After the irradiation it is lowered in the flask for storage. This flask is located in a concrete pit. The specification of the flask that is used in PANBIT Facility is shown in the Table 1 given below. Overview of the batch type Irradiator and Sectional view of PANBIT Facility is shown in Figure 1.
Table 1: The specification of flask used in PANBIT Facility is as follows

<table>
<thead>
<tr>
<th>Specification</th>
<th>Details</th>
</tr>
</thead>
<tbody>
<tr>
<td>Max. Co-60 source capacity</td>
<td>100 kCi (3700 TBq)</td>
</tr>
<tr>
<td>Irradiation floor area</td>
<td>95 Sq. mts</td>
</tr>
<tr>
<td>Minimum built up area of facility</td>
<td>160 sq. mts</td>
</tr>
<tr>
<td>Cylindrical Source cage</td>
<td>O.D. 144 mm X I.D. 110 mm, ht. 446 mm</td>
</tr>
<tr>
<td>Product box conveyor</td>
<td>Shuffle dwell conveyor (Batch type)</td>
</tr>
<tr>
<td>Product box size</td>
<td>390 mm x 390 mm x 400 mm (ht)</td>
</tr>
<tr>
<td>Dose uniformity</td>
<td>1.4 at 0.1 gm/cc of product density</td>
</tr>
<tr>
<td>Throughput</td>
<td>425 Lts / batch every 6 hrs at 25 kGy dose at</td>
</tr>
<tr>
<td>Processing rate</td>
<td>425 Lts per batch</td>
</tr>
<tr>
<td>Operation</td>
<td>Remote operation through electrical control (Manual &amp; automatic)</td>
</tr>
<tr>
<td>Ventilation</td>
<td>20 Air changes / hr</td>
</tr>
<tr>
<td>Safety</td>
<td>Provided with interlock and fail safe mechanism</td>
</tr>
<tr>
<td>Power supply</td>
<td>440 V, 3 phase, 50 cycles, AC – 4 wire system</td>
</tr>
</tbody>
</table>
Brief Communication

Overview of Batch Type Irradiator

Sectional view of PANBIT Facility

Figure 1: Overview and Sectional view of Package

Figure 2: General Features of the Flask
The general features of the Flask are shown in Figure 2. The Flask is fabricated from SS 304L material for better structural integrity. The characteristic of material enables it to withstand all services, transportation and storage conditions. The outer shell is provided with dished ends to avoid corner joints. The fins provided are angled so as to enhance the shock absorbing capacity of the flask as well as to dissipate heat. The outermost dimension of the Flask is 920 mm and length is 1082 mm. The outer shell of the Flask comprises of three parts Top Dish (12mm thick), Cylindrical part (10mm thick) and Bottom dish (12mm thick). The Outer shell is welded to the square Base plate at bottom. The flask has 36 nos longitudinal fins of size 100 mm (w) x 1057 mm (l) x 5mm thick. The fins help dissipate the decay heat to ensure that the temperature of outer surface of the flask remains within prescribed limits. The inner shell forms cylindrical cavity to house the Source cage. The source cage, (Dwg. BRIT/ENGG/PAN/SC/04) in Figure 3, comprises of 12 nos. source holder pipes at 126mm P.C.D and four lifting bars at the top to facilitate lifting of the source cage during the exposure of source. The flask is provided with lead plug weighting 170 kgs to enable loading and unloading of the Source in the flask. During transportation lead plug is secured in position with the help of 12 nos. of M 20 hex bolts. Flask has two lifting lugs. The Flask will be lifted with the help of Dia 25 mm, 6 X 19 Construction, two legs mechanically split wire rope sling which is attached to the flask with the Dee shackle. The flask has a finned transportation cap screwed to the outer shell by M20 hex bolts, 12 nos. The cap protects the screwed end of the lifting bars. The weight of flask is 5000 Kg.

For source replenishment the flask along with decayed source is transported to hot cell. The transportation of the flask requires that it qualifies the TYPE B(U) requirements as stated in national and international regulations. This report covers response of PANBIT PACKAGE when subjected to routine, normal & accident condition as stipulated in IAEA/AERB regulations for the safe transport of the Type B(U) Package. The primary shielding material used in the flask is lead. The shipping flask with approximate weight of 6.650 Tons will accommodate a maximum of 100 kCi of C0-60. Features of PANBIT Package are shown in Table 2.
A 3D model (45° sector) is generated for the simulation. The Flask along with the shock absorbers is modeled for analysis as per Figure 4A and 4B.

Figure 4A: 3D Model of PANBIT Package
Simulation of Fire Test & Post Fire Test

Transient (fire test) analysis has been carried out using the temperature distribution of steady state analysis as the initial conditions. The fire test analysis consists of exposure of the Package to 800°C temperature on all sides. The flame emissivity is taken as 0.9 and surface absorptivity is taken as 0.8 for radiative boundary condition. The convective heat transfer coefficient is taken as per regulatory guidelines. Basically the lead is contained in the belly (space enclosed by inner shell and outer shell) and the lead plug. At the end of fire test there is no melting of lead. The maximum temperature encountered on the outer surface of the transportation package is 800°C. (Refer Figure 5A). The maximum temperature encountered by the outer shell and inner shell at the end of fire test is 199°C and 190°C. The Maximum temperature of lead at the end of fire test is 234°C.

For Post Fire test the analysis was continued after the fire test until the temperatures started decreasing in all the components. This occurred after 2 hrs. 30 minutes from the start of post fire analysis. The temperatures observed at the end of post fire analysis are shown in the Figure 5A and 5B whereas transient time-temperature variations for a range of components of the PANBIT Package is shown in Figure 5C. No lead melting is observed in lead components during the fire and post fire scenario.
The end of fire test showed the maximum temperature encountered in the outer surface of Package as 798°C. The maximum temperature encountered in the outer shell and inner shell at the end of fire test is 199°C and 190°C respectively. At the end of post fire analysis the maximum temperature reached by lead in belly is 234°C. The maximum temperature approached by lead in lead plug is 230°C. The maximum temperatures attained by lead in belly and lead plug during the fire and post fire analysis are well within the lead melting temperature. There is no lead melting occurring. A detailed analysis of the differential expansion of steel and lead components was performed to evaluate the internal pressure generated in the flask. As the gradation in temperature of components and overall temperature of the components is low the rise in internal pressure can be sustained by inner shell so as to enable easy retrieval of the source from the source cage.

Conclusion

Figure 5A: Temperature contours at end of Fire

Figure 5B: Temperature contours (Post Fire)

Figure 5C: Temperature transients on the components of the Package
References


Preparation of ready-to-use Therapeutic doses of Clinical Grade $^{177}$Lu-DOTA-TATE for PRRT

Anupam Mathur,1 P. C. Vrinda,1 Navin Sakhare,1 Yuvraj Nitin,2 R. Krishna Mohan,1 Chanda Arjun,1 Barkha Karkhanis1, N.C. Joseph,1 B.K. Tiwary,1 Ravi Seshan,1 S. S. Sachdev1

1Radiopharmaceuticals Program, Board of Radiation and Isotope Technology, 2Radiation Medicine Centre, Parel, Mumbai-400012, India.

Abstract

$^{177}$Lu-DOTA-TATE is a clinically useful and promising therapeutic radiopharmaceutical for peptide receptor radionuclide therapy (PRRT) of neuroendocrine tumors (NETs) over-expressing somatostatin receptors. Currently, the radiopharmaceutical is prepared in-house at Nuclear medicine centers thereby restricting its use to limited centers only. In the present article we describe systematic studies toward bulk scale formulation of ‘ready-to-use’ $^{177}$Lu-DOTA-TATE using medium specific activity $^{177}$Lu (740-1110 GBq/mg) at a centralized radiopharmacy facility.

Introduction

$^{177}$Lu labeled [DOTA$^{0}$,Tyr$^{3}$] Octreotate (DOTA-TATE) is a clinically useful therapeutic radiopharmaceutical for the treatment of neuroendocrine tumors (NETs) over-expressing somatostatin receptors (sst$R$) for more than a decade.1,2 In this preview, a large number of clinical studies using $^{177}$Lu-DOTA-TATE formulation prepared with carrier added $^{177}$Lu of specific activities > 740 GBq/mg have been reported demonstrating the therapeutic utility of the product.1-4 In most of the cases, the synthesis and quality control of the radiopharmaceutical are carried out at the hospital radiopharmacy and used for captive consumption. However, a number of nuclear medicine centers lack the suitable infrastructure for formulation of the finished radiopharmaceutical in bulk scale as per the cGMP regulations at hospital premises. To circumvent the above limitations, a ready-to-use formulation of $^{177}$Lu-DOTA-TATE radiopharmaceutical has been proposed which is produced as per the existing cGMP regulations and also amenable for transport and use in patient for next two days from the date of production without compromising the clinical efficacy of the product.
The present work includes optimization of labeling reaction with different specific activities of $^{177}$Lu radiometal by varying peptide/metal ratios. A purification protocol using C18 cartridge was standardized in addition to shelf-life determination studies at different storage conditions. This detailed systematic study was undertaken in view of regular production, supply and safe transport of $^{177}$Lu-DOTA-TATE to different Nuclear Medicine Centers throughout India.

**Experimental**

**General**

The peptide $[^0$DOTA$^3$Tyr$^3$] Octreotate (DOTA-TATE) was procured from M/s Pi-CHEM, Austria. Lutetium-177 was produced in Dhruva research reactor and supplied by Radiochemical Section, RPhD, Bhabha Atomic Research Center with specific activity varying in the range 555-925 GBq/mg and is of clinical grade. The HPLC of the radiolabeled preparation was carried out on a JASCO PU 2080 Plus dual pump HPLC system, Japan, with a JASCO 2075 Plus tunable absorption detector and Gina Star radiometric detector system, using a C18 reversed phase HiQ Sil (5 μm, 4 ‘ 250 mm) column. Sep-pak® Vac C18 3 cc (500 mg) cartridges was obtained from M/s Waters, USA. Sterile 0.22 μm membrane syringe filter was procured from M/s Millipore, India.

**Synthesis**

The bulk doses were formulated by proportionately varying the amount and volume of the constituents according to the total dose requirements. A single patient dose of $^{177}$Lu-DOTA-TATE was synthesized by following procedure. $^{177}$Lutetium activity (0.2 mL $^{177}$LuCl$_3$ solution in 0.01 M HCl; 9.25 GBq $^{177}$Lu) was added to a sterile glass vial containing mixture of DOTA-TATE solution (0.25 mL of 1 mg/mL solution in HPLC grade water; 250 μg, 2.5 equivalent of Lu) and gentisic acid (33 mg, 0.1 mmol) dissolved in ammonium acetate buffer (0.2 M; 2.2 mL). The vial was crimped and heated at 95°C using a metal heating block for 1 h. Subsequently, the reaction vial was allowed to cool and attain ambient temperature. The contents of the reaction mixture were loaded onto pre-conditioned C18 Sep-pak® cartridge and washed with 6-8 mL saline to remove unreacted free $^{177}$Lu. The column was further washed with 0.6 mL ethanol to remove the solvent in void volume. Finally the labeled product was eluted out in absolute ethanol (1mL) and the activity of the labeled product ($^{177}$Lu-DOTA-TATE) was assayed using a Dose calibrator. The assayed product was further diluted using ammonium acetate buffer (0.2 M) containing gentisic acid (1.5% w/v) (pH 4-5) to final radioactive concentration of 740 MBq/mL (Ethanol content < 10%). The diluted formulation is then filtered through a 0.22 μm syringe filtration assembly into a 100 mL sterile glass vial. Finally the sterile formulation $^{177}$Lu-DOTA-TATE is dispensed aseptically into sterile glass vial of 15 mL capacity and crimped with sterile combiseal.

The synthesis protocol described above was finalized after extensive radiochemical optimization studies.

**Quality Control Studies**

*High Performance Liquid Chromatography*

HPLC analysis of the radiolabeled formulation was carried out on C18 reversed phase column using gradient elution with
water (A) and acetonitrile (B) as mobile phase. About 20 μL (~1.85 MBq) of the test solution is injected into the column and the following gradient elution program was followed to effect separation [0-4 min 95% A, 4-15 min 95% to 5% A, 15-20 min 5% A, 20-25 min 5% to 95% A and 25-30 min 95% A] of free \(^{177}\text{Lu}\) from labeled \(^{177}\text{Lu}-\text{DOTA-TATE}\) preparation.

**Sterility Test**

The \(^{177}\text{Lu}-\text{DOTA-TATE}\) samples were tested for sterility using sterile, disposable needles and syringes, in a laminar flow bench equipped with adequate precautions for safe handling of radioactivity. The product was aseptically transferred into sterile Fluid Thioglycollate Medium (for detection of aerobes and anaerobes) and sterile Soyabean Casein Digest Medium (for detection of fungi). The inoculated media was incubated at 30-35°C and 20-25°C respectively for a period of 14 days. At intervals during the incubation period and at its conclusion the media was examined for macroscopic evidence of microbial growth by visual examination.

The appearance of the medium was compared with Negative Control Test containers during this entire period. Negative Control Test containers were prepared by aseptic inoculation of sterile Fluid Thioglycollate Medium and Soyabean Casein Digest Medium with sterile saline in the same work area. If no evidence of microbial growth was found, the samples were deemed to be complying with the test for sterility.

**Bacterial Endotoxin Test (BET)**

BET was carried out using a standard commercially available kit (Charles River, India). Sterile, depyrogenated glassware and material was used to conduct the test. The \(^{177}\text{Lu}-\text{DOTA-TATE}\) sample was diluted and incubated with LAL (Limulus Ameobocyte Lysate) reagent at 37±1°C for 60±2 minutes. The lysate sensitivity used was 0.125 I (I denotes the minimum level of endotoxin that can be detected by the lysate). The endotoxin limit for the samples was defined at 25 EU/ml. [1 ng endotoxin (E. coli - EC) = 10 Endotoxin Units (EU) of USP Reference Standard EC-6]. At the end of the incubation period the tube was inverted by 180° to detect gel-clot formation. This was compared to negative and positive Water Control Tests and Product Control Tests. The former contained endotoxin-free water and the latter contained the \(^{177}\text{Lu}-\text{DOTA-TATE}\) sample in the absence and presence of standard endotoxin respectively.

**Biodistribution Studies**

All procedures performed herein were in accordance with the national laws pertaining to the conduct of animal experiments. Normal Wistar rats (200–250 g body weight) were used for the *in vivo* distribution studies. Four sets of Wistar rats (each set containing 4 animals) was intravenously administered with about 0.1 mL of the \(^{177}\text{Lu}-\text{DOTA-TATE}\) (~100 μCi/ 3.7 MBq). The animals were sacrificed at different time points, viz. 30 min, 60 min, 3 h and 24 h post-injection, after which the relevant organs and tissue were excised for measurement of associated activity. Radioactivity measurements were carried-out in a flat-bed type NaI(Tl) scintillation counter with optimal energy window for \(^{177}\text{Lu}\). The accumulated activity was expressed
in terms of % ID/organ. Blood, bone and muscle were taken as 7%, 10% and 40% of total body weight.

Results and Discussion

Easy availability of ready-to-use injectable radiopharmaceutical preparation at nuclear medicine clinic is one of the key factors in the wide-spread applicability of therapeutic radiopharmaceuticals. The present work successfully demonstrates the bulk scale formulation of ready-to-use $^{177}$Lu-DOTA-TATE at a centralized radiopharmacy and its subsequent distribution and clinical utilization at various nuclear medicine clinics.

Therapeutic doses of $^{177}$Lu-DOTA-TATE were directly synthesized upon addition of $^{177}$Lu in 0.01 M HCl to a solution of DOTA-TATE in ammonium acetate buffer containing gentisic acid. The pH of the reaction mixture is a key factor determining the radiolabeling yield. Therefore, the molarity of the buffer and gentisic acid content should be adjusted such that the final pH of reaction mixture should be 4.0-4.5 on addition of acidic $^{177}$Lu activity. The total volume of the reaction mixture also has an effect on the overall yield of the radiolabeled product. The radiochemical yield of the synthesized labeled complex using the protocol standardized was 98.4 ± 0.4% ($n = 6$). Uncomplexed $^{177}$Lu was efficiently removed by C18 Sep-pak purification. The recovery of the purified product from the column in ethanol was ~95% and its radiochemical purity was >99.5%. In order to minimize the purification losses, the purification step could be avoided in cases where the radiolabeling yield is >97%. Hence, each time QC before purification step is a mandatory step and is a part of methodology developed.

The yield and radiochemical purities were ascertained by PC and HPLC. In PC, $^{177}$Lu-DOTA-TATE was found to move towards the solvent front ($R_f = 0.8$ to 1.0) whereas unreacted $^{177}$Lu remains near the point of spotting ($R_f = 0.0$-0.1). HPLC pattern of the purified $^{177}$Lu-DOTA-TATE formulation showed the labeled complex eluted out as a sharp peak at 16.3 ± 0.3 min and free $^{177}$Lu eluted out early between 3-4 min. The radiochemical yields ascertained by these two methods were near identical in all the experiments.

The specific activity of $^{177}$Lu used in all the formulations for this study was ~ 740 MBq/ïg. It is evident from the results that 9.25 GBq $^{177}$Lu could be labeled using 200 mg of DOTA-TATE with high labeling yield of ~97%. High labeling yield is indeed advantageous in maintaining the specific activity of the labeled formulation around 37 MBq/ïg, even after 2 d from the date of preparation. The radiochemical purity was consistently >98% when the peptide/metal ratio was 2.5. Although [DOTA-TATE] / [Lu] ratio of 2.0 is optimal for high yield of the labeled product, use of higher peptide/metal ratio of ~2.5 would completely avoid need for Sep-pak® purification thereby reducing purification losses and production time. In addition, the final product formulation would be in aqueous buffer, thereby completely avoiding the use of ethanol. The specific activity of the labeled product would be ~29.6 MBq/μg of peptide in this case.
Since the ready-to-use formulation of $^{177}$Lu-DOTA-TATE is intended to be used for therapy even 2 d after its preparation, *in vitro* stability of the formulation is a very important parameter. Therapeutic radiopharmaceutical preparations formulated in aqueous medium are susceptible to radiolytic degradation. The gentisic acid present in higher concentrations in the final radiopharmaceutical preparation (4.0 %w/v) was found to precipitate out under refrigerated conditions and does not re-dissolve on attainment of ambient room temperature conditions. Hence, lower content of gentisic acid (1.5% w/v) was found appropriate as no solubility issues were observed at this concentration even when stored at -70°C.

The radiochemical purity was found to degrade below 95% after 3 d when stored at temperatures above 4°C. However, the high radiochemical purity of >98% was maintained up to a period of 9 d for a preparation stored at -70°C. At radioactive concentrations $^{3}925$ MBq/mL, the radiolabeled formulation undergoes radiolytic degradation and the radiochemical purity falls below 95% post 3 d preparation. Hence, a concentration of 740 MBq/mL was found to be optimal for the prolonged shelf-life of product and considered suitable for its deployment. In routine clinical practice, $^{177}$Lu-DOTA-TATE is generally infused into the patients through saline over a period of time, hence there are no stringent norms for radioactive concentration supplied to hospital radiopharmacy.

**Table 1** shows biodistribution data of $^{177}$Lu-DOTA-TATE therapeutic dose preparation in normal Wistar rats. The labeled preparation was found to clear rapidly from the normal circulation with more than 50% of the activity getting excreted over a period of 1 d. A small percentage of the activity was found associated with the kidneys highlighting the affinity of the labeled formulation towards kidney. The activity found associated with the bones was negligible indicating inertness of the complex *in vivo*.

**Conclusion**

A protocol for bulk scale formulation of ready-to-use $^{177}$Lu-DOTA-TATE for therapy of neuroendocrine tumors was developed using moderate specific activity $^{177}$Lu. The specific activity of the radiolabeled peptide was $^{3}29.6$ MBq/μg of the peptide conjugate on the date of its formulation. The product retained its radiochemical purity to the extent of >98% when stored at -70°C with concentration of 740 MBq/mL, ensuring that there would be no constraint in its deployment from central radiopharmacy to distant nuclear medicine centres.
Table 1: Biodistribution data of the therapeutic dose formulation of $^{177}$Lu-DOTA-TATE in normal Wistar rats

<table>
<thead>
<tr>
<th>Organs</th>
<th>% ID/organ (s.d) (n = 3)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>30 min</td>
</tr>
<tr>
<td>Liver</td>
<td>3.95 (0.14)</td>
</tr>
<tr>
<td>Intestines</td>
<td>5.79 (0.14)</td>
</tr>
<tr>
<td>Stomach</td>
<td>0.95 (0.04)</td>
</tr>
<tr>
<td>Kidney</td>
<td>5.09 (0.47)</td>
</tr>
<tr>
<td>Heart</td>
<td>0.18 (0.03)</td>
</tr>
<tr>
<td>Lung</td>
<td>0.59 (0.07)</td>
</tr>
<tr>
<td>Spleen</td>
<td>0.13 (0.01)</td>
</tr>
<tr>
<td>Blood</td>
<td>8.36 (0.58)</td>
</tr>
<tr>
<td>Bone</td>
<td>0.11 (0.02)</td>
</tr>
<tr>
<td>Muscle</td>
<td>10.35 (1.34)</td>
</tr>
<tr>
<td>Urine + Stools</td>
<td>21.12 (9.10)</td>
</tr>
</tbody>
</table>

References


Studies on the Development of Blood Irradiator (BI-1000 EM)

Manoj Sharma, Vaishali Yadav, Kuldeep Sharma, Vinay Bhave, K. Bijju*, Piyush Srivastava,

Engineering Division, Board of Radiation & Isotope Technology
*Radiological Physics and Advisory Division (RP&AD), BARC

Abstract

Blood and blood products are irradiated to avoid the graft versus host diseases (GVHD) by decreasing the number of viable T Lymphocytes. This article describes a prototype model BI_1000 EM which is a Category–I BLOOD IRRADIATOR specially designed for irradiating blood products. This device contains CO-60 radioactive material and hence when it is transported in public domain it has to be safely transported. The Transportation of such device involves proper packaging so that it qualifies all the regulatory national and international requirements stipulated in relevant AERB and IAEA Code. The Codes and regulatory guidelines are being designed with the concept that safety is vested principally in the package which has to provide shielding to protect workers, the public and the environment against the effects of radiation, to prevent an unwanted chain reaction, to prevent damage caused by heat and also to provide protection against dispersion of the contents. The article includes two sections. First DEVICE section deals with the configuration and functionality of the blood irradiator and second transportation section, which deals with the design of Type B (U) PACKAGE that is used during transportation of the irradiator.

I. Device (Blood Irradiator_1000 EM)

Blood Irradiator-1000 EM is compact Economical Model of self shielded CO-60 Gamma irradiator which is ideally suited for Blood Irradiation. The design meets all the requirements of AERB Safety guideline No. AERB/RF-RPF/SG-2 and American National Standard Institute (ANSI N 433.3-1977). A prototype is fabricated to house 300 Ci of Co-60 source in the form of pencils. The Device comprises of Flask (Biological shield for safe exposure of product to Gamma radiation) and the Drive System. A sectional view of General assembly of the unit is shown in Figure 1.
A. Biological Shield:

The prototype units shielding integrity was checked by radiometry. A report of the radiometry is attached. A point kernel based evaluation of radiation shielding and dose distribution of the device was carried out by RP&AD, BARC**. (Figure 2) First time the biological shielding used was a combination of lead and tungsten enclosed in SS 304L Shells. This enabled designing a compact unit. The sample chamber is sandwiched between top and bottom lead shielded drawer is conveyed to irradiation position by drive system. The product is placed in rotating chamber and is conveyed to irradiation position by drive system which safely conveys the sample chamber in the irradiation volume inside the biological shield. The rotating chamber is provided with a motor on the top to enable rotation of sample to provide better dose uniformity. Dosimetry will be carried out by dosimetry group to ascertain the theoretical dose delivery capacity of the unit.
B. Drive System:

Pneumatic drive system will be used as an alternate arrangement for raising and lowering of the drawer to expose the irradiation volume, i.e. sample chamber in irradiation position. This will enable us to transport the Flask with only bottom drawer. Air consumption of Pneumatic Cylinder [DGPL-80-750-PPV-A-KF-B] will be 54 litres per cycle at 6 Bar. Air consumption of complete pneumatic system will be approx. 65 litres/cycle. Through past experiences it was found that the breakdown of the Gamma chamber unit is due to two reasons, first is the jamming of wire rope and second is improper winding or unwinding of the wire rope on the grooved wire rope drum. Instead of cumbersome wire rope mechanism which requires frequent maintenance such as checking the tightness of wire rope, lubrication of the four pulleys used to bring about the movement of the drawer, etc., it was proposed to replace the wire rope mechanism by the pneumatic system. Prime advantage of incorporating this mechanism is that the drawer can be brought back to the safest position that is the lowest position. This is achieved by choosing the cylinder which will automatically lower the drawer in safe position in case of emergency, i.e. power failure. Presently used Gamma Chamber unit has a cranking mechanism to bring back the drawer in lowest position in case of power failure. Also travel time can be reduced by using high speed pneumatic cylinder. This will reduce transit dose which will lead to more accuracy in doses given to the sample for low dose irradiation. This system has minimum settings to be done while installing the system at site, once the setting for accurate positioning of the sample chamber with respect to source cage is done.

II. Transportation of BL_1000EM Package

While transportation, the flask with bottom shielded drawer will be packaged in shock absorber. The transportation of the flask when loaded with radioactivity will be packaged and categorized as TYPE B (U) Package. A sectional view of the BL_1000 EM flask along with shock absorber (SA) is shown in Figure 3 The geometry and sizing of the components of SA are arrived after several design iteration considering the response of the flask in all critical drop simulation. The Shock absorber is designed to withstand the accident scenarios stipulated in the mandatory regulatory national and international codes for safe transportation of TYPE B (U) flask. The safe transport of radioactive material mentioned in Safety Code No. AERB/NRF/TS/SC-1, Rev. 1, of Atomic Energy Regulatory Board, India (Edition 2016) and international standard code mentioned in IAEA Safety Standards, ‘Regulations for the Safe Transport of Radioactive Material (Edition 2012), Specific Safety Requirements No. SSR-6, are the two safety codes which were referred. The two test scenarios stipulated represent the Package in following accident events -

a. IMPACT TEST
b. THERMAL TEST

Figure 3: BL_1000 EM Type B (U) Package (Flask with shock absorber)
Details of the BI_1000 EM Package:

<table>
<thead>
<tr>
<th>Overall size of Package</th>
<th>1042 mm (O.D.) x 1220 mm(H)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Package weight with source pencils</td>
<td>2440 kgs approx.</td>
</tr>
<tr>
<td>Overall size of Flask</td>
<td>570 mm OD x 870 mm Ht.</td>
</tr>
<tr>
<td>Flask weight with Source</td>
<td>1700 kgs approx.</td>
</tr>
<tr>
<td>Flask material of construction</td>
<td>S.S. 304L</td>
</tr>
<tr>
<td>Biological Shielding</td>
<td>Lead and Tungsten</td>
</tr>
<tr>
<td>Shock Absorber (S A ) weight</td>
<td>700 kgs approx.</td>
</tr>
<tr>
<td>S A  material of construction</td>
<td>S.S. 304L</td>
</tr>
<tr>
<td>Source Holder Size</td>
<td>O.D. 11.5 mm X 209 mm lg.</td>
</tr>
<tr>
<td>Maximum No. of Sealed sources.</td>
<td>16 Nos.</td>
</tr>
<tr>
<td>Maximum strength of Co-60</td>
<td>300 Ci</td>
</tr>
<tr>
<td>Dose variation in radial &amp; axial direction</td>
<td>+ / - 12 %</td>
</tr>
<tr>
<td>Over dose ratio (Dmax/Dmin)</td>
<td>1.5</td>
</tr>
</tbody>
</table>

a. **Impact Test**

The regulatory AERB code, AERB / NRF/TS/SC-1 for Safe transportation of radioactive material, requires that the package withstands the following relevant Impact test, maintaining its structural and shielding integrity.

1. DROP I: 9m Drop of Package on unyielding target.
2. DROP II: 1m Drop of Package on Punch

The structural drop test I & II is a dynamic phenomenon featuring complex interaction between structural and internal forces. Drop test impact simulation using FE focuses on the evaluation of nonlinear response of the structural components subjected to impact. In the event of impact, structures are subjected to loadings of high intensity, which induces transient deformations ranging from small deflections to large strains resulting in geometrically nonlinear behaviour. This complex behaviour of structure is further complicated by a nonlinear material behaviour resulting from plastic flow of materials. To simulate these large deformations and inelastic material response, nonlinear FE treatment is required.
Flask components are discretized with 8-noded solid brick elements. The pipe structure and steel plates used for fabrication of the shock absorber are discretized as shell elements. One-dimensional beam elements are used to model the bolts. The nodes of the bolts are merged with coincident nodes of corresponding parts. All other structures are modelled as shell elements or brick elements depending on thickness. Also nodes are merged at welded joints.

Mesh convergence study was performed considering two average mesh sizes 8 mm and 4 mm. The mesh convergence study is based on reducing the elements without changing the order. The process requires user intervention as the simulation is to be performed refining the mesh to estimate the convergence errors. The stress intensity induced in various lead containment boundaries of the packaging for different model during worst orientation case, i.e. inverted drop case was checked. Based on this further simulation were carried out.

1. DROP I: Nine Meter Drop Impact Test of Package on Unyielding Target

The Package has been analysed using PAM-Crash software for four different critical orientations.

Drop orientations which are considered to be critical considering the Package geometry are as follows:-

1. Upright vertical drop (Base of package impacting first the unyielding target)
2. Inverted vertical drop (Top of the Package impacting first the unyielding target)
3. Horizontal Drop
4. Corner (Inclined) Drop

The simulations were performed ensuring the conservation of both, energy and momentum. The SOP detailed by RTD, BARC has been followed in the entire simulation process. The results are evaluated for shielding integrity of the Package by considering the deformed configuration of the package after drop test in all the four orientation. The lead slumping and the maximum induced tresca stresses in critical components during the drop test simulation are shown in the figures below. The stresses induced and the safety margins available after drop are considered to evaluate the structural integrity of the package. The end results of the simulation i.e. deformed configuration is depicted in following Figures 5A, 6A, 7A & 8A and the maximum stress intensity in critical components are depicted in Figures 5B, 6B, 7B & 8B respectively and also tabulated in Table1 where the values of containment bolt loading are mentioned.
Figure 5A: 9m Drop in Upright Orientation

Figure 5B: Stress Contour in the Flask

Figure 6A: 9m Drop In Inverted Orientation

Figure 6B: Stress Contour in the Flask

Figure 7A: 9m Horizontal Drop on Lugfig

Figure 7B: Stress Contour in the Flask
Table 1: Summary of the Stress Qualification of Critical Component in 9M Drop Orientation.

<table>
<thead>
<tr>
<th>Sr. No.</th>
<th>Critical Drop Orientation</th>
<th>Max Allowed</th>
<th>Observed Max S.I (MPa) in various drop Orientation</th>
<th>Safety Margins</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>UPRIGHT</td>
<td>436.5</td>
<td>382</td>
<td>12 %</td>
</tr>
<tr>
<td>2.</td>
<td>INVERTED</td>
<td>436.5</td>
<td>375</td>
<td>16%</td>
</tr>
<tr>
<td>3.</td>
<td>HORIZONTAL</td>
<td>436.5</td>
<td>398</td>
<td>8%</td>
</tr>
<tr>
<td>4.</td>
<td>CORNER</td>
<td>436.5</td>
<td>387</td>
<td>11%</td>
</tr>
</tbody>
</table>

B. LEAD PLUG BOLTS (M12) \[ \frac{F_{tb}^+}{F_{vb}} + \frac{F_{tb}^-}{F_{vb}} \leq 1 \]

<table>
<thead>
<tr>
<th>Sr. No.</th>
<th>WORST DROP (INV.)</th>
<th>Max Allowed</th>
<th>Observed Max S.I (MPa)</th>
<th>Safety Margins</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>WORST DROP (INV.)</td>
<td>1</td>
<td>0.014</td>
<td>98%</td>
</tr>
</tbody>
</table>

2. DROP II: One Meter Drop Test of Package on Unyielding Target

The drop II test shows the results where the Package was subjected to fall with a velocity equivalent to 1m free fall on a punch of Diameter 150 mm and length 200 mm in various orientations. Out of which the case of worst orientation is mentioned below.
Table 2: Summary of the Stress Qualification of Critical Component in 1M Drop in worst orientation

<table>
<thead>
<tr>
<th>Orientation</th>
<th>Max Allowed (MPa.) in various drop Orientation</th>
<th>Observed Max S.I.</th>
<th>Safety Margins</th>
</tr>
</thead>
<tbody>
<tr>
<td>436.5</td>
<td>394</td>
<td></td>
<td>8%</td>
</tr>
</tbody>
</table>

**B. LEAD PLUG BOLTS (M12)**

\[
\frac{ft^2}{Ftb} + \frac{fy^2}{Fyb} \leq 1
\]

<table>
<thead>
<tr>
<th></th>
<th>0.001</th>
<th>&gt; 99%</th>
</tr>
</thead>
</table>

Discussion and Results:

1. The results of the Simulation performed on finite element model for checking the structural integrity of the package under the accident mechanical conditions (DROP I & II) are presented in this report. Four different drop orientations were considered as critical and simulations were performed on the Package.

2. The maximum stress Intensity and combined normalized stress induced in the critical components were presented in Table 1 for each drop orientation & Table 2 for worst drop orientation.

3. Considerable safety margins are observed in the critical components of the flask. This suggests that design of the package is qualified to withstand the impact.
4. The lead slumping observed at the end of the Impact simulations are shown in the figures for the respective drop orientation. The maximum lead slumping does not exceed 3 mm.

5. The shock absorber is designed with two arrays of pipe structure concept to enhance the thermal performance of the shock absorber. It is seen that a minimum air gap of more than 100 mm around the flask and outer cover of shock absorber is maintained even after the impact in worst orientation. This air gap will act as thermal insulation enhancing the flasks thermal performance. Hence, the designed shock absorber, apart from limiting the accident impact to the flask, it also enhances the thermal performance of Flask.

b. Thermal Test

Due to geometric complexity of transport package, a simplified 2Dax_symmetry model is generated, for the simulation. The BL_1000 EM FLASK along with the shock absorber is modelled for analysis (Figure 10). The flask with inner and outer cover of shock absorber is modelled for simplicity. The analysis is carried out as per AERB/IAEA Safety standards [1, 2] for normal (steady state) as well as accident (fire test) condition including post fire. Commercial CFD software, CFD ACE+, is used to simulate the fire and post fire accident event. Temperature dependent thermal conductivity has been considered for all the components. As per regulatory guidelines, solar flux of 800 W/m² is considered on the top surface and solar flux of 400 W/m² is considered on the side surface with solar absorptivity of 0.3. The convective and radiative boundary conditions were applied on the outer surface while taking ambient temperature as 38°C.

A surface emissivity of 0.3 is assumed for steady state analysis. Since the heat flux due to the heat generated by radiation source of strength 300 Ci is less than 4.8 watts, it is negligible and hence not considered in the analysis. The fire test analysis consists of exposure of the Package to 800°C temperature on all sides. The flame emissivity is taken as 0.9 and surface absorptivity is taken as 0.8 for radiative boundary condition. The convective heat transfer coefficient is taken as per regulatory guidelines. The post fire analysis involves simulation of package subjecting it to ambient condition with temperature profile at the end of 30 min fire test as its initial conditions. The simulation is continued until the temperature in the critical components starts dropping.

Following simulations were performed to analyse the response of the package in the normal and accident conditions of transport. The results of simulation are plotted in terms of temperature profiles in the package components:

A. Temperature contour during Normal condition of transport. (Figure 11)

B. Temperature contour at the end of Fire Test analysis. (Figure 12)

C. Temperature contour at the end of Post fire Analysis. (Figure 13)
Discussion and Results

1. The temperature on the surface of the package at the end of normal condition of transport without solar insolation is 48°C and with solar insolation it is 75°C.

2. At the end of fire test the maximum temperature encountered in the outer surface of package is 791°C.

3. The maximum temperature encountered in the outer shell and inner shell is 220°C and 205°C respectively.

4. At the end of fire test and post fire analysis the maximum temperature reached by lead in belly is 218°C.

5. As the melting point of lead is 327°C, and the maximum temperature of lead reached during the fire and post fire analysis are less than 220°C, no lead melting is observed.
Conclusion

• The basic design of flask with respect to radiation shielding is adequate as seen from the radiometry report. The radiation field on the surface of the cabinet will be within the permissible limits. The dose evaluation study also reveals that the unit will be delivering dose rate of 8 Gy/ min** with an over dose ratio of 1.56. Hence the prototype unit qualifies all the criteria required for Category I irradiator as per AERB safety Guidelines No. AERB/RT-RPF/SG-2 (EDITION 2015).

• As the flaks is located inside the Shock absorber the radiation levels on the surface of the package will be less than 10 mr /hr which is 20 times less than the permissible radiation level stipulated in AERB Safety Code AERB/NRF-TS/SC-1 , Rev. 1, Atomic Energy Regulatory Board, India (Edition 2016).

• The stress Intensity induced in the critical components are well within the limits stated in ASME Code Section III, Div1, Appendix F. Also deformation induced is negligible hence no loss of shielding is observed for the DROP I & II test. The impact test has negligible effect on reduction of shielding of the package hence the radiation levels on the surface of the package remains unchanged.

• The thermal simulation results reveal that there is no lead melting occurring in the flask during the fire and post fire analysis.

• The Package design for transportation as TYPE B (U) Package qualifies the entire requirements as stated in the national and international regulatory codes.

References


New ways of producing Tc-99m Generators: Purification & Recycling methodology of Molybdenum

Sankha Chattopadhyay*, Sujata Saha Das, Luna Barua, Madhusmita, Md. Nayer Alam, Asit Kumar Pal, Umesh Kumar, Arup Hudait

Regional Centre, Board of Radiation & Isotope Technology, VECC, 1/AF, Kolkata
E-mail: sankha@vecc.gov.in

*This work is a part of Coordinated Research Project of IAEA and has been submitted during December 2017.

Abstract

The reliable supply of Molybdenum-99 (\(^{99}\text{Mo}\)) and its decay product, Technetium-99m (\(^{99m}\text{Tc}\)), is a vital component of modern medical diagnostic practices. The most popular \(^{99}\text{Mo}^{99m}\text{Tc}\) generator for the production of \(^{99m}\text{Tc}\) radioisotope is the Alumina Column Generator which involves obtaining high specific activity Mo-99 from highly enriched \(^{235}\text{U}\) target through \(^{235}\text{U}(n,f)^{99}\text{Mo}\) reaction. The other method of obtaining \(^{99m}\text{Tc}\) is by solvent exchange generator where low/medium specific activity \(^{99}\text{Mo}\), which is much less expensive, is produced in research reactor by \(^{98}\text{Mo}(n,\gamma)^{99}\text{Mo}\) reaction. Disruption in the supply chain of these radioisotopes, which cannot be effectively stored, can suspend important medical testing services to mankind. Unfortunately, supply reliability has declined over the past decade, due to unexpected shutdowns at the few ageing, \(^{99}\text{Mo}\)-producing, research reactors and processing facilities. These shutdowns have created global supply shortages and compelled to find new ways of producing \(^{99m}\text{Tc}\) generators. The present study includes the development of portable, simple and low cost \(^{99m}\text{Tc}\)-generator based on MEK solvent extraction technique, where low specific activity Mo-99 source obtained from linear accelerator by \(^{100}\text{Mo}(\gamma,n)^{99}\text{Mo}\) or \(^{nat}\text{Mo}(\gamma,n)^{99}\text{Mo}\) reaction, to obtain very high concentration of no-carrier added (nca) \(^{99m}\text{Tc}\) solution.
Introduction

Technetium-99m \( [t_\text{1/2} = 6.02 \text{h}; 140.51 \text{ keV (89\%)}] \) is the most important and useful radioisotope till today in diagnostic nuclear medicine, thanks to its optimal characteristics - short half-life, low-energy \( \gamma \) emission, its versatile chemistry, and relatively low cost. Technetium-99m is generally obtained from \( ^{99}\text{Mo}/^{99m}\text{Tc} \) alumina column generator where \( ^{99}\text{Mo} \) decays by \( \beta^- \) emission (87.5 \%) with a half-life of 66 h to \( ^{99m}\text{Tc} \). The high specific activity Molybdenum-99 is generally produced using highly enriched \( ^{235}\text{U} \) targets through \( ^{235}\text{U} (n, f) ^{99}\text{Mo} \) reaction. Recently, the issues have been raised for the conversion of fission \( ^{99}\text{Mo} \) targets from highly enriched uranium to low enriched uranium to produce high specific activity \( ^{99m}\text{Mo} \). There are other alternative routes to produce \( ^{99}\text{Mo} \) such as \( ^{98}\text{Mo} (n, \gamma) ^{99}\text{Mo} \) in a research reactor, \( ^{100}\text{Mo} (p, x) ^{99m}\text{Tc} \) or \(^{99m}\text{Tc} \) directly through \( ^{100}\text{Mo} (p, 2n) ^{99m}\text{Tc} \) in a cyclotron, \( ^{100}\text{nat}\text{Mo} (\gamma, n) ^{99}\text{Mo} \) in a linear electron accelerator. These routes produce \( ^{99}\text{Mo} \) with an appreciable amount of inactive molybdenum which in turn reduces specific activity of \( ^{99}\text{Mo} \).

The theme of the Coordinated Research Project (CRP) is to identify new technical aspects of the production and quality control of \( ^{99}\text{Mo} \) and \( ^{99m}\text{Tc} \), respectively and \( ^{99m}\text{Tc} \) generators with focus on photodynamic route through the reaction \( ^{100}\text{Mo} (\gamma, n) ^{99}\text{Mo} \), as well as generator development using high capacity adsorbents for Molybdenum to be employed with low to medium specific activity \( ^{99}\text{Mo} \) for application and distribution among Member States. Although the fission based Mo-99 remains the major source for \( ^{99}\text{Mo}^{-99m}\text{Tc} \) generators, in recent years Mo-99 supply crisis was raised by different factors such as the extension of planned outage, the prolonged or unplanned shutdown of many research reactors around the world. These unplanned events, combined with other planned outages created a worldwide Mo-99 supply crisis. In response to the crisis, producers increased the production of Mo-99 to the maximum extent possible and also alternative production routes were suggested for availability of Mo-99 and/or Tc-99m radionuclides.

A relatively new option for Mo-99 production has been a choice to exploit the photo-neutron reaction, i.e., \( (\gamma, n) \) reaction on \( ^{100}\text{Mo} \) or \( ^{nat}\text{Mo} \) target, using high-power electron linear accelerator (LINAC) where the required high-energy photons are available. It was reported that about 80 GBq of \( ^{99}\text{Mo} \) is accumulated in a 10g \( ^{100}\text{Mo} \) target (100\% enrichment) after irradiation for 100 h at a current of 25 \( \mu\text{A} \) and electron energy of 25 MeV. It was also reported that irradiation of 10 g of natural Molybdenum (9.6\% \( ^{100}\text{Mo} \)) under the same conditions yields 7.7 GBq of \( ^{99}\text{Mo} \). The \( ^{99}\text{Mo} \) yield can be increased by optimization of the geometric parameters of the photon flux and the target. However, when natural molybdenum is exposed to \( \gamma \)-ray irradiation, not only \( ^{99}\text{Mo} \) but also long-lived impurities of \( ^{90,91,91m,93m}\text{Mo} \) are accumulated in the target. Moreover, the niobium impurities such as \( ^{97}\text{Nb}, ^{93}\text{Nb}, ^{95}\text{Nb}, \) and \( ^{91m}\text{Nb} \) are formed, respectively by the \( ^{nat}\text{Mo} (\gamma, p) ^{97,96,95,91m}\text{Nb} \) reaction. The niobium impurities like Niobium-99\(m \) and Niobium-99 are also formed in the irradiation of a \( ^{100}\text{Mo} \) target. In both the cases after irradiation of natural Mo and enriched Mo-100, Niobium isotopes should be removed from Molybdenum which can then be safely used for the preparation.
brief communication

of $^{99}$Mo-$^{99m}$Tc generators. To isolate $^{99m}$Tc from highly enriched Molybdenum targets, new procedures allowing efficient separation of expensive pure $^{100}$Mo should also be developed. To get high purity pharmaceutical grade $^{99m}$Tc-pertechnetate at hospital radiopharmacies from low specific activity $^{99}$Mo along with co-produced non-isotopic radioactive and nonradioactive non-isotopic impurities, produced from ($\gamma$, n) reaction on $^{100}$Mo or nat$^{99}$Mo, we need to develop an user-friendly and low cost $^{99m}$Tc generator satisfying the quality of $^{99m}$Tc as per standard pharmacopeia and also we have to give an attention to recover costly enriched $^{100}$Mo target when $^{99}$Mo is produced via enriched Mo-100 ($\gamma$, n) reaction in the purest form from the spent $^{99}$Mo/$^{99m}$Tc generator.

There are several methods for routine separation of $^{99m}$Tc from $^{99}$Mo depending on the specific activity of $^{99}$Mo. The most popular one is the alumina column chromatography. The capacity of adsorption of molybdenum on alumina (20 mg/g of alumina) restricts its use to high specific activity $^{99}$Mo such as the one obtained by thermal fission of $^{235}$U$^{2}$. The low specific activity $^{99}$Mo cannot be used for this purpose since it requires a very large alumina column to adsorb about a gram quantity of Mo needed for preparing generators containing $\leq$18.5 GBq (500 mCi) $^{99}$Mo at the reference time. The large size of the alumina column, in turn, requires large eluate volumes to recover the $^{99m}$Tc and the radioactive concentration (RAC) of the Pertechnetate becomes unacceptably low for radiopharmaceutical use$^{4}$. A new series of micro-porous/meso-porous materials, such as low-cost naturally occurring materials, nanocomposites, etc., having high capacity for adsorbing Mo$^{10-13}$, has also been demonstrated to prepare $^{99m}$Tc-generators at multi-Curie levels. The separation of pertechnetate (from the bulk Molybdate with low specific activity $^{99}$Mo) can be achieved using one of the many strategies (e.g. liquid-liquid extraction$^{14}$, ion-exchange chromatography$^{4}$, zirconium molybdate gel$^{15,16}$, electrochemical method$^{17}$). The automated solvent-extraction based modules have been described by Chattopadhyay et. al.$^{14,18}$ and Martini et al.$^{19}$. The automated Solid Phase Extraction (SPE) based modules have been developed and described by Schaffer et. al.$^{20}$, Das et al.$^{6}$, Morley et al.$^{21}$, McAlister and Philip Horwitz$^{22}$.

The production of $^{99}$Mo through the $^{100}$Mo ($\gamma$, n) $^{99}$Mo or nat$^{99}$Mo ($\gamma$, n) $^{99}$Mo reactions in LINAC results low specific activity $^{99}$Mo as well as isotopic and non-isotopic impurities$^{8,9}$, therefore, the techniques for effective extraction of $^{99m}$Tc from low specific activity $^{99}$Mo as well as a suitable recycling process to recover the costly enriched $^{100}$Mo after irradiation and $^{99m}$Tc separation for future targets needs to be developed.

The MEK based solvent extraction technique is a well known method for the separation of $^{99m}$Tc from low specific activity $^{99}$Mo produced either through $^{98}$Mo ($n,\gamma$)$^{99}$Mo or $^{100}$Mo ($\gamma$,n)$^{99}$Mo and $^{99m}$Tc produced directly through $^{100}$Mo ($p, 2n$) $^{99m}$Tc nuclear reaction$^{14,18,23}$.

Here we report a new, portable solvent extraction based $^{99}$Mo/$^{99m}$Tc generator in tandem with alumina column utilizing ($n,\gamma$) $^{99}$Mo to produce highly purified, concentrated, clinical grade $^{99m}$Tc which also may be applicable for preparation of $^{99m}$Tc generator where $^{99}$Mo is produced from $^{100}$Mo($\gamma$, n) reaction in LINAC. We also report a novel, indigenous methodology for purification of $^{99/100}$Mo produced in LINAC to
remove co-produced Nb isotopes (a simulation radiochemical study) required for preparation of Tc-99m generator. It is also reported the strategies to be followed for recycling of enriched Mo-100.

**Target Recovery**

The target recovery is a very important step to complete the $^{99m}$Tc production cycle where costly enriched Mo-100 is used. Since the natural abundance of $^{100}$Mo (9.63%) is very low and there is a need for a highly enriched target, to achieve high Radionuclidic Purity (RNP) of the final $^{99m}$Tc and high yield of production of $^{99}$Mo in photonuclear reaction, makes the target very expensive, thus target recycling is a very important task. An exhaustive evaluation of the $^{100}$Mo recovery strategy needs to be attempted in order to keep the $^{99m}$Tc production costs within affordable range and the RNP of the final product, from recycled target, within the Pharmacopoeia limits.

In this process, the group of Argonne National Laboratory, Los Alamos National Laboratory, and North Star Medical Technologies has extensively studied and developed the methodologies in the recovery of enriched Mo-100 from spent $^{100/99}$Mo-$^{99m}$Tc generator where $^{99}$Mo has been produced through $^{100}$Mo ($\gamma$, n) photonuclear reaction. They have recovered molybdenum oxide by precipitation of Mo by adding glacial acetic acid or by adding Tetra Butyl Ammonium Bromide from molybdenum in 5N KOH solution. The group has achieved a high purification factor from potassium, and typical Mo recovery yields were about 95%. Due to the high cost for the enriched target material, potential manufacturers require a means to efficiently recycle the enriched Mo into new targets as part of the complete production scheme. Impurities present in the enriched material, as well as those introduced during post-irradiation processing or recycling, need to be closely monitored.

1. **Development of a portable, simple and low cost $^{99m}$Tc-generator based on MEK solvent extraction technique**

Solvent extraction using methyl ethyl ketone (MEK) is effective, simple and cost effective and has a long history of validation. It has been widely used for reactor sources of Mo-99 and would be especially useful for preparation of $^{99}$Mo-$^{99m}$Tc generator from low specific activity Mo-99 source as this is obtained from linear accelerator by $^{100}$Mo ($\gamma$,n) $^{99}$Mo or natMo ($\gamma$,n) $^{99}$Mo reaction. Whether the particular equipment (module) uses evaporation of MEK step or small column trapping of Tc-99m from organic solvent in the latter steps, these purification steps offer the advantage onto the user who has the flexibility on the volume of the saline to be added to the Tc-99m for reconstitution of pertechnetate injections. The user therefore can obtain very high concentration of no-carrier-added Tc-99m solutions even from low activity of Tc-99m in the useful form and also make use of low specific activity $^{99}$Mo.

To be most useful to hospitals and radiopharmacies, the generator should be in automated configuration. An automated solvent extraction generator which has been used by various countries gives enhanced radiological and pharmaceutical safety and offers convenient and very effective chemical separation and yield of sodium [${^{99m}}$Tc] Tc-pertechnetate. In most cases, solvent extraction equipments that have been reported are more appropriate for Centralized Radiopharmacy or large Radiopharmacy operations than smaller nuclear clinic operations (hospital
radiopharmacy). This is necessitated by the higher level of expertise, needed to operate the equipment effectively and safely, ensure that extractions are carried out in cGMP conditions, and also because solvent extraction generators are generally not portable. The production of pharmaceutical grade $^{99m}$Tc from low specific activity $(n, \gamma)^{99}$Mo from $^{98}$Mo or $(\gamma, n)^{99}$Mo from $^{100}$Mo utilizing solvent extraction technology to spread to hospitals like conventional column generator (small size and simplicity in operation). For that we must need some logistic changes in few parameters of the MEK solvent extraction methodology. We have reduced the size of the solvent extractor & minimize the volume of the $^{99}$Mo aqueous and organic solvent [Figure 2-5].

We have designed, developed and fabricated a portable and more user friendly $^{99}$Mo/$^{99m}$Tc generator [Figure 1] based on solvent extraction technique utilizing $(n, \gamma)^{99}$Mo in sodium chloride solution for usage in hospital radiopharmacy$^{28}$. Some hospitals are still using the conventional manual solvent extraction method to extract radiopharmaceutical grade $^{99m}$Tc from large volume of alkaline $^{99}$Mo-molybdate solution in 5N or 6N NaOH/KOH, resulting generation of large radioactive waste. An extraction strict to 5N/6N NaOH condition increases the probability of induced aldol condensed impurities in the final product during evaporation of MEK. In view to utilize low specific activity $^{99}$Mo we have used $(n, \gamma)^{99}$Mo produced in BARC reactor, India. In view to make a mobile, portable, compact and ready to use conventional $^{99}$Mo-$^{99m}$Tc alumina column generator, we may need some logistic changes in few parameters of the solvent extraction principle and usage. The size and shape of the newly developed generator of 500mCi would be similar to commercial alumina column generator.

**Experiment:** The new $^{99}$Mo/$^{99m}$Tc generator system utilized $(n, \gamma)^{99}$Mo and was MEK based solvent extraction technique in absence of NaOH. An aqueous solution of $(n, \gamma)^{99}$Mo (200-500mCi) in dilute NaOH solution (6-15 ml) obtained from BARC/BRIT, Mumbai was taken in an extraction tube (50ml) kept in a lead shielding arrangement. The pH of the solution was adjusted to 7 and saturated with solid NaCl. The pertechnetate ($^{99m}$TcO$_4^-$) was selectively extracted in MEK from aqueous sodium chloride saturated $^{99}$Mo solution and passed through two small alumina column (neutral alumina to remove traces of $^{99}$Mo and acidic alumina to hold $^{99m}$Tc selectively from MEK) in tandem with the extraction tube. Finally, highly pure $^{99m}$Tc was recovered from acidic alumina column with 3-5 ml saline in high concentration. The performance of this new $^{99}$Mo/$^{99m}$Tc generator based on MEK solvent extraction in absence of NaOH was evaluated by studying the recovery yield of $^{99m}$Tc, physico-chemical tests and radiolabelling with the standard radiopharmaceutical kits like MDP, DTPA & MIBI. The pertechnetate obtained as above was checked for clarity, pH, radiochemical purity (RCP) and chemical purity.

**Results & Discussion:** Different batches of $^{99}$Mo-Moly (Batch size: 200mCi or 500mCi) in absence of NaOH & in presence of saturated NaCl were processed to recover pharmaceutical grade $^{99m}$Tc. The $^{99m}$Tc-pertechnetate solution was clear having pH 6-7. The average yield of separation of $^{99m}$Tc was about 90% and $^{99}$Mo breakthrough in $^{99m}$Tc pertechnetate was <0.0002% ($n=10$). The final $^{99m}$TcO$_4^-$ fraction has the Mo and Al content <10ppm and MEK content <0.1 % v/v, with RC Purity >99% and RN Purity >99.9%. The efficacy of labeling specific compounds was assessed using standard
radiopharmaceutical kits, such as $^{99m}$Tc-MDP, $^{99m}$Tc-DTPA and $^{99m}$Tc-MIBI and the RC Purity was above 95% (n = 6). The newly developed portable $^{99m}$Mo/$^{99m}$Tc generator based on MEK solvent extraction in absence of NaOH can provide the highly concentrated and purified $^{99m}$Tc-pertechnetate using indigenously produced $^{99}$Mo from our research reactor and finds its application for hospital radiopharmacy. This method may directly be applicable in separation of pharmaceutical grade $^{99m}$Tc from low specific activity Mo-99 source produced in linear accelerator by $^{100}$Mo ($\gamma$, n) $^{99}$Mo or nat$^{99}$Mo ($\gamma$, n) $^{99}$Mo reaction.

**2. Simulation Experiment for the Recovery and purification of irradiated Mo**

Pre-purification of $^{99}$Mo before preparation of $^{99}$Mo-$^{99m}$Tc generators: This chemical separation step is required in the preparation of $^{99}$Mo-$^{99m}$Tc generator from low specific activity $^{99}$Mo produced through the $^{100}$Mo ($\gamma$, n) $^{99}$Mo or nat$^{99}$Mo ($\gamma$, n) $^{99}$Mo reactions in LINAC$^{8,9}$. There are several methods like sublimation, precipitation, liquid-liquid extraction and anion exchange column methods$^{8,9,24,26}$ to recover molybdenum with
least radioactive and non-radioactive (Zr, Nb, Y etc.) impurities. We have developed a method by simulation tracers methods which may be adapted for large scale purification of $^{99}$Mo produced in LINAC from Mo-100 through photodynamic reaction [Figure 5].

![Figure 5: Schematic of purification of $^{99/100}$Mo to remove co-produced Nb isotopes in ($\gamma$, n) reaction with Mo-100](image)

**Figure 5:** Schematic of purification of $^{99/100}$Mo to remove co-produced Nb isotopes in ($\gamma$, n) reaction with Mo-100

1. **Study to remove radioisotope of Nb, Y & Zr from $^{99m}$Tc [co-produced impurities in $^{100}$Mo (p, 2n) & $^{nat/100}$Mo ($\gamma$, n) reaction] in inactive Mo (By MEK Solvent Extraction method):**

The radioactive tracers (Nb, Y, Zr & Ru) were produced in traces especially in VECC, Kolkata, cyclotron. A solution of inactive Mo (in NaOH, KOH, NH$_3$OH) was doped with tracer isotopes & Tc-99m and neutralized to pH-7 and saturated either with NaCl, KCl or Ammonium Carbonate (Test solution). A simulation experiment was carried out to study if Nb, Y, Zr and Ru radioisotopes are eliminated by the adopted MEK solvent extraction method. An aliquot of samples from the test solution as well as from all the solution obtained after separation were taken and counted in the detector.

**Result:** Non-isotopic impurities were absent in the organic fraction of $^{99m}$Tc obtained in the MEK solvent extraction process.

**Conclusion:** The MEK solvent extraction can directly be used in preparation of $^{99m}$Tc generator produced from $^{99}$Mo via $^{nat}$Mo ($\gamma$, n) reaction.
Acknowledgment

The work was performed in part under a research contract between the International Atomic Energy Agency (IAEA) and the Board of Radiation and Isotope Technology (BRIT), India (Research Contract no. 22475). The investigators thank Shri G. Ganesh, Chief Executive, BRIT and Dr. Sharmila Banerjee, Head, RMC, BARC & Senior General Manager, BRIT for encouragement and support on the project. The investigators are grateful to Shri Amitava Roy Director, VECC, Kolkata and Dr. Tapas Bandyopadhyay, Officer in Charge, Heath Physics Unit, BARC, VECC, Kolkata, for supporting the work.

References


23. Cyclotron based Production of Technetium-99m: Publisher: International Atomic Energy Agency: http://www-pub.iaea.org/books/IAEABooks/10990/Cyclotron-Based-Production-of-Technetium-99m, Date Published: June, 2017


Calibration Laboratory - A Necessity

S.B. Kamble¹, B.K. Pathak¹, A.S. Gengje², L.N. Bandi², P. Srivastava²

¹Calibration Services Group, Marketing & Services Programme, BRIT
²Engineering & Corporate Planning Programme, BRIT

General

Since time immemorial man and radiation have co-existed¹. With advances in science and development of modern technology, man-made radiation has become part and parcel of human life. Starting with simple diagnostic X-ray unit, nuclear medicine centers, radiography devices, nucleonic gauges used in different industries, nuclear research/power reactors, particle accelerators etc; the presence of radiation cannot be avoided. Man has immensely benefited, directly or indirectly from various applications involving radiation. But one has to take into account the risk involved while working with radiation. There is always a trade-off between risk and benefit. Radiation dose below certain level does not cause any harm to humans. As radiation is invisible and otherwise also, we have to monitor and record the radiation levels/dose for radiological safety of the direct beneficiary, staff and also general public.

For radiation monitoring and records, various type of instruments are available. These instruments indicate/record instantaneous radiation levels/radiation dose for a given time interval. In India, Atomic Energy Regulatory Board (AERB) is the authorized body for licensing and regulating the use of radiation sources/equipments and the facilities.

In today’s scenario of whole world, there is possibility of radiation source/s falling into wrong hands and also the likely hood of it being used to carry out terror activities in public places. An unfortunate accident took place at Mayapuri in Delhi, in the year 2010, where a disused radiation source landed in the hands of a scrap dealer; which caused one death and radiation injury to some more people. In order to investigate such cases, to trace missing/orphan radiation sources, survey of the places where radioactivity is being handled, a properly working radiation monitoring instrument is one of the important tools to ensure radiological safety. It helps to access the radiation risk/damage on the site as well as in surrounding areas.

Any instrument is prone to faults due to aging of components, component failure and/or improper handling and hence needs to be tested at regular intervals by the authorized personnel under controlled conditions. This is where the role of Calibration Laboratory comes into picture. The laboratory should have the necessary infrastructure and trained staff. Also, it should be recognized by appropriate authority; in this case AERB.
**Introduction**

BRIT has set up a Calibration Laboratory for calibrating Radiation Monitoring Instruments (RMIs) like Survey meters and Pocket Dosimeters, with aim to provide calibration service to various customers using RMIs for their applications. This adds to the list of services provided by BRIT. The Laboratory is approved by AERB in accordance with Rule-29 of Atomic Energy (Radiation Protection) Rules, 2004 and started functioning since August-2014. Calibration service is offered to in-house users, various divisions in BARC and most importantly to private customers like nuclear medicine centers, radiography firms and various industries using nucleonic gauges etc. The customers from far and wide places like Srinagar to Chennai and from Rajkot to Nazira, in Assam have availed the services. We observed about 15% of meters received do not comply calibration criteria, such cases if not detected and corrected will mislead the user, which is an unsafe situation and must be avoided.

**Details of the Facility**

The laboratory essentially consists of two parts: a control room and a calibration room. The calibration room has a radiation exposure device, specially fabricated for calibration purpose and contains Cs-137 source$^4$ of 2 Curie nominal activity. Also, there is a digital camera which is to be focused on the meter under test to record meter response, a calibration bench with distancing and alignment arrangement to place meter under test at appropriate distance/height from the source/floor. The exposure device is remotely operated from control room. The laboratory also has radiation monitors, radiation interlock facility for automatic locking of entry door upon radiation exposure, a TV monitor to display the image for noting observations of meter under test and audio-visual alarms to know the radiation exposure status.

A view from control room

Calibration set-up

The reference radiation field is standardized by the Radiation Standards Section/RSSD/BARC, which is the National Standards Laboratory in India for ionizing radiations; using Standard ion-chamber maintained by them and traceable to international standards. The radiation field on reference date is known with accuracy of...
± 5%. By applying decay calculations and inverse square law, we can calculate the dose rate at a particular distance and vice-versa. Considering the strength of the source and other specifications of exposure device, RMIs in the range from 0.1 mR/h to 5 R/h and other equivalent units can be calibrated.

Conclusion

As we inch forward as fully developing nation, there will be an increase in the number of industries, applications, new radiation techniques, nuclear medicine centers, radiography firms etc; the number of RMIs/Types required by them are bound to increase steadily, which will need calibration in due course of time. In order to provide efficient service to customers and to cover wider calibration range, the facility will have to be augmented suitably.

References


2. AERB document on setting up of calibration laboratory


4. ISO 4037
Report on RADIOTRACER STUDY of Fly-ash disposal into mine void in Quarry No. 4 of Jagannath OCP for M/s. Bhushan Steel Ltd.

V.N. Yelgaonkar, M.C. Panicker, Gaurav Agrahari, C.B. Tiwari, Vikrant Dhakar, B.K. Pathak

Isotope Application Services, Board of Radiation & Isotope Technology,

Introduction

Board of Radiation and Isotope Technology (BRIT) was carved out of Bhabha Atomic Research Centre, Department of Atomic Energy, Government of India to undertake commercial activities of radioisotope and radiation applications along with other applications. In other words it is a commercial wing of Department of Atomic Energy. Few of the industrial applications of radioisotopes like Gamma column scanning, blockage, void, corrosion detection in pipelines, identification of location of leakage in underground pipelines, residence time distribution analysis in reactor vessels of any kind, flow rate estimation and flow-meter calibration, effluent dispersion studies in surface waters and sediment transport studies on river/sea bed, bore-well interconnection studies for groundwater, studies for the enhanced recovery of oil from the oil wells, reservoir development, interconnection between oil wells, monitoring secondary recovery of oil and its effectiveness, etc are undertaken by BRIT. BRIT also supplies industrial irradiators for the irradiation of surgical items for sterilization, food grains for removal of pests and enhancement of their shelf life, etc. Gamma chambers of various capacities are supplied for research purposes. Indigenous radiography cameras (Industrial gamma radiography exposure device) are supplied for industrial radiography and the radioisotopes are provided for the imported radiography camera. For diagnosis and therapy, radioisotopes are produced and supplied to the hospitals in India and abroad. BRIT has laboratories for radiopharmaceutical distribution at various locations throughout India.

Out of the above activities undertaken by BRIT/BARC, radiotracer studies for dilution and dispersion of pollutants in surface waters is helping various agencies to decide upon the outfall design and its efficacy.

Theory of Radiotracer Study

The basic principle of tracer investigation is to label a substance, an object or a phase and then to follow it through a system or to carry out a quantitative assay of the tracer after it has left the system. The requirements of tracer are that: it should
behave in the same way as the material under investigation, it should be easily detectable at low concentrations, detection should be unambiguous, injection, detection and/or sampling should be performed without disturbing the system, the residual concentration in the system after the study period should be minimal. All these criteria can be met using radioisotopes as tracer and by careful selection of the most appropriate tracer for a particular application. Factors which are important in the selection of radiotracer are: Half life – should be long enough to allow time to transfer the tracer from the nuclear reactor to the work site, prepare the tracer for use and complete the measurements. In order to reduce the level of residual tracer in the system short or optimum half-life tracer is desirable. Type and energy of radiation – should be detectable at lower concentrations either by sampling or in-situ detection, will have direct bearing on the total amount of activity which can be accommodated safely within given system. After injection, self-absorption by water present in the system may reduce the level of radiation to the levels which should be within the legal limits. Physicochemical form – should be compatible with the material being traced both in physical form and chemical form and preferably behave same as the material being traced in the system. The ideal tracer in these circumstances is undoubtedly the irradiated material itself i.e. irradiated fly ash.

The final choice of radiotracer for an investigation is made after consideration of all of the above factors, many of which may be mutually exclusive.

Preferably, highly sensitive detectors which are pre-calibrated are used to track the progress and strength of the radiotracer. In the current scenario, in order to understand and establish the transport of heavy metals and other trace elements from the fly-ash to the surrounding environment, Scandium-46 (with half-life of 84 days and emitting 0.887MeV and 1.119MeV gamma rays) is selected as a radiotracer.

Radioactive methods can help in investigating suspended sediment dynamics, providing important parameters for better designing, maintaining and optimizing disposal of suspended load into the surface water bodies. Radioisotopes as tracers and sealed sources have been useful and often irreplaceable tools for such studies.

For the study of the behaviour of the suspended load, the material being disposed is labelled with the radioactive isotopes such as Au-198/Sc-46 and injected in the water body. In this study the fly ash was labelled with Sc-46 isotope in chloride form. The detection of the radioactive cloud is achieved by towing immersed detectors at different depths.

There are three main transport mechanisms active in the transport of suspended particles:

1. Advection(currents)
2. Dispersion(turbulences)
3. Decantation(specific weight and volume of particles)
Dispersion Coefficient

Dispersion coefficient can be calculated by the method of moments:

\[ D_y = \frac{\sigma y_2^2 - \sigma y_1^2}{2(t_2 - t_1)} \text{m}^2/\text{s} \]

Where \( \sigma y_1 \) and \( \sigma y_2 \) are the variance of cross plume concentration profiles at the sections 1 & 2 and \( t_1 \) & \( t_2 \) are time elapsed from the discharge point to the corresponding section.

Similarly, longitudinal dispersion coefficient can also be estimated using method of moments.

Decantation rate

The quantity of suspended matter at any moment can be obtained from

\[ M(t) = M_0 e^{-\frac{w(t-t_0)}{H \Phi}} \]

Where: \( t \) = time
\( t_0 \) = time of injection
\( w \) = sinking speed of suspension particles
\( H \) = water depth
\( M_0 \) = total mass of suspension tracer
\( \Phi \) = dimensionless function in Rouse’s theory

\[ \Phi = \int \left[ \frac{a}{H} \left( 1 - \frac{z'''}{z''} \right) \right] \frac{w}{z''''} \text{dz} \]

\( z'' = z/H \) reduced height above bed
\( a \) = height of detector above bed
\( k \) = van Karman coefficient
\( u \) = shear stress velocity

Variation of \( M \) with respect to time gives decantation rate:

\[ \theta = \frac{1}{M} \left| \frac{dM}{dt} \right| \text{gm/sec-ton suspension} \]

Case studies show that flocks are formed during slack water. When currents induced due to wind are active, flocks disintegrate and tend to become homogenous.

Transport Velocity

From the iso-count contours, a plot between cumulative of product of count rate and length of lateral spread (Y) \( C_1 = \sum_c \cdot y \) for different locations(x) along the axis of movements is plotted against x, so that contour map is reduced to one dimension. The count distribution diagrams so generated are called as ‘transport diagrams’. For each diagram, the location of the weighted centre of gravity along the axis movements is found out using

\[ X = \frac{\int C_1 \cdot x \cdot dx}{\int C_1 \cdot dx} \]

Successive tracking in time make it possible to establish many centres of gravity and the mean velocity of movement (V_m) is calculated from the shifts in the centre of gravity between two successive tracking.
Radioactive tracers are the only unequivocal method of direct real time assessment of distribution of suspended matter in the surface water as well as ground water. Radiotracers are more sensitive and provide more accurate parameters than conventional tracers. In recent decades, many radiotracer studies for the investigation of suspended sediment transport in natural systems have been conducted worldwide, and various techniques for tracing and monitoring the suspended sediment have been developed by Isotope Application Services of BRIT. In addition to radiotracers, sealed source techniques can provide information on the density of suspended sediments in a channel of navigation as well as on the concentration of sediments circulating in suspension.

The environmental, economic and social benefits from the application of radiotracer and sealed source techniques can be enormous.

Advantages and Disadvantages of using radiotracer technique

Radiotracer technique is carried out without disturbing the system i.e. online. The radiotracer as the name suggests is used in trace quantity in comparison with the material in the system as it can be detected at very low concentrations using the highly sensitive radiation detectors. The detection does not depend upon physical or chemical changes during the study period as the nuclear properties of the radiotracer do not change during the course of the study. Since the properly selected radiotracer either in the same form of the material being traced or labelled on the traced material follows intended flow paths and undergoes same changes as the material being traced, ideally it follows the same flow dynamics of the mother material including leaching, sorption, desorption, flocculation, de-flocculation, floatation and settling. The conventional tracers like dyes, salts, fluorescents, etc. often are interfered by other physical or chemical parameters but radiotracers have no such adverse effect of the suppressing parameters.

Disadvantage of using radiotracers is, it requires trained manpower, additional training for handling of radioisotopes and knowledge of radiation safety. Contamination due to the use of radiotracers in powder as well as liquid form requires huge efforts to deal with.

Safety Issues

Since the water body of mine void is huge and the labelled fly ash being disposed should truly represent the bulk fly ash, the quantity of radiotracer theoretically arrived at is about 5 Ci. The selected radiotracer i.e. scandium (powder of $Sc_2O_3$) in sealed aluminium can needs to be brought to the site by road in a lead container weighing about 800kg with proper regulatory approvals of transportation and usage. The vehicle transporting this will be properly labelled with necessary safety signs.

After it arrives at the site it will be kept secured in a locked room. The handling for making it in to chloride form will be done using long handled tongs. The radioactive scandium chloride will be remotely transferred to the fly ash conditioner using a peristaltic pump. After sufficient time given for labelling, the fly ash will be disposed off in to mine void.

In general principle of ALARA (as low as reasonably achievable) will be strictly
followed while performing the entire operation. Similarly the operations of handling the radiotracers will be carried out in minimum possible time, keeping the safe distance between source and personnel and using maximum possible shielding wherever required.

**Scope of the Work**

**Description about the Site**

M/s. Bhushan Steel limited (BSL) and M/s. Bhushan Energy limited (BEL) are located at Narendrapur village in Meramandali, approx 20 km from Angul town, under the jurisdiction of Dhenkanal district and about 140 km from Bhuvaneshwar in Odisha. Figure 1).

![Figure 1: Location map of study area](image)

M/s. Bhushan Energy limited has in-house thermal power plant (TPP), generating 883MW of electricity. Burning of coal in the plant generates 5000 tons per day of fly-ash (1.65 MTPA) which is proposed to be disposed off in a mine void about 25 km from the plant. The fly-ash disposal site is located in the micro watershed covered between latitudes 20°52′00" N and 20°59′00" N and longitudes 85°07′30" E and 85°15′30" E. The area of study experiences tropical climate with mild winter and hot summer with an average rainfall of 1250mm during June-Sept (monsoon). It is characterized by uneven topography, some scattered hillocks, forest blocks and rocky outcrops. The altitude ranges from 58m to 139m AMSL and the slope is towards the south east direction.

**Narrative of the problem**

Safe disposal of fly ash is a major issue as it contains several toxic chemical constituents which may pollute the environment. Although, utilizing fly-ash for manufacturing bricks and cement could take care of this issue partially, the cost of transporting fly-ash to concerned factories limits its utility.

Ministry of Environment and Forest (MoEF) has accorded permission for disposal of fly ash from the BSL TPP at the disused quarry No. 4 of Jagannath opencast mines of Mahanadi Coalfields limited (MCL). The mine void is located 25km from the TPP and covers an area of 119Ha. The fly-ash generated in the TPP is brought to the quarry in closed vehicles (bulkers) in dry form and is disposed into the mine void after conditioning and making 60% slurry with water. BSL has been disposing fly-ash to the mine void since March, 2014. In order to assess the environmental impact of the fly-ash disposal in the vicinity of the mine void, National Environmental Engineering Research Institute (NEERI (CSIR)) undertook a project to survey the underground and surface water quality in the pre and post monsoon seasons.
Their findings for the ground water showed that the current concentrations of all the cations, anions except the nitrate and fluoride concentrations were within the allowable limits prescribed by Bureau of Indian Standards (BIS). The concentration of all heavy metals except Al, Mn and Ni were also found to be within the permissible limits of BIS. The petrography study indicated presence of Fluoride and Aluminium containing minerals in the rocks, hence it was concluded that the higher concentrations of fluorides and Al was geogenic in nature. A Toxicity characteristic leaching procedure (TCLP) and water extraction test was used to study the leaching of heavy metals from the fly ash. The findings of TCLP and water extraction test showed that the leaching was well within the acceptable international limits. They also concluded that the plume movement is at a pace of maximum 700m in 30 years starting from March 2014 using a groundwater mass transport model MT3D. However, to comply with condition No.-3 (Incorporation of Radioactive tracer studies for heavy metal) of Environmental Clearance, Bhushan Steel Limited approached Board of Radiation and Isotope Technology (BRIT) to carry out the Radiotracer study to understand the leaching of heavy metals from fly ash in to the surface water and surrounding ground water.

Geology and Hydrogeology

The area is largely covered by sedimentary rocks of karhabari and barakar permocarboniferous formation having large deposits of coal belonging to Gondwana group overlying Talchir formation consisting of very thick sandstone and shale sequence. These Gondwana group are overlain by recent alluvium and valley fill materials mainly along the river courses. A small part of granitoid rock of the Eastern Ghats is also exposed in the S-E and S-W part (Figure 2). The sandstones are pale brownish yellow in color, massive, medium to coarse grained and contains Talchir shale, all held together loosely by a clay matrix bearing a slightly greenish tint. The barakar formation which overlies karhabari, is characterized by a thick and conspicuous conglomerate horizon at its base. The conglomerate members form low ridges in the southern and northern part parts of the coalfield. The basal conglomerate unit is overlain by a thick sequence (more than 500m) of medium to coarse grained greyish feldspathic sandstone, grey to dark grey shale, carbonaceous shale, thick coal seams mostly inter bedded with shale.

![Figure 2: Geological map of study area](image)

The area falls under the Brahmani tributary. The ground water reservoir in the area is semi-consolidated Gondwana formations comprising mainly of sandstone,
shale and crystalline rock of Precambrian age. The weathered and fractured sandstone constitute good aquifer and rainfall and seepage from the river Brahmani are the major sources of groundwater replenishment in this area. Groundwater occurs under water table condition in the weathered zone and under semi confined and confined condition in the fracture zone. The depth of dug wells in these formations ranges from 7.2m to 10.5m BGL.

**Targeting the task**

A preliminary site visit was carried out by BRIT scientists to understand the problem and to observe the study area as well as surroundings. There was a mine void filled with water surrounded by thick vegetation (Figure: 3).

![Figure 3: Picture of mine void (west half and east half joined together)](image)

On the upstream side (south side of the void) there was an approach road to the void area. On the south bank of the void towards the western corner on a flat plain ground there were fly-ash conditioners and auxiliary water storage facility and various pumping facilities (Figure: 4).

![Figure 4: Fly ash conditioner](image)

When bulker used to arrive with a load of fly ash, it was connected to the conditioner. The fly ash used to get transferred to the conditioner from bulker pneumatically. In the conditioner fly ash was properly made in to the slurry using water flow with the help of an agitator (Figure: 5).
The homogenized slurry in the proportion of 70% water + 30% fly ash was disposed into the mine void at a constant outflow rate. The additional water jets were spread on the outlet through the nozzles to increase the fluidity of the slurry.

Since the fly ash is disposed off in to the void water, the fly ash may leach in to the void water. The leachates may contain heavy metals which could get percolated in to the ground water contaminating the ground water in the surrounding bore wells. To study the extent of leached heavy metals with respect to time, it was proposed to label the fly ash with a suitable radiotracer while disposing it in to the mine void and to study its spatial and temporal distribution in the water of mine void and its subsequent progress in to the surrounding bore wells.

Similarly to estimate ground water velocity in the surrounding formations, a study to determine the time taken by radiotracer to travel short distance between two bore wells in the southern bank of the void was proposed.

**Experimental**

**Selection of Radiotracer**

Ideally, irradiated fly ash would serve as the best tracer for the proposed study. For this fly ash would be irradiated in nuclear reactor to generate various radioisotopes and disposing this irradiated fly ash into the water of mine void to study dynamics of leachates, both in surface and ground water. However, irradiation of the fly ash would generate radioisotopes of various heavy metals present in the fly-ash and would pose a serious environmental problem of long lived radio-isotopes like Zn-65 (half life: 244 days). Please see the following Figure: 6, for the nuclear properties of Zn-65.
Hence, it was decided to carry out the radiotracer study by fly-ash labelled with Sc-46 (Gamma energies: 0.89, 1.1 MeV, Half-life: 84 days).

For studying ground water movement in the two close bore wells, Mo-99 (average gamma energies: 730 KeV, Half life: 2.7 days) as sodium molybdate was proposed.

**Laboratory studies**

Fly ash is labelled efficiently with Scandium as scandium chloride in acidic medium. However, Scandium being rare element is available in its oxide form. Also, the Scandium in oxide form is preferred for the irradiation in nuclear reactor and is a very popular radiotracer for studying bed load movement in ports and harbours.

Therefore in the laboratory, a study was carried out to estimate the molarity and quantity of HCl required dissolving a specific amount of scandium oxide powder to make it scandium chloride.

It was observed that 12 mg of Sc$_2$O$_3$ powder was dissolved in 30 mL of boiling concentrated HCl. Furthermore, scandium chloride thus formed was not precipitated after cooling it to the room temperature.

**Neutron Irradiation**

The optimum quantity of radioactivity of Sc-46 was estimated to be 5 Ci which would be sufficient to be detected after the leaching and dilution in both water in the mine void and ground water. The Weight of Sc$_2$O$_3$ powder was calculated to get the required activity (5 Ci of Sc-46 at the time of injection) after irradiation for one week and pile factor of 13 in Dhruva research reactor at BARC.

\[
W = \frac{A \cdot M_c e^{-\gamma t_d} \cdot 100}{\sigma \phi N_a N_e I_n (1 - e^{-\gamma t_s})}
\]

Where, $A =$ required activity

$M_c =$ molecular weight

$\gamma =$ decay constant

$t_d =$ duration of irradiation

$\sigma =$ neutron absorption cross section

$\phi =$ neutron flux

$N_a =$ Avogadro no.

$N_e =$ enrichment factor

$I_n =$ no. of atoms in one molecule

1.1 grams of Sc-45 in the form of Sc$_2$O$_3$ amorphous powder was irradiated for one week to obtain approximately 5 Ci of activity.

**Preparations at Site**

In order to monitor the percolation of radiotracer through the groundwater, water samples could be drawn from the available bore wells. However, to get the better understanding about the groundwater movement additional bore wells were requested to be drilled.

In the upstream area on the south bank of the mine void already there was a bore well on one side of the road (BPZ1). Another bore well was drilled on the other side of the road (BPZ3) about 20.3 meters away from the first bore well to study
velocity of groundwater in the vicinity of study area (Figure: 7).

On the flat plain ground i.e. the injection point of the study area, there were 3 fly ash conditioners out of which the east side conditioner was used for the proposed activities.

A temporary laboratory was formed at the site to carry out the radiotracer handling activities. All the material required to carry out the activities like opening of the lead container, removal of can from the container, cutting the can, remotely transferring the radioactive powder into the boiling HCl, remote transfer of scandium chloride solution to the conditioner, agitation of the slurry and its disposal were arranged and safety precautions were taken to tackle any spillage or contamination due to radiotracer if it occurs. The area was covered with polythene sheets and absorbent sheets above them. The radiotracer lead container was kept in the vicinity of the laboratory.

About 2175kgs of fly ash was transported to the site and loaded into the conditioner (Figure: 8). Water was filled in the conditioner in 70:30 ratios to make the slurry.

A rowing boat was also procured to monitor the injected plume movement in the water of mine void.

**Labelling bulk fly ash**

A temporary laboratory (Figure: 9) was formed at the site to carry out the radiotracer handling activities.
All the material required to carry out these activities were arranged and necessary safety precautions were taken. 4 Litres of concentrated HCl was taken in a 5 litre beaker and heated till boiling using gas burner. Before actual handling of the radiotracer, cutting of dummy aluminium can, operation of peristaltic pump, related transfers were practiced. The entire operation was imitated by transporting the water into the conditioner. After conducting all the dummy trials, actual handling was done.

A can containing radioisotope Sc-46 as scandium oxide powder was removed from the transport container using long handled tongs (Figure: 10).

Figure 10: Removal of radioactive can from lead container

The can was placed in a lead die and sheared at the lid of the can remotely, using a long handled cutting tool, as shown in Figure: 11.

Figure 11: Cutting the aluminium can containing radioisotope

The irradiated scandium oxide powder was transferred from the cut open can into the beaker containing boiling concentrated hydrochloric acid as shown in Figure: 12.

Figure 12: Transferring radioisotope powder into the beaker

The powder was allowed to dissolve completely in the concentrated HCl for about half an hour. The peristaltic pump was kept ready with its one end having stainless steel tubing to be inserted in HCl in the beaker and another end already inserted in the slurry in conditioner.
The peristaltic pump was started and the prepared radioactive scandium chloride was slowly transferred in to slurry in the conditioner by keeping agitator rotating so that the fly ash remains in suspension and comes in maximum contact with the radiotracer molecules for effective labelling of fly ash with Sc-46 (Figure: 13).

![Installing the peristaltic pump](image)

Figure 13: Installing the peristaltic pump

The fly ash slurry was made in the conditioner. It was having screw type agitator and water sprinkler to make slurry of desired concentration and at the same time agitator will push the slurry ahead for the disposal.

**Disposal of radiotracer**

After 2 hours of mixing, the batch of labelled fly ash slurry was ready for the disposal. Flap of the conditioner was opened to dispose the Scandium-46 labelled slurry (radiotracer) into the mine void (Figure: 14).

![Disposal of labelled slurry](image)

Figure 14: Disposal of labelled slurry

While disposing the slurry, constant water flushing was done and in addition sprinklers were used to further enhance the disposal. Water flushing was continued till the radioactivity level on the surface of conditioner was brought down to the background level.

**Radiation Safety monitoring**

Before starting the preparations, all the personnel present and would be involved in the operations were given thorough briefing of the total activity planned. Wherever the radioactivity was in use, the polythene sheets were spread and covered with absorbent sheets. From beginning to the end of the entire operation, radiation monitoring was continuously carried out. After the radioactive handling job was finished all the personnel and area under use was specifically monitored (Figure: 15).
The area was thoroughly flushed with copious amount of water so that there are no traces of radioactivity (Figure: 16).

As shown in Figure: 6, a bore well BPZ1 was available on the southern bank of the void which was used to inject radiotracer. A new monitoring bore well BPZ3 of about 153 meters feet depth was drilled at about 20.3 meters from BPZ1 to perform double well tracer study in which the radiotracer Mo-99 as sodium molybdate was injected as radiotracer in BPZ1 (Figure: 17).

After injection of radiotracer in BPZ1, water samples were drawn from the depth of about 30 meters every day from BPZ3. The samples were sent to Radio Analytical Laboratory (RAL) of BRIT for assay of radiotracer content (Figure: 18).

Radiotracer monitoring in surface water

Since the radiotracer was disposed off in the void, the monitoring of its spread in the water of mine void was carried out next day. 1" diameter x1" height sodium iodide scintillation detector connected by about 25 meters cable to a scaler-ratemeter.
was inserted in a PVC pipe of about 3 meters length. The pipe was lowered in the void water from a rowing boat. The detector was moved from surface to the different depths at various locations in the water and the corresponding count rate was recorded with respect to the position of the monitoring boat (Figure: 19). The position was determined by a GPS device. Thus the count-rate data was obtained at various lateral and longitudinal locations and at various depths.

Schedule of ground water sampling

For double well tracer study sampling was scheduled for everyday till the breakthrough (appearance of Mo-99 in bore well samples). Once the breakthrough is achieved the frequency of sampling could be increased to thrice a day. This would give the information on the time required for the tracer injected in BPZ1 to travel up to the monitoring well BPZ3 which is 20.3 meters apart (Figure: 20).

The bore wells are specially drilled for monitoring Sc-46 radiotracer injected while the frequency of sampling was decided to be every week, till the breakthrough is achieved. After the breakthrough, this frequency could be increased to once in a day till the depletion in the count rate is observed in each of the bore well sample.

Analysis of bore well samples

The samples received from each bore well were filtered and filled in a standard counting container. The container was placed in a highly shielded 3" x3" Sodium Iodide scintillation detector coupled to a multichannel analyser at RAL, Vashi, as shown in Figure 18. It was ensured that the background count rate is minimal. Each bore well sample was assayed for 10,000 seconds. In multichannel analyser a spectrum of all the available energies is obtained. In the spectrum few peaks like K-40, Pb-210 are obtained as these are naturally present in the background. Existence and Location of the peaks ensures the quality of counting. When the injected radiotracer will appear in the sample, distinct peaks corresponding to the respective energies to those radioisotopes will be observed. Area under the specific peaks is recorded and compared with the standards to estimate the quantity of
radiotracer present in the sample. Various samples collected so far from the date of injection are tabulated and subsequently their assay results are also attached.

**Results**

**Outcomes from the double well study**

This is a short term study to determine the progress of radiotracer injected in one bore well i.e. BPZ1 to another bore well BPZ3. This will give us the ground water velocity in the topography and geology present in the vicinity of mine void. From the samples drawn for 40 days, Mo-99 could not be observed in any of the samples. The distance between the two bore wells is 20.3 meters. Thus the velocity of the tracer in this area could be less than 0.5 meters per day or even less. Further sampling will be continued till the arrival of the tracer in the monitoring well.

**Isocount contours in the mine void water**

The count-rate data received at given latitude and longitude location was plotted with respect to various depths to get isocount contours for that depth. From the isocount contours, the spread of radiotracer at various depths can be estimated. The isocount contours shown below are for monitoring one each at surface (Figure: 21), 1 meter depth (Figure: 22) and 2 meters depth (Figure: 23).

The spread of radiotracer (velocity of transport of radiotracer in the surface water) in the mine void with respect to time could be obtained when the second tracking after about 50 days is completed.
Progress of radiotracer in the ground water

Water samples are drawn as per schedule from all the designated bore wells weekly for the assay of Scandium-46. As soon as the samples are received, those are assayed on gamma ray spectrometer (multichannel analyser) at RAL, Vashi, to determine the radiotracer content. However, the samples did not show the tracers of the presence of Sc-46.

Discussion

Aim of the double well study was to determine the movement of heavy metal like Mo-99 (molybdate form) in the geology and topography around the mine void. Selection of the tracer for this purpose was on the basis of its similarity with other heavy metals, with respect to their mobility and movement in aqueous environment. Short half-life of the radiotracer was selected because the modelling carried out by other study groups, indicated its rapid movement. However, after the results obtained from this study so far, the postulate has not been verified, meaning that heavy metals do not follow the normal water movement characteristics underground. It appears that they are retarded or slowed down due to repelling charges on the soil matrix available below the ground surface.

As regards Sc-46 as a radiotracer, it needs to be homogenously spread throughout the water in the mine void, so that it can have maximum surface contact with the rocks and its fractures surrounding the void and thus percolating through them to reach to the observation bore wells in the same manner as the groundwater. It has been observed that the injected radiotracer follows the form of a plume in the void water near the injection point, which is evident from the isocount contours obtained at various depths. Heavier particles from the fly ash could be settling on the bottom surface with a localized spread. Column of the plume rises to the surface with confined dimensions and the good amount of spread of the radiotracer is observed on the surface, indicating lighter particles tend to float on the surface.

However, leaching process of the radiotracer (labelled fly ash) may be in action at various depths in the water due to physical churning of the fly ash with water molecules. The same could be in place when the finer particles escape through the cracks in the surrounding rock of the mine void and fractures. Due to their (of finer particles) physical friction with the soil particles, Sc-46 may appear in the water samples drawn from the bore wells. In the reported period of nearly 45 days, no water samples could show the presence of Sc-46.

Conclusion

Double well tracer study has shown that the radiotracer did not reach to the monitoring well, indicating very low permeability of the underground soil matrix in the mine void region of quarry number 4 of Jagannath OCP.
Scandium-46 leached out from labelled fly ash could not be detected in the bore wells surrounding the quarry number 4 of Jagannath OCP, indicating no leachates are reaching the groundwater aquifers from the time of injection.

Further dumping of fly ash could be continued into the mine void to push particulate matter towards the boundaries of the void, forcing the labeled fly ash towards the bore wells and to ascertain the impact of leachates in future. Sampling of the water in the designated bore wells should be continued for about five half-lives of the radiotracer.

While regular dumping, subsequent radiotracer study can be repeated after every two years.
Scientific Highlights during the Calendar Year 2017

1. During January to March 2017, Radiation Physics Group officers co-ordinated the BRIT visit of DRP trainees, DAE-NUJ participants, NFC-TSO’s and NDRF personnel’s along with lecture on various activities of BRIT.

2. First Training Course for Radiological Safety Officers for ‘Gamma and Electron beam Irradiation Facilities’ was conducted at BRIT, Vashi Complex and lectures as well as requisite training on Radiation Safety, Calibration of survey meters and pocket dosimeters, were imparted by the officers of Radiation Physics Group, Calibration services Group and Radiation Processing Plant.

3. A Senior Scientific Officer of Regional Centre for Radiopharmaceuticals, Kolkata was part of the team of authors to write a book on ‘Cyclotron Based Production of Technetium-99m’ which was published by ‘International Atomic Energy Agency’ during June 2017.

4. Radiopharmaceutical Programme, BRIT has launched new ready-to-use $^{177}$Lu-DOTA-TATE injection (Code: LUM-3) useful for therapeutic treatment of somatostatin receptor positive neuroendocrine tumors during 2017. Also, a therapeutic product, $^{131}$I-Lipiodal Injection (Code: IOM-40) useful for treatment of liver cancer was launched by BRIT in association with RPhD, BARC.

5. Scientific Officers of Radiopharmaceutical Programme, Quality Control Programme were involved in co-ordinating BRIT visit and provided lectures/practical demonstration for DRP, DRM/DMRIT students and students from Kundanani College of Pharmacy during 2017.

6. Two MOUs were signed with private entrepreneurs for setting up of new Gamma Radiation Processing Plant, one in Madhya Pradesh and the other in Andhra Pradesh.


8. Cobalt-60 Teletherapy source (CTS) (180RMM) was exported to Kenyatta National Hospital, Kenya, while Co-60 Irradiator Source (125KCi) was exported to Srilanka Atomic Energy Board for source replenishment in Multipurpose Gamma Irradiator at Biyagama.


10. BRIT was successful to design and fabricate new Type-A Packages for transport of radioactive material in liquid form in bulk (18Ci of I-131), which was approved by AERB and is in regular use for transport of I-131 from BARC to BRIT, Vashi Complex. This has reduced the number of packages during transport by a factor of 4.
Labelled Compounds Programme

Compiled, Edited and Published by
Dr. Tarveen Karir
Manager, Scientific Information Resources & Publications,
Corporate Planning Division,
Board of Radiation & Isotope Technology, DAE

Printed by
M/S Sundaram Art Printing Press
Wadala, Mumbai