Development of ^{106}Ru bearing Sealed Source for Eye Cancer Treatment Applications

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Abstract

Use of ^{106}Ru plaque in brachytherapy is a proven technique for treatment of different eye cancers. However, ubiquitous availability of this treatment modality is limited due to highly expensive imported sources. The present communication showcases indigenous development of ^{106}Ru based plaque source, starting from separation of the fissiogenic radionuclide from High Level Waste (HLW) followed by its immobilization onto silver substrate and finally its encapsulation into silver plaque. Evidently, availability of indigenous sources will greatly help in reducing the cost of brachytherapy and help save vision.

Keywords : ^{106}Ru plaque, eye cancer, ocular tumor, radioactive waste, electrodeposition, sealed source.

Introduction

Choroidal melanoma and retinoblastoma are the most commonly occurring cancers of the eye in adults and in children, respectively [1–3]. As the first solution, ophthalmic oncologists prefer plaque radiotherapy owing to the simplicity and flexibility associated with the technique [1, 3]. Indeed, depending upon the location and extent of the melanoma, plaques with different geometries and varying strength are being used for irradiation [1]. Various radionuclides such as ^{60}Co, ^{106}Ru, ^{125}I, ^{103}Pd, ^{90}Sr, and ^{131}Cs were used in making ophthalmic plaques. However, most commonly used ophthalmic plaques are ^{125}I and ^{106}Ru [1, 4]. ^{106}Ru decays by emission of high energy β-rays, while ^{125}I decays by electron capture to an excited state of Tellurium-125 and thereby emits low energy gamma radiation. Indigenous development of ^{125}I plaque has been discussed elsewhere [5]. Present efforts were directed to develop ^{106}Ru plaque indigenously.

The use of ^{106}Ru based plaque is more convenient to doctors. This may possibly due to relatively longer half life of ^{106}Ru. Further, β-radiation has a limited range, leading to a steep dose fall-off, allowing tumours (upto 5mm) to be treated while minimizing inadvertent irradiation of sensitive ocular structures such as the fovea or the optic disc [1 – 4].

Unlike ^{125}I, which can easily be synthesized in chemically pure form, ^{106}Ru is available only in the nuclear waste generated from reprocessing of spent fuel. Therefore, isolation of the fission product in chemically pure form is a herculean task. This may be one of the reasons that no Ru plaque has been prepared after separation of the fission products from High Level Waste (HLW). In India, the management of HLW is being carried out by partitioning of long-lived minor actinides and valuable fission products such as ^{137}Cs and ^{90}Sr. The activity lean solution is a treasure trove of ^{106}Ru. Processing a cubic meter of the solution will be adequate for the preparation of thousands of ophthalmic plaques.

In view of the abundant availability of the radioisotope and its potential use in eye cancer treatment, concerted efforts were made, culminating in the fabrication of ^{106}Ru plaque suitable for brachytherapy applications. In this paper, salient features of process development, including flow sheet finalization for recovery of radio-chemically pure ^{106}Ru, fixation of the recovered ^{106}Ru on silver substrate by electro-deposition and fabrication of sealed source have been highlighted.

Experimental

The actual HLW of research reactor origin was initially subjected to separation of ^{137}Cs by Calix crown, followed by co-extraction of minor actinide and Sr by TEHDGA. The resulting activity lean solution is used as the feed in the reported study. Purification of ^{106}Ru in radio chemically pure form was carried out as per the flow diagram given in **Fig.1**. In the first step, KIO_4 (s) was added to convert Ru species to RuO_4, which was then extracted using CCl_4 pre-equilibrated with Cl_2. The extracted ruthenium was stripped back using 0.1 M hydrazine in 1 M nitric acid. The strip solution was dried and the residue after dissolving in 4 % sulphamic acid was used as catholyte. The ^{106}Ru was electrochemically deposited on Ag cathode using Pt anode and sulphamic acid as anolyte. The Ru deposited cathode was further used for plaque preparation.

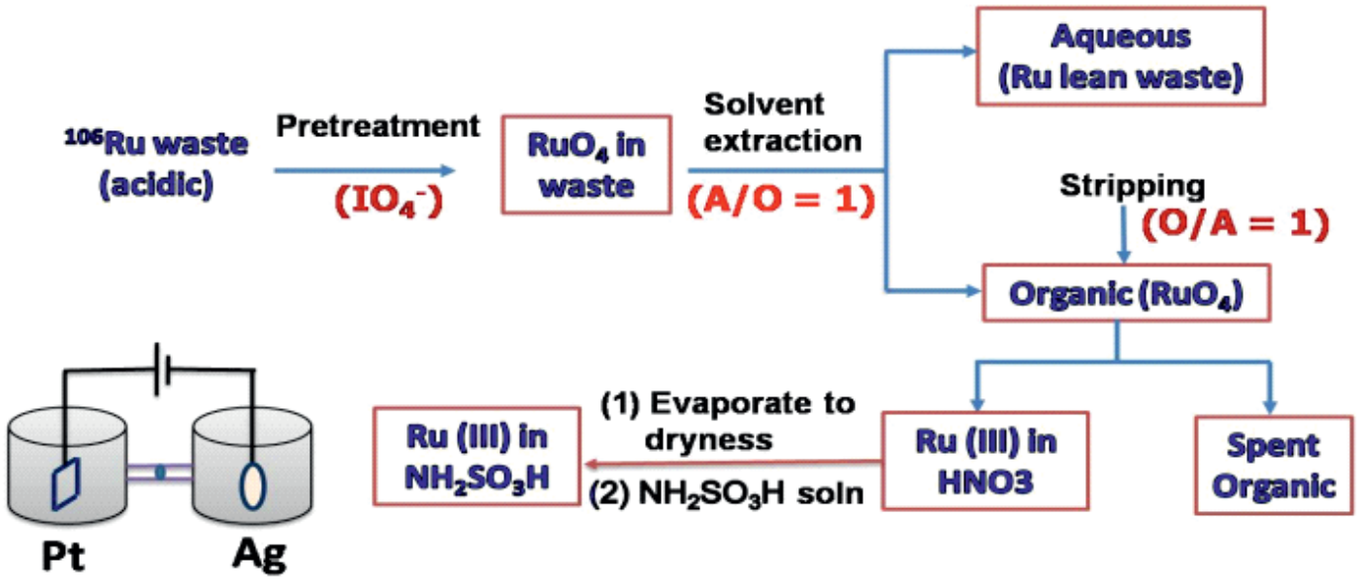

Fig. 1: Schematic flow sheet depicting ^{106}Ru extraction from HLW and subsequent electrodeposition on Ag

The plaque consists of a 0.9 mm thick Ag backing plate (dia 15.8 mm), the 0.2 mm thick Ru coated Ag substrate and a 0.1mm thick Ag window (dia 16 mm). The entire assembly was sealed using an Ag based brazing alloy. For the sealing operation, the plaque was assembled on a graphite jig, which was then placed in an all quartz bell jar assembly maintaining an Ar atmosphere. This assembly was introduced into the furnace preheated at 870°C in a controlled manner and withdrawn after about 2 minutes, for cooling under ambient condition. Post cooling, the plaque was bent to the required radius using a die.

Results and Discussions

The waste feed used in this study was relatively clean and contains only traces of ^{137}Cs (0.1 mCi/l) and almost equal amount of ^{106}Ru and ^{125}Sb (30 mCi/l). For the selective separation, Ru species were oxidized to RuO_4 and extracted in CCl_4. The extent of extraction was studied by varying aqueous to organic (A:O) ratio. It was found that almost 80% of ^{106}Ru can be extracted using A:O ratio of 1 in a single contact. The decrease of organic volume reduced the extent of extraction, but not significantly. Almost 73% of ruthenium was extracted to the organic phase in a single contact using A:O ratio of 3. It was observed that extraction of RuO_4 in CCl_4 is quite rapid and complete equilibration was attained within 10 minutes. This is possibly due to very high solubility of the RuO_4 in CCl_4. As RuO_4 is highly volatile, extraction was carried out under closed conditions to minimize the volatilization loss of the radioelement. Further, RuO_4 is highly unstable and can be reduced easily to RuO_2 upon contact with any organic medium. An oxidative condition is therefore mandatory to maintain RuO_4 in CCl_4. This was achieved using Cl_2 gas equilibrated CCl_4. Stripping of the RuO_4 from CCl_4 phase was carried out after reducing the RuO_4 to Ru(III).

Considering the source preparation by electro-deposition method, a medium directly suitable for electrolysis such as sulphamic acid, was considered to be the ideal choice. The use of sulphamic acid, alone and with hydrazine was attempted. Due to poor stripping efficiency of the previously mentioned processes, hydrazine in HNO_3 was used. Under these conditions also, a long equilibration time of at least 1 hour was required to achieve quantitative stripping. The stripped solution was analyzed using HPGE detector coupled with 8K MCA. No peaks other than gamma peaks for ^{106}Ru are seen, indicating that the process is effective in separating Ru in radiochemically pure form. By adjusting, aqueous organic ratio, both in extraction and stripping steps, it is possible to get a concentrated solution of ^{106}Ru.

Prior to electrodeposition of ^{106}Ru, the strip solution was evaporated to mitigate nitrate interference during deposition. The residue was dissolved in 4% (wt.) sulphamic acid and used as catholyte. It is to be noted that the Ru concentration in the catholyte is quite low (< 300 ppm) and about 300 μCi of ^{106}Ru needs to be deposited on a substrate of diameter 12.7 mm. To achieve this, while obtaining a uniform and adherent coating, several inactive coating trials were carried using simulated waste. The coated samples were analyzed using SEM and EDS, with the results being presented in **Fig. 2**.

An inspection of **Fig. 2** indicates that in case of high current density of 65 $mA.cm^{-2}$ (left), the coating is thick and layered. This coating is not uniform and quite rough. Further, the colour of the coating in case of high current density is black indicating some oxidation of the deposited Ru.

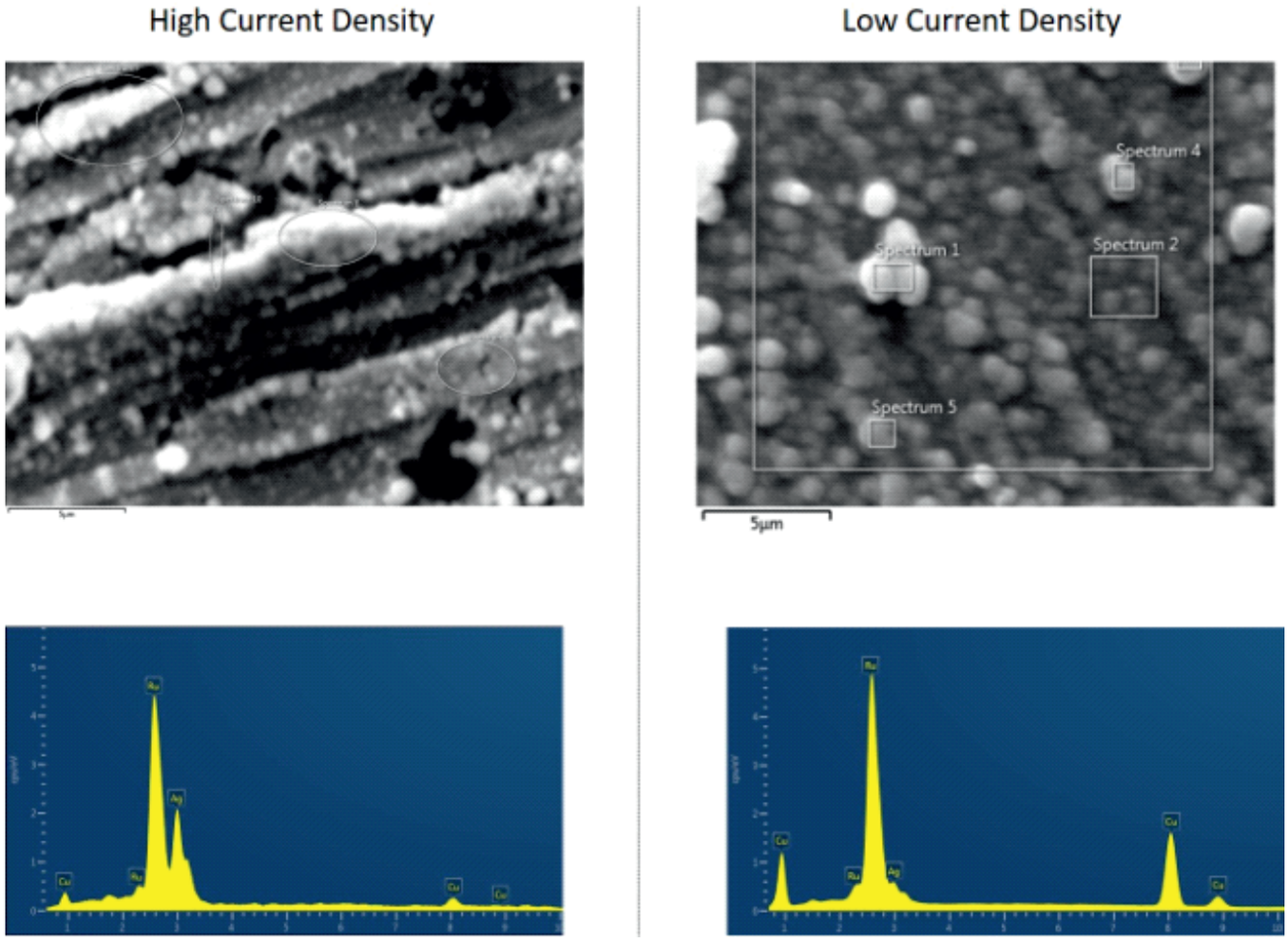

Fig. 2: SEM micrograph of ^{106}Ru deposited electrolytically on Ag at different current densities. The lower images are the EDS spectra of these samples

In comparison, at low current density of 5 mA.cm^{-2}, (right), a more uniform coating of Ru metal on the substrate is obtained. Oxidation is also lesser than the high current density case, as evident from the silvery grey colour of the coating. The optimized current density of 5 mA.cm^{-2} was selected and coating studies were carried out with active Ru containing solutions for various periods up to ~ 80 h. The optimized electrodeposition conditions are in good agreement with the earlier study [6]. In all cases, the coated source was washed thoroughly in boiling water before dispatch for activity measurement. **Table 2** collects the activity and dose rate profile of the sources.

The final stage is the fabrication of the ^{106}Ru sealed source. Briefly, the sealed source comprises the 0.2 mm thick source piece in a sandwiched geometry between a 0.9 mm thick backing plate and a 0.1 mm thick window. As mentioned previously, sealing was carried out under Ar atmosphere. This served the twin objectives of minimization of Ru volatilization and preservation of the graphite jig.

After preparation of more than 20 inactive plaques, first active source sealing was carried out using a 25 µCi source. During the sealing air activity was constantly monitored to check for Ru escape. Almost negligible Ru counts were recorded on the sample. Finally, the sealed plaque was bent to the required radius on a die. A file photograph of a Ru plaque is presented in **Fig. 3**.

Table 2: Activity and dose profiles of various ^{106}Ru plaque sources

Sample no	^{106}Ru activity (µCi)	Dose rate at 1 cm (mR/h)
Ru-1	80	100
Ru-2	50	50
Ru-3	25	40
Ru-4	210	400
Ru-5	282	600
Ru-6	367	600

The active Ru plaque was tested for surface contamination by swipe samples and sealing integrity was ascertained by boiling water test for 3 cycles of 20 minutes each. Counting revealed no activity in the immersion liquid. All inactive plaques qualified type classification tests carried out as per AERB SS3 procedure. The plaque design has got approval from AERB for clinical trials at hospitals.

Conclusion

The present study demonstrates isolation of radiochemically pure ^{106}Ru from HLW. An electrodeposition

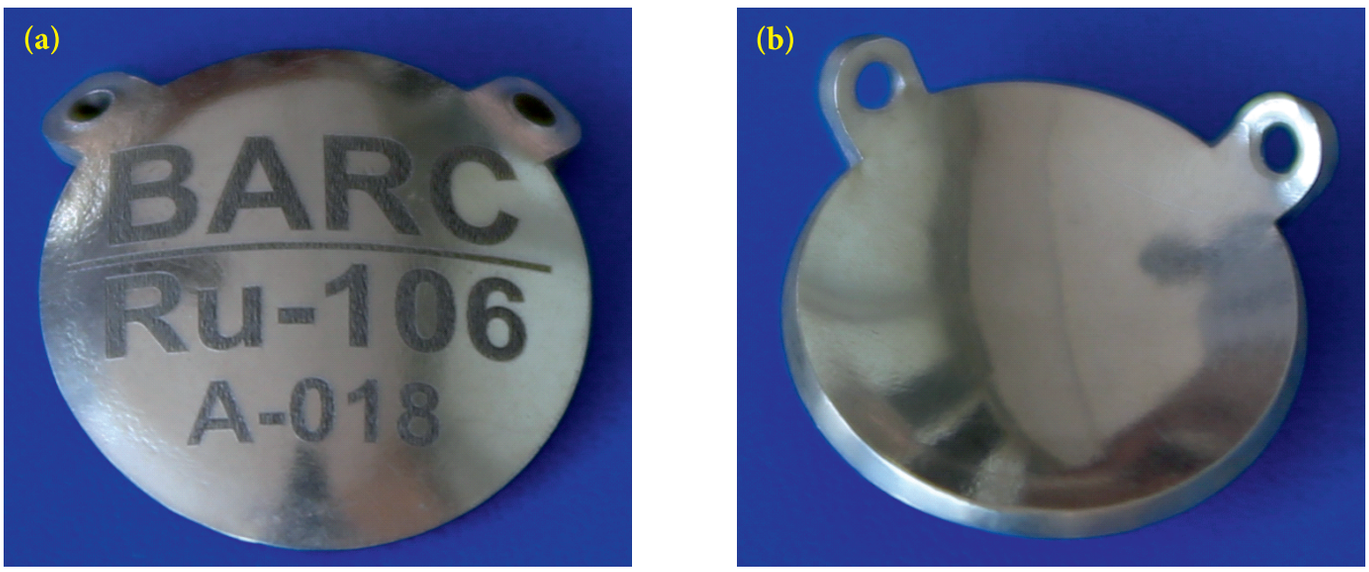

Fig. 3: File photograph of ^{106}Ru plaque (a) Top view and (b) Rear view

process for immobilization of metallic ^{106}Ru on Ag substrate has been optimized and used for source preparation. An innovative sealing process led to the realization of ^{106}Ru brachytherapy plaque. Indeed, this is the third radionuclide, after ^{137}Cs and ^{90}Sr, to be isolated from HLW for societal benefit.

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Acknowledgements

The authors wholeheartedly thank Messer Ajay Manjrekar, Sunil Babu Shinde, S. Langhi, V. M. Phalke, V. K. Kulkarni and Anil Kumar for their involvement in various stages of this work. Sincere thanks to Drs. R. K. Mishra, Amar Kumar and Darshit Mehta for providing Cs lean waste solution used in this study.

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