RADIOISOTOPES FOR AFFORDABLE HEALTHCARE

Recovery of Valuable Radionuclides from High Level Waste

C. P. Kaushik

Utilization of the radioisotopes in medical applications is one of the prime objectives of the Indian nuclear program. Towards this, radioisotopes have been produced in nuclear reactors through the activation route. Of late, with the development of advanced separation techniques, it is possible to recover the radioisotopes from high level nuclear waste for medical applications. The high level waste generated from reprocessing the spent fuel contains many radioisotopes in significant quantities as produced during the fission of ^{235}U.

Table 1 lists some useful fission products present in the high level waste. Successful recovery and utilization of these isotopes in medical applications have led to the concept of treating nuclear waste as a source of wealth[1].

Towards utilization of radioisotopes, India has taken the lead role for their bulk scale recovery from high level waste. Further, the recovered isotopes are purified in a radio-chemically pure form meeting stringent regulatory limits.

Recovery of ^{137}Cs is accomplished through a novel solvent extraction-based process utilizing 1, 3-di-n-octyloxy Calix[4] arene-Crown-6 (CC6) in isodecayl alcohol (IDA)-dodacane solvent system. Stripping of Cs from the loaded organic phase is carried out using dilute nitric acid. The cesium rich product stream is further concentrated and used to make Cs glass pencils for gamma irradiation applications.

^{90}Y ($t_{½}$=64 h), produced from the beta decay of ^{90}Sr ($t_{½}$=30 y), has unique application as a radiopharmaceutical in treating liver cancer and neuroendocrine tumours. A series of separation processes has evolved towards isolation of the radioisotope to meet stringent purity requirements for nuclear medicine. The bulk separation of ^{90}Sr from Cs-lean HLW is carried out by solvent extraction using Tetra Ethyl Hexyl Di-Glyco Amide (TEHDGA) in IDA-Dodecane. It is to be noted that TEHDGA is used for the co-extraction of actinides (An) and Lanthanides(Ln) along with Strontium(Sr) from high active acidic streams. The stripping of the radionuclides from the loaded TEHDGA phase is carried out using dilute HNO_3(0.01 M).The product, Sr-An-Ln rich, is further concentrated and used for recovery of bulk amount of Strontium. Multi-step separation processes for the purification of Sr deployed involving solvent extraction by CMPO followed by chemical precipitation. The recovered Strontium product is used for milking out Yttrium-90 (^{90}Y).

^{106}Ru is in secular equilibrium with its daughter ^{106}Rh. It is a source of high energy beta radiation emitted from the decay of 106Rh to stable Pd and is useful for brachytherapy applications, particularly for eye cancer treatment. Separation of ^{106}Ru is carried out from post TEHDGA cycle raffinate as discussed above. The process utilizes oxidation of Runitrosyl nitrate to RuO_4 by KIO_4 followed by extraction of RuO_4 in chlorinated CCl_4. Finally, the extracted Ru is stripped from CCl_4 using acidic hydrazine solution. Preparation of brachytherapy source from purified ^{106}Ru solution includes electro-deposition of ^{106}Ru on a silver substrate followed by the production of sealed sources in the form of a plaque.

Subsequent articles of this segment highlight the medical applications of these three fission products.

Table 1: A list of valuable radionuclides present in the HLW

Radioisotopes	Half - life	Radiation type	Energy (MeV)	The primary area of applications
Cesium ^{137}Cs	30 y	Gamma (after emitting a Beta radiation)	0.66	Blood irradiation
Strontium/Yttrium $^{90}Sr/^{90}Y$	28 y/64 d	Beta	0.5 and 2.28	90Y used as a Radio-pharmaceutical for treatment of liver cancer, bone pain palliation
Ruthenium ^{106}Ru	365 d	Beta	3.54	Eye cancer (Brachytherapy)

Cesium-137 Glass Pencil Source for Blood Irradiators

C. P. Kaushik

Production of ^{137}Cs based blood irradiators is one of the examples of the successful deployment of radioisotopes recovered from High-Level Radioactive Waste (HLW) in medical industry. With this development, India has become the first country in the world to deploy ^{137}Cs in non-dispersible glassy form as the source in blood irradiators. This development will eventually replace the existing 60Co based blood irradiators in the country and abroad. The use of ^{137}Cs in blood irradiators is advantageous due to the its longer half-life, which reduces source replenishment frequency and man-rem consumption. Further, it is possible to build compact irradiators due to lower shielding requirement. Irradiation of blood to prevent Transfusion Associated–Graft Vs Host Disease (TA-GVHD) has to be done just before its transfusion to immuno-deficient patients like newborn babies, patients undergoing heart or bone-marrow transplant and immune-competent patients who have 1st degree relation with donor. This necessitates placement of irradiator within the hospital premises. The smaller footprint for housing the blood irradiator will, thus, be beneficial especially to small hospitals.

Production of ^{137}Cs-glass source for blood irradiator envisages industrial scale recovery of this radio element from high-level radioactive waste and its subsequent immobilization into a specially formulated glass matrix. Such non-dispersive glassy form of caesium is then metered into the small-sized stainless-steel pencils and sealed by remote welding. Post sealing, pencils are subjected to over packing, surface decontamination and stringent quality assurance checks, at par with international standards (Fig.1). Each pencil contains about 200 g of Cs-glass, amounting to about 300 Ci of ^{137}Cs. More than 200 numbers of Cs-glass pencils have been produced successfully till date

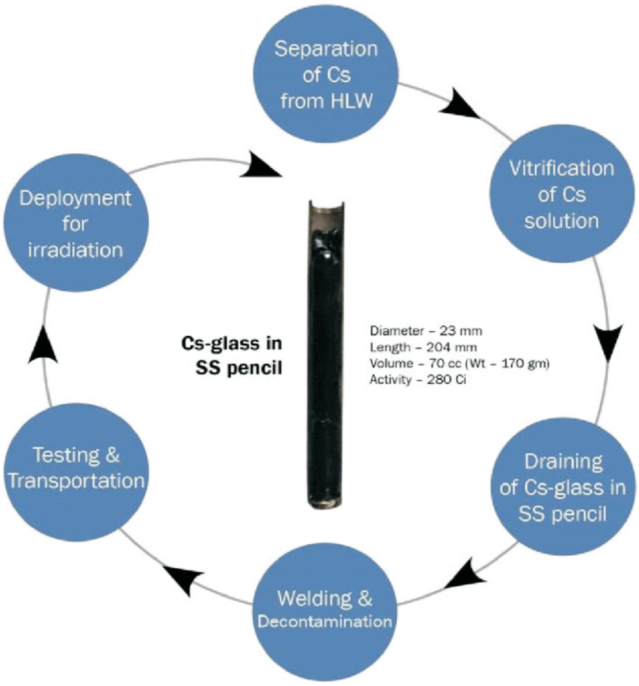

Fig.1: A thumbnail view of ^{137}Cs glass-pencil making process.

Fig.2: Stock image of ^{137}Cs based Blood Irradiators (BI-2000).

Qualified Cs-pencils are transported to BRIT for deployment in blood irradiators and subsequent supply to different hospitals. Each blood irradiator houses ten such pencils in a circular cage. This provides a dose-rate of about 11 Gy/minute, suitable for irradiating a standard blood bag within 3 minutes, with an estimated absorbed dose of about 30 Gy. Fig.2 shows the photograph of the ^{137}Cs based irradiator (BI-2000). At present, more than fifteen such irradiators are in use at leading hospitals of the country.

Clinical Grade ^{90}Y-acetate from Ultra-pure ^{90}Sr

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Yttrium-90, a pure β-emitter (E_{max}=2.28MeV, $T_{½}$ = 64.1h) formed by β- decay of ^{90}Sr, is a potential therapeutic radionuclide. It is widely used in the treatment of hepatocellular carcinoma (HCC), leukaemia, lymphoma, and a range of tumours. Application of ^{90}Y is dependent on the supply of carrier-free, clinical-grade ^{90}Y, which can be separated from highly pure ^{90}Sr.

Ultra-pure ^{90}Sr is being separated from the product streams generated during the partitioning process of HLLW employing a multi-step separation technique using solvent extraction, extraction chromatography, radiochemical precipitation and membrane separation. An indigenously developed two-stage SLM based ^{90}Sr-^{90}Y generator system is utilized for milking carrier-free ^{90}Y [Fig. 1]. The radionuclide impurity of the separated ^{90}Y-acetate product has a β-content of ^{90}Sr less than 10^{-6}Ci/ Ci of ^{90}Y and gross α-activity less than 10^{-9}Ci/Ci of ^{90}Y, which is well within the permissible limit as per the European Pharmacopia. The separated clinical-grade carrier-free ^{90}Y is supplied to Radiation Medicine Centre (RMC), Parel, for cancer therapy.

Fig.1: ^{90}Sr-^{90}Y generator.

Extensive studies were undertaken to optimize various parameters for the transport rate and purity of ^{90}Y products in the generator system. Based on these studies, clinical-grade ^{90}Y-acetate in lots of ~140 mCi are regularly (about 15 lots per year) separated and transported to RMC, Parel, following BSC approved transport guidelines, for therapeutic applications. The feedback received from RMC on the purity and effectiveness of the separated ^{90}Y was excellent. Developmental studies for scaling up are under progress to meet the rising demand for ^{90}Y supply.

RuBy plaque and Simulation Software for Eye Cancer Treatment

C. P. Kaushik

Nuclear Recycle Group of Bhabha Atomic Research Centre has indigenously developed Ruthenium Brachytherapy (RuBy) Plaque for the treatment of eye cancers. RuBy plaque contains Ruthenium-106 radioisotope. The unique radiochemical characteristics of the radioisotope such as emission of high energy beta radiation (3.54 MeV) and rapid beta radiation dose rate fall off make it useful for selective killing of cancer cells without damaging the healthy organs of the eye.

RuBy plaques of two different configurations (round and notched) have been successfully deployed for the treatment of eye cancers. Round plaque is used commonly whereas notched configuration is suitable for the treatment of eye cancers located adjacent to the optic nerve. The first use of the indigenous plaque was carried out on 21st August 2019 by Dr. Santosh Honavar at the Centre for Sight Hospital, Hyderabad for treating a patient with ocular surface squamous neoplasia and the results were very encouraging with no evidence of local tumor recurrence, well-maintained and complete vision. Presently, RuBy plaques are being used at seven hospitals in the country (see Table 1) and more than fifty patients have been treated. All post-treatment results are highly satisfactory, confirming the fact that RuBy plaques have gained the trust of doctors and patients alike. More hospitals are progressively adopting RuBy plaques for better and cost-effective treatment of eye cancer.

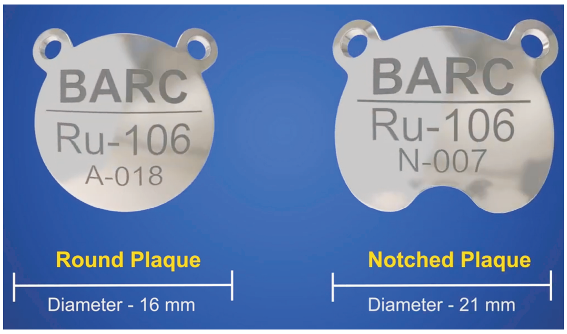

Fig.1: RuBy plaques for the treatment of eye cancers.

Table 1: List of hospitals using RuBy plaques

1	Centre for Sight Hospital, Hyderabad
2	Sankara Eye Hospital, Bangalore
3	RP Centre, AIIMS, Delhi
4	Army Hospital, Delhi
5	Ramakrishna Eye Hospital, Coimbatore
6	Hinduja Hospital, Mumbai
7	Fortis Hospital, Haryana

Complementing the RuBy plaques, NRG, BARC has also developed Plaque Simulation software to assist doctors with optimal treatment planning. The software provides an easy-to-use interface for calibration and dose rate distribution of the RuBy plaques. Fig.2 shows software predicted dose-rate distribution for a given tumor irradiated by a RuBy plaque. It predicts treatment duration and dose received by other healthy parts of the eye. This information is useful for the convenient and optimal planning of treatment for eye cancer.

Indigenous development of RuBy plaques and the Plaque Simulator are breakthroughs in the area of AtmaNirbhar healthcare and providing affordable treatment to eye cancer patients in India. RuBy plaques are marketed by the Board of Radiation & Isotope Technology (BRIT). Detailed information about the products is available at *"www.britatom.gov.in"*.

Input parameters	
Plaque ID	: A59 (Round plaque)
Activity	: 31.5 MBq as on Jan 27, 2021
Tumor radial dimension	: 8 mm
Tumor thickness	: 6 mm
Results	
Date of implant	: Sep 5, 2021
Ave dose rate	: 148 mGy/min
Apex point dose	: 100 Gy
Implant duration	: 35 h
Foveola point dose	: 26 Gy

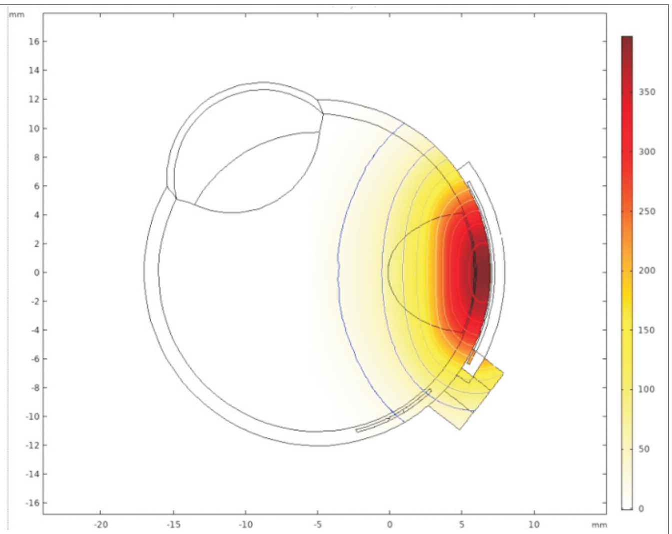

Fig.2: A typical results generated from RuBy Plaque simulator for a given tumor treatmnet by a RuBy plaque.